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Radioactivity of soil in Croatia I: naturally occurring decay chains

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The assessment of environmental radioactivity much relies on radionuclide content in soil. This stems from the significant contribution of soil to both external and internal exposure to ionising radiation via direct emission of gamma radiation and soil-to-plant radionuclide transfer, respectively. This motivated us to carry out a systematic research on the radioactivity of soil in Croatia to obtain relevant data that can be used as a basis for understanding the related effects of geomorphological, biogeographical, and climatological properties of the environment. We collected samples of the surface layer of uncultivated soil (0–10 cm) at 138 sites from all over the country and measured them for radionuclide activity concentrations by means of high-resolution gamma-ray spectrometry. This resulted in radioactivity maps containing data on activity concentrations of representative radionuclides in the environment. In this paper, which is the first in our two-part presentation, we focus on the naturally occurring ^{232}Th and ^{238}U decay chains and their correlations with the diversity of Croatian regions. For both of the chains, activity concentrations were the highest in the Dinaric region, the lowest in the Pannonian region, and intermediate in the Adriatic region. Relatively high concentrations of ^{226}Ra in the soil of the Dinaric region implied a possibility of an enhanced emanation of its progeny ^{222}Rn into the air. Activity concentrations of ^{210}Pb were additionally elevated in areas with dense vegetation, most probably due to an atmospheric deposition of airborne ^{210}Pb onto the surface of plants and their eventual decomposition on the ground.

KEY WORDS: ^{210}Pb; ^{222}Rn; ^{226}Ra; ^{232}Th; ^{238}U; gamma radiation; high-resolution gamma-ray spectrometry; representative radionuclides

Radioactivity is an inherent property of nature, and life on Earth has evolved in the presence of moderate ionising radiation. Furthermore, the use of radioactive sources in science, technology, and medicine implies their possible leakage into the environment, which has piqued research interest in environmental radioactivity. Concentrations of radionuclides in close-to-the-ground media (water, air, and soil) differ considerably, and the same applies to their impact on humans and biota. In the absence of accidental atmospheric releases of anthropogenic (1, 2) or naturally occurring (3) radionuclides, most of external exposure to ionising radiation originates from soil. Moreover, soil-to-plant radionuclide transfer leads to internal exposure. In case of an atmospheric release, concentrations of emitted radionuclides in air invariably decrease with the passing of time. This, however, does not occur in soil, which is a consequence of the eventual deposition of airborne radionuclides onto the ground. Since the atomic and molecular diffusion in solid media is slow, long-lived radionuclides remain in the topmost layer of soil for a long time after they have been deposited, which may have long-term consequences. Therefore, every assessment of the impact of ionising radiation on the environment and living organisms must take into account the complexity of radionuclide content in soil.

Even though soil radioactivity research has a long history, the interest in this matter has not diminished. In recent years, for instance, activity concentrations of radionuclides in soil have been studied for agricultural soil (4, 5), in mining areas (6), close to industrial facilities (7, 8) or deposits of radioactive matter (9, 10), and after the Fukushima accident (11). Other studies focus on soil that is not directly affected by human activities (12, 13) not only to asses the related radiological risk but also obtain baseline data as reference for the future. Our study belongs to this type of research. More precisely, we analysed radionuclide activity concentrations in uncultivated soil sampled at 138 sites, which covered the entire Republic of Croatia and resulted in an overview of soil surface layer radioactivity throughout the country.

Even though soil radioactivity in Croatia has been measured for decades (14), until our study these

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measurements lacked a systematic approach. The goal of the presented research was to collect and analyse samples by taking into account that the obtained results should: (a) cover the entire country, (b) provide enough information for establishing correlations at the level of Croatian regions, (c) provide the basis for further research at a local level, and (d) provide data for contemporary projects on environmental radioactivity at an international level. The last goal may prove useful for the recent initiative for augmenting and improving the European Atlas of Natural Radiation developed and maintained by the Joint Research Centre (15).

Although Croatia is rather small (56,594 km^2), it is quite diverse with respect to various factors that influence environmental radioactivity, such as geological properties, climate, vegetation type and density, and human activities). The country has three distinct regions. The north belongs to the Pannonian Plain and adjacent hilly regions with the same underlying geology and continental climate and vegetation. The Adriatic coast and islands are mostly karstwith Mediterranean climate and vegetation. These two regions are separated by highlands and mountains belonging to the Dinaric Alps, having a cold continental climate and hosting subalpine vegetation on limestone and dolomite. As the climatological, biogeographical, and geomorphological properties of these three regions are shared by other regions in Europe, our study may have implications, at least to some extent, for a more general identification of the effects of these properties on soil radioactivity.

Our experiment was carried out by means of high-resolution gamma-ray spectrometry, which allowed us to obtain comprehensive data on the most abundant naturally occurring (^{232}Th and ^{238}U decay chains, ^{40}K) and anthropogenic (^{137}Cs) radionuclides in the environment. Due to the abundance of data and the specificities of the three regions, we present our results in two separate papers. In this one we focus on the ^{232}Th and ^{238}U decay chains. In the second paper we shall concentrate on ^{40}K and ^{137}Cs (which have different origins but are chemically similar), as well as on absorbed dose rate for external exposure to gamma radiation due to the overall soil radioactivity. We use maps to present our results, since this makes it easier to compare measured activity concentrations with the environmental diversity of Croatia. Detailed numerical data can be found elsewhere (16) and are also available from the authors upon request. Consequences of soil radioactivity for living organisms are discussed throughout the two papers, e.g., we identify regions where elevated concentrations of ^{222}Rn might be expected, those where the atmospheric deposition of its decay product ^{210}Pb is significant, or where ^{137}Cs propagation through food chains is more likely than elsewhere.

GEOMORPHOLOGICAL, BIOGEOGRAPHICAL, AND CLIMATOLOGICAL CHARACTERISTICS OF CROATIA

Figure 1 shows the common division of Croatia in three regions with respect to geomorphological, biogeographical, and climatological characteristics which influence environmental radioactivity. Each of the regions can further be divided into two subregions.

Region I, covering the north of the country, belongs to the Pannonian Plain that extends beyond Croatian borders and covers a significant part of Central Europe. The climate in this region is continental. The western part, denoted as Ia, contains both flat (<200 m.a.s.l.) and hilly (200–500 m.a.s.l.) areas together with a few small mountains (500–1100 m.a.s.l.) (17). The vegetation is mainly grassland, broadleaf forests, orchards, and crops. Subregion Ib is completely flat (<100 m.a.s.l.) (17), containing no significant hills, and agronomic vegetation prevails. The annual precipitation decreases steadily from the east to the west of region I, ranging from ~1200 mm to ~500 mm (17). The most widespread soil types in region I are loess and pseudogley (18). Close to the major rivers (Sava, Drava, and Danube) clay and sand are found, whereas chernozem predominates in subregion Ib (18).

Region II is situated in the Dinaric Alps, which are part of a range of mountains and highlands stretching from the Julian Alps deep into the western part of the Balkan Peninsula. Geologically, the entire region is dominated by

Figure 1 Main geomorphological, biogeographical, and climatological regions of Croatia. Region I belongs to the Pannonian Plain, with subregion Ia comprising hilly areas and subregion Ib being a flat lowland. Region II belongs to the Dinaric Alps. In subregion IIa, the climate is cold continental and vegetation is subalpine, whereas in subregion IIb, karst prevails, and the climate is Mediterranean. Region III is Mediterranean in both climate and vegetation. In subregion IIIa the influence of regions I and II is stronger than in subregion IIIb. Sizeable areas of dense forests are indicated by letter F

limestone and dolomite (18), and this has a significant influence on the properties of the soil. Subregion IIa consists of valleys (500–1000 m.a.s.l.) surrounded by mountains (up to 1757 m.a.s.l.) (17). The vegetation is subalpine and dense, consisting of meadows and forests (broadleaf and coniferous, at low and high altitudes, respectively). The annual precipitation is the highest in the northwest (Gorski Kotar) and along the Velebit Mountain (which stretches parallel to the coast), amounting to 2000–3500 mm, whereas in the eastern part it is about 1500 mm (17). The climate is cold continental, with a limited influence of the Mediterranean climate.

Although subregions IIa and IIb are similar and share the same underlying geology, they differ in several characteristics. Subregion IIb also comprises high mountains (up to 1831 m.a.s.l.), but the terrain is mostly below 500 m.a.s.l. (17), while the vegetation is less abundant (shrubs and bushes, meadows in valleys, coniferous trees) and partly Mediterranean. There are elements of both the Mediterranean and continental climates, with colder spots only at high altitudes. The annual precipitation is 1000–1500 mm (17).

Region III is Mediterranean in most of its properties, especially regarding the climate and vegetation. The bedrock is again predominantly limestone and dolomite, with marl and flysch appearing locally (18). Subregion IIIa comprises the Istrian Peninsula in the northwest, a narrow coastline (most of it at the foot of the Velebit Mountain) east of it, and North Adriatic islands in between. In spite of the prevailing Mediterranean characteristics, the climate is to some extent influenced by the vicinity of subregion IIa. The annual precipitation ranges from ~700 mm in western Istria to ~1500 mm northeast of the peninsula (17). Subregion IIIb consists of the Dalmatian coast and most of the hinterland and includes Dalmatian islands. The climate is completely Mediterranean, with little influence of the continental part of the wider area. The annual precipitation is generally higher than in region I and lower than in region II, ranging from ~700 mm in the northwest to ~1500 mm in the southeast (17). The predominant soil type in region III is *terra rossa*, especially in Istria.

Regions I and II contain sizable areas of dense forests, and these are in Figure 1 indicated by letter F.

MATERIALS AND METHODS

In sampling and measurements we followed a recommended procedure (19) to ensure the compatibility of our results with those obtained elsewhere. Soil was sampled at 138 locations throughout Croatia in 2015 and 2016. We sampled the surface layer (0–10 cm) of uncultivated soil that had not been disturbed by human activities such as agriculture. Most of external exposure to gamma radiation from soil is related to this layer, as radiation from deeper layers is attenuated. Collected soil sampled randomly from ten spots within a 1 m^2 area was mixed to make a representative sample, cleaned from organic material, dried, sieved, ground, put in a 100 mL cylindrical plastic container, and sealed tightly.

The activity of several radionuclides of interest was determined from the activity of a decay product with a shorter half-life ($T_{1/2}$) under the assumption of a secular equilibrium between them. In undisturbed soil, the equilibrium between ^{238}U and ^{234}Th was established naturally, and the same applies to the equilibrium between ^{232}Th and ^{228}Ac. However, the loss of gaseous ^{222}Rn from the surface layer of soil and during sample preparation leads to a disequilibrium between ^{226}Ra and ^{214}Pb. In order to restore the equilibrium, sealed samples were left to rest for more than 30 days.

Radionuclide activity concentrations A were determined by means of high-resolution gamma-ray spectrometry. The setup was based on a high-purity germanium coaxial detector (Ortec GMX; relative efficiency of 74.3 % and energy resolution of 2.23 keV, all at ^{60}Co 1.33 MeV) calibrated using a certified calibration source obtained from the Czech Metrology Institute. We accounted for self-attenuation and coincidence summing effects by using methods described in (20) and (21), respectively.

Our focus was on representative, most widely studied naturally occurring and anthropogenic radionuclides in soil: ^{232}Th and ^{238}U (parent radionuclides of the respective decay chains), ^{226}Ra and ^{210}Pb (radiologically significant, long-lived members of the ^{238}U chain), ^{40}K, and ^{137}Cs (the most abundant long-lived anthropogenic radionuclide in the environment). Their activities were determined by analysing peaks at: 338.3, 911.2, and 969.0 keV (^{228}Ac emissions) for ^{232}Th; 63.3 keV and 92.4–92.8 keV (^{234}Th emissions) for ^{238}U; 295.2 and 351.9 keV (^{214}Pb emissions) for ^{226}Ra; 46.5 keV for ^{210}Pb; 1460.8 keV for ^{40}K; 661.7 keV for ^{137}Cs. Measurements were carried out for 24 h, which resulted in the following typical values of the detection limit (DL): 0.3 Bq/kg for ^{137}Cs, 1 Bq/kg for ^{232}Th and ^{226}Ra, 2 Bq/kg for ^{40}K, 3 Bq/kg for ^{210}Pb, and 4 Bq/kg for ^{238}U.

RESULTS AND DISCUSSION

Our focus was on the ^{232}Th and ^{238}U naturally occurring decay chains. Since ^{235}U is much less abundant than ^{238}U (99.3 % of total U), its decay chain is usually disregarded in analyses of soil radioactivity, and we followed this approach. We shall first address ^{232}Th and ^{238}U as the parent radionuclides of their respective chains. After that, we shall turn to ^{226}Ra and ^{210}Pb, which belong to the ^{238}U chain and are of a special radioecological concern due to their adverse effects on living organisms (^{226}Ra as the parent nuclide of gaseous ^{222}Rn, and ^{210}Pb as a long-lived radionuclide close to the end of the chain).

^{232}Th and ^{238}U

Figure 2 shows the distribution of the A of ^{232}Th ($T_{1/2}$=14 billion years) in Croatian soil, and Figure 3 that of ^{238}U ($T_{1/2}$=4.5 billion years). The two main naturally occurring decay chains – comprising numerous radionuclides – start with ^{232}Th and ^{238}U. Therefore, these results correspond to the radioactivity levels of the whole chains (peculiarities related to the ^{238}U chain will be discussed later).

The average values ($\bar{A}$) of A for ^{232}Th and ^{238}U were similar, being 41 Bq/kg and 45 Bq/kg, respectively. The range of A for ^{232}Th was 8–85 Bq/kg and for ^{238}U, 0–140 Bq/kg (0 means that measured A was below the corresponding DL). Globally, the reported medians (and ranges) are 30 Bq/kg (11–64 Bq/kg) for ^{232}Th and 35 Bq/kg (16–110 Bq/kg) for ^{238}U. Our results, therefore, do not depart from those obtained elsewhere (including the rest of Europe) (22).

A comparison of results in Figures 2 and 3 with the sketch of Croatian regions (Figure 1) reveals a similar trend for ^{232}Th and ^{238}U. Activity concentrations were about average or lower in most of region I and sizable parts of region III. In region II and parts of subregion IIIa (western Istria), they were mainly above average. At a few spots, there were deviations from this trend, but the above conclusion still holds well.

In order to explain these differences, at least qualitatively, we ought to address factors that generally affect the formation of soil. Of these, the most important are the geological background of bedrock and substratum, climate, living organisms close to the surface, and relief (18). In region II, the bedrock is a mixture of limestone and dolomite, the living organisms typical of subalpine ecosystems, and the relief varies from highland valleys to steep mountains. These characteristics seemingly favour the presence of U and Th in soil. In region III, the bedrock is also limestone and dolomite, but the other mentioned factors are markedly different, which reduces the presence of Th and U, despite the same geological background. In region I, limestone and dolomite are scarce, whereas the climate, relief, and exchange of matter between living organisms and soil differ substantially from those in regions II and III. These conditions seem to be less favourable for the presence of Th and U in the surface layer of soil. An elucidation of the above findings requires a focused study that would take into account the complexity of the behaviour of U and Th in soil (23).

^{226}Ra and ^{210}Pb

Figure 4 shows the distribution of the A of ^{226}Ra ($T_{1/2}$=1600 years) in Croatian soil, and Figure 5 that of ^{210}Pb ($T_{1/2}$=22.4 years). For both radionuclides, the spatial distribution of A followed the trend observed for ^{238}U, which was not surprising, since all of them belong to the same decay chain. However, there were differences in the magnitudes of A. For ^{226}Ra, A varied from 14 to 281 Bq/kg, and $\bar{A}$=57 Bq/kg. For ^{210}Pb, the range was 0–288 Bq/kg, and $\bar{A}$=63 Bq/kg. Globally, the reported median for ^{226}Ra is 35 Bq/kg (the same as for ^{238}U) and the range 17–60 Bq/kg (narrower than for ^{238}U) (22). However, there are considerable variations of these quantities in the data for Europe, and our finding, therefore, does not represent an

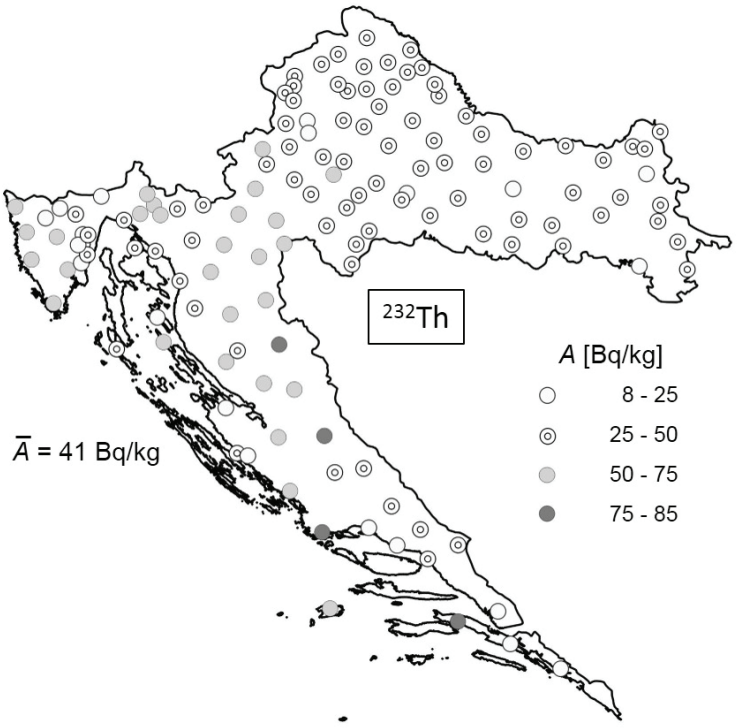

Figure 2 Distribution of the A of ^{232}Th in Croatian soil

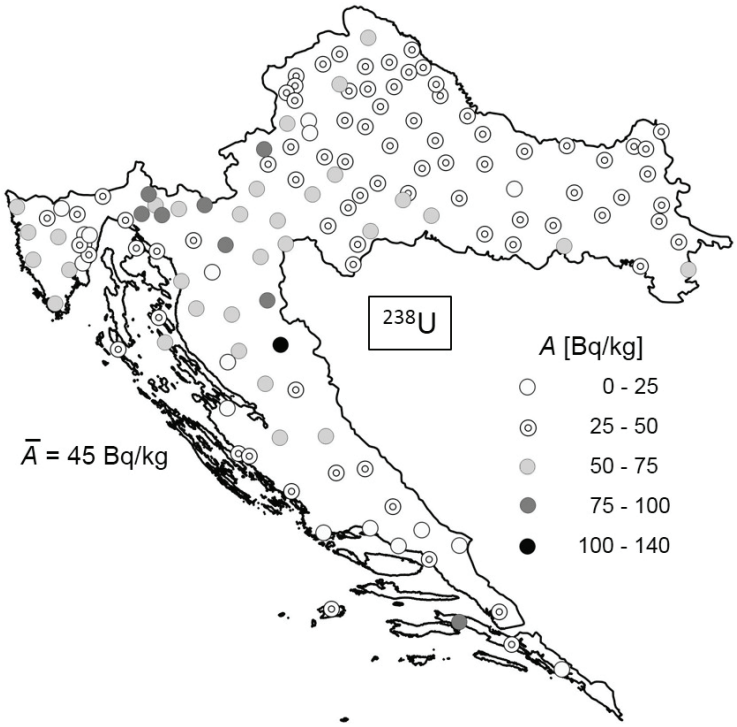

Figure 3 Distribution of the *A* of ^{238}U in Croatian soil

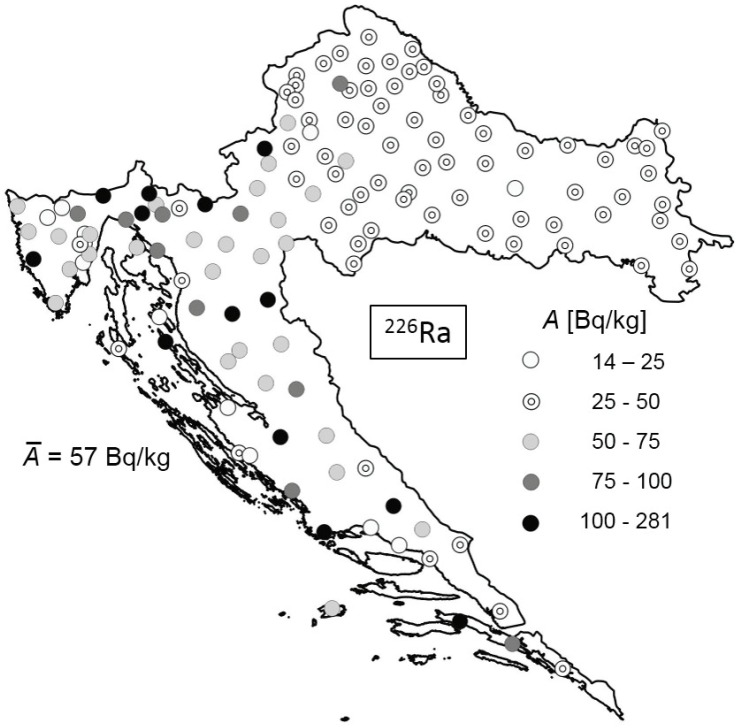

Figure 4 Distribution of the *A* of ^{226}Ra in Croatian soil

anomaly. Since ^{226}Ra and ^{210}Pb appear in the chain later than ^{238}U and have shorter $T_{1/2}$, their activities were expected to be lower or equal to that of ^{238}U (16).

In order to explain the departure from this expectation, we turn to chemical and physical properties that affect the propagation of Ra and Pb through the environment and their accumulation in soil. Let's start with ^{226}Ra, which is a product of five decays, starting with that of ^{238}U. Chemically, Ra is similar to other alkaline earth metals. The highest A of ^{226}Ra was measured at locations in region II, i.e., limestone and dolomite bedrock abundant in $CaCO_3$ and $MgCO_3$. Due to the chemical similarity of Ca, Mg, and Ra, a substitution of Ca or Mg by ^{226}Ra is more favourable than the same process for any other element of the ^{238}U chain. Since the formation of soil depends on the parent material, the concentration of Ra may be enhanced in soil that originates from limestone and dolomite. This is, however, not the only mechanism which can lead to ^{226}Ra content in surface soil being different from that expected solely from the decay of ^{238}U. Since Ca and Mg are biogenic elements, they are exchanged between the surface layer of soil and vegetation. Given the mentioned chemical properties of Ra, the participation of its isotopes in the exchange exceeds that of other radionuclides in the decay chains (24, 25). This process might at least partly account for the highest activity concentrations of ^{226}Ra in the samples from the highly forested northwestern part of subregion IIa (Gorski Kotar). Like with U and Th, a focused study based on the known properties of Ra in soil (23) is required for a full clarification of the observed phenomenon.

^{226}Ra decays into ^{222}Rn, a noble gas with $T_{1/2}$=3.8 days. Radon is the only gaseous radionuclide appearing in the naturally occurring decay chains. Of its isotopes, only ^{222}Rn is of interest for environmental radioactivity, because the other ones (^{219}Rn and ^{220}Rn) have $T_{1/2}$ under one minute. The gaseous nature of ^{222}Rn and its relatively long $T_{1/2}$ lead to its appearance in the air close to the ground. This is a consequence of its diffusion through soil and release into the air above (26, 27). Hence, the ^{238}U chain differs from the ^{232}Th and ^{235}U chains in the appearance of ^{222}Ra, which can easily migrate out from its matrix (in our case, from soil). By consequence, decay products of ^{222}Rn are subject to airborne decay and atmospheric deposition onto different surfaces (28). Of these, ^{210}Pb has the longest $T_{1/2}$ and it therefore deserves a special consideration.

The distribution of the A of ^{210}Pb followed the trend observed for ^{238}U and ^{226}Ra, but its $\bar{A}$ and maximum A were higher than those of the mentioned radionuclides from the same decay chain. In particular, by comparing Figures 1, 4, and 5, one can note that the A of ^{210}Pb tended to be higher than that of ^{226}Ra in forested areas. Radionuclides in the part of the ^{238}U chain between ^{222}Rn and ^{210}Pb have short $T_{1/2}$ (less than 30 minutes), and this results in the appearance of airborne ^{210}Pb (16). Since the migrations of gaseous ^{222}Rn and aerosols (containing Pb) are influenced by atmospheric processes, concentrations of ^{210}Pb in soil might depart from those of ^{226}Ra. Airborne ^{210}Pb not only falls onto the ground but also accumulates on various surfaces and ends up in soil through the decomposition of plants (e.g., leaves) (28–32). The latter process might be extended over a number of years, and it is not surprising that the concentration

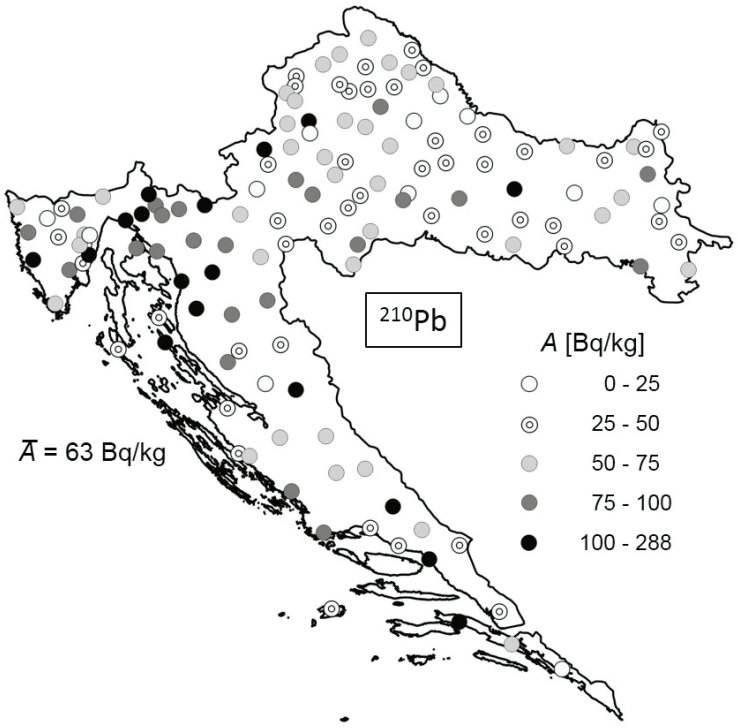

Figure 5 Distribution of the A of ^{210}Pb in Croatian soil

of ^{210}Pb in soil could be higher in areas with rich vegetation (29, 31). These effects provide a plausible explanation of the fact that the *A* of ^{210}Pb in our samples exceeded that of ^{226}Ra mainly in forested areas or elsewhere where the vegetation was dense enough (16).

Areas of potentially higher risk from radon

The ^{232}Th and ^{238}U decay chains in soil contribute significantly to the external exposure of humans and biota to ionising radiation, as many of the members of the chains are gamma emitters (33). When their activity concentrations are small to moderate – which is our case – this contribution usually does not pose a threat. However, the presence of airborne ^{222}Rn is a different matter. High-energy charged particles – especially gamma particles – are more dangerous in internal exposure than gamma photons, which makes the inhalation of ^{222}Rn (an gamma emitter) a serious health hazard (34). Actually, there have been substantiated arguments which relate the inhalation of ^{222}Rn to the risk of lung cancer (35). In turn, radioecological and radiotoxicological concerns with regard to the naturally occurring decay chains are in most cases related to the concentration of ^{222}Rn in air, especially in dwellings.

Areas of potentially higher risk from radon are usually termed "radon prone areas". Their identification, however, is a complex issue, as it must take into account numerous factors that affect the appearance of ^{222}Rn in air and its accumulation in dwellings. There are different approaches to this issue, and some of them include potential formation of ^{222}Rn as a starting point (36). Our data allow us to only determine areas in Croatia where the formation of ^{222}Rn in the topmost layer of soil is comparatively enhanced because ^{226}Ra concentrations are above average. From our results, it follows that these areas in Croatia are mainly in region II and, to a lesser extent, region III. Preliminary results of direct measurements of indoor radon in different parts of Croatia within the Action Plan for Radon 2019–2024 suggest that elevated radon concentrations have indeed been found mainly in these regions (37).

CONCLUSIONS

Our results for activity concentrations of the naturally occurring ^{238}U and ^{232}Th decay chains in the surface layer of uncultivated soil followed the variations of the environmental conditions in Croatia. Measured activity concentrations of the ^{232}Th and ^{238}U decay chains were generally the highest in the Dinaric region, the lowest in the Pannonian region, and intermediate in the Mediterranean region. Possibly the most significant radioecological consequence of this distribution is a comparatively high potential for the formation of ^{222}Rn in the soil of the Dinaric region. Moreover, activity concentrations of ^{210}Pb, which is the longest-living product of the decay of ^{222}Rn, were additionally elevated in areas with dense vegetation. We attribute this enhancement to atmospheric deposition of airborne ^{210}Pb onto the surface of plants and their decomposition on the ground.

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Radioaktivnost tla u Hrvatskoj I. – prirodnopojavni lanci raspada

U radioaktivnosti okoliša velika se pažnja posvećuje radionuklidima u tlu. Taj interes slijedi iz značajnih doprinosa tla i vanjske i unutarnje izloženosti ionizirajućem zračenju izravnom emisijom gama-zračenja i transferom radionuklida iz tla u biljke. To nas je motiviralo da provedemo sustavno istraživanje radioaktivnosti tla u Hrvatskoj kako bismo dobili relevantne podatke kao osnovu za razumijevanje pripadnih učinaka geomorfoloških, biogeografskih i klimatoloških svojstava okoliša. Prikupili smo uzorke površinskoga sloja (0–10 cm) nekultiviranoga tla s 138 lokacija diljem zemlje te smo mjerili koncentracije aktivnosti u njima koristeći se visokorezolucijskom gamaspektrometrijom. To je rezultiralo mapama radioaktivnosti hrvatskoga tla, koje sadržavaju podatke o koncentracijama aktivnosti reprezentativnih radionuklida u okolišu. U ovom radu, koji je prvi u našoj dvodijelnoj prezentaciji, fokusirali smo se na prirodnopojavne ^{232}Th i ^{238}U lance raspada i njihove korelacije s raznolikošću hrvatskih regija. Za oba su lanca koncentracije aktivnosti bile najviše u dinarskom području, najniže u panonskom području i srednje u jadranskom području. Posebice, relativno visoke koncentracije ^{226}Ra u tlu dinarskoga područja impliciraju mogućnost povišene emanacije njegova potomka ^{222}Rn u zrak. Koncentracije aktivnosti ^{210}Pb bile su dodatno povišene u područjima s gustom vegetacijom, vrlo vjerojatno zbog atmosferske depozicije iz zraka na biljke te na njihovu konačnu dekompoziciju na tlu.

KLJUČNE RIJEČI: ^{210}Pb; ^{222}Rn; ^{226}Ra; ^{232}Th; ^{238}U; gama zračenje; reprezentativni radionuklidi; visokorezolucijska gamaspektrometrija