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Some Problems of the Radiometric Mapping of Carbonate Terrains - An Example From Istria (Croatia)

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Ključne riječi: radiometrijsko kartiranje, karbonatni tereni, tlo, podinska stijena, gama-spektrometrija, faktori konverzije, brzina gama doze, Istra, Hrvatska.

Abstract

The gamma dose rates were calculated from the results of gamma-spectrometric measurements of radionuclide concentrations in soils in the region of the Istrian peninsula, Croatia. Calculated gamma dose rate conversion factors (GDRCF) for external exposure above ground, give external dose rates per unit of natural radionuclide concentration in the soil and bedrock. GDRCF in air at a height of 100 cm above ground are tabulated for ^{40}K, ^{232}Th decay series, ^{238}U - ^{230}Th subseries and ^{226}Ra - ^{210}Pb subseries for various source depths in soil or bedrock up to a maximum depth of 120 cm. The radiometric maps must take into consideration gamma doses from soils as well as from underlying rocks in the first metre of ground profile. This is of crucial importance especially for carbonate terrains, where the differences between natural radionuclide concentrations in carbonate derived soils and carbonate bedrock could be of two orders of magnitude. Furthermore, for soils themselves the role of the drainage network in migration processes is undoubtedly very important. All these dependencies make us believe that gamma-spectrometric measurements and related dose rate conversions in connection with *in situ* dose measurements could be the best method for radiometric mapping at least for carbonate terrains with numerous rock outcrops, poor drainage and lithological variability on a local scale. Recalculation of natural background dose rate data, collected by airborne or carborne surveys, into radionuclide concentration in the ground in carbonate terrains is possible in two cases only: when soil is completely absent or where soils are well developed (thicknesses of 25 cm or more).

Sažetak

Na temelju rezultata gama-spektrometrijskog mjerenja koncentracija radionuklida u tlima na području poluotoka Istra, Hrvatska, izračunate su brzine pripadnih gama doza. Proračunati konverzioni faktori gama doze (GDRCF) vanjskog ozračenja iznad površine zemlje daju brzinu vanjske doze zračenja koja potječe od jediničnih koncentracija prirodnih radionuklida prisutnih u tlima i stijenama podine. GDRCF u zraku na visini od 100 cm iznad tla su tabelarno dati za ^{40}K, ^{232}Th raspadnu seriju, ^{238}U - ^{230}Th raspadnu podseriju i ^{226}Ra - ^{210}Pb raspadnu podseriju za izvore smještene u tlu ili podini na različitim dubinama do 120 cm. Radiometrijske karte moraju uvažavati kako gama doze koje potječu iz tala tako i doze koje potječu iz stijena podine u prvom metru dubine profila. Ova je konstatacija izuzetno značajna za karbonatne terene u kojima razlike u koncentracijama prirodnih radionuklida između karbonatnih stijena i tala koja su na njima razvijena mogu biti i dva reda veličine. Nadalje, uloga mreže odvodnjavanja u procesima migracije i redistribucije elemenata u tlima nesumnjivo je veoma bitna. Spomenute ovisnosti nas upućuju na zaključak da gama-spektrometrijska mjerenja i konverzija rezultata u pripadne brzine doze, u kombinaciji s mjerenjima doza *in situm*, predstavlja u najmanju ruku najbolji mogući način radiometrijskog kartiranja. To pogotovo vrijedi za karbonatne terene s brojnim izdancima, slabom mrežom odvodnjavanja i velikim litološkim razlikama na relativno malom prostoru. Preračunavanje brzine prirodne gama doze, a koja je dobivena snimanjima gama spektara iz zraka ili automobila, u pripadne koncentracije radionuklida u tlu, u karbonatnim terenima je moguće samo u dva slučaja: kada tala uopće nema, te u slučaju relativno dobro razvijenih tala debljine 25 cm ili više.

1. INTRODUCTION

This work is part of the national project entitled "General Geochemical Map of Croatia" and also lies within the scope of the UNESCO International Geological Correlation Programme (IGCP) Project 259 - International Geochemical Mapping (IGM), which was terminated and replaced by a new IGCP Project - International Geochemical Baselines. The IGM project was launched to address the problem of the distribution of chemical elements, including radioactive ones, in the earth's surface materials. Current research is based on the aerial gamma-ray spectrometry measurements (airborne or carborne surveys) and on data conversion into radionuclide concentrations in soil, generally. In the context of the IGM project, the main function of ground gamma-ray spectrometry measurements with a portable gamma-ray spectrometer is to provide a link between data gathered by conventional geochemical sampling and related airborne gamma-ray profiles. Many useful details about aerial gamma-ray spectrometry measurements have been published (INTERNATIONAL ATOMIC ENERGY AGENCY, 1990), and regional geochemical maps of some primordial radionuclides have been documented (DUVAL, 1990, 1991).

Airborne gamma surveys which have been applied and recommended for radiometric mapping of most

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national territories are not always suitable, mostly because of the complexity of the terrains especially in areas predominantly covered by carbonate rocks. Additionally, the high cost and other technicalities make this technique unsuitable for radiometric mapping of the national territory of the Republic of Croatia. In 1969, when part of the Republic of Slovenia was covered by airborne measurement, only a negligible part along the Croatian-Slovenian border was included. In the 1970's, during uranium exploration in Slovenia and Croatia, part of Croatia, mostly covered by clastic rocks (Lika, Gorski Kotar) and terrains predominantly built of eruptive and metamorphic rocks (Slavonia) were covered by ground gamma ray spectrometry. Unfortunately, this territory is, in general, not accessible for the field work currently, which together with the unavailability from previous investigations makes any checking of the reliability and usefulness of these data impossible.

Radiation from the soil is the most important contributor to external terrestrial radiation because the bedrock is covered with soil in most areas of the world. It is a well known fact that the absorbed gamma dose rate in air is the function of radionuclide concentration in the first metre of the soil profile. Conversion factors for the calculation of dose rates in the air are known for unit specific activity in soil for potassium, uranium and thorium (UNITED NATIONS, 1982). They assume the following: radiochemical equilibrium between radionuclides in ^{232}Th decay series as well as in ^{238}U decay series, and homogeneous concentration in the first metre of soil. The same assumptions are valid in the case of recalculation of aerial gamma-ray spectrometry data into equivalent radionuclide concentrations in the soil (INTERNATIONAL ATOMIC ENERGY AGENCY, 1990).

In carbonate terrains especially, the soil thickness ranges from a few centimetres up to several metres or more, and it may be completely absent in some areas. Although all ^{232}Th decay products are often in radiochemical equilibrium with ^{232}Th, ^{238}U decay products are not in equilibrium with their precursors. In the first few centimeters of carbonate derived soil, disequilibrium between ^{238}U and ^{226}Ra can be very high (GREEMAN et al., 1990). ^{226}Ra is precursor of noble gas ^{222}Rn and its decay products, among which aerial gamma-ray spectrometry determination of ^{214}Bi is the base for ^{238}U assessment in the ground. Differences between natural radionuclide concentrations in carbonate derived soils and carbonate bedrock could be more than 20-fold (GREEMAN et al., 1990). In the case of ^{232}Th the difference can be two orders of magnitude (BARIŠIĆ, 1993). A mass equivalent to a water column of 8 cm would reduce the exposure rate by about 50%. Soil moisture and vegetation have a similar shielding effect (INTERNATIONAL ATOMIC ENERGY AGENCY, 1990).

Aerial gamma-ray spectrometry provides very good quality data of natural background dose rate under any given conditions. But it seems that conclusions about the concentration of natural radionuclides in soils could be less valuable, even erroneous, especially in carbonate terrains. Conventional geochemical sampling of the first 15 cm of the A soil horizon or upper regolith, below the 01 and Of horizons (if they exist), and laboratory gamma-ray spectrometry could be useful and provide a quality data-base for calculation of the resulting absorbed dose rates in air. Models and data-bases for estimating the distribution of radioactivity in soil and their associated external doses above the soil surface have been presented in the literature (BECK & DE PLANQUE, 1968; BECK, 1980; KOCHER & SJOREEN, 1985), but the methods have not yet been used routinely in environmental dose assessment.

The main goal of this paper is to give an initial insight into the radionuclide distribution and resulting external natural gamma dose rates in a typical karst terrain. However, we studied the parameters and factors which could influence the radionuclide concentrations in soils overlying carbonate rocks together with the resulting gamma doses. Different lithological units, the drainage net, thickness of the soil profile, etc., pointed out the influence of various factors upon the radionuclide concentrations in Istrian soils. It is a well known fact that the resulting gamma dose in air is a function of the radionuclide concentration in the first metre of the ground surface profile (in soils as well as in the underlying bedrock). So, it is necessary to use different gamma dose rate conversion factors (dependent on soil thickness and radionuclide specific activity in soils and underlying rocks) in carbonate terrains. This paper also assesses the influence of soil thickness on the external dose rate assessment in the air (one metre) above the ground for a unit primordial radionuclide concentration existing in the soil or bedrock. The calculations of DRCF are derived using the following assumptions: radionuclide concentrations at any depth in soil or underlying rock are uniform over an infinite surface parallel to the ground surface plain, homogenous dry soil density is 1.4 g cm^{-3}, and the distribution of radionuclides in soil or in bedrock is homogeneous.

2. GEOLOGY AND HYDROGEOLOGY OF ISTRIA

The Istrian peninsula is considered to be the western part of a vast Mesozoic Adriatic carbonate platform. This is the classic karst area, where karst phenomena were first studied and from which the name originated. The relief consists of plains, hills and mountains rising eastwards from the coast to heights of about 1400 m. The belt is characterized by distinct climatic zones, soil development and vegetation. The area is composed of Jurassic, Cretaceous, Tertiary and Quaternary sediments (Fig. 1). While western and southern Istria are mainly characterized by shallow-marine Jurassic and Cretaceous carbonate rocks, the rest of the peninsula is

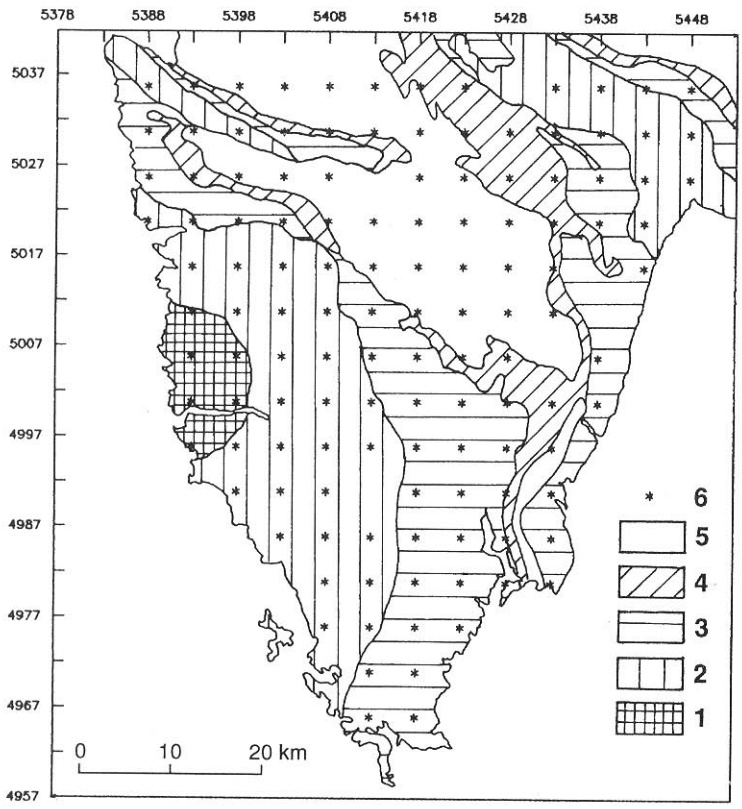

Fig. 1. Geological map of Istria (after the Geological Map of the SFRY, 1:500.000). Legend: 1 - Jurassic carbonates; 2 - Lower Cretaceous carbonates; 3 - Upper Cretaceous carbonates; 4 - Palaeocene-Eocene carbonates; 5 - Eocene Flysch; 6 - Position of samples.

composed of Tertiary foraminiferal limestones and flysch.

The oldest strata exposed in Istria are Middle and Upper Jurassic in age (VELIĆ & TIŠLJAR, 1988). A very short emergence took place after this period, causing local deposition of bauxites. The Cretaceous sequence consists of carbonate rocks deposited from the Tithonian to Lower Campanian. These sediments were raised above sea level by the Laramian orogeny and since then they have been intensively eroded and karstified. The final results of these processes are numerous small bauxite deposits covered by Palaeogene sediments.

The Early Palaeocene was marked by deposition of fresh-water and brackish "Liburnian strata" with coal in the isolated basin. These strata are specially important to this study due to the high content of radionuclides in the associated coal. These layers are discordant with the intensively karstified Cretaceous base. A major and significant transgression invaded this region in the Early Eocene causing the marine sedimentation of foraminiferal limestones and flysch (conglomerates, sandstones, marls and clayey limestones) throughout this period (TIŠLJAR et al., 1983).

The total and final emersion occurred either at the end of Eocene or at the beginning of Oligocene. Emersion from the Early Oligocene until the Recent has caused continuous denudation of the Eocene and Palaeocene sediments from the greater parts of Istria.

The Quaternary consists of different deposits along the river valleys and in the extensive karst poljes. These are black and brown soils, terra rossa, talus deposits, various stream deposits, coastal deposits and lacustrine clays and sands.

The main feature of the hydrogeology of the study area is connected to the presence of carbonate rocks. A regular surface-drainage system is absent or fragmentary (except in the flysch area) because most of the rainfall percolates underground.

3. RADIATION SOURCES IN THE GROUND

The radiation sources that occur in the ground can be classified into two main categories: the natural, primordial radionuclides with their decay products which are present in the earth's crust, and, the radionuclides of various origin which are deposited on the ground surface by dry and/or wet fallout. The latter are generally from one of two sources: natural i.e. cosmogenic radionuclides (^{3}H, ^{7}Be, ^{10}Be, ^{14}C, ^{22}Na, ^{24}Na) and ^{222}Rn decay products and, artificial (man-made) radionuclides. Fallout deposited man-made radionuclides can be divided into two main groups: globally dispersed radionuclides originating from atmospheric nuclear explosions (weapon testing-derived) and, radionuclides originating from accidents in nuclear power facilities (Chernobyl-derived generally, in practice, mainly ^{137}Cs). Primordial terrestrial radiation sources are the long-lived radionuclides that have existed within the earth since its formation. The most important are ^{40}K, ^{87}Rb, and also ^{232}Th, ^{235}U and ^{238}U as heads of their accompanying decay series. The disintegration of radionuclides is followed by particle emission and the

daughter nuclei, often in excitation states, which lose their energy as gamma-ray emissions. Behaviour of the beta emitter ^{87}Rb, which has no gamma lines, is almost unknown. Naturally occurring potassium, one of the major elements in the earth's crust, contains 0.0117% of the ^{40}K isotope with a half-life of 1.28×10^9 years. Gamma ray at 1.46 MeV (10.7% emission intensity) follows ^{40}K decay into stable isotopes of ^{40}Ca and ^{40}Ar.

Thorium and uranium occur in the earth's crust as trace elements. Their soil content depends upon their content in the parent rocks and migration governed by chemical or physical processes. Natural uranium is the mixture of ^{234}U, ^{235}U and ^{238}U isotopes. ^{238}U comprises 99.275% of natural uranium with a $^{238}U/^{235}U$ activity ratio of 0.04603. As a long-lived ^{238}U daughter, ^{234}U comprises only 0.004% of natural uranium. Primordial ^{232}Th decays through daughter radionuclides into the stable lead isotope ^{208}Pb, and similarly, ^{238}U into stable ^{206}Pb as well as ^{235}U into stable ^{207}Pb.

The ^{232}Th, ^{235}U and ^{238}U decay series includes the same element isotopes (radium, noble gas radon, polonium, bismuth, lead) and contains a large proportion of alpha emitters. The main difference between the ^{238}U series on one side, and the ^{232}Th and ^{235}U series on the other, is the fact that ^{232}Th and ^{235}U are the only long-lived radionuclides in the decay series. The relatively long half-life of some daughters results in radiochemical disequilibrium in the ^{238}U decay series. ^{238}U decay series can be divided into two main parts: ^{238}U - ^{230}Th subseries and ^{226}Ra - ^{210}Pb subseries. ^{238}U - ^{230}Th subseries contribute to the ^{238}U decay series and consequently their proportion of the natural background dose rate is very low, less than 1% in the case of radiochemical equilibrium. Possible errors of the ^{238}U - ^{230}Th subseries contribution to the natural background dose rate assessment, caused by radiochemical disequilibrium occurring in this subseries, are insignificant as long as the assessment is based on ^{238}U concentrations.

The ^{226}Ra - ^{210}Pb subseries comprises more than 99% of the ^{238}U decay series proportion of the natural background dose rate in the case of radiochemical equilibrium. Radiochemical equilibrium between ^{238}U and ^{226}Ra in rocks mainly exists, but disequilibrium usually occurs in soil, as a consequence of various physical and chemical processes induced by water migration during rock weathering and pedogenesis, as well as those occurring in the soil itself. Various degrees of general radiochemical disequilibria in the ^{226}Ra - ^{210}Pb subseries are caused by noble gas radon migration and the relatively long half-life of some ^{222}Rn disintegration products.

When ^{226}Ra decays in soil or rock particles, the ^{222}Rn daughter atoms are able to escape from particles to the air or water-filled pores, move through them and escape the ground, entering into the atmosphere. Micropores of nannometric dimensions, alpha-recoil damage in particles and a position close to the surface of individual grains play significant roles in ^{222}Rn ejecting into intragranular pores (FLEISCHER, 1980, 1982). The average radon exhalation rate into atmosphere is less than 0.001 Bq m^{-2} sec^{-1} per ^{226}Ra concentration in soil of 1 Bq kg^{-1} (recalculated according to UNITED NATIONS, 1988). However, this is absolutely insignificant in terms of the natural background dose rate contribution of ^{226}Ra - ^{210}Pb subseries assessment based on ^{226}Ra concentration.

The influence of fallout deposited radionuclides on the ground surface (including long-lived ^{222}Rn disintegration products) is not taken into account in gamma dose rate assessment presented in this paper (only gamma dose rates of ^{137}Cs are given)

4. SAMPLING AND METHODS

4.1. SAMPLING PROCEDURE

Soil was sampled by channel sampling the upper 15 cm of the soil profile (below the O1 and Of horizons, if present). The fraction of air-dried material that passed through a 2-mm sieve was reduced by repeated comminution and quartering to a grain size less then 0.5 mm. The soil types sampled included rendzinas of the A - C soil profile; brown soil on limestone and dolomite, and terra rossa of the A - (B) - C soil profile (calcocambisol); leached brown soils on limestone and dolomite, and terra rossa and rendzina on flysch bedrock (PIRC et al., 1991). Samples were taken in a regular 5x5 km grid with sample points in the middle of the square with the randomly selected position of the grid on the General Geochemical Map of Croatia. The content of inorganic constituents was measured for the same samples (contributing to the geochemical map of Istria). This gives us additional possibilities for comparing the element and radionuclide content in the same samples. In this way a total of 131 samples were collected across Istria and 125 of them were analyzed by the gamma-ray spectrometry.

4.2. GAMMA-SPECTROMETRY

Soil samples were placed in the counting vessels, sealed and stored for at least 4 weeks to allow an ingrowth period of gaseous ^{222}Rn. At the end of the ingrowth period, the samples were counted on a Ge-Li semiconductor detector joined to a 4096 channel analyzer "Canberra". The detector system was calibrated using standards supplied by both the National Bureau of Standards (USA) and Amersham International (UK). Precision and accuracy were checked by simultaneous measurement of International Atomic Energy Agency standards (IAEA-306, IAEA-313 and IAEA-314). Depending on radionuclide activities, spectra were recorded for times ranging from 80,000 to 150,000 seconds.

Activities of ^{40}K were calculated from the 1460.75 keV-peak, ^{137}Cs from the 661.6 keV-peak, ^{226}Ra (as a mean value) from the 609.3 keV-peak of its ^{214}Bi progeny and 352 keV-peak of its ^{214}Pb progeny, ^{228}Ac from

the 911.1 keV-peak and ^{235}U from the 186 keV-peak (after subtraction of the overlapping ^{226}Ra peak). Concentrations of ^{238}U were calculated from the ^{235}U activity assuming the $^{235}U/^{238}U$ activity ratio of 0.04603 (MURRAY & AITKEN, 1988).

4.3. CALCULATION OF GAMMA DOSE RATE

The radiation effects in the air can be expressed in terms of the exposure rate or the absorbed dose rate in air. Conversion factors from radionuclide activity in soil or bedrock to the absorbed dose rate in air have been estimated from photon transport calculations applied to infinite soil or bedrock and air media. Dose rate at reference points for discrete photon energy strongly depend on the attenuation effects in material between the source and the reference point. The numerical solution of this complex mathematical problem evaluates the effect of primary and scattered gamma rays in the ground and in the air.

The natural background dose rate in air is the product of radionuclide specific activities in soil and/or bedrock with the sum of respective products between unit dose rates for each emitted photon energy and their emission intensity. Primordial radionuclides can be considered to be relatively uniformly distributed in the first 100 cm of the vertical soil profile and the natural radionuclides concentration at any depth in soil is assumed to be uniform over an infinite surface parallel to the ground plane. The dose rate, at a point on the ground surface from a monoenergetic gamma source, can be found by integrating the plane source over the depth of soil (DICKSON & KERR, 1975). The unit dose rate factors for all of the gamma-rays and x-ray energies taken into account were obtained from a polynomial least-squares fit of data published by KOCHER & SJOREEN (1985). Data were previously recalculated from volume activity units into specific activity units, assuming a soil density of 1.4 g cm^{-3}. Dose rate factors for material with a density of 2.4 g cm^{-3} (e. g. rocks) are not calculated (according to the INTERNATIONAL ATOMIC ENERGY AGENCY (1990) the factors for converting natural radionuclide concentrations into radiation exposure rates are nearly identical for rocks and soils). The expected higher unit dose rates from rocks, caused by higher activity in the same volume, are reduced by attenuation in more dense rock material.

Intensity of emission and emitted photon energies are taken according to ERDTMAN & SOYKA (1979) and the INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION (1983). All x-ray and gamma-ray energies with emission intensities >0.05% are taken into account, in the case of ^{214}Bi even gamma-rays with the intensity of emissions >0.005%. The increase of absorbed dose rates on higher photon energies indicates the importance of high energy photons, even in the cases of very small emission intensities. The 0.2% energy intensity photon emitted at 3 MeV results in a dose rate in air equal to the 100% energy intensity photon emitted at 0.015 MeV when the soil thickness is 0.5 cm. If the soil thickness is 1 m, only 0.011% of the energy photon emitted at 3 MeV contributes equally to the dose rate as 100% of the energy photon emitted at 0.015 MeV.

Dose rate conversion factors in the air 1 m above ground are given in Table 1 (for soil and bedrock separately) for unit ^{40}K activity, as well as for ^{232}Th decay series, ^{238}U - ^{230}Th subseries, and ^{226}Ra - ^{210}Pb subseries unit head radionuclide activity, in dependence on soil thickness (radiochemical equilibrium in decay series or subseries was assumed). Empty spaces in columns indicate that the last value entered in the column is repeated for all deeper lower boundaries.

Soil thickness (cm)	^{40}K	(a)	(b)	(c)
		(pGy h^{-1} per Bq kg^{-1})		
Dose rate conversion factors for soil				
Outcrop	0.00	0.00	0.00	0.00
0.5	2.77	44.96	0.42	32.80
1	5.43	88.64	0.79	64.23
2	9.66	157.29	1.35	114.41
3	13.22	212.18	1.78	154.46
4	15.92	258.53	2.13	187.64
5	18.43	298.26	2.42	216.34
7.5	23.44	377.13	2.97	273.80
10	27.19	435.04	3.35	315.27
15	32.55	515.07	3.84	372.37
20	35.83	564.40	4.10	405.08
25	38.13	595.86	4.27	428.18
30	39.61	616.57	4.36	441.20
40	41.39	641.16	4.47	457.81
50	42.12	652.92	4.51	464.72
60	42.54	659.71	4.53	468.36
80	42.74	663.88	4.53	470.40
100	42.86	665.44	4.54	471.24
120			665.73	
Dose rate conversion factors for bedrock				
Outcrop	42.86	665.73	4.54	471.24
0.5	40.09	620.77	4.12	438.44
1	37.43	577.09	3.75	407.02
2	33.20	508.44	3.19	356.83
3	29.64	453.55	2.75	316.79
4	26.94	407.21	2.40	283.60
5	24.43	367.47	2.11	254.91
7.5	19.42	288.60	1.57	197.44
10	15.67	230.69	1.19	155.97
15	10.31	150.67	0.70	98.88
20	7.03	101.33	0.44	66.16
25	4.73	69.87	0.27	43.06
30	3.25	49.17	0.17	30.04
40	1.47	24.57	0.07	13.43
50	0.74	12.82	0.03	6.52
60	0.32	6.02	0.00	2.88
80	0.12	1.86		0.84
100	0.00	0.29		0.01
120		0.00		0.00

Table 1. Dose rate conversion factors in the air one meter above ground in dependence on soil thickness. (a) - ^{232}Th series; (b) - ^{238}U - ^{230}Th subseries; (c) - ^{226}Ra - ^{210}Pb subseries.

In the ^{232}Th decay series, the main contribution to the total dose rate arises from ^{208}Tl and ^{228}Ac. Contribution of ^{212}Bi and ^{212}Pb is significant (>10% generally), while the influence of all other radionuclides in series is about 1%. The x-ray radiation contribution in the ^{232}Th decay series (about 1% of dose rate for soil thickness of 0.5 cm and less than 0.4% for soil thickness of 100 cm) is insignificant. It is very important to notice that ^{228}Ac and ^{208}Tl (easily detectable by laboratory gamma-spectrometry) can be used for ^{232}Th decay series dose rate assessment in air. Laboratory measured ^{208}Tl concentrations must be multiplied by factor 0.3593, and then ^{232}Th dose rate factors can be applied for calculation of the ^{232}Th decay series proportion of the natural background dose rate.

The main contribution to the total dose rate in the ^{238}U decay series arises from ^{214}Bi (depending on soil thickness, about 85%), and ^{214}Pb disintegration, about 13% in average. A small contribution arises from ^{234}Th, ^{234m}Pa, ^{234}Pa and ^{226}Ra, while the collective contribution of ^{238}U, ^{234}U, ^{230}Th, ^{222}Rn, ^{214}Po and ^{210}Pb is absolutely insignificant, less than 0.1%. The contribution to the dose rate in air from x-ray radiation in ^{238}U decay series, about 0.4% in average, can be ignored.

It has to be noticed that the main dose rate contribution from radionuclides in the ^{238}U decay series arises from decay products of noble gas ^{222}Rn. It is a daughter of the relatively long-lived ^{226}Ra parent and, in the case of radiochemical equilibrium between them, the dose rate in air is a function of the ^{226}Ra concentration in the soil or bedrock. ^{226}Ra (via its ^{214}Bi or ^{214}Pb decay products) is easily detectable by gamma-spectrometry and can be used for the whole ^{238}U decay series dose rate assessment in air in the cases of radiochemical disequilibrium between ^{226}Ra and ^{238}U. Recalculation of the ^{238}U concentration in soil, based on the ^{238}U decay series dose rate in air, is incorrect in the cases of radiochemical disequilibrium between ^{238}U and ^{226}Ra. The contribution of radionuclides occurring in the ^{235}U decay series (about 1% of the whole ^{238}U decay series on the average) to the total dose rate in air is insignificant.

The values presented in Table 1 are in excellent agreement with previously published data (UNITED NATIONS, 1982, 1988) in the cases of ^{40}K and ^{232}Th decay series, while in the case of ^{238}U decay series, calculated values are about 9.5% higher. It seems that the difference arises from the fact that almost all of the numerous ^{214}Bi gamma rays (including gamma rays with very small emission intensities) are taken into account. The most recent published data of dose rate conversion factors for ^{226}Ra subseries of 461 pGy h^{-1} per Bq kg^{-1} (UNITED NATIONS, 1993) supports this assumption.

Conversion factors for converting dose rates in the air (pGy sec^{-1}) into ppm equivalent uranium (eU), ppm equivalent thorium (eTh) or % K (^{40}K activity of 309.28 Bq kg^{-1} = 1% K, ^{232}Th activity of 4.06 Bq kg^{-1} = 1 ppm eTh and ^{238}U activity of 12.44 Bq kg^{-1} = 1 ppm eU, all recalculated according to SEELMANN-EGGEBERT et al., 1981) are given in Table 2. Some interesting facts have emerged. The published transfer factor values for converting dose rate in air into ppm eU, ppm eTh or % K (INTERNATIONAL ATOMIC ENERGY AGENCY, 1990), were verified by experimental measurements of the exposure dose over various geological materials with known natural radionuclide concentrations. They are lower generally, but in very good agreement with the conversion factors presented in Table 2 for ^{40}K (1.3% lower) and for ^{238}U (3.4% lower). At the same time they are 8.2% lower for ^{232}Th. It is possible that this is caused by the shielding effect of soil moisture especially in the case of high-energy ^{208}Tl gamma rays emitted from deeper soil layers.

GDRCF, for exponentially distributed ^{137}Cs in soils (112.5 pGy h^{-1} per Bq kg^{-1}) was found on the base of ^{137}Cs dose-rate conversion factor of 4.5 x 10^{-13} Gy h^{-1} per Bq m^{-2}, published by LEUNG et al. (1990), and assumption that ^{137}Cs specific activity of 1 Bq kg^{-1} in the first 15 cm of soil is equivalent to ^{137}Cs contamination of 250 Bq m^{-2}.

5. RESULTS AND DISCUSSION

By referring to the gamma dose rate conversion factors (GDRCF) shown in Table 1, the absorbed dose-rates at 1 m above ground from the natural radionuclides and from ^{137}Cs were calculated and are given in Fig. 2. Before any discussion one must be aware that all calculated values are based on the assumption that, at every sample point, the soil thickness is 1 m. We intentionally included this erroneous assumption in our calculation, largely because we have not yet solved the problem of averaging the soil thickness in carbonate terrains by using a regular grid. Data from the pedological map of Istria (ŠKORIĆ et al., 1987) reveal that the soil thickness is ranges from a few centimetres to several metres, but the majority of the area under investigation (excluding flysch) is very probably covered by soil less than 50 cm in thickness. About 90% of the natural

	^{40}K	^{232}Th	^{238}U
		Bq kg^{-1}	
CF(a)	43	662	427
CF(b)	42.86	665.44	475.78(c)
	% K	ppm eTh	ppm eU
CF(d)	3.633	0.693	1.576
CF(e)	3.682	0.750	1.631(c)

Table 2. Conversion factors (CF) for converting radionuclide concentration in the ground into absorbed dose rate in the air and vice versa for case of soil thickness of 1 meter or more. (a) - CF pGy h^{-1} into Bq kg^{-1} (UNITED NATIONS, 1988); (b) - CF pGy h^{-1} into Bq kg^{-1} (this work); (c) - ^{238}U in radiochemical equilibrium with ^{226}Ra; (d) - CF pGy sec^{-1} into 1% K, 1 ppm eTh and 1 ppm eU (INTERNATIONAL ATOMIC ENERGY AGENCY, 1990); (e) - CF pGy sec^{-1} into 1% K, 1 ppm eTh and 1 ppm eU (this work).

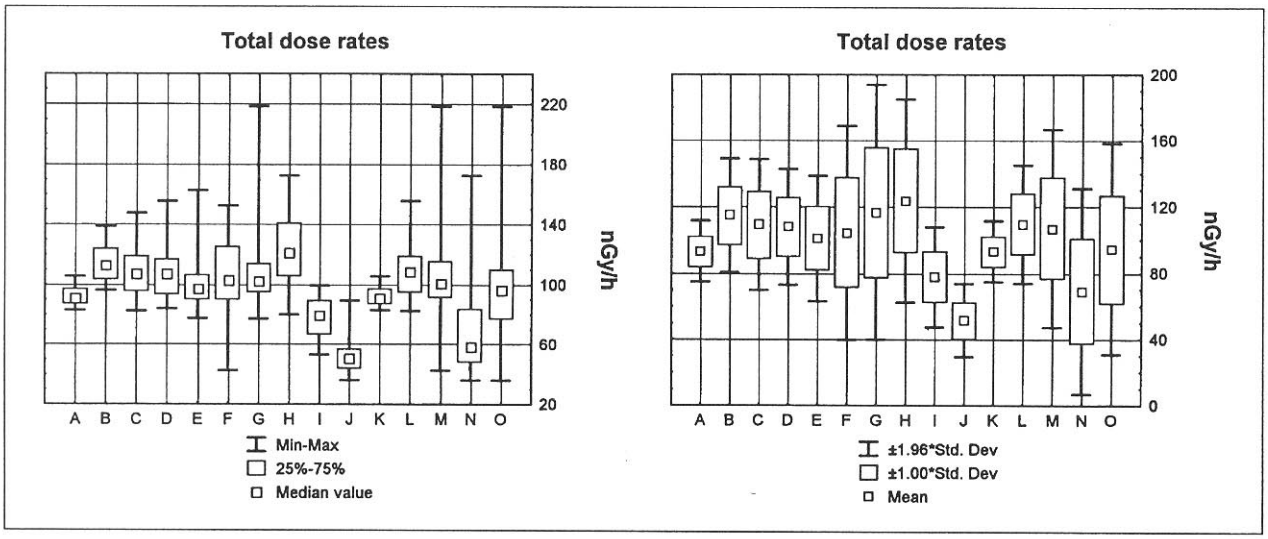

Fig. 2. Total absorbed dose rates (^{137}Cs and natural radionuclides) at one meter above ground according to underlying geological units. Legend: A - Jurassic; B - Berriasian - Barremian; C - Barremian - Aptian; D - Albian; E - Cenomanian; F - Cenomanian - Turonian; G - Turonian - Senonian; H - Palaeocene - Lower Eocene; I - Lower - Middle Eocene; J - Middle - Upper Eocene; K - Jurassic; L - Lower Cretaceous; M - Upper Cretaceous; N - Palaeogene; O - Istria.

background dose rate in air is the result of the primordial radionuclide concentrations in the first 25 cm of the soil (about 55% from the first 7.5 cm in well developed soils). In carbonate terrains, in areas of generally poorly-developed soils, primordial radionuclide concentrations in soils are significantly higher than radionuclide concentrations in underlying carbonate rocks. In such circumstances, soil thickness is the main factor controlling the natural background dose rate. The average activities of ^{40}K, ^{226}Ra, ^{228}Ac and ^{238}U in 37 soil samples developed on Lower Cretaceous carbonate rocks of Istria as well as their average activities in 3 limestone bedrock samples, are used for an example of natural background dose rate assessment. The resulting natural background dose rate in air increases more than 10-fold in the cases of relatively well developed soils (> 25 cm thickness), while in terra rossa a thickness of only 4 cm results in an approximately 5-fold increase in dose rate than in the cases when soil is completely missing. So, the reported dose rates, as a first approximation, are overestimated, especially for the hilly and mountainous areas. In the flysch terrains and areas at lower elevation (valleys, karst poljes, etc.) where soil thicknesses exceed 1 m, the reported values are correct.

The best example for studying the sources and various factors contributing to the absorbed dose rates in air is the Palaeogene beds and associated soils. The Palaeogene bedrock includes Middle and Upper Eocene flysch, Lower Eocene foraminiferal limestones and Liburnian strata of the Palaeocene. This complex comprises the areas with the lowest absorbed dose rates (flysch terrains) as well as the areas with the highest absorbed dose rates (Palaeocene terrains) in the whole of the study area (Fig. 3). The increased absorbed dose rates in air in the vicinity of Raša are probably caused by increased radioactivity in soils associated with (underlying) Palaeocene coal-bearing strata. Poor surface outflow in that area, as in some other karst areas, additionally increases the possibility of the relative accumulation of natural radionuclides in soils vs. soil parent materials.

As is well known, the natural radionuclide concentrations in strata with prevailing clastic rocks, as in flysch, are generally higher than in limestones and dolomites. In soils overlying flysch and carbonate terrains in Istria, the radionuclide distribution pattern is completely the opposite (BARIŠIĆ, 1993). Consequently, this is reflected in the absorbed gamma dose rate in air. Undoubtedly, it is the result of differences in migration due to existing differences in drainage characteristics between the flysch (having a very high coefficient of outflow) and limestone and dolomite terrains where the surface drainage network is poorly developed, or even completely missing. In carbonate terrains, recalculation of natural background dose rates into primordial radionuclide concentrations in the ground is only possible in two cases: when the soil is completely missing and where soils are well developed soil (thickness >25 cm). In the first case, the recalculated value represents radionuclide concentrations in bedrock, while in the second case they represent the concentrations in the soil. Recalculation of dose rates arising from the ^{238}U decay series into ^{238}U concentration in soil or bedrock is incorrect, since the recalculated value presents the ^{226}Ra, but not the ^{238}U concentration. If soil thicknesses range ranging between 0.5-25 cm, recalculated radionuclide concentrations in the ground cannot represent concentrations in the soil or bedrock. Correct correlations with other element concentrations, determined strictly in the soil or bedrock, are also impossible. Consequently, geochemical map construction based on data which are recalculated from aerial gamma-spectrometry surveys in carbonate terrains is questionable.

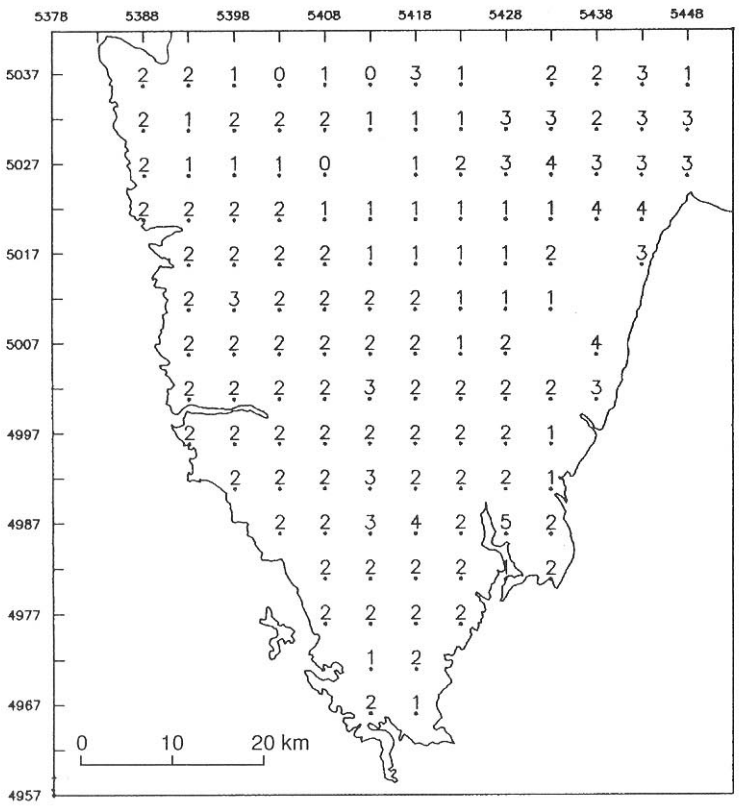

Fig. 3. Total absorbed dose rates in Istria in air 1 m above ground surface. Legend: 0 - < 40 nGyh^{-1}; 1 - 40-80 nGyh^{-1}; 2 - 80-120 nGyh^{-1}; 3 - 120-160 nGyh^{-1}; 4 - 160-200 nGyh^{-1}; 5 - > 200 nGyh^{-1}.

A contribution from various radionuclides to the total absorbed dose-rate in air is given in Fig. 4. ^{40}K contribution is practically the same in all studied geological units. However, the highest differences are found for the radionuclides of the ^{238}U decay series, which was expected.

6. CONCLUSIONS

The natural background dose rate in air above ground surface is the sum of the doses arising from the soil and bedrock. The resulting dose rate in air above the ground surface is the product of characteristic radionuclide specific activity in soil or bedrock with the accompanied DRCF. It is suggested that, for carbonate terrains, various factors should be considered in order to make a radiometric map of good quality. The main problems are: averaging the soil thickness for the entire sampling grid (which the particular sampling point represents); significant differences in radionuclide concentrations between soils and underlying carbonate rocks and the high degree of radiochemical disequilibrium between ^{238}U and ^{226}Ra in soils derived from carbonate rocks.

The laboratory gamma-spectrometry measurement of samples collected during conventional geochemical sampling could provide the data for potassium, thorium, radium and uranium radiometric map construction. Additionally, determination of ^{40}K, ^{226}Ra, ^{228}Ac or ^{208}Tl and ^{238}U specific activity in soil and rock samples provides a characteristic radionuclide data-base of good quality for resulting natural background dose rate assessment. Identical sampling grids and determination of all elements in a single sample provide the correct conditions for element correlation. Possible errors in natural background dose rate assessment, caused by radiochemical disequilibrium occurring in primordial radionuclide decay series or subseries, are insignificant. Correlations between different areas are also possible as the maps of radioactive elements could be constructed under the same conditions.

Recalculation of natural background dose rates into radionuclide concentrations in the ground in carbonate terrains is possible in two cases. If the soil is completely missing, the recalculated value represents the radionuclide concentrations in the bedrock. If the soil thickness is 25 cm or more, the recalculated value represents the radionuclide concentrations in soil relatively well. Recalculation of the dose rate arising from the ^{238}U decay series into ^{238}U concentration is incorrect because recalculated values represent ^{226}Ra activity. Recalculation of radionuclide concentrations in soil or bedrock, based on the aerial gamma-ray surveys in the carbonate terrains, is practically impossible in the cases of poorly-developed soils with thicknesses ranging between 0.5-25 cm.

The airborne gamma survey in carbonate terrains could provide only a very general picture of the radiometric characteristics of these terrains. In our opinion, the radiometric mapping of carbonate terrains should include data from in situ gamma dose-rate measurements together with laboratory gamma ray spectrometry of samples taken at the same locality. Due to the high variability of soil types and thicknesses and litho-

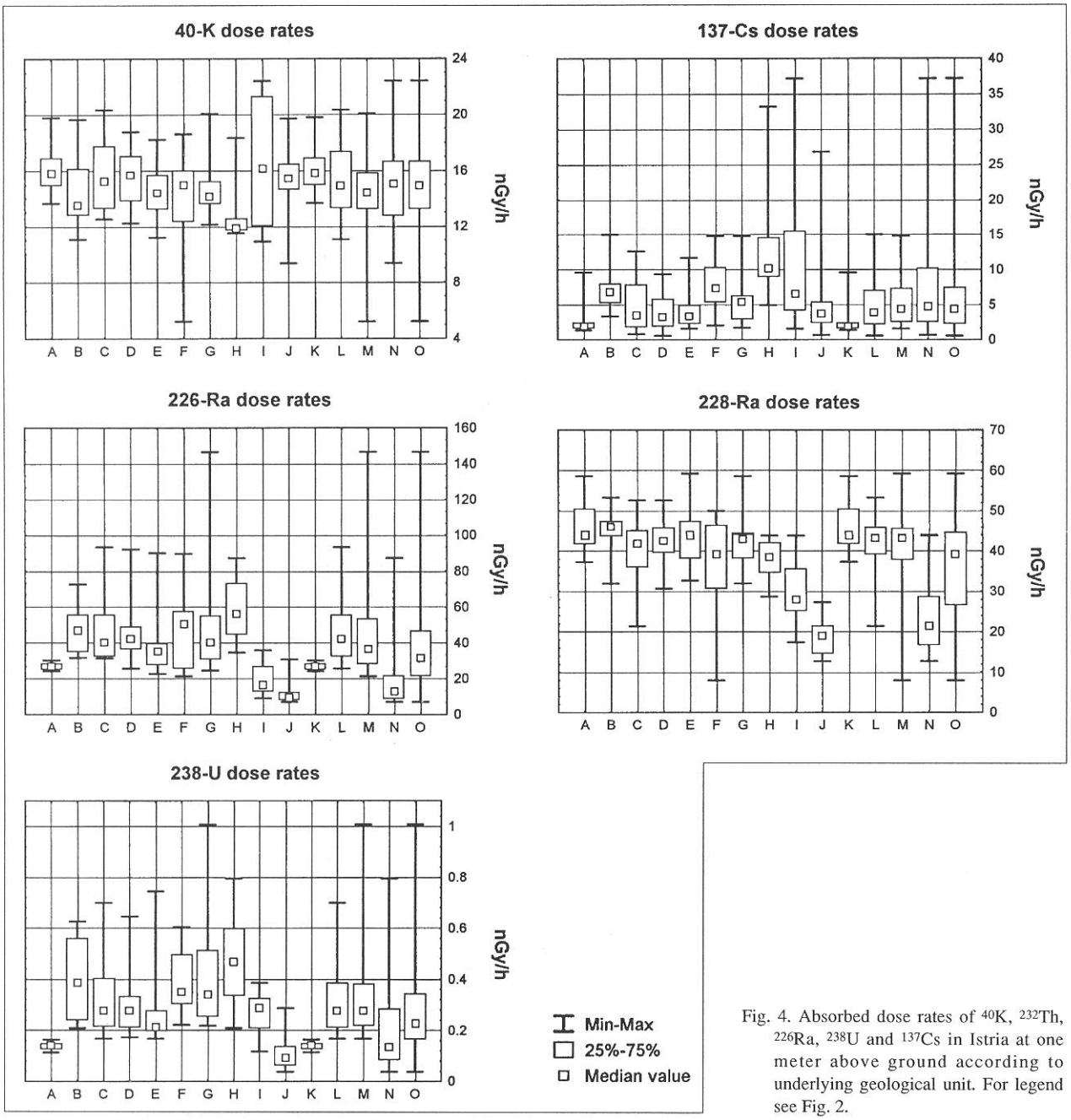

Fig. 4. Absorbed dose rates of ^{40}K, ^{232}Th, ^{226}Ra, ^{238}U and ^{137}Cs in Istria at one meter above ground according to underlying geological unit. For legend see Fig. 2.

logical types of bedrock on a local scale, a much denser sampling grid must be applied.

For the proposed study, as the model area for carbonate terrains, or karst sensu stricto, the area of the Istrian peninsula seems to be highly promising. This paper represents the beginning of a complex study under the project of the Radiometric Map of Croatia.

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