^{137}Cs And Naturally Occurring Radionuclides In Soil In Croatia

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ABSTRACT

Soil contributes significantly to both external and internal exposure to ionising radiation, via direct emission of gamma radiation and soil-to-plant radionuclide transfer, respectively. Next to primordial radionuclides, soil contains radionuclides dispersed during nuclear weapon testing and Chernobyl and Fukushima accidents, especially ^{137}Cs. This motivated us to carry out a systematic research on the radioactivity of soil in Croatia, with a goal to obtain relevant data about the spread of ^{137}Cs in Croatia, as well as about the primordial radionuclides. We had 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 maps of the radioactivity of Croatian soil, containing data on activity concentrations of representative radionuclides in the environment. We focused on ^{137}Cs, and also mapped ^{40}K and radionuclides from ^{232}Th and ^{238}U decay chains. We found that the concentrations of ^{137}Cs tended to increase with altitude, annual precipitation, and vegetation density. Activity concentrations of ^{40}K were the highest in the Pannonian region. The ratio of the concentrations of ^{137}Cs and K in soil, representing the potential for ^{137}Cs entering food chains via uptake by plants, was the lowest in agriculturally important areas in the east of the Pannonian region.

For both ^{232}Th and ^{238}U decay chains, activity concentrations were the highest in the Dinaric region, the lowest in the Pannonian region, and intermediate in the Adriatic region. In particular, 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.

We used the obtained results on activity concentrations to calculate the related absorbed dose rate as a measure of external exposure to ionising radiation from soil. The sum of the absorbed dose rates for naturally occurring radionuclides and ^{137}Cs showed that the external exposure was generally the highest in the Dinaric region and the Istrian Peninsula.

Keywords: *gamma radiation, high-resolution gamma-ray spectrometry, radioecology, representative radionuclides, ^{137}Cs ^{40}K ^{210}Pb ^{222}Rn ^{226}Ra ^{232}Th ^{238}U*

1 INTRODUCTION

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, medicine, etc., implies their possible leakage into the environment, which has increased the 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.

For primordial radionuclides, concentrations depend on geomorphological, biogeographical, and climatological characteristics. Anthropogenic radionuclides mostly came into soil as a result of atmospheric nuclear weapons testing and Chernobyl and Fukushima accidents.

^{137}Cs is considered particularly harmful to living organisms due to its long half-life of 30.1 years, large production rate, and chemical properties similar to those of other alkali metals. In fact, activity concentrations of ^{137}Cs in the environment are – together with those of ^{222}Rn - in the focus of radioecology [1]. The ecological impact of ^{137}Cs is determined not only by its radioactivity (it emits both β and γ radiation) but also by the physical and chemical properties that govern its propagation through the environment and biota. The Cs^+ ionic state facilitates the substitution of potassium and sodium (biogenic elements) with ^{137}Cs in living organisms, which implies that the concentration of ^{137}Cs in a given medium should be compared at least with that of K – which is possible via measurements of radioactive ^{40}K.

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 the radioactivity of the surface layer of soil throughout the country. Even though the radioactivity of soil in Croatia has been measured for decades [2], this has so far lacked a systematic approach. Hence, 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. In particular, the realisation of the objective (d) may be useful for the recent initiative for augmenting and improving The European Atlas of Natural Radiation that has been developed and maintained by the Joint Research Centre [3], as well as to improve on work presented in the Atlas of caesium deposition on Europe after the Chernobyl accident [4].

Croatia comprises three distinct regions. The north belongs to the Pannonian Plain and adjacent hilly regions with the same underlying geology, whereas the climate and vegetation are continental. Along the Adriatic coast and on the islands, karst prevails, the climate and vegetation being Mediterranean. Between these two regions are highlands and mountains belonging to the Dinaric Alps, having a cold continental climate and hosting subalpine vegetation on limestone and dolomite. Therefore, three regions of different climatological, biogeographical, and geomorphological properties – all of them found elsewhere in Europe too – are present in Croatia over a relatively small area, and our study may have implications, at least to some extent, for a more general identification of the effects of the mentioned characteristics on soil radioactivity. The regions are presented in Figure 1. 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 the vegetation is subalpine, whereas in subregion IIb, karst prevails and the climate is Mediterranean. Region III is Mediterranean in both the climate and vegetation. In subregion IIIa, the influence of regions I and II is stronger than in subregion IIIb. Sizable areas of dense forests are indicated by a letter F.

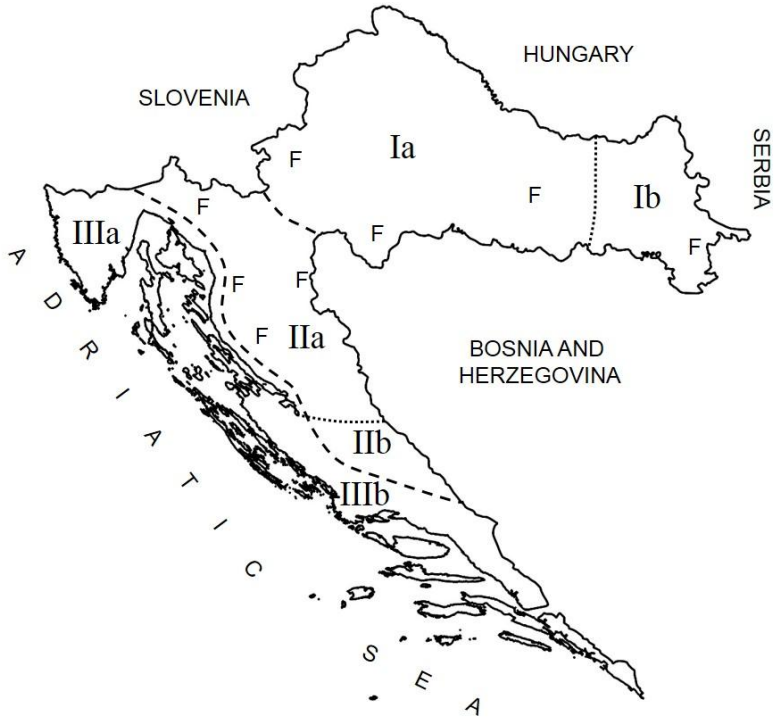

Figure 1: Main Geomorphological, Biogeographical, and Climatological Regions of Croatia

2 MATERIALS AND METHODS

In sampling and measurements, we followed a procedure recommended by the IAEA [5] in order to ensure the compatibility of our results with those obtained elsewhere. Soil was sampled during 2015 and 2016 at 138 locations throughout Croatia. 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 γ radiation from soil is related to this layer due to the attenuation of the radiation from the soil underneath. Collected soil (sampled randomly at ten spots within a $1m^2$ area) was mixed, in order to form a representative sample, and then cleaned from organic material, dried, sieved, and ground. A 100 mL cylindrical plastic container was then filled with thus processed soil and sealed tightly. As explained below, the activity of several radionuclides of interest was determined from the activity of a decay product with a shorter and under the assumption of a secular equilibrium between them. In undisturbed soil, the equilibrium between ^{238}U and ^{234}Th has been established naturally, and the same applies to the equilibrium of ^{232}Th and ^{228}Ac.

However, the loss of gaseous ^{222}Rn from the surface layer of soil, and also during the preparations of samples, 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 γ-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 [6] and [7], respectively.

Our focus was on representative, most widely studied anthropogenic and naturally occurring radionuclides in soil, these being ^{137}Cs (the most abundant long-lived anthropogenic radionuclide in

the environment), ^{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), and ^{40}K. Their activities were determined by analysing peaks at: 661.7 keV for ^{137}Cs; 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. Measurements were carried out for 24 h, which resulted in the following typical values of the detection limit (DL): 0.3 Bq kg^{-1} for ^{137}Cs, 1 Bq kg^{-1} for ^{232}Th and ^{226}Ra, 2 Bq kg^{-1} for ^{40}K; 3 Bq kg^{-1} for ^{210}Pb, and 4 Bq kg^{-1} for ^{238}U.

3 RESULTS AND DISCUSSION

3.1 ^{137}Cs

As outlined before, ^{137}Cs represents a serious radioecological threat. The origin of most of the present-day ^{137}Cs in Croatia is the radionuclide release during the Chernobyl disaster in 1986, with smaller contribution of nuclear weapons tests and much smaller contribution of the Fukushima accident [8], [9]. The distribution of the A of ^{137}Cs in Croatian soil (in 2015-16) is displayed in Figure 2. The measured values ranged from 0 (more precisely, less than the detection limit) to 205 Bq kg^{-1}, with 25 Bq kg^{-1}.

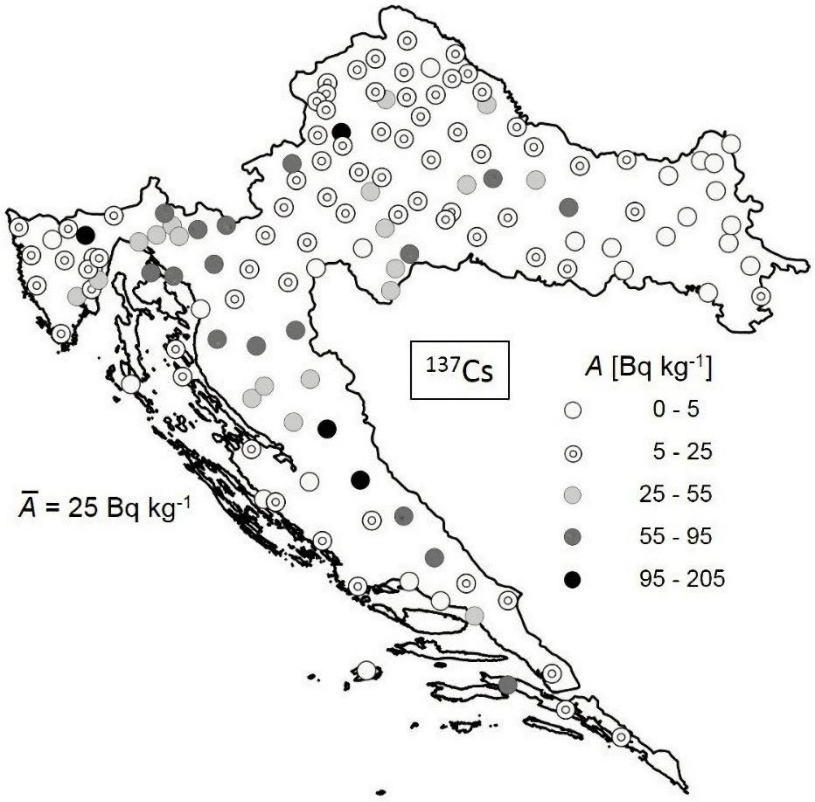

Figure 2: Distribution of the A of ^{137}Cs in Croatian Soil

Generally, the A of ^{137}Cs in our samples was lower than that for ^{232}Th, ^{238}U, ^{226}Ra, ^{210}Pb, and ^{40}K. There was also no dependence on distance from Chernobyl, as the lowest values (~4 Bq kg^{-1} in average) were measured in subregion Ib, i.e., the closest to Chernobyl. In order to understand results in Figure 2, one has to address several factors which might have affected the deposition of (initially airborne) ^{137}Cs onto the ground and its fixation in soil. These factors are expected to be primarily related to atmospheric phenomena and geomorphological properties, with a possible influence of physical and chemical properties of soil [10]. ^{137}Cs is still abundant in the upper atmosphere, from

whence it has been deposited onto the ground mainly via precipitation. When we compare results in Figure 2 with the altitude and annual amount of precipitation [11] at sampling locations, we notice a trend that higher altitudes and more precipitation favoured . The effect of altitude might be a consequence of the pinning of clouds by high mountains, so that ^{137}Cs could be deposited onto the ground directly from the moist air, without actual precipitation. However, we can only identify the mentioned trend, since there are other variables involved. For instance, specific soil properties may have a strong effect on the fixation of ^{137}Cs in the surface layer, as discussed for chernozem in subregion Ib [10].

3.2 ^{40}K

Figure 3 shows the distribution of the A of ^{40}K (= 1.3 billion years) in the surface layer of soil in Croatia. The range of measured A was 62-769 Bq kg^{-1}, and the average value was 423 Bq kg^{-1}. Globally, the reported median and range are 400 Bq kg^{-1} and 140-850 Bq kg^{-1}, respectively, and our results, therefore, do not depart from those obtained elsewhere (including the rest of Europe) [12]. Apart from some local deviations, there was a trend of A being above average in region I and below average in regions II and III. Hence, the A of ^{40}K was generally lower in the regions underlain by karst, where limestone and dolomite bedrock dominated [13]. This behaviour was opposite to that found for the ^{238}U and ^{232}Th decay chains.

^{40}K represents 0.012% of total K, thus a distribution of ^{40}K is actually the same as that of K. Since K is a biogenic element, its concentration in soil depends not only on underlying geology but also on the exchange of matter between soil and living organisms. Generally, ^{40}K contributes significantly to absorbed dose rate for external exposure to ionising radiation from soil, see later, but, in our case, no health hazard can be associated with measured activity concentrations.

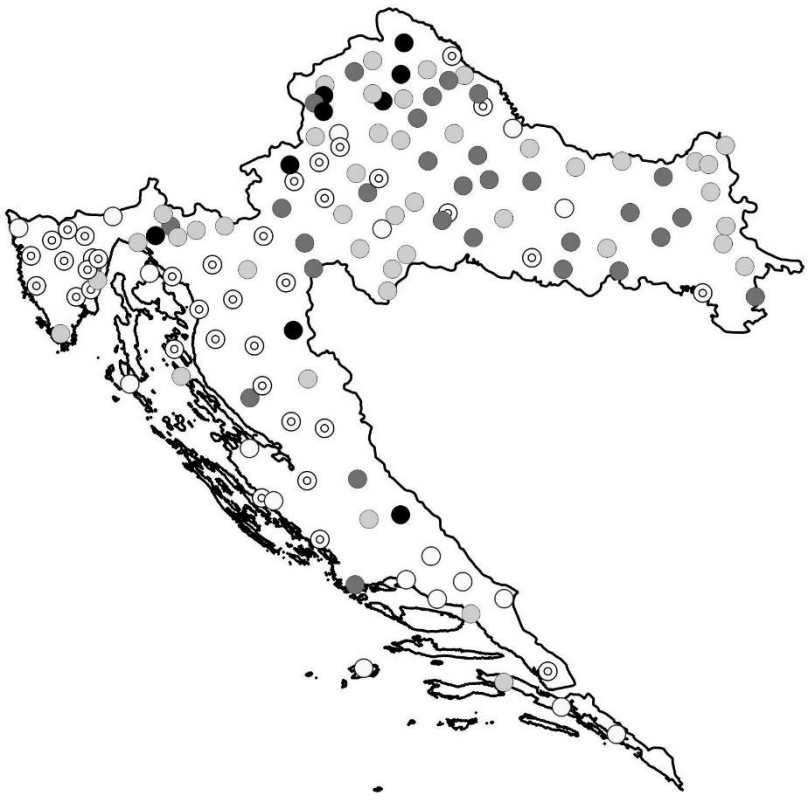

Figure 3: Distribution of the A of ^{40}K in Croatian Soil

Due to their similar electronic structures, K and Cs have similar chemical properties. At insufficient concentrations of K in soil, plants tend to take up chemically similar elements, which includes ^{137}Cs as well [14]. In fact, fertilisers rich in K have been used for the reduction of the uptake of ^{137}Cs in heavily polluted areas [15]. Therefore, the ratio R of atomic concentrations of ^{137}Cs and total K can be defined. This ratio can be considered a measure of the potential of ^{137}Cs to enter food chains via uptake by plants. This ratio can be calculated as (1)

(1)

where and are activity concentrations of ^{137}Cs and ^{40}K, respectively, and $N(^{137}Cs)$ and N(K) are atomic concentrations (total number of atoms per unit of mass) of ^{137}Cs and K, respectively. Results of this calculation are displayed in Figure 4. On average, R is in region I lower than in regions II and III, which is a consequence of relatively high and low in region I compared to the regions II and III. It is of particular importance that R is the lowest in subregion Ib, where the majority of Croatian intense agricultural production is taking place.

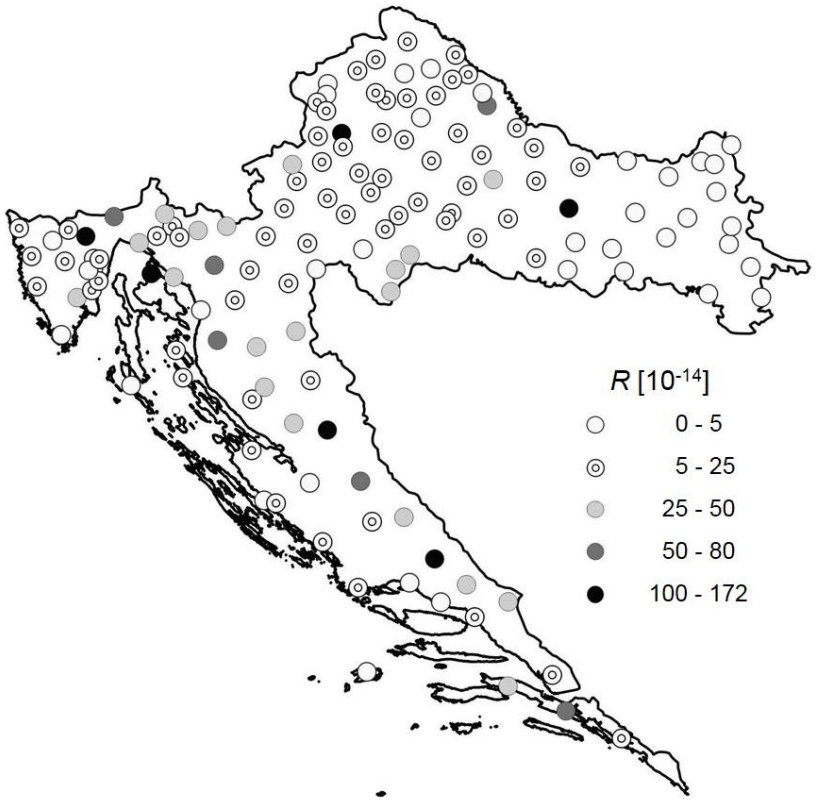

Figure 4: Distribution of the Ratio of Atomic Concentrations of ^{137}Cs and K in Croatian Soil

3.3 ^{232}Th and ^{238}U

Figure 5 shows the distribution of the A of ^{232}Th (= 14 billion years) in Croatian soil, and Figure 6 that of ^{238}U (= 4.5 billion years). The two main naturally occurring decay chains - comprising numerous radionuclides - start with ^{232}Th and ^{238}U. Therefore, these results signify the level of the radioactivity of the whole chains (peculiarities related to the ^{238}U chain will be discussed later).

The average values ( ) of A for ^{232}Th and ^{238}U were similar, being 41 Bq kg^{-1} and 45 Bq kg^{-1}, respectively. The range of A for ^{232}Th was 8–85 Bq kg^{-1}, and for ^{238}U, 0-140 Bq kg^{-1} (here and henceforth, 0 means that measured A was below the corresponding DL). Globally, the reported medians and ranges are 30 Bq kg^{-1} and 11-64 Bq kg^{-1} for ^{232}Th, and 35 Bq kg^{-1} and 16-110 Bq kg^{-1}

for ^{238}U, respectively. Our results, therefore, do not depart from those obtained elsewhere (including the rest of Europe) [12].

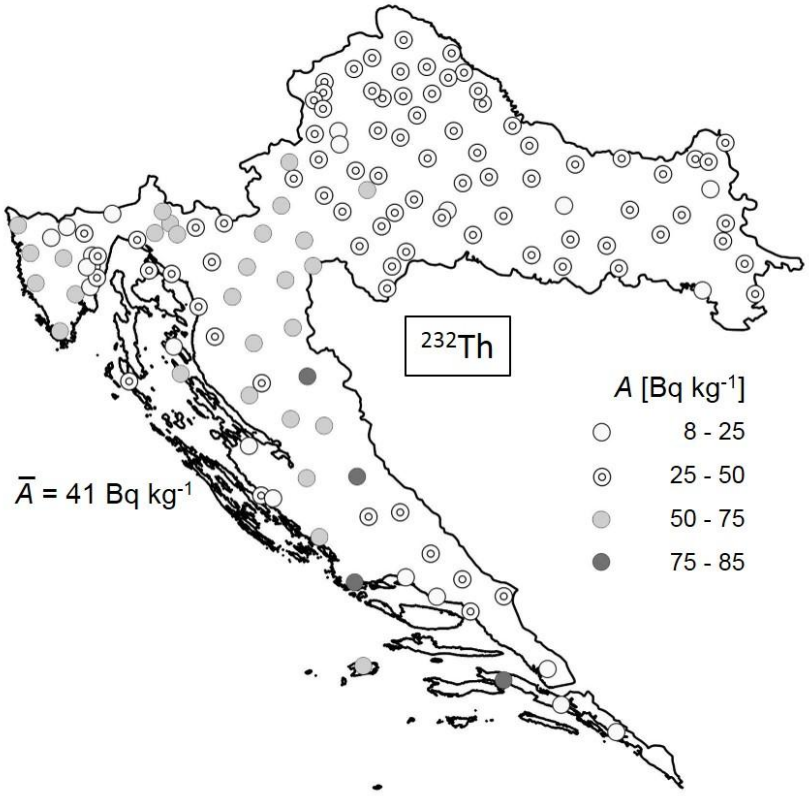

Figure 5: Distribution of the A of ^{232}Th in Croatian Soil

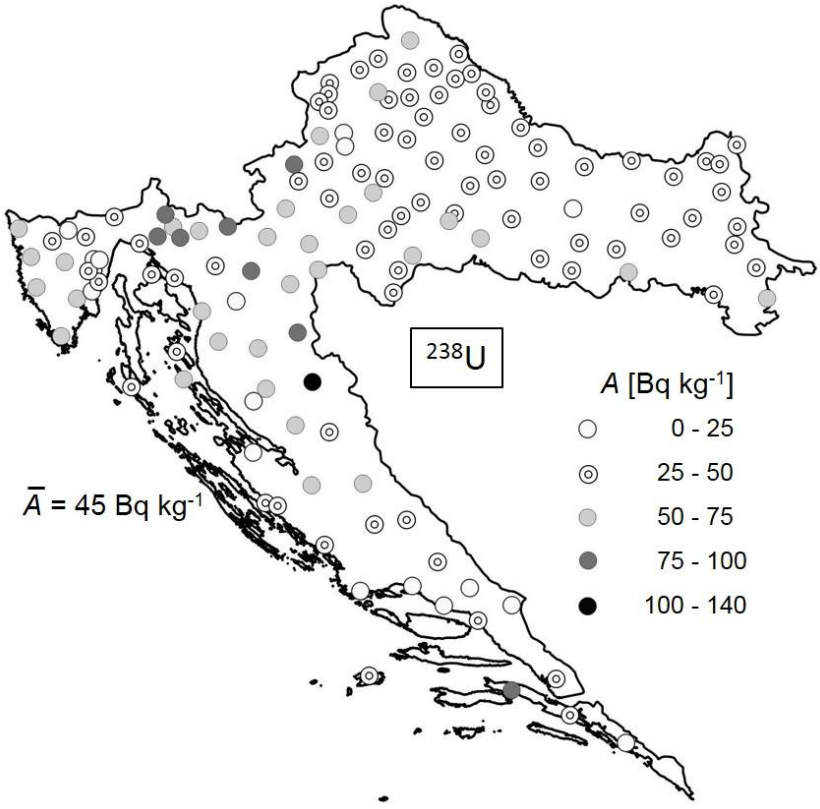

Figure 6: Distribution of the A of ^{238}U in Croatian Soil

A comparison of results in Figure 5 and Figure 6 with the sketch of Croatian regions (Figure 1) reveals a certain trend that was similar 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 [13]. 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.

3.4 ^{226}Ra and ^{210}Pb

Figure 7 shows the distribution of the A of ^{226}Ra (= 1600 years) in Croatian soil, and Figure 8 that of ^{210}Pb (= 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^{-1}, and = 57 Bq kg^{-1}. For ^{210}Pb, the range was 0-288 Bq kg^{-1}, whereas = 63 Bq kg^{-1}.

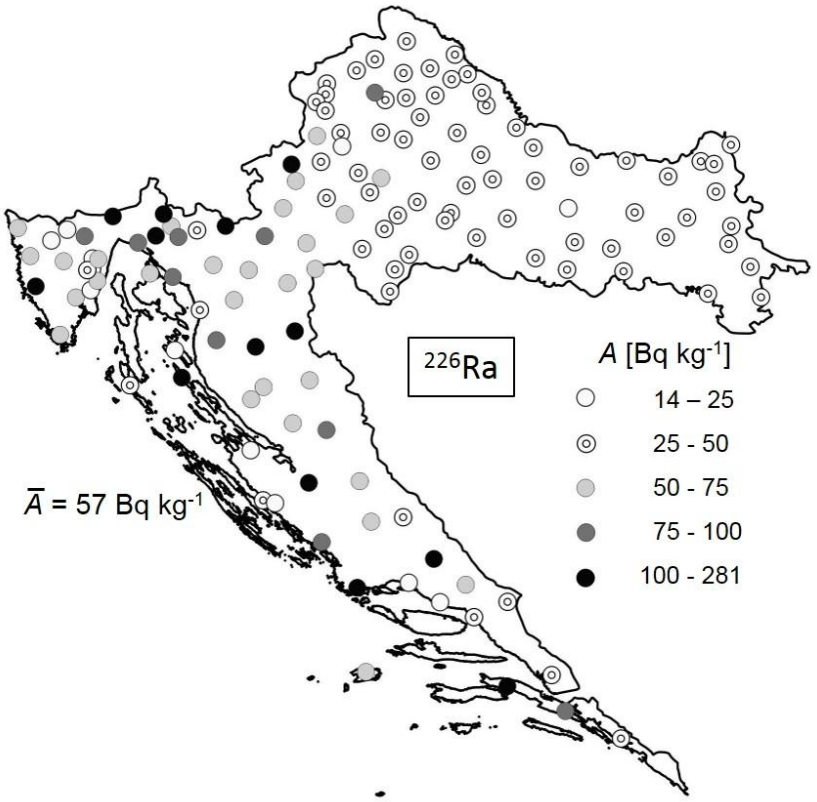

Figure 7: Distribution of the A of ^{226}Ra in Croatian Soil

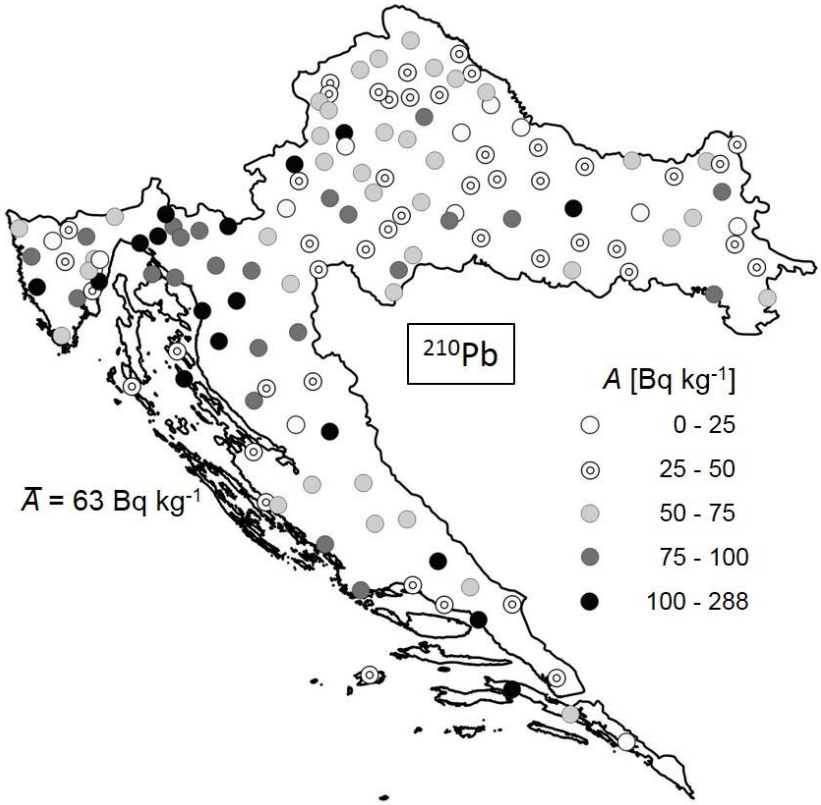

Figure 8: Distribution of the A of ^{210}Pb in Croatian Soil

Globally, the reported median and range for ^{226}Ra are 35 Bq kg^{-1} (the same as for ^{238}U) and 17-60 Bq kg^{-1} (narrower than for ^{238}U), respectively [12]. However, there are considerable variations of these quantities in the data for Europe, and our finding, therefore, does not represent an anomaly. Since ^{226}Ra and ^{210}Pb appear in the chain later than ^{238}U and have shorter , their activities were expected to be smaller or equal to that of ^{238}U [17]. 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.

We first concentrate on ^{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 measured values of the A of ^{226}Ra corresponded to locations in region II, i.e., to 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 the ^{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 the 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 [18], [19]. This process might at least partly account for the fact that the largest activity concentrations of ^{226}Ra were measured in samples from the highly forested northwestern part of subregion IIa (Gorski Kotar).

^{226}Ra decays into ^{222}Rn, a noble gas with = 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 under one minute. The gaseous nature of ^{222}Rn and its relatively long lead to its appearance in the air close to the ground. This is a consequence of its diffusion through soil and emanation into the air

above [20], [21]. Hence, the ^{238}U chain differs from the ^{232}Th and ^{235}U chains by 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 [22]. Of these, ^{210}Pb has the longest 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 and maximum A were larger than those of the mentioned radionuclides from the same decay chain. In particular, by comparing Figure 1, Figure 7, and Figure 8, one can note that the A of ^{210}Pb tended to be larger 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 (less than 30 minutes), and this results in the appearance of airborne ^{210}Pb [17]. 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 by the decomposition of plants (or some of their parts, e.g., leaves) [22], [23], [24], [25]. The latter process might be extended over a number of years, and it is not surprising that the concentration of ^{210}Pb in soil could be enhanced in areas with rich vegetation [23], [24]. 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 [17].

3.5 Absorbed dose rate

Internal exposure to ionising radiation due to the radioactivity of soil depends on numerous factors and has to be estimated on the basis of specific conditions. On the other hand, there is a well-established method of assessing external exposure from measured activity concentrations of radionuclides in soil and their γ emissions. For naturally occurring radionuclides, absorbed dose rate at 1 m above the ground can be calculated from [26]:

(2)

where is in nGy h^{-1}, and , and are activity concentrations of ^{40}K, ^{238}U, and ^{232}Th, respectively, expressed in Bq kg^{-1}. Therefore, our results are sufficient for calculating the distribution of for Croatia. These results are shown in Figure 9. ranged from 23 to 131 nGy h^{-1}. Its average was 63 nGy h^{-1}, exceeding the global median value of 51 nGy h^{-1} moderately [12].

It the case of ^{137}Cs, there is no general expression for calculating the related absorbed dose rate from . Hence, we have to rely on estimates based on other results and models. There are arguments that 1 Bq m^{-2} due to ^{137}Cs in soil results in about 10^{-3} nGy h^{-1} at 1 m above the ground [27], [28]. Other studies indicate that only the topmost ~10 cm layer of soil contributes to , since decreases with depth significantly [29], [30] and also because of the self-attenuation effects [6]. Total ^{137}Cs activity in the upper 10 cm of a 1 m^2 surface layer of soil therefore equals numerically, where ρ is soil mass density in kg m^{-3}. This implies that

(3)

in nGy h^{-1}. By taking into account measured and ρ, we obtained results shown in Figure 10. ranged from 0 to 24 nGy h^{-1}, and the average value was 2.7 nGy h^{-1}, which was considerably smaller than . Therefore, while ^{137}Cs in Croatia may still pose a certain threat regarding internal exposure – due to its competition with K in food chains – its contribution to external exposure is comparatively small.

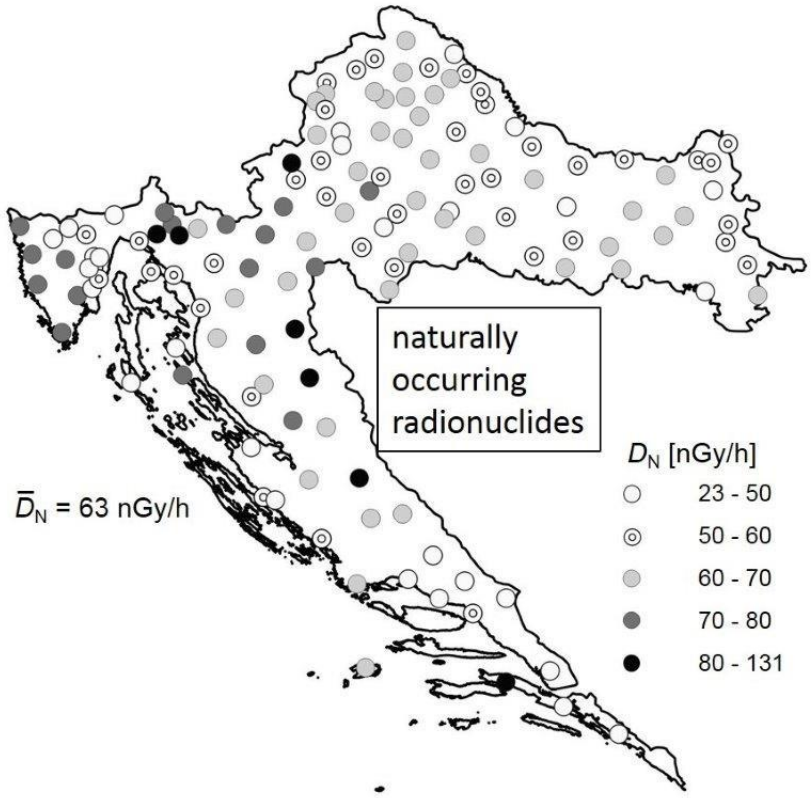

Figure 9: Absorbed Dose Rate Due to Naturally Occurring Radionuclides in Croatian Soil

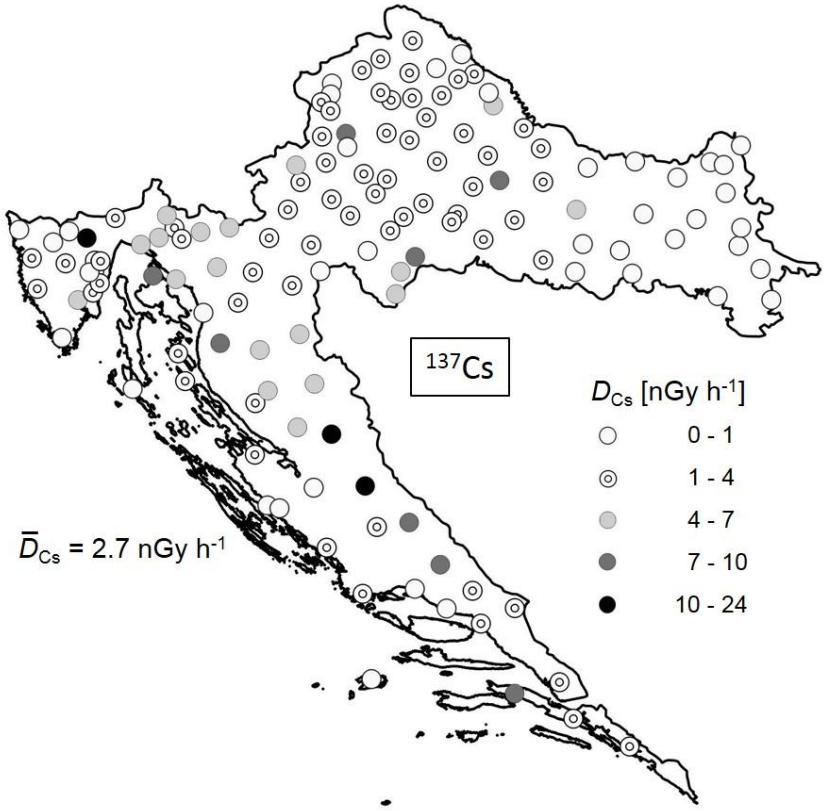

Figure 10: Absorbed Dose Rate Due to ^{137}Cs in Croatian Soil

In Figure 11, we present the distribution of the total absorbed dose rate due to soil radioactivity in Croatia. ranged from 24 to 136 nGy h^{-1}, and its average value was 66 nGy h^{-1}. In most of region II, was above average, which was not surprising because only activity concentrations of ^{40}K were not elevated there. However, the values of in the northwest of subregion IIIa, i.e., on the Istrian Peninsula, were also above average at most of the sampling spots (along the western coast in particular).

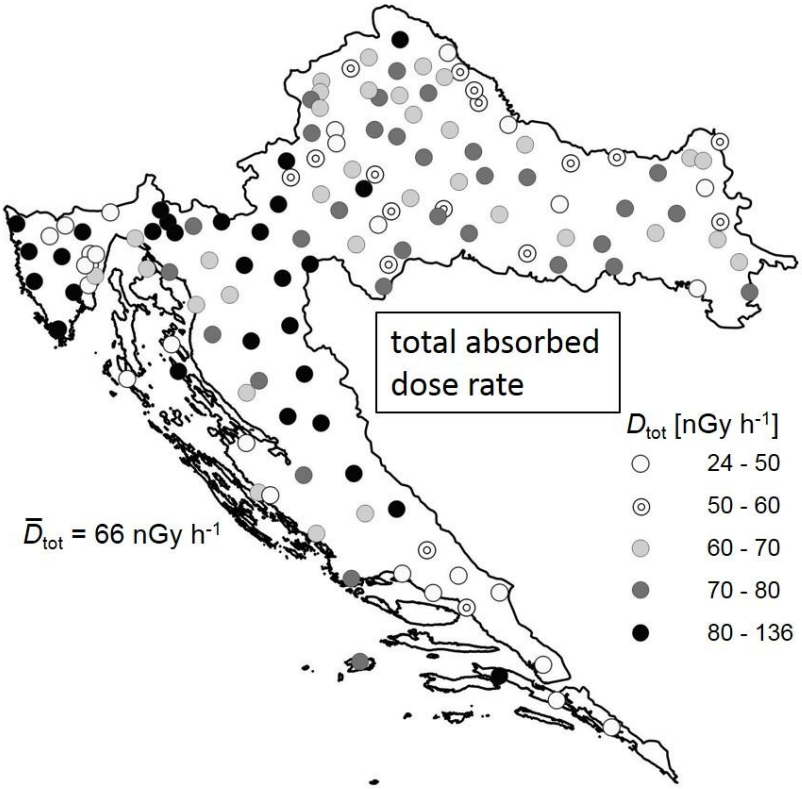

Figure 11: Total Absorbed Dose Rate Due to Radionuclides in Croatian Soil

4 CONCLUSION

Since ^{137}Cs and K have similar chemical properties, they compete in uptake by plants, which implies that their relative concentrations in soil are of interest for the propagation of ^{137}Cs through food chains. Measured activity concentrations of ^{40}K were the highest in the Pannonian part of Croatia, and there was an opposite trend for ^{137}Cs. Their ratio was, consequently, the smallest in the Pannonian region, especially in its eastern part where the majority of Croatian intense agricultural production has been taking place. Activity concentrations of ^{137}Cs tended to be above average at high altitudes, in areas where the annual precipitation was above average and the vegetation dense. 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 assign this enhancement to an atmospheric deposition of airborne ^{210}Pb onto the surface of plants and their decomposition on the ground. By combining the results for ^{137}Cs and ^{40}K with those for the ^{232}Th and ^{238}U decay chains, we calculated the distribution of the absorbed dose rate (external exposure) due to the radioactivity of soil in Croatia. The highest values were in the Dinaric region and on the Istrian Peninsula. Overall,

since this study relied on long-lived radionuclides, our findings may remain viable for a prolonged period of time, and they also might be useful as a reference in case of an accidental appearance of extra radioactivity in the environment of Croatia.

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