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DETERMINATION OF RADIONUCLIDES AND POTENTIALLY TOXIC ELEMENTS ASSOCIATED WITH THE SPOIL BANK REMEDIATION ON THE ENVIRONMENT AT THE ABANDONED URANIUM MINING AREA

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Title: Determination of radionuclides and potentially toxic elements associated with the spoil bank remediation on the environment at the abandoned uranium mining area

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LIST OF ACRONYMS AND CODES

EU	-	European Union
OECD	-	Organisation for Economic Co-operation and Development
IAEA	-	International Atomic Energy Agency
NEA	-	Nuclear Energy Agency
ICRP	-	International Radiation Safety Committee
UNSCEAR	-	The United Nations Scientific Committee on the Effects of Atomic Radiation
WHO	-	World Health Organization
UNESCO	-	United Nations Educational, Scientific and Cultural Organization United
UNEP	-	Nations Environment Programme
WNA	-	World Nuclear Association
FAO	-	Food and Agriculture Organization of the United Nations
BCR	-	European Community Bureau of Reference
AMD	-	Acid mine drainage
PTE	-	Potentially toxic elements
TERNOM	-	Technologically enhanced naturally occurring radioactive material
K_d	-	Solid-liquid distribution coefficient or partition coefficient
MF	-	Mobility factor
TF or TC or CR	-	Transfer factor or Transfer coefficient or Concentration ratio
CEC	-	Cation exchange capacity
SOM	-	Soil organic matter
OM	-	Organic matter
ICP-OES	-	Inductively coupled plasma optical emission spectrometry
ICP-MS	-	Inductively coupled plasma mass spectrometry
FAAS	-	Flame atomic absorption spectrometry
AFS	-	Atomic fluorescence spectroscopy

AAS - Atomic absorption spectrometry

AES/AFS - Atomic emission/ fluorescence spectrometry

NAA - Neutron activation analysis

XRF - X-ray fluorescence

AVS - Anodic striping voltammetry

HPGe - High Purity Germanium

FWHM - Full width at half maximum

MDA - Minimum Detectable Activity

1. BACKGROUND OF THE WORK AND ITS AIMS

This chapter begins with the background of the research study, where the existing data is discussed. It highlights the history of uranium mining in South Africa and Hungary. It follows with the research problem where the issue is identified with supporting data. The aim of the study is clearly stated, and the steps that were taken to achieve the aim are listed on the research objectives. Finally, the rationale highlights the importance of this research study.

1.1. Background

The European Union (EU) defines the proper management of mines and mining waste through a combination of economic, health and safety, and human rights regulations, including requirements for mine operation and closure criteria. The EU member states are bound by these rights and charters and their institutions, ensuring the overall security of some human rights that mining activities could impair. Similar procedures were adopted to boost uranium mining regimes in countries that are not part of EU member states, such as Australia and South Africa (Organisation for Economic Co-operation and Development (OECD), 2014). The Organisation for Economic Co-operation and Development and the International Atomic Energy Agency (IAEA) (1999) reported environmental activities in uranium mining and milling. In this report, an overview of ecological activities related to uranium production was presented. The report also reflected on the increasing awareness in all countries of the need for environmental protection. According to OECD/IAEA (1999), large programs have been underway in several countries to remediate closed uranium mines and mills. The activities and concerns raised in this report were based on survey responses from 29 countries, including South Africa and Hungary.

1.1.1. Uranium mining in South Africa

The mining industry played an essential role in South Africa’s economic, social and political environment since the discovery of gold resources in 1886 (The Atomic Energy Corporation of South Africa Limited, 1988; Olowoyo *et al.*, 2013). Since the late 19th century, the Witwatersrand area near Johannesburg in South Africa have been mined for gold, with the associated uranium deposited as mine waste (primarily tailings), until the second half of the 20th century when uranium was extracted as a by-product of gold mining (Merkel and Hasche-berger, 2008; Dasnois, 2012). In the 1980s, South Africa had a 14.9% and 12.4% share of the world's top three in uranium wealth and production (The Atomic Energy Corporation of South Africa Limited, 1988). Owing to mining activities in South Africa, environmental degradation has become a subject of intense public discussion and scrutiny in recent years (Waggitt, 2008). Unfortunately, this is just an overview of the inheritance of South Africa mine waste (Trangos and Bobbins, 2015).

According to Trangos and Bobbins (2015), mining firms in South Africa had little regard for the social and environmental effects before 1991. Due to the old mines being unprofitable and abandoned, the mine waste was often left untreated and damaged landscapes were left unrehabilitated (Trangos and Bobbins, 2015). South Africa produced vast amounts of tailings during more than a century of gold mining, now covering large areas in densely populated regions. Such tailings are susceptible to generating acid mine drainage (AMD) (Rösner and Van Schalkwyk, 2000). Acid mine drainage and the release of radionuclides and potentially toxic elements (PTEs) such as radon (Rn), aluminum (Al), manganese (Mn), and arsenic (As) are key environmental impacts related to uranium (U) mining (Matshusa and Makgae, 2017). Numerous studies performed in Witwatersrand, gold mining areas, have provided growing evidence that mines often contaminate neighboring habitats with potentially toxic elements, salts, and radionuclides which consist primarily of uranium isotopes and their radioactive daughter products (Rösner and Van Schalkwyk, 2000; Winde and Sandham, 2004; Liefferink, 2008). Owing to the long-ignored transportation of dissolved uranium from gold mines, the discharge containing large concentrations of dissolved pollutants migrated from tailing deposits into neighboring aquifers and eventually joined neighboring streams (Pretorius and Hattingh, 2009). Thus, gold mine tailings can be considered a primary source of soil and groundwater contamination in South African mining areas (Rösner and Van Schalkwyk, 2000). Hence, mine closures and, in particular, environmental restoration must be dealt with urgently (Durand, 2012).

In the study conducted by Rösner and Van Schalkwyk (2000), it was recommended that since environmental pollution is expected to be prolonged due to gold mining activities, it is necessary to understand the mobility and thus the bioavailability of contaminants and the capacity of the soil to retain and mitigate such pollutants in the long term.

1.1.2. Uranium mining in Hungary

Numerous pilot projects for the closure and rehabilitation of the uranium mines from various Central and Eastern European countries (Czech Republic, Slovakia, Albania, Estonia, Hungary, Romania, and Bulgaria) were suggested in the late 1990s (George-Laurentiu *et al.*, 2016). The industrial uranium mining area in Hungary is in the Mecsek mountains in the Southern Transdanubian region, approximately 10–18 km west of Pécs, the country's third-largest town (IAEA, 2005). Uranium mining and processing had been part of Hungary since 1958 (International

Atomic Energy Agency, 2011). Since 2006, several mining licenses have covered four project areas for uranium mine discovery, namely: Mecsek, Bátaszék, Dinnyeberki, and Máriakéménd. However, the projects in Bátaszék, Dinnyeberki, and Máriakéménd were finalized without any notable success (OECD, 2014).

The Mecsekérc Ltd (also known as Mecsek uranium mine was the only producer of uranium in Hungary until 1997 when it was closed due to being uneconomical (OECD, 2005; Malovics, 2014; Hungarian Atomic Energy Authority, 2017). According to the International Atomic Energy Agency (2011), the Hungarian Government decided to shut down uranium ore mining in Mecsek based on a study of environmental effects as well as an investment plan produced on this basis. In 1998 the clean-up of the uranium mining legacy started: closing of the underground mines, remediation of the rock heaps, the sedimentation ponds, and the contaminated water and of the uranium mill. The key remediation activities at the Mecsek uranium mine were concluded in 2009, where 62 ha of land needed to be cleaned and 700,000 m^3 of contaminated soil disposed of (Wallner and Stein, 2012). After closing measures were planned on the remediated site, there were plans and implementations of technical intervention for continuous operation of the established radiological-hydrogeological-geodynamic monitoring system (International Atomic Energy Agency, 2011).

The Mecsekérc Environmental Protection Public Limited Company was founded after 2014 as a state-owned company to continue the tasks of the state mine remediation. This organization committed to the solution and realization of environmental protection, including planning and fulfillment of the remediation activity for uranium industry in Middle and Central European countries; environmental damage assessment; and planning, implementing, and licensing of environmental damage remediation activities (International Atomic Energy Agency (IAEA), 2016). Appropriate Hungarian, as well as international laws and guidelines, IAEA recommendations, and other countries' practices, have been instrumental in setting environmental protection criteria in the remediation process (Hungarian Atomic Energy Authority, 2017). To follow the environmental protection criteria, Mecsek uranium mine conducted a long-term land reclamation which included the monitoring of air, groundwater and surface waters, soil, rocks, and vegetation as endangered and contaminated compartments in the period of mine closure, remediation, and post-closure operation (Mecsekérc Zrt., 2017). Mecsekérc Zrt further stated that regarding remediated waste rock dumps and tailings ponds, it is imperative that the performance of the radon barrier is enough and that the migration of radionuclides is constantly monitored.

1.2. Research problem

Due to the presence of numerous elements and radionuclides, mine tailings cause severe pollution problems and may pose risks to humans (Skipperud *et al.*, 2013; Fernández-Ondoño *et al.*, 2017). Spoil deposits are high in these elements and can often be prone to water and/or wind erosion, spreading the contamination to surrounding areas. These issues have risen over the last few decades (Fernández-Ondoño *et al.*, 2017)). The question of environmental contamination occurs when potentially toxic elements (PTEs) and radionuclides are mobilized from the deposits of uranium-containing waste (waste rock pile, tailings ponds, etc.) into the soil and absorbed by plants or transferred to groundwater (Waggitt, 2008; Shiva Kumar and Srikantaswamy, 2014).

This study is part of the Mecsek uranium mine ongoing environmental monitoring program to monitor the endangered and contaminated compartments in the period of mine closure, remediation, and post-closure operation (Mecsekérc Zrt., 2017). Every three to five years, soil vegetation and water samples are taken from all spoil deposits and from covering soil to monitor the migration of radionuclides and PTEs. In this monitoring program, the radiometric survey is conducted to measure radon-222 (Rn-222), uranium content, and radioactive element migration. Additionally, the pseudo total concentrations of elements are measured in the soil, water, and plants. Based on the results of the monitoring program, spoil deposit No. I was deemed to require further investigation on the mobility of radionuclides and PTEs. However, it is now generally understood that the distribution, mobility, and bioavailability of PTEs and radionuclides in the environment depends not only on their total concentration but also on the form of association in the solid phase they are bound to (Filgueiras *et al.*, 2002). Sequential extraction is an essential and commonly used method for obtaining knowledge about possible mobility (such as potential bioavailability and toxicity) of PTEs and radionuclides (Rauret *et al.*, 1999; Filgueiras *et al.*, 2002; Rosado *et al.*, 2016; Walther and Gupta, 2015; Heltai *et al.*, 2018; Mittermüller *et al.*, 2016; Fernández-Ondoño *et al.*, 2017; Sheppard, 2011; Pérez-Moreno *et al.*, 2018; Bacon and Davidson, 2017). These procedures have not been previously included in the monitoring program for the migration of PTEs and radionuclides in the Mecsek uranium mine. Therefore, to investigate a long-term environmental impact, this research study focused on estimating the migration of these elements using sequential extraction procedure recommended by the Community Bureau of Reference (BCR) procedure (Rauret *et al.*, 1999; Rauret *et al.*, 2001).

1.3. Aim of the study

The main aim of this research was to study the effectiveness of the soil covering layer in retardation of the migration of PTEs and radioactive substances on the covering soil of spoil deposit No. I in Mecsek uranium mine. The goal is to use the same methodologies used in this research study to address similar problems in South Africa.

1.4. Study objectives

Based on the problem statement, the following objectives were identified:

1. To determine the total concentration levels of PTEs and radionuclides in soil, plant, and water samples collected from spoil deposit No. I and to determine if these concentrations are within the regulatory limits. Elaboration of the sampling plan based on the detected anomaly during the previous gamma activity monitoring results according to the topography of spoil deposit No. I.
2. To evaluate the mobility of PTEs and uranium in covering soil by adopting BCR sequential extraction methodology to the specific Mecsek mine conditions. To study the influence of covering soil characteristics (pH, CEC, SOM) on mobility.
3. To study the bioavailability and uptake of PTEs and radionuclides by plants in the spoil deposit No. I using the pseudo total methodology.
4. To evaluate the efficacy of the soil covering layer retarding the migration of the PTEs, radon-222, and radionuclides in the remediated spoil deposit No. I.

1.5. Rationale

During the formation of the soil covering the process of spoil deposits in the Mecsek uranium mine, there was an erosion wounding occurrence in spoil deposit No. I. Consequently, there was a question of the adequacy of the 1-meter thickness soil covering layer used to cover the spoil deposit. The lack of soil cover stability induces acute downward migration of radionuclides and PTEs (Korychenskyi *et al.*, 2018). The ongoing environmental monitoring program in the Mecsek uranium mine identified that the spoil deposit No. I required further investigation on the mobility and vegetation uptake of PTEs and radionuclides. Previous researches have revealed that the mobility of these elements depends on the forms in which they occur in the soils (Nemati *et al.*, 2011; Bielicka-Giełdoń *et al.*, 2013; Sungur *et al.*, 2014). These forms, which can be distinguished by the BCR sequential extraction procedure, include (Rauret *et al.*, 1999):

a) Exchangeable and acid-soluble fraction,
b) Bound to reducible species (e.g., Fe and Mn oxides, oxyhydroxides),
c) Oxidizable - forms bound to organic matter or sulfides, and
d) Strong oxidative acid-soluble residual contents (*aqua regia* and/or H_2O_2/HNO_3)

According to Bielicka-Giełdoń, *et al.* (2013), plants can readily access the first two types of forms, while the second two types are potentially accessible in the long term. A sequential extraction is an essential tool of knowing the conditions of these elements in soils and hence the possible mobility, bioavailability, and chemical nature of a given element that can thus offer a more accurate estimation of the actual environmental effect (Umoren, *et al.*, 2007; Yang *et al.*, 2009; Saleem *et al.*, 2018). Thus, there was a need to include the BCR extraction technique in the monitoring program to provide the information needed for effective planning and management of applications in the Mecsek uranium mine.

2. LITERATURE OVERVIEW

This chapter gives an overview of uranium remediation processes and the potential health and environmental impacts of PTEs and radionuclides because of uranium mining legacy issues. The Ecosystem transfer and the factors influencing the distribution of PTEs and radionuclides are discussed. Analytical methods for the determination of active gamma radionuclides, radon gas in soil, total concentrations of PTEs in plants and soils, and the mobility of PTEs using the BCR method were discussed in detail. Instrumental analytical methods that could be used for the analysis are listed in this chapter, and the regulatory measures to be followed are discussed.

2.1. Uranium mine remediation

The contemporary uranium mining industry started in the late 1940s at a period when little consideration had been given to protecting the environment (Waggitt, 2008). When uranium mining began in the late 19th century in the Czech Republic, it was for the purpose of extracting ores for use in Marie Curie's radium isolation studies (Atomic Heritage Foundation, 2018). Worldwide, uranium ore is mined using three primary procedures which generate some kind of waste: underground mining (27%), open-pit mining (33%), and other processes (40%, including in situ uranium leaching and uranium processing as a by-product of other mines) (Carvalho, 2011; EURATOM Supply Agency, 2018). According to Gavrilescu *et al.* (2009), U is more prevalent than gold (Au), silver (Ag), mercury (Hg), antimony (Sb), or cadmium (Cd), and more or less as common as tin (Sn), cobalt (Co), lead (Pb), molybdenum (Mo), and arsenic (As). Uranium is a chemical element that is silver-white and weakly radioactive, with an atomic number 92 (Závodská *et al.*, 2008). Uranium and its progenies occur naturally in the Earth’s crust and areas with acid igneous rocks, such as granites (Carvalho *et al.*, 2007). Natural uranium consists of three isotopes, U-238 (99.276%), U-235 (0.718%), and U-234 (0.004%), all of which are radioactive (Garnier–Laplace *et al.*, 2001; Závodská *et al.*, 2008). Uranium mining remains controversial mainly due to the legacy of environmental and health problems produced during the industry's early phase (Organisation for Economic Co-operation and Development, 2014). These legacy sites are of particular concern worldwide because of the possible presence of waste components with enhanced concentrations of radionuclides and their potential effects on environmental health (Carvalho *et al.*, 2014). Existing data show that most environmental pollution concerns are likely to originate from the past rather than from current operations in the uranium industry, and thus the concerns are considered legacy issues (Skipperud *et al.*, 2013). Soils near mines are exposed not only to PTE stress but also to radionuclide contamination that can pose a possible soil ecological hazard (Dikinya, 2015).

Mining activities consist of numerous ecological imprints in the environment, but mine tailings and AMD are of significant and global importance (Carvalho, 2017). Carvalho (2017) further states that since the recognition of the mining impacts, environmental and sanitary legislation has been substantially introduced, and some efforts have begun to tackle the industrial heritage through clean-up, remediation, and rehabilitation programs. In addition, Selvakumar *et al.* (2018) state that efforts have been made to remediate the uranium polluted environment through physical, chemical, and biological processes. Any soil remediation strategy has the ultimate goal of providing a final solution that protects human health and the environment (Wuana and Okieimen, 2011). Furthermore, Dikinya (2015) states that polluted soil must be carefully maintained for efficient usage to protect human health and the environment. Generally, the prominent roles of the remediation program are, in general, to delay the release, oxidation, and erosion frequency of contaminants (IAEA, 2009).

Environmental remediation aims to reduce pollution, including decontamination of radioactive soil or water, which may result from uranium mining activities (Gupta and Voronina, 2019). The remediation of these uranium mining legacy sites has been thoroughly assessed by the International Atomic Energy Agency (IAEA) and other organizations (OECD, 2014; Collier, 2016). Most uranium mining sites were largely abandoned in the 1960s due to a decrease in uranium mining activity without any effort at remediation (Waggitt, 2008). In recent years, hundreds of uranium mining/milling sites have been closed down (IAEA, 2004). Several problems occur as a result of uranium mining activities which range from unnatural-looking mining deposits to the migration of PTEs and radionuclides into the environment, thereby contaminating soil and water (Hiller *et al.*, 2016). Similarly, Dikinya (2015) state that PTEs and radionuclides are the natural components of the Earth's crust that generally occurs in the subsurface environment and anthropogenic activities cause problems by enhancing their pollution in the soil environment. These problems, which are generic to the mining industry, are tackled during the rehabilitation of uranium mining sites (Collier, 2016). However, the majority of the countries most affected by the problem of uranium mining do not have sufficient funds or services and technology in their regulatory networks for preparing, implementing, and maintaining rehabilitation programs (Waggitt, 2008).

Soils can be rehabilitated using different techniques of either physical, chemical, and/or biological remediation (Smičiklas and Šljivić-Ivanović, 2016). The choice of methods to be used, however, depends on the mobility of contaminants, the distribution pattern in different fractions of the soil, and future land use (Lourenço *et al.*, 2019*)*. At times the conventional chemical and some physical remediation methods are often expensive, generating toxic and non-eco-friendly products, and sometimes, are ineffective when the contamination is elevated (Bakatula *et al.*, 2012). Any effort to remediate contaminated soils will require information on the source of contamination, introductory chemistry, and the environmental and related health consequences (risks) of such contamination, while soil characterization may provide information on the speciation and bioavailability (Wuana and Okieimen, 2011). Therefore, radionuclide and PTE concentrations and distributions worth a comprehensive study (Yi *et al.*, 2020).

It has been established that there are different known techniques of remediation for soils contaminated with radioactive elements, as shown in (Figure 1.), but very few have been used in field conditions (Gavrilescu *et al.*, 2009). According to Gupta and Walther (2014), remediation of the areas contaminated with radionuclides and PTEs should get proper attention. However, the traditional remediation techniques used in polluted areas have demonstrated many drawbacks, including high costs, soil properties modification, and disruption in soil indigenous microflora (Gupta and Walther, 2014). It is important to note that the Mecsek uranium mine employed the soil cover remediation process for the study area in question. Furthermore, the Mecsek uranium mine has been monitoring the radon barrier function and the migration of radionuclides to check the adequacy of the soil cover layers in remediated waste rock dumps and tailings ponds (Mecsekérc Zrt., 2017).

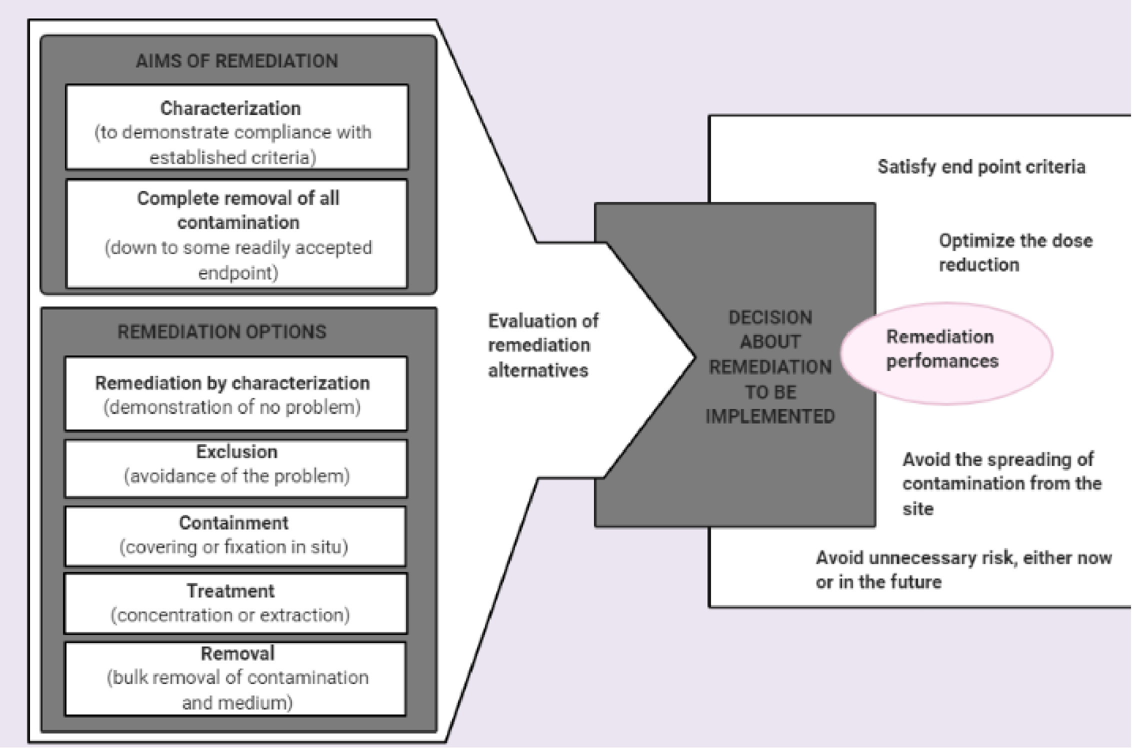


Figure 1. Remediation alternative evaluation in relation to aims and performance (Gavrilescu *et al.*, 2009)

Violante *et al.* (2010) noted that most regulations or guidelines used for protecting soil from elemental pollution, such as setting maximum permissible PTE concentrations used for identifying contaminated sites and for setting goals for remediation clean-up, are still based on evaluating the total concentration of metal present in the soil. When mining soils are remediated, soil and rock that is covering a deposit of ore, such as uranium, usually contain at least trace amounts of the ore plus radioactive decay products (Gavrilescu *et al.*, 2009). Therefore, during remediation processes, it is recommended to apply a multi-layer soil cover on the waste dump in order to minimize the effects on the environment that could result from the toxic effluent releases and the exhalation of radon gas (Rn-222) (Biehler *et al.*, 2002). According to OECD/NEA (2014), the remediation concept consists of stabilizing and protecting the environment by using a covering layer of 1 m thickness to cover the uranium mine waste rock pile/deposit, as shown in (Figure 2.). In some cases, a 2 m thick layer of different materials is used to prevent radon exhalation from the waste pile and infiltration of precipitation into it (Štrok and Smodiš, 2013). The OECD/NEA (2014) further states that the installation of this type of cover brings about a reduction in the Rn-222 exhalation rate and, in turn, a significant reduction in Rn-222 concentration by a factor greater than ten. According to the IAEA (2010), depending on site unique conditions, the cover systems may have any or all of the following characteristics:

- Limiting radon release,
- Radiation shielding,
- Limiting moisture infiltration to a predetermined amount,
- Limiting oxygen diffusion to a predetermined amount,
- Long term erosion resistance,
- Containment of the covered material within the structure,
- Support for vegetation communities, and
- Prevention of capillary rise of oxidation products to the surface.

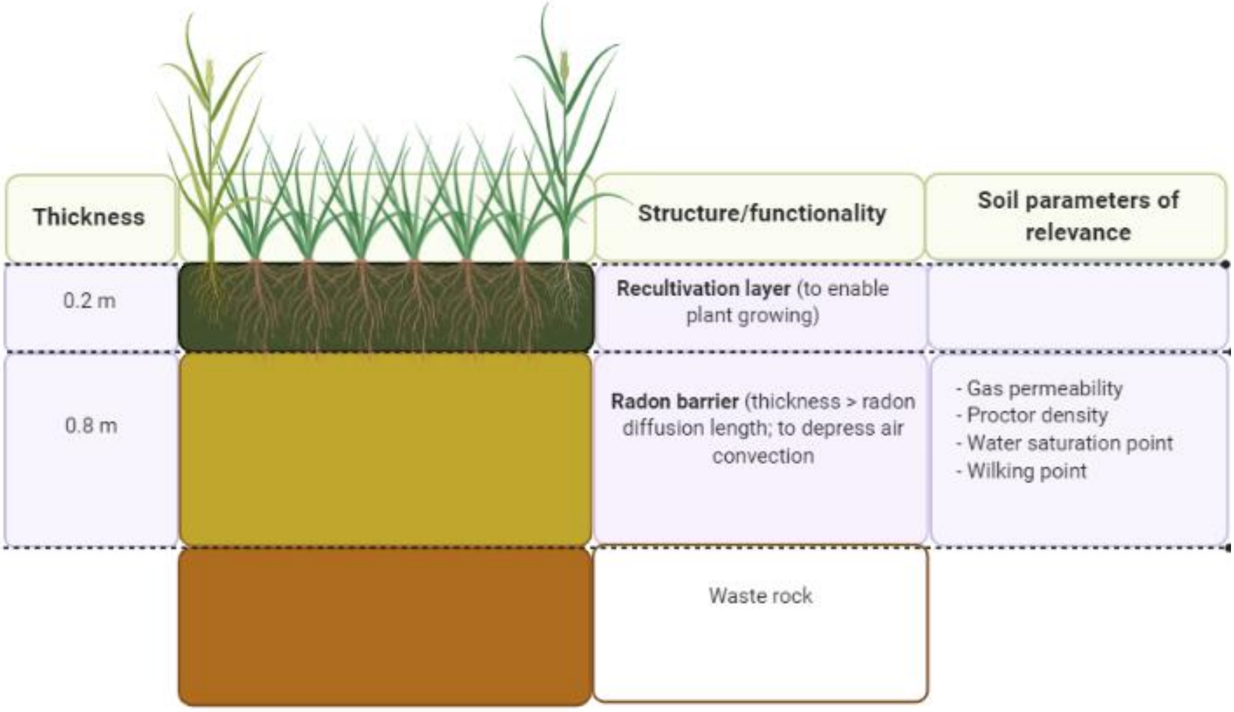


Figure 2. The soil cover design for uranium waste deposit (adapted from OECD/NEA, 2014; and created with BioRender.com)

Richon and Beneito (2002) conducted a study on the evaluation of the effect of a cover layer on Rn-222 exhalation from uranium mill tailings. In this study, a 1 m layer of cover material was placed on top of the first layer to study its effectiveness on Rn-222 flux emanating from the uranium mine tailing (Richon and Beneito, 2002). Similarly, Kuterbekov (2019) conducted a study on rehabilitation events and an estimation of their effectiveness at control sites in the southern part of the tailing ponds of Koshkar-Ata (Mangystau Region, Kazakhstan). In this study, a sand-clay soil-sealing coat covering technique was chosen for rehabilitation to prevent dusting and serve as an additional barrier to radon emanation. The author stated that the advantage of using the sand-clay soil covering technique is stimulating the development of vegetation cover as additional protection against wind erosion. The author concluded that the results of the assessment of the effectiveness of this rehabilitation technique demonstrated considerable improvement of the radiation characteristics of the sites and the gamma concentration was decreased down to the background values of the Aktau city vicinity.

It is critical to evaluate whether the remediation processes used are effective for reducing the impact on the environment by measuring the concentrations of dissolved solids (especially U and Ra-226), the contents of environmental isotopes in precipitation, surface runoff, seepage water, and groundwater (Biehler *et al.*, 2002). Environmental monitoring plays a huge role in determining the effectiveness of soil cover. According to IAEA (2004), the closure and rehabilitation activities involve exercising surveillance over a number of parameters, including monitoring numerous radiological and other parameters.

2.2. Potential health impacts

The mining sector is one of the most polluting business activities, which can have direct consequences on the health of people and animals, resulting in exposure to environmentally harmful agents, or indirectly resulting from the latter's activity on the water, air, soil, and vegetation (George-Laurentiu *et al.*, 2016; Ogundiran and Osibanjo, 2009). Due to these potential risks to human and environmental health, soil contamination is a global ecological problem (Bielicka-Giełdoń *et al.*, 2013). Uranium mining, processing, reclamation, and waste handling may expose living organisms, including humans, to chemical and radiological materials through inhalation, ingestion, absorption through the skin, and gamma radiation (National Research Council of the National Academies, 2012; Public Health Department of the Cree Board of Health and Social Services of James Bay, 2014). Uranium mining legacy results in contamination that does not only come from U and its daughters but also from associated trace metals that may pose a risk to man and the environment (Skipperud *et al.*, 2013). Some of the major pathways for PTEs and radionuclide transfer to humans are through the food chain (soil – plant – human or soil – plant – animal – human), contact with contaminated soil, and drinking of contaminated groundwater (Lema *et al.*, 2014; Wuana and Okieimen, 2011). Likewise, Alharbi and El-Taher (2013) noted that these radionuclides are obtained by plants through their roots or leaves, and animals obtain them through the consumption of plants and are ultimately transferred to man by consuming animal meat, milk, and/or directly from plants by consuming them. Moreover, elements may accumulate in animal and human body tissue to toxic levels once they get to be part of this cycle (Igwe *et al.*, 2005).

2.2.1. Radionuclides

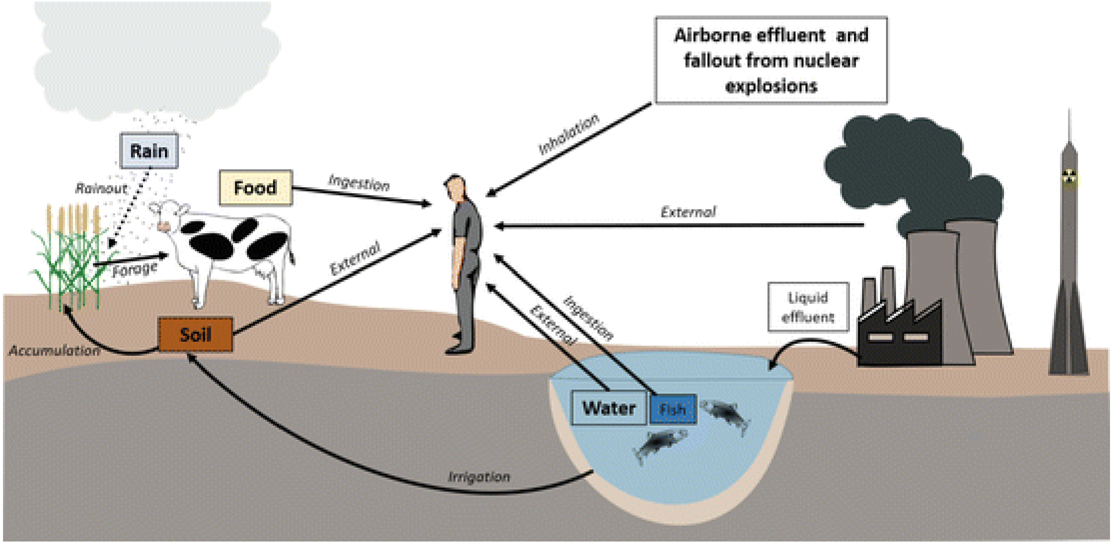


Figure 3. Major exposure pathways of radionuclides (Nieder et al., 2018)

Soil is one of the major sources of radiation exposure to a population via the transfer of radionuclides into the environment (Durusoy and Yildirim, 2017). Recently, owing to the radionuclide toxicity, health hazards, and potential environmental risks, general interest in radioactivity and radionuclide distribution in the environment has considerably augmented (Ahmed *et al.*, 2019). Knowing the nature of radionuclides in the environment, their possible stability, and bioavailability linked to long-term survival, radiological hazards, and human health effects are essential (IAEA, 2016). According to UNSCEAR (2000), K-40, Th-232, and U-238 are principal primordial radionuclides with half-lives comparable to the age of the Earth, and only Th-232 and U-238 head the series of radionuclides that produce significant human exposures. According to Nieder *et al.* (2018), radionuclide toxicity has two main pathways into the human body, as shown in (Figure 3.) (external contamination by exposure to anthropogenic and natural sources, and internal contamination by ingestion or inhalation). United Nations Scientific Committee on the Effects of Atomic Radiation (2000) reported that most leukemia and cancers of several organs, such as lungs, breasts, and thyroid gland, have been linked to radiation exposure, although not certain other organs such as the prostate gland. It has been established that ionizing radiation is strong enough to alter molecular composition like DNA within the body's cells (National Research Council of the National Academies, 2012).

A long-standing radiation protection debate continues over the risk of cancer from low levels of ionizing radiation (Gulan and Milenkovic, 2018). Radiation injuries happened shortly after the humans’ discovery of radiation when the first case emerged in the United States as skin burn just after few months from November 1895 when Roentgen W.C. discovered X-rays (Kang, 2016). At least since the late 1950s, it has been discovered that exposure to radiation is a risk factor for enhanced leukemia (Fairlie, 2009). Other effects of radionuclides exposure or radiation are acute health effects that begin with nausea, vomiting, and headaches (Prakash *et al.*, 2013). It has been established that the radioactive contamination of soil, water, and air can be transferred to humans via plants or through inhalation of radon gas (e.g., Rn-222 and Rn-220) (Ademola *et al*., 2015). The dosage of radon on the general public and miners is a significant health issue due to elevated levels of radon that can arise unabated (Shahrokhi *et al.*, 2017). Radon gas is radioactive, colorless, odorless, and tasteless; the only way to establish the presence of a radon health risk is through analytical measurement (Tanaka *et al.*, 2017). Furthermore, Deng *et al.* (2020) state that radon is a natural carcinogen resulting from the three primordial radionuclides of the uranium series (U-238 and U-235) and thorium series (Th-232). Uranium, like other non-essential PTEs, is chemo-toxic for humans and induces permanent damage to kidneys (nephrotoxic) if ingested above a particular concentration limit (Winde, 2010).

2.2.2. Potentially Toxic elements

The mining areas may put the quality of life of the surrounding communities at a higher risk of exposure to PTEs (Olowoyo *et al.*, 2013). Potentially toxic elements are separated into three main classes; the macronutrients, the micronutrients, and the toxic elements. The macronutrients are needed in large quantities by plants; the micronutrients are needed in small quantities; however, in large amounts, they can be toxic; and lastly, the toxic elements can indicate toxicity even in small quantities. In several studies, it has been established that Ca, K, Mg, and P are macronutrients and Co, Mn, Cu, Fe, Na, and Zn are micronutrients (FAO, 1982; Mahler, 2004; Lohry, 2007; Heidak *et al.*, 2014). Lohry (2007) further explains that the macronutrients are found in two groups, as the main macronutrients (N, P, and K) and as the secondary nutrients (Mg, S, and Ca), and all are needed by plants in large quantities. Furthermore, Lohry (2007) indicated that Na is probably essential for only a few plants indigenous to saline soils. At the same time, Jones and Jacobsen (2001) describe Na as beneficial to the plants. Moreover, Kronzucker *et al.* (2013) emphasized that even though Na has accomplished a reputation for its toxic qualities, it remains beneficial to many plant species at lower levels of supply, and in some species, it is indeed essential.

The toxicity of PTEs in humans has been associated with numerous physiological disruptions following exposure to elevated levels (Ogundiran and Osibanjo, 2009). Furthermore, PTEs can pose a threat to human health, plant growth, and animal life by affecting major biochemical processes (Ogundele *et al.*, 2015). Jolly *et al.* (2013) state the key mechanism of human exposure to metal contamination in the soil to plants transfer of PTEs. According to Pandey *et al.* (2016), pollution from PTEs is hidden, persistent, and irreparable; it not only takes away the eminence of the environment, water sources, and food crops but also risks human health prospects. Similarly, contaminants can be passed to other parts of the ecosystem in polluted soils, which potentially endanger human health (Rastegari Mehr *et al.*, 2017). The protection and sustainability of soil resources are therefore of paramount importance as the problem of the consumption of PTE-contaminated crops (Forghani *et al.*, 2014). The PTEs in these soils can be transmitted to humans by processes of ingestion, inhalation, or dermal absorption (Rastegari Mehr *et al.*, 2017). The toxicity of PTEs can lead to lower energy levels and damage to the brain, lung, kidney, liver, blood supply, and other essential organ functions (Jaishankar *et al.*, 2014). Similarly, Jolly *et al.* (2013) state that elevated metal content above the Maximum Permissible Level (MPL) results in various physiological, cardiovascular, renal, neurological, bone, and many other health disorders. In addition, recurring long-term exposure to some elements may also cause cancer (Jaishankar *et al.*, 2014). Cadmium has been considered to be the second most hazardous trace element after mercury as it is toxic in all its forms (metal, steam, salts, and organic compounds) (Khelifi *et al.*, 2019).

2.3. Potential environmental impacts

Many of the causes of stress on ecological systems are not unique to uranium mining but can be correlated with any mining operation or remediation (National Research Council of the National Academies, 2012). Mining activities and metal production are some of the critical processes by which natural radionuclides in the terrestrial ecosystem are accumulated (Isinkaye *et al.*, 2018). Biological elements, land and water, physical elements, fauna and flora, atmosphere, and social and cultural elements are interacting systems that form part of the environment (OECD Nuclear Energy Agency and International Atomic Energy Agency, 1999).

Reported environmental effects of uranium mining and processing include high levels of potentially toxic elements and radionuclides in water; exposure of marine and terrestrial biota species to high levels of radionuclides; localized reduction of groundwater levels; and other hazardous substances (National Research Council of the National Academies, 2012; Skipperud *et al.*, 2013; Dikinya, 2015). Nevertheless, there have not been many studies conducted on the concentrations and distributions of radionuclides and PTEs in surface water and sediments, particularly in the rivers downstream of uranium mining (Yi *et al.*, 2020).

Environmental metal pollution and technologically enhanced naturally occurring radioactive material (TENORM), as well as radiation exposures to humans, are considered dangerous measures that result from mining and tailing industries (Popic *et al.*, 2011). Similarly, Matshusa and Makgae (2017); Voitsekhovitch *et al.* (2006) suggest that The potential impact of uranium mining on the environment and human health are AMD and the leaching and seepage of the radionuclides from tailings and eventual transport it into the water; and the pollution of groundwater and surface water with highly radioactive elements and chemicals used in mining. Unless properly handled, these elements can become significant off-site pollutants in waterways and reach the biological food chain (Heard, 2017). Acid mine drainage has the potential to be one of the most severe environmental problems caused by uranium mining if it is not appropriately managed and mitigated (National Research Council of the National Academies, 2012). While much is already understood about mining's environmental impacts both on-site and off-site, there are also poorly established long-term threats and limited data available to support the long-term efficacy of uranium mine deposits management facilities (National Research Council of the National Academies, 2012).

Huge quantities of contamination caused by PTEs and radionuclides have been identified in mining environments in recent years (Yi *et al.*, 2020). Both PTEs and radionuclides cannot deteriorate naturally or synthetically, and thus, they become a risk factor to public health when exposed and/or deposited in soil and water (Gupta and Walther, 2014). For plants, uranium deposition begins with the root system and gets accumulated in different sections (Selvakumar *et al.*, 2018). Elements such as chromium (Cr), cobalt (Co), nickel (Ni), copper (Cu), arsenic (As), cadmium (Cd), mercury (Hg), etc.; radioactive elements uranium-235 (U-235), U-238, Th-232, and associated daughter products were listed as potential concern pollutants, with U and Cd potentially having the highest environmental impacts (Hook, 2000; Wade and Coetzee, 2008). Impacts may be defined as those effects that either temporarily or permanently alter the existing system and the defining and measuring of the impacts depends heavily on location, country, and social and economic factors (OECD, 1999). Data collected from previous research, combined with analytical data obtained during the scoping process of the current analysis, resulted in selecting uranium as the most significant contaminant in terms of surface and groundwater pollution (Waggitt, 2008). The content of uranium in groundwater is usually poor; however, U is released from anthropogenic sources of groundwater mainly as a result of mining, milling, and processing practices and the recycling of solid waste (Canadian Council of Ministers of the Environment, 2007). Any possible sources of radiation and radioactive material associated with uranium mining have the potential to degrade the environment and impact the local population unless appropriate safety measures are taken (Tripathi *et al.*, 2008). The environmental degradation (mobility and bioavailability) and toxicity strongly depend on the specific chemical form and binding methods of PTEs and radionuclides (Perez-Moreno *et al.*, 2018). Since the release of radionuclides from contaminated sites frequently occur along with PTEs and persistent organic contaminants, thus generating multiple stressors acting in nature, these stressors need to be investigated together in order to obtain a more comprehensive assessment of possible environmental pollution risks (Popic *et al.*, 2011).

2.3.1. Mobility of radionuclides

Under normal environmental conditions, only a moderate proportion of the total radionuclide content will be mobile and/or bioavailable, therefore establishing the availability of natural radionuclides in environmental conditions is necessary to assess their toxicity (Pérez-Moreno *et al.*, 2018). Radionuclides occur naturally in the environment due to processes such as volcanic activity, erosion, and weathering; these radionuclides include isotopes of uranium, thorium, radium, radon, lead, and polonium (Po) (Yan *et al.*, 2021). Although radionuclides occur naturally in soils and rocks as a result of radioactive decay, most environmental releases of radionuclides are due to industrial activities such as mining and milling (Canadian Council of Ministers of the Environment, 2007; Dudu *et al.*, 2018). Mining and metal production methods have been established as a significant route to the aggregation of radionuclides in terrestrial environments (Isinkaye *et al.*, 2018). The majority of radionuclides released into the environment can accumulate in both the upper soil layer and in aquatic systems sediments, resulting in threats to ecosystems, agro-system, and health (Gavrilescu *et al.*, 2009). The radionuclides' migration is dependent on several factors, such as radionuclide speciation, soil composition (e.g., organic matter, soil water pH), microbial activity, presence of vegetation, and animal grazing/human consumption (Salbu *et al.*, 2004).

Terrestrial radionuclide potassium-40 (K-40) and radionuclides of the U-238 and Th-232 series (also known as parent radionuclides) are the main contributors to natural radiation (Stevanović *et al.*, 2018). Radioactivity results from the spontaneous disintegration of a parent radionuclide and from the process of creating a daughter nuclide by releasing gamma, beta, and/or alpha radiation (Saint-Fort, 2016). Uranium (U^{6+}) and radon are the radionuclides that present a reasonable degree of mobility in the soil out of all the natural radioactive elements (Júnior *et al.*, 2006). According to Payne and Edis (2012), solubility is a significant factor in the mobility of radionuclides. The mobility of a specific mineral phase places an upper limit on solution concentration (Payne and Edis, 2012). Similarly, Walther and Gupta (2015) state that the chemical form and speciation of radionuclides largely determine their behavior and ultimate radiological impact in soils, which significantly affects their mobility, the length of stay in the soil rooting zone, and the availability of biota absorption. Therefore, the information on source terms, including radionuclide speciation, mobility and, biological uptake, is needed to assess the long-term environmental impact of radioactive contamination of ecosystems (Salbu *et al.*, 2004; Sungur *et al.*, 2014; Pérez-Moreno *et al.*, 2018). According to Saint-Fort (2016), when addressing the mobilization and remobilization of radionuclides, it is crucial to consider weathering releases and concurrent speciation to water, sediment, soil, and air with potential for further distribution and dispersion via dust and biota uptake.

The environmental stability and bioavailability are regulated by the physical and chemical state of the radionuclides (Konoplev *et al.*, 2020). The release scenarios such as temperature, pressure, air presence may affect the speciation of deposited radionuclides in an ecosystem (Skipperud and Salbu, 2015). Environmental impact and risk assessments associated with radioactive contamination are typically extracted from total radionuclide concentrations, assuming that radionuclides released into the environment are homogeneously distributed (Skipperud and Salbu, 2015).

2.3.2. Mobility of potentially toxic elements

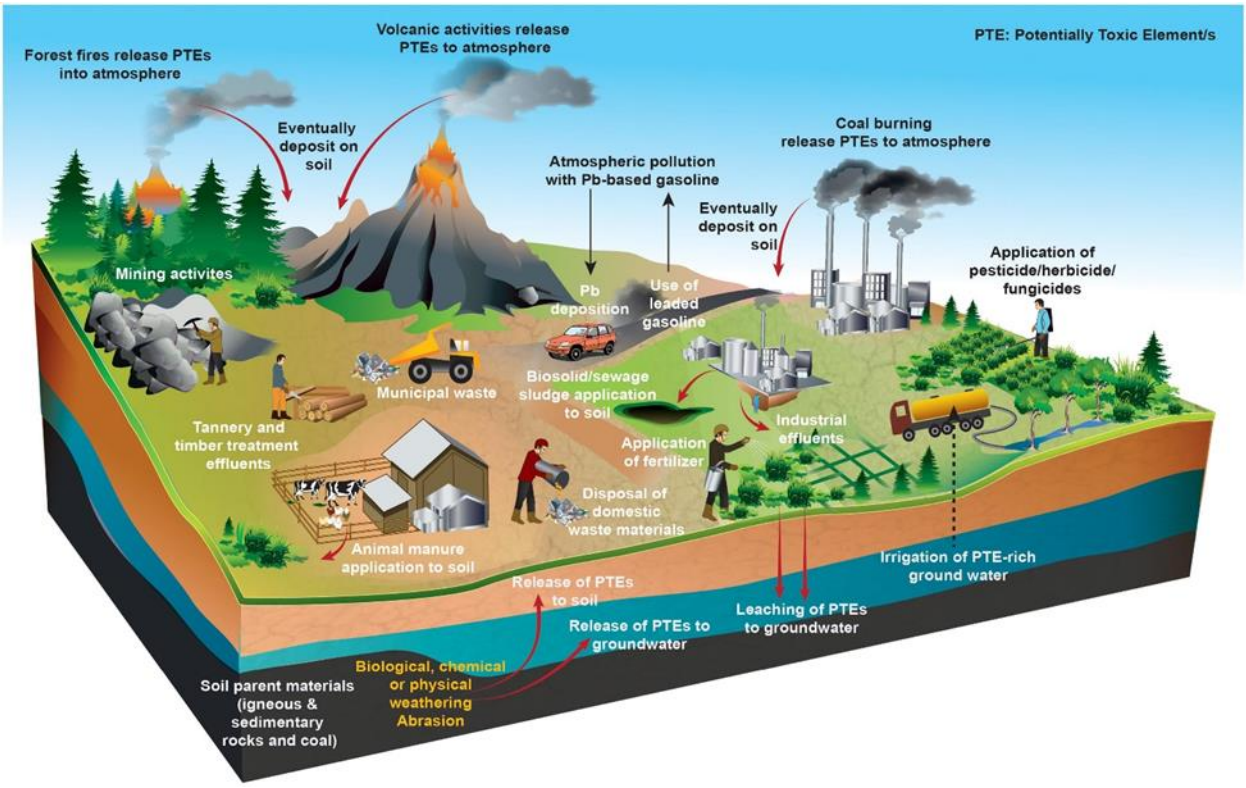


Figure 4. Potentially toxic element sources in soil ecosystems (Palansooriya *et al.*, 2020)

Potentially toxic elements are a group of elements that have been associated with contamination, toxicity, and pollution as a result of natural or anthropogenic sources (such as mining) (Stevanović *et al.*, 2018). While natural PTEs exist in soils, anthropogenic activities help increase the rates of redistribution of these elements through different environmental segments (de Souza Braz *et al.*, 2013). These elements have also been named toxic metals, trace elements, trace metals, or heavy metals, which is the most popular term (Hooda, 2010). Hooda (2010) further state that whilst none of the above-mentioned terminologies are utterly accurate from a chemical point of view, PTEs terminology is more inclusive and appropriate than toxic or heavy metals. High soil degradation in the world is due to the deposition of PTEs in soils by anthropological activities, such as fossil fuel burning, mining, smelting ores, and other activities as shown in (Figure 4.) (Cui *et al.*, 2016). During the mining activities, the pollution of the ecosystem by PTEs poses serious concern due to their non-degradability and persistence in the ecosystem; their physical, chemical, and biological processes may combine to concentrate metals rather than dilute them under certain circumstances; and finally, they have a significant effect in polluting the environment (He *et al.*, 2015; Igwe *et al.*, 2019). In a soil environment, PTEs can exist in both solid and solution phases (Ogundiran and Osibanjo, 2009). Ogundiran and Osibanjo (2009) further state that when these elements are present in the aqueous phase, they are mobile and toxic, while in the solid phase, they are immobile, inert, and harmless. Previous research has demonstrated that if the soil is not very acidic, soil components typically bind these elements so that they do not leach freely from the soil, and they are not readily available to plants (Igwe *et al.*, 2019).

In order to identify, monitor, and assess the potential source of contamination in an area, it is essential to determine the content of PTE and its spatial distributions in soils (Stevanović *et al.*, 2018). However, the substantial migration of these elements down the profile can be identified only in mildly to highly acidic soils (Igwe *et al.*, 2019). Ogundiran and Osibanjo (2009) studied the mobility and speciation of heavy metals in soils impacted by hazardous waste, where the results indicated that the highest levels of PTEs are present in the uppermost layer with significant migration down the soil depth, thereby posing a threat to groundwater quality. Potentially toxic elements are currently of great environmental concern because if their accumulation in soils is excessive, they may be harmful to humans and animals (Dikinya, 2015). However, most elements are needed in small quantities in living organisms (Keshavarzi *et al.*, 2015). Since the toxicity level of some PTEs can be just above the background concentrations that are being present naturally in the environment, therefore, a thorough knowledge of PTEs is essential to enable proper defensive measures to be taken against their excessive contact (Jaishankar *et al.*, 2014). The importance of PTEs in soil may be presented in three ways, e.g., macronutrients, micronutrients, and toxic elements (Khumalo *et al.*, 2020). Plants require essential macronutrients (such as nitrogen (N), phosphorus (P), potassium (K), (calcium) Ca, sulfur (S), and magnesium (Mg)) and micronutrients (such as iron (Fe), zinc (Zn), manganese (Mn), nickel (Ni), and copper (Cu)) from soil to grow and complete the cycle of life (Kabata-pendias and Pendias, 2001). Micronutrients are essential for plants in low concentrations, but in high concentrations, they can be very toxic (Stanojković-Sebić *et al.*, 2017); on the other hand, macronutrients are required in abundance for the plants' growth (Wuana and Okieimen, 2011). Whereas Hg, Pb, Cd, and As are nonessential nutrients that are recognized as harmful and have caused significant health problems (Shiva Kumar and Srikantaswamy, 2014). Al-hwaiti and Ranville (2010) are of the opinion that Cd is of the most significant concern of all the toxic PTEs due to its toxicity, it's capacity to accumulate in soils, and its bioaccumulation in plants and animals.

Increased levels of PTEs in soils can also lead to increased uptake of plants with subsequent human health effects, and these absorptions typically depend on the PTE concentration in the soils, the chemical and final types of the elements (Sungur *et al.*, 2014). Some toxic elements (e.g., Mo) can contribute to trace metal deficiency, such as Zn, Cu, etc. (Dikinya, 2015). Furthermore, soil properties, as well as an increase in the pollution loads of nitrogen and phosphorus, can be altered by biochemical activity associated with PTEs (Dikinya, 2015).

2.4. Ecosystem transfer

It has been established that research studies conducted on soil contamination near mines often do not take into account downstream impacts on plants (Soltani *et al.*, 2017). It is crucial to study the transfer process and plant mineral uptake of natural radionuclides and PTEs through the natural environment in terms of their presence and persistence complexity (Lema *et al.*, 2014). Potentially toxic elements in soils are, on the one hand, the primary source of micronutrients since they contribute to the plant health and nutritional value of food and, on the other, they can accumulate in soils as toxins due to anthropogenic inputs (Kabata-Pendias, 2004; Schneider *et al.*, 2019). Kabata-Pendias (2004) further state that some exceptions are in cases of flooding by contaminated waters or from heavy atmospheric deposition of pollutants. According to Greger (2004), radionuclides' availability can be high in a specific soil or plant. Therefore it is necessary to know the plant type, soil type, and which radionuclides are of interest before making any environmental recirculation calculations of the radionuclide in question (Greger, 2004).

Different factors such as the concentration of each element in soil, soil pH, climate, radionuclide speciation in soil solution, soil organic content, and soil type have been known to impact the radionuclide and PTE accumulation in plants (Adesiji and Ademola, 2019). The transfer of trace elements between soil phases can be seen as the fundamental processes regulating their behavior and bioavailability (Kabata-Pendias, 2004). According to Popic (2014), transfers of the radionuclides and trace elements will change over time due to various parameters and processes such as the ecosystem composition, the essential chemical composition of the elements, soil physical and chemical features, the temperature and hydrological conditions, element migration in the soil, changes in intake/uptake levels, biological half-lives. The most critical problem in agricultural and environmental studies has been the bioavailability of trace elements and radionuclides (Kabata-Pendias, 2004; Strok, 2012). Soil-to-water or sediment-to-water interactions are often described by distribution (also known as a partition) coefficients (K_d), whereas soil-to-plant transfer is usually described by the transfer factors (TFs) (Skipperud *et al.*, 2013).

2.4.1. Mobility factor

Since PTEs are associated with various soil components in different ways, these associations determine their mobility and availability (Kabala and Singh, 2001). Similarly, the estimation of the possibility for bioconcentration and further bioaccumulation of radionuclides in the food chain can be determined by assessing the plants' uptake of these elements (Popic *et al.*, 2011). Moreover, the mobility of these PTEs elements in soil may be evaluated based on the absolute and relative contents of fractions weakly bound to soil components during the sequential extraction procedure (Kabala and Singh, 2001). Therefore, some metal forms are strongly bound to soil components than those extracted in Fraction 1 (F1), Fraction 2 (F2), and Fraction 3 (F3) (Topcuoğlu, 2016). Hence, the relative index of metal mobility may be calculated as a mobility factor (MF) based on the following Equation (1) (Topcuoğlu, 2016):

$$MF = \frac{F1+F2+F3}{F1+F2+F3+F4} \qquad (1)$$

An MF is used to differentiate the natural concentration of PTEs from anthropogenic pollution at various soil depths (Shaheen and Iqbal, 2018). A high percentage of MF has been attributed to the potential of metal to become environmentally mobile and bioavailable (Ogundiran and Osibanjo, 2009).

2.4.2. Distribution coefficient

The speciation of radionuclides and the chemical type have a significant effect on their movement through environmental media and biota uptake (Calmon *et al.*, 2009). According to the IAEA

(2011), the extent of radionuclide sorption in the solid phase is often quantified by the solid-liquid distribution equilibrium (K_d), which can be used when determining the overall mobility and possible residence times of radionuclides in soils. Similarly, K_d plays a crucial role in identifying PTE guidelines in soils and in evaluating environmental risks, and its importance depends on the soil influence and climate patterns of the sites where the soils occur (de Souza Braz *et al.*, 2013). Furthermore, the United States Environmental Protection Agency (1999) asserts that the K_d parameter is exceptionally significant in estimating the potential adsorption ability of dissolved contaminants in contact with the soil. The values of Soil K_d can differ significantly with soil type, and where the soil characteristics are unknown, a generic soil K_d value can be adopted (Calmon *et al.*, 2009). Since the quality and quantity are among the fundamental soil properties that affect sorption, an excellent approach to determining K_d values for a large number of radionuclides and PTEs is the classification of K_d values by soil groups based on soil texture and organic matter content (IAEA, 2011; Sohlenius *et al.*, 2013). Sohlenius *et al*. (2013) further state that a high K_d value means that most of the element is adsorbed to solids and that its mobility due to water flow is poor.

2.4.3. Transfer factor

The radionuclides and PTEs uptake from the soil by plants are generally expressed as soil-to-plant transfer factor (TF) or transfer coefficient (TC) or concentration ratio (CR) (Laţo *et al.*, 2012; Adesiji and Ademola, 2019). Transfer factors or concentration ratios are classified as the ratio of radionuclide or element concentrations in an organism to that in soil (Popic, 2014). Transfer factors (TFs) are widely used to determine the bioavailability of radionuclides and metals and ecological transfer from soil to biota, as shown in (Figure 5.) (Calmon *et al.*, 2009; Popic, 2014). According to IAEA (2010), the transfer of radionuclides is usually in larger concentrations to leaves and stems and in much lower levels to the generative parts of plants.

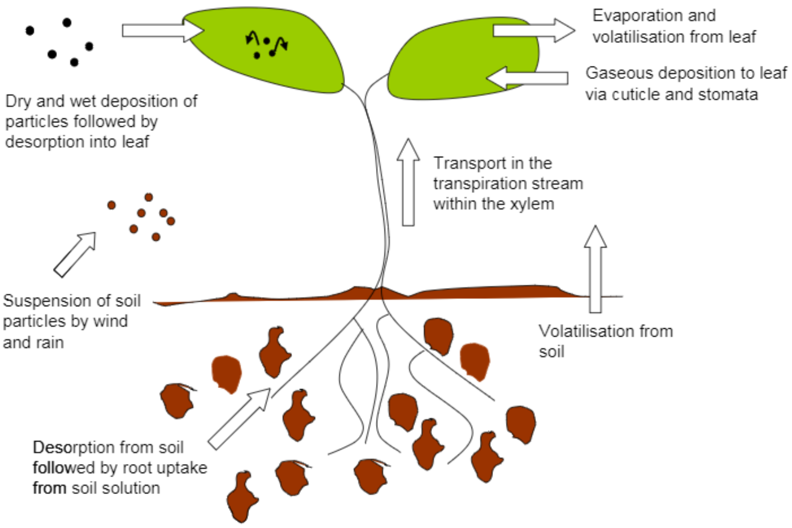


Figure 5. Principle uptake pathways for the uptake of soil contaminants by plants (Collins et al., 2006; McLaughlin, 2018)

The soil-to-plant TF is considered to be one of the most significant parameters required for nuclear industries in environmental safety assessment (El-taher, 2015). Chakraborty *et al.* (2013) noted that the soil-to-plant TF is used for environmental transfer models, which play an essential role in predicting the radionuclide concentration for estimating dose impact to a human being. Furthermore, (Skipperud *et al.*, 2013) concur that TF (kg/kg) are used for modeling purposes and are based on many factors (e.g., soil characteristics, microbial processes, plant and animal species, dietary behaviors, trophic levels) and, in particular, radionuclide and metal speciation. Previous studies indicate that by changing various soil properties and environmental conditions, there can be a transformation of radionuclides from a potential mobile form to an immobile form or vice versa, with a direct impact on plant uptake (Iurian *et al.*, 2015). The TF for the uptake of any radionuclide or PTE from soil to plant is defined as the ratio of the activity concentration in a plant part (in dry weight) to the activity concentration in the specified soil layer (in dry weight) (IAEA, 2011). According to Gerzabek *et al.* (1998); Smičiklas and Šljivić-Ivanović (2016), soil-to-plant TFs have been commonly used in radioecology to measure the availability of radionuclides in soil for plant uptake using Equation (2). Laţo *et al. (2012)* concur that the content of PTEs in soils can be established in a similar way.

$$TF = \frac{Plant\ activity\ concentration\ (\frac{Bq}{kg})}{Total\ soil\ activity\ concentration\ (\frac{Bq}{kg})}\ or\ TF = \frac{Metal\ content\ in\ plant\ (\frac{mg}{kg})}{Metal\ content\ in\ soil\ (\frac{mg}{kg})} \qquad (2)$$

Given that soil-to-plant transfer varies significantly between different plant species and seasons, this approach also provides rough estimates of the potential bioavailability of elements (Smičiklas and Šljivić-Ivanović, 2016). Similarly, Jolly *et al.* (2013) state that the TF values may vary significantly between plant species and locations. Higher TF values (≥1) indicate higher absorption of elements from the soil by the plant and higher suitability of the plant phyto-extraction and phytoremediation, and lower values indicate the inadequate response of plants towards element absorption, and the plant can be used for human/animal consumption (Agic *et al.*, 2015; Mirecki *et al.*, 2015). Furthermore, high transfer or mobility factors demonstrate that anthropogenic PTEs can disperse and be bioavailable easily within the environment, if not limited to the setting (Ogundiran and Osibanjo, 2009). Smičiklas and Šljivić-Ivanović (2016) state that TFs, despite limitations, are currently accepted as the most practical way of describing plant uptake. The amount of radionuclides in the soil solution that is available for root uptake is affected by the changes in soil physicochemical properties and biological processes (Hegazy *et al.*, 2013). Kabata-Pendias (2004) noted that there had been an increase in a number of researches associated with both the understanding of the processes involved with plant absorption of elements (nutrients and non-nutrients) and to determine the most accurate techniques for the prediction of a given element's availability to plants. The potential toxicity of PTEs is limited by three protective mechanisms of soil-plant transfer systems which may reduce the problems they might cause in animals and human beings (Laţo *et al.*, 2012). Laţo *et al.* (2012) further state that these protective mechanisms include: elements that are insoluble in the soil and that do not accumulate in plants; elements that are absorbed in the root but are insoluble and come in small quantities; elements that cause phytotoxicity when added in abundance, so plants are not ingested by humans or animals. Therefore, to measure radiation doses for humans, it is vital to evaluate and estimate the concentration of different elements present in the soil and their transfer factor to different plant samples (El-taher, 2015). Furthermore, all the significant variables that govern the behavior of these elements in soils, particularly those that control their mobility and soil-plant transfer, should be considered for environmental protection criteria (Kabata-Pendias, 2004).

2.5. Factors influencing the distribution of PTEs and radionuclides

The distribution of elements in the environment is determined by the properties of each element and by various environmental factors (Jaishankar *et al.*, 2014). Soil properties are primarily grouped into physical, chemical and biological, microflora and microfauna (Smičiklas and Šljivić-Ivanović, 2016). After radionuclides and PTEs deposition on the soil surface, the relocation and movement of these elements in soil depend on the soil properties, i.e., pH, cation exchange capacity (CEC), texture, interchangeable calcium and potassium, clay content, grain size, as well as organic matter (Salbu *et al.*, 2004; Skipperud and Salbu, 2015; Gupta and Voronina, 2019; Shiva Kumar and Srikantaswamy, 2014). Similarly, these properties also contribute to the pollution of PTEs and radionuclides in soils and affect solubility (Igwe *et al.*, 2005). Furthermore, the physicochemical properties (pH, soil organic matter, cation exchange capacity, moisture content, clay content, nutrient status, and mineralogy) might influence the migration of these elements in soil (Adesiji and Ademola, 2019). Therefore, it is necessary to characterize the properties of soils and plants to investigate the nature of radionuclides and PTEs in a mining environment, such as the transfer from soils to plants (Soltani *et al.*, 2017).

According to Skipperud and Salbu (2015), the understanding of mobility data requires knowledge of soil physical properties. If mobility factors are very high, it indicates that anthropogenic radionuclides and PTEs have great potential to spread and be rapidly bioavailable in the environment if measures are not put in place (Ogundiran and Osibanjo, 2009). There is a correlation between the CEC values and the values of the pH and organic matter (OM) content (Bielicka-Giełdoń *et al.*, 2013). According to Bielicka-Giełdoń *et al.* (2013), higher pH values and organic matter content result in higher values of the CEC measured. As a result, CEC is considered a good indicator of soil quality and productivity (Ross and Ketterings, 2004). In addition, Agic *et al.* (2015) state that when the pH, organic matter content, CEC, and clay increase, the percentage, and availability of the metals reduce. As a consequence, it is of utmost importance to identify the factors that influence the bioavailability, leaching, and possible toxicity of elements in soils (Violante *et al.*, 2010).

2.5.1. The pH

Soil chemistry plays a major role in nutrient availability for the plants, where nutrient availability is determined by acidity or alkalinity (basicity) of the soil solution (Steed and Reed, 2000). Changes in soil pH (either acidification or alkalization) are regarded as a sensitive indicator of anthropogenic factors (Kabała *et al.*, 2016). There is a possibility for higher mobility of metallic elements when the pH is acidic, and thus greater bioavailability for living organisms and hence more tremendous potential for toxicity (Bielicka-Giełdoń *et al.*, 2013). The pH is one of the factors that have an influence on the mobility of radionuclide and PTEs in the soil (Shiva Kumar and Srikantaswamy, 2014; Ogundiran and Osibanjo, 2009; Dowdall *et al.*, 2008). According to Igwe *et al.* (2005), pH is the most crucial characteristic in determining the chemical environment of plants and soil. Additionally, Tomašić *et al.* (2013) state that the soil pH is a vital soil parameter that is positively correlated with CEC. Furthermore, the metal content associated with carbonates is prone to changes in pH and can be mobilized if pH is reduced (Filgueiras *et al.*, 2002).

It is therefore presumed that the concentration of an element in the soil solution is dependent on the equilibrium between the soil solution and the solid phase, with pH playing a decisive role where the metal mobility decreases with increasing pH (Bielicka-Giełdoń *et al.*, 2013). Similarly, Rieuwerts (2007) states that the separation of trace metals in soils is most often and consistently influenced by the pH of the soil property. Rieuwerts (2007) further says that pH often leads as the parameter that can better estimate trace metal solubility in the opposite association. According to Fite and Leta (2015), the pH is either directly or indirectly affects the migration, transformation, and many reaction pathways of metal retention by soils. Savosko *et al.* (2020) concur that the pH is the predominant soil factor in determining the mobility and availability of radionuclides. Since the pH is the most critical factor controlling the distribution of PTE and radionuclides concentrations, it is consequently possible to predict the future mobility of different elements by predicting the future changes in pH (Sohlenius *et al.*, 2013). According to Kabała *et al.* (2016), a number of soil reaction measures are used worldwide depending on the need or objective, such as distilled water, one (1) mol/L KCl, and 0.01 mol/l $CaCl_2$. Kabała *et al.* (2016) further state that KCl solution has been the most common eluent for soil pH measurement at a soil: solution ratio 1:2.5, which is used to determine soil fertility, soil liming needs, and soil contamination.

2.5.2. The soil organic matter

The organic matter content of soils (SOM) is highly heterogeneous; it is decomposed rapidly in an arid environment and degraded soils (Iurian *et al.*, 2015). However, its deposition is gradual and is determined by climate, soil type, vegetation, and soil organisms (Iurian *et al.*, 2015). According to Rieuwerts (2007), the extensive soil literature shows that SOM is well known to have a significant effect on trace metal behavior. Similarly, El-taher (2015) and Khelifi *et al.* (2019) concur that the OM is one of the factors controlling the bioavailability and the content of PTEs and radionuclides in the soil. Furthermore, Bielicka-Giełdoń *et al.* (2013) state that organic matter is one of the most significant aspects of soil fertility because it is the natural protective barrier against metals as it decreases their mobility. The differences in organic matter content have a substantial effect on the distribution of radionuclides and PTEs, and also, the amount of organic carbon has an essential determinant of element mobility (Sohlenius *et al.*, 2013). According to Hernandez-Soriano and Jimenez-Lopez (2012), organic matter content in soil plays a vital role in the proper uptake for certain elements that are essential for healthy crop development, e.g., Cu and Zn. Calmon *et al.* (2009) state that there is a possibility of different impacts because of changes in soil OM content, depending on whether the respective radionuclide can form organic complexes. According to Roper *et al.* (2019), SOM consists of various C compounds, and many methods have been developed to quantify SOM and its constituent compounds.

2.5.3. The cation exchange capacity

The soil's cation exchange capacity (CEC) is a measure of the amount of negatively charged sites on soil surfaces that can hold cations by electrostatic forces (Jaremko and Kalembasa, 2014). The function of the CEC is to assess soil fertility, nutrient retention capacity, and the capacity to preserve groundwater from cation contamination (Bielicka-Giełdoń *et al.*, 2013). It is essential to know the intended use of the data because of the different methods used to estimate the CEC (Ross and Ketterings, 2004). The CEC influences the mobility of metals, where silicate minerals may provide cation exchange (CE) sites for adsorption/desorption of the metals (Rodgers *et al.*, 2015). Furthermore, CEC is necessary to maintain a sufficient amount of plant-available calcium (Ca^{2+}), magnesium (Mg^{2+}), and potassium (K^{+}) in soils (Jaremko and Kalembasa, 2014). Similarly, for radionuclides, the properties of the soil absorbing complex are the determining factor from soil to plants (Savosko *et al.*, 2020). Savosko *et al.* (2020) furthermore state that if the strength of sorption of radionuclides in the soil is higher, the greater is its CEC and the sum of exchange bases. Similarly, Calmon *et al.* (2009) state that low CEC leads to the increased mobility of radionuclides within the soil profile. Whereas a high CEC decreases metal mobility and availability as well as improves preservation of metal cations, as many studies reveal the significant relationships between the contents of CEC, clay, or organic matter (Du Laing *et al.*, 2009). It is believed that the agrochemical properties of soils play a significant role in the behavior of radionuclides (Savosko *et al.*, 2020).

Different methods have been proposed to measure the CEC and sum of exchangeable cations in soils and many yields in very different results (Jaremko and Kalembasa, 2014). Several studies have been conducted to compare these methods (Ciesielski and Sterckeman, 1997; Ross and Ketterings, 2004; Tomašić *et al.*, 2013; Jaremko and Kalembasa, 2014; Zgorelec *et al.*, 2019; Mustapha *et al.*, 2020).

Ciesielski and Sterckeman (1997) conducted a comparison study between three methods for the determination of CEC and exchangeable cations in soils. In this study, the comparison of the single extraction cobalt hexamine chloride (Cohex chloride) method, the barium chloride method, and the ammonium acetate method was done. The researchers found the barium chloride method and the single extraction Cohex method as equivalent. The researchers suggested that the Cohex chloride can favorably replace the barium chloride method. The results of this study indicated that the specificities of the ammonium acetate method make it difficult to choose an alternative procedure *(Ciesielski and Sterckeman, 1997)*. Likewise, Jaremko and Kalembasa *(2014)* compared four methods for the determination of CEC of soils; Kappen's method, sodium acetate (pH = 8.2), barium chloride, and hexaamminecobalt (III) chloride (Cohex chloride). The barium chloride method and the Cohex chloride method were found to be equivalent. The values obtained with the sodium acetate method (especially for acid soils reach in organic matter and very calcareous soils) and Kappen's method gave an overestimated result. In addition, the comparison of two different CEC determination methods (barium chloride and ammonium acetate) regarding the soil properties was made by Zgorelec *et al. (2019)*. In this study, it was observed that the values of CEC obtained by barium chloride were more corresponded to the actual sorption capacity of soils than the values of CEC obtained by ammonium acetate. Furthermore, the authors point out that even though the pH seven (7) buffered ammonium acetate solution is the widely used method, its use for some acid and/or salty and/or high OM content soils is questionable. Consequently, the ammonium acetate method may result in an overestimation of CEC in soils with pH-dependent charges. In contrast, the barium chloride method resulted in accurate and precise CEC values measured with reference materials and actual samples (Zgorelec *et al.*, 2019). Moreover, the barium chloride method is adopted and accepted as the international standard (Tomašić *et al.*, 2013). In another study, Ross and Ketterings (2004) recommended the process for determining the CEC in soil depending on the intended use of the data. The authors suggested that for regulatory and/or soil classification purposes, ammonium acetate buffered at pH 7 is the recommended procedure. Whereas, to accurately measure the CEC of a soil under field conditions, the barium chloride compulsive method is suggested. Ross and Ketterings (2004) described the advantages and disadvantages of using the barium chloride CEC determination method.

Advantages for $BaCl_2$ CEC determination method

- It is a highly repeatable, precise, direct measure of a soil's capacity to retain non-acid cations (CEC_b).
- It determines CEC_b at the pH and ionic strength of the soil.

Disadvantages for $BaCl_2$ CEC determination methods

- It is very time-consuming and generates hazardous waste ($BaCl_2.2H_2O$).
- It is unlikely to be well-suited for most routine soil testing laboratories and might only be offered by University laboratories.

2.5.4. Meteorological conditions

Meteorological conditions such as rainfall and temperature affect the physicochemical parameters of the soil since they regulate leaching intensity and soil mineral weathering (Isimekhai *et al.*, 2017). In addition, environmental floods and droughts may have a substantial impact on the quality and quantity of water bodies, with direct or indirect effects on ecosystems (Valencia-Avellan *et al.*, 2017). Valencia-Avellan *et al.* (2017) further state that the rainfall (e.g., acid rain) may cause pH changes, disrupting the buffering ability of the river chemistry, causing metals to be desorbed from sediments or soils. Similarly, Hurel *et al.* (2017) assert that the dissolution of minerals as a result of frequent and extreme rainfall, pH variation, and aerobic conditions increase the mobility of trace elements. The mining areas may be affected by the increasing frequency and magnitude of rainfall series events by re-mobilizing and transferring metal-rich sediments and increasing metal-rich runoff, impacting the quality of river water (Valencia-Avellan *et al.*, 2017). A similar approach applies to radionuclides, Walther and Gupta (2015) state that the total amount of rainfall and the intensity of the precipitation are some of the key factors regulating the uptake of radionuclides. Radionuclide migration to deeper soil layers may be enhanced by high-intensity rainfalls. According to Smičiklas and Šljivić-Ivanović (2016), the rain and the soil's ability to immobilize radionuclides are essential factors for regulating activity concentrations available to biota. Therefore, it is necessary not to neglect climatic conditions when studying the mobility of elements (Hurel *et al.*, 2017).

2.6. Analytical methods

Soil is the fundamental predictor of any possible future long-term contamination and helps hazardous areas for human health to be detected (Gad *et al.*, 2019). Several analytical methods such as gamma-ray spectrometry, alpha-particle spectrometry, liquid scintillation emanation counting, gross alpha counting, inductively coupled plasma mass spectrometry (ICP-MS), inductively coupled plasma optical emission spectrometry (ICP-OES), and kinetic phosphorescence analysis have been applied in order to determine the concentrations of uranium and radium-226 (Waggitt, 2008). Similarly, for PTEs, many studies have been carried out to determine the content of PTEs using total or pseudo-total sample preparation procedures (de Souza *et al.*, 2015). de Souza *et al.* (2015) further state that a variety of extraction methods to assess PTE bioavailability in the soil have been proposed; however, there is no agreement about the most effective extractor.

2.6.1. Determination of gamma active radionuclide

Monitoring of ambient radioactivity is vital, and the information on radiation levels in the rock and soil is crucial for the estimation of natural radiological risk resulting from these isotopes (Júnior *et al.*, 2006). In order to assess the health risks posed by radiation exposure, environmental radiation techniques are used to determine the dosage of radiation exposure from environmental sources (Durusoy and Yildirim, 2017). Several radiochemical analytical procedures have been developed based on extraction with ion-exchange column chromatography, liquid organic solvents, and packed columns that are ready-to-use (Oliveira & Carvalho, 2006). Oliveira & Carvalho (2006) further state that, nonetheless, most of these procedures are time-consuming, generate high amounts of toxic waste, such as organic solvents, and do not always allow for high chemical recovery yields. Gamma-spectrometry using a sodium iodide detector has been used for the analysis of radionuclide concentrations (Ademola *et al.*, 2015; Al-Jundi, 2002; Manigandan and Chandar Shekar, 2014). Additionally, according to Júnior *et al.* (2006), gamma-spectrometry with high purity germanium detector is a very effective method for analyzing radionuclides in environmental samples.

2.6.2. Determination of radon gas in soil

Different techniques have been used for measuring the concentrations of radon (Rn-222) and its daughters in the environment, where most common methods rely on the detection of alpha particles. (Baskaran, 2016; Agency for Toxic Substances and Disease Registry, 2013). The Agency for Toxic Substances and Disease Registry (2013) further states that although some methods are based on the detection of emitted gamma rays during the decay process. One of the methods that have been used for the determination of radon is a volume trap detector using an alpha spectrometer, where the material is dissolved in nitric acid followed by additional digestion in hydrochloric acid (Oberstedt & Vanmarcke, 1996). Furthermore, the polonium is auto deposited on a silver plate during drying with an infrared source (Oberstedt & Vanmarcke, 1996*)*. Similarly, Carvalho *et al.* (2007) state that analyses made by alpha spectrometry are performed on less than 63 µm solid material samples in mud from acid water neutralization. These samples are electroplated on stainless or silver foil discs following the complete dissolution of sample material with acids (Carvalho *et al.*, 2007).

Other radon determination methods have been evaluated by Tanaka *et al.* (2017), where the authors studied the accurate measurement of indoor radon concentration using a low-effective volume radon monitor. In this study, a gas-flow ionization chamber, a trace environmental level detector with a photomultiplier tube, and AlphaGUARD was used. In this study, the measurement time and net count required for indoor radon measurements were evaluated. The results indicated that AlphaGUARD being a low-effective volume detector and one of the most popular portable radon monitors, which is currently available, can provide accurate measurements. However, when AlphaGUARD is evaluated against a gas-flow ionization chamber and a trace environmental level detector with a photomultiplier tube, the two monitors were found to be ~10 times more sensitive than alphaGUARD (Tanaka *et al.*, 2017).

2.6.3. Total and pseudo total element determination in soils, plants, and water

Total element concentrations can be determined by total mineralization (digestion of soils and plants). In practice, the hardly dissoluble silicates contents are frequently neglected by strong oxidative acid mixture digestion; in this case, it can be called a pseudo total element content. Later in the dissertation, total and pseudo total procedures will be applied according to the technique used.

Many studies that include the determination of total PTE concentrations have been conducted (Opaluwa *et al.*, 2012; Baran and Tarnawski, 2015; Rosado *et al.*, 2016; Zhang *et al.*, 2017; Isimekhai *et al.*, 2017; Stevanović *et al.*, 2018). The determination of total PTE concentration and its spatial distributions in soils could be partially beneficial in determining, monitoring, and evaluating the potential source of contamination in the area of concern (Stevanović *et al.*, 2018). Additionally, the determination of total PTE concentration has been a valuable indicator for the appropriate assessment of sediment contamination (Baran and Tarnawski, 2015). However, total concentrations might provide inadequate information about the potential mobilization and associated risk of PTEs in soils (Sungur *et al.*, 2014; Rinklebe and Shaheen, 2017). Although the mobility, bioavailability, and eco-toxicity of PTEs depend more on their chemical speciation rather than on their total content (Soltani et al., 2017), it is helpful to determine the total concentrations of PTEs. Risk assessment management and regulatory procedures, in general, have prioritized the soil total content of PTEs as a possible source of human, cattle, and wildlife exposure (Adamo *et al.*, 2014). Intawongse and Dean (2007) acknowledge that not all methods are helpful to study all PTEs under different soil conditions.

Ogundele *et al.* (2015) state that studies on PTE accumulation in the soils is well documented. However, studies focused on soil contamination near mines often pay little attention to downstream impacts on plants (Soltani *et al.*, 2017). The soil is the primary source of PTEs for plants, animals, and humans (Hooda, 2010). Since soil to plant transfer is a significant process that exposes humans to PTEs through the food chain (Keshavarzi *et al.*, 2015), it is crucial to determine the total concentration in various environmental mediums to understand the level of accumulation. On the other hand, different plant species have been used to remove the PTEs from the soil and have demonstrated the ability to extract and store these PTEs in roots or aerial parts for remediation purposes (Kede *et al.*, 2014). According to Ojuederie and Babalola (2017), bioremediation is an eco-friendly and productive way of using the biological processes of microorganisms and plants to restore environments contaminated with PTEs and eliminate harmful contaminants. However, the plants that are not intended for remediation purposes can uptake these PTEs from the soil and be part of the food chain, thereby posing a danger to human and environmental health (Shaheen *et al.*, 2016). Although the majority of these PTEs have essential biological functions in plants as essential micronutrients (Jaishankar *et al.*, 2014), they are needed in minimal quantities (Lohry, 2007). Therefore, elevated levels of PTEs in the soil due to anthropogenic activities pose environmental and health risks (Hooda, 2010).

Additionally, anthropogenic activities such as uranium mining have been a significant source of contamination to aquatic environments due to environmental problems associated with deactivated mines (Ferrari *et al.*, 2017). Contaminants such as U can leach or be mobilized from the uranium spoil deposits into the groundwater or surface water. Waterborne stream contamination is primarily due to the transport of uranium from mine tailings as an aqueous phase with a possibility of uranium transported in colloid form (Winde *et al.*, 2004). Quesada-González *et al.* (2018) state that the U that exists naturally in granite and other mineral deposits may also contribute to groundwater contamination. The aquatic environment usually is the ultimate destination of contaminants from problematic areas, where the surrounding biota and humans can be affected (Ferrari *et al.*, 2017). According to Winde (2016), in order to quantify a contamination hazard in the environment, there are two significant aspects to be considered (the concentration of the pollutant and its quantity). Furthermore, maximum permitted concentrations for different contaminant types are legislated to minimize environmental and health risks (Winde, 2016). It is the mine’s prerogative to ensure that any of the materials do not surpass the thresholds in place and to ensure that the drinking water is protected by conducting routine environmental monitoring. Goulet *et al.* (2015) state that further toxicity studies with water chemistry downstream from uranium mines are required to help predictive evaluations of the impacts of U discharge into the surrounding environment.

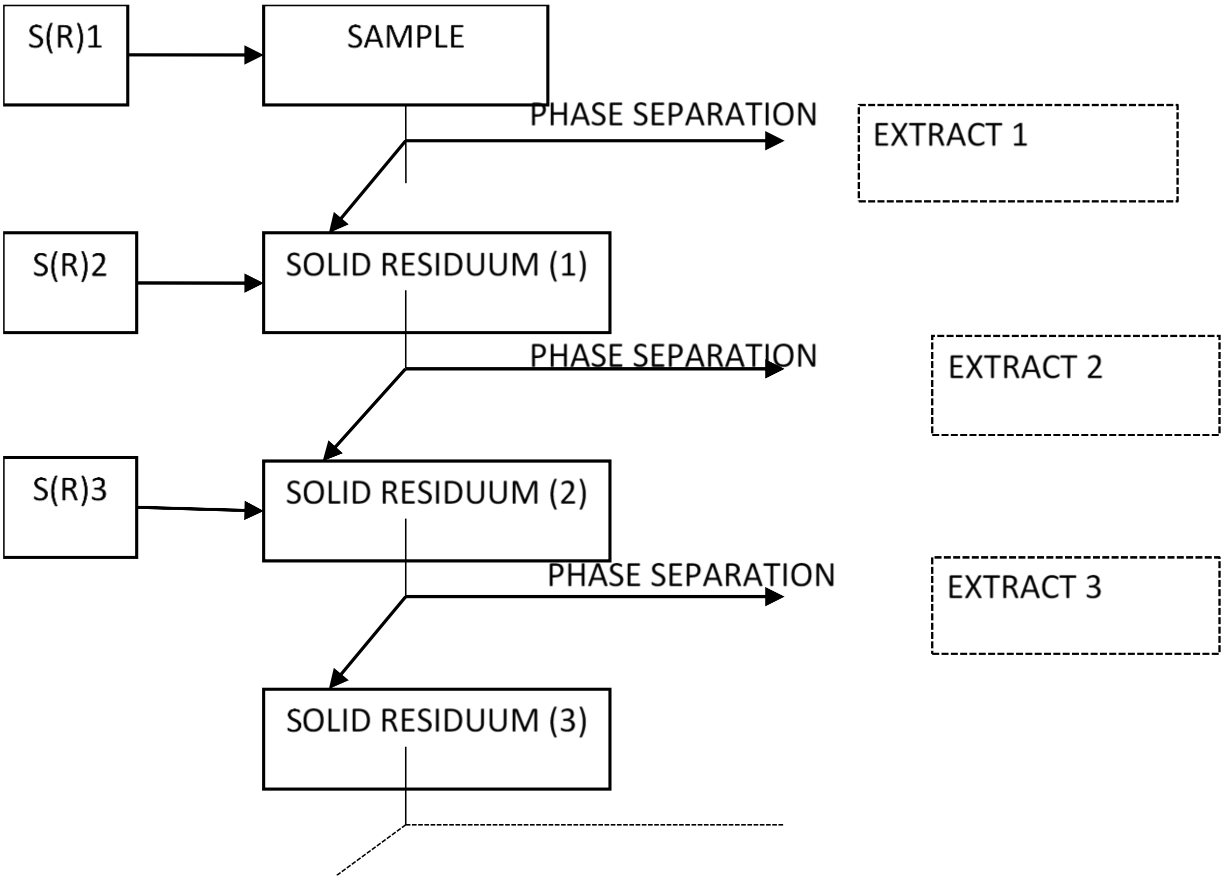
To address the environmental problems, during the Mecsek uranium mine operation (1956 – 1997), the town of Pecs and the surrounding countryside, including its hillside vineyards and groundwater, were given high priority regarding environmental protection (IAEA, 2009). As part of the remediation process, frequent monitoring of radioactivity and PTEs in groundwater near the spoil deposits, seepage water from the waste rock pile, mine water, and treated mine water was conducted. According to Mecsekérc Zrt. (2017), this hydrogeological monitoring is to ensure that the Pellérd-Tortyogó and the northern karstic water resources are protected against any contamination and to predict any process indicating the distribution of contamination to be able to plan for any intervention. Amongst the elements being monitored, U has been a priority due to its potential chemotoxicity and high mobility. Iurian *et al.* (2015) state that although U is a radioactive element, it is also a PTE whose primary effect on human health is through a chemical toxic impact on the kidneys. Iurian *et al.* (2015) further state that uranium is commonly managed as a chemical toxic species due to its low radiological risk compared with chemical toxic risk. Therefore, as part of the monitoring program, the concentrations of U in water from the Mecsek uranium mine were determined in this study. It has been indicated that the most critical areas to focus on regarding hydrological monitoring include contamination impacts resulting from waste rock dumps No. I, No. III, and also the monitoring related to the remediation of several ponds (Mecsekérc Zrt., 2017). Mecsekérc Zrt. (2017) further states that this process is predicted to continue for at least 15 to 30 years.

2.6.4. BCR sequential extraction

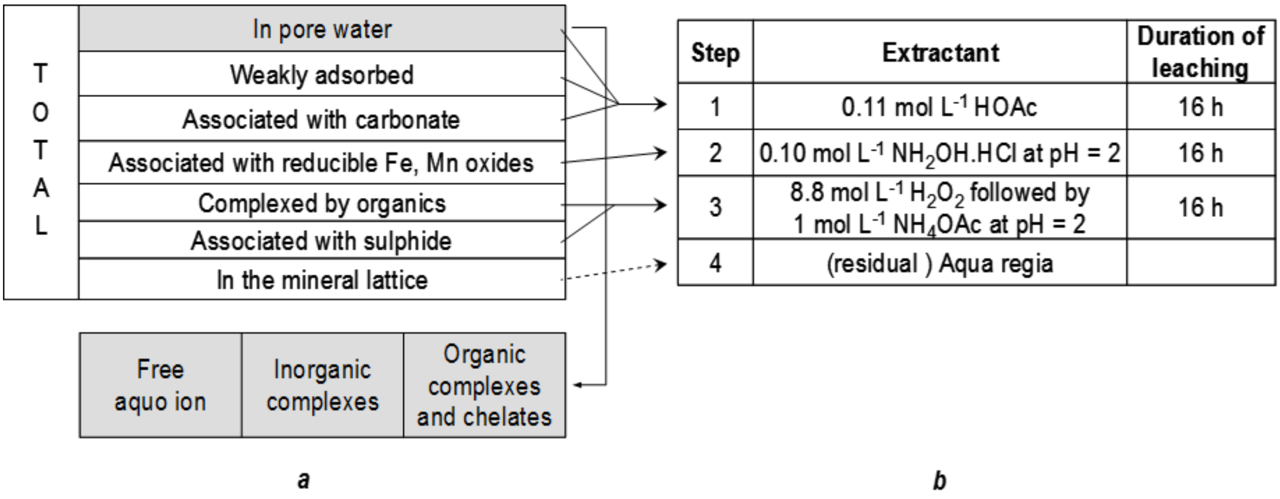
It has been established that total concentration cannot provide the required information for the bioavailability of PTEs; hence speciation is a useful tool for obtaining this information (Geography and Labor, 2008; Sungur *et al.*, 2014). Moreover, the total metal content alone does not provide adequate knowledge about the dangers caused by metals because they are accessible to plants to a different degree depending on their type of occurrence (Geography and Labor, 2008). Okoro *et al.* (2017) emphasize that the characteristics and toxicity for the total concentration of PTEs are often not accurately represented. Okoro *et al.* (2017) further state that to address these challenges, analyzing the individual fractions of the soil to which the metals are attached to better understand their actual and potential environmental effects is helpful. According to Qasim and Motelica-Heino (2014), additional data on the concentrations of particular physicochemical forms of the PTE is required as the behavior of PTE in the environmental system relies on chemical speciation. The method of fractional leaching is traditionally applied for the migration of radionuclides and PTEs investigation in soil, including subsequent soil processing with different reagents to determine mobile forms: water-soluble, exchangeable, acid-soluble; and firmly fixed (Kuterbekov, 2019). According to Gleyzes *et al.* (2002), sequential extraction is one of the most widely-applied procedures proposed by Tessier *et al.* (1979). The sequential extraction studies have been providing an effective way of assessing the metals associated with the main accumulative phases in sedimentary deposits (Filgueiras e*t al.*, 2002).

In the study by Heltai *et al*. (2015), the development of the BCR sequential extraction procedure and the methodological problems connected to its practical application are discussed in detail as follows:

"The environmental mobility and biological impact of heavy metals in environmental systems are determined by their chemical species forms. Biological risk assessment of contaminated areas, therefore, in principle, should require the total speciation of all chemical elements present in the system. However, due to the complex structure of solid environmental samples (e.g., soils, sediments, gravitation dust, mining waste, etc.), this analytical task in practice cannot be completely solved. As a compromise fractionation of heavy metal content according to environmental mobility and estimation of biological availability by sequential extraction procedures became the main tool of primary risk assessment of heavy metal contamination in the soil/water/atmosphere system. The term "fractionation" was recommended by the IUPAC in 2000 (Templeton *et al.*, 2000). Mobility of an element according to different solubility of different binding classes can be estimated by solvents sequentially applied which contain increasingly more and more aggressive reagents (Caroli, 199; Kersten and Förstner, 1995). The strategy of sequential extractions can be presented on the following scheme, where the expression of SOLVENT(REAGENT) is abbreviated by S(R).



The first sequential extraction procedure for characterization of mobility of heavy metals in aquatic sediments was developed by Tessier *et al.* (1979) who classified the metal content of sediments according to solubility and binding forms of metallic species as it is presented in the following scheme (part "a"). Based on this essential work several 5-8-step sequential extraction schemes were developed by which the original chemical structures (binding forms) are gradually dissolved and/or decomposed (Gleyzes *et al.*, 2002). The total time demand of these procedures is 5-6 days, while duration of the applied batch leaching steps is determined by long setting up time of partition and other heterogeneous equilibria. The proposal for a 3(+1)-step sequential extraction procedure issued by EU Community Bureau of Reference (BCR) in 1993 is a simplified version of the above-mentioned schemes (Ure *et al.*, 1993), but it is still time consuming and labour intensive (scheme part "b"). As it can be seen in the scheme, in this method the water soluble, the weakly adsorbed and the carbonate-associated metal fractions are extracted simultaneously by acetic acid in the 1st leaching step, in 2nd step the metal fractions associated with reducible Fe and Mn oxides are determined after reduction by hydroxil-amin and then the fraction of metals complexed with organics and associated with sulphides is extracted after oxidization by H_2O_2. In the (+1) step the residual fraction can be obtained by means of digestion with aqua regia or HNO_3/ H_2O_2.



Step	Extractant	Duration of leaching
1	0.11 mol L^{-1} HOAc	16 h
2	0.10 mol L^{-1} $NH_2OH.HCl$ at pH = 2	16 h
3	8.8 mol L^{-1} H_2O_2 followed by 1 mol L^{-1} NH_4OAc at pH = 2	16 h
4	(residual) Aqua regia	

Re-adsorption of extracted metals during the phase separation can lead to analyte losses. In the extracts the high reagent’s concentrations may cause strong matrix effects during the spectrochemical element detection. The CRM 601 sediment reference material issued in 1997 was certified only for five elements (Cd, Cr, Ni, Pb, Zn) to this procedure (López-Sánchez *et al.*, 1998, Quevauviller *et al.*, 1997). During the elaboration of the next (BCR 701) sediment materials, the original BCR procedure was modified to avoid the analyte losses during the second leaching step due to the pH instability (Rauret *et al.*, 1999; Sahuquillo *et al.*, 1999; Pueyo *et al.*, 2001; Rauret *et al.*, 2001)”.

Sequential extraction methods may provide valuable knowledge about the binding mechanisms and the possible mobility of radionuclides and metals in the environment (Skipperud and Salbu, 2015). Sungur *et al.* (2014) state that using several sequential extractions to identify the different fractions of PTEs is the key to the understanding of the geochemical processes and bioavailability. Through these methods, the sample is treated with a series of extraction reagents with the explicit purpose of dissolving the various phases (or fractions) of the sediment and thus allowing the associated metals to be determined (Dos Santos *et al.*, 2012). Furthermore, these methods enable measurement of broader aspects or phases (e.g., bioavailability) and, in the majority of cases, it is adequate for environmental policy purposes (Rauret *et al.*, 1999). Therefore, a validated technique is required to ensure the reliability and accuracy of the different results obtained (Perez-Moreno *et al.*, 2018).

Numerous extraction techniques have been employed to mimic various environmental scenarios, using strong acids to test the overall content and bioavailability of PTEs (Qasim and Motelica-Heino, 2014). However, comparing the data obtained from various sequential extraction schemes is difficult due to the lack of uniformity in these techniques since the results rely on the extraction method used (Rauret *et al.*, 1999; Pavlović *et al.*, 2018). Hence, the BCR sequential technique is recommended to determine the fractionation or distribution of PTEs in sediments and soils (Kede *et al.*, 2014). The BCR technique was developed when the Standards, Measurements, and Testing Program (SM&T), formerly known as the Reference Bureau of the European Community (BCR), initiated a joint project in 1992 aimed at harmonizing and standardizing the various procedures used in soil and sediment to fractionate metals (Rauret *et al.*, 1999; Zhu *et al.*, 2015; Pavlović *et al.*, 2018). Since then, various studies have been successfully conducted to study the PTEs in the soils and/or sediments using the BCR technique (Pueyo *et al.*, 2008; Sutherland, 2010; Wang *et al.*, 2010; Nemati *et al.*, 2011; Bielicka-Giełdoń *et al.*, 2013; Adamo *et al.*, 2014; Barać *et al.*, 2016; Mittermüller *et al.*, 2016; Rosado *et al.*, 2016; Rinklebe and Shaheen, 2017; Pérez-Moreno *et al.*, 2018; Heltai *et al.*, 2019).

Sutherland (2010) conducted a review study on the BCR® -701 methodology. The author observed forty sets of data from the literature. The outcome of the observed data was described as generally of high quality and encouraging. The author noted that there were concerns regarding the sequential extraction procedures in general, there have been reports expressed over the lack of specificity in element removal, re-adsorption, and subsequent redistribution among phases, the absence of comparability as experimental parameters between studies differs widely, and the potential for contamination increases as the number of steps increases, and the length of time required to process successive steps which typically take over several days.

It has been proven in recent studies that the three-step BCR sequential extraction scheme is not without problems and that work is needed to identify the causes of poor reproducibility (Rauret *et al.*, 1999). While the use of sequential extraction schemes may include problems such as non-selectivity, re-adsorption, etc., these methodologies are useful for metal fractionation in order to predict metal mobility and bioavailability (Filgueiras *et al.*, 2004). While these extraction procedures are not necessarily precise, they may help assess the relative contribution of mixed sources of pollution to this mobility and may provide additional data on the mobility of elements in soils with changing environmental conditions (Pueyo *et al.*, 2008). On the other hand, Tlustos *et al.* (2005) contend that even though these procedures are usually time-consuming and require skilled personnel and good analytical instrumentation techniques, they give the most accurate information about fractionation and transformation of elements in soil, especially in relation to soil pollution and long-term effect of soil amendments. Furthermore, these methods fulfil all the required criteria for wide-ranging implementation: quick, applicable to all types of soils and sediments, low cost, and easy to understand and comparable results (Rosado *et al.*, 2016).

Moreover, Heltai *et al.* (2019) suggest that there is a need for the certification for other environmental matrices since the BCR certified reference material sample is only certified for the fractionation of six elements (Cu, Cd, Cr, Ni, Pb, Zn) in freshwater sediments. Whilst initially envisioned for use in sediments; the BCR scheme has been successfully applied in the analysis of a wide range of other substrates, including several different soil types (Bielicka-Giełdoń *et al.*, 2013).

Sequential extraction technique may be time-consuming; however, it offers comprehensive information on the origin, mode of occurrence, biological and physio-chemical availability, mobilization, and transport of trace metals (Filgueiras *et al.*, 2002). Similarly, Violante *et al.* (2010) point out that the bioavailability of PTEs and metalloids, their biological uptake, as well as their ecotoxicological impacts on the soil biota, can be better understood in terms of their chemical speciation. Different factors such as extractant pH, temperature, length of extraction, extraction in an inert atmosphere, and the form and concentration of reagents can be studied to determine sources of uncertainty in the BCR three-step sequential extraction procedure (Rauret *et al.*, 1999). Heltai *et al.* (2018) reiterate that considering these methodological problems, the use of BCR sequential extraction continues to be the traditional technique for evaluating environmental risks of PTE pollution, thus offering the potential for quantitative analysis of environmental mobility.

Environmental pollution studies on soils are often based on the use of leaching or extraction procedures (e.g., sequential extraction procedures) that allow for the measurement of mobility and bioavailable forms of elements (Rauret *et al.*, 1999). In environmental research, the fractionation of PTEs is critical in determining the amount of PTEs that can be potentially soluble, mobile, and accessible to the ecosystem that cannot be calculated with the total concentration of these elements alone (Ogundiran and Osibanjo, 2009). Sequential extraction procedures help to provide essential knowledge about the fraction distribution of these PTEs in soils (Saffari *et al.*, 2009) and have become the standard methodology in risk evaluation on PTE contaminations (Heltai *et al.*, 2018). As far as environmental degradation is concerned, a BCR sequential extraction technique offers critical knowledge about metal mobility and bioavailability and assures further use in various terrestrial and aquatic fields (Sungur *et al.*, 2014). Moreover, these techniques fulfill the required criteria for implementation across a broad range of applications: quick, cost-effective, easy-to-use, and provide comparable outcomes for any form of sediment or soil (Rosado *et al.*, 2016).

2.7. Instrumental analytical methods

Potentially toxic elements may be found in all types of ecosystems, and therefore, several instrumental analytical techniques may be used to evaluate the concentration levels of PTEs in different kinds of samples (Helaluddin *et al.*, 2016). Determinative methods that can be used for PTEs are flame atomic absorption spectrometry (FAAS) (Ademola *et al.*, 2015), atomic fluorescence spectroscopy (AFS); anodic stripping voltammetry, inductively coupled plasma optical emission spectrometry (ICP-OES) and inductively coupled plasma mass spectrometry (ICP-MS) are among several analytical methods which have been used for the determination of PTEs contents in marine environments (Okoro *et al.*, 2012). Atomic absorption spectrometry (AAS), atomic emission/fluorescence spectrometry (AES/AFS), ICP-MS, ICP-OES, neutron activation analysis (NAA), X-ray fluorescence (XRF), and anodic striping voltammetry (AVS) have been listed as the most predominant analytical techniques by Helaluddin *et al.* (2016). According to Feist *et al.* (2008), a significant number of elements may be determined simultaneously or sequentially using instrumental techniques such as ICP-OES or AAS.

Sereshti *et al.* (2012) state that ICP-OES provides fast multi-element simultaneous analysis, leading to various applications with a wide variety of complex and organic matrices. Furthermore, even the most refractory elements are atomized and stimulated with comparatively low interference effects as the samples undergo an inert and high-temperature medium (Sereshti *et al.*, 2012). Similarly, Fassel and Kniseley (1974) assert that the ICP-OES has become an important methodology for assessing metals in various sample matrices. Fassel and Kniseley (1974) further state that this method utilizes a range of nebulizers or sample introduction methods for injecting fluid samples through radiofrequency (RF)-induced argon plasma. Most laboratories use ICP-OES techniques for research and environmental control (Bettinelli *et al.*, 2000). According to Bettinelli *et al*. (2000), the successful use of these techniques depends on various conditions such as good detection limits, linear curve calibration, acid dissolution compatibility, and low matrix sensitivity.

Heltai *et al.* (2019) studied the application of flexible multi-elemental ICP-OES detection in fractionation of potentially toxic element content of solid environmental samples by a sequential extraction procedure. The main goal of this study was to elaborate on the multi-elemental ICP-OES method, which is a flexible and reliable application for the evaluation of PTE mobility in soils and in sediments by BCR fractionation. In this study, the authors recommended that the flexible multi-elemental ICP-OES method should be applied for analysis of various BCR-extractant-solvent media for the reliable detection of all PTE contamination, in the case of appropriate matrix-matched calibration and internal standard application and yttrium standard application. The authors’ conclusion stated that the ICP-OES method was successfully applied for controlling the remediation of PTE contaminated lakes like the Gödöllő-lake system, and it can also be applied successfully for characterization of environmental mobility of PTE and other micro-and macro-elements content in soils. The authors further stated that among the simultaneous multi-elemental capacity methods, ICP-OES has adequate detection power for PTEs in environmental studies, and its robustness is better than that of the ICP-MS technique, which has better detection power than ICP-OES.

2.8. Regulatory measures

Uranium mining is currently being undertaken under radically different conditions and is now the most regulated and one of the world's safest forms of mining (OECD, 2014). International Atomic Energy Agency (IAEA), United Organisation for Economic Co-operation and Development (OECD)/Nuclear Energy Agency (NEA), World Nuclear Association (WNA), and International Commission on Radiological Protection (ICRP) as well as national regulatory bodies and companies, support important platforms that collect data and provide information for regulators and operators to better manage uranium mining and milling to mitigate any adverse industrial impacts (Collier, 2016), especially with regard to potential radiological effects. Apart from some water resource conservation regulations, there was virtually no regulation on environmental protection (Waggitt, 2008). Amid CRP guidelines, also accepted by the IAEA and OECD/NEA among other international organizations, several countries in the last decade have implemented new legislation on radiological safety, such as the EU (Carvalho, 2011). Carvalho and IAEA (2004) further state that these radiation safety laws, in conjunction with other environmental protection legislation, contribute to the remediation of old uranium mining and milling sites.

The legacy problems of uranium mines today arose because of the lack of legislation earlier on (Waggitt, 2008). Furthermore, many of the countries most affected by these legacy issues have not developed sufficient environmental protection laws and resources (Waggitt, 2008). Regulatory standards for PTE levels for soils have been established; however, there is a vast inconsistency between the different countries as to the critical value of each contaminant (He *et al.*, 2015). Some countries have recently begun to incorporate the bioavailability principle in their environmental protection and remediation legislation (Violante *et al.*, 2010). Following these developments, Hungary introduced the soil toxic element contamination limit that is regulated by the Hungarian Government (6/2009. (IV. 14.), 2009) based on the regulation of other EU member countries (Rékási and Filep, 2012).

The International organizations and national authorities responsible for radiological protection, such as the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), the World Health Organization (WHO), and the IAEA, have placed regulatory measures to follow for the protection against ionizing radiation. A UNSCEAR scientific committee of the General Assembly, the body in the United Nations system with a mandate to assess and report levels and effects of exposure to ionizing radiation, has reported on the permissible limits and average radionuclides worldwide (UNSCEAR, 2000). The data from United Nations Educational, Scientific and Cultural Organization (UNESCO)/WHO/United Nations Environment Program (UNEP) (1996), WHO (2004, 2006), UNSCEAR (2000), and Food and Agriculture Organization of the United Nations (FAO)/WHO, (2011) reports are used as a guideline to evaluate the results of the radiological and PTE monitoring programs against the permissible limits recommended by these organizations in various environmental matrices around the world.

3. MATERIALS AND METHODS

This chapter explains the detailed steps taken to achieve the results. At the beginning of this chapter, a summarized methodology is shown in a flowchart, and a description of a study area is outlined, followed by the description of the types and numbers of samples collected. This chapter presents the analysis of soil chemical properties that might affect the mobility of PTEs and radionuclides. Methodologies that are used to determine radionuclide concentrations and total concentrations of PTEs are mentioned in detail, and the transfer factors of these elements could be determined as a result. Finally, this chapter concludes by detailing the BCR sequential extraction procedure used to assess the mobility of PTEs in Mecsek uranium mine spoil deposit No. I.

3.1. Methodology flowchart

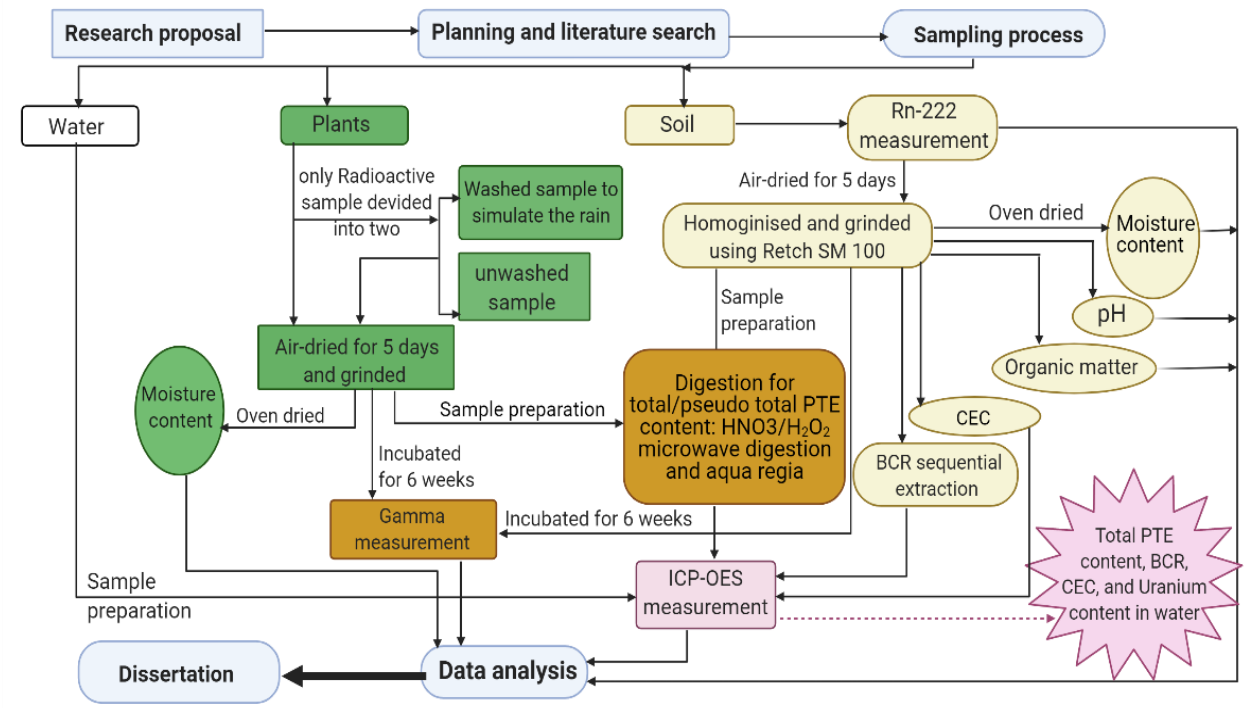


Figure 6. Flow chart summarizing the methodology of this research (created with BioRender.com)

3.2. Study area

Figure 7. The actual Mecsek uranium mine deposit No. I

Soil and covering plant samples were collected on the 11th of July 2018 from four sampling locations in Mecsek uranium mine deposit No. I in Pecs, South-west of Hungary (Figure 7.) and (Figure 8.): (1) at the top of the deposit - Rn-M11, (2) on the slope of the deposit – Rn-M12, (3) at the bottom field – Rn-M13 and (4) at the bottom edge of the slope of the deposit - radioactive point (Figure 8.) illustrates deposit No. I with gamma dose rates that were measured previously as part of the monitoring program. The radioactive point was chosen based on the knowledge of its high gamma dose rate ranging between 100 and 200 nGy/h to give an indication of whether PTE concentrations would also be high in this location. Spoil deposit No. I was covered with different types of soils taken from various locations, mixed together to form a soil covering layer, and it was also covered with different kinds of vegetation, as indicated in (Figure 7.). Underneath the soil covering layer, there were natural rocks.

Eleven water samples were collected between the 20th of October 2020 and the 27th of October 2020 during one of the routine monitoring processes conducted by the Mecsek uranium mine. The purpose for collecting these water samples was to determine the potential mobility of uranium from three spoil deposits No. (I, II, III). Water samples were collected from various areas, including groundwater, seepage mine water, treated mine water, and mine water from different deposits.

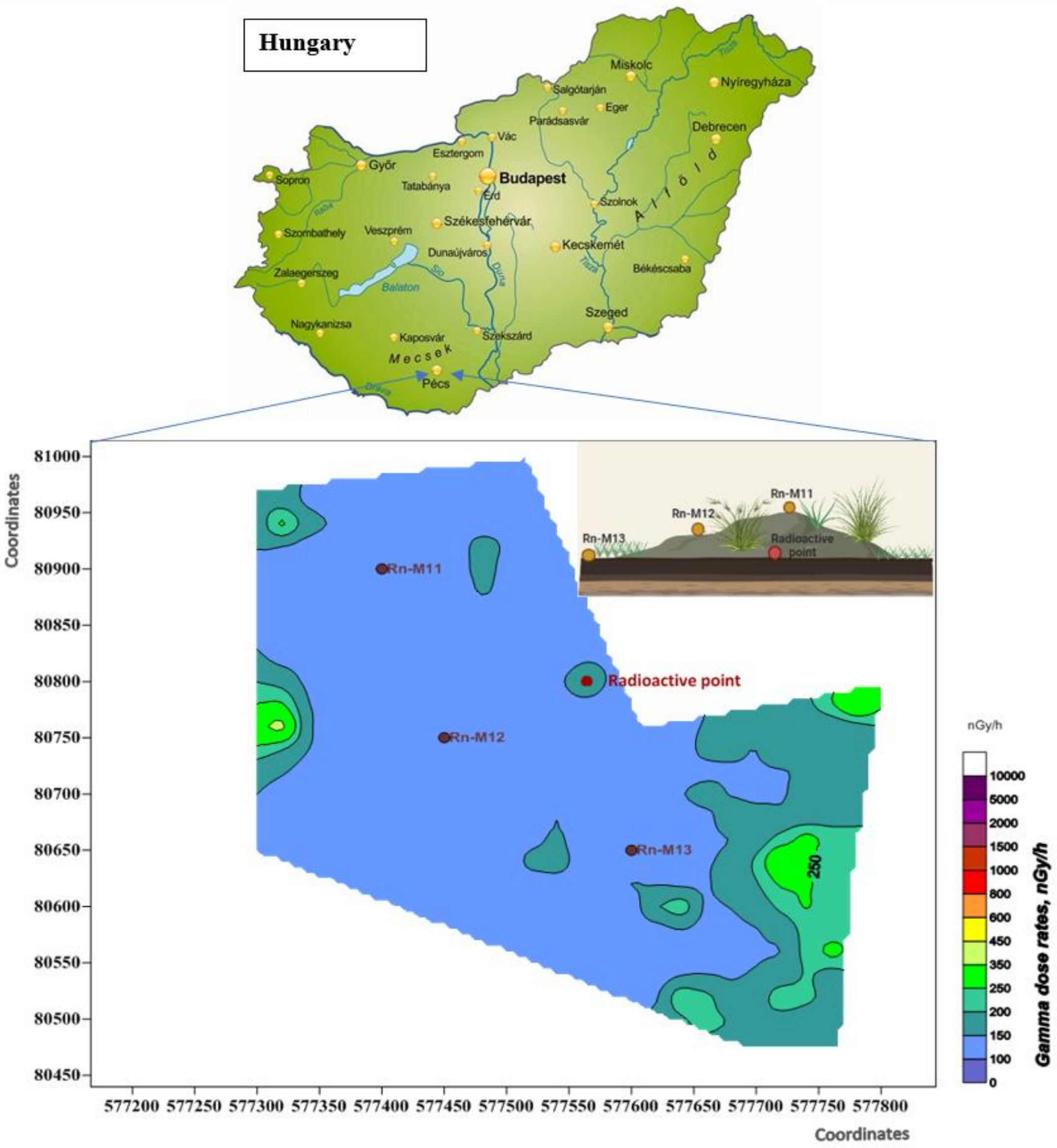


Figure 8. Site location map of Hungary (OrangeSmile.com, n.d.) *and the soil and plants sampling points in Mecsek uranium mine from spoil bank No. I, Pécs, 2017 showing the gamma dose rates distribution with an insert of a schematic diagram of spoil deposit No. I*

Geological information about the study site

Regarding the geology and geography, the mining site is located in the hilly countryside, and the main part of the milling site is a flat area (Juhasz and Erdi-Krausz, 1993). Juhasz and Erdi-Krausz, (1993) further state that the geological formation of the mining site mainly consists of limestone, sandstone, and marl.

3.3. Soil, plant, and water sampling

3.3.1. Soil sampling

Soil samples were collected using a hand auger instrument labeled with different measurements. Each soil core sample was taken from different depths: 0 – 25 cm, 25 – 50 cm, 50 – 75 cm, and 75 – 100 cm. For sample location Rn-M11, 75 – 100 cm depth could not be sampled as this was hard rock. Radioactive soil was possible to sampling only from the top 0-25 cm layer. All samples collected were carefully placed in clean polyethylene bags before transported to the laboratory.

3.3.2. Plant sampling

Covering plant samples (a mixture of Asteraceae and Quackgrass - Elytrigia repens) were collected by cutting from the surface of the plants with a knife. The reason for the plant samples to be collected by cutting them from the surface is because when determining the potential risk that may impact the animals and humans, it is important to look at how the animals graze in the field. Under normal circumstances, animals graze on the surface and rarely eat roots; eventually, humans eat animals. Therefore, to determine the true reflection of the potential risk, only the part of the plants that are above the surface was taken into consideration.

Plant samples were collected from a 1.5 m^2 area of the same locations where the soil samples were also collected from (Rn-M11, Rn-M12, Rn-M13, and radioactive point) to determine if there is any biological uptake of PTEs and radioactivity from the soil.

3.3.3. Water sampling

A total of eleven water samples were collected from various places around the Mecsek mine. From these samples, six samples were groundwater samples, of which four samples (1) Pk-29/1, (2) Pk-33/1, (3) Pk-44/3, and (4) 1504/1 were collected from monitoring well near the spoil deposit No. I and two samples (5) P-2/5 and (6) P-2/6 were collected from the water pumping well at former percolation area No. II.). One sample: (7) Északi-táró was mixed water: mine water from the spoil deposit No. III and leaking water from precipitation collected from the gravitational effluent mine water (north mines). Two samples were seepage water; one sample: (8) CS-0 collected from the spoil deposit No. II and another sample: (9) IIIm. Gyűjtő collected from spoil deposit No. III. One sample: (10) 6/11. szint was mine water from the mine No. I collected from water pumping well. The last sample (11) Elfolyó was treated mine water collected from the mining water treatment plant. The samples were labeled based on the mine description.

3.4. Characteristics that may affect elemental mobility

3.4.1. Meteorological conditions

The average precipitation at Pécs was obtained from the meteorological data extracted from the Hungarian Central Statistical Office Websites (https1://www.ksh.hu/docs/eng/xstadat/xstadat_annual/i_met002cc.html) and (https2://www.ksh.hu/docs/eng/xstadat/xstadat_infra/e_met006.html).

3.4.2. Determination of soil and plant moisture content

Approximately 25 g of soil were dried in an oven at 105°C for 72 hours, cooled in a desiccator, and weighed for the determination of the soil moisture content. The amount of water present in the soil (% moisture content) was determined (Nielsen, 2019).

Similar to soil samples, the plant samples were dried in an oven at 105°C for 72 hours, cooled in a desiccator, and weighed for the determination of the soil moisture content (Ismail, 2017).

3.4.3. Determination of the soil pH

In order to determine the pH of the soil collected from the Mecsek uranium mine, the procedure below was followed:

The soil pH was determined in 1 mol/l KCl (1:2.5 soil to KCl ratio) with the aid of a Glass-electrode pH meter. The pH meter was calibrated with pH 7.0 and pH 4.0 buffer standards before use. Ten grams of homogenized soil was weighed into a centrifuge tube, and 25 mL of 1 mol/l KCl added to it. This could stand for 1 hour and be shaken occasionally. Following this, the electrodes of the pH meter were inserted into the partly settled suspension and the pH recorded. The soil pH was measured in triplicate.

3.4.4. Determination of cation exchange capacity

The CEC in soil was determined according to the Hungarian Standard MSZ 1484-3:2006 (2006). Approximately 4 g of each homogenous soil was weighed in duplicate, and the mass was recorded. 0.1 M $BaCl_2$ solution was prepared and buffered by adjusting the pH to 8.2 with triethanolamine ($C_6H_{15}NO_3$). 40 mL of 0.1 M $BaCl_2$ solution was added to each soil sample. The samples were shaken for 2 hours using a Yellow Line OS5 shaker at 560 rpm. After 2 hours, the sample solid and liquid phases were separated by centrifugation for 10 minutes at 5000 rpm. The samples were filtered using Munktell filter paper grade 389. The samples were diluted by pipetting 0.1 mL of a sample into a 10 mL volumetric flask and topped with 9.9 mL of distilled water. The Ba was then measured in samples using ICP-OES.

3.4.5. Determination of the soil organic matter

Approximately 1 g of each homogenized and sieved soil sample was weighed in duplicate. Total carbon (% C) was determined using a Vario MAX cube CNS Analyzer instrument. Furthermore, the CaCO3 content (%) was determined by employing the Lloyd Kahn procedure using LECO CR-412 Carbon Analyzer with an IR detector (Bernard *et al.*, n.d.). From the combination of these results, it was possible to determine the total organic carbon (%TOC). Then the organic matter (% OM) was calculated from % TOC.

The LECO CR-412 Carbon Analyzer was calibrated prior to the analysis of samples. Different amounts of high purity calcium carbonate standard (99.95% purity, carbon content of 12.0%) were used to calibrate the instrument. The approximate amounts of calcium carbonate used for the six-point calibration were 0.01 g, 0.05 g, 0.10 g, 0.25 g, and 0.50 g. An empty carbon-free combustion boat was analyzed as a blank for the calibration curve. The calibration curve provided an analysis range of approximately 0.0 to 0.06 g total carbon. Each calibration standard fell within 3% of the known percent carbon value to meet acceptance criteria. A continuing calibration check standard (mid-level standard) was analyzed for every ten samples and had to be within 5% of the known value of the standard. Total carbon was analyzed by placing approximately 0.350 g of dried, grinded, and homogenized sample into a clean, carbon-free combustion boat. The sample boat was placed on the autosampler rack assembly and loaded onto the LECO carbon analyzer. Total organic carbon was analyzed by placing approximately 0.350 g of dried, grinded, and homogenized samples into a clean, carbon-free combustion boat. Each sample boat was treated with phosphoric acid drop by drop until the sample stops “bubbling,” and the samples were completely moist with acid. The samples were placed into an oven set at 40°C for 24 hours and then transferred to an oven set at 105°C. Once the samples were dry, the boat was placed on the autosampler rack assembly and loaded onto the LECO carbon analyzer.

3.5. Radioanalytical procedure

3.5.1. Soil sample preparation

Before the soil homogenizing process, Rn-222 was measured for all soil samples. After which, all soil samples were air-dried for five days. After five days, the samples were physically homogenized and grinded using Retch SM 100 with a 0.25 mm sieve instrument. Approximately

30 g of homogeneous soil samples were weighed and airtight sealed in plastic containers for the gamma-ray measurements. Samples were kept in storage for six weeks before gamma-ray analysis took place; this incubation period allows Ra-226 and its short-lived progenies to achieve secular equilibrium noting the exact dates and times from sealing was recorded.

3.5.2. Plant sample preparation

Only the radioactive plant sample was separated into two portions, half of the sample was not washed, and the other half was washed first with low to medium pressure water for five minutes to simulate the rain. This washing process was conducted on this sample to determine whether the radioactivity concentration is due to the contamination through air particles/dust or biological uptake through the soil.

After this process, all plant samples were air-dried for five days and grinded into a powder form. Approximately 8 g of grinded plant samples were weighed and airtight sealed in plastic containers, and kept for six weeks to allow the growth of Ra-226 and its short-lived progenies before gamma-ray measurements.

3.5.3. Radon measurement by AlphaGUARD

The calibration for the AlphaGUARD is done every ten years at the manufacturing company. The specified calibration tolerance by the factory is ± 3 %. The current calibration values are shown in (Table 1.) below.

Table 1. AlphaGUARD PQ2000 / MC50 calibration values

Calibration for:	**Target value (Bq/m^3)**	**Measured value (Bq/m^3)**	**Radon mean deviation (%)**
1 kBq/m3	1175	1171.1	-0.33
10 kBq/m3	11142	11042	-0 9

Sample analysis

Radon (Rn-222) was measured for each soil sample for 24 hours using Genitron Instruments AlphaGUARD PQ2000/MC50 Multiparameter Radon Monitor before the samples were counted for 24 hours each. The results were recorded every hour. The average results were recorded for each sample. The background was counted and recorded as 40.00 ± 7.00 Bq/m^3. The background was then subtracted from each sample to give the final results reported in this study.

3.5.4. Gamma spectroscopy

Both homogenous soil and plant samples were measured for 3600 seconds each using CANBERRA HPGE high purity semiconductor gamma detector. (Figure 9.) indicates the geometry parameters used for the measurements. The instrument was calibrated for energy, full width at half maximum (FWHM), and efficiency (η) (geometry calibration and efficiency calibration function and calibration coefficients) before analysis to ensure the reliability of the instrument, that it can be trusted. Energy calibration was conducted using the following sources Co-60, Ba-133, Cs-137, Na-22, and K-40. The same sources were used for specifying the efficiency calibration procedure, and the calibration curve is shown in (Figure 10.). Geometry calibration was completed by simulation using CANBERRA Geometry Composer v.4.2 software.

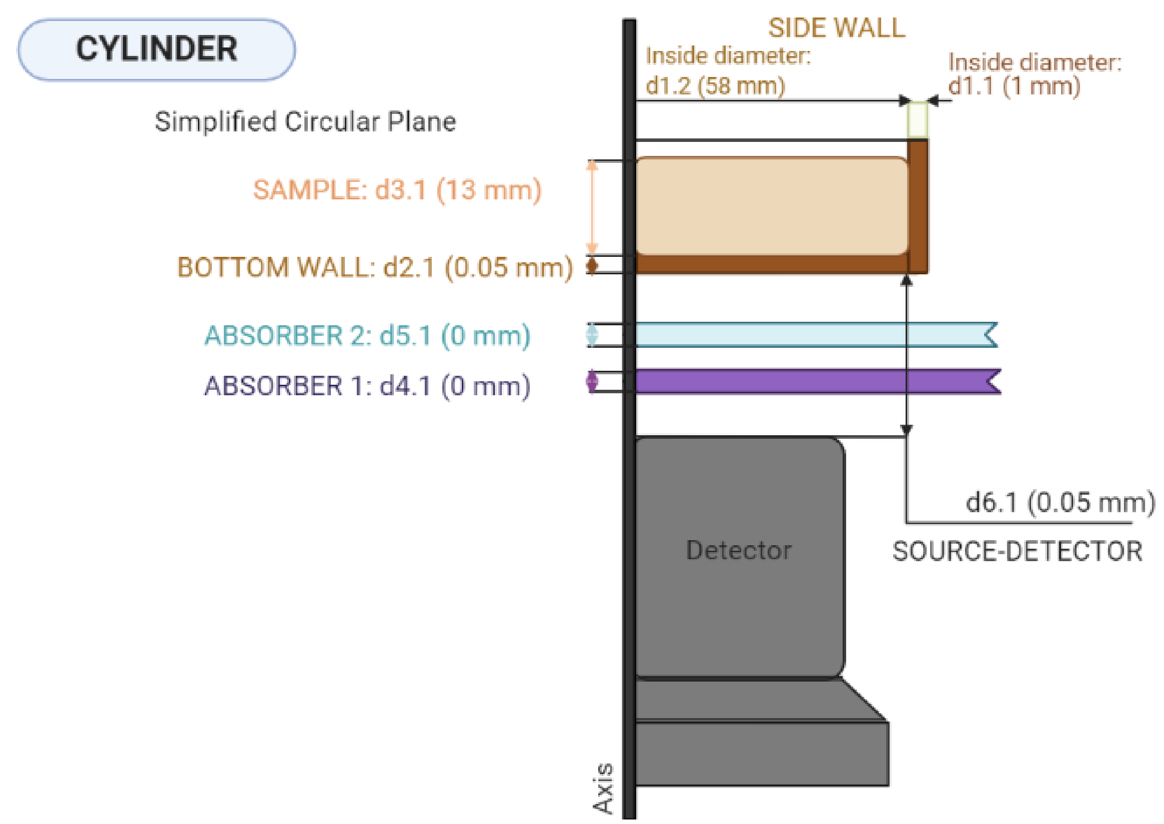


Figure 9. Geometry model for gamma measurements

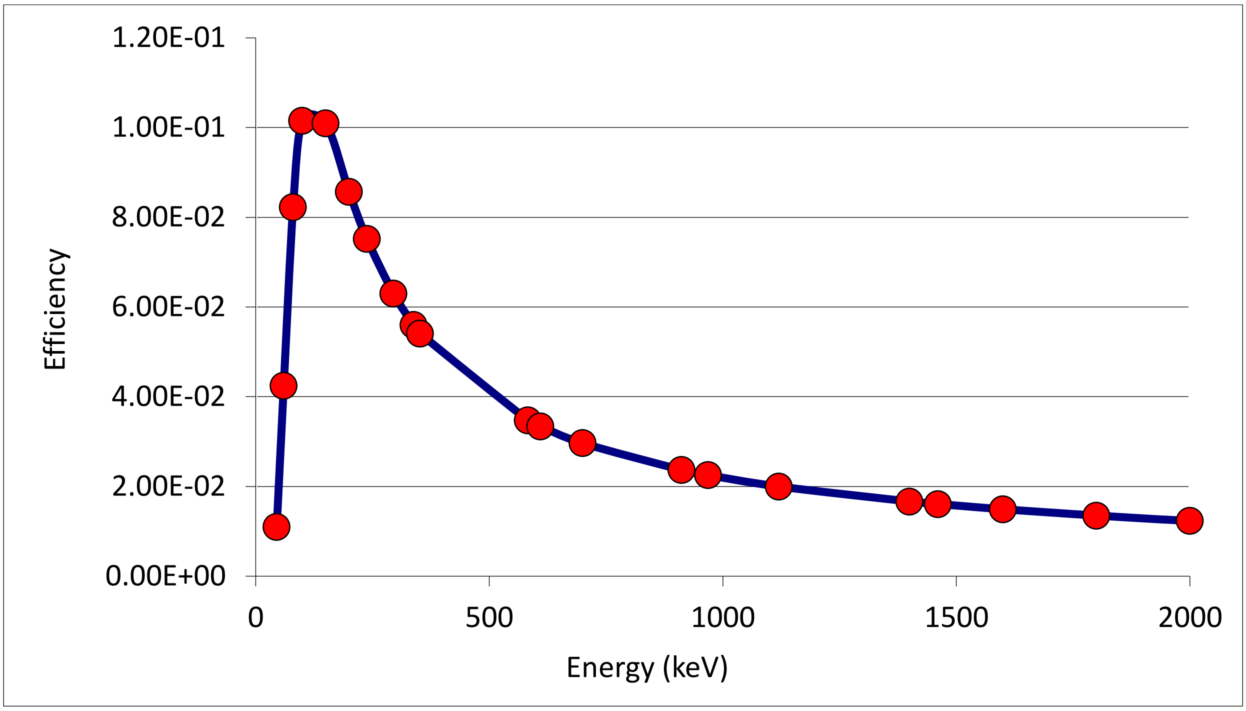


Figure 10. Efficiency calibration curve

The background was measured for the duration of 108000 seconds. The indoor background spectrum within the collimator is assumed to be constant. The background was quite low due to using a proper collimator. The background spectrum was normalized and then subtracted from the live time and energy-calibrated observed spectra, obtaining background compensated spectra channel as shown in Equation (3):

$$I_c = I - I_0 \qquad (3)$$

Where: I_0 - background intensity, I – intensity, and I_c - background compensated intensity.

Compensated spectra were processed, and the nuclide activities were determined from the predefined nuclide peaks using the Total Peak Area method described in the ORTEC GammaVision 6 (5.2.1.1.). The method was implemented in a Visual BASIC code running under an EXCEL environment. The Minimum Detectable Activity (MDA) is a measure of how small activity could be present and not be detected by the analysis. The MDA calculation was completed by ORTEC Critical Level (GammaVision 6, 5.7.2.).

Sample analysis

The soil and plant samples were kept in storage for the duration of six weeks to attain the radioactive equilibrium between Ra-226 and the decay products with short half-lives (Júnior *et al.*, 2006). All samples have had enough time for establishing secular equilibrium between decay series products, so determining the activity of parent isotopes was possible through measuring daughter isotopes having significant gamma lines. The reference isotopes were used for quantitative analysis: actinium-228 (Ac-228) and Pb-212 for Th-232 decay chain and Pb-214 and bismuth-214 (Bi-214) for U-238 (Ra-226) decay series. Measured radionuclides were compared to identify the radionuclides that had the highest activity and to identify the trends of all the radionuclides as the depths of soil samples change. The activity concentrations for each soil sample were compared with the plant sample collected at the same sampling point to identify if there is any biological uptake of radionuclides. The parameters for gamma measurements were as follows: Outer diameter – 60 mm, Side wall – 1 mm, Bottom wall – 0.5 mm, sample density - ~1.7 g/cm^3 (sand/soil approximated), and Sealed: foil – ~0.3 – 0.4 mm, hermetic sealing.

Generally, environmental radioactivity that is specifically predominant in the soil is due to the decay of the primordial radionuclides of U-238, U-235, Th-232, rubidium (Rb-87), and K-40. However, the highest contribution to this radioactivity is due to the great number of decay products of U-238 and Th-232 (Júnior *et al.*, 2006). These primordial radionuclides were measured, and the activity concentration for each was determined per depth of the soil sample and plant samples.

3.6. Pseudo total element analysis

3.6.1. Plant sample preparation

Approximately 0.5 g of homogenized plant samples were weighed into the Teflon vessel using an analytical balance. 5 mL of nitric acid (65% Merck), 6 mL of hydrogen peroxide (30% Emsure®), and approximately 9 mL of distilled water were added into the Teflon vessel containing the samples to make it up to 20 mL. The reaction could subside completely before capping the vessel.

3.6.2. Soil sample preparation

All soil samples were air-dried for five days. After five days, the samples were physically homogenized and grinded using Retch SM 100 with a 0.25 mm sieve instrument. Approximately 0.5 g of homogenized soil samples were weighed into the Teflon vessel using an analytical balance. 5 mL of nitric acid (65% Merck), 3 mL of hydrogen peroxide (30% Emsure®), and approximately 2 mL of distilled water were added into the Teflon vessel containing the samples to make it up to 10 mL. The reaction could subside completely before capping the vessel.

3.6.3. Microwave digestion

The microwave digestion method was conducted according to the Hungarian Standard MSZ 21470-50:2006 (2006). The microwave digestion vessels containing samples were assembled and placed in a CEM Mars 5 Xtraction 230/60 Microwave Accelerated Reaction System 907501. The microwave digestion system was set at the following parameters: Ramp time – 20 minutes (450 Psi or 180 °C), Hold time – 18 minutes, Still time – 5 minutes (0 Watt), Hold time – 10 minutes (800-Watt, 450 Psi and/or 180 °C) and Cooling time – 20 minutes. (Figure 11.) indicates the changes in parameters as the microwave operational time changes during the digestion.

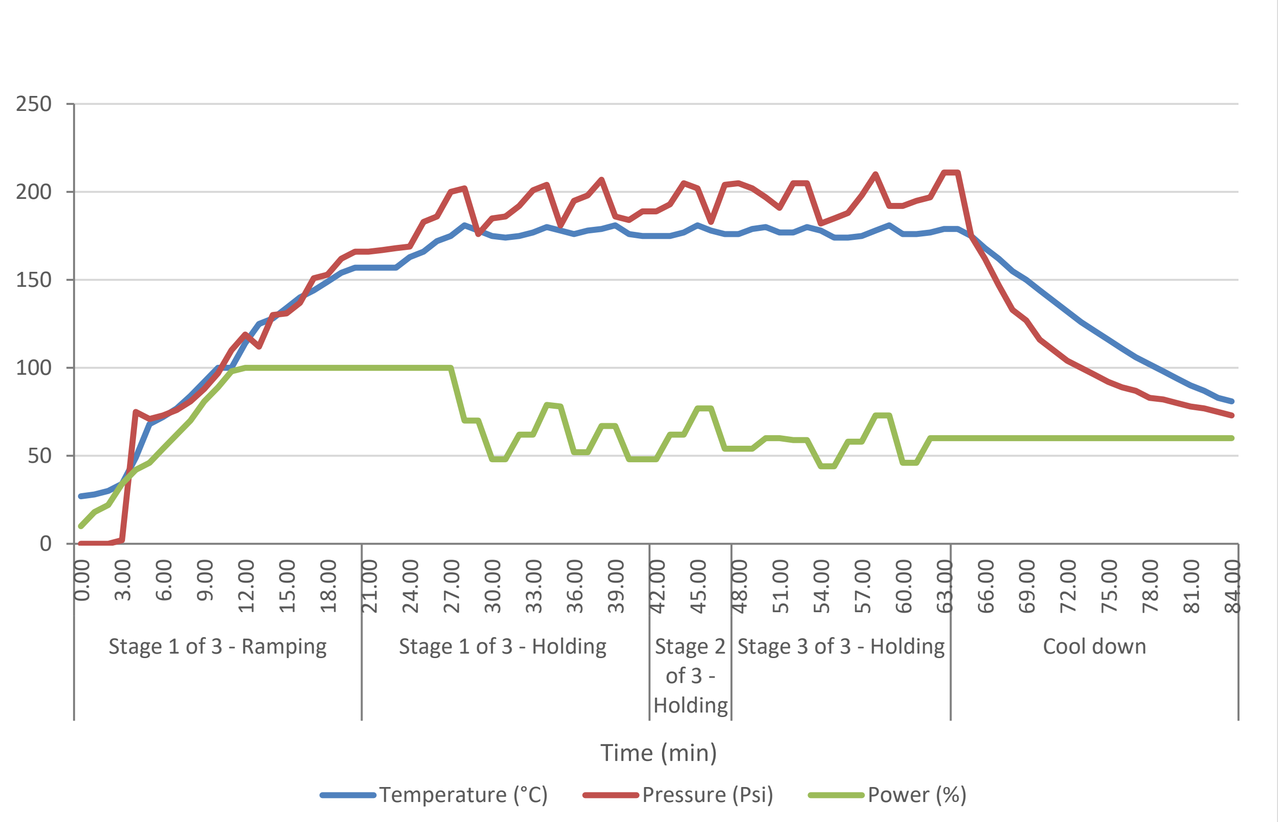


Figure 11. Microwave parameters

After digestion, each sample was filtered using a 90 mm Filter Discs (Quant.) Grade: 389 into a 25 mL volumetric flask and made up to 25 mL with distilled water. Each sample was physically homogenized by shaking it and transferred into a centrifuge tube for the trace element analysis.

3.6.4. Water sample preparation

Water samples were prepared using the Hungarian Standard MSZ 1484-3:2006 (2006). Water samples were filtered into a 40 mL centrifuge tube using Munktell filter discs grade 389. The samples were measured first before adding anything to determine if they will need dilution. The

1000 mg/L yttrium standard solution $(YNO_3)_3$ in HNO_3 0.5 mol/L was added into the samples as an internal standard. 4.90 mL of each sample and 0.1 mL of yttrium standard solution were added into a 10 mL volumetric flask, and the sample solution was made up to 10 mL with 4.90 mL distilled water.

Four blank solutions were prepared: 0 mL yttrium solution and 10 mL distilled water, 1 mL yttrium solution and 9 mL distilled water, 5 mL yttrium solution and 5 mL distilled water, and 10 mL yttrium solution and 0 mL distilled water.

Three external standards were used:

- Uranium ICP standard $(UO_2NO_3)_2$ in HNO_3 2-3 % 10 mg/L – U,
- Yttrium standard solution $(YNO_3)_3$ in HNO_3 0.5 mol/l – 1000 mg/L Y, and
- Certified Elements Standard – Uranium
  Concentration: 1000 ± 3 µg/mL, 20°C
  Matrix: 2.5 % HNO_3
  Density: 1.0152 g/mL, 20°C

Samples were measured using an Inductively Coupled Plasma – Optical Emission Spectrometer (ICP-OES) to determine the uranium element in water.

3.6.5. Instrumental analysis

The total elemental content of digested solutions was determined by a HORIBA Jobin Yvon ACTIVA M Inductively Coupled Plasma – Optical Emission Spectrometer (ICP-OES) using operation parameters proposed by the manufacturer and yttrium internal standard. (Table 2.) indicates the ICP-OES operational parameters used to determine the following elements (with ICP-OES emission lines): Ca (315.887 nm), Cd (228.802 nm), Co (228.615 nm), Cr (205.571 nm), Cu (324.754 nm), Fe (240.489 nm), K (766.490 nm), Mg (279.078 nm), Mn (257.610 nm), Na (589.592 nm), Ni (231.604 nm), P (253.560 nm), Pb (220.353 nm), Zn (213.857 nm), Ba (455.403 nm), and U (398.957 nm).

Table 2. ICP-OES operational parameters for trace elemental analysis

Operational parameters	**Settings**
Incident RF-power	1200 W
Outer gas flow (Argon)	16 L/min
Sheath gas flow (Argon)	0.3 L/min

Auxilary gas flow (Argon)	0.6 L/min
Nebulizer gas pressure (Argon)	2.86 bar
Nebulizer solution uptake (Meinhardt-cyclonic spray chamber)	0.85 mL/min

3.7. The soil/plant transfer factor determination

The soil/plant TFs for radionuclides and PTEs were calculated using Equation (2) as indicated in chapter 2.4.3 to determine the soil's effects for each element on the plants, consequently predicting the plant uptake. The TF was calculated only from the total concentrations obtained from the 0 – 25 depth of soil sample because the grass does not have deep roots to reach a deeper level of the soil. The higher the TF value, the more mobile/available the element is (Intawongse et al., 2007; Laţo *et al.*, 2012).

3.8. BCR sequential extraction procedure

Three independent replicates were performed for homogenous soil samples collected at 0 – 25 cm depths only. Blanks were measured in parallel for each set of analyses using the three-step BCR sequential extraction procedure with an additional (+1) *aqua regia* step. The Certified Reference Material BCR-701 was prepared in triplicate in parallel with the samples to ensure the accuracy of the analysis. The BCR three-step sequential extraction scheme was proposed by the European Community Bureau of Reference in 1992 (the Standards Measurement and Testing Program) (Zhu *et al.*, 2015). The extractable contents of Cd, Cr, Cu, Ni, Pb, and Zn were determined using the procedure below (also refer to chapter 2.6.4) as described in detail by (Rauret *et al.*, 1999; Rauret *et al.*, 2001; Heltai *et al.*, 2019).

Blanks

i. *Vessel blank*

   To one vessel from each batch, taken through the cleaning procedure, 40 mL of solution A was added. This blank solution was analyzed along with the sample solutions from step 1 (described below).

ii. *Reagent blank*

   A sample of each batch of solutions A (acetic acid, 0.11 mol/L), B (hydroxylammonium chloride, 0.5 mol/L), C (hydrogen peroxide, 300 mg/g, 8.8 mol/L) and D (ammonium acetate, 1.0 mol/L) was analysed.

iii. *Procedural blank*

With each batch of extractions, a blank sample (i.e., a vessel without a soil sample) was carried through the complete procedure and analyzed at the end of each extraction step.

Step 1

40 mL of solution A was added to 1 g soil in an 80 to 100 mL centrifuge tube, then extracted by shaking for 16 h at 22 ± 5 °C (overnight) and at a speed of 30 ± 10 rpm. The delay did not occur between the addition of the extractant solution and the beginning of the shaking. The extract was separated from the solid residue by centrifugation at 3000 g for 20 minutes and decanted the supernatant liquid into a polyethylene container. The container was closed, and the extract was stored in a refrigerator at about 4 °C prior to analysis. The residue was washed by adding 20 mL of distilled water, shook for 15 minutes on the end-over-end shaker, and centrifuged for 20 minutes at 3000 g. The supernatant was decanted and discarded while taking care not to discard any of the solid residues.

Step 2

40 mL of freshly prepared solution B was added to the residue from step 1 in the centrifuge tube and resuspended by manual shaking. The extraction was conducted by mechanical shaking for 16 hours at 22 ± 5 °C (overnight) and at a speed of 30 ± 10 rpm. No delay occurred between the addition of the extractant solution and the beginning of the shaking. The extract was separated from the solid residue by centrifugation and decantation as in step 1. The extract was retained in a polyethylene container, as before, for analysis. The residue was washed by adding 20 mL of distilled water, shaken for 15 minutes on the end-over-end shaker, and centrifuged for 20 minutes at 3000 g. The supernatant was decanted and discarded while taking care not to discard any of the solid residues.

Step 3

10 mL of solution C was added carefully to the residue in the centrifuge tube in small aliquots to avoid losses due to possible violent reaction. The vessel was covered loosely with its cap and digested at room temperature for 1 hour with occasional manual shaking. The digestion continued for 1 hour at 85 ± 2 °C in a water bath, and then the volume was reduced to less than 3 mL by further heating of the uncovered tube. A further aliquot of 10 mL of solution C was added. The covered vessel was heated again to 85 ± 2 °C and digested for 1 hour. The cover was removed, and the volume of liquid was reduced to about 1 mL. Care was taken not to bring the residue to complete dryness. 50 mL of solution D was added to the cool, moist residue and shaken for 16

hours at 22 ± 5 °C (overnight) and at a speed of 30 ± 10 rpm. No delay occurred between the addition of the extractant solution and the beginning of the shaking. The extract was separated from the solid residue by centrifugation and decantation as in step 1 and retained as before for analysis.

+ 1 Step (Aqua regia)

Approximately 3 g of the residue from step 3 of the BCR sequential extraction of the air-dried material was weighed into the reaction vessel. 0.5 mL to 1.0 mL of water was added to obtain a slurry, and while mixing, 21 mL of 12.0 mol/L HCL was added, followed by 7 mL of 15.8 mol/L HNO_3, added drop by drop to reduce foaming. 15 mL of 0.5 mol/L HNO_3 was added to the adsorption vessel, and the vessel was connected to the reflux condenser, and both were placed on the top of the reaction flask. The solution could stand for 16 h at room temperature to allow for slow oxidation of the organic matter of the soil. The temperature of the reaction mixture was raised slowly until reflux conditions were reached and maintained for 2 hours, ensuring that the condensation zone was lower than 1/3 of the height of the condenser. The reaction mixture could cool slowly to room temperature.

After cooling down, the content of the absorption vessel was added through the condenser tube into the reaction vessel, and both were rinsed with 10 mL of 0.5 mol/L NHO_3. The extract was filtered using a cellulose-based membrane filter with a medium pore size of 8 µm to remove particulates (silicates and other insoluble materials) while collecting the filtrate in a 100 mL graduated flask. All the initial filtrate passed through the filter paper then the insoluble residue was washed onto the filter paper with a minimum of 0.5 mol/L HNO_3. The graduated flask was filled up to the mark with 0.5 mol/L HNO_3 and homogenized by shaking. The trace elements could be determined by an appropriate ICP-OES method.

3.9. Quality control

The quality control for this study was conducted using the Certified Reference Material (CRM) BCR$^{®}$-701. This CRM is certified for only six elements (Cd, Cr, Cu, Ni, Pb, and Zn). The percentage recovery for each element was calculated.

In order to validate the BCR sequential procedure, the sums of the cumulated concentrations for each element in Step 1 (F1) + Step 2 (F2) + Step 3 (F3) + Aqua regia step (F4) were compared to the pseudo total concentrations obtained from the same samples (0 – 25 cm depth) and given in mg/kg units.

3.10. Statistical analysis

The statistical analysis was conducted using SPSS Statistics 28 for basic descriptive statistical analysis, a one-sample t-test to determine the differences between the means of one variable and the linear regression analysis for the determination of the soils' effect on the plants.

The descriptive statistical analysis was conducted to determine whether the data distribution would be normal or non-normal for physicochemical characteristics (variables) in soil samples. The skewness and kurtosis have to be within the accepted threshold of +/- 2 to suggest normal distribution (Alshahri, 2019). For the data that is normally distributed, the Pearson correlation matrix was selected to determine the mutual relationship to identify the degree of association between pairs variables and describe the interdependency.

One-sample t-test analysis was used to determine whether the specific value of each element is different from the unknown population mean at a 95 % confidence interval. The linear regression analysis was used to determine whether the soil has an effect on the plant, consequently determining whether the plant uptake of each element is due to the soil concentration of the same element. To facilitate the comparison of the results, the corresponding values of the soil and plant analyses are presented in regression graphs in order to combine the results of the two techniques. The higher the correlation coefficient (r) indicates, the better the link between the soil and plant analyses (FAO, 1982).

The principal component analysis (PCA) biplot was utilized using Analyse-it for Microsoft excel 2010 version 5.80.2 software to examine the relationships among the radionuclides, PTEs, and soil characteristics in a multidimensional dataset. The Analyse-it software was also used to plot the correlation figures and linear regression figures. To determine the common characteristics between PTEs, radionuclides, and physicochemical properties in the soil and plant samples, Pearson's correlation coefficient (r) was determined. The regression statistics were also conducted to verify the significance of possible differences between PTE and radionuclide concentrations and the soil characteristics variables in soil samples at a 95 % confidence level ($p \leq 0.05$). When the r-value is close to 1, it is considered that the two variables have a relationship and $p \leq 0.05$ state that there is sufficient evidence to conclude that the correlation coefficient is significantly different from zero and therefore reject the null hypothesis (Althouse and Soman, 2017; Statstutor, 2015).

4. RESULTS AND DISCUSSION

The characteristics that may affect the mobility of elements in the soil are discussed. The radionuclide, PTE total concentrations in soil and plant samples, and uranium in water samples are displayed and compared to the Hungarian regulatory limits, WHO, and UNSCEAR recommended limits. The transfer factors were calculated to evaluate the plant uptake. Additionally, the mobility of radionuclides and PTEs in soil was evaluated using the BCR sequential extraction procedure, and the results are discussed in this chapter. Finally, the correlations and the relationship between the PTEs, the radionuclides, and the chemical properties of the soil were determined through statistical analysis.

4.1. Characteristics that may affect the mobility of elements

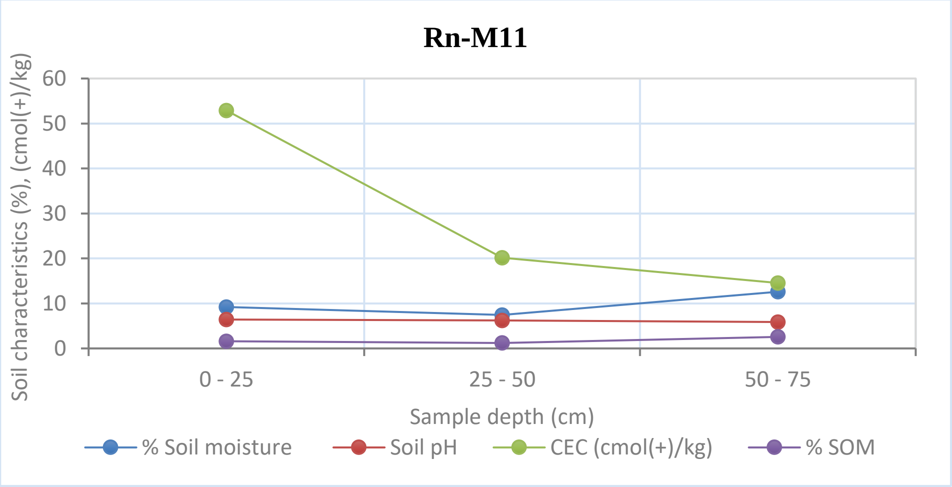
4.1.1. Meteorological conditions

The meteorological data were retrieved from the data published on the Website (https1://www.ksh.hu/docs/eng/xstadat/xstadat_annual/i_met002cc.html) of the Hungarian Central Statistical Office. During the remediation period, from the time when the mine closed to a sampling of soil (1997 – 2018), the average precipitation was 674 mm with a minimum of 405 mm in 2011 and a maximum of 981 mm in 2014. During the sampling month of July 2018, the precipitation intensity was 58 mm over thirteen precipitation days; the average temperature was 22. 1 ℃ with a minimum of 10.1 ℃ and a maximum of 31.3 ℃, and the days with wind greater than or equal to 10 m/s were eleven as indicated on the Website (https2://www.ksh.hu/docs/eng/xstadat/xstadat_infra/e_met006.html) (Hungarian Central Statistical Office, n.d.). It should be noted that in 2018, the highest precipitation intensity (134 mm) was observed in June, while sampling took place in July.

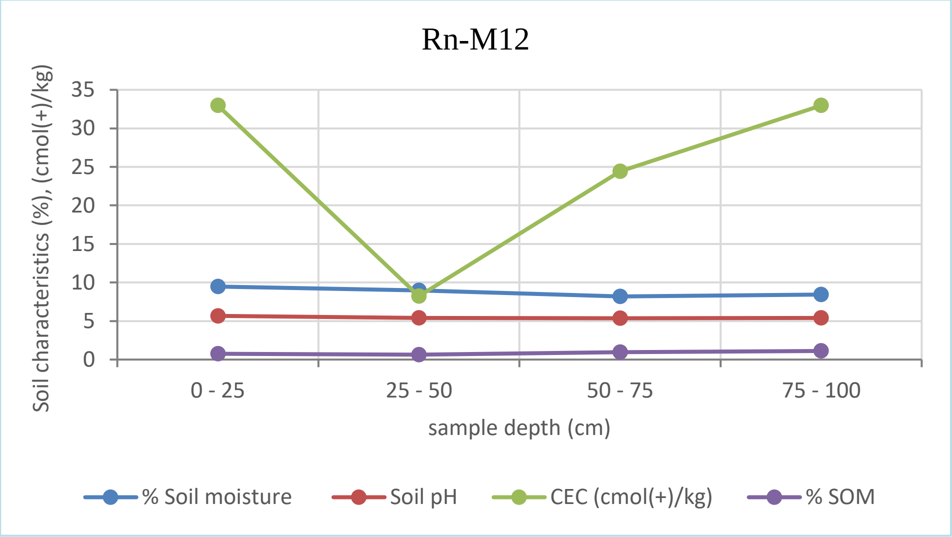
4.1.2. Physicochemical characteristics

The physicochemical characteristics were measured to determine their content in soil and plants. These parameters were eventually evaluated for their possible effects on plant uptake of elements and radionuclides from the soil. The raw data for the CEC and SOM is presented in (Appendix A2. and A3.) respectively. The behavior of the physicochemical properties in the function of depth is indicated in (Figure 12. a, b, c, and d) and in the function of topography is indicated in (Figure 12. e). It should be noted that the Radioactive sample was sampled from 0 – 25 cm depth only due to the hard rock beneath the surface, therefore (Figure 12. d) was plotted using the results for the soil characteristics measured in 0 – 25 cm depth. Additionally, the soil moisture content (% SM), the average soil organic matter (% SOM), pH, CEC, and plant moisture content are indicated in (Table 3.).

a)



b)



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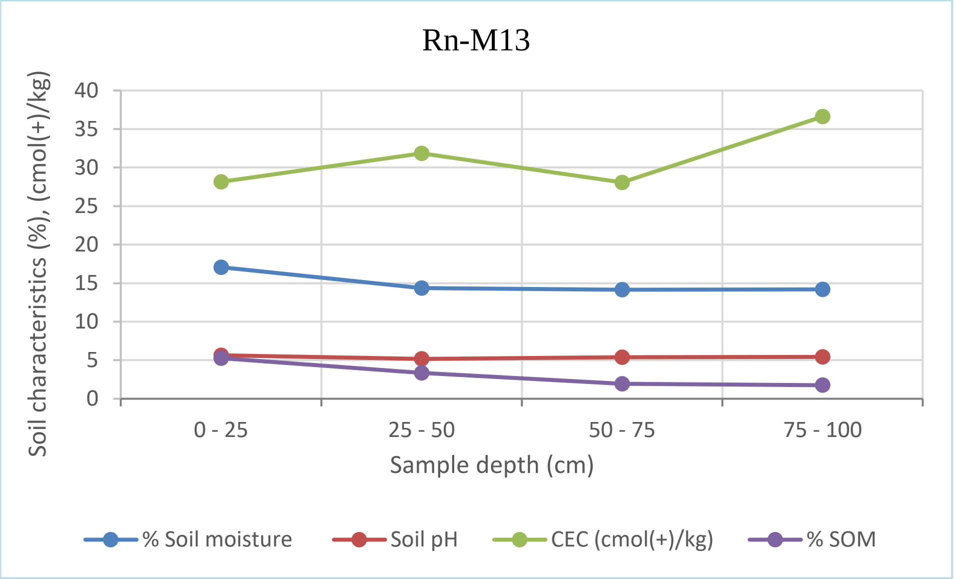

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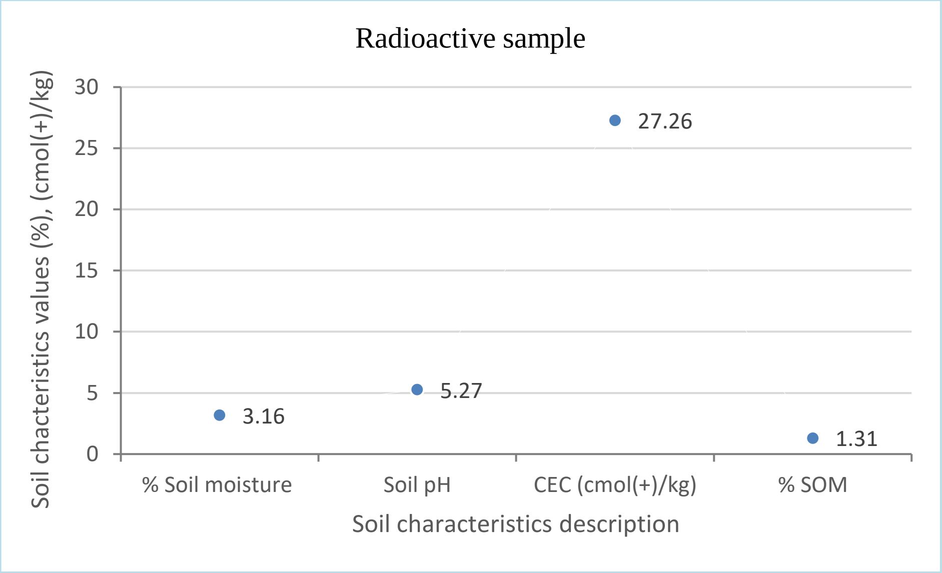


e)

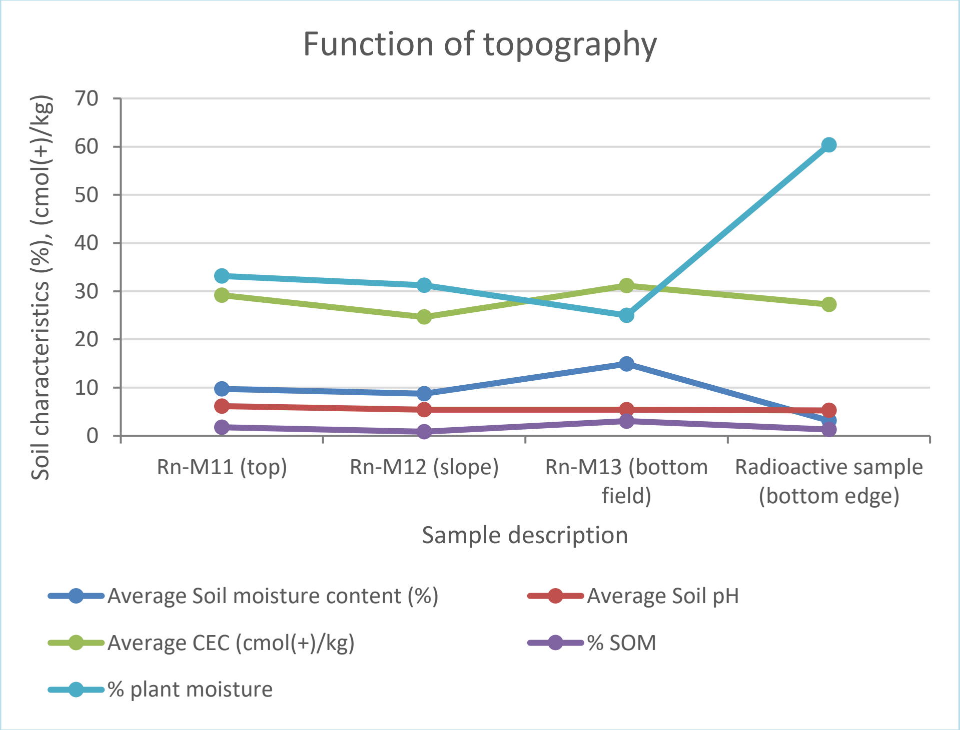


Figure 12. Comparison for the physicochemical characteristics in terms of depth in a) Rn-M11; b) Rn-M12; c) Rn-M13; d) Radioactive sample; and e) The averages of all the samples in terms of location

Table 3. Properties that may affect the mobility of elements

Sample description	**Depth (cm)**	**Soil moisture content (%)**	**Average Soil pH**	**CEC (cmol(+)/kg)**	**SOM (%)**	**Plant moisture content (%)**

Rn-M11	0 – 25	9.24	6.22	52.9 ± 6.71	1.61	33.1
	25 – 50	7.44	6.12	20.2 ± 0.06	1.21	
	50 – 75	12.6	5.99	14.6 ± 4.55	2.56	
	75 – 100	Hard rock (not sampled)				
Rn-M12	0 – 25	9.47	5.79	33.0 ± 0.69	0.75	31.2
	25 – 50	8.99	5.50	8.23 ± 1.09	0.64	
	50 – 75	8.19	5.47	24.4 ± 1.31	0.96	
	75 – 100	8.43	5.38	33.0 ± 0.01	1.11	
Rn-M13	0 – 25	17.1	5.61	28.2 ± 0.44	5.27	25.0
	25 – 50	14.3	5.27	31.8 ± 4.61	3.34	
	50 – 75	14.2	5.38	28.1 ± 2.67	1.92	
	75 – 100	14.2	5.36	36.6 ± 0.01	1.76	
Radioactive sample	0 - 25	3.16	5.27	27.3 ± 0.01	1.31	60.3

The soil moisture ranged from 3.16 % to 17.1 %, with the radioactive sample having the lowest soil moisture content. This might be due to the degraded quality of the soil, which exhibited poor ability for plant growth, as indicated in (Figure 13.). Rn-M13 sample exhibited the highest soil moisture and soil organic matter content with an average of 15.0 % and 3.07 %, respectively. This sample was taken from the area that is completely covered with grass, as shown in (Figure 7.) which might have influenced the high percentages of moisture and organic matter in the soil. According to Thlaspi *et al.* (2003), plants accumulate large amounts of metals from the soil when their moisture content is high. A similar scenario could be expected in the case of organic matter since these two variables correlate with each other ($p \leq 0.05$). Additionally, it should be noted that the samples were collected a month after the high precipitations (134 mm) were recorded in June 2018. It was observed that the soil moisture content had an opposite behavior to the plant moisture content for each sample, as shown in (Figure 12. e) ($r = -0.923$; $p \leq 0.05$). The sample location with the lowest soil moisture content resulted in the highest plant moisture content. The descriptive statistics results for all the soil characteristics considered for this study are presented in (Appendix A13.).

According to Dragović *et al.* (2015), the soil moisture content is one of the factors that cause the variations of background concentrations of radionuclides in soil. Furthermore, low moisture content would lower hydrophilic radionuclides since solubility would decrease (Ahmad *et al.*, 2019). Similarly, Thlaspi *et al.* (2003) conducted a study on soil moisture effects on uptake of metals by biota, where the authors concluded that the study conducted did not find any correlation between low soil moisture and enhanced uptake of metals. As a matter of fact, exactly the reverse was observed where enhanced metal absorption at higher soil moisture values was commonly observed (Thlaspi *et al.*, 2003).

Figure 13. Radioactive sampling point

The pH values indicated a narrow range of 5.27 to 6.22. These pH values suggested that the soil was acidic. The observations indicated that the trend of the pH values decreased with the decreasing slope of the spoil deposit. The water hardness is responsible for the pH changes. According to Wilson, (2010), alkalinity is important because it buffers the pH of water within the system and without this buffering capacity, small additions of acids or bases would result in significant changes of pH. The hardness of water in the mining area is due to the dissolution of minerals in soils such as the limestone ($CaCO_3$), and can also be associated with pollution by industrial effluents (Ferrari *et al.*, 2017). When the lime dessolution occurs, the Ca decreases and the dissolved CO_2 increases which results to the pH decrease, consequently the CEC also decreases. This explanation is supported by the PCA statistical analysis in (Figure 23.). The sample collected from the top of the deposit (Rn-M11) had the highest average pH (6.11) and the sample collected from the bottom edge of the slope (Radioactive sample) had the lowest pH (5.27). Chakraborty *et al.* (2013) reported that soil-to-plant TF depends on soil properties such as pH and moisture content.

The CEC values ranged from 8.25 ± 1.09 (cmol(+)/kg) to 52.89 ± 6.71 (cmol(+)/kg). The CEC values did not follow any trend. It was observed that Rn-M12 at 25 – 50 cm depth had the lowest

CEC of 8.25 ± 1.09 (cmol(+)/kg), which is an indication that the soil at this sampling location is more susceptible to cation nutrient loss through leaching.

The SOM ranged between 0.87 % and 3.87 %. It could be observed that the sample collected from the slope of the spoil deposit (Rn-M12) had the lowest average % SOM. Although this sample had a higher moisture content than the Radioactive sample, its water-holding capabilities might be low. This could be due to the water runoffs down the slope instead of infiltrating through the soil in this location. This has been proven by the sample collected at the bottom of the deposit (Rn-M13) with the highest soil moisture and soil organic matter contents.

One-Sample t-test statistical analysis indicated that the CEC values were significantly different ($p > 0.05$) in all three depths (0 – 25 cm, 25 – 50, and 50 – 75 cm) and three samples (Rn-M11, Rn-M12, and Rn-M13) that were considered for the function of topography. In contrast, the SOM was only significantly different in 25 – 50 cm depth. In terms of depth, the CEC and the SOM were found to be significantly different in Rn-M11. The statistical results for the soil characteristics are discused in detail in section 4.4.1.

4.2. Radioanalytical results

There have not been known limits placed for natural radionuclides (K-40, U-238, U-235, and Th-232) in the soil since K-40, Th-232, and U-238 are principal primordial radionuclides that have existed since the beginning of Earth. However, UNSCEAR (2000) placed international median values for each radionuclide in soil and the mean values for each country based on previously reported activity concentrations in the world. Therefore, the activity concentration obtained in this study were compared to the mean concentrations for Hungary. The UNSCEAR did not report on the mean values for U-235; this might be due to that the natural uranium contains 0.7% of the U-235 isotope, and the remaining 99.3% is mostly the U-238 isotope (World Nuclear Association, 2020). For Rn-222, it is the obligation for each country to place a limit not to be exceeded following the regulations of the International Commission on Radiological Protection (ICRP), IAEA, and/or UNSCEAR based on the safety of the people. Hence, Hungary set the Rn-222 action level value of 1000 Bq/m^{3,} which is documented in the Hungarian Regulation 10 16/2000 (2000) (Shahrokhi *et al.*, 2017). Following the closure of the Mecsek uranium mine, the Republic of Hungary (2008) indicated the limit values (Table 4.) for the release and environmental load, which had to be complied with within the course of mine closure and remediation projects according to the environmental protection license and the specifications. The Rn-222 activity concentrations obtained in this study were compared to the Hungarian Regulation 10 16/2000 action level value, radiation protection requirement for radon reported by the Republic of Hungary, and the activity concentrations of natural radionuclides in soil samples were compared to the international values reported by UNSCEAR (2000).

Table 4. Radiation protection requirements (the Republic of Hungary, 2008)

For the remediation of waste rock piles, heap leaching piles, and tailings ponds		For the remediation of surface facilities and their immediate surroundings	
Radon		**Radon**	
Rn exhalation	0.74 Bq/m^2/s	Rn exhalation	0.74 Bq/m^2/s
Rn concentration	background + 30 Bq/m^3	Rn concentration	1000 Bq/m^3
Activity concentration of soil		**Activity concentration of soil**	
In the uppermost 0-15 cm layer	background + 180 Bq/kg	In the uppermost 0-15 cm layer	background + 180 Bq/kg
In the next 15 cm layer	background + 550 Bq/kg	In the next 15 cm layer	background + 550 Bq/kg

4.2.1. Radon activity concentrations

The summary of the results for Rn-222 activity concentration measurements in the soil samples is indicated in (Table 5.).

Table 5. Rn-222 activity concentration in soil

Sample name	**Sample depth (cm)**	**Activity (Bq/m^3)**
Rn-M11	0 – 25	5.08 ± 1.34
	25 – 50	25.6 ± 1.81
	50 – 75	< background
	75 – 100	Hard rock (not sampled)
Rn-M12	0 – 25	29.3 ± 1.35
	25 – 50	29.1 ± 1.79
	50 – 75	14.3 ± 1.95
	75-100	23.0 ± 0.77
Rn-M13	0 – 25	13.5 ± 2.33
	25 – 50	49.3 ± 2.27
	50 – 75	40.2 ± 2.16
	75 – 100	44.3 ± 2.16
Radioactive sample	0 – 25	80.3 ± 1.34

Activity concentrations of Rn-222 for each soil sample were investigated to determine the behavior of Rn-222 as the depth of the sample increases and to determine if the activity concentrations were within the set limit. The average Rn-222 activity concentrations for the soil depths of Rn-M11, Rn-M12, and Rn-M13 samples were 15.3 ± 1.58 Bq/m^3, 23.9 ± 1.47 Bq/m^3, and 36.8 ± 2.23 Bq/m^{3,} respectively. The Radioactive sample exhibited a higher activity concentration as compared to all the samples collected. This sampling point already indicated a higher gamma dose rate, as shown in (Figure 8.) from the previous monitoring report conducted by the Mecsekérc Zrt. (2017).

The activity concentrations for Rn-222 did not have a particular pattern regarding the vertical distribution of sample depth. Statistically, the sample depths and the Rn-222 concentrations indicated a weak negative correlation (r = - 0.100; p = > 0.05). The topographic details imply that the activity concentration for Rn-222 might decrease as the slope of the spoil deposit increase. This observation indicates that the sample collected from the top of the deposit had an average of 15.3 ± 1.58 Bq/m^3, while the sample collected from the bottom edge of the deposit had 80.3 ± 1.34 Bq/m^3. The One-Sample t-test statistical analysis results indicated that there was a significant difference ($p > 0.05$) between the sample locations (Topography). In contrast, the t-test indicated that only Rn-M11 had significant differences in terms of depths. The Rn-222 activity concentration for sampling point Rn-M11 (50 - 75 cm) was below the detection limit, as can be seen in (Table 5.). The reason for this might be the hard rock that was below 50 -75 cm layer. The statistical analysis showed that the Rn-222 concentrations were significantly different in the Rn-M11 sample. According to Elzain (2014), lower porosity of the soil and intact rocks above the aquifer reduces the probability of radon gas escape, while higher porosity might allow the radon gas to escape easily. Rn-M13 and Radioactive samples were observed to have an elevated Rn-222 activity concentration compared to Rn-M11 and Rn-M12. This increase in concentration might be due to the fact that the sampling point for Rn-M13 was at the bottom of the slope on the field surface, and the other two points are at the top of the slope. In comparison, the radioactive sample was at the edge of the deposit, which is at the bottom of the deposit slope. The Rn-222 activity concentrations, which ranged between 5.08 ± 1.34 Bq/m^3 and 80.3 ± 1.34 Bq/m^3, were found to be below the action level value of 1000 Bq/m^3 set by the Republic of Hungary (2008) for radiation protection and also stated in the Hungarian Regulation 10 16/2000 (2000) for workplaces like mines, tourists caves, and other underground activities. This action level is set in accordance with the ICRP, where the action level is suggested to be between 500 Bq/m^3 and 1500 Bq/m^3 (ICRP, 2012; Shahrokhi *et al.*, 2017).

4.2.2. Gamma spectroscopy

The soil activity concentrations for natural radionuclides measured were compared to the Hungarian mean value for the natural radionuclide content in the soil as specified by UNSCEAR (2000). The raw data for radionuclides are shown in (Appendix 4.). The activity concentrations for K-40 were presented separately from other radionuclides for better visuals, as shown in (Figure 14.).

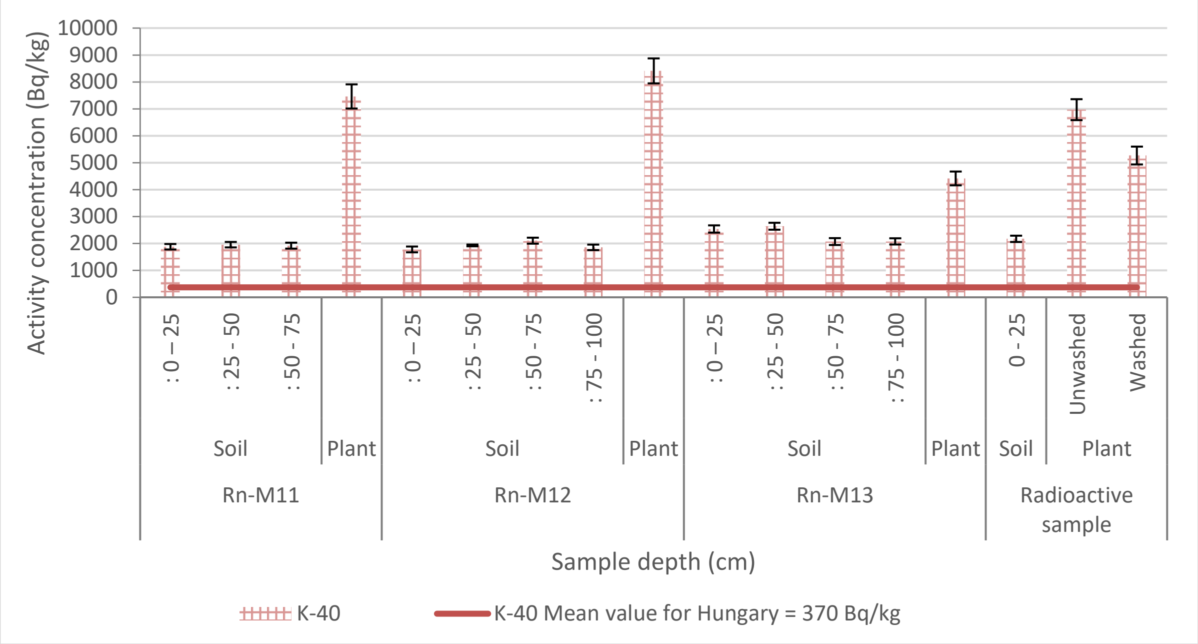


Figure 14. Activity concentrations for K-40 measured in soil and plant samples

The vertical analysis for K-40 results indicated that the soil depth did not have an impact on the concentration since similar concentrations could be obtained for each sample. The average activity concentrations for K-40 in soil samples Rn-M11 and Rn-M12 were 1918 ± 104 Bq/kg and 1917 ± 88.4 Bq/kg, respectively. In contrast, the average activity concentrations in Rn-M13 and Radioactive samples were 2332 ± 126 Bq/kg and 2173 Bq/kg, respectively. These results give an indication that the soil samples (Rn-M11 and Rn-M12) collected from the top of the deposit had lower concentrations of K-40 as compared to the soil samples (Rn-M13 and Radioactive sample) collected from the lower part of the deposit No. I. This might be due to some movement of K-40 radioactivity through water precipitation. The activity concentrations of K-40 in soil were all above the World’s median value of 400 Bq/kg for K-40, and the concentrations were outside the range of 140 – 850 Bq/kg as reported by the UNSCEAR (2000), the lowest activity concentration reported in this study was 1779 ± 108 Bq/kg. The activity concentrations for K-40 sequence was as follows: Rn-M13 > Radioactive > Rn-M11 > Rn-M12. Elevated concentration of K-40 could be the result of high clay content since K-40 is part of a clay minerals component, and its mobility is determined by the solubility in the soil (Ahmad *et al.*, 2019). One-Sample t-test statistical results indicated that the differences in K-40 concentrations were due to chance ($p \leq 0.05$) in terms of depths and in terms of topography.

It was notable that the activity concentration of K-40 in the unwashed plant samples was slightly higher than in the washed sample. This concludes that some of the activity concentration in plants is due to dust particles. The comparison of the results for soil samples with plant samples collected at the same location indicated that K-40 radioactivity concentrations of plant samples were more than double the soil activity concentrations. This implies that the plant samples might have taken up a large amount of K-40 from the soil. These results are in agreement with the study conducted by Manigandan (2009), in which it was discovered that the K-40 activity concentrations were considerably higher than other radionuclides, which suggested higher levels of uptake of this radionuclide.

The activity concentrations for U-238, Th-232, and U-235 radionuclides in the soil samples in each depth; and the concentrations for the plant samples collected at the same location are shown in (Figure 15.). The activity concentration for U-238 and Th-232 in soil were compared to the mean values for Hungary as reported by UNSCEAR (2000). The results for the Radioactive sample are presented separately in (Figure 16.) to show the progenies for U-238 behavior since U-238 was elevated in this location.

The activity concentration for U-238 in soil ranged from 66.6 ± 12.2 Bq/kg to 154.80 ± 12.8 Bq/kg. The average activity concentrations for Rn-M11, Rn-M12, and Rn-M13 were 103.50 ± 11.8 Bq/kg, 85.3 ± 9.54 Bq/kg, and 120.47 ± 13.5 Bq/kg, respectively. These activity concentrations of U-238 were all above the mean values for soils in Hungary (29 Bq/kg) and the median value for soils in the World (35 Bq/kg) as reported by UNSCEAR (2000). These results indicate that Rn-M13 had elevated U-238 activity concentration as compared to other samples. The source of elevated U-238 could be associated with geological material.

Observations for Th-232 indicated that Rn-M13 had the highest average activity concentration (58.86 ± 10.6 Bq/kg) in the soil as compared to Rn-M11 and Rn-M13, which had 42.52 ± 9.28 Bq/kg and 42.8 ± 42.83 Bq/kg, respectively. The Th-232 activity concentrations were above the mean value for soils in Hungary (28 Bq/kg) and the World's median value (30 Bq/kg) (UNSCEAR, 2000). Uranium-235 had the lowest activity concentrations as compared to all-natural radionuclides measured in this study. The U-235 concentrations ranged between 3.21 ± 0.16 Bq/kg and 7.22 ± 0.60 Bq/kg, with sample Rn-M13 displaying a higher average activity concentration (5.62 ± 0.63 Bq/kg) than the rest of the samples. In fact, it was notable that when comparing Rn-M11, Rn-M12, and Rn-M13, Rn-M13 displayed higher activity concentrations for all-natural radionuclides measured. This might be due to the fact that the Rn-M13 sample was collected at the bottom of the deposit. The higher activity concentration might suggest that the source of contamination may be near the sample collection area. The vertical distribution of activity concentrations for all the natural radionuclides in all depth levels did not follow any trend; the results varied throughout for all sample locations. Statistical results indicated that when it comes to the topography, the differences in U-238, U-235, and Th-232 concentrations were due to chance ($p \leq 0.05$). However, in terms of depths, there was a significant difference between the concentrations of U-238 and U-235 among the depths of Rn-M13.

Plant samples were observed to have elevated U-238 concentration (with an average of 170.62 ± 43.4 Bq/kg) compared to all the natural radionuclides measured (Th-232 = 37.19 ± 29.8 Bq/kg and U-235 = 7.96 ± 2.03 Bq/kg). This might be attributed to high concentrations of U-238 in soil. Overall, the activity concentrations for all the natural radionuclides detected in plant samples were above the average concentrations found in soil samples collected from the same location, with an exception for Th-232. Thorium-232 displayed an average concentration that was less than the average concentration of the soil. This could be the indication that there was a biological uptake of these radionuclides by plants from the soil. However, for Th-232, the uptake was less than the uptake of other natural radionuclides.

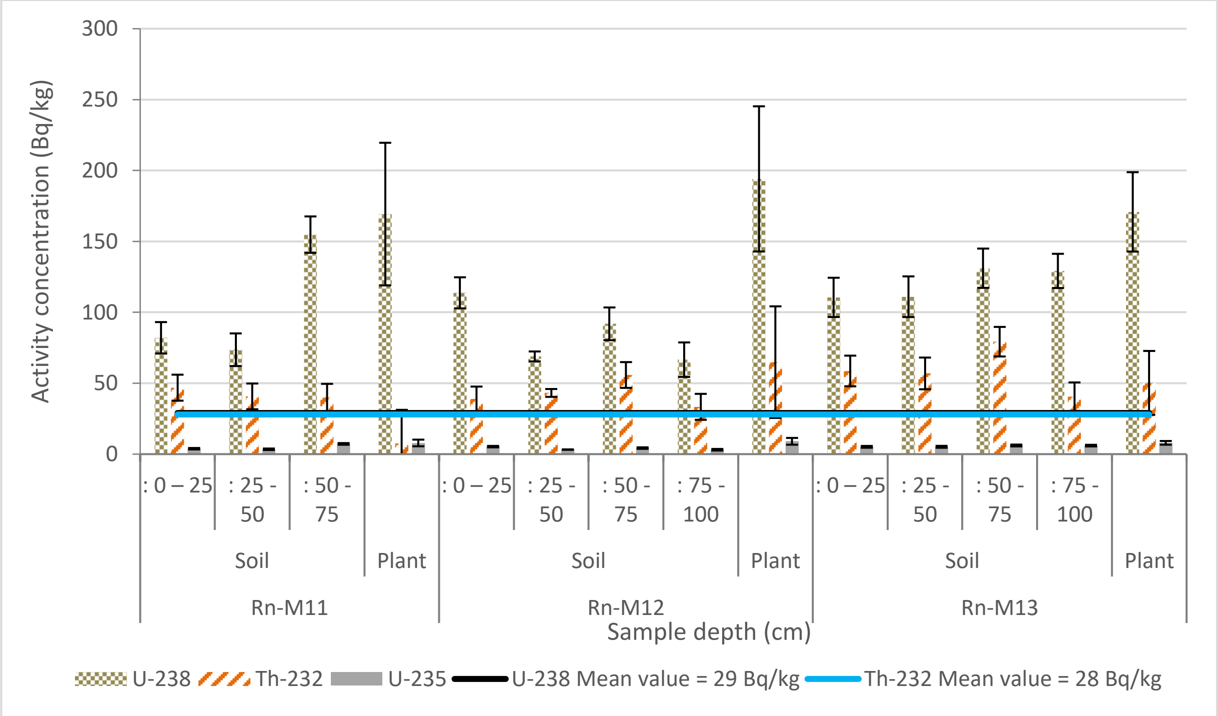


Figure 15. Activity concentrations for the natural radionuclides in soil and plant samples

The results for the activity concentrations for U-238 and Th-232 progenies in soil and plants for Rn-M11, Rn-M12, and Rn-M13 samples are presented in (Appendix A5.) and (Appendix A6). The vertical analysis did not show any trend; however, it could be noted that Ac-228 (969.1 keV) could not be detected in all plant samples, except for the Radioactive washed sample with 40.17 ± 110 Bq/kg. This could be the result of cross-contamination since the concentration was not detectable in the same sample which was unwashed. The observation made that plant samples contained higher activity concentrations of U-238 progenies (Pb-214 at 295.2 keV, Pb-214 at 352 keV, Bi-214 at 609.3 keV, and Bi-214 at 1120.3 keV) and Th-232 progeny (Ac-228 at 911.6 keV) as compared to the soil samples. Furthermore, Pb-212 (238.6 keV) concentration in the Radioactive soil sample was not detectable; however, in plants, both washed and unwashed samples resulted in 39.43 ± 22.6 Bq/kg and 37.12 ± 27.19 Bq/kg, respectively. This observation could be an indication that the plants absorb all the Pb-212 from the soil.

The results for natural radionuclides in (Figure 15.) and their decay products in (Appendix A4.) indicated that the depth of the soil did not have an impact on activity concentrations; there was a variation in activity concentrations. However, the topography of the deposit may have influenced the concentration results. The samples collected on the top of the deposit appeared to have lower concentrations compared to the samples collected at the edge or at the bottom of the deposit. This might be an indication that there is a migration of radionuclides from the top of the deposit to the bottom of the deposit. When comparing the soil samples to the plant samples taken from the same sampling point, there was an indication of higher activity concentrations in plants. This might be an indication of the biological uptake of the radionuclide by the plant samples.

The activity concentrations for the Radioactive soil sample washed and unwashed Radioactive plant samples were compared as illustrated separately to other samples as shown in (Figure 16.). While comparing the activity concentrations for the washed and unwashed plant samples, it was noted that the concentration difference was not significant. The difference in activity concentrations for both washed and unwashed plant samples was unnoticeable.

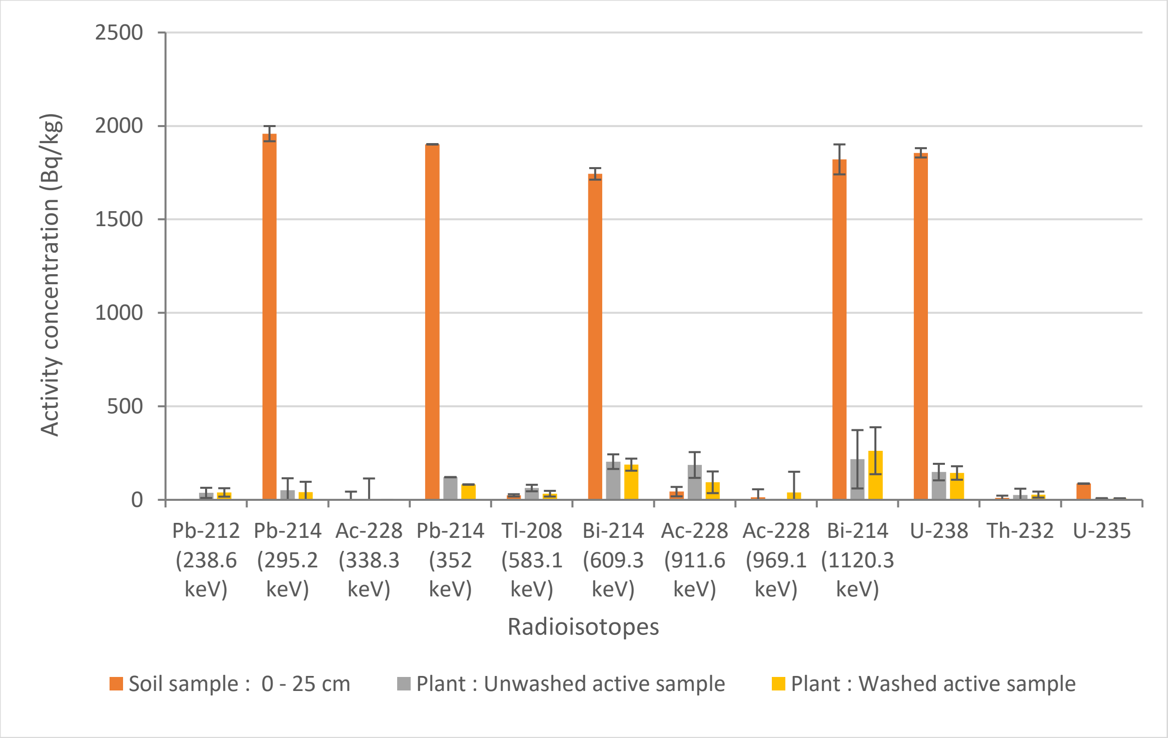


Figure 16. Radioactive sample – activity concentrations for radioactive soil and plant sample before and after washing

The results for the radioactive sample in (Figure 16.) indicated elevated activity concentrations of U-238 and its decay products. The activity concentration of U-238 in soil was observed to be elevated in the Radioactive sample (1856.29 ± 24.9 Bq/kg), whereas other samples had a concentration range of 85.30 ± 9.54 Bq/kg to 120.47 ± 13.5 Bq/kg. The observation was made that U-238 concentration levels in a soil sample were above the mean value specified by UNSCEAR. It was expected for the activity concentration of U-238 to be elevated in soil collected from the Radioactive point due to the high gamma dose rate (between150 - 200 nGy/h), as can be seen in (Figure 8.). The activity concentrations of U-238 and its decay isotopes in the Radioactive sample ranged from 1744± 30.9 Bq/kg and 1959 ± 40.6 Bq/kg. On the other hand, Th-232 in the Radioactive soil sample brought about the lowest activity concentration (10.04 ± 12.3 Bq/kg) as compared to all the samples. However, the plant samples indicated higher activity concentrations (washed: 28.07 ± 16.0 Bq/kg and unwashed: 26.16 ± 33.4 Bq/kg) than the soil samples. The Radioactive soil sample is the only sample that resulted in Th-232 being less than the mean value for soils in Hungary (28 Bq/kg) and the World's median value (30 Bq/kg) (UNSCEAR, 2000).

The sequence for U-238 concentration in soil was as follows: Radioactive > Rn-M13 > Rn-M11 > Rn-M12, the sequence for Th-232 activity concentrations was as follows: Rn-M13 > Rn-M12 > Rn-M11 > Radioactive, and the sequence for U-235 activity conentrations was as follows: Radioactive > Rn-M13 > Rn-M11 > Rn-M12.

The plant uptake of radionuclides in the soil is commonly known as soil to plant TF (Adesiji and Ademola, 2019). Transfer factor values were determined on a mass basis as indicated in (Table 6.); the activity concentration of plant samples (dry matter) was related to the radionuclide activity concentration in the first 25 cm of soil and calculated using Equation (2) as indicated in chapter 2.4.3. The TF values obtained in this study were compared to the mean values for radionuclide TF ratios (grasses) in a temperate environment for all types of soil reported by the International Atomic Energy Agency on their Technical Reports Series no. 472 (IAEA, 2011).

Table 6. Transfer Factors calculated from 0 – 25 cm soil samples

Radionuclides	**Rn-M11**	**Rn-M12**	**Rn-M13**	**Radioactive sample**
Pb-212 (238.6 keV)	n.d.	1.25 ± 4.53	n.d.	n.d.
Pb-214 (295.2 keV)	1.97 ± 4.86	3.27 ± 4.45	1.16 ± 1.97	0.03 ± 1.59
Ac-228 (338.3 keV)	n.d.	1.56 ± 4.68	2.35 ± 2.05	2.44 ± 2.62
Pb-214 (352 keV)	0.16 ± 4.40	2.57 ± 4.40	0.88 ± 1.99	0.06 ± 1.24
Tl-208 (583.1 keV)	1.34 ± 4.75	6.40 ± 4.26	0.22 ± 2.18	2.64 ± 2.81
Bi-214 (609.3 keV)	3.17 ± 4.42	1.72 ± 4.53	1.11 ± 2.03	0.12 ± 1.26
Ac-228 (911.6 keV)	7.95 ± 4.37	1.68 ± 4.19	0.68 ± 2.37	4.26 ± 2.73
Ac-228 (969.1 keV)	n.d.	n.d	n.d	n.d
Bi-214 (1120.3 keV)	2.30 ± 4.51	0.85 ± 4.74	2.44 ± 2.03	0.12 ± 1.95
K-40 (1460.8 keV)	3.97 ± 4.45	4.73 ± 4.31	1.74 ± 1.91	3.21 ± 3.32
U-238	2.06 ± 4.54	1.71 ± 4.67	1.55 ± 2.02	0.08 ± 1.78
Th-232	0.16 ± 2.59	1.67 ± 4.53	0.86 ± 2.08	2.60 ± 2.71
U-235	2.06 ± 4.54	1.71 ± 4.67	1.55 ± 2.02	0.08 ± 1.78
		TF values for grasses (IAEA, 2010)		
	Mean	Minimum	Maximum	
Pb	0.31	0.11	1.0	
K (in pasture grasses)	0.73	-	-	
U	0.017	0.00020	5.5	
Th	0.042	0.00074	0.65	

The n.d. abbreviation represents not detectable.

Rn-M11 and Rn-M12 indicated high TF ratios as compared to Rn-M13 and radioactive samples. Rn-M11 had the highest TF ratio of 7.95 ± 4.37 for Ac-228 (911.6 keV), followed by Tl-208 (583.1 keV) with a TF ratio of 6.40 ± 4.26 in sample Rn-M12. The Pb TF ratios for the radioactive sample were all below the minimum values reported by the IAEA. In comparison, the rest of the samples contained Pb radioisotopes that were above the maximum value. The TF ratios for U-238 and U-235 were the same for all samples, which were all above the mean values but were below the maximum value of 5.5 as reported by IAEA. It could be noted that the TF ratios for the U radioisotopes in the Radioactive sample were lower than the rest of the samples. This could be explained by the high concentrations of U radioisotopes in the Radioactive soil sample. This observation suggests that the plants in this sampling location could uptake fewer U radioisotopes as compared to the plants in other sampling locations. The TF ratios for Th-232 were less in Rn-M11 and Rn-M13. However, the Radioactive sample had an elevated TF value as compared to other natural radionuclides. According to Azeez *et al.* (2019), the mobility for Th-232 may be restricted in plants due to its adsorption by cell wall materials, and it is also highly insoluble in natural waters. The TF ratios for K-40 were observed to be higher than the TF ratios for all the natural radionuclides. These results are harmonious with the K-40 results from the study conducted by Azeez *et al.* (2019), where the K-40 TF ratios were found to be significantly higher in all plants as compared to other radionuclides and the authors are of the opinion that this might be due to the high mobility of K-40 in the soil and its solubility in water. However, there is no data in the IAEA (2000) report regarding the TF ratios for K in normal grasses except for pasture grasses which only shows the mean value of 0.73. The sequence for natural radionuclide TF ratios was as follows:

- K-40: Rn-M12 > Rn-M11 > Radioactive sample > Rn-M13
- U-238 and U-235: Rn-M11 > Rn-M12 > Rn-M13 > Radioactive sample
- Th-232: Radioactive sample > Rn-M12 > Rn-M13 > Rn-M11

Transfer factor results indicated that there is a possibility of radionuclide uptake by plants. All samples were reported to have TF ratios that were very high for almost all radionuclides analyzed in this study (except for Pb-212 at 238.6 keV and Ac-228 at 969.1 keV). According to Fite and Leta (2015), if the TF ratios > 1, the plants have accumulated elements, the TF ratios approximately 1 indicate that the plants are not influenced by the elements, and if the TF ratios < 1, it is the indication that plants avoid the elements from the uptake.

4.3. Potentially toxic elements

The results for total trace elements are discussed in detail in the publication by Khumalo *et al.* (2020). The summary for this paper is given in section 4.3.1.

4.3.1. Pseudo total elements results

The PTE concentrations obtained by the pseudo total analysis were compared to the soil permissible limits regulated by the Hungarian Government (6/2009. IV. 14. 2009) based on the regulation of other European (EU) member countries (Rékási and Filep, 2012). Additionally, the target values (Denneman and Robberse 1990; Ministry of Housing, Netherlands, 1994) and the allowed levels (Commission of the European Communities, 1986) for elements in the soil are included for reference in comparison of the Hungarian limits to the World's limits. (Table 7.) indicates the permissible limits in soil.

Table 7. Permissible limits for PTEs in soil

Element	**[a]Target value of soil (mg/kg)**	**[b]Concentration of elements in soil and contamination limit values (mg/kg)**	**[c]Allowed levels in soil (mg/kg)**	**[d]Intervention values (mg/kg)**
Cd	0.80	1.00	1.00 – 3.00	12.0
Co	-	30.0	-	240
Cr	100	75.0	100 – 150	380
Cu	36.0	75.0	50.0 – 140	190
Fe	-	-	-	-
Mn	-	-	-	-
Ni	35.0	40.0	35.0 – 75.0	210
Pb	85.0	100	50.0 – 300	530
Zn	50.0	200	150 – 300	720

[a]Target values are specified to indicate desirable maximum levels of elements in unpolluted soils (Denneman and Robberse 1990).
[b]Contamination limit values for Hungarian soils (6/2009. IV. 14. 2009; Rékási and Filep, 2012).
[c]Allowed levels in soil (Commission of the European Communities, 1986).
[d]Intervention values (Netherlands Ministry of Housing, Spatial Planning and the Environment (VROM), 2000).

The results were presented according to the elemental importance in soil. The different classifications of these PTEs are discussed in section 2.2.2. The elements that are required by plants in large quantities (Ca, K, Mg, P, and Fe) are illustrated in (Figure 17.); elements that are needed in small amounts are shown in two figures for clear visibility: Co, Mn, and Cu are shown in (Figure 18.) and Na, Zn, and Ni are illustrated in (Figure 19.); and toxic elements (Cd, Cr, and Pb) in (Figure 20.). The pseudo total concentrations of the PTEs in soil and plants are presented in (Appendix A7.) for Rn-M11, (Appendix A8.) for Rn-M12, (Appendix A9.) for Rn-M13, and (Appendix A10.) for the Radioactive sample. According to Hooda (2010), Fe is an essential micronutrient; however, within soils and plants, it is not toxic because its average concentration is generally greater than 100 mg/kg; hence it was presented together with the macronutrients. For visual presentation of each element in the depths of each sample, the PTE figures showing the tendencies for each element are presented in (Appendix A11.).

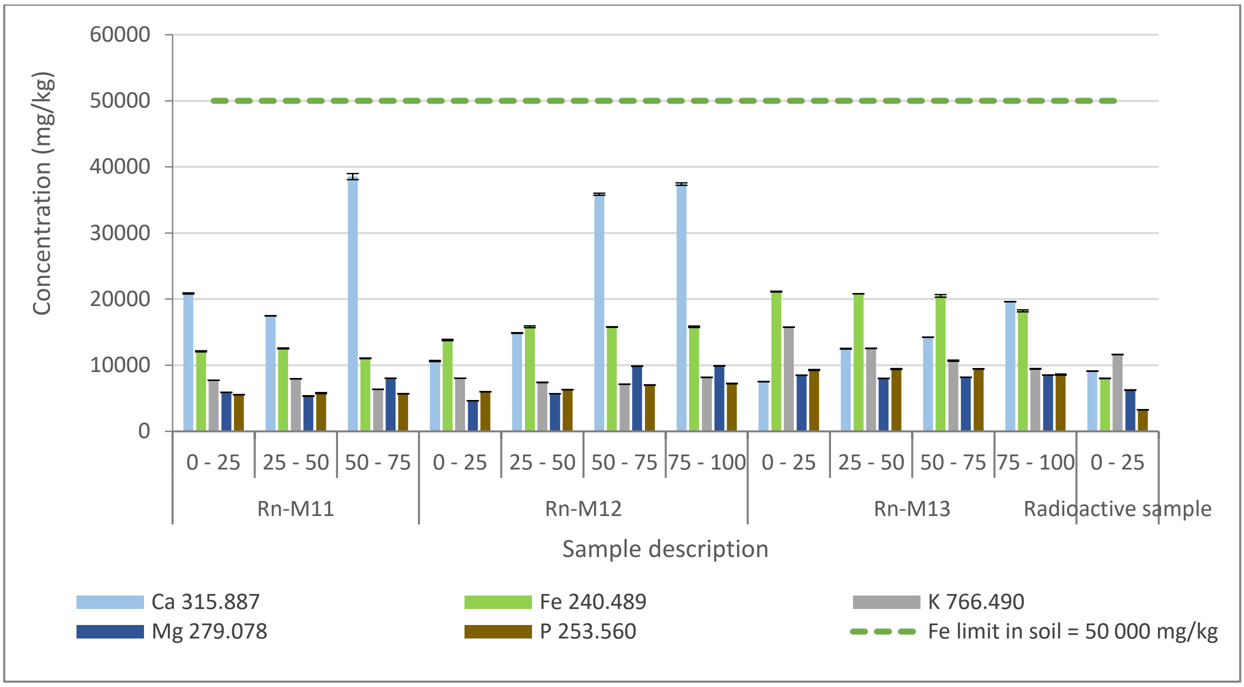


Figure 17. The total concentration of Ca, Fe, K, Mg, and P in soil samples

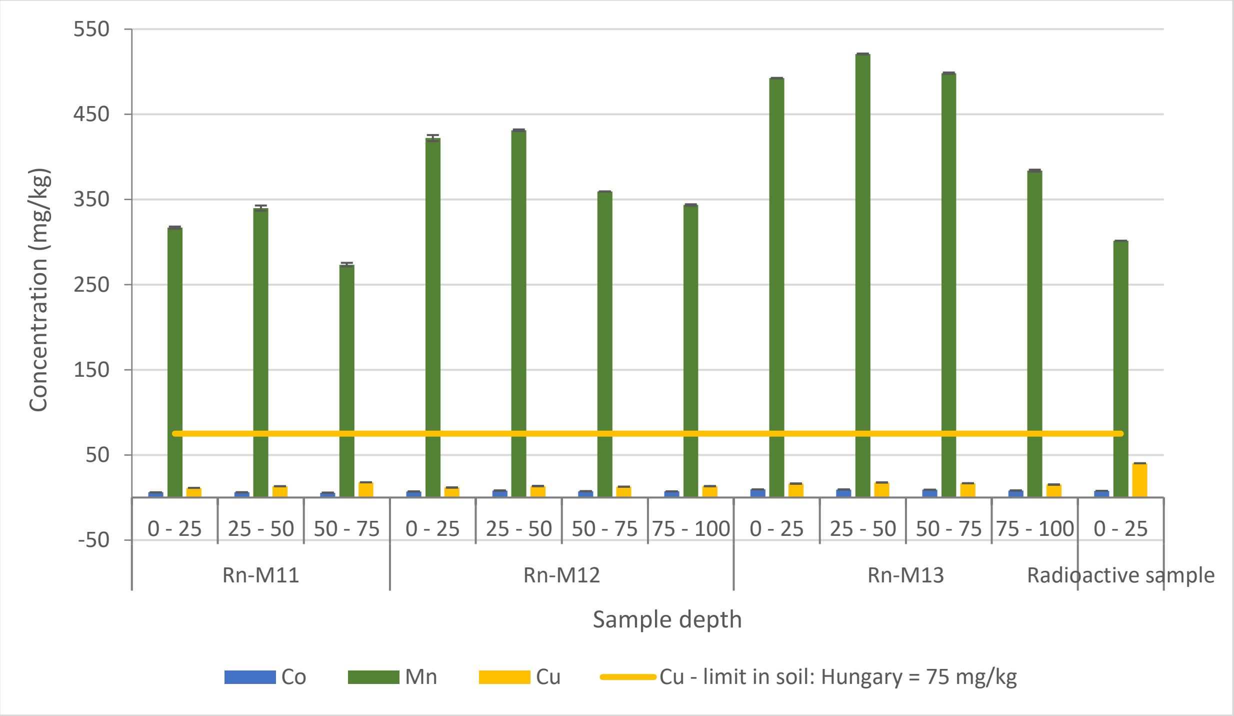


Figure 18. The total concentration of Co, Cu, and Mn in soil samples

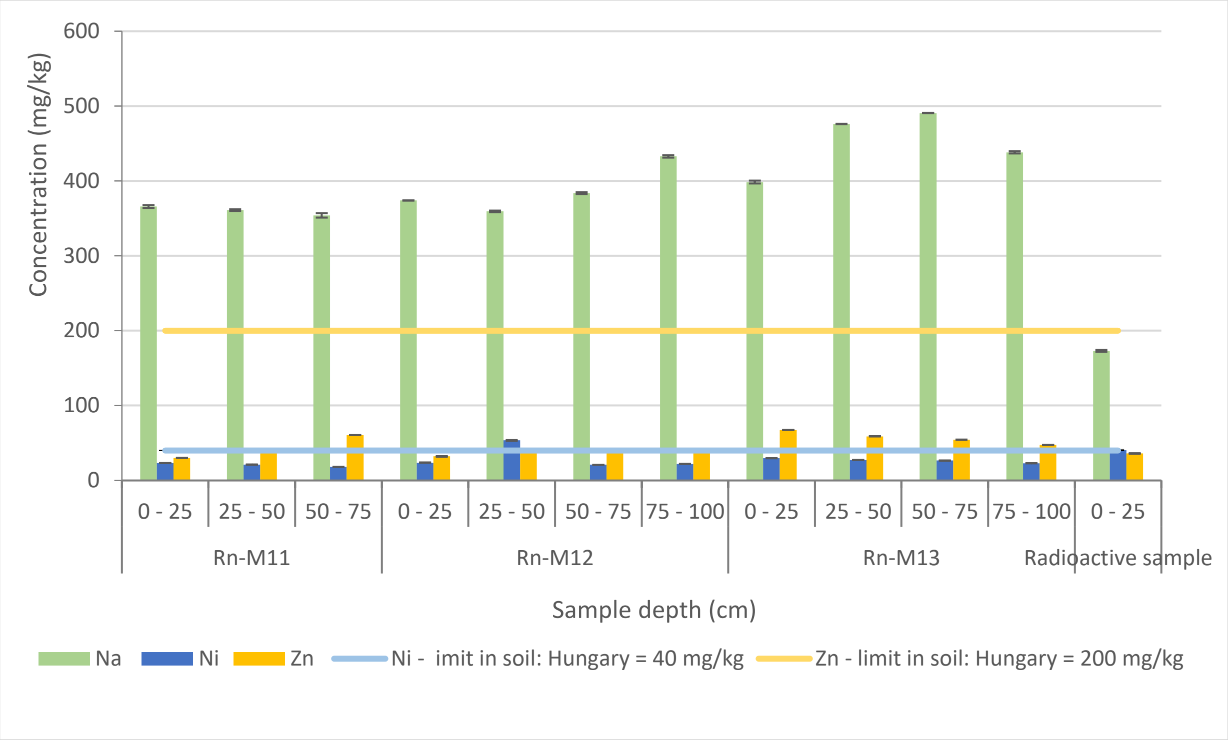


Figure 19. The total concentration of Na, Ni, and Zn in soil samples

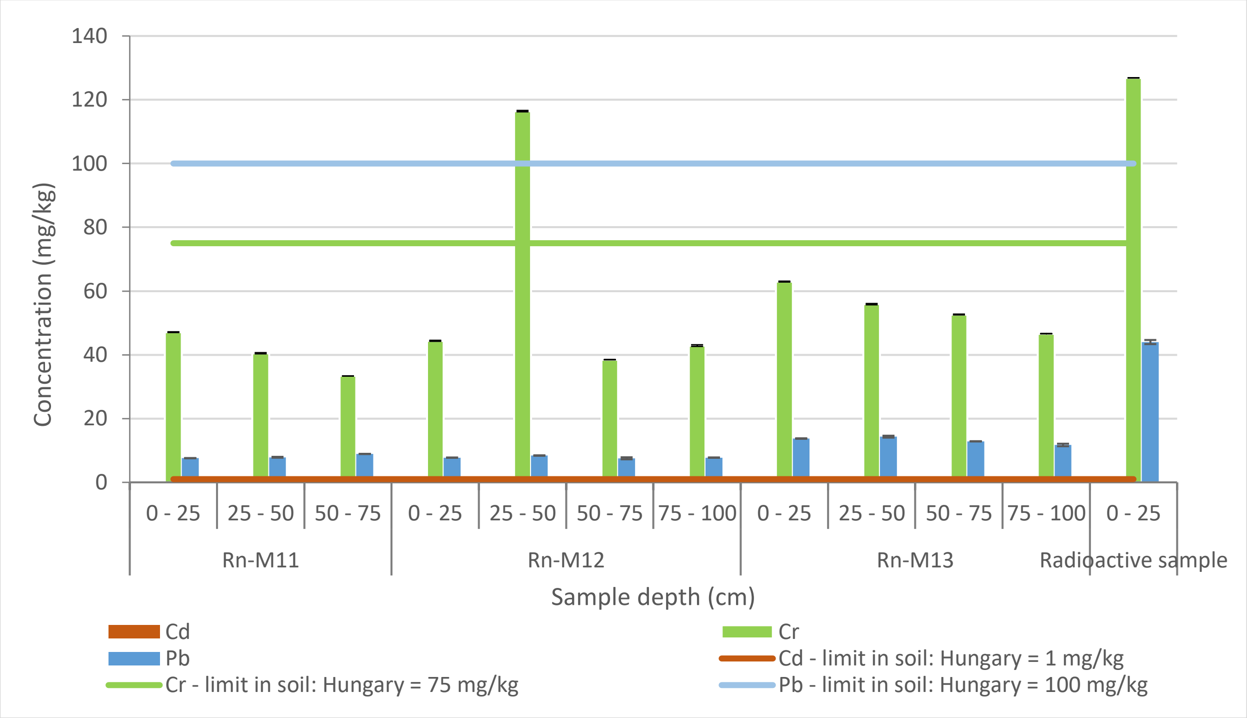


Figure 20. The total concentration of Cr, Cd, and Pb in soil samples

The concentration levels for Co, Cu, and Zn in soil were within the Hungarian limit values of 30 mg/kg, 75 mg/kg, and 200 mg/kg, respectively (6/2009. IV. 14. 2009; Rékási and Filep, 2012). Nickel concentration levels for sample Rn-M12 at 25 – 50 cm and for Radioactive sample were above the limits recommended for soil in Hungary (limit of 40 mg/kg), which were 53.5 ± 0.43

mg/kg and 40.5 ± 0.06 mg/kg, respectively. However, these Ni concentrations were still within the allowed concentrations of 35 – 75 mg/kg recommended by the Commission of the European Communities (1986). Although micronutrients from the soil are essential for plant growth and development (Voss, 1998), high concentration levels can be toxic (Stanojković-Sebić *et al.*, 2017). The results for toxic elements in the soil from spoil deposit No. I indicated that the Cd element was lower than the detection limit, whereas the Pb concentrations were below the Hungarian limits. Overall, the average concentrations for PTEs were all within the Hungarian limits for soils collected in Rn-M11, Rn-M12, and Rn-M13. Although Cr concentrations for the Rn-M12 sample at 25 – 50 cm (116 ± 0.21 mg/kg) and Radioactive sample (127 ± 0.01 mg/kg) were above the Hungarian limit value of 75 mg/kg, these concentrations were still within the allowed levels of 100 – 150 mg/kg (Commission of the European Communities, 1986).

Most PTE concentrations in soil samples collected from the spoil deposit No. I was lower than the contamination limit values for Hungarian soils and within the permissible limits. The results for micronutrients indicated that there was a trend in the increase of concentrations of elements as the slope of the deposit decreased. Soil samples collected from the top of the deposit (Rn-M11) contained less average concentration, followed by the sample collected from the slope (Rn-M12) and the sample collected from the bottom of the deposit (Rn-M13) had the highest average concentration. It should be noted that this sample was also observed to have the highest soil organic matter content. This observation might be an indication that there is a possibility of migration of these elements from the top to the bottom of the spoil deposit. The observations revealed that the Rn-M13 sample had the highest average concentrations for each element among all samples except in cases of Cr, Cu, Ni, and Pb, where the Radioactive sample had the highest average concentrations. It could be noted that the Rn-M13 also had the highest percentage of soil moisture content. This might be an indication that there is the possibility of water infiltrating through the deposit while migrating PTEs. The abundance of Cr, Cu, Ni, and Pb in the Radioactive sample could mean that these elements are correlated to U-238.

It is worth mentioning that the U total concentrations in soil and plants were not measured using the pseudo total method. However, the U concentrations in soil for 0 – 25 cm depth could be obtained from the sum of the BCR sequential method. The U concentrations for the depths 25 – 50, 50 – 75, and 75 – 100 were calculated from U-238 using specific activity concentration, hence the exclusion of U concentrations in the figures above. The calculated U concentrations are presented in (Appendix 7.), (Appendix 8.), (Appendix 9.), and (Appendix 10.) with the rest of the PTEs.

The concentrations for the elements in analyzed plants were compared to the WHO (1996) permissible limits for plants as indicated in (Table 8.).

Table 8. Total concentrations for elements in plant samples

Element	**Detection limits**	**Rn-M11 Concentration (mg/kg)**	**Rn-M12 Concentration (mg/kg)**	**Rn-M13 Concentration (mg/kg)**	**Radioactive unwashed plant Concentration (mg/kg)**	**Radioactive washed plant Concentration (mg/kg)**	***Permissible values of plant (mg/kg)**
Ca	1.30E-05	5710 ± 31.3	4400 ± 9.01	2133 ± 6.89	5369 ± 0.25	5184 ± 24.9	-
Cd	3.45E-05	n.d.	n.d.	n.d.	n.d.	n.d.	0.80
Co	5.64E-05	n.d.	n.d.	n.d.	n.d.	n.d.	-
Cr	5.59E-05	**14.2 ± 0.01**	n.d.	n.d.	n.d.	n.d.	1.30
Cu	3.35E-05	5.89 ± 0.05	**18.3 ± 1.21E-03**	**59.5 ± 0.07**	**21.7 ± 0.13**	**73.1 ± 0.10**	10.0
Fe	3.81E-05	265 ± 2.08	94.93 ± 0.32	301 ± 1.16	52.39 ± 0.21	68.59 ± 0.20	-
K	1.12E-03	7206 ± 11.9	7608 ± 87.7	8861 ± 16.0	13289 ± 7.37	14302 ± 3.17	-
Mg	1.15E-05	1781 ± 7.97	1786 ± 5.06	785 ± 0.51	868 ± 4.30	925 ± 1.69	-
Mn	8.22E-06	34.2 ± 0.07	21.5 ± 0.11	26.1 ± 0.29	3.68 ± 0.02	8.97 ± 0.06	-
Na	7.22E-04	73.9 ± 0.31	43.0 ± 0.12	36.9 ± 0.25	33.5 ± 0.87	52.8 ± 0.29	-
Ni	6.74E-05	5.48 ± 0.03	2.42 ± 0.01	n.d.	n.d.	n.d.	10.0
P	4.12E-03	1411 ± 0.28	1150 ± 2.08	1307 ± 0.34	1489 ± 9.02	1631 ± 3.95	-
Pb	2.21E-04	n.d.	n.d.	n.d.	n.d.	n.d.	2.00
U	9.42E-04	13.6 ± 2.33	15.6 ± 2.38	13.7 ± 1.30	11.9 ± 2.05	11.5 ± 1.67	
Zn	4.20E-05	**20.9 ± 0.03**	**9.41 ± 0.03**	**13.6 ± 0.05**	**8.87 ± 0.02**	**13.3 ± 0.22**	0.60

Results highlighted in **bold** were above the WHO permissible limits and n.d. refers to not detectable.
*****The permissible values in plants (WHO, 1996; Ogundele *et al.*, 2015).

Chromium concentration for the Rn-M11 sample (14.2 ± 0.01 mg/kg) was observed to be above WHO (1996) limit of 1.3 mg/kg. Copper concentrations for Rn-M12, Rn-M13, and Radioactive (both unwashed and washed plant) samples were above the WHO permissible limit of 10 mg/kg. It was also observed that the concentrations for Cu increased as the topography changed from the top of the deposit to the bottom of the deposit. The increase in Cu concentration at the bottom of the deposit might be an indication that Cu was transported down the slope of the deposit and has accumulated at the bottom of the deposit. Zinc concentrations for all the plant samples collected were above WHO permissible limits of 0.6 mg/kg. This observation showed that Zn might have been available from soil to be absorbed in large quantities by plants. Due to metals being bound very tightly to the soil or exist in solid minerals at high pH, Jones and Jacobsen (2001) are of the opinion that Cu, Mn, Zn, Fe, and Ni will be more available at low pH levels than at high pH levels. It could be notable that the plant moisture and average concentrations for plants were having the same trends. The plant sample with the highest % plant moisture was observed to have accumulated elevated PTE concentration and vice versa. When comparing the results for the washed and unwashed Radioactive plant samples, there was no noticeable difference in concentration. This indicated that the concentrations of the elements obtained in plants are not due to surface contamination but are due to biological uptake.

The transfer factor (TF) ratios were calculated from the plant shoots. The results are indicated in (Table 9.) to determine the ability of a metal species to migrate from the soil into plant roots.

Table 9. Soil/Plant Transfer Factor (TF) from pseudo total concentrations

Elements	Rn-M11	Uncertainty	Rn-M12	Uncertainty	Rn-M13	Uncertainty	Radioactive sample	Uncertainty
Ca	0.27	± 0.36	0.41	± 0.11	0.28	± 0.88	0.59	± 0.01
Cd		-		-		-		-
Co	n.d.		n.d.		n.d.		n.d.	
Cr	0.30	± 0.11	n.d.		n.d.		n.d.	
Cu	0.52	± 1.50	**1.56**	**± 0.02**	**3.64**	**± 14.33**	0.54	± 4.58
Fe	0.02	± 0.03	0.01	± 0.00	0.01	± 0.02	0.01	± 0.01
K	0.93	± 2.15	0.95	± 15.80	0.56	± 0.65	**1.14**	**± 1.19**
Mg	0.30	± 0.92	0.39	± 0.35	0.09	± 0.02	0.14	± 0.11
Mn	0.11	± 0.06	0.05	± 0.03	0.05	± 0.84	0.01	± 0.21
Na	0.20	± 0.16	0.12	± 13.04	0.09	± 0.13	0.19	± 0.66
Ni	0.24	± 0.60	0.10	± 0.29	n.d		n.d.	
P	0.25	± 0.01	0.19	± 0.09	0.14	± 4.57E-03	0.46	± 2.05
Pb	n.d.		n.d.		n.d.		n.d.	
Zn	0.69	± 0.12	0.29	± 0.11	0.20	± 0.39	0.25	± 0.09

n.d. represents not detectable.

It was observed that in the soil, the concentrations for Ni and Cr were above the Hungarian limits in some sampling locations. However, the TF values for these elements were very low. This could be the result of the plant having low suitability for phytoextraction and phytoremediation (Mirecki *et al.*, 2015). Mirecki *et al.* (2015) further explain that the availability of PTEs for plants is controlled by the plant's requirement for micronutrients and the capacity to absorb and eliminate toxic elements. Agic *et al.* (2015) emphasized that the availability, uptake, and accumulation of PTEs vary by shoots and roots for different plant species and by their adaptation to the environmental conditions. Hence, low TF values may be due to some elements having low translocation capabilities from the roots to the shoot system of the plant. As a result, higher element concentrations in the soil does not necessarily mean higher element uptake by plants. Furthermore, Fite and Leta (2015) state that the direct deposition and foliar absorption could cause a high level of elements in the plants more than the translocation of the root-to-upper plant portion. Copper TF values indicated an increasing trend from the Rn-M11 with reported TF = 0.52 ± 1.50 , M12 with TF = 1.56 ± 0.02 and to M13 with TF = 3.64 ± 14.33. This increasing trend might imply that there was a migration of Cu from the top of the deposit to the bottom of the deposit. Copper TF values for sampling points Rn-M12 and Rn-M13 were above the limit of 1, whereas the Radioactive sample reported the TF value of K to be 1.14 ± 1.19. This observation implies that Cu and K metals were available to be taken up by plants (Intawongse *et al.*, 2007, Agic *et al.*, 2015).

4.3.2. Uranium concentrations in water

The concentrations of U in water were determined to evaluate the mobility of U from the surrounding spoil deposits into the groundwater and to determine the risks associated with its chemical effects. In addition, the levels of U in effluent water were determined to ensure that when the effluent water is released into the environment, it is within the regulatory limits to protect the environment. Furthermore, the content of U in seepage water was determined to find out how much of U could possibly seep into the groundwater. The U concentrations were measured on the groundwater effluent water and seepage water from the Mecsek uranium mine, and the results were recorded in (Table 10.). The IAEA (2004b) reported on the treatment of liquid effluents from uranium mines and mills during and after the operation. In this report, the concentrations for U in process water, pond water, seepage from the dams, and shallow groundwater measured in the Mecsek mine between 1996 and 2000 were reported and are listed in (Table 10.) below. In Hungary, the research of natural radioactivity of groundwater is an essential issue since both the drinking water supply and the bottled mineral water market rely almost 100 % on groundwater (Erőss *et al.*, 2018). Therefore, the groundwater concentrations obtained in this study were compared to the total uranium guideline value of 0.03 mg/L in drinking water set by WHO (2004). In contrast, the effluent water and the seepage water results were compared to the U_{max} limit of 2 mg/L, which is regulated by the Hungarian Ministerial Decree No. 15/2001 (VI. 6.) KöM (2001). All the water results obtained in this study were also compared to the results that were previously obtained in the Mecsek uranium mine and published by the IAEA (2004b).

Table 10. Uranium concentrations for water samples

Results from the current study			**Results from the IAEA** (2004b) **Study**	
Sample ID	**Sample description**	**U concentration (mg/L)**	**Sample description**	**U concentration (mg/L)**
PK-33/1	Groundwater (No. I)	6.06 ± 0.03	groundwater	0.01 – 0.04
PK-44/3	Groundwater (No. I)	0.23 ± 0.001	pond water	0.03
PK-29/1	Groundwater (No. I)	1.87 ± 0.01	process water	< 0.5
1504/1	Groundwater (No. I)	2.78 ± 0.01	seepage water	2 - 5
P-2/5	Groundwater (No. II)	1.90 ± 0.06		
P-2/6	Groundwater (No. II)	0.52 ± 0.003		
Elfolyó	treated mine water	0.32 ± 0.001		

6/11 Szint	mine water from the spoil deposit No. I	2.46 ± 0.01
Északi-táró	mixed water: mine water from the waste deposit No. III and leaking water from precipitation	6.72 ± 0.04
IIIM. Gyűjtő	seepage water from the waste deposit No. III	6.99 ± 0.02
Cs-0	seepage water from the waste rock pile No. II	0.84 ± 0.003

In this study, the focus was on the groundwater samples collected near spoil deposit No. I, since it is the deposit that the entire study is based on. Groundwater sample PK-33/1 collected near the spoil deposit No. I indicated an elevated U concentration of 6.06 ± 0.03 mg/L which is above the WHO guideline value for U (0.03 mg/L) in drinking water (WHO, 2004). In fact, all four groundwater samples collected near the spoil deposit No. I (PK-33/1, PK-44/3, PK-29/1, and 1504/1) showed a large difference in concentrations (6.06 ± 0.03 mg/L, 0.23 ± 0.001 mg/L, 1.87 ± 0.01 mg/L, and 2.78 ± 0.01 mg/L respectively). It was observed that all groundwater samples collected in this study were above the WHO guideline value. Elevated U concentrations in groundwater are of concern because it indicates active processes of the uranium leaching from the spoil deposits. This increase also indicates the capability of elements migrating into the groundwater. The elevated U levels in groundwater may be due to the seeping of water containing high concentrations of U from the process water. According to Abiye and Shaduka (2017), if the tailings are not lined, the seepage from the processing water that is ponding on the tailing dams cannot be prevented from seeping into the groundwater. The comparison of U concentration in this study and the study conducted by IAEA (2004b) indicated that U concentrations in groundwater for this study ranged from 0.23 mg/L to 6.06 mg/L, whereas the U concentrations from the IAEA study ranged from 0.01 mg/L to 0.04 mg/L. It should be noted that in the Mecsek uranium mine, the groundwater contaminated with U is remediated by extracting it from the area and purifying it before being released to protect the drinking water aquifer (IAEA, 2004b). The uranium decontamination of contaminated water involves ion exchange technology, in which uranium dissolved in water is bound by the anion exchange resin, and the U content of the purified water is reduced to below 1 mg/L to ensure compliance with the U_{max} = 2 mg/L limit value (Mecsekérc Zrt., 2017). This groundwater remediation process has been continuously operating since 2007.

The U concentration in the seepage water from spoil deposit No. III was very high (6.99 ± 0.02 mg/L) as compared to the U concentration range (2 – 4 mg/L) in the seepage water reported by the IAEA. Furthermore, it was observed that both the seepage water (IIIM. Gyűjtő) from spoil deposit No. III and the mixed water (Északi-táró) from the same deposit resulted in high levels of U (6.72 ± 0.04 mg/L and 6.99 ± 0.02 mg/L respectively). Overall, the U concentrations in mine water for this study ranged from 0.32 ± 0.001 mg/L to 6.72 ± 0.04 mg/L, whereas the U concentrations from the IAEA study were < 0.5 mg/L. According to the IAEA (2011), effluent water often contains U, Ra-226, and other PTEs, often in excess of regulatory requirements. These findings explained the reasons for Mecsekérc Zrt. (2017) to focus more on waste rock dumps No. I and No. III to ensure that the remediation process is successful.

4.3.3. BCR sequential extraction

The concentrations of Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, U, and Zn in fractions F1, F2, F3, and F4 were determined from the 0 – 25 cm soil depth, and the results are given in (Table 11.). In the second column, the detection limits of elements are given in mg/kg units for the applied ICP-OES methods.

Table 11. Average concentrations for each fraction of the BCR sequential extraction and pseudo total concentrations with standard uncertainties

Fraction 1 – Acid soluble

Element	Instrumental detection limit (mg/kg)	Concentration ± standard uncertainty (mg/kg)			
		Rn-M11	Rn-M12	Rn-M13	Radioactive sample
Cd	3.45E-05	0.03 ± 4.83E-03	0.01 ± 0.01	0.05 ± 0.01	0.06 ± 0.02
Co	5.64E-05	0.05 ± 0.02	0.02 ± 0.01	0.81 ± 0.03	4.27 ± 1.84
Cr	5.59E-05	0.07 ± 0.01	0.10 ± 0.02	0.07 ± 0.01	0.30 ± 0.11
Cu	3.35E-05	n.d.	n.d.	0.08 ± 0.02	5.08 ± 2.21
Fe	3.81E-05	0.57 ± 0.53	0.30 ± 0.23	7.02 ± 0.57	824 ± 366
Mn	8.22E-06	31.1 ± 0.17	19.5 ± 1.37	166 ± 2.94	202 ± 88.1
Ni	6.74E-05	0.43 ± 0.21	0.32 ± 0.05	1.20 ± 0.02	2.28 ± 0.97
Pb	2.21E-04	0.10 ± 0.06	0.08 ± 0.20	0.20 ± 0.18	4.34 ± 2.16
U	9.42E-04	n.d	n.d	n.d	17.4 ± 7.46
Zn	4.20E-05	0.19 ± 0.25	n.d	2.05 ± 0.42	5.84 ± 2.15

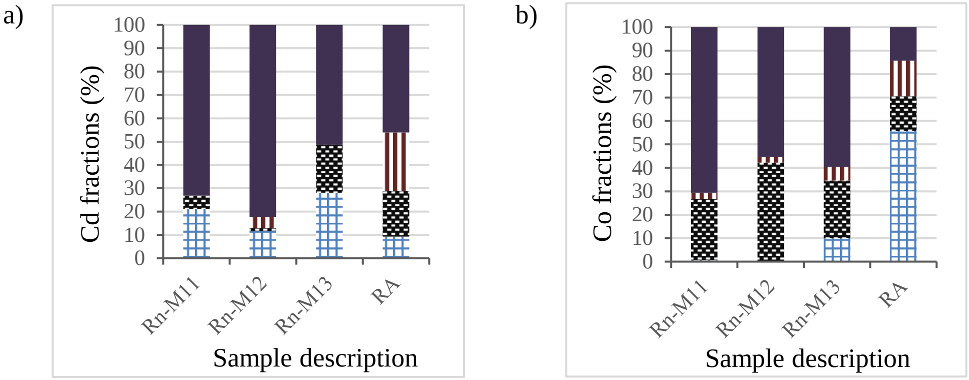
Fraction 2 - Reducible

Element	Instrumental detection limit (mg/kg)	Rn-M11	Rn-M12	Rn-M13	Radioactive sample
Cd	3.45E-05	0.01 ± 3.96E-03	n.d	0.04 ± 2.62E-03	0.12 ± 0.05
Co	5.64E-05	2.23 ± 0.08	3.33 ± 0.11	1.99 ± 0.04	1.14 ± 0.53
Cr	5.59E-05	6.00 ± 0.23	5.09 ± 1.37	2.10 ± 0.74	16.6 ± 9.41
Cu	3.35E-05	1.47 ± 0.02	1.35 ± 0.03	2.07 ± 0.22	8.89 ± 3.54
Fe	3.81E-05	4547 ± 17.9	584 ± 22.5	2805 ± 99.7	1349 ± 567
Mn	8.22E-06	177 ± 4.81	286 ± 11.0	161 ± 4.75	75.0 ± 39.0
Ni	6.74E-05	4.42 ± 0.22	4.09 ± 0.53	3.01 ± 0.37	6.14 ± 3.89
Pb	2.21E-04	3.35 ± 0.04	3.92 ± 0.07	7.18 ± 0.11	28.2 ± 11.7
U	9.42E-04	n.d	n.d	n.d	14.5 ± 5.77

Zn	4.20E-05	2.38 ± 0.36	1.32 ± 0.31	6.32 ± 0.29	5.50 ± 2.46
Fraction 3 - Oxidizable					
Element	Instrumental detection limit (mg/kg)	Rn-M11	Rn-M12	Rn-M13	Radioactive sample
Cd	3.45E-05	n.d	0.01 ± 2.60E-03	n.d	0.16 ± 0.07
Co	5.64E-05	0.22 ± 0.02	0.19 ± 0.02	0.48 ± 0.02	1.17 ± 0.51
Cr	5.59E-05	2.76 ± 0.09	2.87 ± 0.42	2.99 ± 0.50	4.84 ± 2.17
Cu	3.35E-05	0.50 ± 0.11	0.44 ± 0.10	1.05 ± 0.05	15.2 ± 6.98
Fe	3.81E-05	213 ± 4.09	192 ± 5.04	586 ± 27.8	898 ± 376
Mn	8.22E-06	9.78 ± 0.27	13.0 ± 0.32	14.8 ± 0.62	11.3 ± 5.11
Ni	6.74E-05	1.49 ± 0.03	1.64 ± 0.10	1.90 ± 0.10	2.05 ± 0.81
Pb	2.21E-04	0.48 ± 0.33	0.57 ± 0.25	0.77 ± 0.50	1.41 ± 1.00
U	9.42E-04	1.28 ± 0.21	2.07 ± 0.15	3.19 ± 0.12	18.9 ± 7.50
Zn	4.20E-05	1.04 ± 0.14	0.85 ± 0.21	2.72 ± 0.09	5.91 ± 2.28
Fraction 4 – Residual					
Element	Instrumental detection limit (mg/kg)	Rn-M11	Rn-M12	Rn-M13	Radioactive sample
Cd	3.45E-05	0.11 ± 0.03	0.09 ± 0.01	0.10 ± 0.02	0.29 ± 0.03
Co	5.64E-05	6.03 ± 0.30	4.39 ± 0.55	4.84 ± 0.28	1.09 ± 0.13
Cr	5.59E-05	74.5 ± 2.63	57.6 ± 4.58	47.3 ± 7.51	62.1 ± 6.80
Cu	3.35E-05	13.9 ± 0.28	13.7 ± 0.74	16.1 ± 0.90	10.7 ± 2.42
Fe	3.81E-05	26731 ± 4018	19861 ± 3.11	17681 ± 1639	4360 ± 430
Mn	8.22E-06	201 ± 24.4	133 ± 22.8	96.6 ± 5.00	19.7 ± 3.28
Ni	6.74E-05	32.3 ± 1.31	25.9 ± 0.97	19.5 ± 3.16	19.0 ± 3.30
Pb	2.21E-04	8.98 ± 3.22	4.87 ± 1.37	4.36 ± 0.45	10.4 ± 2.22
U	9.42E-04	n.d	n.d	n.d	n.d
Zn	4.20E-05	50.9 ± 1.76	42.5 ± 5.82	54.5 ± 2.98	15.1 ± 2.05

The n.d. refers to not detectable

The average percentage distribution of PTE concentrations for each sample (where RA represents Radioactive sample) in different fractions of the modified BCR is represented graphically in (Figure 21.).



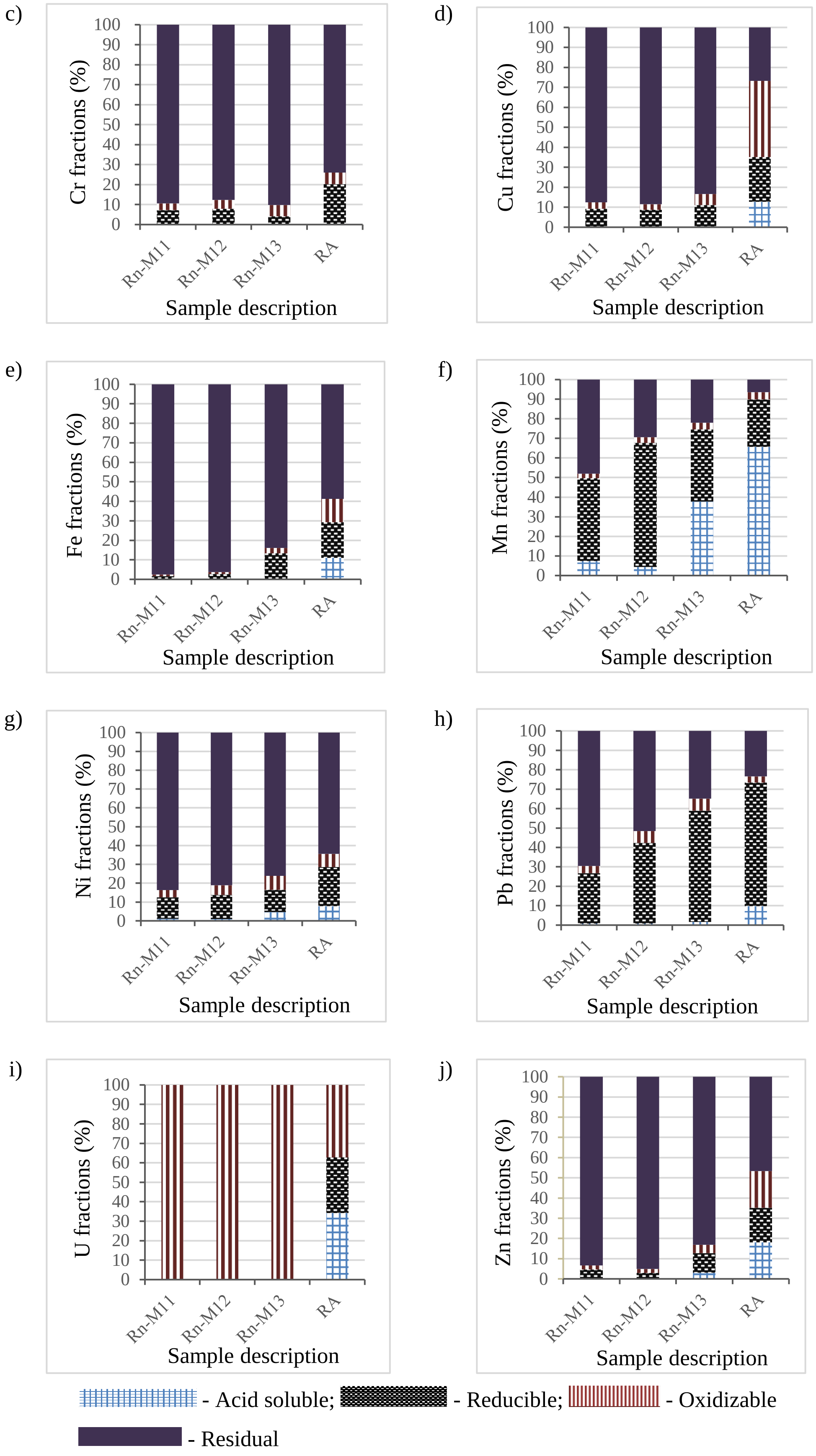


Figure 21. The average percentage for the distribution of extractable content (average %) of a) Cd, b) Co, c) Cr, d) Cu, e) Fe, f) Mn, g) Ni, h) Pb, i) U, and j) Zn in soil from uranium mine spoil bank

The results indicated that the percentages of the extractable amount of U, Mn, Pb, Co, and Cd in the non-residual fractions were subsequently high (ranging from 36.8 % to 100 %). High percentages of these PTEs in mobile fractions might lead to the high bioavailability of these elements, leading to toxicity if not properly handled (Zimmerman and Weindorf, 2010; Okoro *et al.*, 2017). Sungur *et al.* (2014) state that the availability of these elements may lead to mobility and pose a critical toxicity risk in plants, and may cause environmental pollution over time. The average percentage of Cd (17.5 %) in the acid-soluble fraction presents a significant environmental hazard, taking into consideration the high toxicity of Cd (Ma *et al.*, 2011). Although the percentage in the acid-soluble fraction is minimal, this fraction is more mobile and dangerous relative to other fractions, and thus, the pollution may become substantially high (Soltani *et al.*, 2017). The observation made was that most of the PTEs in the residual fraction followed:

either Rn-M11 > Rn-M12 > Rn-M13 > Radioactive sample,

or Rn-M12 > Rn-M11 > Rn-M13 > Radioactive sample descending orders,

except in the case of Cr where the order was Rn-M13 > Rn-M11 > Rn-M12 > Radioactive sample.

The observation made was that the Cr behavior was decreasing with the decreasing soil moisture percentage. Avudainayagam *et al.* (2003) state that adsorption/precipitation reactions, plant uptake, or sub-surfaced layers can temporarily remove Cr from the soil, provided soil pH and soil moisture are favored.

These results imply that the Radioactive sample had the highest quantity of PTEs bound to the non-residual fractions. The topographic conditions and the anthropogenic activities may have contributed to the amount of concentration found in these sample locations and the amount being released into the environment. The Radioactive sample, which was sampled from the bottom of the spoil deposit, had lower pH, lower CEC with a higher PTE distribution percentage, whereas the samples collected from the top of the deposit and on the slope showed slightly higher pH, higher CEC, with less PTE distribution percentage. Furthermore, the study conducted by Khumalo *et al.* (2020) indicated that the soil samples collected from the top of the deposit (Rn-M11) contained less average total concentration, followed by the sample collected from the slope (Rn-M12) and the samples collected from the bottom of the deposit (Rn-M13 and Radioactive sample) had the highest average total concentrations. This could be due to the leaching from the spoil rock and transportation along by the lateral water stream horizontally from the top of the deposit to the bottom of the deposit. Additionally, there was no trend in terms of depths; the PTE concentrations varied throughout different depths of each sample. This could be the result of different soil types that were used to form the soil cover layer of the spoil deposit in question (Khumalo *et al.*, 2020). It was also noted that the Rn-M13 sample had similar trends to the Radioactive sample when it came to the distribution of elements, except the Rn-M13 contained less concentration of U. The reason might be that both these samples were sampled at the bottom of the spoil deposit where the soil had elevated total concentrations of PTEs as per the study conducted by Khumalo *et al.* (2020).

The results of this study are in agreement with numerous studies (Bielicka-Giełdoń *et al.*, 2013; Sungur *et al.*, 2014; Sahito *et al.*, 2015; Pavlović *et al.*, 2018). The study conducted by Bielicka-Giełdoń *et al.* (2013) resulted in an abundance of Fe, Cu, Cr, and Ni in the residual fraction. Similarly, the study conducted by Pavlović *et al.* (2018) indicated that Fe, Cu, Cr, and Ni were predominantly in the residual fraction at most sites investigated. Furthermore, in the study conducted by Sungur *et al.* (2014), the authors stated that it was clear that Mn, Cd, and Pb are the most mobile elements in soil samples, whereas the least-mobile elements are Ni, Zn, and Cu. Additionally, the PTEs that were bound to the non-residual fractions suggest that they are primarily derived from anthropogenic inputs. In contrast, the PTEs in the residual fractions indicate that they are of lithogenic origin (Baran and Tarnawski, 2015). The study conducted by Sahito *et al.* (2015) indicated that the total Cu and Pb were the most mobile elements in the soils investigated as compared to Cd, Fe, and Zn. According to De Almeida *et al.* (2007), Cd, Pb, Cu, and Cr are associated with various bioavailable geochemical fractions, like the water-soluble fraction, the exchangeable fraction, and the non-available fractions like those associated with the crystalline net of clays and silica minerals.

The mobility of PTEs in soil

The fractions weakly bound to soil components (acid-soluble, reducible, and oxidizable) were used to assess the mobility of elements in soil (Kabala and Singh, 2001). The mobility factor was calculated, and the results are illustrated in (Figure 22.). The results indicated the mobility of PTEs was in the following order:

- Rn-M11 sample: U > Mn > Pb > Co > Cd > Ni > Cu > Cr > Zn > Fe,
- Rn-M12 sample: U > Mn > Pb > Co > Ni > Cd > Cr > Cu > Zn > Fe,
- Rn-M13 sample: U > Mn > Pb > Cd > Co > Ni > Zn > Cu > Fe > Cr, and
- Radioactive sample: U > Mn > Co > Pb > Cu > Cd > Zn > Fe > Ni > Cr.

The mobility order for all the samples asserts that U followed by Mn and Pb were highly mobile in this study, while Fe, Zn, and Cr were less mobile. According to Wijaya *et al.* (2018), the dominance of Fe in the residual fraction is an indication that Fe was from natural soil minerals bound to silicate and aluminate. This data reveals that Cd, Mn, Pb, and U were more available for mobility compared to other PTEs in this study. The high mobility of U might be coming from the wounding that occurred during the remediation process of the spoil deposit No. I. Under normal circumstances, Cd is usually more mobile than Pb. However, natural radioactive elements such as uranium break down and form Pb as one of their end products (Kotoky *et al.*, 2003). This explains the reason Pb was found to be more mobile after U and Mn. Based on previous studies, sequential extractions revealed that a significant fraction of U and possibly Pb and Cd could be considered as mobile (Skipperud and Salbu, 2015).

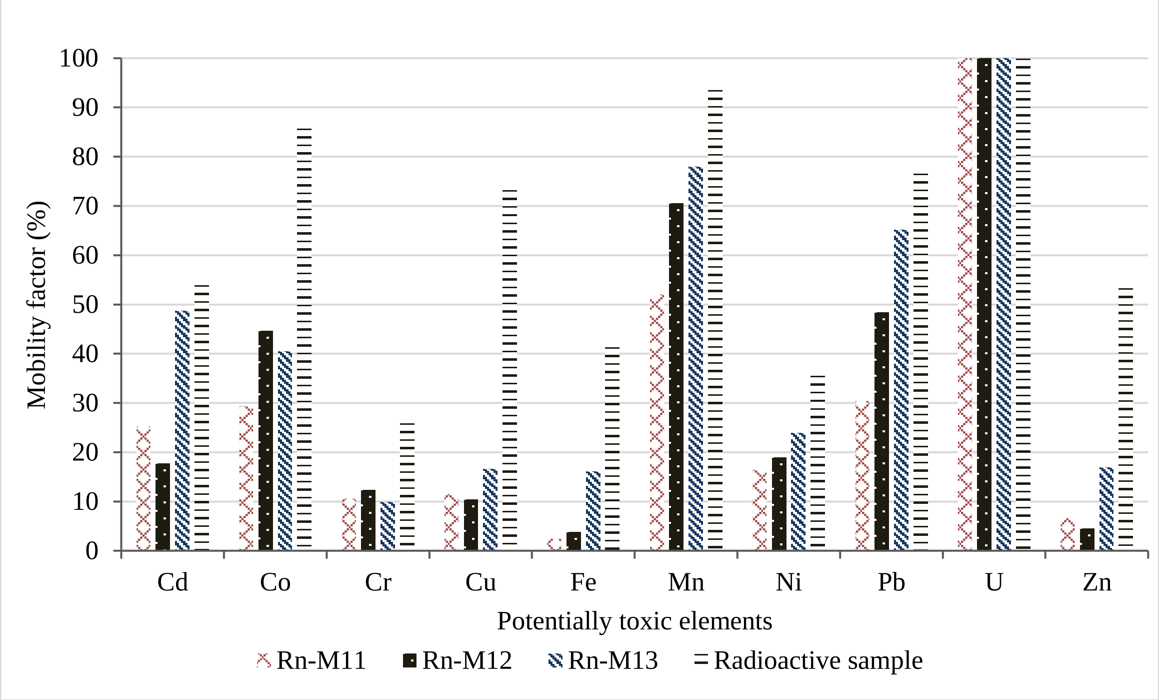


Figure 22. Potential mobility of PTEs in soil

4.3.4. Quality control

The quality control for the BCR sequential procedure was conducted using the Certified Reference Material (CRM) BCR®-701, and the results are presented in (Table 12.).

Table 12. Quality control results obtained for the analysis of BCR®-701

Extraction step	Element	Experimental value (mg/kg of dry mass ± standard deviation)	Certified value (mg/kg of dry mass ± standard deviation)	Recovery (%)
Step 1	Cd	7.20 ± 0.08	7.34 ± 0.35	98.1
	Cr	2.88 ± 0.03	2.26 ± 0.16	127
	Cu	50.69 ± 0.59	49.30 ± 1.70	103

	Ni	11.93 ± 0.11	15.40 ± 0.9	77.5
	Pb	0.06 ± 0.26	3.18 ± 0.21	1.95*
	Zn	176.1 ± 4.12	205.0 ± 6.0	85.9
Step 2	Cd	2.30 ± 0.04	3.77 ± 0.28	99.5
	Cr	39.80 ± 0.30	45.70 ± 2.0	89.3
	Cu	109.3 ± 0.69	124.0 ± 3.0	92.0
	Ni	23.94 ± 0.33	26.60 ± 1.3	90.0
	Pb	114.2 ± 1.48	126.0 ± 3.0	89.7
	Zn	104.7 ± 0.81	114.0 ± 5.0	91.9
Step 3	Cd	0.78 ± 0.01	0.27 ± 0.06	128
	Cr	107.1 ± 0.51	143.0 ± 7.0	76.4
	Cu	48.84 ± 0.14	55.2 ± 4.0	95.9
	Ni	13.44 ± 0.08	15.30 ± 0.9	82.6
	Pb	9.28 ± 0.64	9.30 ± 2.0	92.4
	Zn	38.01 ± 0.23	45.70 ± 4.0	78.4

* The recovery percentage is too low.

The results produced good recoveries except for Pb in the acid-soluble fraction, which indicated a very low yield of 1.95 %. There could be several reasons for the Pb recovery to have a low yield. According to Barać *et al.* (2016), errors during elemental analysis can be caused by improper sampling, storage, sample preparation, and the analysis itself. Furthermore, Barać *et al.* (2016) state that a poor recovery could be the result of washing with water between two extraction steps, however in this study, only Pb had a poor recovery, and therefore, this could not be the reason in this case. On the other hand, Van Herreweghe *et al.* (2003) believe that the overall poor recovery rates in the extraction procedure could be caused by the heterogeneity of the sample. It was observed that Cr in the acid-soluble fraction and Cd in the oxidizable fraction extracted 127 % and 128 %, respectively. This might be due to a cumulative error in each of the extraction measurements (Van Herreweghe *et al.*, 2003).

In addition, the validation of the BCR sequential procedure is presented in (Table 13.) and in (Appendix A12.) indicated that there was a slight difference between the sum of the concentrations cumulated concentrations of fractions (F1+F2+F3+F4) in the BCR procedure and the pseudo total concentrations.

Table 13. Validation for the BCR sequential procedure

Sum of Fraction 1 + Fraction 2 + Fraction 3 + Fraction 4					
Element	Instrumental detection limit (mg/kg)	Rn-M11	Rn-M12	Rn-M13	Radioactive sample
Cd	3.45E-05	0.15 ± 0.05	0.11 ± 0.03	0.20 ± 0.04	0.62 ± 0.16
Co	5.64E-05	8.53 ± 0.42	7.93 ± 0.69	8.12 ± 0.36	7.67 ± 3.01
Cr	5.59E-05	83.4 ± 2.96	65.6 ± 6.40	52.4 ± 8.76	83.9 ± 18.5
Cu	3.35E-05	15.9 ± 0.43	15.28 ± 0.89	19.3 ± 1.19	39.9 ± 15.2
Fe	3.81E-05	27398 ± 4040	20637 ± 3140	21079 ± 1767	7430 ± 1740

Mn	8.22E-06	419 ± 29.6	452 ± 35	439 ± 13.3	308 ± 135
Ni	6.74E-05	38.7 ± 1.77	32.0 ± 1.66	25.6 ± 3.65	29.5 ± 8.86
Pb	2.21E-04	12.9 ± 3.66	9.45 ± 1.90	12.5 ± 1.24	44.4 ± 17.1
U	9.42E-04	1.28 ± 0.35	2.07 ± 0.23	2.72 ± 0.19	50.8 ± 20.7
Zn	4.20E-05	54.5 ± 2.51	44.7 ± 6.52	65.6 ± 3.77	32.3 ± 8.94
Pseudo total concentrations (0 – 25 cm) (Khumalo *et al.*, 2020)					
Element	Instrumental detection limit (mg/kg)	Rn-M11	Rn-M12	Rn-M13	Radioactive sample
Cd	3.45E-05	n.d	n.d	n.d	n.d
Co	5.64E-05	6.16 ± 0.07	7.15 ± 0.02	9.53 ± 0.09	7.87 ± 0.01
Cr	5.59E-05	47.10 ± 0.20	44.39 ± 0.27	62.97 ± 0.22	126.83 ± 0.01
Cu	3.35E-05	11.31 ± 0.06	11.73 ± 0.08	16.35 ± 0.01	40.25 ± 0.05
Fe	3.81E-05	12104 ± 139	13817 ± 169	21139 ± 109	8014 ± 57.1
Mn	8.22E-06	317.0 ± 2.04	422.1 ± 6.21	492.43 ± 0.60	301.52 ± 0.17
Ni	6.74E-05	23.17 ± 0.10	23.91 ± 0.06	29.65 ± 0.03	40.54 ± 0.10
Pb	2.21E-04	7.58 ± 0.01	7.74 ± 0.01	13.73 ± 0.03	44.0 ± 1.11
Zn	4.20E-05	30.13 ± 0.37	32.09 ± 0.45	67.43 ± 0.12	36.05 ± 0.38

The results of the comparison between the sum of the accumulated BCR concentrations and the concentrations obtained from the pseudo total were found to be acceptably close. There were some cases where the concentrations of the BCR were larger than the concentrations obtained by pseudo total. This could be caused by the cumulative error from each extraction measurement (Van Herreweghe *et al.*, 2003).

4.4. Statistical analysis

4.4.1. Statistical results for the physicochemical properties

The mean, standard error, standard deviation, variance, kurtosis, and skewness for the soil characteristics were determined and are shown in (Appendix A13.).

The descriptive statistical analysis was conducted for Rn-M11, Rn-M12, and Rn-M13 samples. The statistical results indicated that the kurtosis of all the variables measures for the Rn-M11 sample was undefined due to less than four data points, whereas the skewness for all the variables was within the acceptable threshold. In addition, the Rn-M12 sample indicated that the kurtosis of the soil moisture (%), pH, and soil organic matter (%) were out of the +/- 2 acceptable threshold, whereas, for Rn-M13, only the soil moisture (%) was out of this threshold. The skewness of all the measured variables in each sample was within the acceptable threshold. The descriptive statistics for all the soil samples combined indicated that the kurtosis and the skewness of the pH, CEC, soil moisture (%), and soil organic matter (%) were all within the +/- 2 acceptable threshold, which indicated normal distribution. However, the kurtosis of the plant moisture (%) and soil organic matter (%) were out of this threshold. This is due to some of the percentages of these variables being the outliers which suggests that the data distribution was not normal in this case. In distribution analysis, the skewness is used to indicate asymmetry and deviation from a normal distribution in nature, where the high values of skewness are caused by the large variation in the concentrations, which resulted in outlier values (Alshahri, 2019).

In the t-test statistical analysis, only Rn-M11, Rn-M12, and Rn-M13 samples were considered. The Radioactive sample was excluded due to having only one depth 0 – 25 cm and also having outliers. One-Sample t-test statistical analysis was conducted to determine if there are differences in each soil characteristic in terms of topography for different samples (Appendix A14.) and in terms of different depths for each sample (Appendix A15.). When considering the topography, it was observed that the SOM values were significantly different in all three depths that were considered for this analysis, whereas the CEC was only significantly different in 25 – 50 cm depth. In terms of depth, CEC and SOM were found to be significantly different in Rn-M11. The differences in SOM percentages for different sampling locations might be due to the different types of soils used to cover the deposit and the topography. Soil texture affects the soils' water holding capacity. The run-offs transport the SOM down the slope of the deposit, and the bottom location is favored by wet conditions as compared to the samples which are on the mid-slope and on top of the deposit.

4.4.2. Correlation matrices and comparison of radionuclides, PTE pseudo total, and physicochemical properties associations

Pearson's correlation coefficients of the radionuclides, total PTEs, and chemical properties of the soil were calculated from the concentrations of each sample to establish inter-relationships of these elements and chemical properties in the soil samples. The results of the correlation matrix are presented in (Table 14.), and the visual presentation of the correlation is shown in (Appendix A16.) The outcome of the correlation relationship conducted was used to prove the null hypothesis which states that the correlation coefficient is not significantly different from zero. When the *r*-value is close to 1, it is considered that the two variables have a strong relationship; and $p \leq 0.05$ indicate that there is sufficient evidence to conclude that the correlation coefficient is significantly different from zero and therefore reject the null hypothesis (Althouse and Soman, 2017; Statstutor, 2015). To describe the correlation between the two variables, the following terms were used: weak correlation for $r = 0.20$ to 0.40, moderate correlation for $r = 0.41$ to 0.60 and strong correlation for $r > 0.60$ (Gulan *et al.*, 2017).

H_0: null hypothesis – the Pearson's correlation coefficient (*r*) is not significantly different from 0; the results are due to chance; and **H_1: alternative hypothesis** – the Pearson's correlation coefficient (*r*) is significantly different from 0; the *r*-value is closer to 1.

Table 14. The correlation matrix (r) for the concentrations of the natural radionuclides, total PTEs, and the chemical properties of the soil

Pearsons' (*r*)	U-238	Th-232	U-235	K-40	Rn-222	Ca	Co	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	P	Pb	U	Zn	CEC	pH	% Soil moisture	% SOM
U-238	-																						
Th-232	**-0.65**	-																					
U-235	**1.00**	**-0.65**	-																				
K-40	0.13	0.31	0.13	-																			
Rn-222	**0.73**	-0.35	**0.73**	0.31	-																		
Ca	-0.30	-0.06	-0.30	-0.42	**-0.54**	-																	
Co	0.05	0.45	0.05	**0.75**	0.47	**-0.57**	-																
Cr	**0.69**	-0.43	**0.69**	0.16	**0.62**	**-0.51**	0.33	-															
Cu	**0.97**	**-0.54**	**0.97**	0.29	**0.74**	-0.30	0.18	**0.67**	-														
Fe	**-0.55**	**0.78**	**-0.55**	**0.57**	-0.05	-0.22	**0.79**	-0.23	-0.42	-													
K	0.26	0.22	0.26	**0.84**	0.40	**-0.66**	**0.80**	0.27	0.38	**0.53**	-												
Mg	-0.19	0.32	-0.19	0.34	-0.15	**0.56**	0.30	-0.34	-0.08	0.48	0.17	-											
Mn	-0.33	**0.69**	-0.33	**0.60**	0.18	**-0.57**	**0.86**	0.05	-0.22	**0.88**	**0.62**	0.10	-										
Na	**-0.81**	**0.79**	**-0.81**	0.19	-0.31	0.13	0.37	**-0.61**	**-0.70**	**0.84**	0.09	0.45	**0.62**	-									
Ni	0.39	-0.22	0.39	0.12	0.45	-0.50	0.39	**0.94**	0.38	-0.04	0.20	-0.35	0.23	-0.39	-								
P	**-0.59**	**0.80**	**-0.59**	**0.54**	-0.11	-0.11	**0.70**	-0.38	-0.44	**0.98**	0.48	**0.54**	**0.80**	**0.89**	-0.20	-							
Pb	**0.97**	-0.52	**0.97**	0.32	**0.80**	-0.42	0.26	**0.70**	**0.98**	-0.36	0.46	-0.12	-0.14	**-0.69**	0.42	-0.41	-						
U	**1.00**	**-0.65**	**1.00**	0.13	**0.73**	-0.30	0.05	**0.69**	**0.97**	**-0.55**	0.26	-0.20	-0.33	**-0.81**	0.39	**-0.59**	**0.97**	-					
Zn	-0.20	0.49	-0.20	**0.68**	-0.12	-0.05	**0.50**	-0.19	0.04	**0.60**	**0.58**	**0.50**	0.47	0.43	-0.16	**0.66**	-0.03	-0.20	-				
CEC	-0.03	0.05	-0.03	0.02	0.00	-0.11	-0.01	-0.32	-0.12	0.07	0.17	0.03	-0.01	0.18	-0.42	0.11	-0.01	-0.03	-0.25	-			
pH	-0.28	-0.05	-0.28	-0.44	**-0.60**	0.11	**-0.71**	-0.39	-0.37	-0.41	-0.36	-0.45	-0.46	-0.13	-0.38	-0.35	-0.38	-0.28	-0.26	0.28	-		
% Soil moisture	**-0.57**	**0.71**	**-0.57**	**0.50**	-0.30	-0.13	**0.51**	-0.40	-0.39	**0.81**	0.46	0.38	**0.64**	**0.74**	-0.25	**0.86**	-0.39	**-0.57**	**0.81**	0.08	-0.10	-	
% SOM	-0.11	0.38	-0.11	**0.78**	-0.15	-0.27	**0.50**	-0.12	0.07	**0.53**	**0.78**	0.33	0.46	0.26	-0.13	**0.55**	0.07	-0.11	**0.86**	0.08	-0.04	**0.75**	-

Note: SM is the soil moisture, and the values highlighted in **bold** are Pearson's correlation coefficient (*r*) significant at 95 % confidence level; $p \leq 0.0$

The observations for the physicochemical properties of the soil indicated:

A weak correlation between the pH and the radionuclides with the range $-0.03 > r < 0.05$ at $p > 0.05$.

Similarly, there was a weak correlation between the pH and the PTEs where the range was $-0.25 > r < 0.18$ at $p > 0.05$ except in the case of the pH – Ni pair where the moderate negative correlation was observed ($r = -0.42$).

On the other hand, the % SM demonstrated strong correlations with Th-232, Fe, P, Mn, Na, and Zn at ($p \leq 0.05$).

The strong correlation between soil moisture and these elements could predict their behavior in soil.

Additionally, the results indicated a weak positive correlation between the pairs pH – % SM and CEC – pH; however, the pair CEC – % SM showed a weak negative correlation. The results indicated a strong correlation between the % SOM and the % SM ($r = 0.75$; $p \leq 0.05$). Additionally, % SOM had a strong correlation ($p \leq 0.05$) with Zn ($r = 0.86$), K-40 ($r = 0.78$), and K ($r = 0.78$); and a moderate correlation ($p \leq 0.05$) with Fe (r = 0.53), P ($r = 0.55$), Co ($r = 0.50$), and Mn ($r = 0.46$). However, the % SOM was observed to have a weak relationship with the radionuclides, the pH, and the CEC. It could be noted that the SOM had a significant relationship ($p \leq 0.05$) with the macronutrients. This confirms that when the soil contains a higher % SOM, it is likely to contain a large number of nutrients. The results of this study indicated that the % SOM and % plant moisture had a negative correlation ($r = - 0.923$) which was significant at $p \leq 0.05$. This could be noted in (Figure 12. e) where the trend of these two variables is inversely proportional.

The correlation matrix between radionuclides and PTEs indicated a significant relationship ($p \leq 0.05$) between the following pairs:

- U-238 – U-235 ($r = 1.00$), U-238 – Rn-222 ($r = 0.73$) and U-238 – Cu ($r = 0.97$)
- Th-232 – Fe ($r = 0.78$), Th-232 – P ($r = 0.80$), Th-232 – Na ($r = 0.79$), and Th-232 – Mn ($r = 0.69$).
- U-235 – Rn-222 ($r = 0.73$) and U-235 – Cu ($r = 0.97$)
- K-40 – K ($r = 0.84$) and K-40 – Co ($r = 0.75$), K-40 – Mn ($r = 0.60$), K-40 – Zn ($r = 0.68$), K-40 – Fe ($r = 0.57$), and K-40 – P ($r = 0.54$).

The correlation between U-238 and U-235 could be because these isotopes are both primary isotopes of uranium, and the correlation between U-238 and Rn-222 could be because Rn-222 is a progeny of U-238. A strong positive correlation between these radionuclides and PTEs suggests their common origin (Bai *et al.*, 2017). Whereas the observations among PTEs resulted in the strong significant correlation matrices ($r > 0.60$; $p \leq 0.05$) between the following pairs:

- Fe – P, Fe – Co, Fe – Mn, Fe – Na and Fe – Zn and Zn
- K – Co and K – Mn
- P – Co, P – Zn, P – Mn, and P – Na
- Co – Mn
- Mn – Na

It could be noted that Co and Mn were common in these correlation matrices. According to Gulan *et al.* (2017), Co is widely dispersed in the Earth's crust, and its correlation with Mn shows that the occurrence in the soils is mostly due to similar sources.

4.4.3. Principal component analysis for PTE pseudo total, radionuclides, and soil characteristics

The correlation matrix for the PTEs in soil, radionuclides in soil, and soil characteristics were confirmed by the PCA correlation biplot in (Figure 23.). The correlation biplot indicated that most of the elements contribute to the positive side of the principal component two (PC 1) and the negative side of the PC 2. The radioactive elements (U-238, U-235, and Rn-222) indicated a strong correlation with the PTEs (U, Ni, Pb, Cu, and Cr), which were all contributing to the negative side of the PC 1 and PC 2. It could be observed that the CEC and the pH were diagonally opposite the U-238, U-235, and Rn-222, including all the PTEs that are correlated with these elements. This observation emphasizes that the diagonally opposite variables are negatively correlated; in this case, it can be pointed out that to minimize the mobility of these radionuclides, the pH and CEC should increase. Furthermore, Ca was observed to be strongly associated with the pH. According to Panhwar *et al.* (2016), when the Ca content increases in the soil, the pH will also increase. The CEC vector is short due to the large variance in the data.

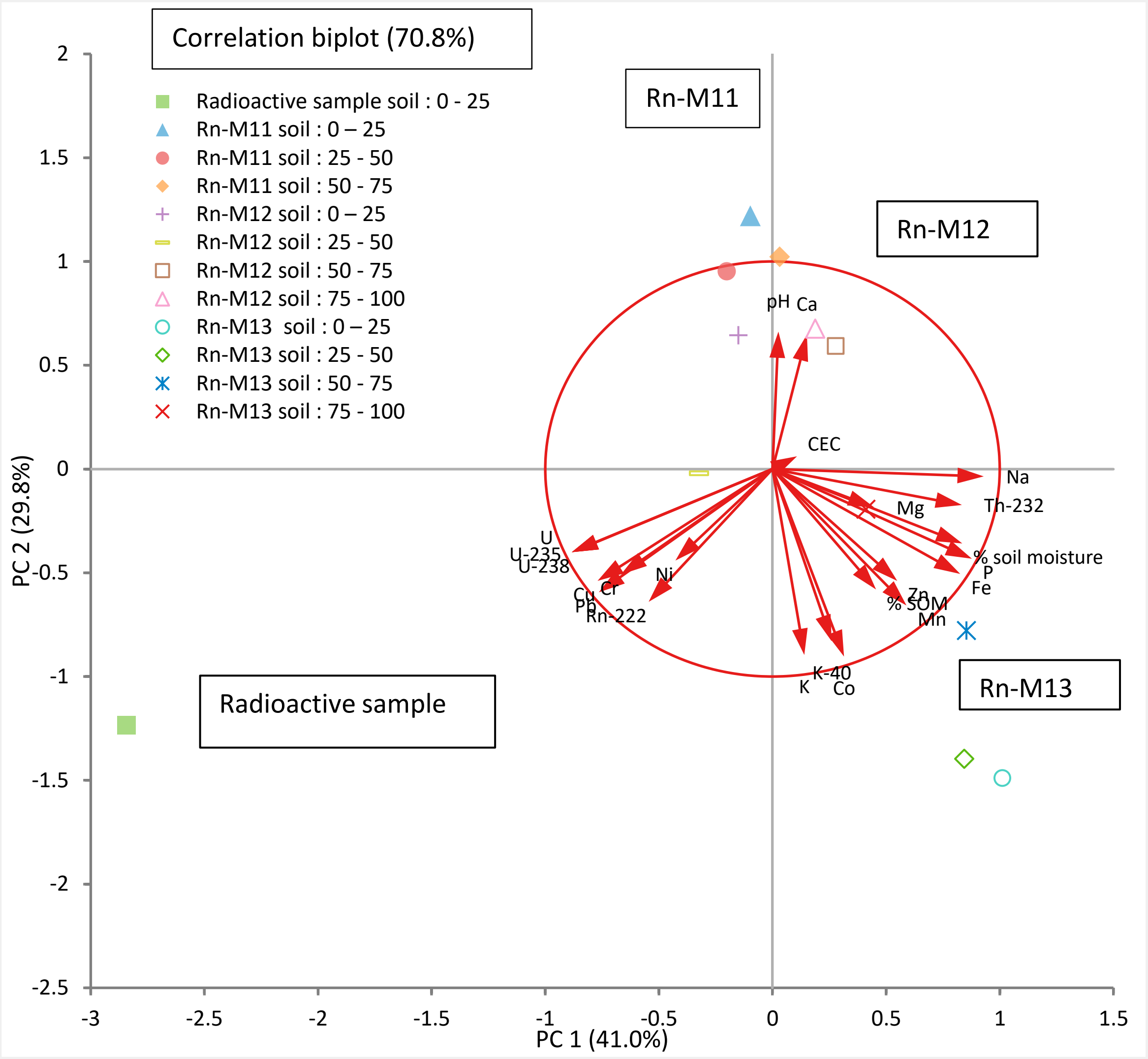


Figure 23. Principal component analysis plot for the soil characteristics, radionuclides, and PTEs in soil

The results of the correlation biplot are displayed in (Table 15.) and (Table 16.). (Table 15.) shows that PC 1 contributed 41.0 % while PC 2 contributed 29.8 % to the proportion. In (Table 16), PC 1 has a strong positive association with Th-232, Fe, Na, P, and % soil moisture. Whereas the PC 2 has a strong negative association with K-40, Co, and K.

The PCA correlation biplot for the elements in plants is shown in (Figure 24.), and the component proportions, as well as the coefficient distributions, are presented in (Appendix A17.). It was observed that the larger percentage was negatively associated with PC 1 and positively associated with PC 2 with a large percentage associated with the Rn-M11 sample. The Rn-M12 had an association with the radioactive elements (U-238, U-235, Th-232), and U. Uranium-238, U-235, and U were all negatively correlated with P in plants

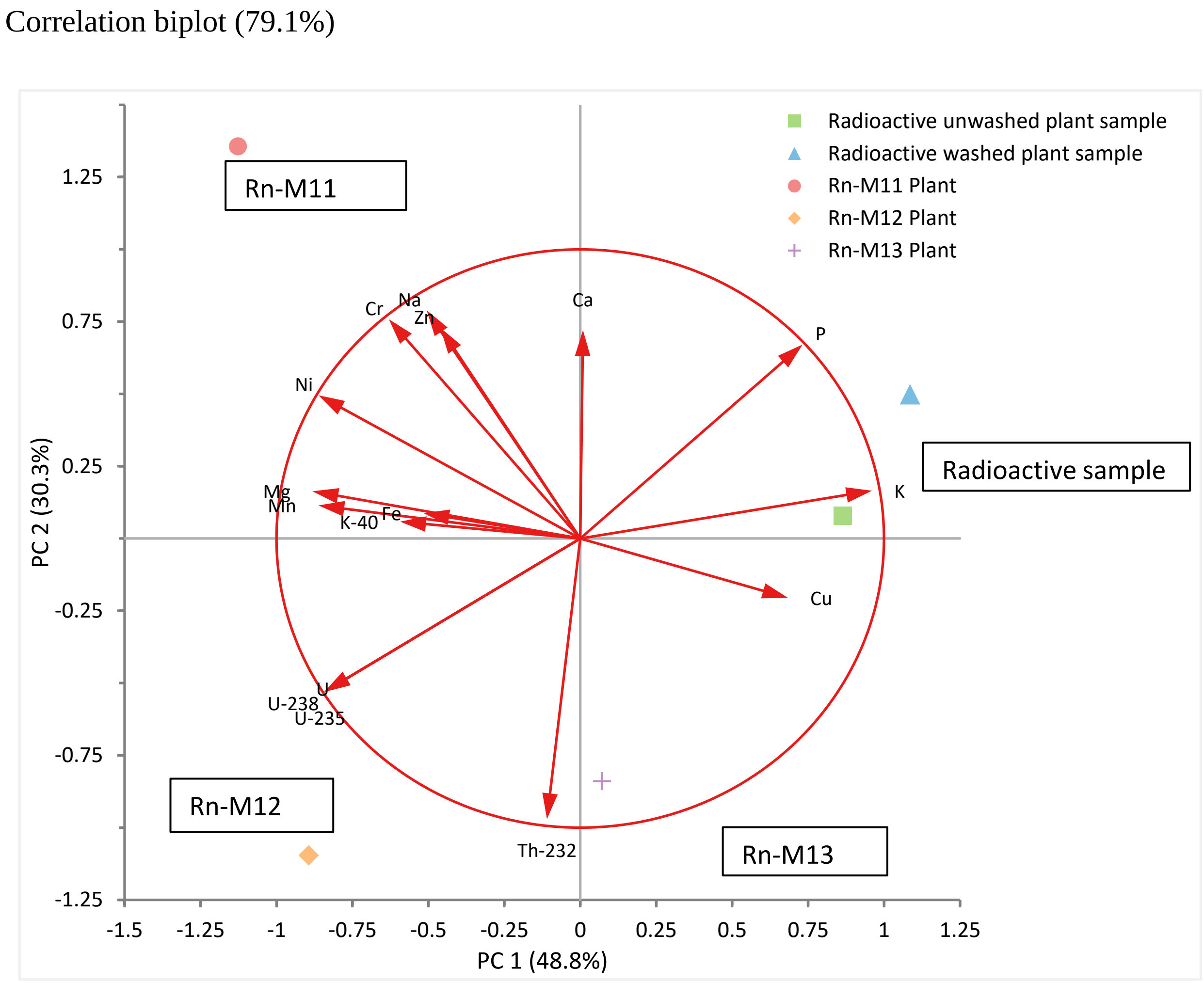
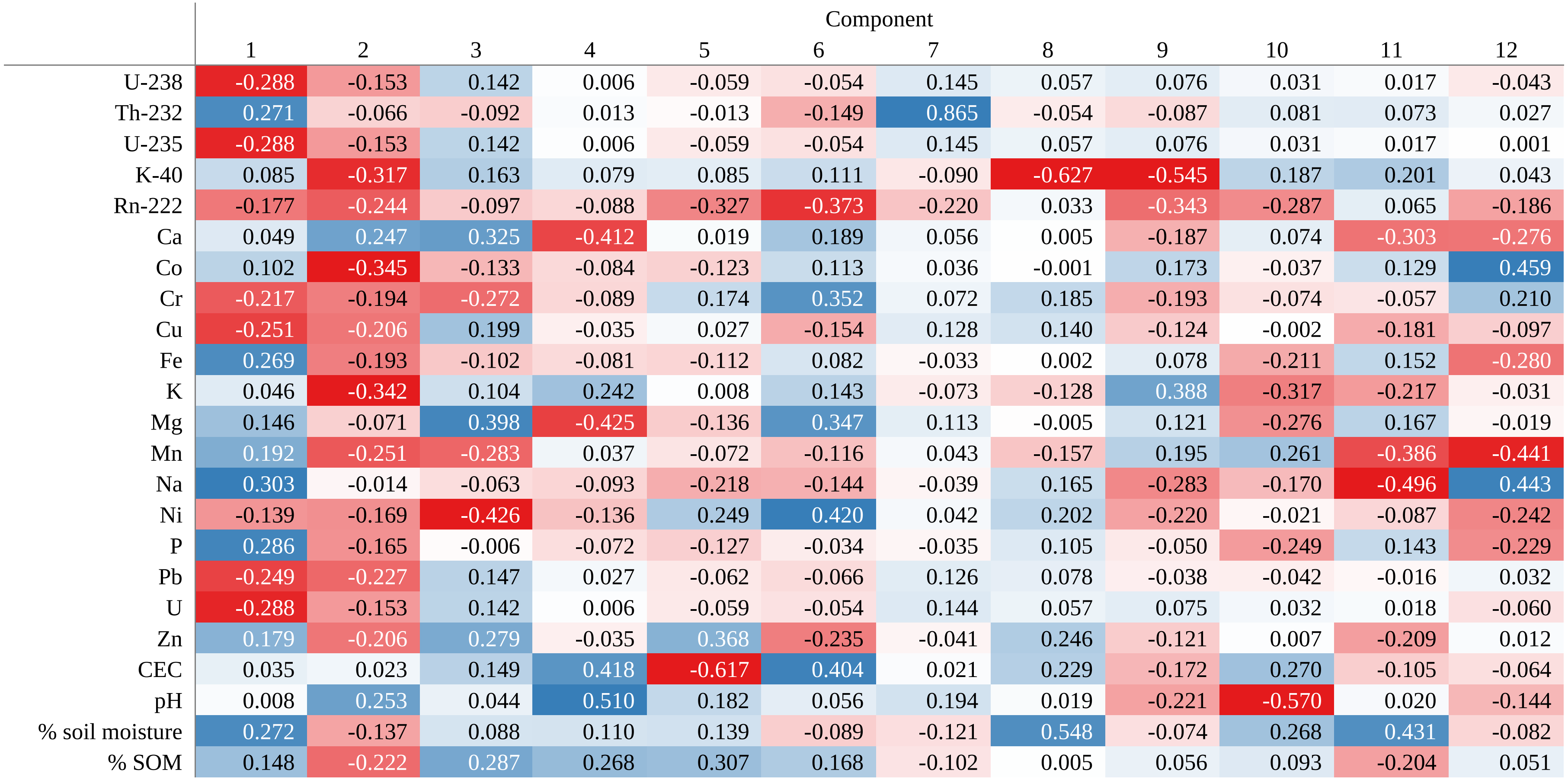


Figure 24. Principle Component Analysis plot for radionuclides and PTEs in plants

Table 15. Principal component analysis for soil characteristics, radionuclides, and PTEs in soil

Component	Variance	Proportion	Cumulative proportion
1	9.429	0.410	0.410
2	6.854	0.298	0.708
3	2.189	0.095	0.803
4	1.748	0.076	0.879
5	1.365	0.059	0.938
6	0.510	0.022	0.961
7	0.324	0.014	0.975
8	0.275	0.012	0.987
9	0.142	0.006	0.993
10	0.111	0.005	0.998
11	0.052	0.002	1.000
12	0.000	0.000	1.000

Table 16. Coefficients for radionuclides, PTEs, and soil characteristics in soil



	Component											
	1	2	3	4	5	6	7	8	9	10	11	12
U-238	-0.288	-0.153	0.142	0.006	-0.059	-0.054	0.145	0.057	0.076	0.031	0.017	-0.043
Th-232	0.271	-0.066	-0.092	0.013	-0.013	-0.149	0.865	-0.054	-0.087	0.081	0.073	0.027
U-235	-0.288	-0.153	0.142	0.006	-0.059	-0.054	0.145	0.057	0.076	0.031	0.017	0.001
K-40	0.085	-0.317	0.163	0.079	0.085	0.111	-0.090	-0.627	-0.545	0.187	0.201	0.043
Rn-222	-0.177	-0.244	-0.097	-0.088	-0.327	-0.373	-0.220	0.033	-0.343	-0.287	0.065	-0.186
Ca	0.049	0.247	0.325	-0.412	0.019	0.189	0.056	0.005	-0.187	0.074	-0.303	-0.276
Co	0.102	-0.345	-0.133	-0.084	-0.123	0.113	0.036	-0.001	0.173	-0.037	0.129	0.459
Cr	-0.217	-0.194	-0.272	-0.089	0.174	0.352	0.072	0.185	-0.193	-0.074	-0.057	0.210
Cu	-0.251	-0.206	0.199	-0.035	0.027	-0.154	0.128	0.140	-0.124	-0.002	-0.181	-0.097
Fe	0.269	-0.193	-0.102	-0.081	-0.112	0.082	-0.033	0.002	0.078	-0.211	0.152	-0.280
K	0.046	-0.342	0.104	0.242	0.008	0.143	-0.073	-0.128	0.388	-0.317	-0.217	-0.031
Mg	0.146	-0.071	0.398	-0.425	-0.136	0.347	0.113	-0.005	0.121	-0.276	0.167	-0.019
Mn	0.192	-0.251	-0.283	0.037	-0.072	-0.116	0.043	-0.157	0.195	0.261	-0.386	-0.441
Na	0.303	-0.014	-0.063	-0.093	-0.218	-0.144	-0.039	0.165	-0.283	-0.170	-0.496	0.443
Ni	-0.139	-0.169	-0.426	-0.136	0.249	0.420	0.042	0.202	-0.220	-0.021	-0.087	-0.242
P	0.286	-0.165	-0.006	-0.072	-0.127	-0.034	-0.035	0.105	-0.050	-0.249	0.143	-0.229
Pb	-0.249	-0.227	0.147	0.027	-0.062	-0.066	0.126	0.078	-0.038	-0.042	-0.016	0.032
U	-0.288	-0.153	0.142	0.006	-0.059	-0.054	0.144	0.057	0.075	0.032	0.018	-0.060
Zn	0.179	-0.206	0.279	-0.035	0.368	-0.235	-0.041	0.246	-0.121	0.007	-0.209	0.012
CEC	0.035	0.023	0.149	0.418	-0.617	0.404	0.021	0.229	-0.172	0.270	-0.105	-0.064
pH	0.008	0.253	0.044	0.510	0.182	0.056	0.194	0.019	-0.221	-0.570	0.020	-0.144
% soil moisture	0.272	-0.137	0.088	0.110	0.139	-0.089	-0.121	0.548	-0.074	0.268	0.431	-0.082
% SOM	0.148	-0.222	0.287	0.268	0.307	0.168	-0.102	0.005	0.056	0.093	-0.204	0.051

The correlation matrix indicates the correlation coefficient for each pair of variables. Positively correlated variables are blue, and negatively correlated variables are red, with the intensity-dependent on the magnitude of the correlation.

4.4.4. The comparison of the pseudo total concentrations of each element in terms of topography and depths

The comparison for each element concentration in terms of topography was made. In this comparison, only three samples (Rn-M11, Rn-M12, and Rn-M13) were taken into consideration. The radioactive sample was excluded since it was collected from the edge of the deposit and mostly contained outliers in terms of concentrations.

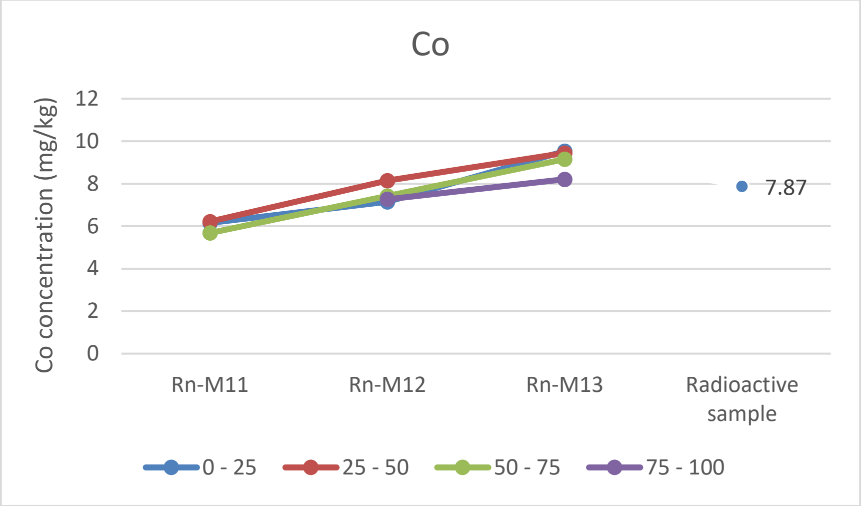
Before conducting the One-Sample t-test statistical analysis, the concentrations for each element were plotted, comparing each element for different samples in the same depth (Figure 25.) (e.g., comparing Co in 0 – 25 cm for Rn-M11, 0 – 25 cm for Rn-M12, and 0 – 25 cm in Rn-M13 to see if there are differences in concentrations of Co as the slope decreases). This comparison was made to give an indication of the behavioral tendencies for each element in the same depth but in different positions of the deposit. It could be noted that U-238, U-235, and Pb had similar trends. Additionally, Cr and Ni also indicated similar behavioral patterns. Likewise, Co, Fe, and P also showed similar behavioral patterns. There was a noticeable increase in tendencies for Co, Mn, Fe, P, and Rn-222 elements as the topographic conditions change.

To determine if these visuals were statistically correct, a One-Sample t-test analysis was conducted for each element in Rn-M11, Rn-M12, and Rn-M13 considering 0 – 25 cm depth (Table 17.), 25 – 50 cm depth (Table 18.), and 50 – 75 cm depth (Table 19.). The t-test statistical analysis results for 0 – 25 cm depth for all four samples, including the Radioactive sample, are presented in (Appendix A26.) The null hypothesis for the t-test was a follows:

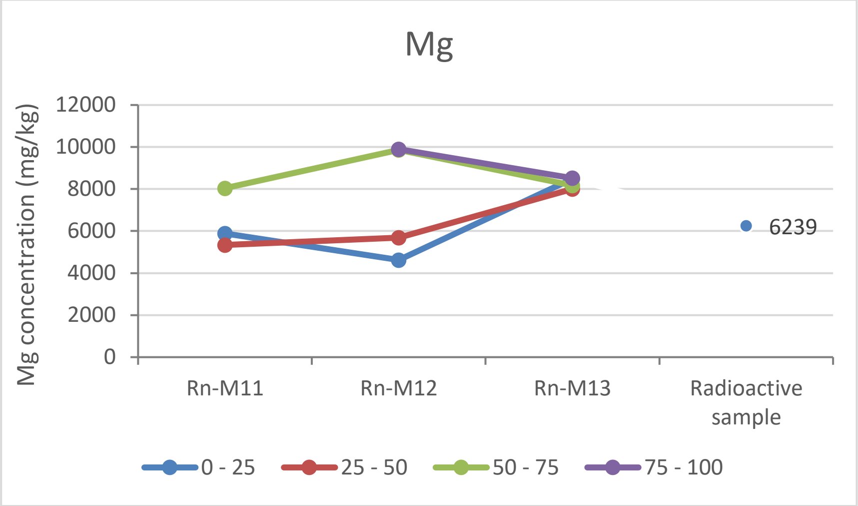
H_0: The concentrations of each element are <u>not equal</u> as the topographic locations of the deposit change. If $p \leq 0.05$, the null hypothesis is rejected.

In 0 – 25 cm depth (Table 17.), it was observed that the Rn-222, Ca, K, and Zn concentrations were significantly different. Whereas, in 0 – 25 cm depth (Appendix A26.) which includes the Radioactive sample, indicated that U-238, U-235, Rn-222, U, Pb, and Cu were significantly different in different sampling locations. These results confirm that the majority of the concentrations of these elements come from the Radioactive sample. A similar observation was made in 25 – 50 cm depth (Table 18.) and 50 – 75 cm depth (Table 19.) where Cr and Ni; and Rn-222 and Ca were all significantly different, respectively. These observations suggest that as the slope changes, the concentrations of these elements will significantly change. In this case, it is apparent that the water movement down the slope moves some elements in the water direction.

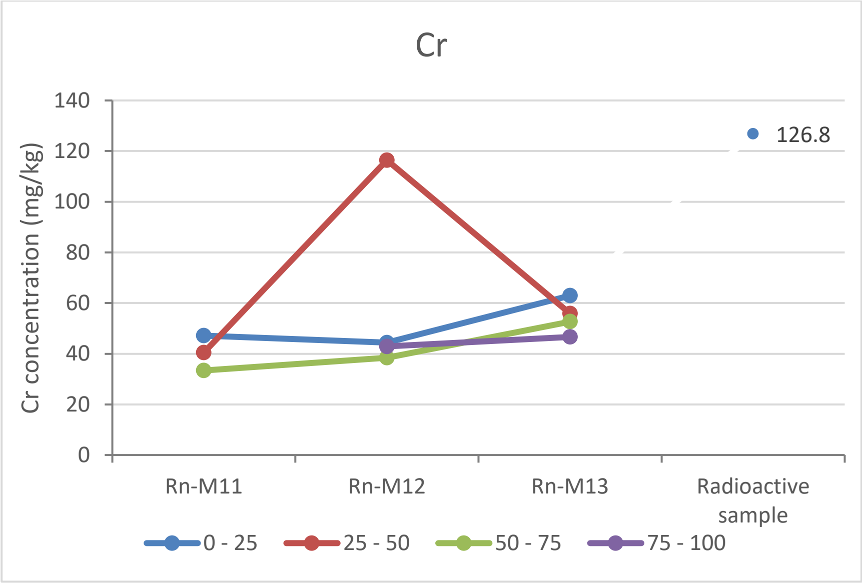
One-Sample t-test statistical analysis was also conducted to determine if there are differences in concentrations between the depths of each sample. The t-test results for Rn-M11 are shown in (Appendix 18.), Rn-M12 are shown in (Appendix 19.), and Rn-M13 are shown in (Appendix A20.). The results indicated that only the Rn-M11 sample had elements that were significantly different as the depth deepens. In this sampling location, U-238, U-235, Rn-222, and Ca were significantly different. These results suggest that the insufficient cover layer affects the concentrations of these elements from the spoil rock to the top layer of the deposit. If the soil cover layer is not sufficient, these elements can easily become mobile.


Co
Co concentration (mg/kg)
12
10
8
6
4
2
0
7.87
Rn-M11
Rn-M12
Rn-M13
Radioactive sample
0 - 25
25 - 50
50 - 75
75 - 100

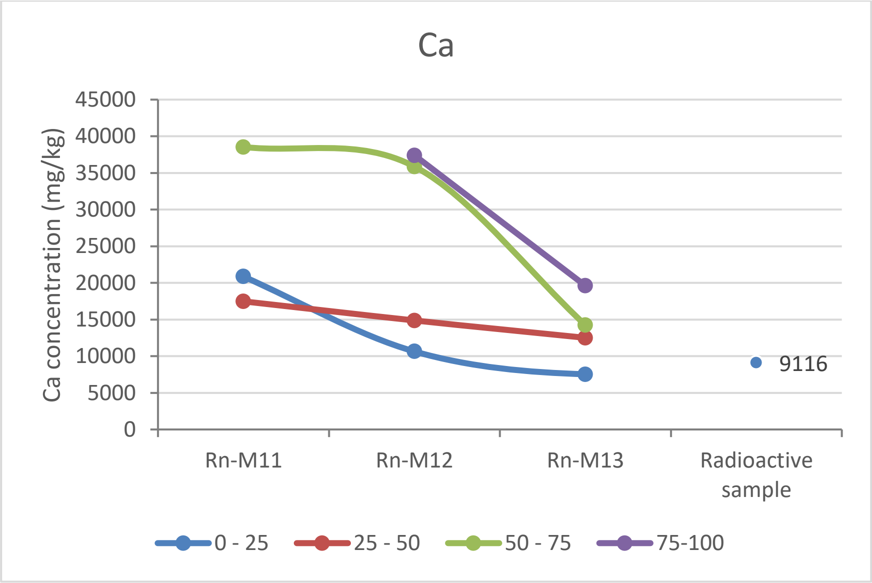
a)


Mg
Mg concentration (mg/kg)
12000
10000
8000
6000
4000
2000
0
6239
Rn-M11
Rn-M12
Rn-M13
Radioactive sample
0 - 25
25 - 50
50 - 75
75 - 100

b)

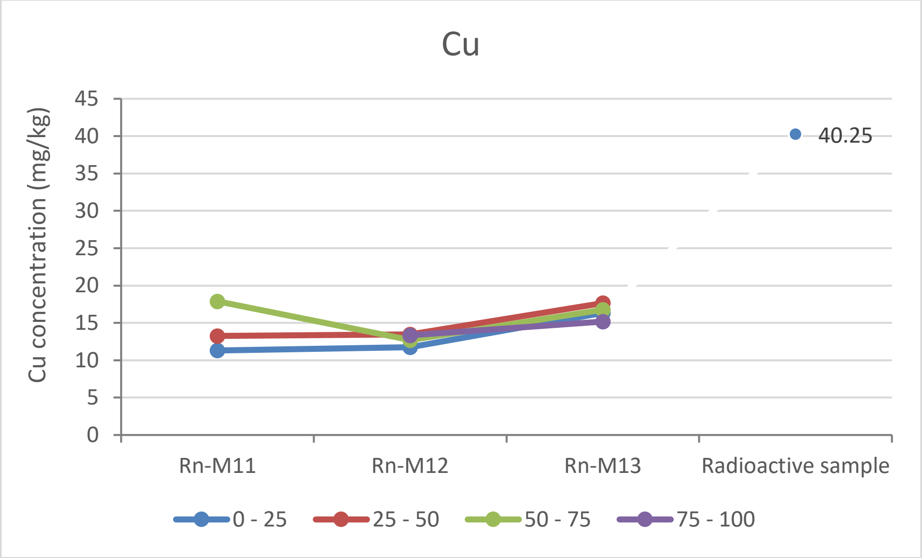

Cr
Cr concentration (mg/kg)
140
120
100
80
60
40
20
0
126.8
Rn-M11
Rn-M12
Rn-M13
Radioactive sample
0 - 25
25 - 50
50 - 75
75 - 100

c)

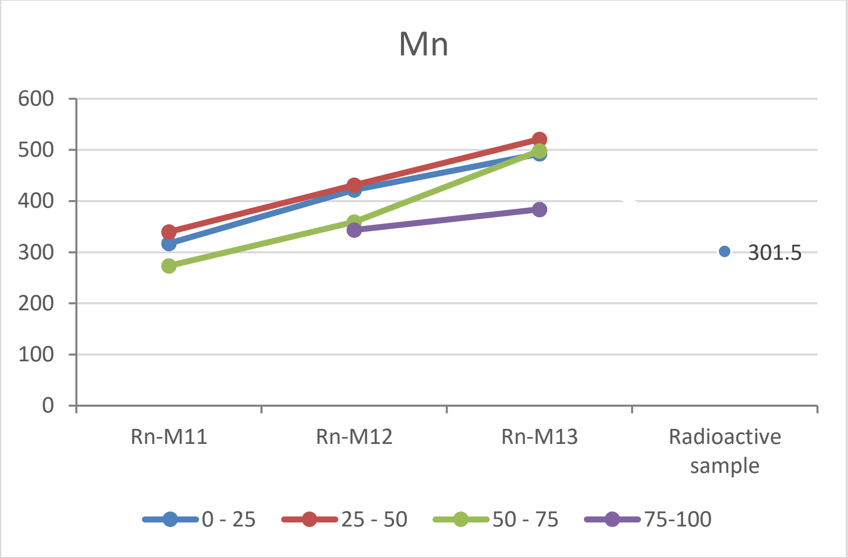

Ca
Ca concentration (mg/kg)
45000
40000
35000
30000
25000
20000
15000
10000
5000
0
9116
Rn-M11
Rn-M12
Rn-M13
Radioactive sample
0 - 25
25 - 50
50 - 75
75-100

d)

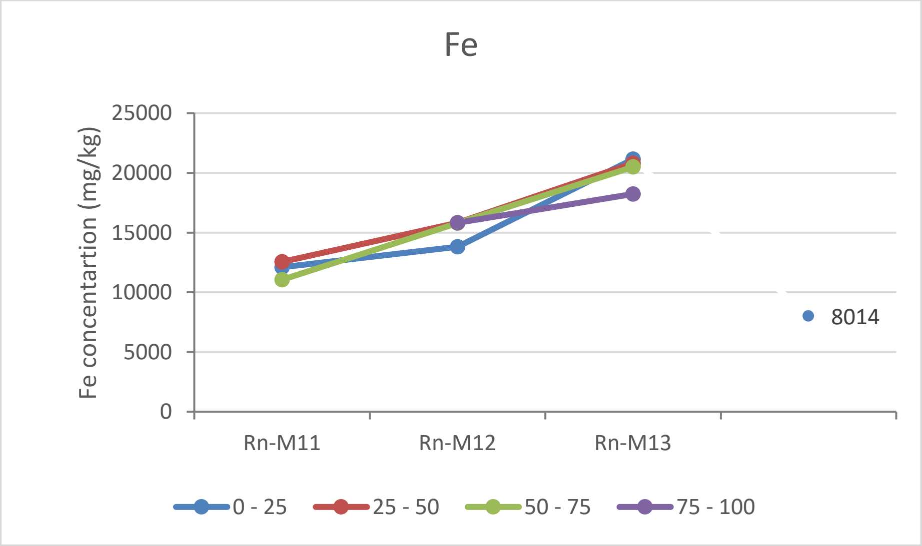
e)


Cu
Cu concentration (mg/kg)
45
40
35
30
25
20
15
10
5
0
Rn-M11
Rn-M12
Rn-M13
Radioactive sample
40.25
0 - 25
25 - 50
50 - 75
75 - 100

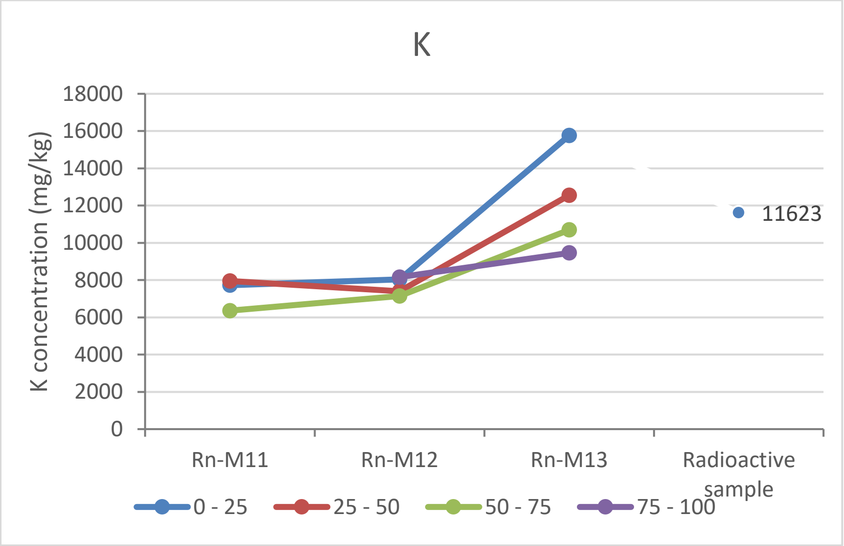
f)

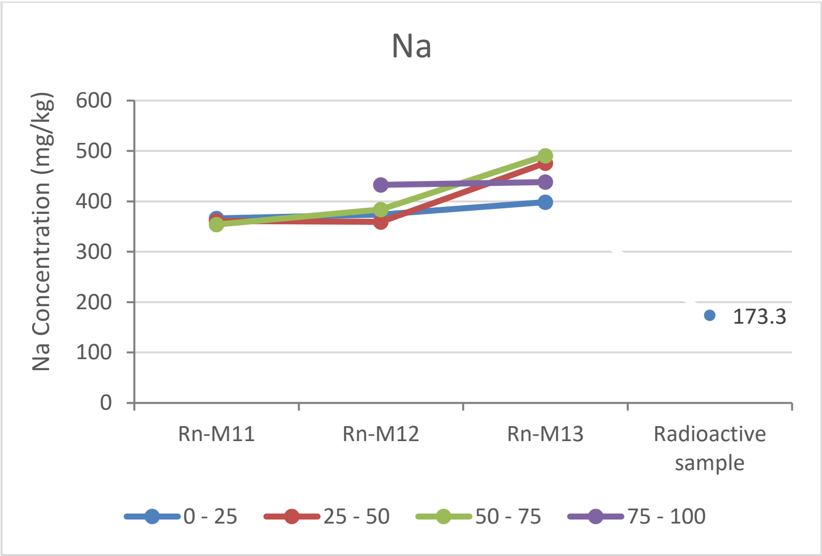

Mn
600
500
400
300
200
100
0
Rn-M11
Rn-M12
Rn-M13
Radioactive sample
301.5
0 - 25
25 - 50
50 - 75
75-100

g)

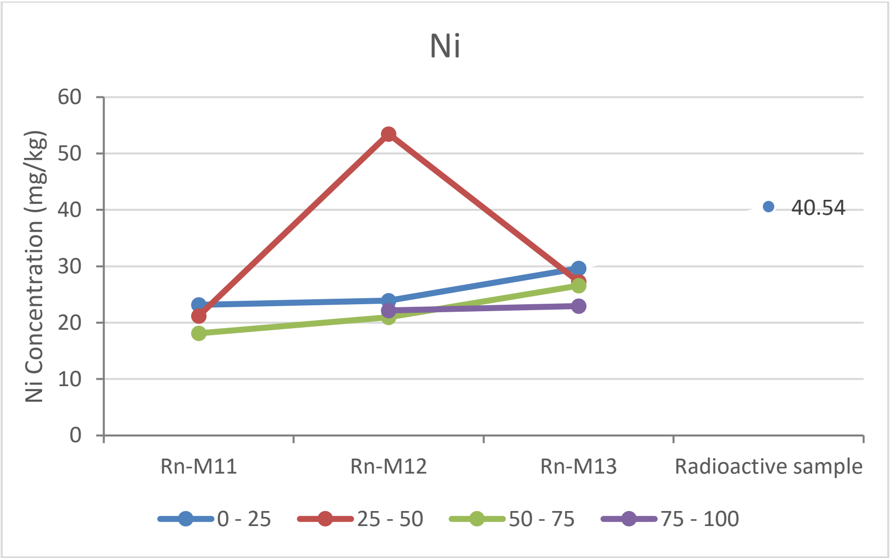

Fe
Fe concentartion (mg/kg)
25000
20000
15000
10000
5000
0
Rn-M11
Rn-M12
Rn-M13
8014
0 - 25
25 - 50
50 - 75
75 - 100

h)

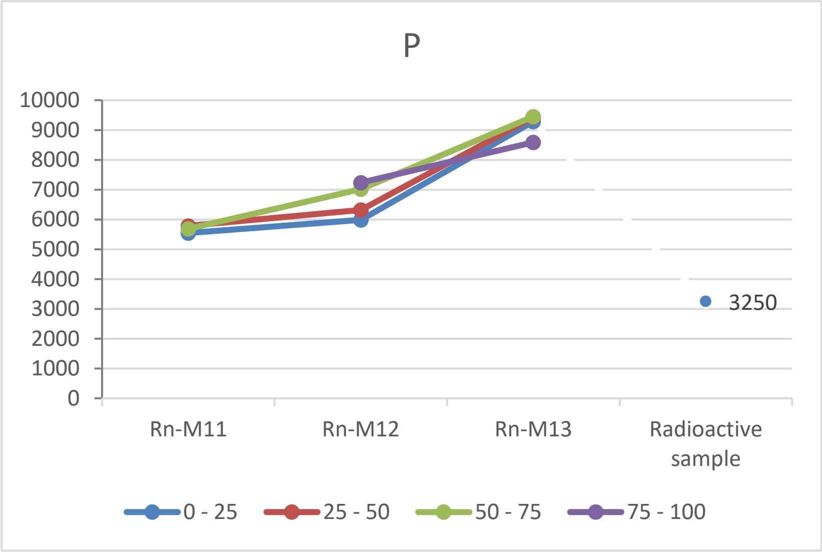

K
K concentration (mg/kg)
18000
16000
14000
12000
10000
8000
6000
4000
2000
0
Rn-M11
Rn-M12
Rn-M13
Radioactive sample
11623
0 - 25
25 - 50
50 - 75
75 - 100


Na
Na Concentration (mg/kg)
600
500
400
300
200
100
0
173.3
Rn-M11
Rn-M12
Rn-M13
Radioactive sample
0 - 25
25 - 50
50 - 75
75 - 100

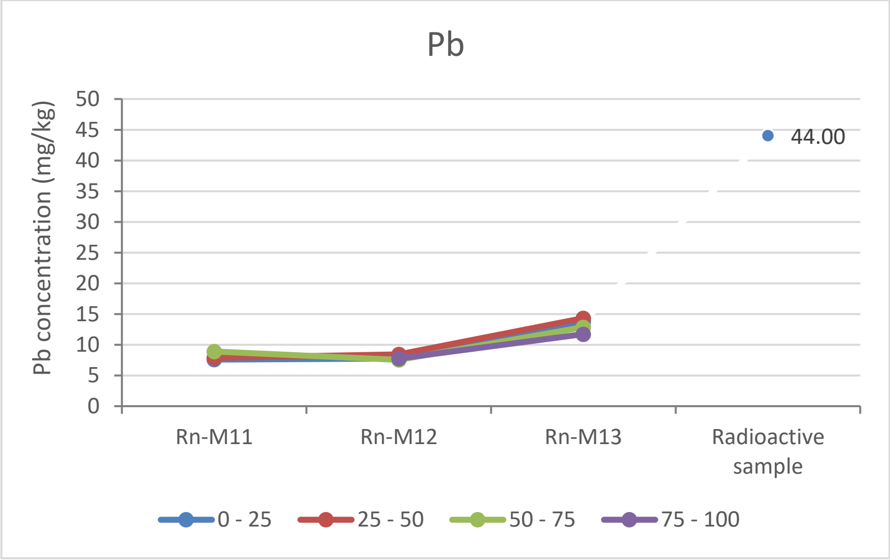
i)


Ni
Ni Concentration (mg/kg)
60
50
40
30
20
10
0
40.54
Rn-M11
Rn-M12
Rn-M13
Radioactive sample
0 - 25
25 - 50
50 - 75
75 - 100

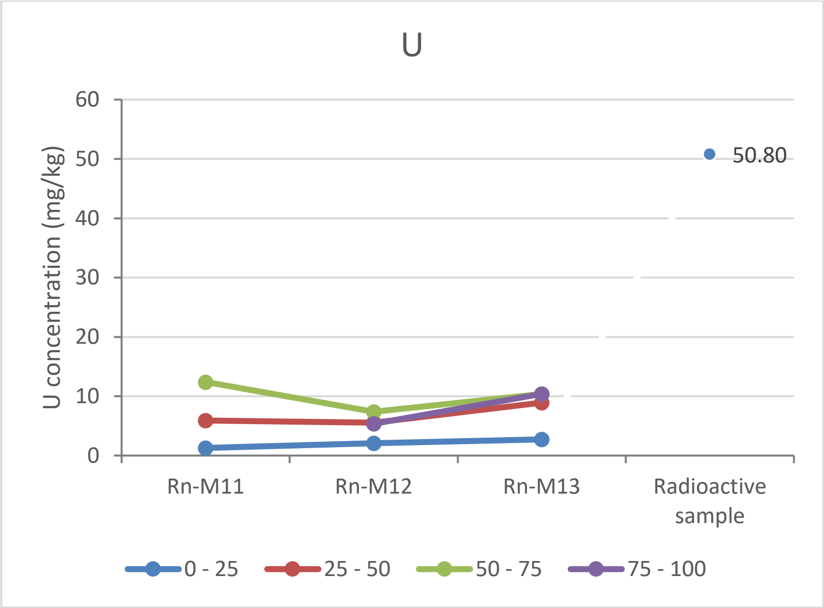
j)


P
10000
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
3250
Rn-M11
Rn-M12
Rn-M13
Radioactive sample
0 - 25
25 - 50
50 - 75
75 - 100

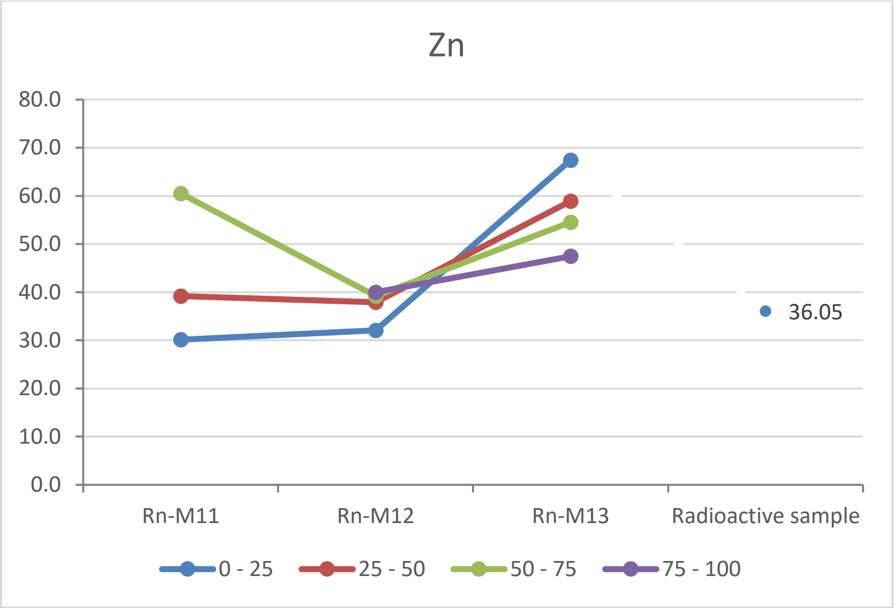
K)


Pb
Pb concentration (mg/kg)
50
45
40
35
30
25
20
15
10
5
0
44.00
Rn-M11
Rn-M12
Rn-M13
Radioactive sample
0 - 25
25 - 50
50 - 75
75 - 100

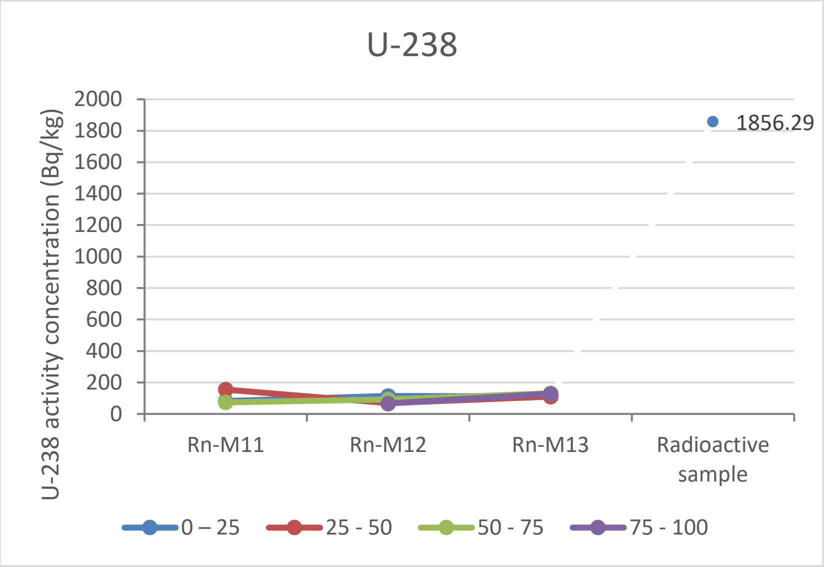
l)


U
U concentration (mg/kg)
60
50
40
30
20
10
0
50.80
Rn-M11
Rn-M12
Rn-M13
Radioactive sample
0 - 25
25 - 50
50 - 75
75 - 100

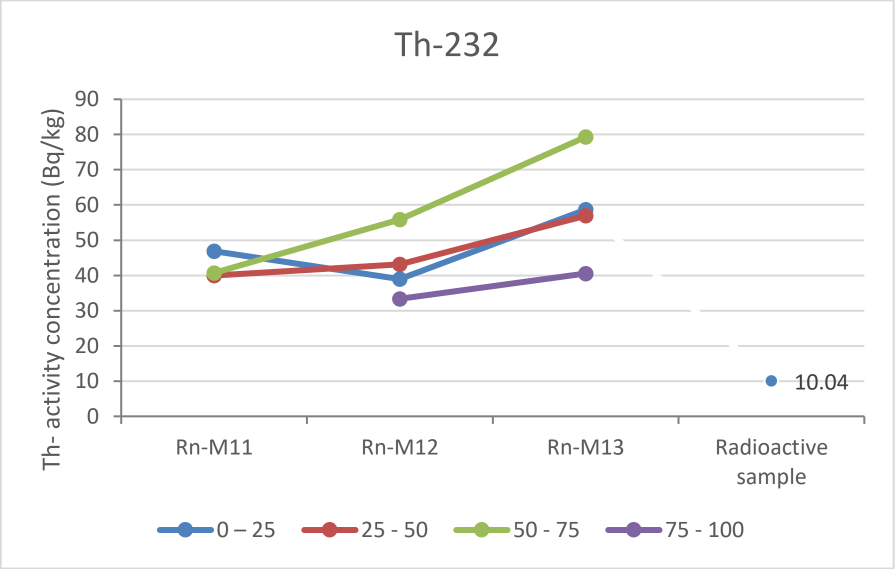
m)


Zn
80.0
70.0
60.0
50.0
40.0
30.0
20.0
10.0
0.0
36.05
Rn-M11
Rn-M12
Rn-M13
Radioactive sample
0 - 25
25 - 50
50 - 75
75 - 100

n)


U-238
U-238 activity concentration (Bq/kg)
2000
1800
1600
1400
1200
1000
800
600
400
200
0
1856.29
Rn-M11
Rn-M12
Rn-M13
Radioactive sample
0 – 25
25 - 50
50 - 75
75 - 100

o)


Th-232
Th- activity concentration (Bq/kg)
90
80
70
60
50
40
30
20
10
0
10.04
Rn-M11
Rn-M12
Rn-M13
Radioactive sample
0 – 25
25 - 50
50 - 75
75 - 100

p)

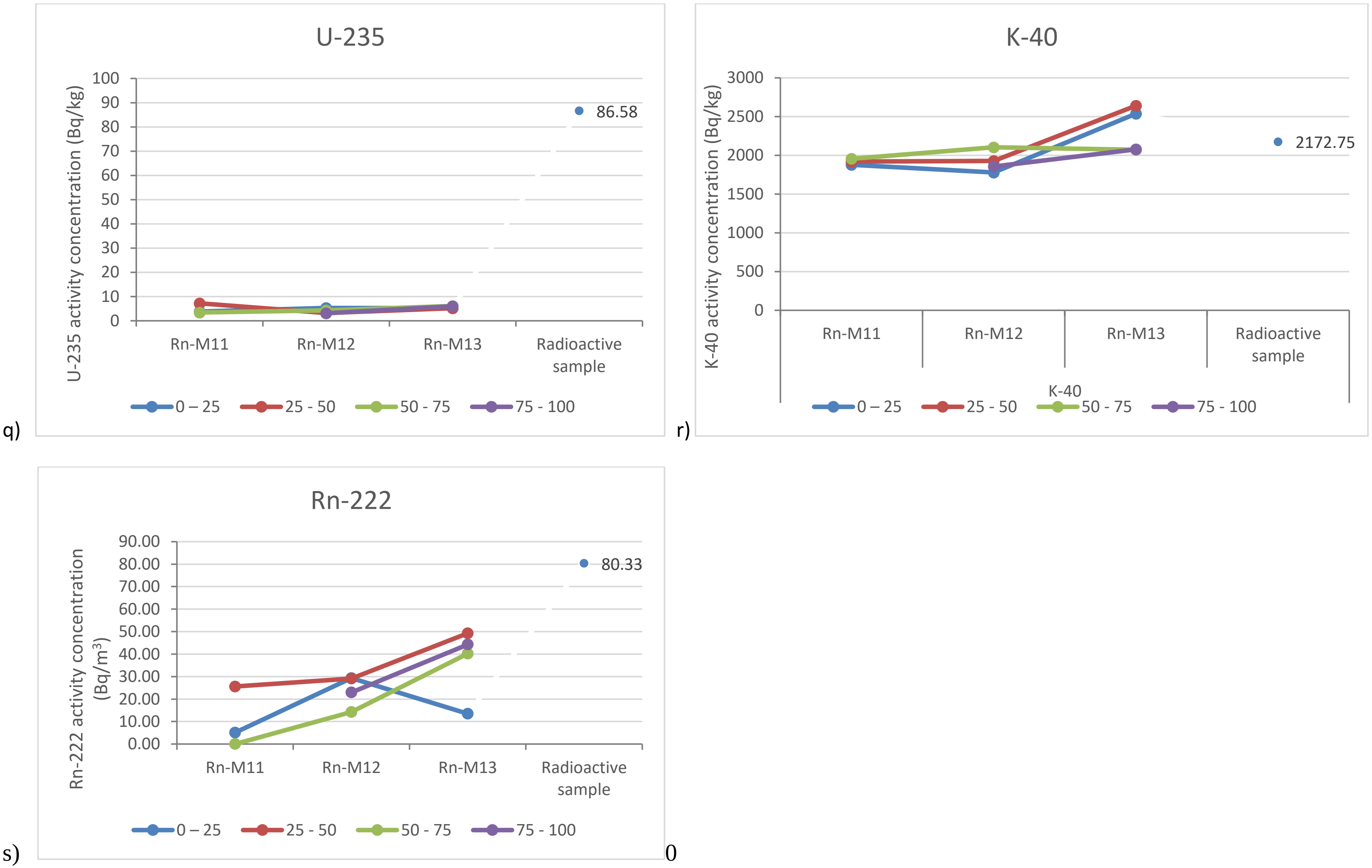


Figure 25. The comparison of the pseudo total concentrations of each element in the same depth for different soil samples

Table 17. One-Sample t-test analysis for each element in Rn-M11, Rn-M12, and Rn-M13 for 0 – 25 cm depth (function of topography)

One-Sample Statistics					**One-Sample Test**							**One-Sample Effect Sizes**			
							Significance			95% Confidence Interval of the Difference				95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean	t	df	One-Sided p	Two-Sided p	Mean Difference	Lower	Upper	Standardizer[a]	Point Estimate	Lower	Upper
U_238	3	102.367	17.615	10.170	10.065	2	0.005	0.010	102.367	58.608	146.126	17.615	5.811	0.741	11.256
Th_232	3	48.200	9.914	5.724	8.421	2	0.007	0.014	48.200	23.572	72.828	9.914	4.862	0.553	9.451
U_235	3	4.767	0.815	0.470	10.135	2	0.005	0.010	4.767	2.743	6.790	0.815	5.851	0.749	11.332
K_40	3	2065.000	412.610	238.220	8.668	2	0.007	0.013	2065.000	1040.020	3089.980	412.610	5.005	0.582	9.722
Rn_222	3	15.9600	12.29597	7.09908	2.248	2	0.077	**0.154**	15.96000	-14.5849	46.5049	12.29597	1.298	-0.385	2.878
Ca	3	13010.333	6974.659	4026.821	3.231	2	0.042	**0.084**	13010.333	-4315.679	30336.346	6974.659	1.865	-0.184	3.864
Cd	3	0.000	.00000[a]	0.000											
Co	3	7.613	1.732	1.000	7.613	2	0.008	0.017	7.613	3.311	11.916	1.732	4.395	0.456	8.567
Cr	3	51.500	10.050	5.803	8.875	2	0.006	0.012	51.500	26.533	76.467	10.050	5.124	0.606	9.949
Cu	3	13.133	2.836	1.637	8.021	2	0.008	0.015	13.133	6.088	20.179	2.836	4.631	0.506	9.013
Fe	3	15686.667	4798.911	2770.653	5.662	2	0.015	0.030	15686.667	3765.511	27607.822	4798.911	3.269	0.202	6.444
K	3	10511.000	4551.179	2627.624	4.000	2	0.029	**0.057**	10511.000	-794.755	21816.755	4551.179	2.310	-0.049	4.667
Mg	3	6333.667	1969.684	1137.198	5.570	2	0.015	0.031	6333.667	1440.700	11226.634	1969.684	3.216	0.189	6.345
Mn	3	410.333	88.081	50.854	8.069	2	0.008	0.015	410.333	191.527	629.140	88.081	4.659	0.511	9.065
Na	3	379.667	17.214	9.939	38.201	2	0.000	0.001	379.667	336.904	422.429	17.214	22.055	3.464	42.385
Ni	3	25.573	3.549	2.049	12.480	2	0.003	0.006	25.573	16.756	34.390	3.549	7.205	1.000	13.915
P	3	6939.333	2041.943	1178.916	5.886	2	0.014	0.028	6939.333	1866.865	12011.802	2041.943	3.398	0.233	6.687
Pb	3	9.673	3.488	2.014	4.803	2	0.020	0.041	9.673	1.008	18.338	3.488	2.773	0.078	5.521
U	3	2.023	0.721	0.416	4.860	2	0.020	0.040	2.023	0.232	3.815	0.721	2.806	0.086	5.581
Zn	3	43.200	20.982	12.114	3.566	2	0.035	**0.070**	43.200	-8.921	95.321	20.982	2.059	-0.123	4.212

The *p* values highlighted in **bold** are above the 0.05 significance level.

Table 18. One-Sample t-test analysis for each element in Rn-M11, Rn-M12, and Rn-M13 for 25 – 50 cm depth (function of topography)

					One-Sample Test							**One-Sample Effect Sizes**			
One-Sample Statistics							Significance			95% Confidence Interval of the Difference				95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean	t	df	One-Sided p	Two-Sided p	Mean Difference	Lower	Upper	Standardizer^{a}	Point Estimate	Lower	Upper
U_238	3	84.50	23.07	13.32	6.344	2	0.012	0.024	84.50	27.19	141.81	23.07	3.663	0.295	7.184
Th_232	3	46.93	8.72	5.04	9.321	2	0.006	0.011	46.93	25.27	68.60	8.72	5.381	0.657	10.438
U_235	3	3.94	1.08	0.62	6.322	2	0.012	0.024	3.94	1.26	6.62	1.08	3.650	0.292	7.160
K_40	3	2174.67	403.23	232.81	9.341	2	0.006	0.011	2174.67	1172.98	3176.35	403.23	5.393	0.660	10.460
Rn_222	3	34.67	12.79	7.39	4.694	2	0.021	0.043	34.67	2.89	66.45	12.79	2.710	0.061	5.404
Ca	3	14950.67	2487.41	1436.11	10.411	2	0.005	0.009	14950.67	8771.60	21129.73	2487.41	6.011	0.779	11.636
Cd	3	0.00	.00000^{a}	0.00											
Co	3	7.94	1.62	0.94	8.462	2	0.007	0.014	7.94	3.90	11.97	1.62	4.885	0.558	9.495
Cr	3	70.80	39.89	23.03	3.074	2	0.046	**0.092**	70.80	-28.30	169.90	39.89	1.775	-0.214	3.703
Cu	3	14.83	2.48	1.43	10.340	2	0.005	0.009	14.83	8.66	21.01	2.48	5.970	0.771	11.559
Fe	3	16391.33	4156.74	2399.90	6.830	2	0.01	0.021	16391.33	6065.41	26717.25	4156.74	3.943	0.358	7.712
K	3	9301.00	2828.90	1633.26	5.695	2	0.015	0.029	9301.00	2273.63	16328.37	2828.90	3.288	0.207	6.480
Mg	3	6342.33	1449.35	836.78	7.579	2	0.008	0.017	6342.33	2741.95	9942.71	1449.35	4.376	0.452	8.530
Mn	3	430.67	90.50	52.25	8.242	2	0.007	0.014	430.67	205.85	655.48	90.50	4.759	0.532	9.255
Na	3	398.67	66.98	38.67	10.309	2	0.005	0.009	398.67	232.28	565.05	66.98	5.952	0.768	11.524
Ni	3	33.99	17.14	9.90	3.435	2	0.038	**0.075**	33.99	-8.59	76.57	17.14	1.983	-0.147	4.075
P	3	7177.00	1968.21	1136.35	6.316	2	0.012	0.024	7177.00	2287.69	12066.31	1968.21	3.646	0.291	7.153
Pb	3	10.19	3.57	2.06	4.950	2	0.019	0.038	10.19	1.33	19.05	3.57	2.858	0.100	5.678
U	3	6.79	1.85	1.07	6.344	2	0.012	0.024	6.79	2.18	11.40	1.85	3.663	0.295	7.183
Zn	3	45.33	11.77	6.79	6.673	2	0.011	0.022	45.33	16.10	74.56	11.77	3.853	0.338	7.541

The *p* values highlighted in **bold** are above the 0.05 significance level.

Table 19. One-Sample t-test analysis for each element in Rn-M11, Rn-M12, and Rn-M13 for 50 – 75 cm depth (function of topography)

One-Sample Statistics					**One-Sample Test**							**One-Sample Effect Sizes**			
							Significance			95% Confidence Interval of the Difference		Standardizer[a]	Point Estimate	95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean	t	df	One-Sided p	Two-Sided p	Mean Difference	Lower	Upper			Lower	Upper
U_238	3	125.967	31.850	18.388	6.850	2	0.01	0.021	125.967	46.848	205.086	31.850	3.955	0.361	7.734
Th_232	3	58.367	19.775	11.417	5.112	2	0.018	0.036	58.367	9.242	107.491	19.775	2.951	0.124	5.852
U_235	3	5.873	1.479	0.854	6.877	2	0.01	0.02	5.873	2.199	9.548	1.479	3.970	0.364	7.763
K_40	3	2031.333	98.673	56.969	35.657	2	<.001	<.001	2031.333	1786.216	2276.450	98.673	20.587	3.227	39.566
Rn_222	3	18.167	20.377	11.765	1.544	2	0.131	**0.263**	18.167	-32.453	68.786	20.377	0.892	-0.559	2.224
Ca	3	29550.333	13324.603	7692.963	3.841	2	0.031	**0.062**	29550.333	-3549.815	62650.481	13324.603	2.218	-0.076	4.500
Cd	3	0.000	.00000[a]	0.000											
Co	3	7.417	1.735	1.002	7.404	2	0.009	0.018	7.417	3.107	11.727	1.735	4.275	0.430	8.338
Cr	3	41.500	9.945	5.742	7.228	2	0.009	0.019	41.500	16.794	66.206	9.945	4.173	0.408	8.146
Cu	3	15.800	2.740	1.582	9.986	2	0.005	0.01	15.800	8.992	22.608	2.740	5.766	0.732	11.169
Fe	3	15775.333	4721.006	2725.674	5.788	2	0.014	0.029	15775.333	4047.704	27502.962	4721.006	3.342	0.220	6.581
K	3	8063.000	2309.689	1333.500	6.046	2	0.013	0.026	8063.000	2325.415	13800.585	2309.689	3.491	0.255	6.861
Mg	3	8691.000	1016.165	586.683	14.814	2	0.002	0.005	8691.000	6166.707	11215.293	1016.165	8.553	1.239	16.491
Mn	3	376.667	113.536	65.550	5.746	2	0.014	0.029	376.667	94.629	658.705	113.536	3.318	0.214	6.536
Na	3	409.667	72.016	41.579	9.853	2	0.005	0.01	409.667	230.769	588.565	72.016	5.689	0.717	11.022
Ni	3	21.893	4.297	2.481	8.825	2	0.006	0.013	21.893	11.219	32.567	4.297	5.095	0.600	9.894
P	3	7383.333	1908.639	1101.953	6.700	2	0.011	0.022	7383.333	2642.012	12124.655	1908.639	3.868	0.341	7.571
Pb	3	9.783	2.785	1.608	6.085	2	0.013	0.026	9.783	2.866	16.701	2.785	3.513	0.260	6.903
U	3	10.060	2.527	1.459	6.895	2	0.01	0.02	10.060	3.782	16.338	2.527	3.981	0.366	7.783
Zn	3	51.400	10.983	6.341	8.106	2	0.007	0.015	51.400	24.116	78.684	10.983	4.680	0.516	9.106

The p values highlighted in **bold** are above the 0.05 significance level.

4.4.5. The correlation matrices and comparison of elemental associations for the BCR procedure

The statistical analysis for the BCR procedure, as shown in (Appendix A21.) indicated that:

The pH had a strong correlation with Co ($r = 0.965$), Fe ($r = 0.891$), and Ni ($r = 0.806$) at $p \leq 0.05$; a moderate correlation with Mn ($r = 0.512$) and Zn ($r = 0.500$) at $p > 0.05$; a low correlation with Cr ($r = 0.208$, $p > 0.05$); and a negative correlation with Cd, Cu, Pb, and U at $p \leq 0.05$.

The CEC had a strong correlation with Ni ($r = 0.812$), Co ($r = 0.861$) at $p \leq 0.05$; a moderate correlation with Cr ($r = 0.511$, $p > 0.05$) and Fe ($r = 0.618$, $p \leq 0.05$); a low correlation with Zn ($r = 0.290$) and Mn ($r = 0.093$) at $p > 0.05$; and a negative correlation with Cd, Cu, Pb, and U at $p > 0.05$.

The % SOM had a strong correlation with Zn ($r = 0.942$, $p \leq 0.05$), and a significant negative correlation with U ($r = 0.581$) and Cr ($r = 0.789$). The correlations between the pH, CEC, Co, Ni, and Fe were demonstrated by the behavior of these elements. When the pH and CEC decreased, these elements were increasing in the non-residual fractions.

In this statistical analysis, there was a significant relationship among Cd, Cr, Pb, U, and Cu at $p \leq 0.05$. Consequently, the null hypothesis was rejected. The pairs Cd-Cu ($r = 0.994$), Cd-Pb ($r = 0.995$), Cd-U ($r = 0.981$), Cu-Pb ($r = 0.999$), Cu-U ($r = 0.995$), and Pb-U ($r = 0.995$) at $p \leq 0.05$ had a strong correlation which might be an indication that the anthropogenic conditions could be the common factor among these elements. These results imply that where U is mobile, there is a high possibility that Cd, Cu, and Pb will also be available. This has been demonstrated by the Radioactive sample where 100 % of U was bound to all the non-residual fractions. Similarly, Cd (63.86 %), Cu (80.40 %), and Pb (83.10 %) including Co (90.02 %), Mn (87.63 %), and Zn (63.48 %) in Radioactive sample were in abundance in the non-residual fractions.

One-Sample t-test statistical analysis was conducted to determine if there are differences for each element in each fraction of the BCR method considering different samples. The results for the statistical analysis for Fraction 1 are shown in (Appendix A22.), for Fraction 2 are shown in (Appendix A23.), for Fraction 3 are shown in (Appendix A24.), and for Fraction 4 are shown in (Appendix A25.). In Fraction 1, all elements were significantly different except for Cd, while in Fraction 2, Cd, Co, Cu, U, and Zn were significantly different. Furthermore, Cd, Co, Cu, U, and Zn were significantly different in different samples, and in Fraction 4, the Cd and Mn elements were significantly different.

Linear regression analysis was performed to determine the soil's effect on the plants using the soil concentrations from the sum of the mobilized fractions (obtained using a BCR method) and the plant concentrations (Table 20.). The results were statistically insignificant for most of the elements. The results indicated that the correlations between the soil and the plants for each element were not strong except for U, which showed R = 0.794, $p > 0.05$. In fact, all the p values in this analysis were above the 0.05 significance level. The unstandardized coefficient B resulted in negative values except for Fe, which showed a positive but low value. In summary, these observations suggest that the mobilized element from the BCR method are not being taken up by the plants. In this case, the BCR method could not be considered to determine the elemental plant uptake from the soil. This is due to a large number of factors that control the PTE accumulation and bioavailability, such as soil and climate conditions, plant genotype, and agronomic management; this includes active/passive transfer processes, redox states, sequestration and speciation, the type of plant root system and the response of plants to elements in relation to seasonal cycles (Olszewski *et al.*, 2016). In this study, most of these factors were not taken into consideration since the plant analysis.

Hence the transfer factor values calculated using pseudo total concentrations are useful in this case for the determination of elemental plant uptake as it has already been established by several researchers (as discussed in section 2.4.3).

Table 20. Regression analysis for PTEs to determine the soil's effect on the plants using the sum of the mobilized fractions of the BCR method

Model Summary					**ANOVA[a]**						**Coefficients[a]**					
												Unstandardized Coefficients		Standardized Coefficients		
Element	R	R Square	Adjusted R Square	Std. Error of the Estimate	Model	Sum of Squares	df	Mean Square	F	Sig.		B	Std. Error	Beta	t	Sig.
Cr	.192[a]	0.037	-0.445	8.52768	Regression	5.575	1	5.575	0.077	.808[b]	Soil	-0.184	0.666	-0.192	-0.277	0.808
Cu	.092[a]	0.008	-0.487	28.20403	Regression	13.491	1	13.491	0.017	.908[b]	Soil	-0.158	1.212	-0.092	-0.130	0.908
Fe	.044[a]	0.002	-0.497	150.73046	Regression	87.131	1	87.131	0.004	.956[b]	Soil	0.004	0.060	0.044	0.062	0.956
Mn	.309[a]	0.095	-0.357	15.05023	Regression	47.697	1	47.697	0.211	.691[b]	Soil	-0.074	0.161	-0.309	-0.459	0.691
Ni	.471[a]	0.222	-0.167	2.80946	Regression	4.498	1	4.498	0.570	.529[b]	Soil	-0.560	0.742	-0.471	-0.755	0.529
U	.794[a]	0.631	0.446	1.12533	Regression	4.327	1	4.327	3.417	.206[b]	Soil	-0.049	0.027	-0.794	-1.849	0.206
Zn	.435[a]	0.189	-0.217	6.13069	Regression	17.515	1	17.515	0.466	.565[b]	Soil	-0.345	0.505	-0.435	-0.683	0.565
a. Predictors: (Constant), Soil					a. Dependent Variable: Plant b. Predictors: (Constant), Soil						a. Dependent Variable: Plant					

Regression analysis for Cd, Co, and Pb was not done due to the concentrations in plants being undetectable

5. CONCLUSION AND RECOMMENDATIONS

It has been established that radioactivity should be controlled, and the limitation of discharges to the environment should ensure that the levels in the environment are below the limits derived from the ICRP's (Valkovic, 1993). On the other hand, according to (Almeida *et al.*, 2011), there is no limitation to the concentration of naturally occurring radioisotopes in the soil (Laţo *et al.*, 2012). However, the activity concentrations for the soil can be compared to the World median values reported by UNSCEAR (2000).

It was observed that the activity concentrations of K-40, U-238, and Th-232 radionuclides indicated high activity concentrations in all soil and plant samples collected from the Mecsek uranium mine at Pecs. The concentrations for these three natural radionuclides were above the Hungarian mean values and the World's median values as specified by UNSCEAR. The vertical distribution of activity concentrations for all the natural radionuclides in all depth levels did not follow any trend; the results varied throughout for all sample locations. However, It could be noted that the samples collected from the bottom of the deposit (Rn-M13 and Radioactive samples) had elevated activity concentrations as compared to the samples collected from the top and on the slope of the deposit. This increase in concentrations is due to the leaching from the spoil matter through the cover soil layer along the slope of the deposit.

Most PTE concentrations in soil cover samples collected from deposit No. I were lower than the contamination limit values for Hungarian soils and within the permissible limits. The results for some PTEs indicated a trend in the increase of concentrations of elements as the slope of the deposit decreased. Soil samples collected from the top of the deposit (Rn-M11) contained less average concentration, followed by the sample collected from the slope (Rn-M12) and the sample collected from the bottom of the deposit (Rn-M13) had the highest average concentration. The results for Rn-M13 were observed to have the combination of the highest average for the soil moisture content, soil organic matter content, and PTE concentrations. These results suggest that when the soil organic matter content and/or soil moisture content is high, there is a possibility for the PTEs to be available in abundance. This was proven by statistical analysis, where there was a strong correlation among these variables ($p \leq 0.05$).

The results for PTEs in plants indicated that there was a possibility that these elements were available for uptake by the plants. Even though some of these elements are essential to plants, elevated concentrations may cause several problems, including the toxicity of the environment. Similarly, radionuclides activity concentrations were observed to be elevated in plants for all the samples collected from spoil deposit No. I. The unnoticeable difference in the results for the washed and unwashed radioactive samples eliminated the possibility that the concentrations detected in plant samples might be due to the surface contamination caused by the erosion. The transfer factor values calculated from the pseudo total concentrations indicated a likelihood of radionuclide and PTE uptake by plants. All samples were reported to have TF ratios that were very high for almost all radionuclides analyzed and for some of the PTEs, which indicated a higher absorption of these elements from the soil by the plant. The linear regression analysis for the determination of soil's effect on plants using the sum of the BCR mobile fractions indicated that there is no evidence that the plants have taken up the elements from these mobile fractions. This could be due to the plants having different plant genotypes, and agronomic management, and different type of plant root systems, and the response of plants to elements. In this case, the BCR method could not be used successfully to predict the possibility of plant uptake. However, the transfer factor calculations using pseudo total could be successfully applied for the prediction of plant uptake.

It was observed that all groundwater samples collected in this study were above the WHO guideline value. Elevated U concentrations in groundwater are of concern because it indicates active processes of the uranium leaching from the spoil deposits. It also shows the capability of elements migrating into the groundwater. The elevated U levels in groundwater may be due to the seeping of water containing high concentrations of U from the process water. These findings explained the need for Mecsekérc Zrt. (2017) to focus more on waste rock dumps No. I to ensure that the remediation process is successful. Based on the high levels of U in groundwater, it could be concluded that the soil cover layer requires an additional step to ensure that its integrity is intact. According to IAEA (2004a), one of many techniques to monitor the integrity of the soil cover layer is to monitor the radiological and chemical leachate collected in the vault drainage system and in groundwater to provide an indication of degradation in cover performance.

The possibility of migration of PTEs was confirmed using fractionation by sequential extraction for PTEs. The high average percentage of U, Mn, and Pb and a high percentage of U, Mn, Pb, and Cd in non-residual fractions indicates that there is a possibility being released may become a threat to the environment by subsequently becoming available to be taken up by plants (Pavlović *et al.*, 2018; Pérez-Moreno *et al.*, 2018). Although these elements exhibit high percentages in non-residual fractions, a large part of these percentages come from the Radioactive sample, which is radioactive and highly contaminated with U. Therefore, the location where Radioactive sample was taken from should be considered as a potential hazard to the environment. The fractional distribution of the Radioactive sample shows that the mobility, availability, and vertical transport of metals are surprisingly different (Fedotov *et al.*, 2018). The U, Cd, Co, Mn, Cu, Pb, and Zn fractional distribution in the Radioactive sample indicates that some parts of the spoil deposit require additional steps to protect the environment.

Based on the statistical analysis, a significant correlation between the pH and the CEC existed; therefore, it was concluded that these soil chemical properties have an impact on each other. Additionally, the pH had a significant relationship with Co, Fe, and Ni; and the CEC had a significant relationship with Co, Fe, and Ni, whereas the relationship between the CEC and Cr was not significant). It was observed that the pH and CEC relationship with the radionuclides was not significant. However, soil moisture had a significant relationship with Th-232. This strong positive correlation between these radionuclides and PTEs suggests their common origin (Bai *et al.*, 2017). When considering the topography, it was observed that the SOM values were significantly different in all three depths that were considered for this analysis, whereas the CEC was only significantly different in 25 – 50 cm depth. In terms of depth, CEC and SOM were found to be significantly different in Rn-M11. There was a strong significant relationship between radionuclides and some PTEs. The principal component analysis biplot analysis for soil indicated that when the pH, CEC, and Ca are high, the mobility of U, U-235, U-238, Cu, Cr, Pb, Rn-222, and Ni decrease. Furthermore, there was a very strong relationship among Cd, Cr, Pb, U, and Cu, which is significant at a 95 % confidence level. These results imply that where U is mobile, there is a high possibility that Cd, Cu, and Pb will also be available. The main trend of the concentration changes along the slope (considering only Rn-M11, Rn-M12, and Rn-M13), One-Sample t-test indicated that there was an increase of element concentrations from the top to bottom direction. The difference was significant in the case of Rn-222, U, Ca, Ni, Zn, Cr, and Ni. However, when the Radioactive sample was considered, an increase of U-238, U-235, and Pb was significant. The vertical distribution of PTEs and radionuclides did not show unambiguous trends; it was different for element to element and sampling position on the slope. The results indicated that only the Rn-M11 sample had elements that were significantly different as the depth deepens. In this sampling location, U-238, U-235, Rn-222, and Ca were significantly different. These results show that two samples (Rn-M11 and Radioactive samples) which did not have sufficient (1 m) soil cover, showed significant differences in concentrations for some elements. Moreover, these results suggest that the insufficient cover layer affects the concentrations of these elements from the spoil rock to the top layer of the deposit.

Therefore, as Sungur et al. (2014) stated, the BCR sequential extraction procedure has proven to be useful in providing information regarding the mobility of PTEs for the planning and management of recultivation applications. The concentrations for each PTE obtained using a BCR sequential extraction procedure were significantly different in each fraction for Cd, Cr, Co, Cu, U, Pb, Mn, and Fe. Based on the plant radionuclidic and PTE uptake; and on the results from the BCR sequential extraction, it is apparent that one meter (1 m) intact of the soil cover layer for spoil deposit No. I is enough for effective retardation of migration of PTEs and radionuclides. However, where the soil cover layer thickness is low (Radioactive sample), it is not compelling enough. Therefore, additional soil cover is needed to stabilize and protect the environment to ensure environmental safety. Additionally, since the sampling of soil sample in 75 – 100 cm depth in Rn-M11 and 25 – 100 cm for Radioactive sample was not possible due to the rocks underneath, it is an indication that the cover layer was not 1 m in these locations as recommended by the OECD/NEA (2014).

As a summary, it can be concluded that the BCR sequential extraction is an appropriate tool for the identification of leakages through the wounded remediation cover soil layer. Despite the relatively low pseudo-total concentration of the PTEs, the sequential extraction proves additional information on their mobility by water infiltration. These results of the BCR sequential extraction confirm the establishment based on the measuring of pseudo total concentration and plant uptake. This methodology has proven to be appropriate for application in similar circumstances in South Africa.

6. NEW SCIENTIFIC RESULTS

1. Radionuclide and PTE concentrations in the soil cover layer of the spoil deposit No. I take their origin from the soil's own matter and from the spoil rock due to the leaching and migration by water infiltration vertically and along the slope. The distribution of PTE and radionuclide concentrations in the soil cover layer depends on the chemical character of an element and soil characteristics (pH, CEC, SOM, Soil moisture, etc.) and has shown different patterns for different elements. In general, it could be stated that most PTE concentrations were lower than the contamination limits values for the Hungarian Soils. On the contrary, K-40, U-238, and Th-232 radionuclides were above the Hungarian mean values and the World median values. The main trend of the concentration changes along the slope (considering only Rn-M11, Rn-M12, and Rn-M13), there was an increase of element concentrations from the top to bottom direction. In the case of Rn-222, U, Ca, Ni, Zn, Cr, and Ni, the difference was significant. However, when the Radioactive sample was considered, an increase of U-238, U-235, and Pb was significant. The soil samples at the bottom of the deposit (Rn-M13) had the highest average SOM, soil moisture, and the least pH values; however, the CEC varied for all samples. The vertical distribution of PTEs and radionuclides did not show unambiguous trends; it was different for element to element and sampling position on the slope. At the wounded sampling location (Radioactive sample), where the soil cover layer was only 25 cm, significantly higher PTE and radionuclide concentrations were measured.
2. The mobility of different elements in the soil cover layer was detected by the BCR fractionation. In the upper (0-25 cm) covering soil layer, the BCR fractionation results have shown that the abundance of U (100%), Mn (73.5 %), Pb (55.1%), and Cd (36.8 %) was present in mobile fractions and this is an indication that these elements may move together along the slope of deposit by water. The mobility of PTEs at all sampling locations (average mobility factor) followed the order: U > Mn > Pb > Co > Cd > Cu > Ni > Zn > Fe > Cr. The mobility of U was confirmed by total concentrations in all four groundwater samples collected near spoil deposit No. I; PK-33/1 (6.06 ± 0.03 mg/L), PK-44/3 (0.23 ± 0.001 mg/L), PK-29/1 (1.87 ± 0.01 mg/L), and 1504/1 (2.78 ± 0.01 mg/L) which were all above the WHO guideline value for U (0.03 mg/L) in drinking water.
3. The concentrations of U-238, Th-232, K-40, Cr, Cu, and Zn in plant samples and the TF transfer factor values for U-238, U-235, Th-232, K-40, Cu, and K indicated that these elements were easily available for uptake from soil to plants. Using the pseudo total soil

concentrations for TF calculation, problem areas could be identified, and applying the BCR sequential extraction method helped to specify the elements prone to mobility and bioavailability according to environmental conditions in the Mecsek uranium mine.

4. The combination of the BCR sequential extraction and pseudo total methodologies used in this study discovered some problems regarding the efficacy of the recultivation soil in uranium mine areas; thus, it is possible to make additional proposals for more supplementary remediation processes which are necessary. It was proved that the soil cover layer satisfactory retard the migration of the PTEs and radionuclides from the spoil leaching. However, when it is wounded, it may cause the release of these contaminants into the groundwater and surrounding environment. The BCR sequential extraction has proven to identify mobile elements in uranium mine deposits. Therefore, this methodology should be an appropriate tool in evaluating environmental contamination. As a result, this fractionation method should be included in the monitoring program.

7. SUMMARY

This study is part of the ongoing environmental monitoring program of the abandoned Mecsek uranium mine in the period of mine closure, remediation, and post-closure operations. During this monitoring program on the recultivated spoil deposit No. I, the radionuclide and the potentially toxic element (PTE) contents in the covering soil had shown some anomalies that refer to possible migration alongside the slope through the covering soil. Consequently, there was a question of the adequacy of the 1-meter thickness soil covering layer used to cover the spoil deposit. Therefore, to further investigate this issue, the soil and plant samples were collected from four sampling locations following the top to the bottom position of the slope. The total metallic element content of soil and plant transfer was determined by multi-elemental inductively coupled plasma—optical emission spectrometry (ICP-OES). In contrast, Gamma-spectrometry and AlphaGUARD determined the activity concentrations for radionuclides. The natural radionuclide (U-238, Th-232, and K-40) concentrations in soil were all above the mean values for Hungary and the World median values as specified by The United Nations Scientific Committee on the Effects of Atomic Radiation, however, there was no trend in terms of soil depths. The study indicated a correlation between soil organic matter, PTE, and radionuclide content in plants and soil, and this suggested a high possibility for PTEs and radionuclides to be mobile and available for uptake by plants.

To confirm the indication of PTE mobility in the present study for the soil samples, the BCR sequential extraction procedure was applied to characterize the environmental mobility of PTEs, and it was compared with the soil pH and the cation exchange capacity (CEC). The results indicated that the ratio of Cd, Co, Mn, Pb, and U in the non-residual fractions ranged between 36.8 % to 100 % and increased from top to the bottom direction. The results of the BCR sequential extraction confirmed the establishment based on the measuring of pseudo total concentration and plant uptake.

Based on the radionuclides and PTE plant uptake and on the results from the BCR sequential extraction, it is apparent that one meter (1 m) intact of the soil cover layer for spoil deposit No. I is enough for effective retardation of migration of PTEs and radionuclides. However, where the soil cover layer thickness is low (Radioactive sample), it is not compelling enough. The distribution of U, Cd, Mn, Co, and Pb in fractions indicated that some parts of the spoil deposit require additional steps to hinder the migration through the covering soil layer. Therefore, the BCR sequential extraction procedure has proven to be useful in providing information for the planning and management of remediation operations. Consequently, this methodology can be useful in addressing similar problems in South Africa.

8. ÖSSZEFOGLALÁS

PhD kutatómunkám a Magyarországon a felhagyott Mecsek uránbánya bezárási és remediációs környezeti monitoring programjához kapcsolódott. Az eddigi monitoring során a rekultivált No. I jelű meddőhányót fedő talajrétegben a radionuklidok és a poteciálisan toxikus elemek (PTE) koncentrációjában mutatkozó anomáliák felvetették a a medőhányó lejtő irányában a környezet felé történő migráció lehetőségét. Ezért felmerült a kérdés, hogy ezen a területen megfelelő-e az egy méter vastag talajtakaró a migráció visszattartására. PhD kutatásaim célja ennek a kérdésnek a megválaszolására irányult a meddőhányó területén a lejtő irányában felülről lefelé vett talaj- és növényminták továbbá vízminták vizsgálatával. A mintavételi tervet az eddigi radiológiai monitoring gamma aktivitás eredményei alapján dolgoztam ki. A talaj, növény és vízminták mineralizálható teljes elemkoncentrációinak mérésére a doktori munkahelyemen kidolgozott flexibilis multielemes induktív csatolású plazma—optikai emissziós spektrometriás (ICP-OES) módszert alkalmaztam, a radionuklidok aktivitás koncentrációját gamma spektrometriával, a radont AlphaGUARD segítségével mértem. A természetes radionuklidok (U-238, Th-232, K-40) aktivitás koncentrációi a teljes fedőtalaj rétegben meghaladták a Magyar- és a világátlagokat (The United Nations Scientific Committee on the Effects of Atomic Radiation), s nem változtak szignifikánsan a mélység függvényében. A talaj és a növények föld felettirészének PTE és radionuklid tartalma és a talaj szervesanyag tartalma között kimutatott korreláció utalt a PTE és radionuklid tartalom jelentős mobilitásának lehetőségére a lejtő irányában, amelyet talaj/növény átviteli faktorok is alátámasztottak. A No. I. medőhányóval szomszédos területről származó vízmintákban ugyancsakészlelheő volt a raradionuklid és PTE koncentrációk növekedése a korábbi értékekhez képest.

A teljes elemkoncentrációk mérése alapján feltételezhető jelentős környezeti elem-mobilitás alátámasztására elvégeztem a felső 0-25 cm talajrétegben az elemtartalom frakcionálását a BCR által javasolt szekvens extrakciós eljárással, s a mobilitási faktorokat összehasonlítottam a talaj pH és kation cserélőképesség változásával (CEC). Eredményeim szerint a Cd, Co, Mn, Pb és U könnyen mobilizálható frakcióinak részaránya a teljes extraháható koncentrációk 37 és 100 %-a közé esett és a lejtőn lefelé haladva növekedett.

A növények radioaktív elem felvétele és a BCR szekvens extrakció eredményei igazolták, hogy az 1 mr vastagságú fedőtalaj réteg a rekultivált No. I. meddőhányón elegendő hatékonysággal gátolta a PTE és radinuklidok migrációját. Azonban azokon a helyeken, ahol a fedőréteg vastagsága kisebb (radioaktiv mintapont körzete) ez a visszatartás nem megfelelő mértékű. Az U, Cd, Mn, Co és Pb nagyarányú jelenléte a könnyen mobilizálható frakciókban azt jelzik, hogy a meddőhányó érintett részein további remediációs lépések szükségesek a fedőtalaj rétegen keresztül történő migráció visszatartására. Az erre vonatkozó tervek kidolgozásához, s az érintett területek körülhatárolásához a monitorozás kiegészítése a BCR szekvens extrakcióval hasznos információkat szolgáltat. Ez a metodika hazámban a Dél-Afrikai Köztársaságban is jól alkalmazható a felhagyott uránbányák remediációja során.

9. LIST OF PUBLICATIONS

1. Publication (Journal Article: Q2 - IF = 3.5)

Heltai, György; Győri, Zoltán; Fekete, Ilona; Halász, Gábor; Kovács, Katalin; Takács, Anita; Khumalo, Lamlile; Horváth, Márk. Application of flexible multi-elemental ICP-OES detection in fractionation of potentially toxic element content of solid environmental samples by a sequential extraction procedure. MICROCHEMICAL JOURNAL 149 Paper: 104029, 7 p. (2019). DOI WoS REAL Scopus.

2. Publication (Journal Article: Q3 - Peer-reviewed)

Lamlile, Khumalo; Gyorgy, Heltai; Mark, Horvath. The migration of potentially toxic elements during the recultivation of the uranium mining deposit in Mecsek. ACTA HYDROLOGICA SLOVACA 20: 2 pp. 210-217, 8 p. (2019). DOI Egyéb URL. Publication:30974882.

3. Publication (Journal Article: Q3 – IF = 1.5) – Accepted on the 11th June 2021

Lamlile Khumalo, György Heltai, András Várhegyi and Márk Horváth. Evaluating the mobility of potentially toxic elements from the uranium mine spoil bank using the BCR sequential extraction procedure. ECOLOGICAL CHEMISTRY AND ENGINEERING S (ECE S).

4. Oral presentation (Conference paper)

Lamlile, Khumalo; Mark, Horvath; Gyorgy, Heltai, Characterization of mobility of potentially toxic elements in soil-water system by sequential extraction procedures In: Jakab, G Tóth A Csengeri E (eds.) Alkalmzakodó vízgazdálkodás: lehetőségek és kockázatok Szarvas, Hungary: Szent István Egyetem Agrár- és Gazdaságtudományi Kar, (2018) pp. 260-266., 7 p.

22 March 2018, Szarvas, Hungary.

5. Oral presentation (Abstract)

Khumalo, Lamlile; Heltai, György; Horváth, Márk, Evaluation of the ecological risk caused by the mobility of radioactive elements and potentially toxic elements during the recultivation of the uranium mining deposit No.1 in Mecsek, 18th Alps-Adria Scientific Workshop. 1– 6 April 2019, Cattolica, Italy.

6. Oral presentation (Abstract)

Lamlile, Khumalo; Mark, Horvath; Gyorgy, Heltai, Sampling procedure for monitoring processes for the mobility of radioactive elements and potentially toxic elements during the recultivation of the uranium mining deposit No.1 in Mecsek, XXI. Századi vízgazdálkodás a tudományok metszéspontjában: II. Víztudományi Nemzetközi - Konferencia. 22 March 2019, Szarvas, Hungary.

7. Poster (Abstract)

Lamlile, Khumalo; György, Heltai; Miklós, Czémán; András, Várhegyi; Gábor, Németh; Gábor, Földing; Csaba, Alföldi; Mark, Horvath, Evaluation of the ecological risk caused by the mobility of radioactive and potentially toxic elements during the recultivation of the uranium mining deposit No. 1 in Mecsek, XVI Hungarian - Italian Symposium on Spectrochemistry: technological innovation for water science and sustainable aquatic biodiversity & 61st Hungarian Spectrochemical Conference. 2– 6 October 2018, Butapest, Hungary.

8. Oral presentation (Abstract)

György Heltai; Zoltán Győri; Ilona Fekete; Gábor Halász; Katalin Kovács; Anita Takács; Lamlile Khumalo; Márk Horváth. Fractionation of potentially toxic elements in sediment/soil/water system by a sequential extraction procedure applying flexible multielemental spectrochemical detection in remediation of contaminated areas. In: Mihucz, Viktor Gábor (eds.) XVI Hungarian - Italian Symposium on Spectrochemistry: technological innovation for water science and sustainable aquatic biodiversity & 61th Hungarian Spectrochemical Conference: October 3-6, 2018, Budapest: programme & book of abstracts. Budapest, Hungary: MKE (2018) 118 p. p. 49 Paper: KL04

9. Oral presentation (Abstract)

Khumalo, L.H.N.; Heltai, Gy.; Horvath, M. Mobility of radionuclides from the spoil deposit No.1 of the abandoned uranium mine in Pécs, Hungary In: 5th International Conference on Environmental Radioactivity (ENVIRA2019): Variations of Environmental Radionuclides. (2019) p. 67 Paper: ID 89. 8 – 13 September 2019, Prague, Czech Republic.

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11. APPENDICES

A1: Bibliography

6/2009. (IV. 14.). (2009), *KvVM-EüM-FVM Common Order about the Standard Limits and Measurement of Contamina- Tion for the Protection Of underground Water and Geological Medium*, Budapest.

Abiye, T. and Shaduka, I. (2017), “Radioactive seepage through groundwater flow from the uranium mines, Namibia”, *Hydrology*, Vol. 4 No. 1, pp. 1–11.

Adamo, P., Iavazzo, P., Albanese, S., Agrelli, D., De Vivo, B. and Lima, A. (2014), “Bioavailability and soil-to-plant transfer factors as indicators of potentially toxic element contamination in agricultural soils”, *Science of the Total Environment*, Elsevier B.V., Vol. 500–501, pp. 11–22.

Ademola, A.K., Olaoye, M.A. and Abodunrin, P.O. (2015), “Radiological safety assessment and determination of heavy metals in soil samples from some waste dumpsites in Lagos and Ogun state, south-western, Nigeria”, *Journal of Radiation Research and Applied Sciences*, Elsevier Ltd, Vol. 8 No. 1, pp. 148–153.

Adesiji, N.E. and Ademola, J.A. (2019), “Soil-to-cassava plant transfer factor of natural radionuclides on a mining impacted soil in a tropical ecosystem of Nigeria”, *Journal of Environmental Radioactivity*, Elsevier, Vol. 201 No. October 2018, pp. 1–4.

Agency for Toxic Substances and Disease Registry. (2013), *Toxicological Profile for Uranium, ATSDR Toxicological Profiles*, Atlanta, Georgia, available at:https://doi.org/http://dx.doi.org/10.1155/2013/286524.

Agic, R., Skopje, F., Milenkovic, L. and Ilic, Z.S. (2015), “Transfer Factor as Indicator”, No. December.

Ahmad, A.Y., Al-Ghouti, M.A., AlSadig, I. and Abu-Dieyeh, M. (2019), “Vertical distribution and radiological risk assessment of 137Cs and natural radionuclides in soil samples”, *Scientific Reports*, Springer US, Vol. 9 No. 1, pp. 1–14.

Ahmed, M.E., Al-Haddad, A., Mydlarczyk, A. and Aba, A. (2019), “The Presence and Distribution of Radioactivity and Radionuclides in Kuwait Wastewater Treatment Plants”, *Arabian Journal for Science and Engineering*, Springer Berlin Heidelberg, Vol. 44 No. 10, pp. 8779–8786.

Al-hwaiti, M.S. and Ranville, J.F. (2010), “Distribution of potentially toxic metal and radionuclide contamination in soils related to phosphogypsum waste stockpiling in the Eshidiya Mine , Jordan Geochemistry : Exploration , Environment , Analysis Distribution of potentially toxic metal and radion”, No. August 2015, available at:https://doi.org/10.1144/1467-7873/09-232.

Al-Jundi, J. (2002), “Population doses from terrestrial gamma exposure in areas near to old phosphate mine, Russaifa, Jordan”, *Radiation Measurements*, Vol. 35 No. 1, pp. 23–28.

Alharbi, A. and El-Taher, A. (2013), “A study on transfer factors of radionuclides from soil to plant”, *Life Science Journal*, Vol. 10 No. 2, pp. 532–539.

Almeida, G.M., Campos, S.S.S., Gennari, R.F. and Souza, S.O. (2011), “Determination of the concentration of radionuclides in soil and water next the uranium mine of Caetité-ba”.

Alshahri, F. (2019), “Uranium and Trace Metals Contamination in Topsoil from Different Zones Around Industrial City, Al Jubail, Saudi Arabia”, *Archives of Environmental Contamination and Toxicology*, Springer US, Vol. 77 No. 2, pp. 308–319.

Althouse, A.D. and Soman, P. (2017), “Understanding the true significance of a P value”, *Journal of Nuclear Cardiology*, Vol. 24 No. 1, pp. 191–194.

Atomic Heritage Foundation. (2018), “Uranium Mining | Atomic Heritage Foundation”, *National Museum of Nuclear Science & History*, available at: https://www.atomicheritage.org/history/uranium-mining (accessed 3 April 2020).

Avudainayagam, S., Megharaj, M., Owens, G., Kookana, R.S., Chittleborough, D. and Naidu, R. (2003), “Chemistry of chromium in soils with emphasis on Tannery waste sites”, in Ware, G.W. (Ed.), *Reviews of Environmental Contamination and Toxicology*, Vol. 178, Springer-Verlag, New York, pp. 53–91.

Azeez, H.H., Mansour, H.H. and Ahmad, S.T. (2019), “Transfer of natural radioactive nuclides from soil to plant crops”, *Applied Radiation and Isotopes*, Elsevier Ltd, Vol. 147 No. October 2018, pp. 152–158.

Bacon, J.R. and Davidson, C.M. (2017), “Is there a future for sequential chemical extraction ? {”, No. October, available at:https://doi.org/10.1039/b711896a.

Bai, H., Hu, B., Wang, C., Bao, S., Sai, G., Xu, X., Zhang, S., *et al.* (2017), “Assessment of radioactive materials and heavy metals in the surface soil around the Bayanwula prospective uranium mining area in China”, *International Journal of Environmental Research and Public Health*, Vol. 14 No. 3, pp. 1–14.

Bakatula, E.N., Cukrowska, E.M. and Tutu, H. (2012), “Proceedings of the Annual Workshop IGCP/SIDA No.594, Windhoek, Namibia, July 5th – 6th, 2012”, in Mapani, B. and Kribek, B. (Eds.), *Environmental and Health Impacts of Mining in Africa*, Czech Geological Survey, Prague, Czech Republic.

Barać, N., Škrivanj, S., Bukumirić, Z., Živojinović, D., Manojlović, D., Barać, M., Petrović, R., *et al.* (2016), “Distribution and mobility of heavy elements in floodplain agricultural soils along the Ibar River (Southern Serbia and Northern Kosovo). Chemometric investigation of pollutant sources and ecological risk assessment”, *Environmental Science and Pollution Research*, Vol. 23 No. 9, pp. 9000–9011.

Baran, A. and Tarnawski, M. (2015), “Assessment of heavy metals mobility and toxicity in contaminated sediments by sequential extraction and a battery of bioassays”, *Ecotoxicology*, Springer US, Vol. 24 No. 6, pp. 1279–1293.

Baskaran, M. (2016), *Radon: A Tracer for Geological, Geophysical and Geochemical Studies*, edited by Baskaran, M., © Springer International Publishing, Switzerland, available at:https://doi.org/10.1007/978-3-319-21329-3.

Bernard, B.B., Bernard, H. and Brooks, J.M. (n.d.). “Determination of total carbon, total organic carbon and inorganic carbon in sediments”, *JTDI-Brooks International/B&B Laboratories Inc.*, Texas, available at:https://doi.org/10.1099/00222615-7-2-223.

Bettinelli, M., Beone, G.M., Spezia, S. and Baffi, C. (2000), “Determination of heavy metals in soils and sediments by microwave-assisted digestion and inductively coupled plasma optical emission spectrometry analysis”, Vol. 424, pp. 289–296.

Biehler, D., Jaquet, O., Croise, J. and Lavanchy, J. (2002), *Hydrogeological Site Investigation for the Efficient Remediation of Uranium Mining Sites — an Integrated Approach, Inis.Iaea.Org*, available at: https://inis.iaea.org/search/search.aspx?orig_q=RN:33032940.

Bielicka-Giełdoń, A., Ryłko, E. and Zamojć, K. (2013), “Distribution, bioavailability and fractionation of metallic elements in Allotment garden soils using the BCR sequential extraction procedure”, *Polish Journal of Environmental Studies*, Vol. 22 No. 4, pp. 1013–1021.

BioRender.com. (n.d.). “BioRender”, available at: https://biorender.com/ (accessed 28 January 2021).

Calmon, P., Fesenko, S., Voigt, G. and Linsley, G. (2009), “Quantification of radionuclide transfer in terrestrial and freshwater environments”, *Journal of Environmental Radioactivity*, Vol. 100 No. 9, pp. 671–674.

Canadian Council of Ministers of the Environment. (2007), *Canadian Soil Quality Guidelines for Uranium : Environmental and Human Health - Scientific Supporting Document, Food Chemistry*, Vol. 277.

Caroli, S. (1996), “Element speciation in bioinorganic chemistry”, New York, Wiley.

Carvalho, F.P. (2011), “Environmental Radioactive Impact Associated to Uranium Production”, *American Journal of Environmental Sciences*, Vol. 7 No. 6, pp. 547–553.

Carvalho, F.P. (2017), “Mining industry and sustainable development: Time for change”, *Food and Energy Security*, Vol. 6 No. 2, pp. 61–77.

Carvalho, F.P., Oliveira, J.M., Malta, M. and Lemos, M.E. (2014), “Radioanalytical assessment of environmental contamination around non-remediated uranium mining legacy site and radium mobility”, *Journal of Radioanalytical and Nuclear Chemistry*, Vol. 299 No. 1, pp. 119–125.

Carvalho, F.P., Oliveira, M. and Lopes, I. (2007), “Radionuclides from past uranium mining in rivers of Portugal”, *Journal of Environmental Radioactivity*, Vol. 98, pp. 298–314.

Chakraborty, S.R., Azim, R., Rezaur Rahman, A.K.M. and Sarker, R. (2013), “Radioactivity concentrations in soil and transfer factors of radionuclides from soil to grass and plants in the chittagong city of Bangladesh”, *Journal of Physical Science*, Vol. 24 No. 1, pp. 95–113.

Chibuike, G.U. and Obiora, S.C. (2014), “Heavy metal polluted soils: Effect on plants and bioremediation methods”, *Applied and Environmental Soil Science*, Vol. 2014, available at:https://doi.org/10.1155/2014/752708.

Ciesielski, H. and Sterckeman, T. (1997), “A comparison between three methods for the determination of cation exchange capacity and exchangeable cations in soils”, *Agronomie*, Vol. 17 No. 1, pp. 9–16.

Collier, D. (2016), *Uranium Mine and Mill Remediation and Reclamation, Uranium for Nuclear Power: Resources, Mining and Transformation to Fuel*, Elsevier Ltd, available at:https://doi.org/10.1016/B978-0-08-100307-7.00016-8.

Commission BCR information reference materials (CRM 601). (1997), ISBN 92-828-0127-6.

Commission of the European Communities. (1986), ‘Council Directive (86/278/EEC) of 12 June on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture'. Official Journal of the European Communities', L 181, pp. 6–12.

Cui, H., Fan, Y., Fang, G., Zhang, H., Su, B. and Zhou, J. (2016), "Ecotoxicology and Environmental Safety Leachability , availability and bioaccessibility of Cu and Cd in a contaminated soil treated with apatite , lime and charcoal : A fi ve-year fi eld experiment", Vol. 134, pp. 148–155.

Dabek-Zlotorzynska, E., Kelly, M., Chen, H. and Chakrabarti, C.L. (2005), "Application of capillary electrophoresis combined with a modified BCR sequential extraction for estimating of distribution of selected trace metals in $PM_{2.5}$ fractions of urban airborne particulate matter", *Chemosphere*, Vol. 58, pp. 1365-1376.

Dasnois, N. (2012), *Uranium Mining in Africa : A Continent at the Centre of a Global Nuclear Renaissance*, Johannesburg.

De Almeida, A.A.F., Valle, R.R., Mielke, M.S. and Gomes, F.P. (2007), "Tolerance and prospection of phytoremediator woody species of Cd, Pb, Cu and Cr", *Brazilian Journal of Plant Physiology*, Vol. 19 No. 2, pp. 83–98.

Deng, D., Zhang, L., Dong, M., Samuel, R.E., Ofori-Boadu, A. and Lamssali, M. (2020), "Radioactive waste: A review", *Water Environment Research*, Vol. 92 No. 10, pp. 1818–1825.

Denneman, C. A. J. and Robberse, J.G. (1990), 'Ecotoxicological risk assessment as a base for development of soil quality criteria, In: Contaminated Soil'90; third international KfK/TNO conference on Contaminated Soil', Arendt, F., Hinselveld, M. and van den Brink, W.J. (Eds) Kluwer academic publishers, Dordrecht/Boston/London.

Dikinya, O. (2015), "Heavy metals and radionuclide status and characterisation of pre-mined soils Heavy metals and radionuclide status and characterisation of pre-mined soils in Serule , North East Botswana", *Environmental Earth Sciences*, Springer Berlin Heidelberg, No. May 2014, available at:https://doi.org/10.1007/s12665-014-3795-3.

Dowdall, M., Standring, W., Shaw, G. and Strand, P. (2008), "Will global warming affect soil-to-plant transfer of radionuclides ?", *Journal of Environmental Radioactivity*, Elsevier Ltd, Vol. 99 No. 11, pp. 1736–1745.

Dragović, S., Petrović, J., Dragović, R., Đorđević, M., Đokić, M. and Gajić, B. (2015), "The Influence of Edaphic Factors on Spatial and Vertical Distribution of Radionuclides in Soil". In: Walther, C. and Gupta, D. (Eds) Radionuclides in the Environment. Springer, Cham. https://doi.org/10.1007/978-3-319-22171-7_3

Dudu, V.P., Mathuthu, M. and Manjoro, M. (2018), "Assessment of heavy metals and radionuclides in dust fallout in the West Rand mining area of South Africa", *Clean Air Journal*, Vol. 28 No. 2, pp. 42–52.

Durand, J.F. (2012), "The impact of gold mining on the Witwatersrand on the rivers and karst system of Gauteng and North West Province, South Africa", *Journal of African Earth Sciences*, Elsevier Ltd, Vol. 68, pp. 24–43.

Durusoy, A. and Yildirim, M. (2017), "Determination of radioactivity concentrations in soil samples and dose assessment for Rize Province, Turkey", *Journal of Radiation Research and Applied Sciences*, Elsevier Taiwan LLC, Vol. 10 No. 4, pp. 348–352.

El-taher, A. (2015), "A Study on Transfer Factors of Radionuclides from Soil to plant", No.

January 2013.

Elzain, A.A. (2014), “Measurement of Radon-222 concentration levels in water samples in Sudan”, Vol. 5 No. 2, pp. 229–234.

Erdi-Krausz, G. (2005), “Problems and solutions for water treatment at the closed Hungarian uranium industry”, in IAEA in cooperation with the OECD Nuclear Energy Agency and DIAMO State Owned Enterprise (Ed.), *Recent Developments in Uranium Exploration, Production and Environmental Issues - IAEA-TECDOC-1463*, IAEA, Staz, Czech Republic, p. 87.

Erőss, A., Csondor, K., Izsák, B., Vargha, M., Horváth, Á. and Pándics, T. (2018), “Uranium in groundwater – The importance of hydraulic regime and groundwater flow system’s understanding”, *Journal of Environmental Radioactivity,* Vol. 195 No. September, pp. 90–96.

EURATOM Supply Ageny. (2018), *EURATOM Supply Agency - Annual Report 2017,* available at:https://doi.org/10.2833/61904.

Fairlie, I. (2009), “Childhood cancer near nuclear power stations”, *Environmental Health*, Vol. 8 No. 1, p. 43.

FAO/WHO. (2011), “Joint FAO/WHO Food Standards Programme Codex Committee on Contaminants in Foods”, *Codex Committee on Contaminants in Foods,* CODEX ALIMENTARIUS COMMISSION, The Hague, The Netherlands, available at: ftp://ftp.fao.org/codex/meetings/CCCF/cccf5/cf05_INF.pdf.

Fassel, V.A. and Kniseley, R.N. (1974), “Inductively Coupled Plasma-Optical Emission Spectroscopy”, *Analytical Chemistry,* Vol. 46 No. 13, pp. 1110A-1120A.

Fedotov, P.S., Dzhenloda, R.K., Dampilova, B. V., Doroshkevich, S.G. and Karandashev, V.K. (2018), “Unexpected behavior of Zn, Cd, Cu, and Pb in soils contaminated by ore processing after 70 years of burial”, *Environmental Chemistry Letters*, Springer International Publishing, Vol. 16 No. 2, pp. 637–645.

Feist, B., Mikula, B., Pytlakowska, K., Puzio, B. and Buhl, F. (2008), “Determination of heavy metals by ICP-OES and F-AAS after preconcentration with 2,2′-bipyridyl and erythrosine”, *Journal of Hazardous Materials,* Vol. 152 No. 3, pp. 1122–1129.

Fernández-Ondoño, E., Bacchetta, G., Lallena, A.M., Navarro, F.B., Ortiz, I. and Jiménez, M.N. (2017), “Use of BCR sequential extraction procedures for soils and plant metal transfer predictions in contaminated mine tailings in Sardinia”, *Journal of Geochemical Exploration,* Elsevier B.V., Vol. 172, pp. 133–141.

Ferrari, C.R., Do Nascimento, H.D.A.F., Rodgher, S., Almeida, T., Bruschi, A.L., Nascimento, M.R.L. Do and Bonifácio, R.L. (2017), “Effects of the discharge of uranium mining effluents on the water quality of the reservoir: An integrative chemical and ecotoxicological assessment”, *Scientific Reports,* Vol. 7 No. 1, pp. 1–10.

Filgueiras, A. V., Lavilla, I. and Bendicho, C. (2002), “Chemical sequential extraction for metal partitioning in environmental solid samples”, *Journal of Environmental Monitoring,* Vol. 4 No. 6, pp. 823–857.

Filgueiras, A. V., Lavilla, I. and Bendicho, C. (2004), “Evaluation of distribution, mobility and binding behaviour of heavy metals in surficial sediments of Louro River (Galicia, Spain) using chemometric analysis: A case study", *Science of the Total Environment,* Vol. 330 No. 1–3, pp. 115–129.

Fite, T. and Leta, S. (2015), "Determination of Levels of As , Cd , Cr , Hg and Pb in Soils and Some Vegetables Taken from River Mojo Water Irrigated Farmland at Koka Village , Oromia State , East Ethiopia", *International Journal of Sciences: Basic and Applied Research (IJSBAR) I*, Vol. 21 No. 2, pp. 352–372.

Flórián, K., Matherny, M., Nickel, H., Pliešovská, N. and Uhrinová, K. (2003), "Environmental characteristics of the atmosphere of residential agglomerations - I", Dustiness of the agglomerations, *Chem. Pap.*, Vol. 57, pp. 369-373.

Flórián, K., Matherny, M., Nickel, H., Pliešovská, N. and Uhrinová K. (2003), "Environmental characteristics of the atmosphere of residential agglomerations - II", Main, minor, and trace elements in the gravitation dust sediments, *Chem. Pap.*, Vol. 57, pp. 374-381.

Forghani, G., Kazemi, G.A. and Qishlaqi, A. (2014), "Bioavailability and speciation of potentially toxic elements in agricultural soils neighboring an industrial complex in northwest Isfahan , Iran", pp. 165–173.

Gad, A., Saleh, A. and Khalifa, M. (2019), "Assessment of natural radionuclides and related occupational risk in agricultural soil, southeastern Nile Delta, Egypt", *Arabian Journal of Geosciences*, Arabian Journal of Geosciences, Vol. 12 No. 6, available at:https://doi.org/10.1007/s12517-019-4356-6.

Garnier–Laplace, J., Colle, C. and Morello, M. (2001), *Radionuclide Fact Sheet: Natural Uranium and the Environment*.

Gavrilescu, M., Pavel, L.V. and Cretescu, I. (2009), "Characterization and remediation of soils contaminated with uranium", *Journal of Hazardous Materials,* Vol. 163 No. 2–3, pp. 475–510.

Geography, E. and Labor, N. (2008), "Heavy metal content of flood sediments and plants", Vol. 2 No. 2, pp. 120–131.

George-Laurentiu, M., Florentina-Cristina, M. and Andreea-Loreta, C. (2016), "The Assessment of Social and Economic Impacts Associated to an Abandoned Mining Site Case study: Ciudanovita (Romania)", *Procedia Environmental Sciences,* Elsevier B.V., Vol. 32, pp. 420–430.

Gerzabek, M.H., Strebl, F. and Temmel, B. (1998), "Plant uptake of radionuclides in lysimeter experiments", *Environmental Pollution,* Vol. 99 No. 1, pp. 93–103.

Gleyzes, C., Tellier, S. and Astruc, M. (2002), "Fractionation studies of trace elements in contaminated soils and sediments: A review of sequential extraction procedures", *TrAC - Trends in Analytical Chemistry,* Vol. 21 No. 6–7, pp. 451–467.

Goulet, R.R., Thompson, P.A., Serben, K.C. and Eickhoff, C. V. (2015), "Impact of environmentally based chemical hardness on uranium speciation and toxicity in six aquatic species", *Environmental Toxicology and Chemistry,* Vol. 34 No. 3, pp. 562–574.

Greger, M. (2004), *Uptake of Nuclides by Plants,* Stockholm, Sweden.

Gulan, L. and Milenkovic, B. (2018), "Environmental risk assessment of radioactivity and heavy metals in soil of Toplica region , South Serbia", Vol. 0, pp. 2101–2118.

Gulan, L., Milenkovic, B., Zeremski, T., Milic, G. and Vuckovic, B. (2017), “Persistent organic pollutants, heavy metals and radioactivity in the urban soil of Priština City, Kosovo and Metohija”, *Chemosphere*, Elsevier Ltd, Vol. 171, pp. 415–426.

Gunn, A.M., Winnard, D.A. and Hunt, D.T.E. (1988), “Trace metal speciation in sediment and soils”. In: Kramer, J.R. and Allen, H.E. (Eds.), *Metal speciation: Theory, analysis and application*, Lewis Publications, Boca Raton, Florida.

Gupta, D.K. and Voronina, A. (2019), *Remediation Measures for Radioactively Contaminated Areas*, Springer International Publishing AG, Switzerland.

Gupta, D.K. and Walther, C. (2014), *Radionuclide Contamination and Remediation through Plants*, edited by Gupta, D.K. and Walther, C.*Radionuclide Contamination and Remediation Through Plants*, Springer International Publishing, Switzerland, available at:https://doi.org/10.1007/978-3-319-07665-2.

Győri, Z., Alapi, K., Sipos, P. and Zubor, Á. (2003a), “Effects of heavy metal on floodplain soils and pastures of the River Tisza, Hungary II”, In *Natural Attenuation of Metals Along the Tisza River–Floodplain–Wetlands Continuum*, pp. 161–163, Debreceni Egyetem, Debrecen.

Győri, Z., Alapi, K. and Szilágyi Sz. (2003b), “Effects of heavy metal on floodplain soils and pastures of the River Tisza, Hungary I”, In *Natural Attenuation of Metals Along the Tisza River–Floodplain–Wetlands Continuum*, pp. 146–160, Debreceni Egyetem, Debrecen.

He, Z., Shentu, Yang, X., Baligar, V.C., Zhang, T. and Stoffella, &. (2015), “Soil pollution in the world (EEC”, *Journal of Environmental Indicators*, Vol. 9 No. Table 2, pp. 17–18.

Heard, B. (2017), *Environmental Impacts of Uranium Mining in Australia History , Progress and Current Practice Environmental Impacts of Uranium Mining in Australia*, Sydney, Australia.

Hegazy, A.K., Afifi, S.Y., Alatar, A.A., Alwathnani, H.A. and Emam, M.H. (2013), “Soil characteristics influence the radionuclide uptake of different plant species”, *Chemistry and Ecology*, Vol. 29 No. 3, pp. 255–269.

Heidak, M.O., Glasmacher, U.A. and Schöler, H.F. (2014), “A comparison of micronutrients (Mn, Zn, Cu, Mo, Ni, Na) within rocks, soils, and leaves, from fallow agricultural lands and natural laurel forest areas (Tenerife, Canary Islands, Spain)”, *Journal of Geochemical Exploration*, Elsevier B.V., Vol. 136, pp. 55–64.

Helaluddin, A., Khalid, R.S., Alaama, M. and Abbas, S.A. (2016), “Main Analytical Techniques Used for Elemental Analysis in Various Matrices”, *Pharm Res Tropical Journal of Pharmaceutical Research Journal Citation ReportsScience Edition*, Vol. 15 No. 152, pp. 427–427.

Heltai, Gy. and Füleky, Gy. (1992), “Soil analysis and nutrition capacity”, *Microchemical Journal*, Vol. 46 No. 1, pp. 4-19.

Heltai, Gy., Fekete, I., Gémesi, Z., Percsich, K., Flórián, K. and Tarr, Zs. (1998), “Environmental evaluation of a local lake chain affected by wastewater by means of spectrochemical analytical methods”, *Microchem. J.*, Vol. 59, pp. 125-135.

Heltai, Gy., Percsich, K., Fekete, I., Barabás, B. and Józsa, T. (2000), “Speciation of waste water sediments”, *Microchem. J.*, Vol. 67, pp. 43-51.

Heltai, Gy., Fehér, B., Percsich, K., Barabás, B. and Fekete, I. (2002), “Application of sequential extraction with supercritical CO_2, subcritical H_2O, and an H_2O/CO_2 mixture for estimation of environmentally mobile heavy metal fractions in sediments", *Anal. Bioanal. Chem.*, Vol. 373, pp. 863-866.

Heltai, G. (2005), "Chemical interpretation of a new sequential extraction scheme based on supercritical CO_2 and subcritical H_2O solvents", *Bulletin of the Szent István University*, pp.107-122.

Heltai, Gy., Percsich, K., Halász, G., Jung, K. and Fekete, I. (2005), "Estimation of ecotoxicological potential of contaminated sediments based on a sequential extraction procedure with supercritical CO_2 and subcritical H_2O solvents", *Microchem. J.*, Vol. 79, pp. 231-237.

Heltai, G., Remeteiová, D., Horváth, M., Széles, É., Halász, G., Fekete, I. and Flórián, K. (2011), "Various fractionation procedures in study of heavy metals mobility in the environment", *Ecological Chemistry and Engineering S*, Vol. 18 No. 1, pp. 55-65.

Heltai, G., Győri, Z., Fekete, I., Halász, G., Kovács, K., Takács, A., Boros, N., *et al.* (2018), "Longterm study of transformation of potentially toxic element pollution in soil/water/sediment system by means of fractionation with sequential extraction procedures", *Microchemical Journal*, Vol. 136, pp. 85–93.

Heltai, G., Győri, Z., Fekete, I., Halász, G., Kovács, K., Takács, A., Khumalo, L., *et al.* (2019), "Application of flexible multi-elemental ICP-OES detection in fractionation of potentially toxic element content of solid environmental samples by a sequential extraction procedure", *Microchemical Journal*, Elsevier, Vol. 149 No. January, p. 104029.

Hernandez-Soriano, M.C. and Jimenez-Lopez, J.C. (2012), "Effects of soil water content and organic matter addition on the speciation and bioavailability of heavy metals", *Science of the Total Environment*, Elsevier B.V., Vol. 423, pp. 55–61.

Van Herreweghe, S., Swennen, R., Vandecasteele, C. and Cappuyns, V. (2003), "Solid phase speciation of arsenic by sequential extraction in standard reference materials and industrially contaminated soil samples", *Environmental Pollution*, Vol. 122 No. 3, pp. 323–342.

Hiller, E., Tóth, R., Kučerová, G., Jurkovič, Ľ., Šottník, P., Lalinská-Voleková, B. and Vozár, J. (2016), "Geochemistry of Mine Tailings from Processing of Siderite–Cu Ores and Mobility of Selected Metals and Metalloids Evaluated by a Pot Leaching Experiment at the Slovinky Impoundment, Eastern Slovakia", *Mine Water and the Environment*, Vol. 35 No. 4, pp. 447–461.

Hooda, P.S. (2010), *Trace Elements in Soils*, edited by Hooda, P.S., 1st ed., John Wiley & Sons, Ltd, Chichester, West Sussex, United Kingdom, available at:https://doi.org/10.13140/RG.2.1.3377.1123.

Hook, R. Van. (2000), "Potential Health and Environmental Effects of Trace Elements and Radionuclides from Increased Coal Utilization", Vol. 33 No. December 1979, pp. 227–247.

Horváth, M., Boková, V., Heltai, G., Flórián, K. and Fekete. I. (2010), "Study of application of BCR sequential extraction procedure for fractionation of heavy metal content of soils, sediments and gravitation dusts", *Toxicol. Environ. Chem.*, Vol. 92 No. 3, pp. 429-441.

Horváth, M., Halász, G., Kucanová, E., Kuciková, B., Fekete, I., Remeteiová, D., Heltai, Gy. amd Flórián K. (2013), "Sequential extraction studies on aquatic sediment and biofilm samples for the assessment of heavy metal mobility". *Microchem. J.,* Vol. 107, pp. 121-125.

Hungarian Atomic Energy Authority. (2017), *NATIONAL REPORT Sixth Report Prepared within the Framework of the Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management*, Budapest.

Hungarian Central Statistical Office. (n.d.). "Meteorological data (1985-)", *Summary Tables (STADAT) - Time Series of Annual Data - Environment*, available at: https://www.ksh.hu/docs/eng/xstadat/xstadat_annual/i_met002cc.html (accessed 7 April 2021a).

Hungarian Central Statistical Office. (n.d.). "Meteorological Data", *Summary Tables (STADAT) - Time Series of Annual Data - Environment: 15.2.2.8. Meteorological Data of Pécs**, available at: https://www.ksh.hu/stadat_files/kor/en/kor0077.html (accessed 30 March 2021b).

Hungarian Regulation 10 16/2000. (2000), "Ministry of Health implementing the provisions of the law No. CXVI. of the year 1996 of nuclear energy", *Hungarian Bulletin No. 55*; Budapest (2000).

Hurel, C., Taneez, M., Volpi Ghirardini, A. and Libralato, G. (2017), "Effects of mineral amendments on trace elements leaching from pre-treated marine sediment after simulated rainfall events", *Environmental Pollution*, Elsevier Ltd, Vol. 220 No. March 2018, pp. 364–374.

IAEA. (2004a), *Safety Report Series No.35 - Surveillance and Monitoring of Near Surface Disposal Facilities for Radioactive Waste,* Vienna, Austria.

IAEA. (2004b), *Treatment of Liquid Effluent from Uranium Mines and Mills - Report of a Co-Ordinated Research Project 1996–2000, IAEA-TECDOC-1419,* Austria.

IAEA. (2009), *Establishment of Uranium Mining and Processing Operations in the Context of Sustainable Development, IAEA Nuclear Energy Series No. NF-T-1.1 - STI/PUB/1401,* Vienna, Austria, available at: https://www-pub.iaea.org/MTCD/Publications/PDF/Pub1401_web.pdf.

IAEA. (2010), *Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments, Technical Reports Series No.472,* Vol. 472, Vienna, Austria, available at:https://doi.org/10.1016/j.jenvrad.2010.10.004.

ICRP. (2007), *The 2007 Recommendations of the International Commission on Radiological Protection, Annals of the ICRP - Publication 103.*

Igwe, J.C., Nnorom, I.C. and Gbaruko, B.C. (2005), "Kinetics of radionuclides and heavy metals behaviour in soils: Implications for plant growth", *African Journal of Biotechnology,* Vol. 4 No. 13 SPEC. ISS., pp. 1541–1547.

Igwe, J.C., Nnorom, I.C. and Gbaruko, B.C. (2019), "Radionuclides and heavy metals in soils in connection to soil contamination and their effects on plant growth", Vol. 7 No. 1, pp. 511–517.

Intawongse, M. and Dean, J.R. (2007), "Uptake of heavy metals by vegetable plants grown on contaminated soil and their bioavailability in the human gastrointestinal tract", *Food Additives and Contaminants,* Vol. 23 No. 1, pp. 36–48.

Interantional Atomic Energy Agency. (2011), *The Uranium Mining Remediation Exchange*

Group.

International Atomic Energy Agency. (2010), “Best Practice in Environmental Management of Uranium Mining”, *International Atomic Energy Agency*, available at: http://www-pub.iaea.org/MTCD/Publications/PDF/Pub1406_web.pdf.

International Atomic Energy Agency. (2016), “Soils Newsletter”, *Newsletter*, IAEA, Vienna, Austria, available at:https://doi.org/10.1163/15700607-00561p01.

International Atomic Energy Agency (IAEA). (2004), “Uranium Mill Tailings”, *The Long Term Stabilization of Uranium Mill Tailings*, Vol. IAEA-TECDO No. August, pp. 75–80.

International Atomic Energy Agency (IAEA). (2005), “Recent developments in uranium exploration, production and environmental issues”, *Proceedings of a Technical Meeting Organized by the IAEA in Cooperation with the OECD Nuclear Energy Agency and DIAMO State Owned Enterprise*, IAEA, Staz, Czech Republic.

International Atomic Energy Agency (IAEA). (2011), *Technical Reports Series No. 472: Review of “Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments”, IAEA*, Vol. 102, Austria, available at:https://doi.org/10.1016/j.jenvrad.2010.10.004.

International Atomic Energy Agency (IAEA). (2016), “Country Nuclear Power Profiles: Hungary”, IAEA, available at: https://www-pub.iaea.org/MTCD/Publications/PDF/cnpp2016/countryprofiles/Hungary/Hungary.htm (accessed 7 May 2020).

Isimekhai, K.A., Garelick, H., Watt, J. and Purchase, D. (2017), “Heavy metals distribution and risk assessment in soil from an informal E-waste recycling site in Lagos State, Nigeria”, *Environmental Science and Pollution Research*, Vol. 24 No. 20, pp. 17206–17219.

Isinkaye, O.M., Adeleke, S. and Isah, D.A. (2018), “Background Radiation Measurement and the Assessment of Radiological Impacts due to Natural Radioactivity Around Itakpe Iron-Ore Mines”, *Mapan - Journal of Metrology Society of India*, Vol. 33 No. 3, pp. 271–280.

Ismail, B.P. (2017), “Ash Content Determination”, in Nielsen, S.S. (Ed.), *Food Analysis Laboratory Manual*, Third Edit., Springer International Publishing AG, Cham, Switzerland, pp. 117–119.

Iurian, A.-R., Phaneuf, M.O. and Mabit, L. (2015), *Radionuclides in the Environment*, edited by Walther, C. and Gupta, D.K., Switzerland.

Jaishankar, M., Tseten, T., Anbalagan, N., Mathew, B.B. and Beeregowda, K.N. (2014), “Toxicity, mechanism and health effects of some heavy metals”, *Interdisciplinary Toxicology*, Vol. 7 No. 2, pp. 60–72.

Jaremko, D. and Kalembasa, D. (2014), “A comparison of methods for the determination of cation exchange capacity of soils”, *Ecological Chemistry and Engineering S*, Vol. 21 No. 3, pp. 487–498.

Jena, V., Gupta, S., Dhundhel, R.S., Matic, N., Frančišković-Bilinski, S. and Devic, N. (2013), “Determination of total heavy metal by sequential extraction from soil”, *Int. J. Res. Environ. Sci. Technol.* Vol. 3 No. 1, pp. 35-38.

Jolly, Y.N., Islam, A. and Akbar, S. (2013), “Transfer of metals from soil to vegetables and possible health risk assessment”, pp. 1–8.

Jones, C. and Jacobsen, J. (2001), “Plant Nutrition and Soil Fertility: Nutrient Management Module No.2”, *Nutrient Management Module*.

Juhasz, L. and Erdi-Krausz, G. (1993), “Consequences of the Hungarian uranium mining and milling”, in IAEA (Ed.), *Proceedings of a Workshop Held within the Technical Co-Operation Project on Environmental Restoration in Central and Eastern Europe in Budapest, Hungary, 4-8 October 1993: Planning for Environmental Restoration of Radioactiviely Contaminated Sites in Cent*, INTERNATIONAL ATOMIC ENERGY AGENCY, Vienna, Austria, pp. 151–156.

Júnior, J.A.S., Cardoso, J.J.R.F., Silva, C.M., Silveira, S. V. and Amaral, R.S. (2006), “Determination of radionuclides in the environment using gamma-spectrometry”, *Journal of Radioanalytical and Nuclear Chemistry*, Vol. 269 No. 2, pp. 451–455.

Kabała, C., Musztyfaga, E., Gałka, B., Łabuńska, D. and Mańczyńska, P. (2016), “Conversion of soil pH 1:2.5 KCl and 1:2.5 H2O to 1:5 H2O: Conclusions for soil management, environmental monitoring, and international soil databases”, *Polish Journal of Environmental Studies*, Vol. 25 No. 2, pp. 647–653.

Kabala, C. and Singh, B.R. (2001), “Fractionation and Mobility of Copper, Lead, and Zinc in Soil Profiles in the Vicinity of a Copper Smelter”, *Journal of Environmental Quality*, Vol. 30 No. 2, pp. 485–492.

Kabata-Pendias, A. (2004), “Soil-plant transfer of trace elements - An environmental issue”, *Geoderma*, Vol. 122 No. 2-4 SPEC. IIS., pp. 143–149.

Kabata-pendias, A. and Pendias, H. (2001), *Trace Elements in Soils and Plants Trace Elements in Soils and Plants*.

Kang, K.W. (2016), “History and organizations for radiological protection”, *Journal of Korean Medical Science*, Vol. 31 No. 3, pp. S4–S5.

Kede, M.L. uiz. F.M., Correia, F. V., Conceição, P.F., Junior, S.F.S., Marques, M., Moreira, J.C. and Pérez, D. V. (2014), “Evaluation of mobility, bioavailability and toxicity of Pb and Cd in contaminated soil using TCLP, BCR and earthworms”, *International Journal of Environmental Research and Public Health*, Vol. 11 No. 11, pp. 11528–11540.

Kersten, M. and Förstner, M. (1995), “Speciation of trace metals in sediments and combustion waste”, in Ure, A.M. and Davidson, C.M. (Eds.), *Chemical speciation in the environment*, Blackie Academic and Professional, London pp. 237.

Keshavarzi, B., Moore, F., Ansari, M., Rastegari Mehr, M., Kaabi, H. and Kermani, M. (2015), “Macronutrients and trace metals in soil and food crops of Isfahan Province, Iran”, *Environmental Monitoring and Assessment*, Vol. 187 No. 1, available at:https://doi.org/10.1007/s10661-014-4113-y.

Khelifi, F., Besser, H., Ayadi, Y., Liu, G., Yousaf, B., Harabi, S., Bedoui, S., *et al.* (2019), “Evaluation of potentially toxic elements’ (PTEs) vertical distribution in sediments of Gafsa–Metlaoui mining basin (Southwestern Tunisia) using geochemical and multivariate statistical analysis approaches”, *Environmental Earth Sciences*, Springer Berlin Heidelberg, Vol. 78 No. 2, pp. 1–14.

Khumalo, L., Heltai, G. and Horváth, M. (2020), “The migration of potentially toxic elements during the recultivation of the uranium mining deposit in Mecsek”, *Acta Hydrologica Slovaca*, Vol. 20 No. 2, pp. 210–217.

Konoplev, A., Kato, K. and Kalmykov, S.N. (2020), *Behavior of Radionuclides in the Environment II - Chernobyl*, edited by Konoplev, A., Kato, K. and Kalmykov, S.N.*Radiochemistry and Nuclear Chemistry*, II., Springer Nature Singapore Pte Ltd., Singapore, available at:https://doi.org/10.1016/b978-075067463-8/50022-4.

Konradi, E.A., Frentiu, T., Ponta, M. and Cordos, E. (2005), “Use of Sequential Extraction to Assess Metal Fractionation in Soils from Bozanta Mare”, *Romania. Acta Universitatis Cibiniensis Seria F Chemia,* Vol. 8 No. 2, pp. 5-12.

Korychenskyi, K.O., Laptev, G. V., Voitsekhovych, O. V., Lavrova, T. V. and Dyvak, T.I. (2018), “Speciation and mobility of uranium in tailings materials at the U-production legacy site in UKRAINE”, *NUCL. PHYS. AT. ENERGY*, Vol. 19, pp. 270–279.

Kotoky, P., Bora, B.J., Baruah, N.K., Baruah, J., Baruah, P. and Borah, G.C. (2003), “Chemical fractionation of heavy metals in soils around oil installations, Assam”, *Chemical Speciation and Bioavailability,* Vol. 15 No. 4, pp. 115–126.

Kronzucker, H.J., Coskun, D., Schulze, L.M., Wong, J.R. and Britto, D.T. (2013), “Sodium as nutrient and toxicant”, *Plant and Soil,* Vol. 369 No. 1–2, pp. 1–23.

Kubová, J., Streško, V., Bujdoš, M., Matúš, P. and Medved’, J. (2004), “Fractionation of various elements in CRMs and in polluted soils”, *Anal. Bioanal. Chem.*, Vol. 379, pp. 108-114.

Kubová, J., Matúš, P., Hagarová, I., Bujdoš, M. and Medved’, J. (2008), “Utilization of optimized BCR three-step sequential and dilute HCl single extraction procedures for soil–plant metal transfer predictions in contaminated lands”, *Talanta,* Vol. 75, pp. 1110-1122.

Kuterbekov, K. (2019), *Environmental Monitoring at a Former Uranium Milling Site*, Springer International Publishing AG, Switzerland.

Du Laing, G., Rinklebe, J., Vandecasteele, B., Meers, E. and Tack, F.M.G. (2009), “Trace metal behaviour in estuarine and riverine floodplain soils and sediments: A review”, *Science of the Total Environment,* Elsevier B.V., Vol. 407 No. 13, pp. 3972–3985.

Laţo, A., Radulov, I., Berbecea, A., Laţo, K. and Crista, F. (2012), “The transfer factor of metals in soil-plant system Mp Ms”, *Research Journal of Agricultural Science,* Vol. 44 No. 3, pp. 67–72.

Lema, M.W., Ijumba, J.N., Njau, K.N., Ndakidemi, P.A., Arusha, P.O.B.N. and Republic, U. (2014), “Environmental Contamination By Radionuclides And Heavy Metals Through The Application Of Phosphate RocksDuring Farming And Mathematical Modeling Of Their Impacts To The Ecosystem”, *International Journal of Engineering Research and General Science,* Vol. 2 No. 4, pp. 852–863.

Liefferink, M. (2008), “ECO-SYSTEMS A contribution to a just transition to a low carbon economy to combat unemployment and climate change”, pp. 1–53.

Lohry, R. (2007), “Micronutrients : Functions , Sources and Application Methods”, *Indiana CCA Conference Proceedings*.

López-Sánchez, J.F., Sahuquillo, A., Fiedler, H.D., Rubio, R., Rauret, G., Muntau, H. and Quevauviller, Ph. (1998). “CRM 601, A stable material for its extractable content of heavy metals”, *Analyst,* Vol. 123, pp. 1675-1677.

Lourenço, J., Mendo, S. and Pereira, R. (2019), “Rehabilitation of Radioactively Contaminated Soil: Use of Bioremediation/Phytoremediation Techniques”. In: Gupta, D. and Voronina, A.

(Eds) Remediation Measures for Radioactively Contaminated Areas. Springer, Cham.

Ma, H., Hua, L. and Ji, J. (2011), “Speciation and phytoavailability of heavy metals in sediments in Nanjing section of Changjiang River”, *Environmental Earth Sciences*, Vol. 64 No. 1, pp. 185–192.

Malovics, G. (2014), “Uranium mine opening in Pecs, Hungary”. Retrieved 09 24, 2017, from Ejatlas.org: http://ejalas.org/conflict/uranium-mine-reopening-in-pecs-hungary.

Manigandan, P.K. (2009), “Activity concentration of radionuclides in plants in the environment of Western Ghats”, Vol. 7 No. 2, pp. 85–90.

Manigandan, P.K. and Chandar Shekar, B. (2014), “Evaluation of radionuclides in the terrestrial environment of Western Ghats”, *Journal of Radiation Research and Applied Sciences*, Elsevier Ltd, Vol. 7 No. 3, pp. 310–316.

Matshusa, K. and Makgae, M. (2017), “Prevention of future legacy sites in uranium mining and processing: The South African perspective”, *Ore Geology Reviews*, Elsevier B.V., Vol. 86, pp. 70–78.

Mecsekérc Zrt. (2017), “Uranium Monitoring”, available at: https://www.mecsekerc.hu/eng-uranipari-monitoring (accessed 10 May 2020).

Ministerial Decree No. 15/2001 (VI. 6.) KöM. (2001), *Environment on Radioactive Discharges to the Atmosphere and into Waters during the Use of Atomic Energy and on Monitoring of the Discharge, The Hungarian Ministry*, Budapest, available at: In Hungarian.

Mirecki, N., Agič, R., Šunić, L., Milenković, L. and Ilić, Z.S. (2015), “Transfer Factor As Indicator”, *Fresenius Environmental Bulletin ·*, Vol. 24 No. 11c, p. 4212.

Mittermüller, M., Saatz, J. and Daus, B. (2016), “A sequential extraction procedure to evaluate the mobilization behavior of rare earth elements in soils and tailings materials”, *Chemosphere*, Elsevier Ltd, Vol. 147, pp. 155–162.

MSZ 1484-3. (2006), “Testing of waters. Part 3: Determination of dissolved, suspended and total metals in water by AAS and ICP-OES”, *Hungarian Standard Association*, Budapest (in Hungarian).

MSZ 21470–50. (2006), “Environmental testing of soils. Determination of total and soluble toxic element, heavy metal and chromium (VI) content”. *Hungarian Standard Association*, Budapest, (In Hungarian).

Mustapha, A.A., Abdu, N., Oyinlola, E.Y. and Nuhu, A.A. (2020), “Comparison of 3 Different Methods of CEC Determination in Nigerian Savannah Soils”, *Journal of Applied Sciences*, Vol. 20 No. 4, pp. 159–165.

National Research Council of the National Academies. (2012), *Uranium Mining in Virginia: Scientific, Technical, Environmental, Human Health and Safety, and Regulatory Aspects of Uranium Mining and Processing in Virginia*, available at:https://doi.org/10.17226/13266.

Nemati, K., Bakar, N.K.A., Abas, M.R. and Sobhanzadeh, E. (2011), “Speciation of heavy metals by modified BCR sequential extraction procedure in different depths of sediments from Sungai Buloh, Selangor, Malaysia”, *Journal of Hazardous Materials*, Elsevier B.V., Vol. 192 No. 1, pp. 402–410.

Nieder, R., Benbi, D.K. and Reichl, F.X. (2018), *Soil Components and Human Health*, edited by

Nieder, R., Benbi, D.K. and Reichl, F.X.*Soil Components and Human Health*, Springer Nature, Dordrecht, The Netherlands, available at:https://doi.org/10.1007/978-94-024-1222-2.

Nielsen, S.S. (2019), "Moisture Content Determination", in Nielsen, S.S. (Ed.), *SpringerBriefs in Applied Sciences and Technology*, Third Edit., Springer International Publishing AG, Cham, Switzerland, pp. 105–115.

Núñez-Delgado, A., Romar-Gasalla, A., Santás-Miguel, V., Fernández-Sanjurjo, M.J., Álvarez-Rodríguez, E., Nóvoa-Muñoz, J.C. and Arias-Estévez, M. (2017), "Cadmium and lead sorption/desorption on non- amended and by-product-amended soil samples and pyritic material", *Water (Switzerland),* Vol. 9 No. 11, available at:https://doi.org/10.3390/w9110886.

OECD/NEA. (2014), *Managing Environmental and Health Impacts of Uranium Mining, NEA No. 7062*, PARIS CEDEX 16.

OECD. (1999), *Environmental Activities in Uranium Mining and Milling : A Joint Report, Nuclear Development - A Joint NEA/IAEAReport*, Paris, France.

OECD. (2014), *U Ranium 2014 : Resources , Production and Demand*, France.

Ogundele, D.T., Adio, A.A. and Oludele, O.E. (2015), "Heavy Metal Concentrations in Plants and Soil along Heavy Traffic Roads in North Central Nigeria", *Environmental & Analytical Toxicology*, Vol. 5 No. 6, pp. 1–5.

Ogundiran, M.B. and Osibanjo, O. (2009), "Mobility and speciation of heavy metals in soils impacted by hazardous waste", *Chemical Speciation and Bioavailability*, Vol. 21 No. 2, pp. 59–69.

Ojuederie, O.B. and Babalola, O.O. (2017), "Microbial and plant-assisted bioremediation of heavy metal polluted environments: A review", *International Journal of Environmental Research and Public Health*, Vol. 14 No. 12, available at:https://doi.org/10.3390/ijerph14121504.

Okoro, H.K., Fatoki, O.S., Adekola, F.A., Ximba, Bhekumusa, J. and Snyman, Re.G. (2012), "A Review of Sequential Extraction Procedures for Heavy Metals Speciation in Soil and Sediments", *Journal of Environmental & Analytical Toxicology*, Vol. 01 No. S1, pp. 1–9.

Okoro, H.K., Ige, J.O., Iyiola, O.A. and Ngila, J.C. (2017), "Fractionation profile, mobility patterns and correlations of heavy metals in estuary sediments from olonkoro river, in tede catchment of western region, Nigeria", *Environmental Nanotechnology, Monitoring and Management*, Vol. 8, available at:https://doi.org/10.1016/j.enmm.2017.04.003.

Olowoyo, J.O., Odiwe, A.I., Mkolo, N.M. and Macheka, L. (2013), "Investigating the concentrations of different elements in soil and plant composition from a mining area", *Polish Journal of Environmental Studies*, Vol. 22 No. 4, pp. 1135–1141.

Olszewski, G., Boryło, A. and Skwarzec, B. (2016), "A study on possible use of Urtica dioica (common nettle) plants as uranium (234U, 238U) contamination bioindicator near phosphogypsum stockpile", *Journal of Radioanalytical and Nuclear Chemistry*, Springer Netherlands, Vol. 308 No. 1, pp. 37–46.

Opaluwa, O.D., Aremu, M.O., Ogbo, L.O., Abiola, K.A., Odiba, I.E., Abubakar, M.M. and Nweze, N.O. (2012), "Heavy metal concentrations in soils , plant leaves and crops grown around dump sites in Lafia Metropolis , Nasarawa State , Nigeria”, *Advances in Applied Science Research*, Vol. 3 No. 2, pp. 780–784.

OrangeSmile.com. (n.d.). “Maps of Hngary”, *Orange Smile*, available at: http://www.https//www.orangesmile.com/travelguide/hungary/country-maps.htm (accessed 3 April 2021).

Organisation for Economic Co-operation and Development. (2014), *Managing Environmental and Health Impacts of Uranium Mining*.

Palansooriya, K.N., Shaheen, S.M., Chen, S.S., Tsang, D.C.W., Hashimoto, Y., Hou, D., Bolan, N.S., *et al.* (2020), “Soil amendments for immobilization of potentially toxic elements in contaminated soils: A critical review”, *Environment International*, Elsevier, Vol. 134 No. June 2019, p. 105046.

Pandey, B., Agrawal, M. and Singh, S. (2016), “Ecological risk assessment of soil contamination by trace elements around coal mining area”, *Journal of Soils and Sediments*, Vol. 16 No. 1, pp. 159–168.

Panhwar, Q.A., Naher, U.A., Shamshuddin, J., Othman, R. and Ismail, M.R. (2016), “Applying limestone or basalt in combination with bio-fertilizer to sustain rice production on an acid sulfate soil in malaysia”, *Sustainability (Switzerland)*, Vol. 8 No. 7, available at:https://doi.org/10.3390/su8070700.

Pavlović, D., Pavlović, M., Čakmak, D., Kostić, O., Jarić, S., Sakan, S., Đorđević, D., *et al.* (2018), “Fractionation, Mobility, and Contamination Assessment of Potentially Toxic Metals in Urban Soils in Four Industrial Serbian Cities”, *Archives of Environmental Contamination and Toxicology*, Springer US, Vol. 75 No. 3, pp. 335–350.

Payne, T.E. and Edis, R. (2012), “Mobility of Radionuclides in Tropical Soils and Groundwater”, *Radioecology in the Environment*, Elsevier Ltd, Australia, pp. 93–121.

Perez-Moreno, S.M., Gazquez, M.J., Perez-Lopez, R. and Bolivar, J.P. (2018), “Validation of the BCR sequential extraction procedure for natural radionuclides”, *Chemosphere*, Vol. 198, pp. 397–399.

Pérez-Moreno, S.M., Gázquez, M.J., Pérez-López, R. and Bolivar, J.P. (2018), “Validation of the BCR sequential extraction procedure for natural radionuclides”, *Chemosphere*, Vol. 198, pp. 397–408.

Popic, J.M. (2014), *Environmental Impact of Radionuclides and Trace Elements in the Thorium Rich Fen Area in Norway*, Norwegian University of Life Sciences, available at:https://doi.org/http://hdl.handle.net/11250/2425285.

Popic, J.M., Salbu, B., Strand, T. and Skipperud, L. (2011), “Assessment of radionuclide and metal contamination in a thorium rich area in Norway”, *Journal of Environmental Monitoring*, Vol. 13 No. 6, pp. 1730–1738.

Prakash, D., Gabani, P., Chandel, A.K., Ronen, Z. and Singh, O. V. (2013), “Bioremediation: A genuine technology to remediate radionuclides from the environment”, *Microbial Biotechnology*, Vol. 6 No. 4, pp. 349–360.

Pretorius, S. and Hattingh, A. (2009), “The impacts of mining activities on the environment and the necessity for an environmental assessment strategy for such activity in South Africa”, Vol. 2009 No. 55.

Public Health Department of the Cree Board of Health and Social Services of James Bay. (2014), *Potential Health and Psychosocial Impacts of Uranium Development in Eeyou Istchee*.

Pueyo, M., Rauret, G., Lück, D., Yli-Halla, M., Muntau, H., Quevauviller, Ph. and López-Sánchez, J.F. (2001), “Certification of the extractable contents of Cd, Cr, Cu, Ni, Pb and Zn in a freshwater sediment following a collaboratively tested and optimised three-step sequential extraction procedure”, *J. Envir. Monit.*, Vol. 3, pp. 243-250.

Pueyo, M., Mateu, J., Rigol, A., Vidal, M. and Lo, J.F. (2008), “Use of the modified BCR three-step sequential extraction procedure for the study of trace element dynamics in contaminated soils”, *Environmental Pollution*, Vol. 152, pp. 330–341.

Qasim, B. and Motelica-Heino, M. (2014), “Potentially toxic element fractionation in technosoils using two sequential extraction schemes”, *Environmental Science and Pollution Research*, Vol. 21 No. 7, pp. 5054–5065.

Quesada-González, D., Jairo, G.A., Blake, R.C., Blake, D.A. and Merkoçi, A. (2018), “Uranium (VI) detection in groundwater using a gold nanoparticle/paper-based lateral flow device”, *Scientific Reports*, Vol. 8 No. 1, pp. 8–15.

Quevauviller, P., Rauret, G., López-Sánchez, J.F., Rubio, R., Ure, A. and Muntau, H. (1997), “Certification of trace metal extractable contents in a sediment reference material (CRM 601) following a three-step sequential extraction procedure”, *Sci. Total Environ.*, Vol. 205, pp. 223-234.

Rastegari Mehr, M., Keshavarzi, B., Moore, F., Sharifi, R., Lahijanzadeh, A. and Kermani, M. (2017), “Distribution, source identification and health risk assessment of soil heavy metals in urban areas of Isfahan province, Iran”, *Journal of African Earth Sciences*, Vol. 132, pp. 16–26.

Rauret, G., López-Sánchez, J.F., Lück, D., Yli-Halla, M., Muntau, H. and Quevauviller, P. (2001), *The Certification of the Extractable Contents (Mass Fractions) of Cd, Cr, Cu, Ni, Pb and Zn in Freshwater Sediment Following a Sequential Extraction Procedure BCR-701*, European Commission, Brussels, available at: https://ec.europa.eu/jrc/en/publication/eur-scientific-and-technical-research-reports/additional-certification-pb-mass-fraction-bcr-320r-channel-sediment.

Rauret, G., López-Sánchez, J.F., Sahuquillo, A., Rubio, R., Davidson, C., Ure, A. and Quevauviller, P. (1999), “Improvement of the BCR three step sequential extraction procedure prior to the certification of new sediment and soil reference materials”, *Journal of Environmental Monitoring*, Vol. 1 No. 1, pp. 57–61.

Rauret, G., López-Sánchez, J.F., Sahuquillo, A., Rubio, R., Davidson, C., Ure, A. and Quevauviller, P. (2001), *The Certification of the Extractable Contents (Mass Fractions) of Cd, Cr, Cu, Ni. Pb and Zn in Freshwater Sediment Following a Sequential Extraction Procedure - BCR-701*, Belgium, available at: https://www.crm.jrc.ec.europa.eu/.

Rékási, M. and Filep, T. (2012), “Fractions and background concentrations of potentially toxic elements in Hungarian surface soils”, *Environmental Monitoring and Assessment*, Vol. 184, pp. 7461–7471.

Remeteiová, D., Smiňčáková, E. and Flórián, K. (2007), “Study of the chemical properties of gravitation dust sediments”, *Microchim. Acta*, Vol. 156, pp. 109–113.

Republic of Hungary. (2008), *National Report, Editor and Publisher*, Budapest.

Richon, P. and Beneito, A. (2002), “Evaluation of the effect of a cover layer on radon exhalation from uranium mill tailings : transient radon flux analysis”, Vol. 63, pp. 49–64.

Rieuwerts, J.S. (2007), “The mobility and bioavailability of trace metals in tropical soils: A review”, *Chemical Speciation and Bioavailability*, Vol. 19 No. 2, pp. 75–85.

Rinklebe, J. and Shaheen, S.M. (2017), “Geochemical distribution of Co, Cu, Ni, and Zn in soil profiles of Fluvisols, Luvisols, Gleysols, and Calcisols originating from Germany and Egypt”, *Geoderma*, Elsevier, Vol. 307 No. August, pp. 122–138.

Rodgers, K.J., Hursthouse, A. and Cuthbert, S. (2015), “The potential of sequential extraction in the characterisation and management of wastes from steel processing: A prospective review”, *International Journal of Environmental Research and Public Health*, Vol. 12 No. 9, pp. 11724–11755.

Roper, W.R., Robarge, W.P., Osmond, D.L. and Heitman, J.L. (2019), “Comparing Four Methods of Measuring Soil Organic Matter in North Carolina Soils”, *Soil Science Society of America Journal*, Vol. 83 No. 2, pp. 466–474.

Rosado, D., Usero, J. and Morillo, J. (2016), “Ability of 3 extraction methods (BCR, Tessier and protease K) to estimate bioavailable metals in sediments from Huelva estuary (Southwestern Spain)”, *Marine Pollution Bulletin*, Elsevier Ltd, Vol. 102 No. 1, pp. 65–71.

Rösner, T. and Van Schalkwyk, A. (2000), “The environmental impact of gold mine tailings footprints in the Johannesburg region, South Africa”, *Bulletin of Engineering Geology and the Environment*, Vol. 59 No. 2, pp. 137–148.

Ross, D.S. and Ketterings, Q. (2004), “Recommended Methods for Determining Soil Cation Exchange Capacity”, in Rajala, A. (Ed.), *Plant Growth Regulators to Manipulate Oat Stands*, Vol. 13, pp. 186–197.

Rusnák, R., Halász, G., Horváth, M. and Remeteiová D. (2010), “Preliminary results on the intensification of the BCR sequential extraction with sonication for sediments, soils, and gravitation dust sediment samples”, *Toxicological & Environmental Chemistry*, Vol. 92 No. 3, pp. 443-452.

Saffari, M., Yasrebi, J. and Karimian, N. (2009), “Evaluation of Three Sequential Extraction Methods for Fractionation of Zinc in Calcareous and Acidic Soils”, *Research Journal of Biological Science*.

Sahito, O.M., Afridi, H.I., Kazi, T.G. and Baig, J.A. (2015), “Evaluation of heavy metal bioavailability in soil amended with poultry manure using single and BCR sequential extractions”, *International Journal of Environmental Analytical Chemistry*, Vol. 95 No. 11, pp. 1066–1079.

Sahuquillo, A., López-Sánchez, J.F., Rubio, R., Rauret, G., Thomas, R.P., Davidson, C.M. and Ure, A.M. (1999), “Use of a certified reference material for extractable trace metals to assess sources of uncertainty in the BCR three-stage sequential extraction procedure”, *Anal. Chim. Acta*, Vol. 382, pp. 317-327.

Sahuquillo, A., Rigol, A. and Rauret, G. (2003), “Overview of the use of leaching/extraction tests for risk assessment of trace metals in contaminated soils and sediments”, *Trends Anal. Chem.*, Vol. 22, pp. 152-159.

Saint-Fort, R. (2016), “Understanding Sorption Behavior and Properties of Radionuclides in the

Environment", *Intech*, Vol. i, Calgary, AB, Canada, p. 13.

Salbu, B., Lind, O.C. and Skipperud, L. (2004), "Radionuclide speciation and its relevance in environmental impact assessments", *Journal of Environmental Radioactivity*, Vol. 74 No. 1–3, pp. 233–242.

Saleem, M., Iqbal, J., Akhter, G. and Shah, M.H. (2018), "Fractionation, bioavailability, contamination and environmental risk of heavy metals in the sediments from a freshwater reservoir, Pakistan", *Journal of Geochemical Exploration*, Elsevier, Vol. 184 No. November 2017, pp. 199–208.

Dos Santos, J.S., Dos Santos, M.J.S. and Dos Santos, M.L.P. (2012), "Influence of the rainfall regime on the mobility of Zn, Cd, Ni, Cu, Mn and Fe in the surface sediments of the Contas River located in the Brazilian semi-arid region", *Journal of the Brazilian Chemical Society*, Vol. 23 No. 4, pp. 718–726.

Savosko, V., Podolyak, A., Komarova, I. and Karpenko, A. (2020), "Modern environmental technologies of healthy soils contaminated by heavy metals and radionuclides", *E3S Web of Conferences*, Vol. 166, pp. 1–9.

Schneider, A., Nguyen, V.X., Viala, Y., Violo, V., Cornu, J.Y., Sterckeman, T. and Nguyen, C. (2019), "A method to determine the soil-solution distribution coefficients and the concentrations for the free ion and the complexes of trace metals: Application to cadmium", *Geoderma*, Elsevier, Vol. 346 No. April, pp. 91–102.

Selvakumar, R., Ramadoss, G., Mridula, P.M., Rajendran, K., Thavamani, P., Ravi Naidu and Megharaj, M. (2018), "Challenges and complexities in remediation of uranium contaminated soils: A review", *Journal of Environmental Radioactivity*, Elsevier, Vol. 192 No. January, pp. 592–603.

Sereshti, H., Entezari Heravi, Y. and Samadi, S. (2012), "Optimized ultrasound-assisted emulsification microextraction for simultaneous trace multielement determination of heavy metals in real water samples by ICP-OES", *Talanta*, Elsevier, Vol. 97, pp. 235–241.

Shaheen, A. and Iqbal, J. (2018), "Spatial distribution and mobility assessment of carcinogenic heavy metals in soil profiles using geostatistics and Random Forest, Boruta Algorithm", *Sustainability (Switzerland)*, Vol. 10 No. 3, available at:https://doi.org/10.3390/su10030799.

Shaheen, S.M., Tsadilas, C.D., Ok, Y.S. and Rinklebe, J. (2016), "Potential Mobility, Bioavailability, and Plant Uptake of Toxic Elements in Temporary Flooded Soils", in Jorg, R., Knox, A. and Michael, P. (Eds.), *Trace Elements in Waterlogged Soils and Sediments*, 1st ed., CRC Press, Boca Raton, Florida, pp. 287–312.

Shahrokhi, A., Vigh, T., Németh, C., Csordás, A. and Kovács, T. (2017), "Radon measurements and dose estimate of workers in a manganese ore mine", *Applied Radiation and Isotopes*, Elsevier Ltd, Vol. 124 No. February, pp. 32–37.

Shiowatana, J., Tantidanai, N., Noobkaew, S. and Nacapricha, D. (2001), "A novel continuous-flow sequential extraction procedure for metal speciation in solids", *J. Environ. Qual.*, Vol. 30, pp. 1195-1205.

Shiva Kumar, D. and Srikantaswamy, S. (2014), "Factors Affecting on Mobility of Heavy Metals in Soil Environment", *International Journal for Scientific Research & Development*, Vol. 2 No. 3, pp. 201–203.

Skipperud, L. and Salbu, B. (2015), “Sequential extraction as a tool for mobility studies of radionuclides and metals in soils and sediments”, *Radiochimica Acta*, Vol. 103 No. 3, pp. 187–197.

Skipperud, L., Strømman, G., Yunusov, M., Stegnar, P., Uralbekov, B., Tilloboev, H., Zjazjev, G., *et al.* (2013), “Environmental impact assessment of radionuclide and metal contamination at the former U sites Taboshar and Digmai, Tajikistan”, *Journal of Environmental Radioactivity*, Elsevier Ltd, Vol. 123, pp. 50–62.

Smičiklas, I. and Šljivić-Ivanović, M. (2016), “Radioactive Contamination of the Soil: Assessments of Pollutants Mobility with Implication to Remediation Strategies”, in Larramendy, M.L. and Soloneski, S. (Eds.), *Soil Contamination - Current Consequences and Further Solutions*, InTech, Rijeka, Croatia, pp. 268–276.

Sohlenius, G., Saetre, P., Nordén, S., Grolander, S. and Sheppard, S. (2013), “Inferences about radionuclide mobility in soils based on the solid/liquid partition coefficients and soil properties”, *Ambio*, Vol. 42 No. 4, pp. 414–424.

Soltani, N., Keshavarzi, B., Moore, F., Sorooshian, A., Ahmadi, M.R., Sciences, A., Ore, I., *et al.* (2017), “Distribution of potentially toxic elements (PTEs) in tailings, soils, and plants around Gol-E-Gohar iron mine, a case study in Iran”, *Environ Sci Pollut Res Int.*, Vol. 24 No. 23, pp. 18798–18816.

de Souza Braz, A.M., Fernandes, A.R., Ferreira, J.R. and Alleoni, L.R.F. (2013), “Distribution coefficients of potentially toxic elements in soils from the eastern Amazon”, *Environmental Science and Pollution Research*, Vol. 20 No. 10, pp. 7231–7242.

de Souza, E.S., Fernandes, A.R., de Souza Braz, A.M., Sabino, L.L.L. and Alleoni, L.R.F. (2015), “Potentially toxic elements (PTEs) in soils from the surroundings of the Trans-Amazonian Highway, Brazil”, *Environmental Monitoring and Assessment*, Vol. 187 No. 1, available at:https://doi.org/10.1007/s10661-014-4074-1.

Stanojković-Sebić, A., Maksimović, J., Dinić, Z., Dobrivoj, P., Iličić, R., Stanojković, A. and Pivić, R. (2017), “Microelements and Heavy Metals Content in Frequently Utilized Medicinal Plants Collected from the Power Plant Area Microelements and Heavy Metals Content in Frequently Utilized Medicinal Plants Collected from the Power Plant Area”, *Natural Product Communications*, Vol. 12 No. February, pp. 185–188.

Statstutor. (2015), “Pearson ’ s Correlation”, available at: http://www.statstutor.ac.uk/resources/uploaded/pearsons.pdf.

Steed, S. and Reed, J. (2000), “Measuring pH of Soils”, *Geoderma*, Florida.

Stevanović, V., Gulan, L., Milenković, B., Valjarević, A., Zeremski, T. and Penjišević, I. (2018), “Environmental risk assessment of radioactivity and heavy metals in soil of Toplica region, South Serbia”, *Environmental Geochemistry and Health*, Vol. 40 No. 5, pp. 2101–2118.

Strok, M. (2012), “Transfer of natural radionuclides from hay and silage to cow ’ s milk in the vicinity of a former uranium mine”, Vol. 110, pp. 64–68.

Štrok, M. and Smodiš, B. (2013), “Soil-to-plant transfer factors for natural radionuclides in grass in the vicinity of a former uranium mine”, *Nuclear Engineering and Design*, Vol. 261, pp. 279–284.

Sungur, A., Soylak, M. and Ozcan, H. (2014), “Investigation of heavy metal mobility and availability by the BCR sequential extraction procedure: Relationship between soil properties and heavy metals availability", *Chemical Speciation and Bioavailability,* Vol. 26 No. 4, pp. 219–230.

Sungur, A., Soylak, M., Yilmaz, S. and Özcan, H. (2014), "Determination of heavy metals in sediments of the Ergene River by BCR sequential extraction method", *Environmental Earth Sciences,* Vol. 72 No. 9, pp. 3293–3305.

Sutherland, R.A. (2010), "BCR®-701: A review of 10-years of sequential extraction analyses", *Analytica Chimica Acta,* Elsevier B.V., Vol. 680 No. 1–2, pp. 10–20.

Tanaka, A., Minami, N., Yasuoka, Y., Iimoto, T., Omori, Y., Nagahama, H., Muto, J., *et al.* (2017), "Accurate measurement of indoor radon concentration using a low-effective volume radon monitor", *Radiation Protection Dosimetry,* Vol. 177 No. 3, pp. 324–330.

Templeton, D.M., Ariese, F., Cornelis, R., Danielsson, L.G., Muntau, H., Van Leeuwen, H.P. and Lobinski, R. (2000), "Guidelines for terms related to chemical speciation and fractionation of elements", Definitions, structural aspects, and methodological approaches. *Pure Appl. Chem.,* Vol. 72, pp. 1453-1470.

Tessier, A., Campbell, P.G.C. and Bisson, M. (1979), "Sequential Extraction Procedure for the Speciation of Particulate Trace Metals", *Analytical Chemistry,* Vol. 51 No. 7, pp. 844–851.

The Atomic Energy Corporation of South Africa Limited. (1988), *URANIUM IN SOUTH AFRICA,* Pretoria.

Thlaspi, by, Scott Angle, B.J., M Baker, A.J., Whiting, S.N. and Chaney, R.L. (2003), "Soil moisture effects on uptake of metals", *Plant and Soil,* Vol. 256, pp. 325–332.

Tlustos, P., Szakova, J., Starkova, A. and Pavlikova, D. (2005), "A comparison of sequential extraction procedures for fractionation of arsenic, cadmium, lead, and zinc in soil", *Central European Journal of Chemistry,* Vol. 3 No. 4, pp. 830–851.

Tomašić, M., Žgorelec, Ţ., Jurišić, A. and Kisić, I. (2013), "Cation exchange capacity of dominant soil types in the Republic of Croatia", *Journal of Central European Agriculture,* Journal of Central European Agriculture, Vol. 14 No. 3, pp. 84–98.

Topcuoğlu, B. (2016), "Heavy Metal Mobility and Bioavailability on Soil Pollution and", *Int'l Journal of Advances in Agricultural & Environmental Engg.,* Vol. 3 No. 1, pp. 208–213.

Trangos, G. and Bobbins, K. (2015), "Gold Mining Exploits and the Legacies of Johannesburg's Mining Landscapes", *Scenario Journal.*

Tripathi, R.M.Ã., Sahoo, S.K., Jha, V.N., Khan, A.H. and Puranik, V.D. (2008), "Assessment of environmental radioactivity at uranium mining , processing and tailings management facility at Jaduguda , India", Vol. 66, pp. 1666–1670.

Umoren, I.U., Udoh, A.P. and Udousoro, I.I. (2007), "Concentration and chemical speciation for the determination of Cu, Zn, Ni, Pb and Cd from refuse dump soils using the optimized BCR sequential extraction procedure", *Environmentalist,* Vol. 27 No. 2, pp. 241–252.

UNEP/OCHA. *Assessment Mission Report,* 2000, Geneva.

United States Environmental Protection Agency. (1999), "Understanding variation in partition coeffcient, Kd, values.", *Epa 402-R-99-004B,* Vol. II No. August 1999.

UNSCEAR. (2000), “Sources and Effects of Ionizing Radiation”, *United Nations Scientific Committee on the Effects of Atomic Radiation*, New York.

Ure, A.M., Quevauviller, Ph., Muntau, H. and Griepink, B. (1993), “Speciation of heavy-metals in soils and sediments - an account of the improvement and harmonization of extraction techniques undertaken under the auspices of the BCR of the Commission of the European Communities”, *Int. J.Envir. Anal. Chem.*, pp. 135-151.

Valencia-Avellan, M., Slack, R., Stockdale, A. and Mortimer, R.J.G. (2017), “Effect of episodic rainfall on aqueous metal mobility from historical mine sites”, *Environmental Chemistry*, Vol. 14 No. 8, pp. 469–475.

Valkovic, V. (1993), “Determination Of Radionuclides In Environmental Samples”, *Techniques and Instrumentation in Analytical Chemistry*, Vol. 13 No. C, pp. 311–356.

Violante, A., Cozzolino, V., Perelomov, L., Caporale, A.G. and Pigna, M. (2010), “Mobility and bioavailability of heavy metals and metalloids in soil environments”, *Journal of Soil Science and Plant Nutrition*, Vol. 10 No. 3, pp. 268–292.

Voitsekhovitch, O., Soroka, Y. and Lavrova, T. (2006), *Uranium Mining and Ore Processing in Ukraine – Radioecological Effects on the Dnipro River Water Ecosystem and Human Health, Radionuclides in the Environment: International Conference on Isotopes in Environmental Studies*, Vol. 8, Elsevier Masson SAS, available at:https://doi.org/10.1016/S1569-4860(05)08014-9.

Voss, R. (1998), “Micronutrients”.

VROM. (2000), “Circular on target values and intervention values for soil remediation”, *Netherlands Government Gazette*, Vol. 2000, p. 51.

Wade, P. and Coetzee, H. (2008), “Risk Assessment of Uranium in Selected Gold Mining Areas in South Africa”. In: Merkel, B.J. and Hasche-Berger, A. (Eds), *Uranium, Mining and Hydrogeology*, Springer, Berlin, Heidelberg.

Waggitt, P. (2008), “Uranium mining legacies remediation and renaissance development: an international overview”, in Merkel, B.J. and Hasche-berger, A. (Eds.), *Uranium, Mining and Hydrogeology*, Saxon State Ministry of Environment and Agriculture U, Freiberg, available at:https://doi.org/10.1007/978-3-540-87746-2.

Wallner, A. and Stein, P. (2012), *Uranium Mining in and for Europe*, Vienna.

WHO. (1996), *Water Quality Assessments - A Guide to Use of Biota, Sediments and Water in Enviromental Monitoring*, edited by Chapman, D.*University Press, Cambridge ISBN*, 2nd ed., E&FN Spon, an imprint of Chapman & Hall, London, Great Britain, available at:https://doi.org/10.4324/9780203476710.

WHO. (2004), *WHO Guidelines for Drinking Water Quality, Vol. 1. Recommendations, Environmental Pollution Series A, Ecological and Biological*, 3rd ed., Vol. 1, Geneva, available at:https://doi.org/10.1016/0143-1471(85)90051-0.

Wijaya, A.R., Farida, I., Wonorahardjo, S., Utomo, Y., Daupor, H., Hossain, M.S. and Kunisue, T. (2018), “BCR Sequential Leaching for Geochemical Fractions and Assessment of Fe, Ni, and Mn in the Coastal Sediments Sendang Biru Port, East Java, Indonesia”, *Journal of Physics: Conference Series 1093 012002*, Vol. 1093 No. 1, pp. 1–8.

Wilson, P.C. (2010), “Water Quality Notes : Alkalinity and Hardness 1 Ecological /

Management", *University of Florida, IFAS Extension,* Department of Soil and Water Sciences, United States of America.

Winde, F. (2010), "Uranium pollution of the wonderfonteinspruit, 1997-2008 part 1: Uranium toxicity, regional background and mining-related sources of uranium pollution", *Water SA,* Vol. 36 No. 3, pp. 239–256.

Winde, F. (2016), "Uranium pollution in South Africa: Past research and future needs", *Hrvatski Geografski Glasnik,* Vol. 77 No. 2, pp. 33–53.

Winde, F. and Sandham, L.A. (2004), "Uranium pollution of South African streams – An overview of the situation in gold mining areas of the Witwatersrand", pp. 131–149.

Winde, F., Wade, P. and Van Der Walt, I.J. (2004), "Gold tailings as a source of waterborne uranium contamination of streams - The Koekemoerspruit# (Klerksdorp goldfield, South Africa) as a case study. Part I of III: Uranium migration along the aqueous pathway", *Water SA,* Vol. 30 No. 2, pp. 219–225.

World Nuclear Association. (2020), "Uranium Enrichment", *World Nuclear Association,* available at: https://www.world-nuclear.org/information-library/nuclear-fuel-cycle/conversion-enrichment-and-fabrication/uranium-enrichment.aspx%0Ahttp://www.world-nuclear.org/information-library/nuclear-fuel-cycle/conversion-enrichment-and-fabrication/uranium-enrichme (accessed 19 February 2021).

Wuana, R.A. and Okieimen, F.E. (2011), "Heavy Metals in Contaminated Soils : A Review of Sources , Chemistry , Risks and Best Available Strategies for Remediation", Vol. 2011, available at:https://doi.org/10.5402/2011/402647.

Yan, L., Le, Q. Van, Sonne, C., Yang, Y., Yang, H., Gu, H., Ling, N., *et al.* (2021), "Phytoremediation of radionuclides in soil , sediments and water", *Journal of Hazardous Materials,* Elsevier B.V., Vol. 407 No. November 2020, p. 124771.

Yang, Z., Wang, Y., Shen, Z., Niu, J. and Tang, Z. (2009), "Distribution and speciation of heavy metals in sediments from the mainstream, tributaries, and lakes of the Yangtze River catchment of Wuhan, China", *Journal of Hazardous Materials,* Vol. 166 No. 2–3, pp. 1186–1194.

Yi, L., Gao, B., Liu, H., Zhang, Y., Du, C. and Li, Y. (2020), "Characteristics and assessment of toxic metal contamination in surface water and sediments near a Uranium mining area", *International Journal of Environmental Research and Public Health,* Vol. 17 No. 2, available at:https://doi.org/10.3390/ijerph17020548.

Závodská, L., Kosorínová, E., Scerbakova, L. and Lesny, J. (2008), "Environmental chemistry of uranium", *HUISSN 1418-7108: HEJ Manuscript No.: ENV-081221-A,* Vol. 17, pp. 1–18.

Žemberyová, M., Barteková, J. and Hagarová, I. (2006), "The utilization of modified BCR three-step sequential extraction procedure for the fractionation of Cd, Cr, Cu, Ni, Pb and Zn in soil reference materials of different origins", *Talanta,* Vol. 70, pp. 973-978.

Zgorelec, Z., Grahovac, B., Percin, A., Jurkovic, V., Gandjaeva, L. and Maurović, N. (2019), "Comparison of two different CEC determination methods regarding the soil properties", *Agriculturae Conspectus Scientificus,* Vol. 84 No. 2, pp. 151–158.

Zhang, G., Bai, J., Xiao, R., Zhao, Q., Jia, J., Cui, B. and Liu, X. (2017), "Heavy metal fractions and ecological risk assessment in sediments from urban, rural and reclamation-affected rivers of the Pearl River Estuary, China", *Chemosphere*, Vol. 184, available at:https://doi.org/10.1016/j.chemosphere.2017.05.155.

Zhu, X., Yang, F. and Wei, C. (2015), "Factors influencing the heavy metal bioaccessibility in soils were site dependent from different geographical locations", *Environmental Science and Pollution Research*, Vol. 22 No. 18, pp. 13939–13949.

Zimmerman, A.J. and Weindorf, D.C. (2010), "Heavy Metal and Trace Metal Analysis in Soil by Sequential Extraction: A Review of Procedures", *International Journal of Analytical Chemistry*, Vol. 2010, pp. 1–7, Article ID 387803 doi:10.1155/2010/387803.

A2. Cation exchange capacity (CEC)

Sample	**Soil depth**	**Ba^{+} conc. (mg/L)**	**Ba^{2+} conc.**	**Soil weight (g)**	**Volume prepared in (mL)**	**Ba^{2+} conc. (mg/kg)**	**Ba^{2+} conc. (g/kg)**	**CEC (g/kg) to (cmol(+)/kg)**	**Average CEC**
Rn-M11	0 – 25	6343.89	12687.79	4.0087	40	126602.53	126.60	60.80 ± 4.88	52.89 ± 3.87
		5566.13	11132.26	4.0433	40	110130.39	110.13	52.89 ± 2.87	
	25 – 50	2201.45	4402.91	4.007	40	43952.15	43.95	21.11 ± 0.07	20.17 ± 0.05
		2008.95	4017.90	4.0129	40	40049.82	40.05	19.23 ± 0.03	
	50 – 75	1434.19	2868.38	4.0025	40	28665.88	28.67	13.77 ± 1.92	14.55 ± 2.63
		1599.63	3199.26	4.0097	40	31915.22	31.92	15.33 ± 3.34	
Rn-M12	0 – 25	468.33	936.65	4.0018	40	9362.32	9.36	4.50 ± 0.49	32.80 ± 0.40
		6372.35	12744.70	4.0069	40	127227.53	127.23	61.10 ± 0.30	
	25 – 50	263.85	527.69	4.0349	40	5231.28	5.23	2.51 ± 0.11	8.23 ± 0.63
		1453.21	2906.41	4.0019	40	29050.33	29.05	13.95 ± 1.15	
	50 – 75	1602.97	3205.94	4.0025	40	32039.40	32.04	15.39 ± 1.51	24.42 ± 0.76
		3487.51	6975.01	4.0056	40	69652.61	69.65	33.45 ± 2.15E-03	
	75 - 100	3443.06	6886.13	4.013	40	68638.21	68.64	32.96 ± 1.32E-03	32.96 ± 1.32E-03
Rn-M13	0 – 25	2336.85	4673.71	4.023	40	46469.88	46.47	22.32 ± 0.51	28.15 ± 0.26
		3550.93	7101.86	4.0134	40	70781.50	70.78	33.99 ± 3.42E-03	
	25 – 50	2728.88	5457.77	4.0066	40	54487.77	54.49	26.17± 5.32	31.83 ± 3.07
		3904.99	7809.99	4.0016	40	78068.64	78.07	37.49 ± 0.81	
	50 – 75	3513.39	7026.77	4.0044	40	70190.53	70.19	33.71 ± 3.42E-03	28.08 ± 1.54
		2348.77	4697.54	4.0181	40	46763.80	46.76	22.46 ± 3.09	
	75 - 100	3931.26	7862.52	4.0037	40	78552.58	78.55	37.72 ± 5.32E-03	36.61 ± 0.03

		3698.30	7396.61	4.0026	40	73918.03	73.92	35.50 ± 0.06	
Radioactive sample	0 - 25	3421.16	6842.31	4.0051	40	68336.01	68.34	32.82 ± 0.01	27.26 ± 0.02
		2260.87	4521.75	4.0009	40	45207.29	45.21	21.71 ± 0.03	

Conc. is for the concentration

A3: Determination of soil organic matter (SOM %)

Sample	**Name**	**Weight (mg)**	**N (%)**	**C (%)**	**S (%)**	**C/N ratio**	**TC (%)**	**$CaCo_3$ (%)**	**Average TOC (%)**	**Average SOM (%)**
Rn-M11	0 – 25	841.70	0.06	1.86	0.02	31.3	1.87	7.8	0.93	1.61
		891.30	0.06	1.88	0.01	31.0				
	25 – 50	785.40	0.06	1.5	0.01	25.9	1.51	6.7	0.70	1.21
		867.90	0.06	1.51	0.01	24.9				
	50 – 75	857.20	0.11	3.67	0.01	33.9	3.69	18.3	1.49	2.56
		875.10	0.11	3.70	0.00	34.1				
Rn-M12	0 – 25	942.20	0.04	0.87	0.01	20.4	0.87	3.6	0.44	0.75
		894.50	0.04	0.87	0.01	20.1				
	25 – 50	854.90	0.03	0.88	0.01	27.4	0.91	4.5	0.37	0.64
		853.00	0.03	0.94	0.00	28.7				
	50 – 75	828.50	0.03	2.10	0.00	77.7	2.10	12.8	0.56	0.96
		837.30	0.03	2.09	0.00	74.7				
	75 – 100	881.90	0.03	2.21	0.00	76.7	2.22	13.1	0.65	1.11
		912.50	0.03	2.23	0.00	75.8				
Rn-M13	0 – 25	841.10	0.23	3.13	0.04	13.8	3.14	0.6	3.06	5.27
		868.50	0.22	3.14	0.04	14.0				
	25 – 50	908.80	0.16	2.20	0.02	13.7	2.21	2.2	1.94	3.34
		943.60	0.16	2.21	0.02	13.8				
	50 – 75	938.00	0.1	1.57	0.01	15.7	1.55	3.6	1.12	1.92
		825.80	0.1	1.53	0.01	15.5				
	75 – 100	839.60	0.08	1.77	0.01	21.2	1.77	6.2	1.02	1.76
		920.40	0.08	1.76	0.01	20.9				
Radioactive sample	0 – 25	918.50	0.02	1.04	0.02	47.3	1.03	2.2	0.76	1.31
		929.50	0.02	1.01	0.03	42.7				

A4.Specific activity concentration for the radionuclides in soil and plants

			Specific activity and uncertainty of the mother isotope of the decay chain (Bq/kg)									Specific activity (Bq/kg)		
			^{238}U			^{232}Th			^{235}U			^{40}K		
Sample description	Sample type	Sample depth	Sample activity density		Sample activity density deviation	Sample activity density		Sample activity density deviation	Sample activity density		Sample activity density deviation	Sample activity density		Sample activity density deviation
Rn-M11	soil	0 - 25	82.07	±	11.07	46.87	±	9.16	3.83	±	0.52	1878.20	±	100.85
		25 - 50	73.64	±	11.51	40.72	±	9.09	3.43	±	0.54	1955.58	±	101.22
		50 - 75	154.80	±	12.83	39.97	±	9.60	7.22	±	0.60	1919.41	±	110.56
	Plant		169.30	±	50.28	7.49	±	23.75	7.90	±	2.35	7461.08	±	448.61
Rn-M12	Soil	0 - 25	113.77	±	10.96	38.96	±	8.69	5.31	±	0.51	1779.02	±	107.66
		25 - 50	68.91	±	3.51	43.16	±	2.77	3.21	±	0.16	1927.76	±	32.55
		50 - 75	91.91	±	11.54	55.82	±	9.10	4.29	±	0.54	2104.21	±	110.75
		75 - 100	66.62	±	12.17	33.36	±	9.15	3.11	±	0.57	1855.70	±	102.52
	Plant		194.10	±	51.20	64.90	±	39.35	9.05	±	2.39	8411.01	±	464.00
Rn-M13	Soil	0 - 25	110.55	±	13.84	58.65	±	10.81	5.16	±	0.65	2537.66	±	134.48
		25 - 50	111.02	±	14.35	56.93	±	11.18	5.18	±	0.67	2640.42	±	128.36
		50 - 75	131.10	±	13.85	79.30	±	10.41	6.11	±	0.65	2070.92	±	127.64
		75 - 100	129.21	±	12.06	40.54	±	10.05	6.03	±	0.56	2077.12	±	114.86
	Plant		170.85	±	28.00	50.23	±	22.54	7.97	±	1.31	4415.99	±	256.30
Radioactive sample	Soil	0 - 25	1856.29	±	24.85	10.04	±	12.33	86.58	±	1.16	2172.75	±	117.22
	Plant	Unwashed sample	148.23	±	44.26	26.16	±	33.37	6.91	±	2.06	6969.89	±	389.13
	Plant	Washed sample	143.46	±	36.06	28.07	±	16.00	6.69	±	1.68	5266.51	±	331.36

A5. Activity concentration for the radionuclides in soil and plants for U-238 and Th-232 progenies

			Pb-212 (238.6 keV)			**Pb-214 (295.2 keV)**			**Ac-228 (338.3 keV)**			**Pb-214 (352 keV)**			**Tl-208 (583.1 keV)**		
Sample Identification	Sample type	Sample depth (cm)	Activity concentration (Bq/kg)		Sample activity deviation (Bq/kg)	Activity concentration (Bq/kg)		Sample activity deviation (Bq/kg)	Activity concentration (Bq/kg)		Sample activity deviation (Bq/kg)	Activity concentration (Bq/kg)		Sample activity deviation (Bq/kg)	Activity concentration (Bq/kg)		Sample activity deviation (Bq/kg)
Rn-M11	Soil	0 – 25	46.13	±	7.95	93.87	±	15.77	19.53	±	28.50	50.88	±	10.09	22.27	±	4.90
		25 - 50	40.49	±	7.94	85.04	±	16.72	65.73	±	29.84	86.08	±	9.62	12.99	±	5.59
		50 - 75	9.87	±	8.67	96.56	±	20.46	135.81	±	29.69	125.81	±	11.99	14.20	±	5.96
	Plant		n.d.			184.74	±	76.58	n.d			7.91	±	44.41	29.95	±	23.26
Rn-M12	Soil	0 – 25	54.53	±	7.23	71.14	±	16.62	59.54	±	27.80	74.21	±	9.62	15.39	±	5.13
		25 - 50	39.22	±	2.44	40.04	±	5.20	55.72	±	8.88	52.08	±	3.01	17.39	±	1.58
		50 - 75	36.97	±	8.32	57.00	±	18.57	84.80	±	29.65	53.66	±	9.97	24.37	±	5.48
		75 - 100	28.21	±	8.01	82.14	±	17.28	81.14	±	28.86	58.16	±	10.21	24.09	±	5.23
	Plant		68.28	±	32.80	232.55	±	74.01	92.88	±	130.16	190.81	±	42.32	98.44	±	21.83
Rn-M13	Soil	0 – 25	33.37	±	10.15	87.91	±	20.87	81.51	±	35.72	89.85	±	11.73	42.13	±	5.78
		25 - 50	57.46	±	9.51	83.37	±	21.22	109.06	±	35.33	82.56	±	11.74	40.62	±	6.09
		50 - 75	24.69	±	9.49	99.88	±	20.87	150.87	±	32.88	110.72	±	12.65	23.38	±	6.04
		75 - 100	35.10	±	8.88	103.77	±	18.56	65.33	±	32.19	69.44	±	11.15	25.35	±	5.42
	Plant		n.d.			102.34	±	41.11	191.84	±	73.39	79.27	±	23.30	9.07	±	12.59
Radioactive sample	Soil	0 - 25	n.d.			1958.69	±	40.62	1.84	±	41.93	1901.77	±	28.93	23.89	±	6.22
	Unwashed plant		37.12	±	27.19	50.80	±	64.55	4.49	±	109.67	121.18	±	35.78	63.03	±	17.48
	Washed plant		39.43	±	22.55	41.65	±	54.73	n.d.			81.66	±	31.46	32.69	±	15.26

A6. Activity concentration for the radionuclides in soil and plants for U-238 and Th-232 progenies

			Bi-214 (609.3 keV)			**Ac-228 (911.6 keV)**			**Ac-228 (969.1 keV)**			**Bi-214 (1120.3 keV)**		
Sample Identification	Sample type	Sample depth (cm)	Activity concentration (Bq/kg)		sample activity deviation (Bq/kg)	Activity concentration (Bq/kg)		sample activity deviation (Bq/kg)	Activity concentration (Bq/kg)		sample activity deviation (Bq/kg)	Activity concentration (Bq/kg)		sample activity deviation (Bq/kg)
Rn-M11	Soil	0 – 25	71.47	±	10.41	34.32	±	21.08	99.54	±	32.12	112.04	±	38.75
		25 - 50	85.51	±	9.99	100.99	±	18.35	43.68	±	35.12	37.94	±	40.57
		50 - 75	111.92	±	12.45	43.89	±	21.97	n.d.			284.92	±	43.78
	Plant		226.51	±	46.04	272.73	±	92.11	n.d.			258.03	±	174.61
Rn-M12	Soil	0 – 25	102.29	±	9.90	105.85	±	18.91	26.38	±	31.21	207.45	±	38.12
		25 - 50	62.38	±	3.17	70.99	±	5.98	60.31	±	10.39	121.15	±	12.30
		50 - 75	43.19	±	10.69	75.98	±	18.62	77.14	±	35.07	213.78	±	39.66
		75 - 100	71.89	±	10.55	77.94	±	20.36	n.d.			54.29	±	43.06
	Plant		176.10	±	44.86	178.36	±	79.21	n.d.			176.93	±	180.72
Rn-M13	Soil	0 – 25	107.28	±	12.80	159.37	±	21.43	77.60	±	39.24	157.16	±	48.23
		25 - 50	109.64	±	13.41	49.17	±	24.98	20.59	±	41.18	168.50	±	50.27
		50 - 75	95.70	±	13.30	128.58	±	22.97	118.25	±	38.77	218.09	±	47.94
		75 - 100	72.15	±	12.44	104.74	±	21.70	36.39	±	39.34	271.46	±	41.29
	Plant		118.81	±	26.03	108.47	±	50.81	0.00	±	0.00	383.00	±	98.13
Radioactive sample	Soil	0 - 25	1743.56	±	30.91	43.73	±	25.21	14.43	±	41.83	1821.13	±	80.22
	Plant Unwashed		204.09	±	39.08	186.26	±	68.91	n.d.			216.85	±	156.10
	Plant Washed		187.99	±	32.32	93.76	±	57.94	40.17	±	110.02	262.54	±	125.60

A7. Rn-M11 pseudo total concentration of PTEs in soil and plants

Rn-M11																
Sample type	**Soil**												**Plant**			
Depth (cm)	**0 - 25**				**25 - 50**				**50 - 75**							
Element	Total concentration (mg/kg)		Standard deviation (SD)	Uncertainty (mg/kg)	Total concentration (mg/kg)		Standard deviation (SD)	Uncertainty (mg/kg)	Total concentration (mg/kg)		Standard deviation (SD)	Uncertainty (mg/kg)	Total concentration (mg/kg)		Standard deviation (SD)	Uncertainty (mg/kg)
Ca	20860.91	±	150.16	86.69	17476.40	±	48.63	28.07	38540.63	±	789.30	455.70	5710.41	±	54.26	31.33
Cd	n.d.				n.d.				n.d.				n.d.			
Co	6.16	±	0.07	0.04	6.22	±	0.04	0.02	5.68	±	0.05	0.03	n.d.			
Cr	47.10	±	0.20	0.11	40.49	±	0.31	0.18	33.39	±	0.01	3.86E-03	14.19	±	0.02	0.01
Cu	11.31	±	0.06	0.03	13.26	±	0.09	0.05	17.89	±	0.14	0.08	5.89	±	0.09	0.05
Fe	12103.95	±	138.58	80.01	12548.33	±	118.12	68.20	11050.04	±	87.35	50.43	265.23	±	3.60	2.08
K	7724.75	±	9.58	5.53	7951.40	±	38.35	22.14	6359.99	±	28.72	16.58	7205.72	±	20.60	11.89
Mg	5888.18	±	15.01	8.67	5338.84	±	85.29	49.24	8040.88	±	13.42	7.75	1781.29	±	13.81	7.97
Mn	316.99	±	2.04	1.18	339.87	±	5.28	3.05	273.48	±	3.63	2.10	34.23	±	0.13	0.07
Na	365.93	±	3.27	1.89	361.10	±	1.94	1.12	354.03	±	5.15	2.98	73.89	±	0.53	0.31
Ni	23.17	±	0.10	0.06	21.22	±	0.08	0.05	18.12	±	0.05	0.03	5.48	±	0.06	0.03
P	5543.60	±	36.61	21.14	5785.68	±	132.20	76.32	5686.71	±	48.97	28.27	1410.64	±	0.48	0.28
Pb	7.58	±	0.01	0.01	7.87	±	0.24	0.14	8.91	±	0.07	0.04	n.d.			
U	6.59	±	0.89	0.51	5.91	±	0.92	0.53	12.40	±	1.03	0.59	13.60	±	4.04	2.33
Zn	30.13	±	0.37	0.21	39.19	±	0.08	0.05	60.52	±	0.08	0.05	20.91	±	0.05	0.03

A8. Rn-M12 pseudo total concentration of PTEs in soil and plants

Rn-M12																				
Sample type	**Soil**																**Plant**			
Depth (cm)	**0 - 25**				**25 - 50**				**50 - 75**				**75 - 100**							
Element	Total concentration (mg/kg)		Standard deviation (SD)	Uncertainty (mg/kg)	Total concentration (mg/kg)		Standard deviation (SD)	Uncertainty (mg/kg)	Total concentration (mg/kg)		Standard deviation (SD)	Uncertainty (mg/kg)	Total concentration (mg/kg)		Standard deviation (SD)	Uncertainty (mg/kg)	Total concentration (mg/kg)		Standard deviation (SD)	Uncertainty (mg/kg)
Ca	10641.42	±	146.31	84.47	14872.52	±	87.26	50.38	35868.25	±	271.38	156.68	37406.47	±	321.48	185.61	4399.59	±	15.61	9.01
Cd	n.d.				n.d.				n.d.				n.d.				n.d.			
Co	7.15	±	0.02	0.01	8.14	±	0.01	4.77E-03	7.42	±	0.05	0.03	7.27	±	0.06	0.03	n.d.			
Cr	44.39	±	0.27	0.15	116.43	±	0.36	0.21	38.47	±	0.05	0.03	42.90	±	0.44	0.25	n.d.			
Cu	11.73	±	0.08	0.05	13.47	±	0.07	0.04	12.71	±	0.02	0.01	13.34	±	0.13	0.07	18.30	±	2.10E-03	1.21E-03
Fe	13816.64	±	168.64	97.36	15822.87	±	233.27	134.68	15784.06	±	74.03	42.74	15819.69	±	187.76	108.40	94.93	±	0.56	0.32
K	8045.29	±	9.62	5.55	7400.34	±	59.60	34.41	7136.54	±	28.95	16.71	8159.24	±	20.43	11.80	7607.97	±	151.90	87.70
Mg	4624.97	±	25.41	14.67	5683.93	±	7.20	4.16	9862.23	±	67.63	39.04	9897.33	±	32.85	18.97	1785.93	±	8.77	5.06
Mn	422.07	±	6.21	3.59	431.24	±	1.72	0.99	359.35	±	0.29	0.17	343.47	±	1.52	0.88	21.54	±	0.20	0.11
Na	373.89	±	0.02	0.01	359.38	±	1.82	1.05	383.83	±	2.19	1.27	432.82	±	2.91	1.68	43.02	±	0.21	0.12
Ni	23.91	±	0.06	0.04	53.47	±	0.75	0.43	20.99	±	0.12	0.07	22.18	±	0.07	0.04	2.42	±	0.02	0.01
P	5990.72	±	41.75	24.11	6315.55	±	29.05	16.77	7012.80	±	48.87	28.21	7234.93	±	67.42	38.93	1150.10	±	3.60	2.08
Pb	7.74	±	0.01	3.79E-03	8.41	±	0.06	0.04	7.54	±	0.56	0.32	7.75	±	0.01	3.75E-03	n.d.			
U	9.14	±	0.88	0.51	5.54	±	0.28	0.16	7.38	±	0.92	0.53	5.35	±	0.98	0.57	15.60	±	4.11	2.38
Zn	32.09	±	0.45	0.26	37.91	±	0.20	0.11	39.22	±	0.22	0.13	40.00	±	0.19	0.11	9.41	±	0.05	0.03

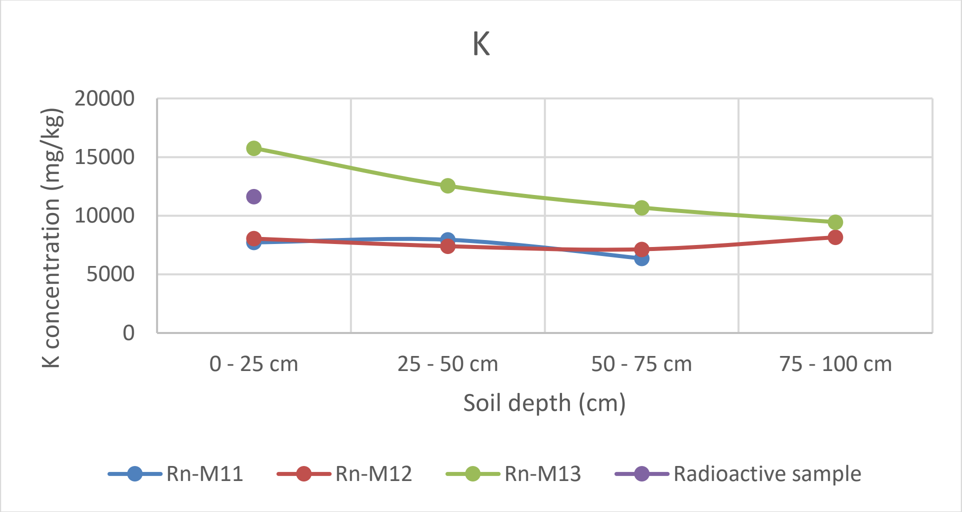
A9. Rn-M13 pseudo total concentration of PTEs in soil and plants

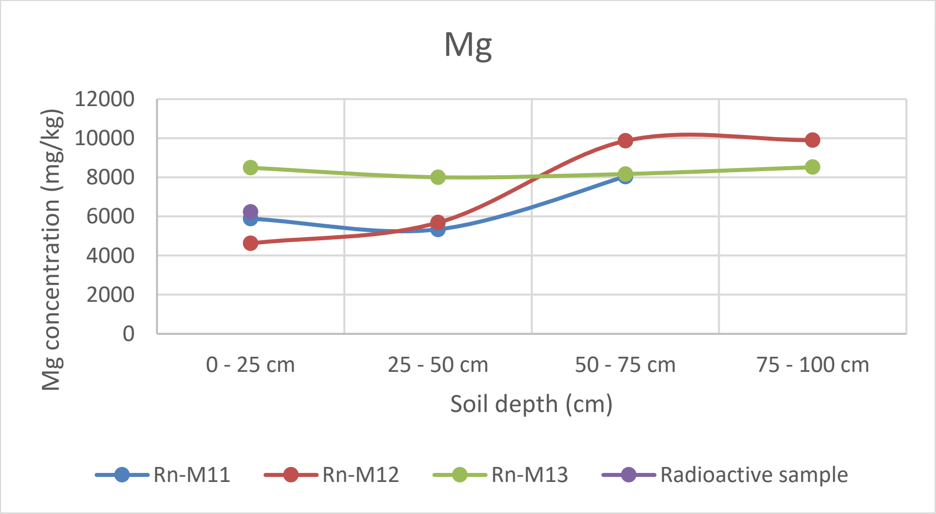
Rn-M13																				
Sample type	**Soil**																**Plant**			
Depth (cm)	**0 - 25**				**25 - 50**				**50 - 75**				**75 - 100**							
Element	Total concentration (mg/kg)		Standard deviation (SD)	Uncertainty (mg/kg)	Total concentration (mg/kg)		Standard deviation (SD)	Uncertainty (mg/kg)	Total concentration (mg/kg)		Standard deviation (SD)	Uncertainty (mg/kg)	Total concentration (mg/kg)		Standard deviation (SD)	Uncertainty (mg/kg)	Total concentration (mg/kg)		Standard deviation (SD)	Uncertainty (mg/kg)
Ca	7528.92	±	13.50	7.79	12502.81	±	100.47	58.01	14241.96	±	42.20	24.37	19613.93	±	31.49	18.18	2132.77	±	11.94	6.89
Cd	n.d.				n.d.				n.d.				n.d.				n.d.			
Co	9.53	±	0.09	0.05	9.45	±	0.03	0.02	9.15	±	0.07	0.04	8.21	±	0.07	0.04	n.d.			
Cr	62.97	±	0.22	0.13	55.89	±	0.34	0.20	52.64	±	0.21	0.12	46.63	±	0.07	0.04	n.d.			
Cu	16.35	±	0.01	0.00	17.65	±	0.36	0.21	16.77	±	0.05	0.03	15.18	±	0.02	0.01	59.50	±	0.11	0.07
Fe	21139.22	±	108.88	62.86	20802.79	±	20.76	11.98	20491.69	±	340.54	196.61	18229.52	±	256.31	147.98	301.45	±	2.01	1.16
K	15762.72	±	42.50	24.54	12551.91	±	51.42	29.69	10692.24	±	155.23	89.62	9455.13	±	86.18	49.76	8860.64	±	27.63	15.95
Mg	8487.81	±	42.52	24.55	8003.76	±	1.48	0.86	8170.15	±	19.23	11.10	8516.88	±	7.30	4.21	784.89	±	0.87	0.51
Mn	492.43	±	0.60	0.35	520.88	±	0.86	0.50	498.26	±	1.39	0.80	383.88	±	1.96	1.13	26.10	±	0.50	0.29
Na	398.55	±	3.34	1.93	476.14	±	0.32	0.19	490.82	±	0.26	0.15	438.29	±	2.77	1.60	36.90	±	0.43	0.25
Ni	29.65	±	0.03	0.02	27.28	±	0.12	0.07	26.57	±	0.19	0.11	22.95	±	0.11	0.06				n.d.
P	9283.47	±	128.48	74.18	9429.05	±	117.15	67.64	9449.72	±	49.47	28.56	8586.58	±	146.44	84.55	1307.24	±	0.59	0.34
Pb	13.73	±	0.03	0.02	14.33	±	0.52	0.30	12.86	±	0.08	0.05	11.72	±	0.69	0.40	n.d.			
U	8.88	±	1.11	0.64	8.92	±	1.15	0.66	10.40	±	1.11	0.64	10.40	±	0.97	0.56	13.73	±	2.25	1.30
Zn	67.43	±	0.12	0.07	58.92	±	0.08	0.04	54.51	±	0.12	0.07	47.46	±	0.24	0.14	13.56	±	0.05	0.03

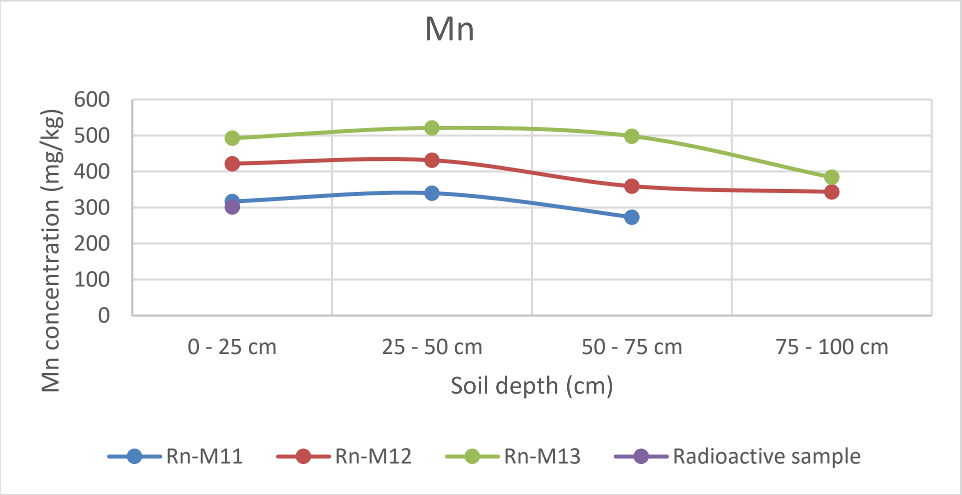
A10. Radioactive sample pseudo total concentration of PTEs in soil and plants

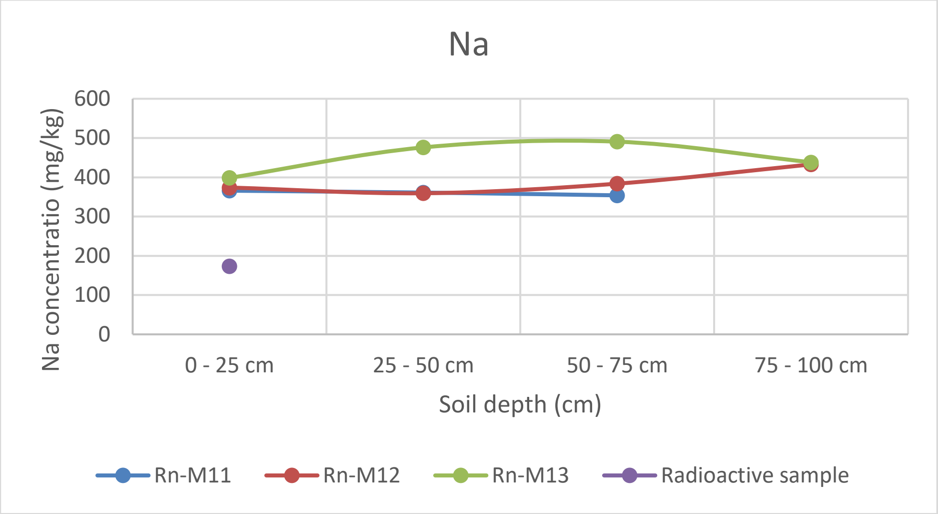
Radioactive sample												
Sample type	**Soil**				**Plant**							
Depth (cm)	**0 - 25**				**Unwashed**				**Washed**			
Element	Total concentration (mg/kg)		Standard deviation (SD)	Uncertainty (mg/kg)	Total Concentration (mg/kg)		Standard deviation (SD)	Uncertainty (mg/kg)	Total concentration (mg/kg)		Standard deviation (SD)	Uncertainty (mg/kg)
Ca	9116.44	±	43.16	24.92	5368.94	±	0.43	0.25	5184.20	±	43.05	24.85
Cd			n.d.				n.d.				n.d.	
Co	7.87	±	3.78E-03	2.18E-03			n.d.				n.d.	
Cr	126.83	±	0.01	0.01			n.d.				n.d.	
Cu	40.25	±	0.05	0.03	21.65	±	0.22	0.13	73.09	±	0.17	0.10
Fe	8013.97	±	57.10	32.96	52.39	±	0.37	0.21	68.59	±	0.35	0.20
K	11623.50	±	10.69	6.17	13289.26	±	12.76	7.37	14301.76	±	5.50	3.17
Mg	6239.30	±	69.62	40.19	868.42	±	7.44	4.30	925.47	±	2.93	1.69
Mn	301.52	±	0.17	0.10	3.68	±	0.04	0.02	8.97	±	0.10	0.06
Na	173.28	±	2.29	1.32	33.53	±	1.51	0.87	52.84	±	0.50	0.29
Ni	40.54	±	0.10	0.06			n.d.				n.d.	
P	3250.14	±	7.63	4.40	1489.16	±	15.61	9.02	1631.12	±	6.84	3.95
Pb	44.00	±	1.11	0.64			n.d.				n.d.	
U	149.00	±	1.20	0.69	11.91	±	3.56	2.05	11.53	±	2.90	1.67
Zn	36.05	±	0.38	0.22	8.87	±	0.03	0.02	13.34	±	0.39	0.22

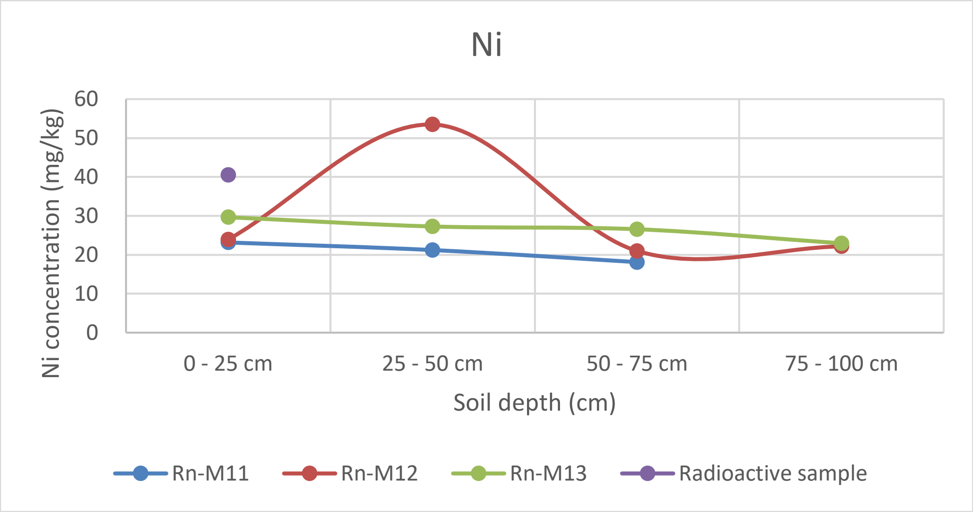
A11. Tendencies for each element in the soil in terms of depths in each sample

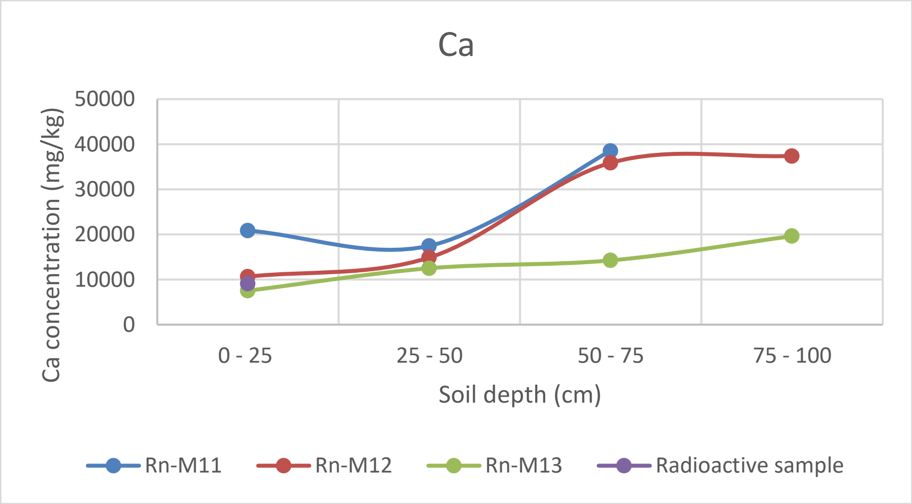


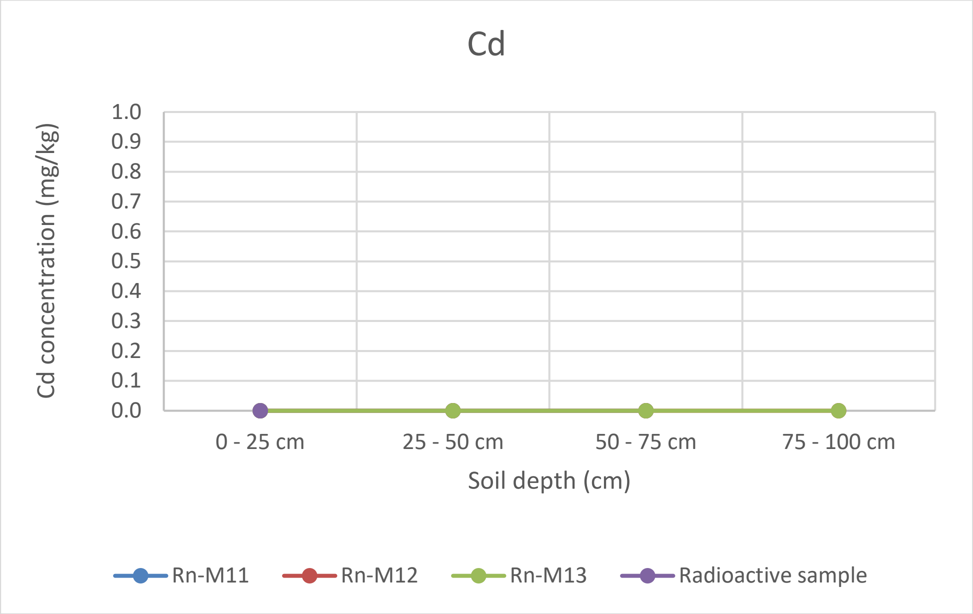


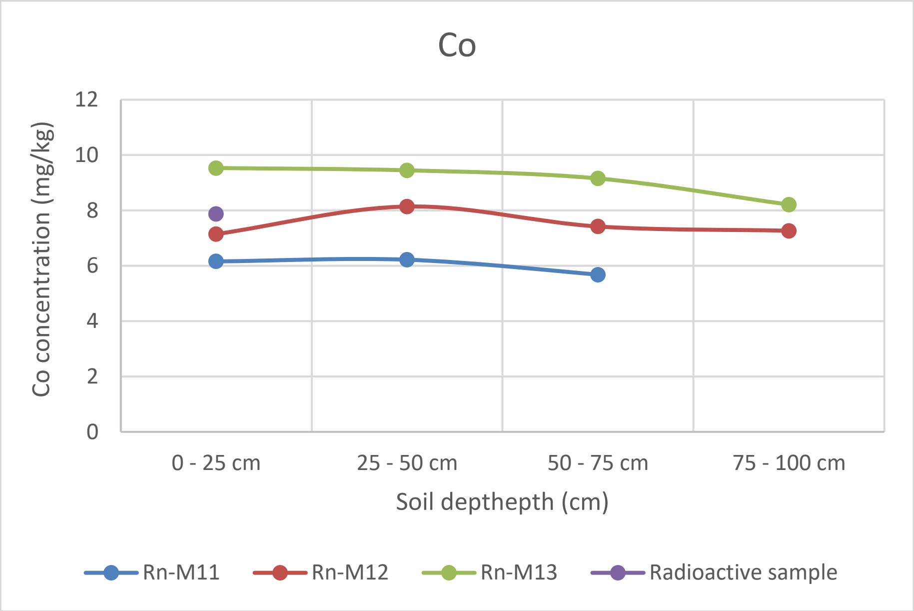


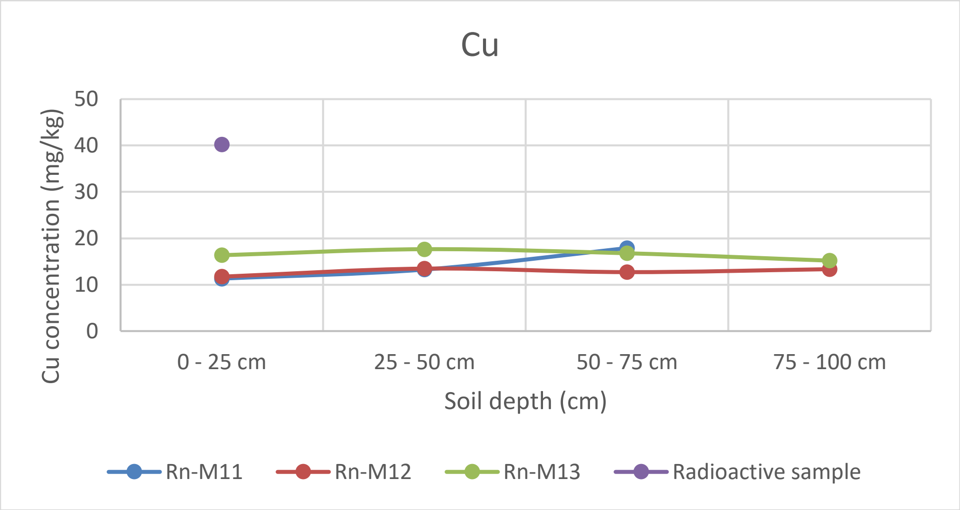

Na
Na concentratio (mg/kg)
600
500
400
300
200
100
0
0 - 25 cm
25 - 50 cm
50 - 75 cm
75 - 100 cm
Soil depth (cm)
Rn-M11
Rn-M12
Rn-M13
Radioactive sample

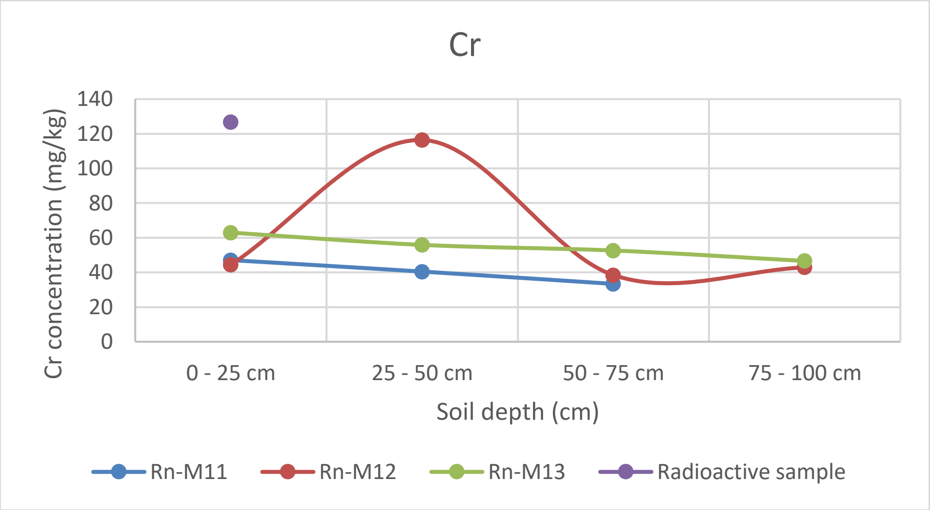

Ni
Ni concentration (mg/kg)
60
50
40
30
20
10
0
0 - 25 cm
25 - 50 cm
50 - 75 cm
75 - 100 cm
Soil depth (cm)
Rn-M11
Rn-M12
Rn-M13
Radioactive sample

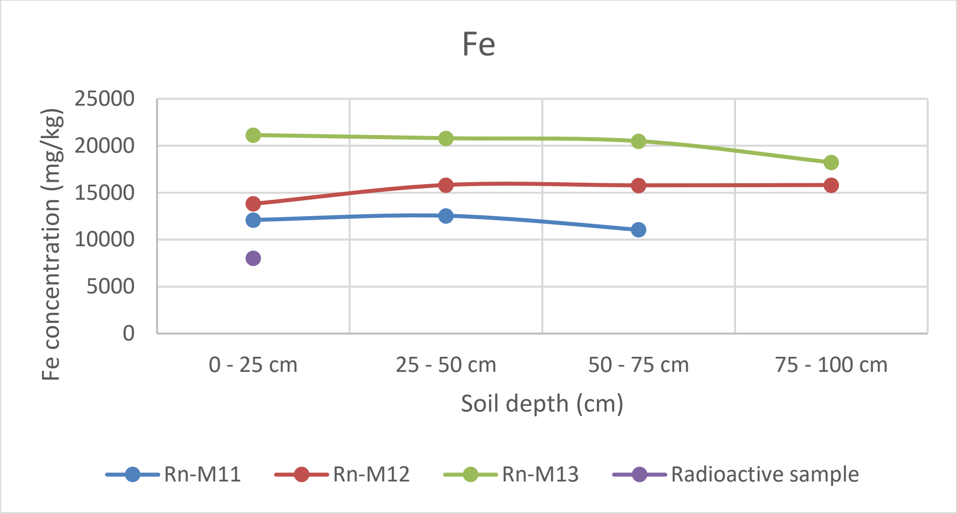

Ca
Ca concentration (mg/kg)
50000
40000
30000
20000
10000
0
0 - 25
25 - 50
50 - 75
75 - 100
Soil depth (cm)
Rn-M11
Rn-M12
Rn-M13
Radioactive sample

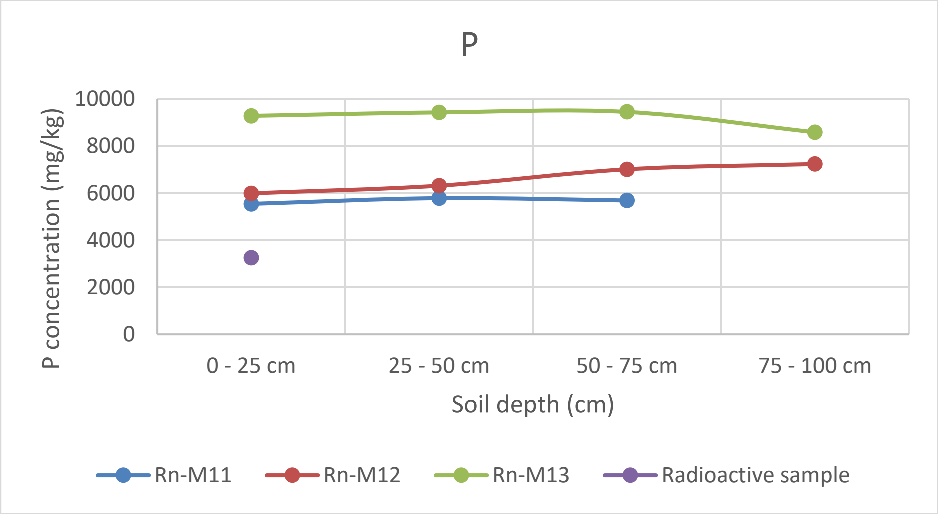

Cd
Cd concentration (mg/kg)
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.0
0 - 25 cm
25 - 50 cm
50 - 75 cm
75 - 100 cm
Soil depth (cm)
Rn-M11
Rn-M12
Rn-M13
Radioactive sample

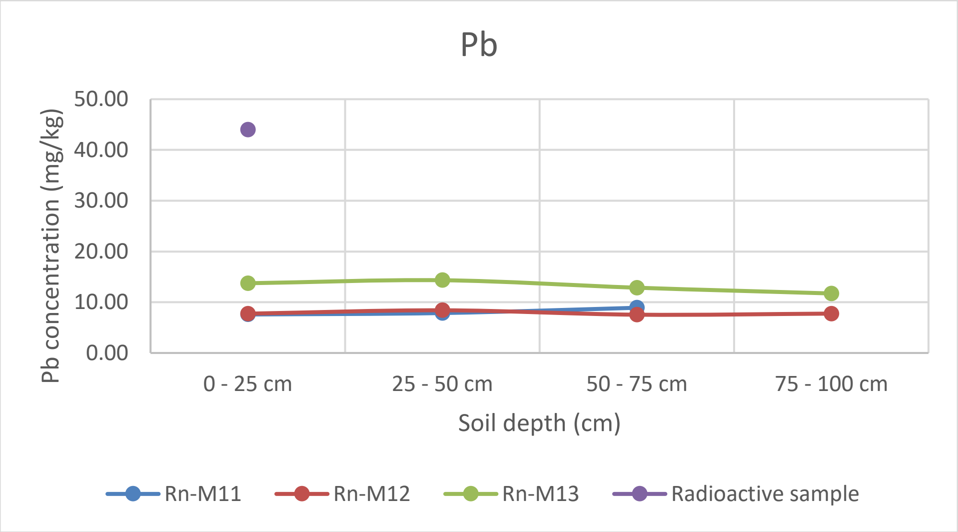

Co
Co concentration (mg/kg)
12
10
8
6
4
2
0
0 - 25 cm
25 - 50 cm
50 - 75 cm
75 - 100 cm
Soil depthepth (cm)
Rn-M11
Rn-M12
Rn-M13
Radioactive sample

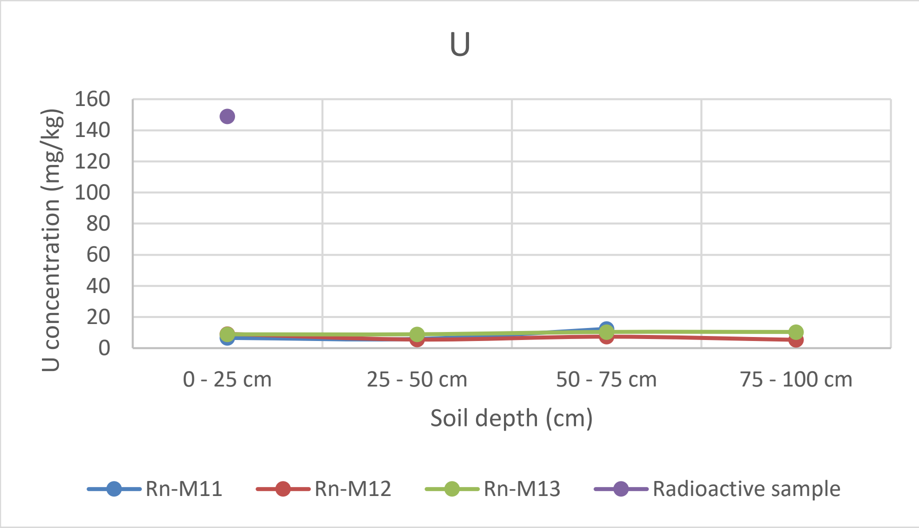

Cu
Cu concentration (mg/kg)
50
40
30
20
10
0
0 - 25 cm
25 - 50 cm
50 - 75 cm
75 - 100 cm
Soil depth (cm)
Rn-M11
Rn-M12
Rn-M13
Radioactive sample

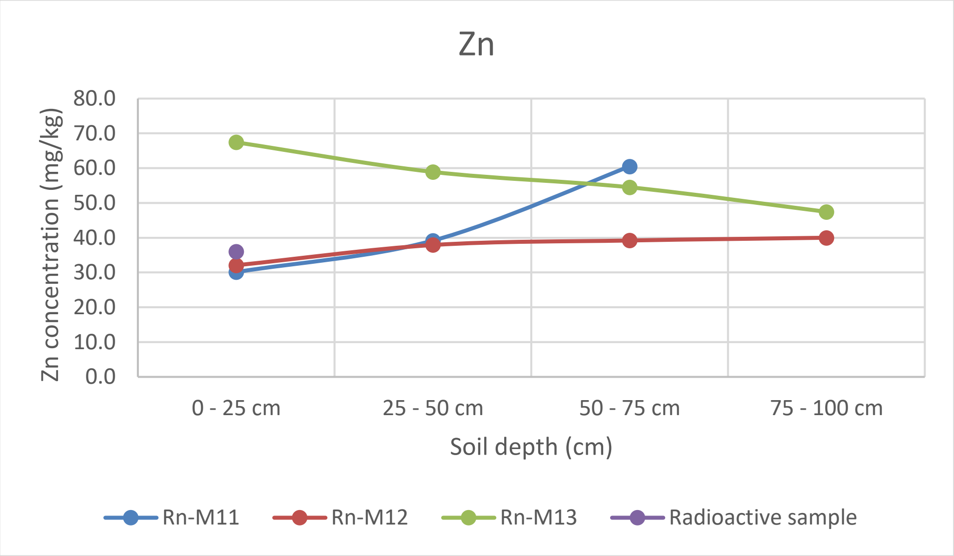

Cr
Cr concentration (mg/kg)
140
120
100
80
60
40
20
0
0 - 25 cm
25 - 50 cm
50 - 75 cm
75 - 100 cm
Soil depth (cm)
Rn-M11
Rn-M12
Rn-M13
Radioactive sample

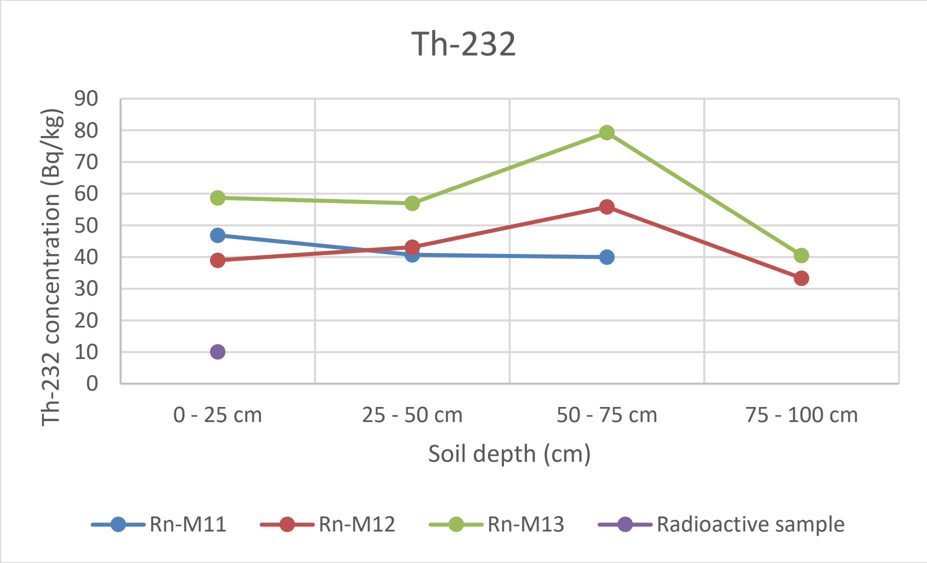

Fe
Fe concentration (mg/kg)
25000
20000
15000
10000
5000
0
0 - 25 cm
25 - 50 cm
50 - 75 cm
75 - 100 cm
Soil depth (cm)
Rn-M11
Rn-M12
Rn-M13
Radioactive sample

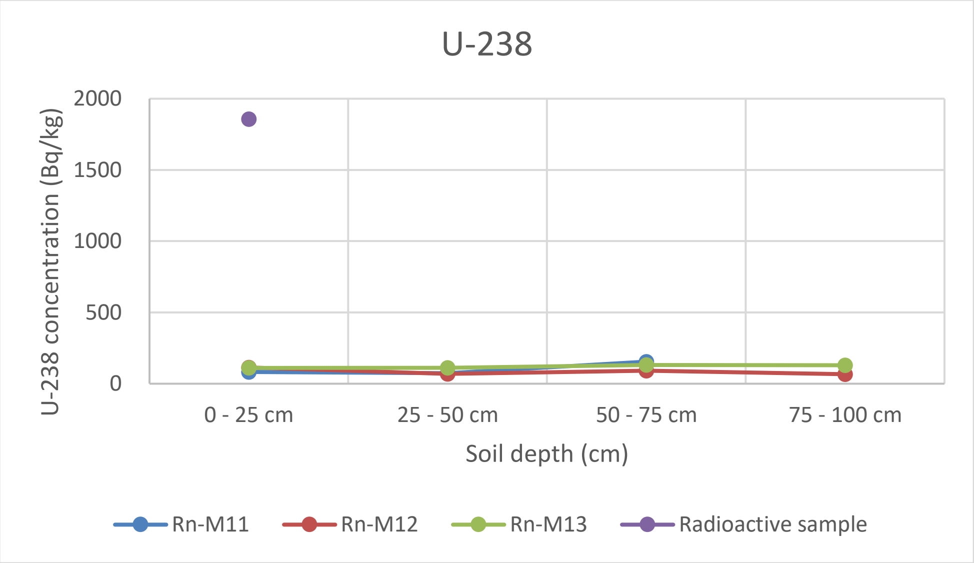

P
P concentration (mg/kg)
10000
8000
6000
4000
2000
0
0 - 25 cm
25 - 50 cm
50 - 75 cm
75 - 100 cm
Soil depth (cm)
Rn-M11
Rn-M12
Rn-M13
Radioactive sample

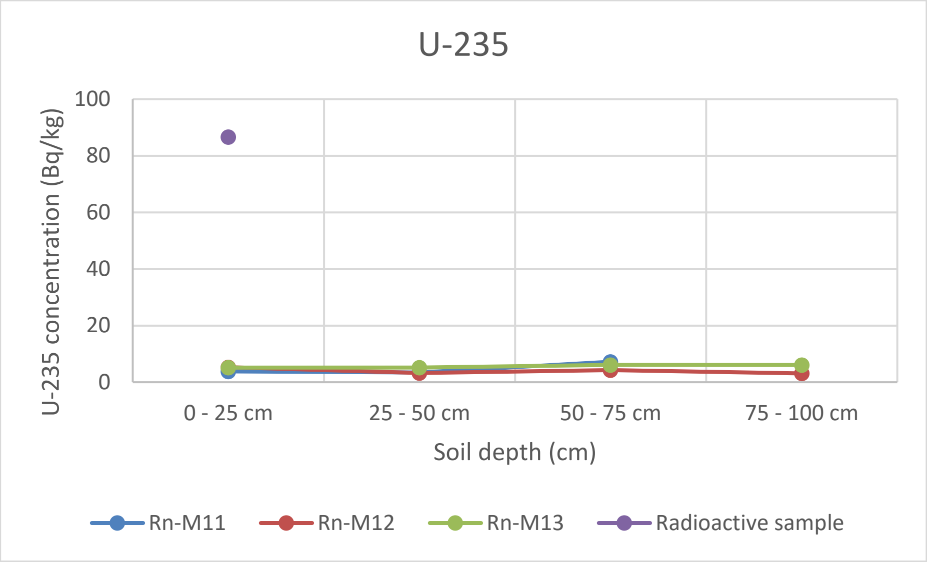

Pb
Pb concentration (mg/kg)
50.00
40.00
30.00
20.00
10.00
0.00
0 - 25 cm
25 - 50 cm
50 - 75 cm
75 - 100 cm
Soil depth (cm)
Rn-M11
Rn-M12
Rn-M13
Radioactive sample

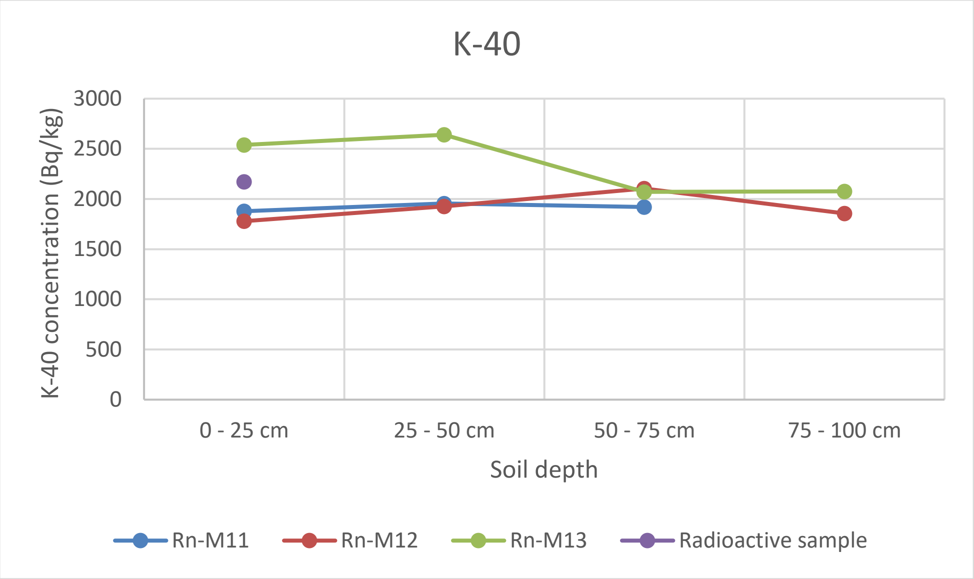

U
U concentration (mg/kg)
160
140
120
100
80
60
40
20
0
0 - 25 cm
25 - 50 cm
50 - 75 cm
75 - 100 cm
Soil depth (cm)
Rn-M11
Rn-M12
Rn-M13
Radioactive sample


Zn
Zn concentration (mg/kg)
80.0
70.0
60.0
50.0
40.0
30.0
20.0
10.0
0.0
0 - 25 cm
25 - 50 cm
50 - 75 cm
75 - 100 cm
Soil depth (cm)
Rn-M11
Rn-M12
Rn-M13
Radioactive sample

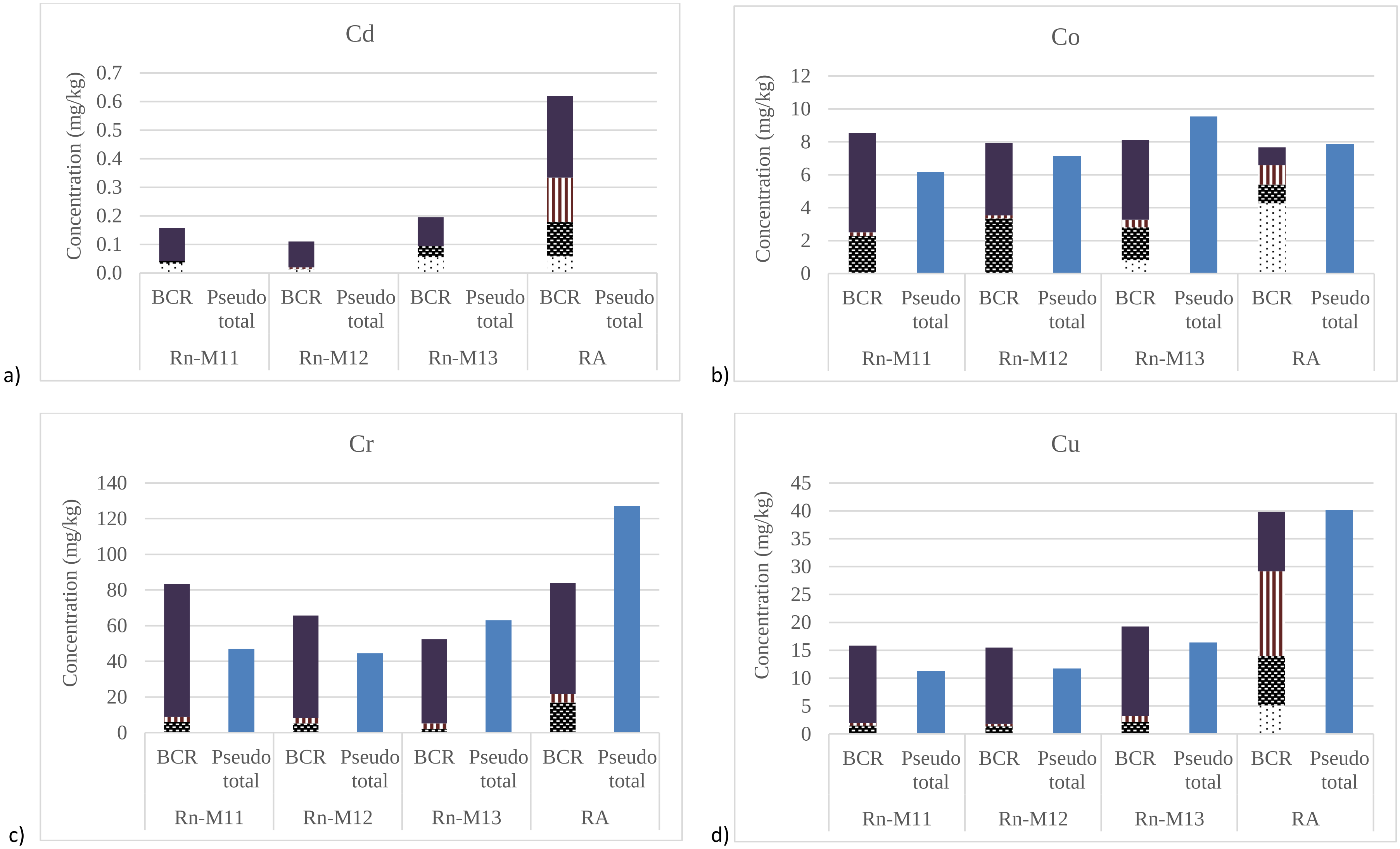

Th-232
Th-232 concentration (Bq/kg)
90
80
70
60
50
40
30
20
10
0
0 - 25 cm
25 - 50 cm
50 - 75 cm
75 - 100 cm
Soil depth (cm)
Rn-M11
Rn-M12
Rn-M13
Radioactive sample

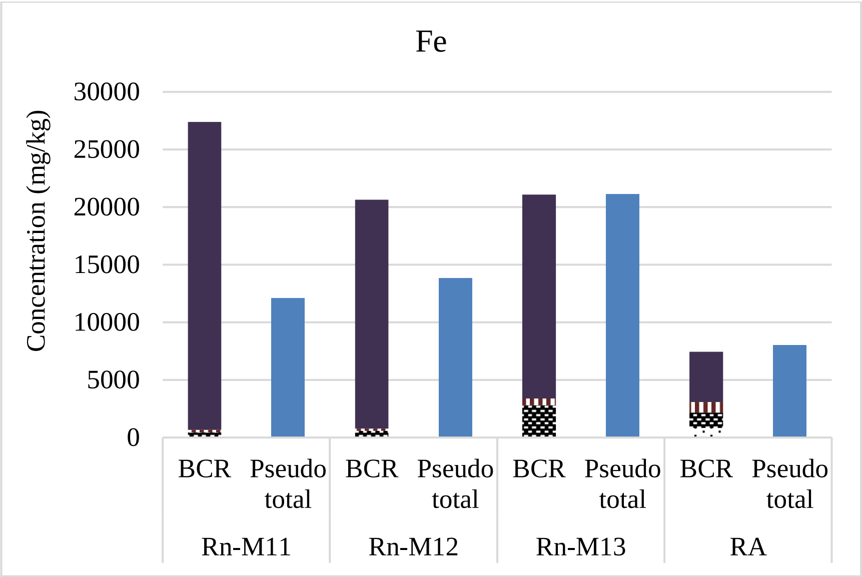

U-238
U-238 concentration (Bq/kg)
2000
1500
1000
500
0
0 - 25 cm
25 - 50 cm
50 - 75 cm
75 - 100 cm
Soil depth (cm)
Rn-M11
Rn-M12
Rn-M13
Radioactive sample


U-235
U-235 concentration (Bq/kg)
100
80
60
40
20
0
0 - 25 cm
25 - 50 cm
50 - 75 cm
75 - 100 cm
Soil depth (cm)
Rn-M11
Rn-M12
Rn-M13
Radioactive sample

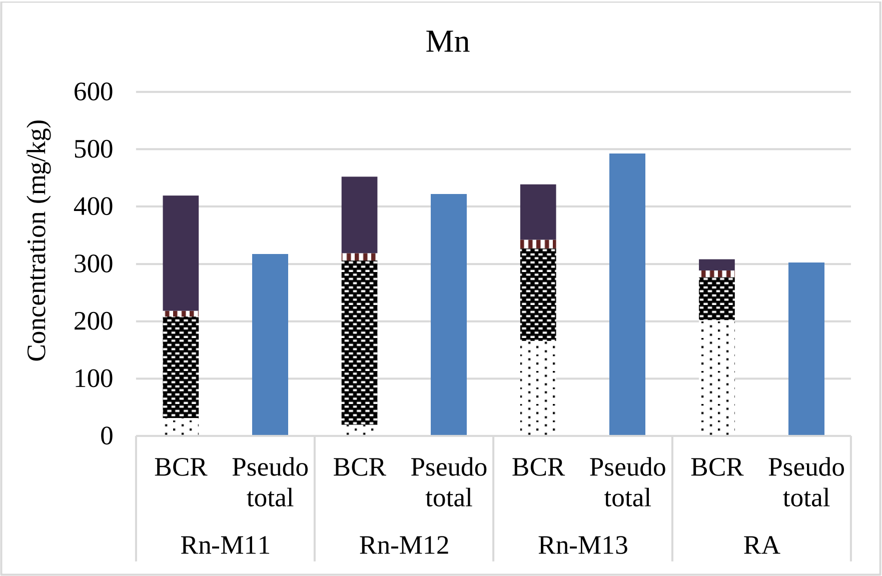

K-40
K-40 concentration (Bq/kg)
3000
2500
2000
1500
1000
500
0
0 - 25 cm
25 - 50 cm
50 - 75 cm
75 - 100 cm
Soil depth
Rn-M11
Rn-M12
Rn-M13
Radioactive sample

A12. A comparison for the sum of the BCR sequential extraction concentrations and the total concentrations of a) Cd, b) Co, c) Cr, d) Cu, e) Fe, f) Mn, g) Ni, h) Pb, and i) Zn in soil

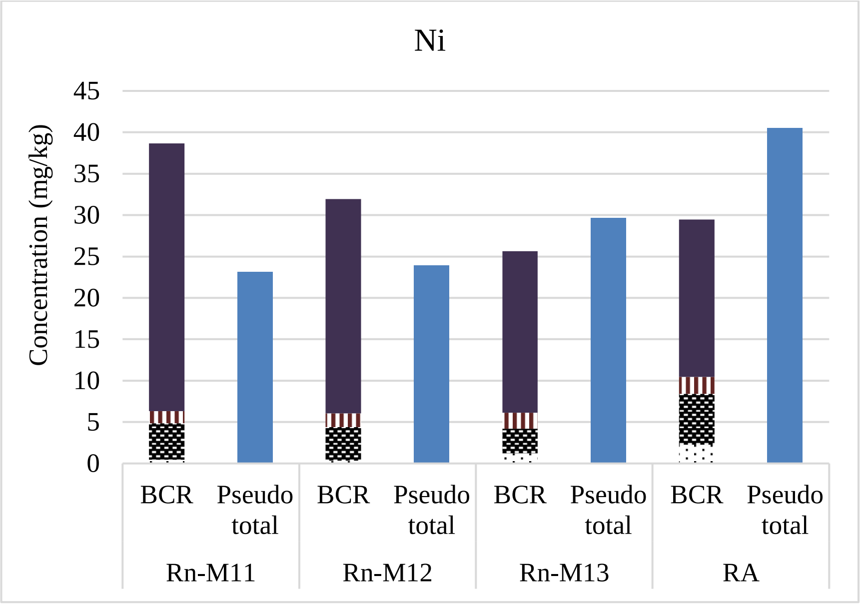



Fe
Concentration (mg/kg)
30000
25000
20000
15000
10000
5000
0
BCR
Pseudo total
Rn-M11
Rn-M12
Rn-M13
RA

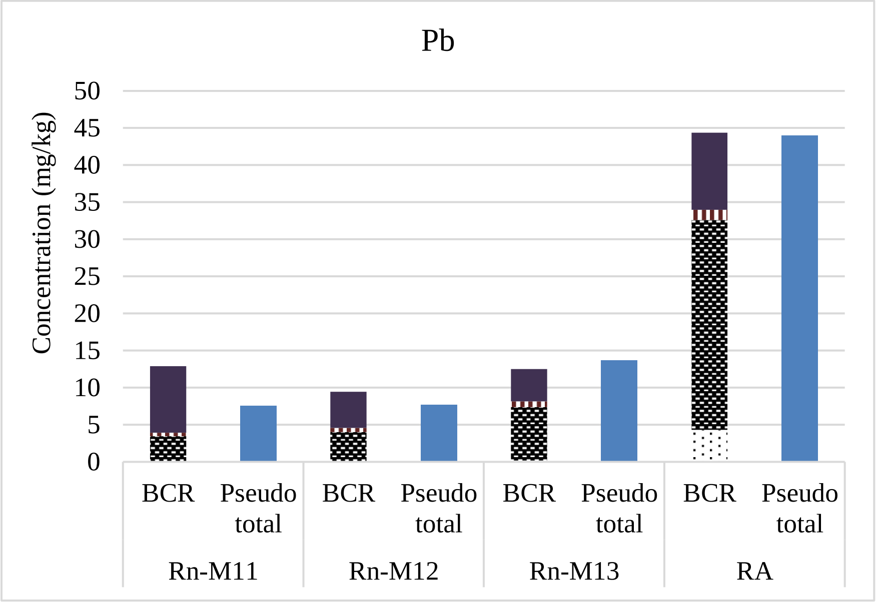
e)


Mn
Concentration (mg/kg)
600
500
400
300
200
100
0
BCR
Pseudo total
Rn-M11
Rn-M12
Rn-M13
RA

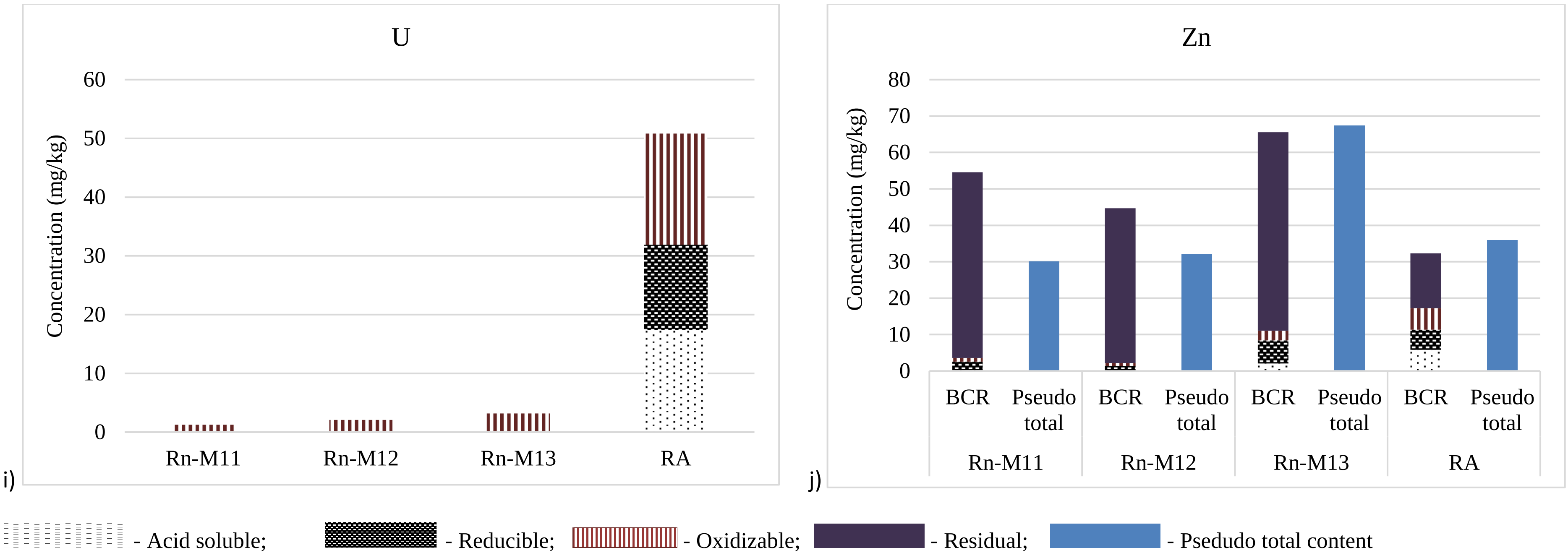
f)


Ni
Concentration (mg/kg)
45
40
35
30
25
20
15
10
5
0
BCR
Pseudo total
Rn-M11
Rn-M12
Rn-M13
RA

g)


Pb
Concentration (mg/kg)
50
45
40
35
30
25
20
15
10
5
0
BCR
Pseudo total
Rn-M11
Rn-M12
Rn-M13
RA

h)


i)
U
Concentration (mg/kg)
0
10
20
30
40
50
60
Rn-M11
Rn-M12
Rn-M13
RA
j)
Zn
Concentration (mg/kg)
0
10
20
30
40
50
60
70
80
BCR
Pseudo total
BCR
Pseudo total
BCR
Pseudo total
BCR
Pseudo total
Rn-M11
Rn-M12
Rn-M13
RA
- Acid soluble;
- Reducible;
- Oxidizable;
- Residual;
- Psedudo total content

A13. Descriptive statistics for soil characteristics

Descriptive Statistics for Rn-M11

	N	Minimum	Maximum	Sum	Mean	Std. Deviation	Variance	Skewness		Kurtosis	
	Statistic	Statistic	Statistic	Statistic	Statistic	Statistic	Statistic	Statistic	Std. Error	Statistic	Std. Error
CEC (cmol(+)/kg)	3	14.55	52.89	87.61	29.2033	20.70482	428.690	1.590	1.225	-	-
pH	3	5.87	6.42	18.51	6.1700	0.27839	0.078	-0.782	1.225	-	-
Soil_moisture (%)	3	7.44	12.60	29.28	9.7600	2.61901	6.859	0.858	1.225	-	-
SOM (%)	3	1.21	2.56	5.38	1.7933	0.69342	0.481	1.107	1.225	-	-

Descriptive Statistics for Rn-M12

	N	Minimum	Maximum	Sum	Mean	Std. Deviation	Variance	Skewness		Kurtosis	
	Statistic	Statistic	Statistic	Statistic	Statistic	Statistic	Statistic	Statistic	Std. Error	Statistic	Std. Error
CEC (cmol(+)/kg)	4	8.23	32.96	98.57	24.6425	11.65878	135.927	-1.376	1.014	1.318	2.619
pH	4	5.36	5.67	21.85	5.4625	0.14056	0.020	1.809	1.014	3.430	2.619
Soil_moisture (%)	4	8.19	9.47	35.08	8.7700	0.57457	0.330	0.419	1.014	-2.215	2.619
SOM (%)	4	0.64	1.11	3.46	0.8650	0.21048	0.044	0.190	1.014	-2.592	2.619

Descriptive Statistics for Rn-M13

	N	Minimum	Maximum	Sum	Mean	Std. Deviation	Variance	Skewness		Kurtosis	
	Statistic	Statistic	Statistic	Statistic	Statistic	Statistic	Statistic	Statistic	Std. Error	Statistic	Std. Error

CEC (cmol(+)/kg)	4	28.08	36.61	124.67	31.1675	4.02896	16.233	1.066	1.014	-0.200	2.619
pH	4	5.17	5.64	21.62	5.4050	0.19399	0.038	0.000	1.014	0.863	2.619
Soil_moisture (%)	4	14.20	17.10	59.80	14.9500	1.43411	2.057	1.994	1.014	3.978	2.619
SOM (%)	4	1.76	5.27	12.29	3.0725	1.62803	2.650	1.057	1.014	-0.188	2.619

Descriptive Statistics for all four samples

	N	Minimum	Maximum	Sum	Mean	Std. Deviation	Variance	Skewness		Kurtosis	
	Statistic	Statistic	Statistic	Statistic	Statistic	Statistic	Statistic	Statistic	Std. Error	Statistic	Std. Error
CEC (cmol(+)/kg)	12	8.23	52.89	338.11	28.1758	11.29603	127.600	0.373	0.637	1.545	1.232
pH	12	5.17	6.42	67.25	5.6042	0.38592	0.149	1.196	0.637	0.584	1.232
Soil_moisture (%)	12	3.16	17.10	127.32	10.6100	3.90144	15.221	-0.100	0.637	-0.253	1.232
SOM (%)	12	0.64	5.27	22.44	1.8700	1.32120	1.746	1.791	0.637	3.398	1.232

A14. One-Sample t-test analysis for each soil characteristic in Rn-M11, Rn-M12, and Rn-M13 in different depths (function of topography)

0 - 25 cm depth (Rn-M11, Rn-M12, Rn-M13, and Radioactive sample)

	One-Sample Statistics				One-Sample Test							One-Sample Effect Sizes			
							Significance			95% Confidence Interval of the Difference		Standardizer[a]	Point Estimate	95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean	t	df	One-Sided p	Two-Sided p	Mean Difference	Lower	Upper			Lower	Upper
CEC (cmol(+)/kg)	4	35.3150	11.98119	5.99060	5.895	3	0.005	0.010	35.31500	16.2502	54.3798	11.98119	2.948	0.522	5.375
pH	4	5.7500	0.48229	0.24114	23.845	3	0.000	0.000	5.75000	4.9826	6.5174	0.48229	11.922	3.128	21.090
Soil_moisture (%)	4	9.7425	5.70932	2.85466	3.413	3	0.021	0.042	9.74250	0.6577	18.8273	5.70932	1.706	0.050	3.295
SOM (%)	4	2.2350	2.05448	1.02724	2.176	3	0.059	**0.118**	2.23500	-1.0341	5.5041	2.05448	1.088	-0.239	2.326

0 - 25 cm depth (Rn-M11, Rn-M12, and Rn-M13) excluding Radioactive sample

	One-Sample Statistics				One-Sample Test							One-Sample Effect Sizes			
							Significance			95% Confidence Interval of the Difference				95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean	t	df	One-Sided p	Two-Sided p	Mean Difference	Lower	Upper	Standardizer[a]	Point Estimate	Lower	Upper
CEC (cmol(+)/kg)	3	38.0000	13.11747	7.57338	5.018	2	0.019	0.038	38.00000	5.4144	70.5856	13.11747	2.897	0.110	5.751
pH	3	5.9100	0.44193	0.25515	23.163	2	0.001	0.002	5.91000	4.8122	7.0078	0.44193	13.373	2.052	25.727
Soil_moisture (%)	3	11.9367	4.47306	2.58252	4.622	2	0.022	0.044	11.93667	0.8250	23.0484	4.47306	2.669	0.050	5.327
SOM (%)	3	2.5433	2.40019	1.38575	1.835	2	0.104	**0.208**	2.54333	-3.4191	8.5057	2.40019	1.060	-0.483	2.487

25 - 50 cm depth (Rn-M11, Rn-M12, and Rn-M13)

	One-Sample Test	One-Sample Effect Sizes

	One-Sample Statistics														
							Significance			95% Confidence Interval of the Difference				95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean	t	df	One-Sided p	Two-Sided p	Mean Difference	Lower	Upper	Standardizer^a	Point Estimate	Lower	Upper
CEC (cmol(+)/kg)	3	20.0767	11.80028	6.81289	2.947	2	0.049	**0.098**	20.07667	-9.2368	49.3902	11.80028	1.701	-0.238	3.573
pH	3	5.5967	0.55194	0.31866	17.563	2	0.002	0.003	5.59667	4.2256	6.9678	0.55194	10.140	1.512	19.530
Soil_moisture (%)	3	10.2433	3.59764	2.07710	4.932	2	0.019	0.039	10.24333	1.3063	19.1804	3.59764	2.847	0.097	5.658
SOM (%)	3	1.7300	1.42313	0.82164	2.106	2	0.085	**0.170**	1.73000	-1.8053	5.2653	1.42313	1.216	-0.417	2.741

50 - 75 cm depth (Rn-M11, Rn-M12, and Rn-M13)

	One-Sample Statistics				One-Sample Test							One-Sample Effect Sizes			
							Significance			95% Confidence Interval of the Difference				95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean	t	df	One-Sided p	Two-Sided p	Mean Difference	Lower	Upper	Standardizer^a	Point Estimate	Lower	Upper
CEC (cmol(+)/kg)	3	22.3500	6.99849	4.04058	5.531	2	0.016	0.031	22.35000	4.9648	39.7352	6.99849	3.194	0.184	6.304
pH	3	5.5333	0.29160	0.16836	32.866	2	0.000	0.001	5.53333	4.8089	6.2577	0.29160	18.975	2.966	36.474
Soil_moisture (%)	3	11.6633	3.11256	1.79704	6.490	2	0.011	0.023	11.66333	3.9313	19.3954	3.11256	3.747	0.314	7.343
SOM (%)	3	1.8133	0.80532	0.46495	3.900	2	0.030	**0.060**	1.81333	-0.1872	3.8138	0.80532	2.252	-0.066	4.562

A15. T-test results for the determination of differences in soil characteristics values for different samples in depths (function of depths)

Rn-M11: differences in terms of depth

One-Sample Statistics					One-Sample Test							One-Sample Effect Sizes			
							Significance			95% Confidence Interval of the Difference				95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean	t	df	One-Sided p	Two-Sided p	Mean Difference	Lower	Upper	Standardizer^{a}	Point Estimate	Lower	Upper
CEC (cmol(+)/kg)	3	29.2033	20.70482	11.95394	2.443	2	0.067	**0.135**	29.20333	-22.2303	80.6370	20.70482	1.410	-0.342	3.068
pH	3	6.1700	0.27839	0.16073	38.388	2	0.000	0.001	6.17000	5.4784	6.8616	0.27839	22.163	3.481	42.593
Soil_moisture (%)	3	9.7600	2.61901	1.51208	6.455	2	0.012	0.023	9.76000	3.2540	16.2660	2.61901	3.727	0.309	7.304
SOM (%)	3	1.7933	0.69342	0.40035	4.479	2	0.023	**0.046**	1.79333	0.0708	3.5159	0.69342	2.586	0.028	5.175

Rn-M12: differences in terms of depth

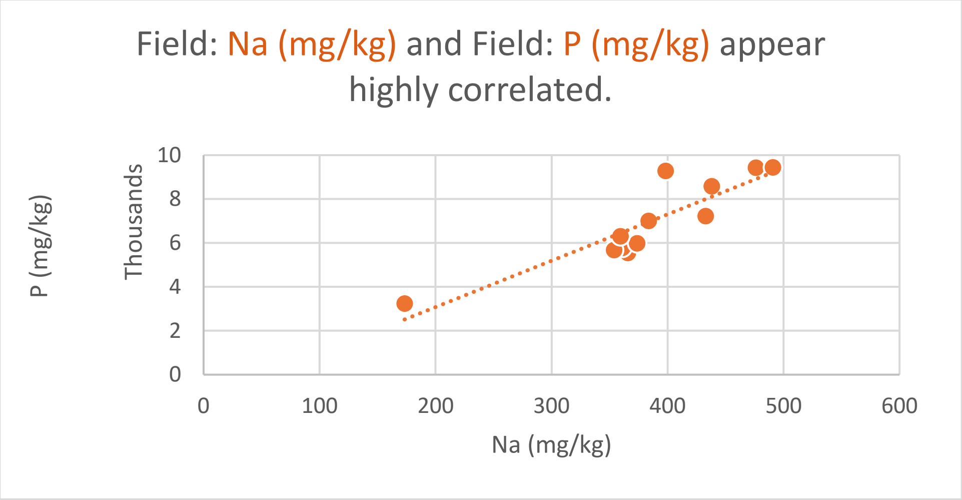
One-Sample Statistics					One-Sample Test							One-Sample Effect Sizes			
							Significance			95% Confidence Interval of the Difference				95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean	t	df	One-Sided p	Two-Sided p	Mean Difference	Lower	Upper	Standardizer^{a}	Point Estimate	Lower	Upper
CEC (cmol(+)/kg)	4	24.6425	11.65878	5.82939	4.227	3	0.012	0.024	24.64250	6.0908	43.1942	11.65878	2.114	0.216	3.965
pH	4	5.4625	0.14056	0.07028	77.722	3	0.000	0.000	5.46250	5.2388	5.6862	0.14056	38.861	10.401	68.613
Soil_moisture (%)	4	8.7700	0.57457	0.28729	30.527	3	0.000	0.000	8.77000	7.8557	9.6843	0.57457	15.264	4.039	26.978
SOM (%)	4	0.8650	0.21048	0.10524	8.219	3	0.002	0.004	0.86500	0.5301	1.1999	0.21048	4.110	0.903	7.380

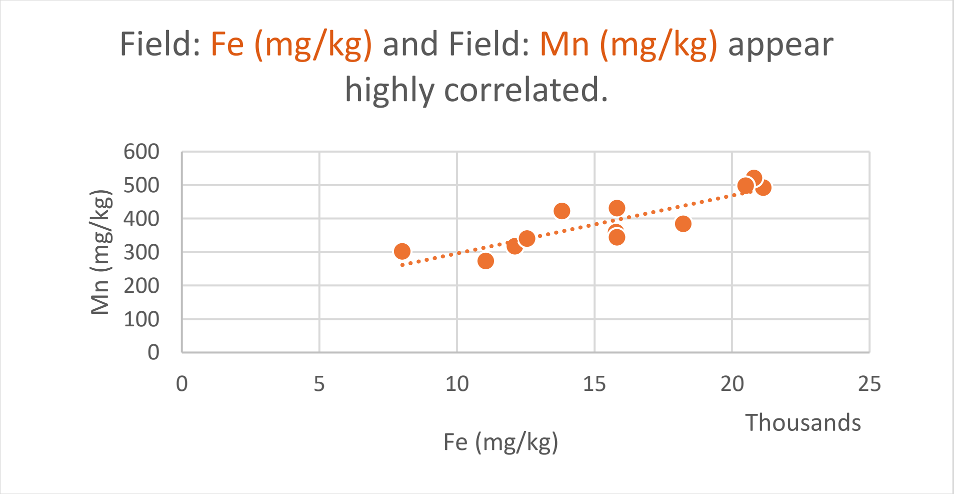
Rn-M13: differences in terms of depth

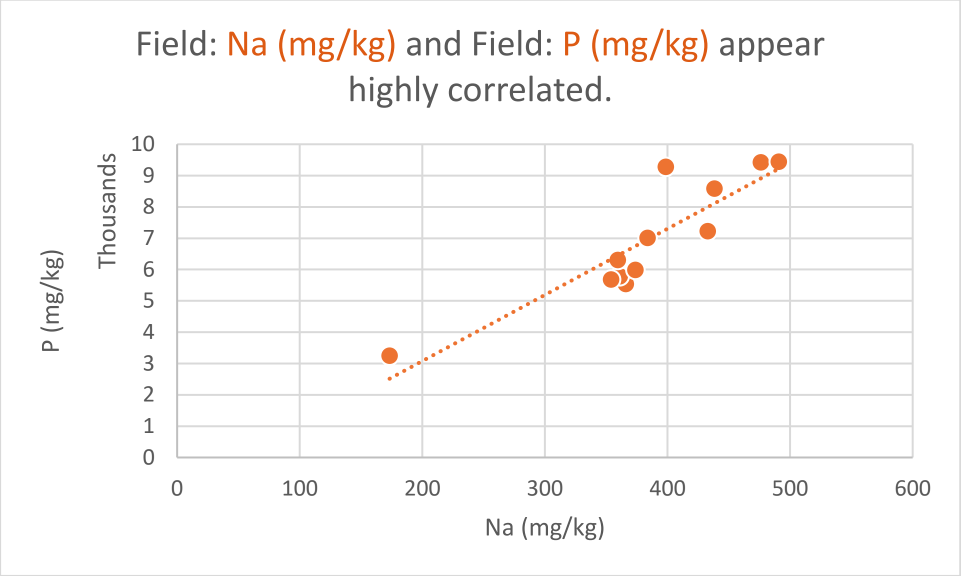
	One-Sample Test	One-Sample Effect Sizes

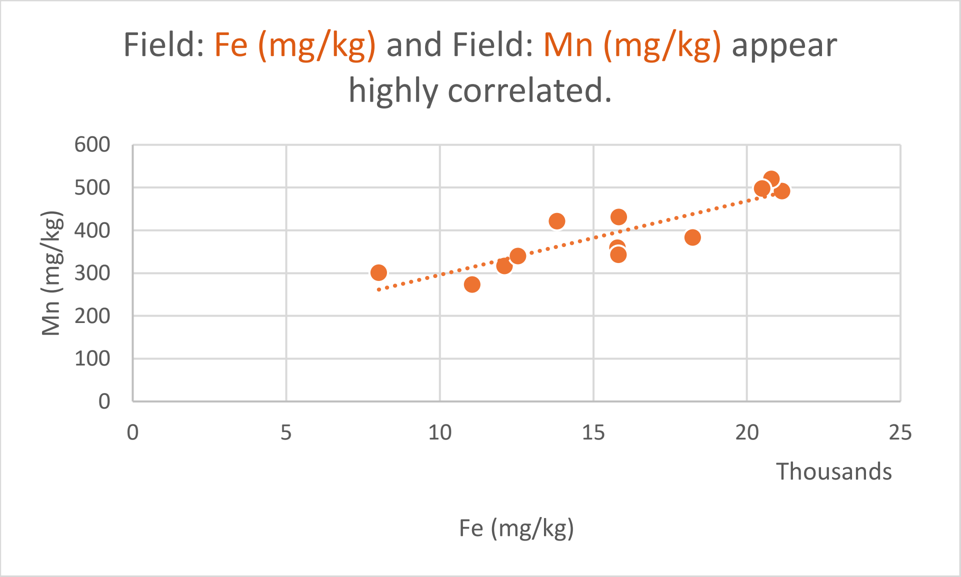
One-Sample Statistics							Significance			95% Confidence Interval of the Difference				95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean	t	df	One-Sided p	Two-Sided p	Mean Difference	Lower	Upper	Standardizer^a	Point Estimate	Lower	Upper
CEC (cmol(+)/kg)	4	31.1675	4.02896	2.01448	15.472	3	0.000	0.001	31.16750	24.7565	37.5785	4.02896	7.736	1.967	13.723
pH	4	5.4050	0.19399	0.09700	55.724	3	0.000	0.000	5.40500	5.0963	5.7137	0.19399	27.862	7.443	49.202
Soil_moisture (%)	4	14.9500	1.43411	0.71705	20.849	3	0.000	0.000	14.95000	12.6680	17.2320	1.43411	10.425	2.716	18.452
SOM (%)	4	3.0725	1.62803	0.81402	3.774	3	0.016	0.033	3.07250	0.4819	5.6631	1.62803	1.887	0.126	3.591

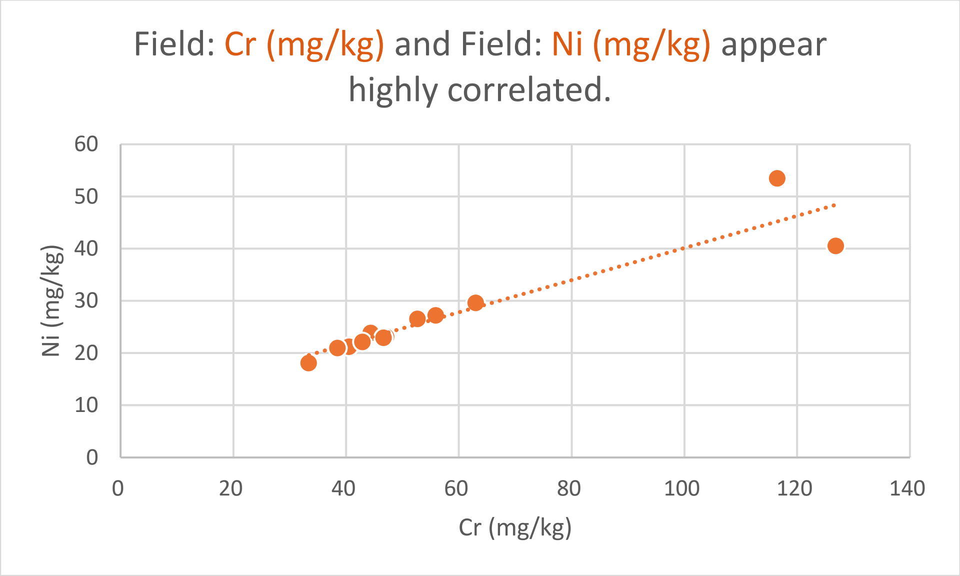
A16. Correlation graphs for the soil characteristics, radionuclides, and PTEs in soil

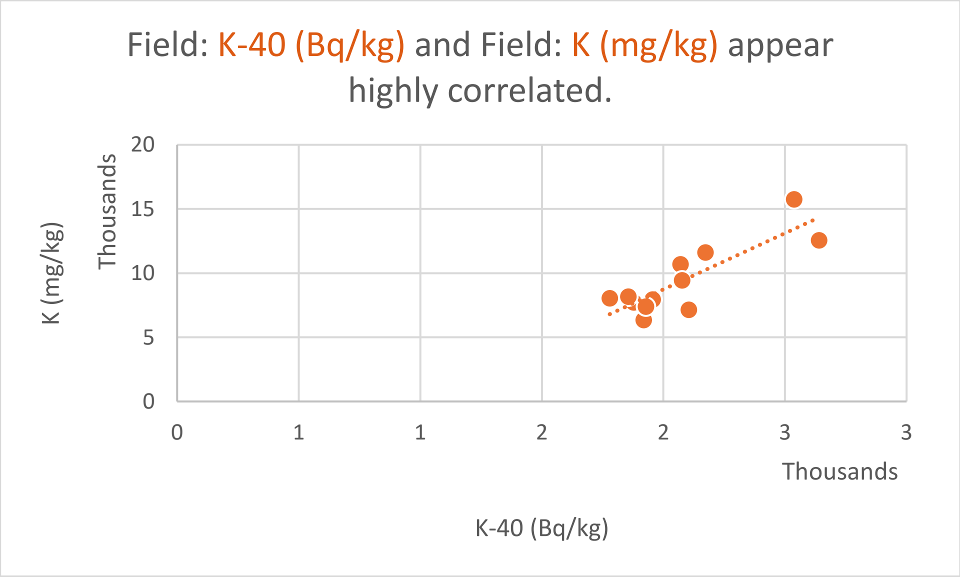


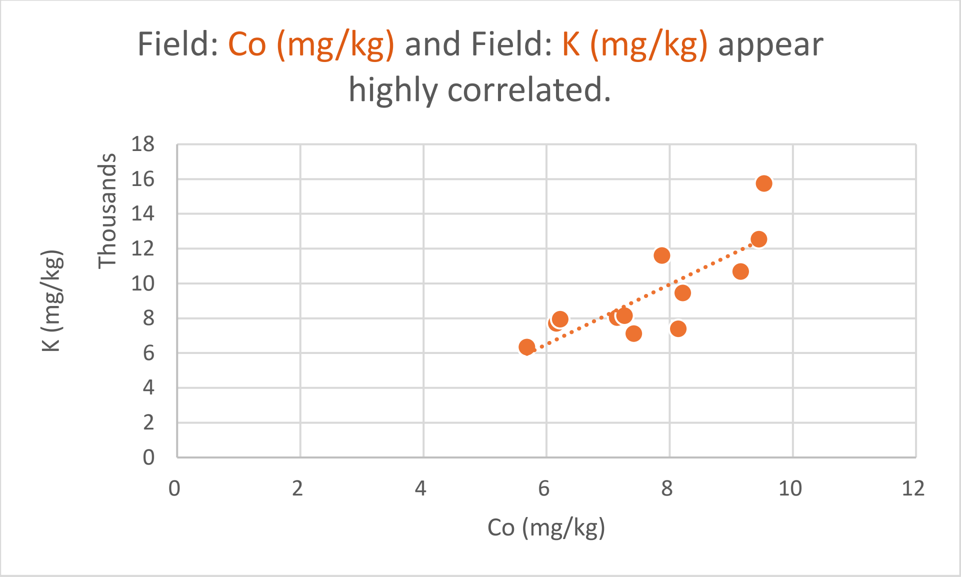


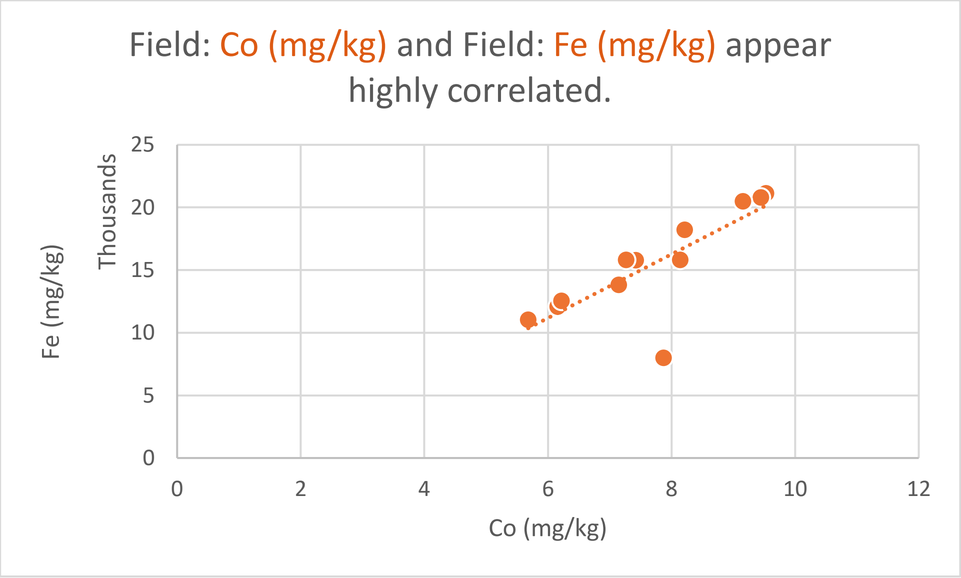

Field: Na (mg/kg) and Field: P (mg/kg) appear highly correlated.
P (mg/kg)
Thousands
10
9
8
7
6
5
4
3
2
1
0
0
100
200
300
400
500
600
Na (mg/kg)

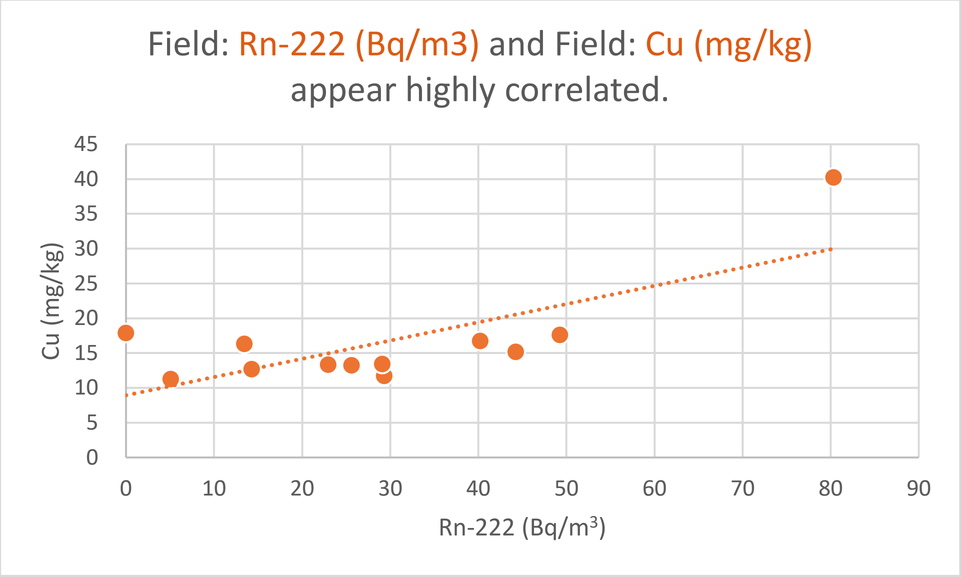

Field: Fe (mg/kg) and Field: Mn (mg/kg) appear highly correlated.
Mn (mg/kg)
600
500
400
300
200
100
0
0
5
10
15
20
25
Thousands
Fe (mg/kg)

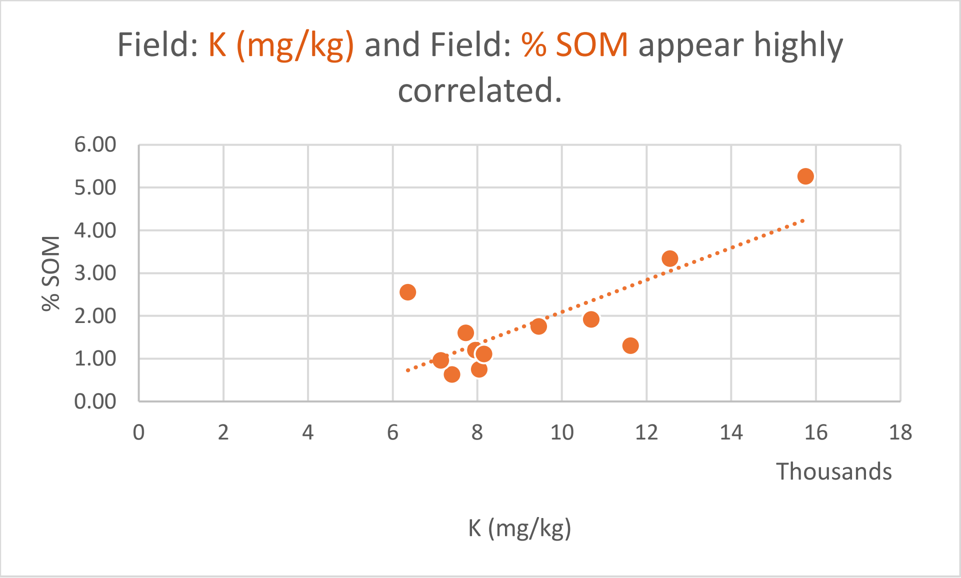

Field: Cr (mg/kg) and Field: Ni (mg/kg) appear highly correlated.

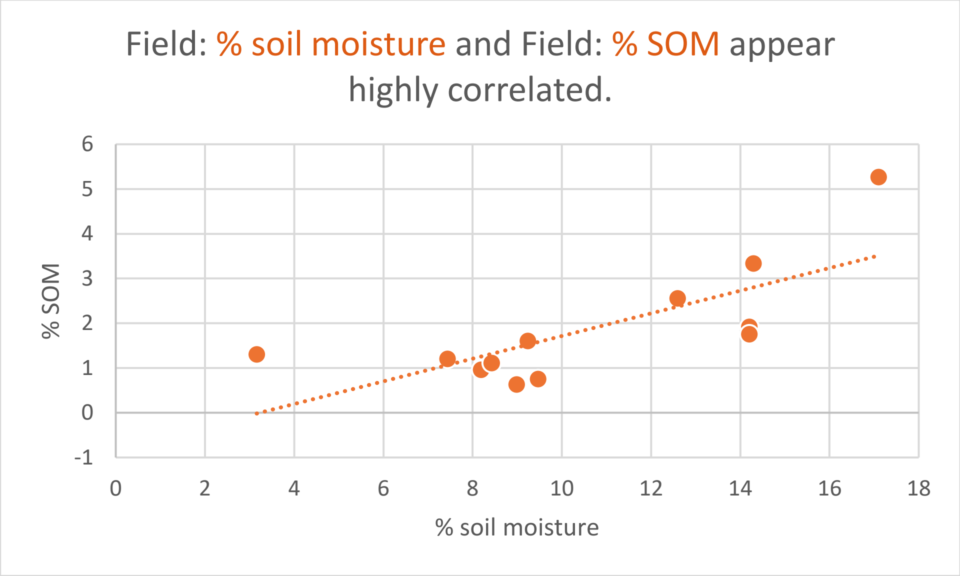

Field: K-40 (Bq/kg) and Field: K (mg/kg) appear highly correlated.

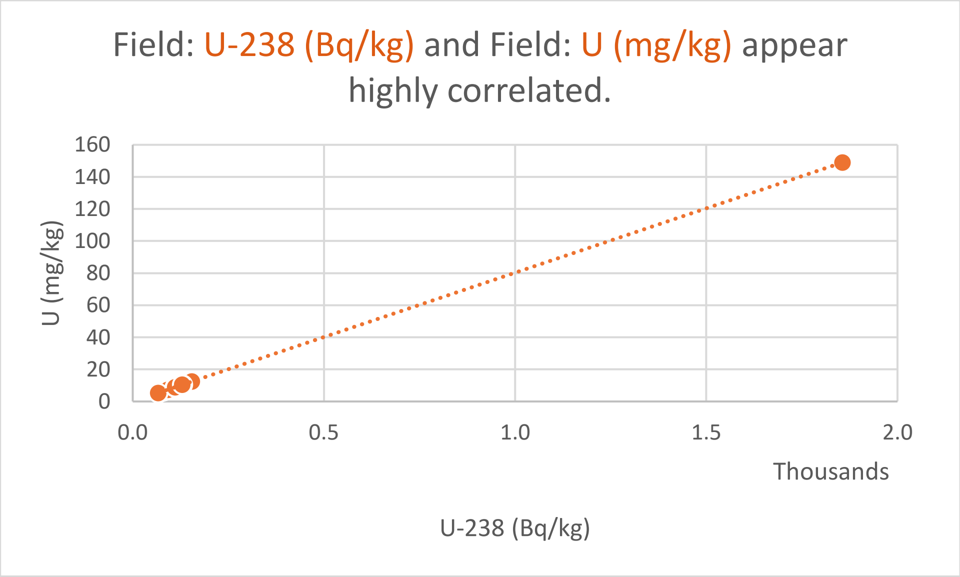

Field: Co (mg/kg) and Field: K (mg/kg) appear highly correlated.


Field: Co (mg/kg) and Field: Fe (mg/kg) appear highly correlated.


Field: Rn-222 (Bq/m3) and Field: Cu (mg/kg) appear highly correlated.


Field: K (mg/kg) and Field: % SOM appear highly correlated.


Field: % soil moisture and Field: % SOM appear highly correlated.


Field: U-238 (Bq/kg) and Field: U (mg/kg) appear highly correlated.

A17. Pseudo total Principal Components Analysis for radionuclides and PTEs in plants

Component	**Variance**	**Proportion**	**Cumulative proportion**
1	7.813	0.488	0.488
2	4.851	0.303	0.791
3	2.791	0.174	0.966
4	0.546	0.034	1.000
5	0.000	0.000	1.000

Coefficients	**Component**				
	1	**2**	**3**	**4**	**5**
U-238	-0.301	-0.241	0.027	0.113	0.698
Th-232	-0.039	-0.441	0.020	0.286	0.045
U-235	-0.301	-0.241	0.027	0.113	-0.098
K-40	-0.212	0.027	0.479	-0.080	-0.084
Ca	0.003	0.327	0.411	0.123	-0.130
Cr	-0.225	0.344	-0.073	-0.155	-0.040
Cu	0.245	-0.093	-0.317	0.620	-0.232
Fe	-0.185	0.040	-0.492	-0.305	-0.003
K	0.344	0.075	0.123	0.117	-0.071
Mg	-0.315	0.074	0.226	0.314	0.059
Mn	-0.308	0.052	-0.295	0.043	-0.182
Na	-0.181	0.358	-0.062	0.450	-0.059
Ni	-0.308	0.225	0.064	0.044	-0.080
P	0.262	0.305	-0.025	0.153	0.581
U	-0.301	-0.241	0.027	0.113	-0.116
Zn	-0.165	0.331	-0.298	0.122	0.138

A18. Pseudo total t-test results for the determination of differences in concentration means for Rn-M11 sample in different depths

One-Sample Statistics					One-Sample Test							One-Sample Effect Sizes			
							Significance			95% Confidence Interval of the Difference				95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean	t	df	One-Sided p	Two-Sided p	Mean Difference	Lower	Upper	Standardizer[a]	Point Estimate	Lower	Upper
U_238	3	103.567	44.745	25.833	4.009	2	0.028	**0.057**	103.567	-7.586	214.719	44.745	2.315	-0.048	4.676
Th_232	3	42.533	3.798	2.193	19.398	2	0.001	0.003	42.533	33.099	51.968	3.798	11.199	1.691	21.560
U_235	3	4.827	2.082	1.202	4.015	2	0.028	**0.057**	4.827	-0.346	9.999	2.082	2.318	-0.047	4.682
K_40	3	1917.667	39.017	22.527	85.129	2	0.000	0.000	1917.667	1820.743	2014.590	39.017	49.149	7.800	94.410
Rn_222	3	10.227	13.554	7.825	1.307	2	0.161	**0.321**	10.227	-23.443	43.896	13.554	0.755	-0.626	2.020
Ca	3	25626.000	11312.050	6531.015	3.924	2	0.030	**0.059**	25626.000	-2474.690	53726.690	11312.050	2.265	-0.062	4.587
Cd	3	0.000	.00000[a]	0.000											
Co	3	6.020	0.296	0.171	35.229	2	0.000	0.001	6.020	5.285	6.755	0.296	20.340	3.187	39.093
Cr	3	40.333	6.852	3.956	10.196	2	0.005	0.009	40.333	23.313	57.353	6.852	5.887	0.755	11.400
Cu	3	14.167	3.384	1.954	7.250	2	0.009	0.018	14.167	5.760	22.574	3.384	4.186	0.411	8.171
Fe	3	11900.667	769.421	444.226	26.790	2	0.001	0.001	11900.667	9989.318	13812.015	769.421	15.467	2.396	29.742
K	3	7345.333	860.773	496.968	14.780	2	0.002	0.005	7345.333	5207.054	9483.612	860.773	8.533	1.236	16.454
Mg	3	6422.667	1428.146	824.541	7.789	2	0.008	0.016	6422.667	2874.954	9970.379	1428.146	4.497	0.478	8.759
Mn	3	310.000	34.044	19.655	15.772	2	0.002	0.004	310.000	225.430	394.570	34.044	9.106	1.335	17.550
Na	3	360.333	6.028	3.480	103.541	2	0.000	0.000	360.333	345.360	375.307	6.028	59.779	9.495	114.824
Ni	3	20.837	2.547	1.470	14.171	2	0.002	0.005	20.837	14.510	27.163	2.547	8.182	1.174	15.782
P	3	5672.333	121.665	70.243	80.753	2	0.000	0.000	5672.333	5370.101	5974.566	121.665	46.623	7.397	89.557
Pb	3	8.120	0.699	0.404	20.110	2	0.001	0.002	8.120	6.383	9.857	0.699	11.611	1.759	22.348
U	3	2.023	0.721	0.416	4.860	2	0.020	0.040	2.023	0.232	3.815	0.721	2.806	0.086	5.581
Zn	3	43.267	15.603	9.008	4.803	2	0.020	0.041	43.267	4.507	82.026	15.603	2.773	0.078	5.521

The values highlighted in **bold** were significantly different.

A19. Pseudo total t-test results for the determination of differences in concentration means for Rn-M12 sample in different depths

One-Sample Statistics					One-Sample Test							One-Sample Effect Sizes			
							Significance		Mean Difference	95% Confidence Interval of the Difference		Standardizer[a]	Point Estimate	95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean	t	df	One-Sided p	Two-Sided p		Lower	Upper			Lower	Upper
U_238	4	85.35	22.26	11.13	7.67	3	0.002	0.005	85.35	49.94	120.76	22.26	3.84	0.82	6.90
Th_232	4	42.85	9.52	4.76	9.00	3	0.001	0.003	42.85	27.70	58.00	9.52	4.50	1.02	8.06
U_235	4	3.98	1.04	0.52	7.69	3	0.002	0.005	3.98	2.33	5.63	1.04	3.85	0.82	6.92
K_40	4	1916.75	138.87	69.44	27.61	3	<.001	<.001	1916.75	1695.78	2137.72	138.87	13.80	3.64	24.40
Rn_222	4	23.93	7.05	3.53	6.79	3	0.003	0.007	23.93	12.70	35.15	7.05	3.39	0.67	6.14
Ca	4	24697.00	13909.13	6954.57	3.55	3	0.019	0.038	24697.00	2564.46	46829.54	13909.13	1.78	0.08	3.41
Cd	4	0.00	.00000[a]	0.00											
Co	4	7.50	0.44	0.22	33.76	3	<.001	<.001	7.50	6.79	8.20	0.44	16.88	4.48	29.83
Cr	4	60.45	37.12	18.56	3.26	3	0.024	0.047	60.45	1.39	119.51	37.12	1.63	0.02	3.17
Cu	4	12.80	0.81	0.40	31.67	3	<.001	<.001	12.80	11.51	14.09	0.81	15.84	4.20	27.99
Fe	4	15311.00	996.16	498.08	30.74	3	<.001	<.001	15311.00	13725.89	16896.11	996.16	15.37	4.07	27.17
K	4	7685.25	495.25	247.62	31.04	3	<.001	<.001	7685.25	6897.20	8473.30	495.25	15.52	4.11	27.43
Mg	4	7517.00	2762.06	1381.03	5.44	3	0.006	0.012	7517.00	3121.94	11912.06	2762.06	2.72	0.44	4.99
Mn	4	388.75	44.23	22.11	17.58	3	<.001	<.001	388.75	318.37	459.13	44.23	8.79	2.26	15.58
Na	4	387.50	32.03	16.01	24.20	3	<.001	<.001	387.50	336.54	438.46	32.03	12.10	3.18	21.40
Ni	4	30.14	15.60	7.80	3.86	3	0.015	0.031	30.14	5.31	54.96	15.60	1.93	0.14	3.66
P	4	6638.75	582.90	291.45	22.78	3	<.001	<.001	6638.75	5711.22	7566.28	582.90	11.39	2.98	20.15
Pb	4	7.86	0.38	0.19	41.46	3	<.001	<.001	7.86	7.26	8.46	0.38	20.73	5.52	36.61
U	4	6.85	1.78	0.89	7.71	3	0.002	0.005	6.85	4.02	9.68	1.78	3.85	0.82	6.93
Zn	4	37.30	3.57	1.79	20.88	3	<.001	<.001	37.30	31.61	42.99	3.57	10.44	2.72	18.48

A20. Pseudo total t-test results for the determination of differences in concentration means for Rn-M13 sample in different depths

One-Sample Statistics					**One-Sample Test**							**One-Sample Effect Sizes**			
							Significance		Mean Difference	95% Confidence Interval of the Difference		Standardizer^a	Point Estimate	95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean	t	df	One-Sided p	Two-Sided p		Lower	Upper			Lower	Upper
U_238	4	120.50	11.00	5.50	21.909	3	<.001	<.001	120.50	103.00	138.00	11.00	10.96	2.86	19.39
Th_232	4	58.85	15.90	7.95	7.401	3	0.003	0.005	58.85	33.54	84.16	15.90	3.70	0.77	6.67
U_235	4	5.62	0.52	0.26	21.586	3	<.001	<.001	5.62	4.79	6.45	0.52	10.79	2.82	19.10
K_40	4	2331.50	300.25	150.12	15.531	3	<.001	<.001	2331.50	1853.74	2809.26	300.25	7.77	1.98	13.77
Rn_222	4	36.83	15.99	7.99	4.606	3	0.01	0.019	36.83	11.38	62.27	15.99	2.30	0.29	4.28
Ca	4	13472.00	4985.82	2492.91	5.404	3	0.006	0.012	13472.00	5538.44	21405.56	4985.82	2.70	0.44	4.96
Cd	4	0.00	.00000^a	0.00											
Co	4	9.09	0.61	0.30	29.992	3	<.001	<.001	9.09	8.12	10.05	0.61	15.00	3.97	26.51
Cr	4	54.53	6.84	3.42	15.95	3	<.001	<.001	54.53	43.65	65.40	6.84	7.98	2.04	14.14
Cu	4	16.53	1.04	0.52	31.864	3	<.001	<.001	16.53	14.87	18.18	1.04	15.93	4.22	28.16
Fe	4	20166.00	1317.43	658.72	30.614	3	<.001	<.001	20166.00	18069.67	22262.33	1317.43	15.31	4.05	27.06
K	4	12115.50	2744.66	1372.33	8.828	3	0.002	0.003	12115.50	7748.14	16482.86	2744.66	4.41	1.00	7.91
Mg	4	8294.75	249.56	124.78	66.475	3	<.001	<.001	8294.75	7897.65	8691.85	249.56	33.24	8.89	58.69
Mn	4	473.75	61.12	30.56	15.501	3	<.001	<.001	473.75	376.49	571.01	61.12	7.75	1.97	13.75
Na	4	451.00	41.22	20.61	21.881	3	<.001	<.001	451.00	385.41	516.59	41.22	10.94	2.86	19.36
Ni	4	26.61	2.77	1.39	19.186	3	<.001	<.001	26.61	22.20	31.03	2.77	9.59	2.49	16.99
P	4	9187.25	407.00	203.50	45.146	3	<.001	<.001	9187.25	8539.62	9834.88	407.00	22.57	6.02	39.87
Pb	4	13.15	1.12	0.56	23.399	3	<.001	<.001	13.15	11.36	14.94	1.12	11.70	3.07	20.70
U	4	9.65	0.87	0.43	22.282	3	<.001	<.001	9.65	8.27	11.03	0.87	11.14	2.91	19.71
Zn	4	57.08	8.33	4.17	13.701	3	<.001	<.001	57.08	43.82	70.33	8.33	6.85	1.72	12.17

A21. Pearson correlation matrix for PTEs obtained using the BCR procedure and the soil characteristics

	Cd	**Co**	**Cr**	**Cu**	**Fe**	**Mn**	**Ni**	**Pb**	**U**	**Zn**	**pH**	**CEC**	**% SOM**
Cd	1												
Co	**-0.674**	1.000											
Cr	0.498	0.024	1.000										
Cu	**0.997**	**-0.723**	0.451	1.000									
Fe	**-0.914**	**0.916**	-0.242	**-0.940**	1.000								
Mn	**-0.982**	0.562	**-0.651**	**-0.968**	**0.840**	1.000							
Ni	-0.299	**0.629**	**0.677**	-0.351	0.524	0.114	1.000						
Pb	**0.996**	**-0.663**	0.563	**0.992**	**-0.904**	**-0.991**	-0.227	1.000					
U	**0.991**	**-0.732**	0.524	**0.993**	**-0.939**	**-0.975**	-0.270	**0.995**	1.000				
Zn	**-0.701**	**0.693**	**-0.669**	**-0.709**	**0.748**	**0.736**	-0.109	**-0.750**	**-0.786**	1.000			
pH	**-0.654**	**0.965**	0.208	**-0.706**	**0.891**	0.512	**0.806**	**-0.621**	**-0.683**	0.500	1.000		
CEC	-0.257	**0.861**	0.511	-0.325	**0.618**	0.093	**0.812**	-0.224	-0.309	0.290	**0.897**	1.000	
SOM	-0.486	0.435	**-0.789**	-0.480	0.485	0.573	-0.424	-0.556	**-0.581**	**0.942**	0.195	0.038	1.000

The **bold** *r* values are significant at $p \leq 0.05$.

A22. BCR t-test results to determine if there are differences between the concentrations of each element in different samples in Fraction 1

Fraction 1					**One-Sample Test**							**One-Sample Effect Sizes**			
One-Sample Statistics							Significance		Mean Difference	95% Confidence Interval of the Difference		Standardizer[a]	Point Estimate	95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean	t	df	One-Sided p	Two-Sided p		Lower	Upper			Lower	Upper
Cd	4	0.038	0.022	0.011	3.382	3	0.022	0.043	0.038	0.002	0.073	0.022	1.691	0.043	3.271
Co	4	1.288	2.022	1.011	1.274	3	0.146	**0.292**	1.288	-1.929	4.504	2.022	0.637	-0.496	1.692
Cr	4	0.135	0.111	0.055	2.435	3	0.046	**0.093**	0.135	-0.041	0.311	0.111	1.217	-0.174	2.522
Cu	4	1.290	2.527	1.263	1.021	3	0.191	**0.382**	1.290	-2.731	5.311	2.527	0.510	-0.578	1.531
Fe	4	207.973	410.697	205.348	1.013	3	0.193	**0.386**	207.973	-445.538	861.483	410.697	0.506	-0.581	1.526
Mn	4	104.650	92.917	46.459	2.253	3	0.055	**0.110**	104.650	-43.202	252.502	92.917	1.126	-0.220	2.384
Ni	4	1.058	0.904	0.452	2.339	3	0.051	**0.101**	1.058	-0.381	2.496	0.904	1.170	-0.198	2.449
Pb	4	1.180	2.107	1.054	1.120	3	0.172	**0.344**	1.180	-2.173	4.533	2.107	0.560	-0.545	1.593
U	4	4.350	8.700	4.350	1.000	3	0.196	**0.391**	4.350	-9.494	18.194	8.700	0.500	-0.585	1.518
Zn	4	2.020	2.709	1.355	1.491	3	0.116	**0.233**	2.020	-2.291	6.331	2.709	0.746	-0.429	1.837

The values highlighted in **bold** were significantly different.

A23. BCR t-test results to determine if there are differences between the concentrations of each element in different samples in Fraction 2

Fraction 2					One-Sample Test							One-Sample Effect Sizes			
One-Sample Statistics							Significance			95% Confidence Interval of the Difference				95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean	t	df	One-Sided p	Two-Sided p	Mean Difference	Lower	Upper	Standardizer[a]	Point Estimate	Lower	Upper
Cd	4	0.043	0.054	0.027	1.563	3	0.108	**0.216**	0.043	-0.044	0.129	0.0544	0.781	-0.408	1.886
Co	4	2.173	0.902	0.451	4.815	3	0.009	0.017	2.173	0.737	3.608	0.9023	2.408	0.329	4.458
Cr	4	7.448	6.325	3.163	2.355	3	0.050	**0.100**	7.448	-2.617	17.512	6.3250	1.177	-0.194	2.461
Cu	4	3.445	3.644	1.822	1.891	3	0.078	**0.155**	3.445	-2.353	9.243	3.6436	0.945	-0.315	2.117
Fe	4	1298.000	1079.472	539.736	2.405	3	0.048	**0.095**	1298.000	-419.681	3015.681	1079.4724	1.202	-0.181	2.499
Mn	4	174.750	86.642	43.321	4.034	3	0.014	0.027	174.750	36.883	312.617	86.6425	2.017	0.178	3.805
Ni	4	4.415	1.298	0.649	6.802	3	0.003	0.006	4.415	2.349	6.481	1.2981	3.401	0.675	6.154
Pb	4	10.675	11.838	5.919	1.804	3	0.085	**0.169**	10.675	-8.161	29.511	11.8375	0.902	-0.339	2.055
U	4	3.618	7.235	3.618	1.000	3	0.196	**0.391**	3.618	-7.895	15.130	7.2350	0.500	-0.585	1.518
Zn	4	3.880	2.407	1.204	3.224	3	0.024	0.048	3.880	0.050	7.710	2.4070	1.612	0.009	3.143

The values highlighted in **bold** were significantly different.

A24. BCR t-test results to determine if there are differences between the concentrations of each element in different samples in Fraction 3

Fraction 3					**One-Sample Test**							**One-Sample Effect Sizes**			
One-Sample Statistics							Significance			95% Confidence Interval of the Difference		Standardizer^a	Point Estimate	95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean	t	df	One-Sided p	Two-Sided p	Mean Difference	Lower	Upper			Lower	Upper
												0.07848	0.542	-0.557	1.570
Cd	4	0.043	0.078	0.039	1.083	3	0.179	**0.358**	0.043	-0.082	0.167	0.456	1.130	-0.218	2.390
Co	4	0.515	0.456	0.228	2.260	3	0.054	**0.109**	0.515	-0.210	1.240	0.630	0.818	-0.157	1.729
Cr	4	3.365	0.988	0.494	6.813	3	0.003	0.006	3.365	1.793	4.937	0.988	3.407	0.677	6.163
Cu	4	4.295	7.269	3.634	1.182	3	0.161	**0.322**	4.295	-7.271	15.861	7.269	0.591	-0.525	1.632
Fe	4	472.250	336.627	168.313	2.806	3	0.034	0.068	472.250	-63.398	1007.898	336.627	1.403	-0.085	2.810
Mn	4	12.230	2.184	1.092	11.202	3	0.001	0.002	12.230	8.755	15.705	2.184	5.601	1.354	9.979
Ni	4	1.770	0.252	0.126	14.044	3	0.000	0.001	1.770	1.369	2.171	0.252	7.022	1.765	12.470
Pb	4	0.808	0.420	0.210	3.849	3	0.015	0.031	0.808	0.140	1.475	0.420	1.925	0.141	3.652
U	4	6.368	8.412	4.206	1.514	3	0.114	**0.227**	6.368	-7.017	19.752	8.412	0.757	-0.422	1.852
Zn	4	2.630	2.343	1.171	2.245	3	0.055	**0.110**	2.630	-1.098	6.358	2.343	1.123	-0.221	2.379

The values highlighted in **bold** were significantly different.

A25. BCR t-test results to determine if there are differences between the concentrations of each element in different samples in Fraction 4

Fraction 4					**One-Sample Test**							**One-Sample Effect Sizes**			
One-Sample Statistics					t	df	Significance		Mean Difference	95% Confidence Interval of the Difference		Standardizer[a]	Point Estimate	95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean			One-Sided p	Two-Sided p		Lower	Upper			Lower	Upper
Cd	4	0.148	0.095	0.048	3.094	3	0.027	**0.054**	0.148	-0.004	0.299	0.095	1.547	-0.020	3.038
Co	4	4.088	2.115	1.057	3.866	3	0.015	0.031	4.088	0.723	7.452	2.115	1.933	0.144	3.666
Cr	4	60.373	11.272	5.636	10.712	3	0.001	0.002	60.373	42.436	78.309	11.272	5.356	1.282	9.551
Cu	4	13.600	2.218	1.109	12.263	3	0.001	0.001	13.600	10.070	17.130	2.218	6.131	1.509	10.908
Fe	4	17158.250	9363.241	4681.621	3.665	3	0.018	0.035	17158.250	2259.243	32057.257	9363.241	1.833	0.103	3.501
Mn	4	112.575	75.536	37.768	2.981	3	0.029	**0.059**	112.575	-7.620	232.770	75.536	1.490	-0.045	2.948
Ni	4	24.175	6.262	3.131	7.722	3	0.002	0.005	24.175	14.211	34.139	6.262	3.861	0.825	6.948
Pb	4	7.153	2.994	1.497	4.778	3	0.009	0.017	7.153	2.388	11.917	2.994	2.389	0.322	4.426
U	4	0.000	.00000[a]	0.000											
Zn	4	40.740	17.843	8.922	4.566	3	0.010	0.020	40.740	12.348	69.132	17.843	2.283	0.282	4.249

The values highlighted in **bold** were significantly different.

A26. Pseudo total One-Sample t-test analysis for each element in Rn-M11, Rn-M12, Rn-M13, and Radioactive sample for 0 – 25 cm depth (function of topography)

One-Sample Statistics					One-Sample Test							One-Sample Effect Sizes			
							Significance			95% Confidence Interval of the Difference				95% Confidence Interval	
	N	Mean	Std. Deviation	Std. Error Mean	t	df	One-Sided p	Two-Sided p	Mean Difference	Lower	Upper	Standardizer[a]	Point Estimate	Lower	Upper
U_238	4	540.775	876.935	438.467	1.233	3	0.153	**0.305**	540.775	-854.624	1936.174	876.935	0.617	-0.508	1.665
Th_232	4	38.650	20.745	10.372	3.726	3	0.017	0.034	38.650	5.641	71.659	20.745	1.863	0.116	3.551
U_235	4	25.225	40.922	20.461	1.233	3	0.153	**0.305**	25.225	-39.891	90.341	40.922	0.616	-0.508	1.665
K_40	4	2092.000	341.195	170.597	12.263	3	0.001	0.001	2092.000	1549.083	2634.917	341.195	6.131	1.509	10.908
Rn_222	4	32.0400	33.72369	16.86184	1.900	3	0.077	**0.154**	32.04000	-21.6219	85.7019	33.72369	0.950	-0.312	2.124
Ca	4	12036.750	6018.474	3009.237	4.000	3	0.014	0.028	12036.750	2460.014	21613.486	6018.474	2.000	0.171	3.777
Cd	4	0.000	.00000[a]	0.000											
Co	4	7.678	1.420	0.710	10.813	3	0.001	0.002	7.678	5.418	9.937	1.420	5.406	1.297	9.639
Cr	4	70.375	38.632	19.316	3.643	3	0.018	0.036	70.375	8.903	131.847	38.632	1.822	0.099	3.483
Cu	4	19.900	13.730	6.865	2.899	3	0.031	**0.063**	19.900	-1.948	41.748	13.730	1.449	-0.064	2.883
Fe	4	13768.500	5483.656	2741.828	5.022	3	0.008	0.015	13768.500	5042.779	22494.221	5483.656	2.511	0.367	4.632
K	4	10789.000	3757.387	1878.693	5.743	3	0.005	0.010	10789.000	4810.159	16767.841	3757.387	2.871	0.496	5.245
Mg	4	6310.000	1608.937	804.468	7.844	3	0.002	0.004	6310.000	3749.822	8870.178	1608.937	3.922	0.844	7.054
Mn	4	383.250	90.035	45.017	8.513	3	0.002	0.003	383.250	239.985	526.515	90.035	4.257	0.949	7.635
Na	4	328.000	104.285	52.142	6.290	3	0.004	0.008	328.000	162.060	493.940	104.285	3.145	0.590	5.714
Ni	4	29.315	8.025	4.012	7.306	3	0.003	0.005	29.315	16.546	42.084	8.025	3.653	0.758	6.588
P	4	6017.000	2486.460	1243.230	4.840	3	0.008	0.017	6017.000	2060.487	9973.513	2486.460	2.420	0.333	4.479
Pb	4	18.255	17.398	8.699	2.099	3	0.063	**0.127**	18.255	-9.429	45.939	17.398	1.049	-0.259	2.269
U	4	14.218	24.395	12.198	1.166	3	0.164	**0.328**	14.218	-24.601	53.036	24.395	0.583	-0.530	1.622
Zn	4	41.400	17.506	8.753	4.730	3	0.009	0.018	41.400	13.545	69.255	17.506	2.365	0.313	4.386

The values highlighted in **bold** were significantly different.