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DETERMINATION OF ACTIVITY CONCENTRATION OF ^{40}K AND GROSS BETA ACTIVITY IN SOIL FROM KAVADARCI AND ITS ENVIRONS

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A b s t r a c t: A survey was carried out to determine the activity concentration and distribution of ^{40}K and gross beta activity in the soil from the city of Kavadarci, Republic of Macedonia, and its environs. A total of 45 surface soil samples were collected from evenly distributed sampling sites over an area of 360 km^2. The activity concentrations of ^{40}K were measured using a high purity germanium (HPGe) gamma-ray detector, while the gross beta activity measurements were made using a low background gas-flow proportional counter. The obtained values for the activity concentrations of ^{40}K were found to be in the range of 286±6 and 801±12 Bq/kg with an average value of 545±118 Bq/kg. The gross beta activities varied between 438±21 and 1052±36 Bq/kg, with an average value of 681±146 Bq/kg. These values allowed the determination of the elemental concentrations of potassium as well as the air absorbed gamma dose rate, which were found to range from 0.92±0.02 to 2.56±0.04% and from 11.9±0.1 to 33.4±0.5 nGy/h, respectively. The mean values of these parameters were 1.74±0.37% and 22.8±4.9 nGy/h. All obtained values fall within the worldwide range as reported in the literature. A strong correlation between the content of potassium in the soils and their geological origin was observed.

Key words: potassium; soil; gamma spectrometry; gross beta activity, activity concentration; absorbed dose rate, lithological units

INTRODUCTION

External exposures outdoors arise from terrestrial radionuclides present at trace levels in all soils and the specific levels are related to the types of rock from which the soils originate. There have been many surveys to determine the background levels of radionuclides in soils, which can in turn be related to the absorbed dose rates in air. The spectrometric measurements indicate that the three components of the external radiation field, namely from the gamma-emitting radionuclides in the ^{238}U and ^{232}Th series and ^{40}K, make approximately equal contribution to the external gamma radiation dose to individuals (UNSCEAR, 2000).

Potassium is an essential element of human metabolism and can be found in all living cells, mainly in the muscular tissue. Natural potassium is composed of three isotopes: ^{39}K, ^{40}K and ^{41}K. Of these naturally occurring potassium isotopes only ^{40}K is unstable, having a half-life of 1.28×10^9 years. It occurs to an extent of 0.012 % in natural potassium, thereby imparting a specific activity of approximately 30 Bq/g potassium. ^{40}K is a beta and gamma emitter (89 % and 11 % of its radiation, respectively) with respective energies of 1.3 and 1.46 MeV (Bowen, 1979).

Because of its relative abundance and its energetic beta emission, ^{40}K is easily the predominant radioactive component in normal foods and human tissues. It is important to recognize that the potassium content of the body is under strict homeostatic control and is not influenced by variations in environmental levels (Eisenbud, 1987).

Radioactivity levels of the environment depend on geological aspects, mainly on rocks and soil, where the radionuclides are found in varying concentrations (Tzortzis and Tsertos, 2004). Representative values of the potassium content of rocks, as summarized by Kohman and Saito (1954) indicate a wide range of values, from 0.1 % for limestones trough 1 % for sandstones and 3.5 % for granite. The potassium content of soils of arable lands is controlled by the use of fertilizers. It is estimated that about 1.11×10^{14} Bq is added annually to the soils of the United States in the form of fertilizer (Guimond, 1978).

The mean natural radionuclide contributions to gross beta activity in soil are ^{40}K and ^{226}Ra beta emitting decay products (Kücükömeroglu et al., 2008).

The aim of the present study is to measure the beta activity levels in the surface soil over the Kavadarci region, to assess the radiation hazard to the population due to ^{40}K and to investigate the connection between the potassium content in the soils and the geology of the terrain. The importance of this work arises from the fact that there is no reference regarding this area concerning the ^{40}K activity concentrations in the soil.

MATERIAL AND METHODS

Study area

The city of Kavadarci is situated in Tikveš valley, about 100 km south of Skopje, the capital of The Republic of Macedonia (Fig. 1). The study area is limited with coordinates (Gauss Krueger zone 7) 7574000 (W) -7592000 (E) and 4582000 (S) – 4602000 (N).

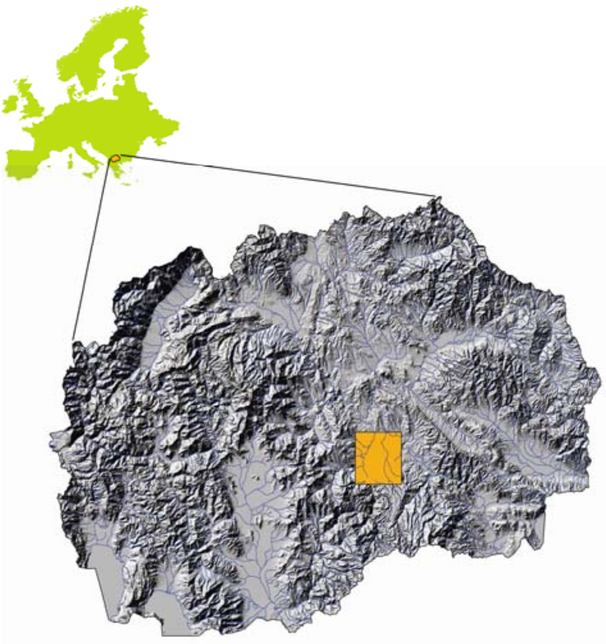

Fig. 1. Location of the study area

The urban area is surrounded by hills from east and south. Of the total 360 km^2 of the study area, rivers and lakes cover 6 km^2, cultivable land 221 km^2, non-cultivable area 120 km^2 and urbanized area 13 km^2 (Fig. 2). The Kavadarci region is the main center of vine production in Macedonia and south-eastern Europe and it is known for its ferro-nickel industrial activity (Stafilov et al., 2008; 2010).

The geological description of the study area (Fig. 3) was made according to Rakićević et al. (1965) and Hristov et al. (1965).

The region of Paleozoic and Mezozoic rocks (Pz-Mz) cover approximately 39 km^2 in the SW and W part of the investigated area and 7 soil samples were taken from this area. The Upper Eocene flisch sediments and yellow sandstones (E-Flis) are developed along Vardar, Crna Reka and Luda Mara valleys, they cover approximately 34 km^2, mainly in the north part of the investigated area, where 4 sampling sites are located. The Pliocene sediments (Pl-sand) fill the Tikveš basin, they cover the biggest part of the study area (about 182 km^2) and from this region 23 soil samples were collected. The Quaternary pyroclastic vulcanites (Q) are found on the south-east from Kavadarci, they are spread over an area of around 25 km^2 and 5 soil samples were taken. Quaternary ages is represented with deluvial (12 km^2), terrace (23 km^2) and aluvial sediments (40 km^2). On this lityhological units 6 sampling sites are located.

Sampling

The soil samples from the city of Kavadarci and its environs were collected for preparing a geochemical atlas of this region and according to the European guidelines for soil pollution studies (Teocharopulos et al., 2001; Šajn, 2004). The investigated region was covered by a sampling grid of 4×4 km^2 (Fig. 4) and a total of 45 soil samples were taken. The samples were collected from the upper 5 cm layer. Each sample represents a composite material collected at the central sample point and four points within the radius of 10 m around it towards N, E, S and W. The mass of such sample was about 1 kg.

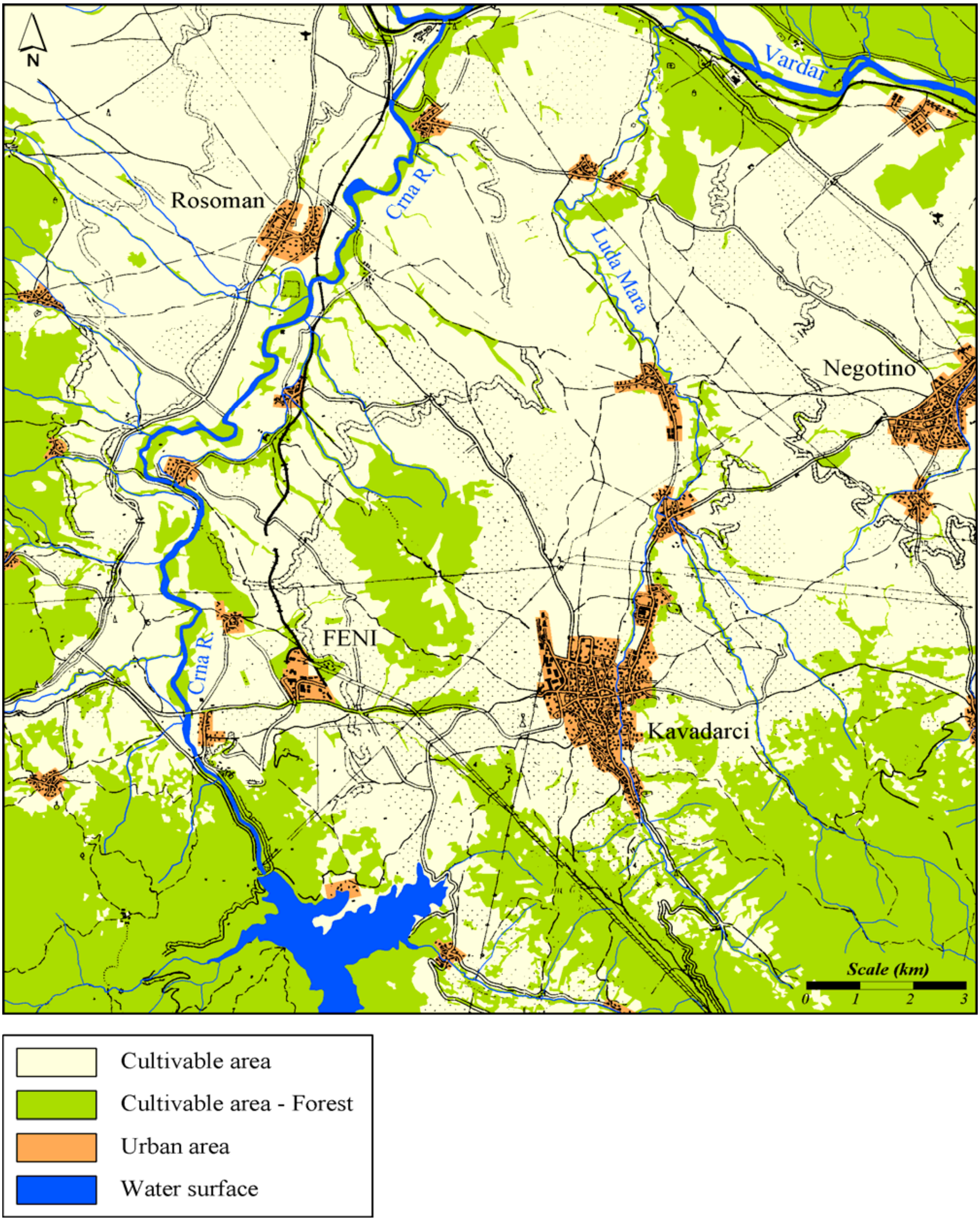

Fig. 2. Land use map of the study area

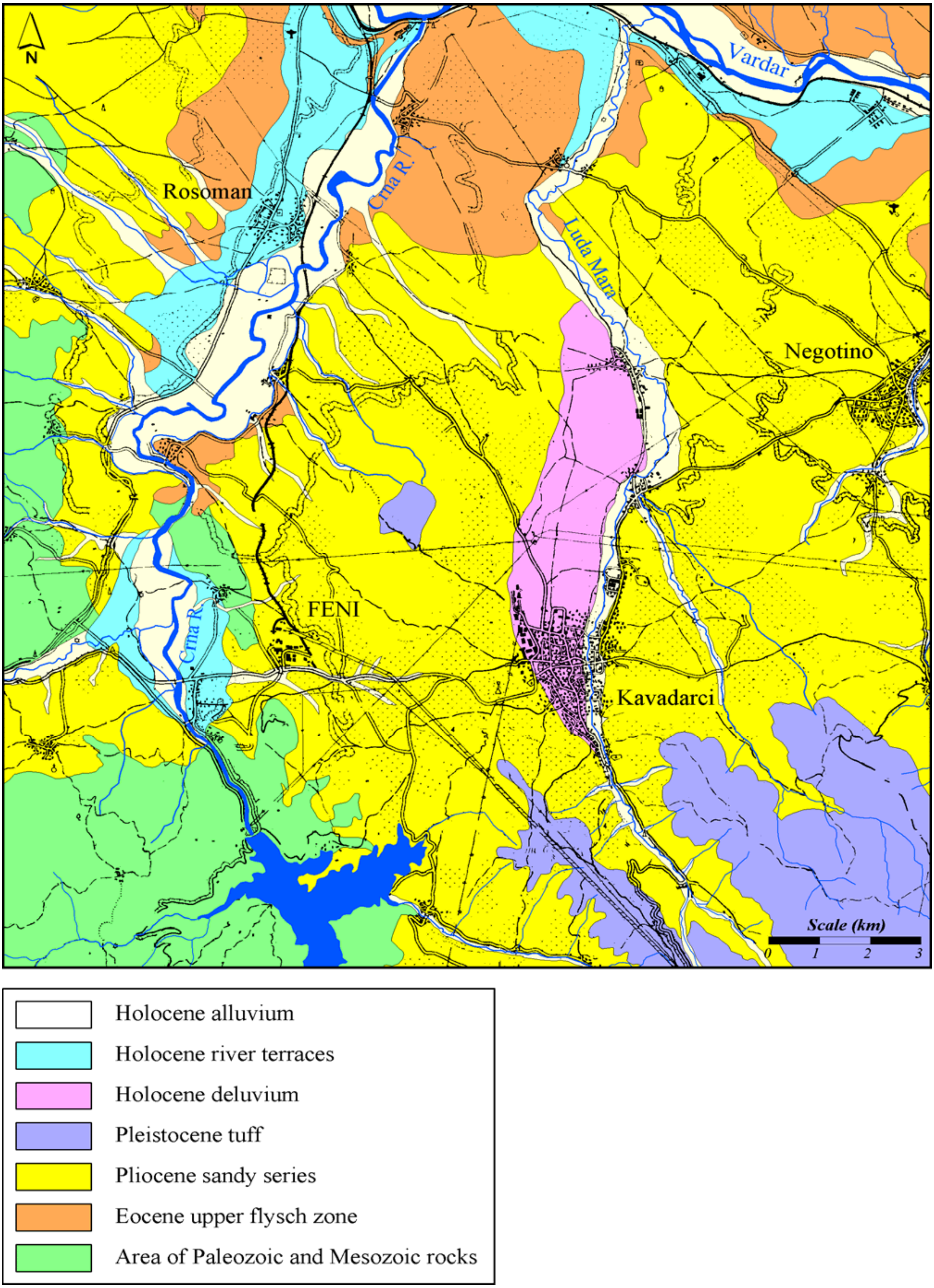

Fig. 3. Lithological map of the study area

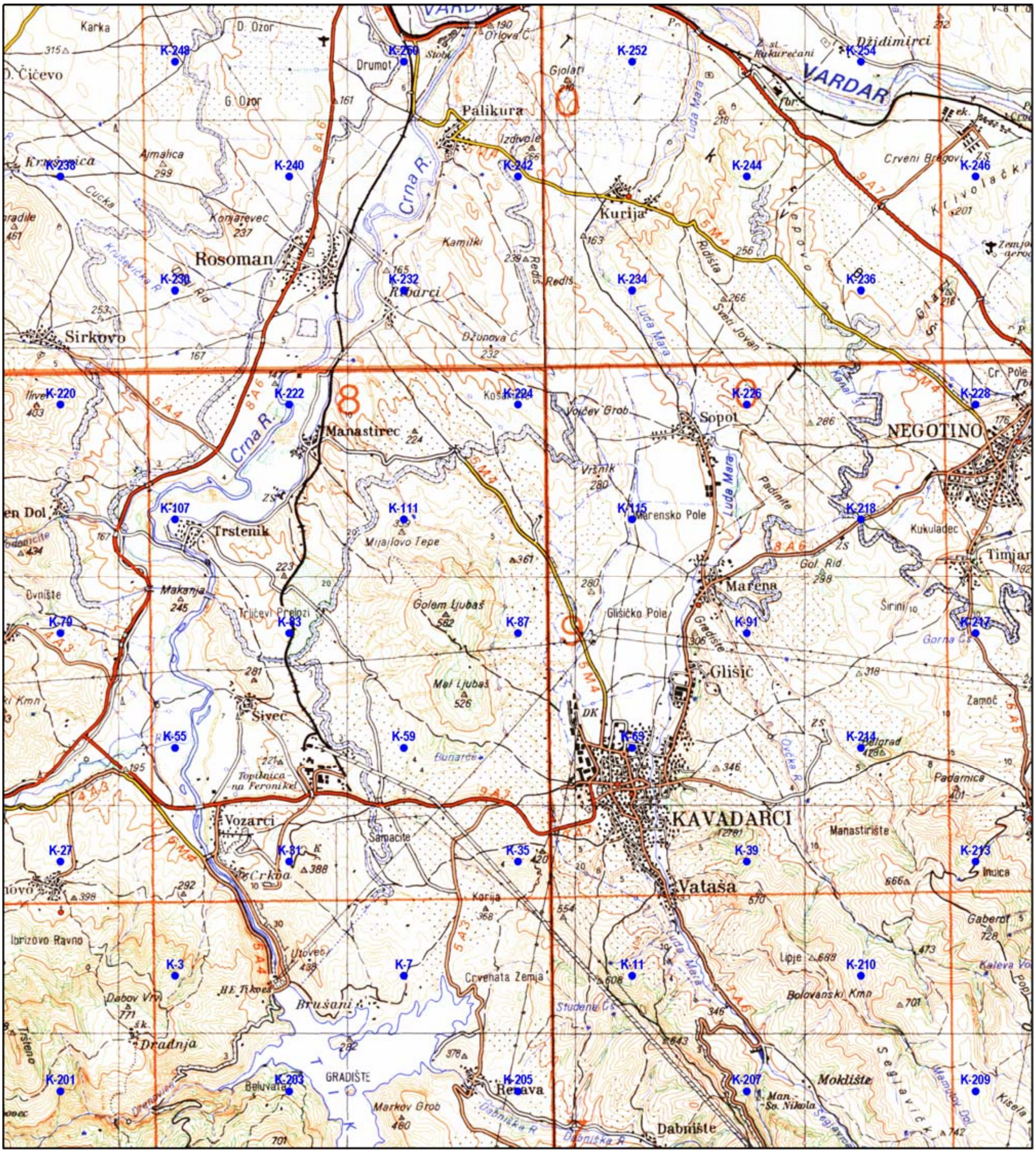

Fig. 4. Soil samples locations in the Kavadarci area

Sample preparation

The samples were mixed thoroughly, collected in plastic bags and labeled properly. In the laboratory, after removing the roots and stones, the samples were air dried for about two weeks, grounded, sieved and homogenized. The fraction below 2 mm was used for the analyses. The data have been expressed on dry weight basis.

About 200 mg of each soil sample were taken for measurement of the gross beta activity, transferred into a 2-in. diameter stainless steel planchete, evenly spread, fixed with acetone and dried under infrared lamp.

For gamma spectrometric analyses of ^{40}K prepared soil samples of about 150 g each were stored in standardized cylindrical plastic containers.

Radiological analyses

The measurement of gross beta activity in all soil samples was performed using MINI 20 Very low background multiple detector counting system for low alpha/beta activities, Eurysis Measures. The detectors used were gas flow proportional counters. The system was calibrated with prepared standard samples which contain known concentration of ^{40}K. The counting time was 60 minutes, by four independent detectors in the system, simultaneously. Each sample was counted for three times in a row and the results were given as an arithmetic mean with the statistical error.

For the measurement of activity concentrations of ^{40}K in the soil samples a high resolution gamma-ray spectrometer consisting of a coaxial P-type HPGe detector with a relative efficiency of 27.1 % was used. The detector was coupled to a Canberra multi-channel analyzer (MCA). The resolution was 2 keV at 1.33 MeV of ^{60}Co. The detector was shielded in an 8 cm lead chamber with an inner lining of 2 cm thick copper plate to reduce the background. The results were analyzed by Genie-2000 software (Canberra). A reference sample (Soil-375) provided by the International Atomic Energy Agency was used for the efficiency calibration of the system. Each sample was counted for 60 000 s.

The activity concentration of ^{40}K was determined using its single 1460.8 keV gamma-ray line and converted to total elemental concentration of potassium, reported in %, using the following equation (Tzortzis and Tsertos, 2004):

$$F_E = \frac{M_E \cdot C \cdot A_E}{\lambda_E \cdot N_A \cdot f_{A,E}}$$

where F_E is the fraction of element E in the sample, M_E, λ_E, $f_{A,E}$ and A_E are the atomic mass (kg/mol), the radioacivity decay constant (s^{-1}), the fraction atomic abundance in nature and measured activity concentration (Bq/kg) of the corresponding radionuclide, respectively, N_A is Avogadro's (6.023×10^{23} atoms/mol) and C is a constant with a value of 100 for potassium.

The absorbed gamma dose rate in air 1 m above the ground (D_K), proceeding from the gamma emissions of ^{40}K, in nGy/h, was calculated on the basis of guidelines provided by UNSCEAR, 2000:

$$D_K = 0.0417 \cdot A_K$$

where A_K is the activity concentration of ^{40}K (Bq/kg).

RESULTS AND DISCUSSION

The obtained results for the minimum, maximum and average values of gross beta activity and the activity contrations of ^{40}K in Bq/kg of dry soil, the calculated content of potassium (in %) and the absorbed gamma dose rate (in nGy/h) are presented in Table 1.

Table 1

The minimum, maximum and average values of gross beta activity, activity concentration of ^{40}K, elemental concentration of potassium and the absorbed gamma dose rate

	Minimum	Maximum	Average
Gross beta activity (Bq/kg)	438±21	1052±36	681±146
^{40}K (Bq/kg)	286±6	801±12	545±118
K (%)	0.92±0.02	2.56±0.04	1.74±0.37
D_K (nGy/h)	11.9±0.1	33.4±0.5	22.8±4.9

The gross beta activity of the soils varied between 438±21 and 1052±36 Bq/kg, with an average of 681±146 Bq/kg. The map of the distribution of the gross beta activity in the studied area is shown in Fig. 5.

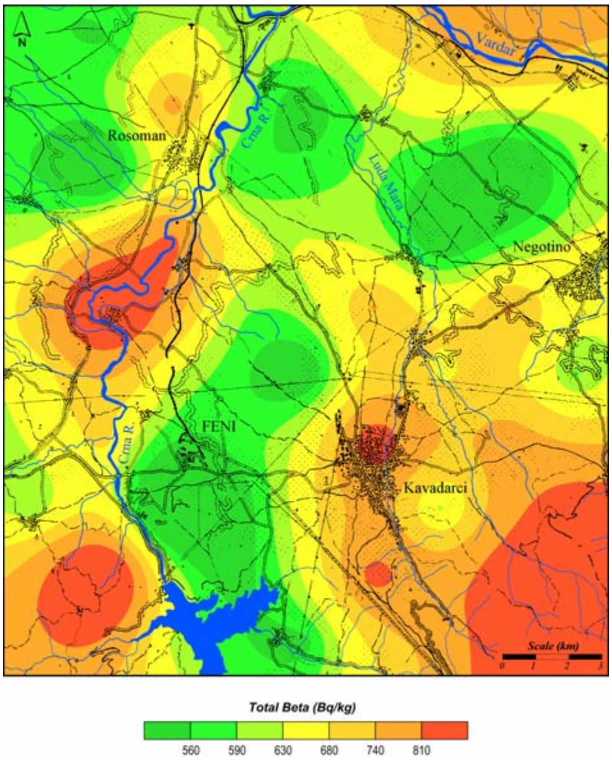

Fig. 5. Map of the distribution of gross beta activity

The activity concentration of potassium in the soil samples was found to be in the range from 286±6 to 801±12 Bq/kg, with an average of 545±118 Bq/kg. The elemental concentration of potassium ranged from 0.92±0.02 to 2.56±0.04%, with an average of 1.73±0.37%. The distribution of potassium in the studied area is shown in Fig. 6.

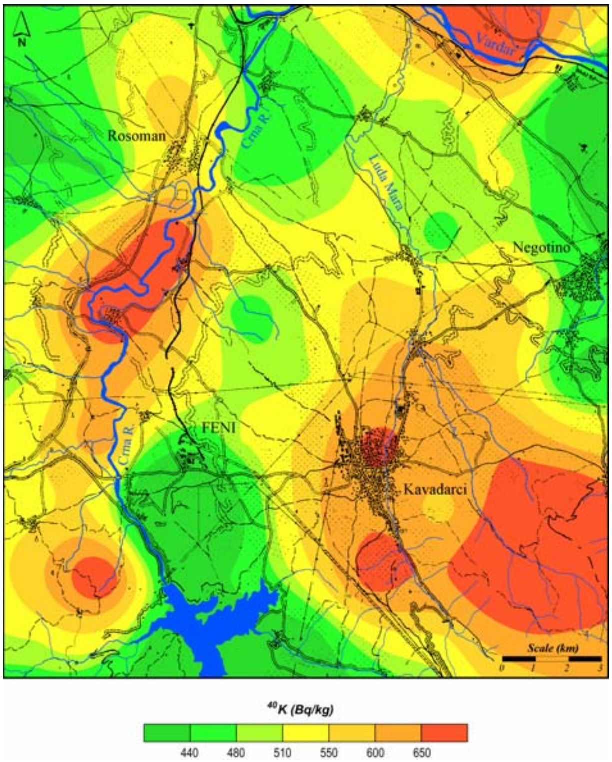

Fig. 6. Map of the distribution of ^{40}K

The measured activity concentration of ^{40}K were compared with the values reported worldwide as shown in Table 2.

Table 2

Activity concentrations of ^{40}K (Bq/kg) measured worldwide

Region	References	^{40}K
Pernambuco, Brazil	Santos Jr., et al. 2005	1827
Thanjavur, India	Senthilkumar et al., 2010	149.5±3.1
Riyadh, Saudi Arabia	Alaamer, 2008	225±63
Ontario, Canada	Vanden Bygaart et al., 1999	461.5±168.4
Marmara Region, Turkey	Kilic et al., 2007	442.5±189.9
West Bank, Palestina	Dabayneh et al., 2008	630
Ptolemais, Greece	Psichoudaki and Papaefthymiou, 2008	496±56
Punjab, Pakistan	Tahir et al., 2005	307±101
Vojvodina, Serbia	Bikit et al., 2005	554±92
Nigeria	Ajayi, 2009	286.5±308.5
Turkey	Kücükömeroglu et al., 2008	51-1605
Veles region, Macedonia	Dimovska et al., 2010	585.7 ± 86.4
Worldwide average	UNSCEAR, 2000	400 (140-850)
This work		545±118

The calculated air absorbed gamma dose rate values varied from 11.9±0.1 to 33.4±0.5 nGy/h, with a mean of 22.8±4.9 nGy/h.

The statistical data for gross beta activity and the activity concentration of ^{40}K, according to the basic lithological units, are given in Table 3.

Table 3

The minimum, maximum and average values of gross beta activity and activity concentration of ^{40}K according to the basic lithological units

	Lithological unit	Minimum	Maximum	Average
Gross beta activity (Bq/kg)	Pz-Mz	438±21	1052±36	681±146
	E-Flis	494±25	688±28	591±91
	Pl-sand	490±27	868±33	625±91
	Q-Tuf	744±29	1030±34	880±102
	Q-Al	612±26	1012±33	790±136
^{40}K (Bq/kg)	Pz-Mz	286±6	756±11	481±144
	E-Flis	374±6	541±7	450±69
	Pl-sand	400±6	754±11	523±77
	Q-Tuf	588±8	780±11	677±73
	Q-Al	469±7	801±12	662±121

The highest average values for the activity concentration of ^{40}K and gross beta activity (677±73 Bq/kg and 880±102 Bq/kg, respectively) are found in the regions of Pleistocene tuff (Q-Tuf), whereas the lowest average values (450±69 Bq/kg and 591±91 Bq/kg, respectively) occur in the areas of the Eocene upper flysch zone (E-Flis) (Figs. 7 and 8).

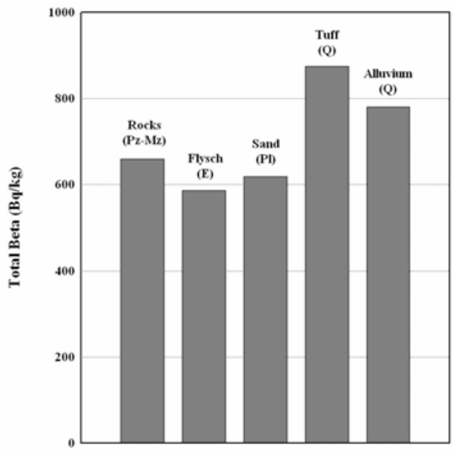

Fig. 7. Average gross beta activity according to the basic lithological units

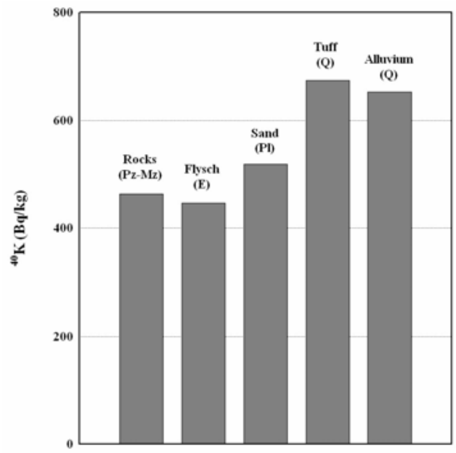

Fig. 8. Average activity concentration of ^{40}K according to the basic lithological units

CONCLUSION

The calculated average value for the gross beta activity in the analyzed soil samples is 681±146 Bq/kg. The common values range from several hundreds to 1000 Bq/kg (ISO 18589-3, 2007). The gross beta activity in environmental samples derives mainly from the presence of ^{40}K and also the other natural beta emitting radionuclides. The high obtained value for the linear coefficient of correlation *r* (0.82) between the gross beta activity and the activity of ^{40}K is in an agreement with the literature data.

The average activity concentration of ^{40}K measured in the soil samples from the region of Kavadarci and its environs (545±118 Bq/kg) is comparable with the values for the activity concentration of ^{40}K in the soils from other region in Macedonia and the neighboring countries (Serbia, Greece). It is slightly higher than the worldwide average, but still in the range of UNSCEAR 2000 report, which is 140-850 Bq/kg.

The highest average values for the investtigated parameters are found in the regions of Pleistocene tuff and Holocene alluvium of the rivers Vardar, Crna Reka and Luda Mara, whereas the lowest average values occur in the areas of the Eocene upper flysch zone, which proves the relation between the content of potassium in soils and their geological origin.

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Р е з и м е

ОПРЕДЕЛУВАЊЕ НА АКТИВНОСТА НА ^{40}K И ВКУПНАТА БЕТА АКТИВНОСТ ВО ПОЧВАТА ОД КАВАДАРЦИ И НЕГОВАТА ОКОЛИНА

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Клучни зборови: калиум; почва; гама спектрометрија; вкупна бета активност, специфична активност, брзина на гама доза, литолошки единици.

Испитување беше извршено со цел да се определи активноста и дистрибуцијата на ^{40}K и вкупната бета активност во почвата од Кавадарци, Република Македонија, и неговата околина. Земени се вкупно 45 примероци од површински почви од рамномерно распоредени локации, на површина од 360 km^2. Специфичните активности на ^{40}K беа мерени со помош на П-тип коаксијален гама детектор од германиум со висока чистота, додека мерењата на

вкупната бета активност беа извршени со користење на нискофонов гасно-проточен пропорционален бројач. Добиените вредности за специфичната активност на ^{40}K се движат од 286±6 до 801±12 Bq/kg, со средна вредност од 545±118 Bq/kg. Вкупната бета активност варира помеѓу 438±21 и 1030±36 Bq/kg, со средна вредност од 681±146 Bq/kg. Овие податоци овозможија да се пресмета концентрацијата на калиум, како и брзината на гама дозата на зрачење, кои изнесуваа од 0,92±0,02 до 2,56±0,04 % и од 11,9±0,1 до 33,4±0,5 nGy/h, соодветно. Средните вредности на овие параметри беа 1,74±0,37 % и 22,8±4,9 nGy/h. Сите добиени вредности се споредливи со просечните во светски рамки, објавени во литературата. Резултатите од анализата укажуваат на силна поврзаност помеѓу застапеноста на калиумот во почвите и нивното геолошко потекло.