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Radionuclide mapping of the Molise region (Central Italy) via gamma-ray spectrometry of soil samples: relationship with geological and pedological parameters

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Abstract Radionuclide maps of the Molise region were created for the first time using gamma-ray spectrometry analysis of 205 soil samples. The geographical distributions of ^{40}K, ^{232}Th and ^{238}U were within the world average values for soils. ^{40}K was distributed homogeneously with a slight enhancement along the coastline. The decay chains of ^{238}U and ^{232}Th were in secular equilibrium with their daughters, also showing a homogeneous distribution except for localized areas of enhanced concentrations close to the borders with the Lazio and the Campania regions. Concentrations of all three radionuclides were correlated with geological and pedological characteristics of soils. The measured external gamma-dose rate in the air due to naturally occurring radionuclides in the soil, and the dose rate due to cosmic rays were in agreement with values measured in other Mediterranean regions. Increased ^{137}Cs levels from atmospheric nuclear weapons tests and the Chernobyl fallout were found at sites above 1,000 m a.s.l.

Keywords Gamma-ray spectrometry · Natural radioactivity · Soil · Radiation exposure

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Introduction

Environmental radioactivity studies of soil have been carried out in many countries with the aim to develop radiation maps of regions important either for mineral exploration, geophysical and geological–investigations, or for terrestrial radioactivity assessment and its impact on population (e.g. [1–6]). Recently, the European Radiological Data Exchange Platform (EURDEP) has provided nonvalidated radiological monitoring data from most European countries available in nearly real-time [7]. The participation of EU member states is regulated by the Council Decision 87/600, and the Recommendation 2,000/473/EURATOM.

In Italy, the soil radioactivity data (usually obtained by in situ and/or laboratory gamma-ray spectrometry), have been collected in rather limited areas, e.g. at the central sector of the Southern Alpine domain [8], at the Alps-Apennine transition region [9], and at the Eolian Islands (Southern Italy) [10]. Larger areas were investigated more recently, e.g. the regions of Piemonte [11], and Abruzzo [12, 13].

In this paper we present results of a survey carried out in the Molise region (Central Italy), which represents the smallest of the Italian regions. The aim of this study has been to assess contents of natural radionuclides in soil, and to investigate their relationship with geological and pedological characteristics of the region. The project was carried out in collaboration with the National Civil Protection of Molise, as a part of the ERMES (Radioactivity Monitoring for Earth Sciences) program of the Instituto Nazionale di Fisica Nucleare (INFN). Several environmental radioactivity studies, carried out in the framework of the ERMES project in the underground laboratories of the Laboratori Nazionali del Gran Sasso (LNGS), have already been published [14–21].

Geology and pedology of the Molise region

Geology background

The Molise region has a rather complex geological structure [22]. The present configuration is a result of a continuous palaeo-geographic evolution accompanied by tectonic events that mainly occurred in the parossistic phase of the orogenesis of the Apennines in the Mio-Pleistocene. The territory is made exclusively by sedimentary formations, a large part of them are of marine origin. More recent formations of continental origin are overlying the old formations. The older marine formations belong to five lithostratigraphic units, which were formed in the various geological periods:

(i) Lazio-Abruzzi and Campania-Lucania units, carbonatic platform of carbonate and undifferentiated calcareous-dolomitic sedimentation (Cretacous and Late Triassic) (area of Matese-Mainarde);
(ii) Sannio-Molise unit, characterized by flysch and marl, with calcareous sedimentation (Late Cretaceous-Early Miocene), containing debris brought from the Lazio-Abruzzi and Campania-Lucania units (areas Monti Venafro-Isernia, Frosolone, Sepino);
(iii) Top-thrust basin Molise, an open environment of sea sedimentation, relatively deep, with conglomerates and claystones on the basis and arenites on the summits (valleys of Trigno and Biferno, up to the Monti Frentani, early Pliocene and Messinian);
(iv) Periadriatic foredeep, a profound depression, parallel to the actual coastline, created during the Pleistocene; it is characterized by subsidence phenomena, together with argillic-sandy sedimentation of the Plio-Pleistocene;
(v) Apulia Foreland calcarenites in the territory of the Regione Molise (which are not outcropping), as covered by the Plio-Pleistocenic sediments of the foredeep.

Pedology background

The Molise region is covered mainly by Dystric Cambisol, and in the mountainous partly also by Leptosol [23]. Calcari-vertic Cambsiol and Calcaric Regosol can also be found in some very localized areas. Along the coastline the situation is far more variable, with presence of Chromic, Calcaric, Eutri-fluvic and Vertic Cambisols, and to a somewhat lesser extent Chromic Vertisol can also be found.

The Cambisol represents a young soil. The pedogenic processes are evident from color development and/or structure formations below the surface horizon. The Cambisol occurs in a wide variety of environments around the world, and under many kinds of vegetation. They are commonly referred to as brown soil. The Leptosol represents a shallow layer over hard rock, and comprises a very gravelly or highly calcareous material. It is found mainly in mountainous regions and in areas where the soil has been eroded to the extent that hard rock comes near to the surface. Because of limited pedogenic development, Leptosols do not have much structure. On a global scale, Leptosols are very extensive. The Regosol is a very weakly developed mineral soil in unconsolidated materials with only a limited surface horizon. Limiting factors for soil development range from low soil temperatures, prolonged dryness, characteristics of the parent material or erosion. The Regosols form a taxonomic rest group containing all soil types that cannot be accommodated in any of the other groups. The Regosols are extensive in eroding lands, in particular, in arid and semi-arid areas and in mountainous regions. Internationally, the Regosols are similar to the Entisols. The Vertisols are rich in swelling clay minerals and occur primarily in landscapes under climates with pronounced dry and wet seasons. The Vertisols shrink and swell upon drying and wetting. Deep wide cracks are formed when the soil dries out, and swelling in the wet season. It creates polished and grooved ped surfaces (slickensides), or wedge-shaped or parallel-sided aggregates in the subsurface vertic horizon. The landscapes of the Vertisol may have a complex micro-topography of micro-knolls and micro-basins.

Materials and methods

Sample collection

Molise is moderately populated (72.11 inhabitants km^{-2}) with 55.3 % living in a mountainous territory, up to 2,241 m a.s.l. The remaining 44.7 % of the country is considered hilly or flat. A grid was applied over the whole Molise region (total area of 4,438 km^2) with regular intervals and a resolution of 0.05° in latitude and longitude (approximately squares of 5 × 5 km). It was a "blind" grid, in order not to bias the survey by any a priori consideration. In each point of the grid (205 in total), a soil sample was collected by spade. The samples were not collected on cultivated land. Moreover samples were not collected down to the same depth, as in some cases the soil layer was rather thin above the underlying rock. The maximum depth was 20 cm below the soil surface.

Laboratory analyses and data evaluation

Gamma-ray spectrometry

The radioactive elements of major interest assessed in this survey were ^{238}U and ^{232}Th (analyzed via their daughters), and ^{40}K. The ^{137}Cs from global fallout and from the Chernobyl accident was also found to be present in the soil samples. Massic activities of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs in 205 soil samples were obtained using gamma-ray spectrometry in the STELLA (SubTerranean Low-Level Assay) facility of the INFN/LNGS. Four ORTEC®-supplied p-type coaxial HPGe detectors were used in analyses, which main characteristics have already been described [24]. The measurement time varied between half an hour and 2.5 days, chosen that at least 100 net counts were in the full energy peak (FEP) of the gamma-line at the highest energy (2,615 keV). The longer measurement times of 1–2.5 days were due to the fact that some samples were measured over night or over the weekend.

The samples were not treated mechanically (only organic material and stones of size >1 cm^3 were removed from the samples), therefore we conducted a series of tests when results of untreated and treated samples (prepared by drying and homogenization) were compared. The difference in the results before and after the treatment was less than 5 %. This is due to the fact that our analysis heavily relies on the calculation of the FEP efficiencies with computer simulation taking into account the apparent density of the sample and possible sample inhomogeneities. Each sample was weighed with a precision of better than 0.7 %. After the measurements aliquots of each sample were dried for 24 h at 110 °C in order to obtain the dry mass, to which the results were normalized. All samples were measured in plastic Marinelli containers with 1,500 cm^3 volume. The average dry sample mass was between 200 and 400 g for 90 % of the samples, 7 % of the samples had mass between 120 and 200 g, and 3 % between 400 and 550 g.

The data acquisition system used a commercial ORTEC® MCA computerized system (ADC and GammaVision® software). The FEP efficiency was calculated using a Monte Carlo simulation program named MaGe [25] based on the GEANT4 software package [26, 27]. The program has recently been validated through international proficiency tests [28]. Simulations were carried out in order to account for the different filling heights and apparent densities in the Marinelli sample containers.

It is also worth mentioning that due to specific conditions of the STELLA facility at the LNGS [24], the HPGe detectors were operating in ultra low background configurations with sophisticated shielding of the detectors. The intrinsic background of the HPGe detectors has been therefore by about 1,000 times lower with respect to the sample activity [29]. This implies that the data are much more robust as they are not subject to any fluctuation in the detector background.

Analysis of gamma-ray spectra

The following radionuclides were measured (all nuclear data referring to gamma-energies were taken from Nucléide [30]:

(i) *^{232}Th-series*. The gamma-ray emitting radionuclides ^{228}Ac, ^{212}Pb, ^{212}Bi and ^{208}Tl were analyzed in soil samples. These radionuclides also gave information whether secular equilibrium was present (or not) within the decay chain for ^{228}Ra (^{228}Ac), and ^{228}Th (^{212}Pb, ^{212}Bi and ^{208}Tl). Within the uncertainty of the measurement, all radionuclides were found to be in equilibrium. The average value for the activity ratio ^{228}Ra/^{228}Th was (0.99 ± 0.08). The activity concentrations of ^{228}Ra and ^{228}Th were therefore combined to give the ^{232}Th value, denoted as Th_{eq}.

(ii) *^{238}U-series*. The gamma-ray emitting radionuclides ^{234}Th, ^{234m}Pa, ^{226}Ra, ^{214}Pb and ^{214}Bi were used for analysis. Also in this case it was possible to check if radionuclides are in secular equilibrium (between the upper part of the decay chain given by ^{238}U (^{234}Th, ^{234m}Pa) and the bottom given by ^{226}Ra (^{226}Ra, ^{214}Pb and ^{214}Bi). The average value for the activity ratio of ^{226}Ra/^{234m}Pa was (0.7 ± 0.3). No significant effect due to a loss of ^{222}Rn or due to any equilibrium effect of this gas with its daughter nuclides was found within measurement uncertainties. The average value for the activity ratio (^{226}Ra/^{214}Pb + ^{214}Bi) was (0.9 ± 0.3). Due to short measurement times, the ^{238}U-part (^{234}Th, ^{234m}Pa) resulted often in upper detection limits only. The gamma-rays of ^{234}Th are below 100 keV, where the p-type HPGe detectors have a very low efficiency due to the thick Cu window of the end-cap, and a relatively thick dead layer (about 1.5 mm). On the other hand ^{234m}Pa has the most intense gamma-rays at 1,001 keV, but with a very low emission probability (0.837 %). Therefore, only ^{226}Ra data were used further for representing the ^{238}U-series, referred as U_{eq} and assuming secular equilibrium in the decay chain.

(iii) *Potassium*. The concentration of this element was determined by measuring its primordial radioactive isotope ^{40}K.

(iv) *Total radiation dose rate in the air.* The total dose rate in air, D_{tot}, due to the naturally occurring elements (D_{nat}) and the cosmic rays (D_{CR}) has been determined according to the following relations based on the UNSCEAR report [31]. For the dose rate due to the gamma-radiation from natural radioactive elements in the ground

$$D_{nat}\left[\mathrm{nGy\ h^{-1}}\right] = 0.460\ U_{eq}\left[\mathrm{Bq\ kg^{-1}}\right] + 0.606\ Th_{eq}\left[\mathrm{Bq\ kg^{-1}}\right] + 0.04224\ K\left[\mathrm{Bq\ kg^{-1}}\right],$$

and for the dose rate due to the cosmic radiation as function of the altitude h (in km) where the samples were collected

$$D_{CR}\left[\mathrm{nGy\ h^{-1}}\right] = 27.0\left(0.21\ e^{-1.649h} + 0.79\ e^{0.4528h}\right).$$

No contributions from anthropogenic radionuclides (except ^{137}Cs) were observed.

In each spectrum for each radionuclide of interest, the most abundant photoelectric peaks (with emission probability larger than 5 %) were analyzed. The net-peak area was calculated subtracting the background continuum on the left and the right side of the photoelectric peak. The contribution of the intrinsic background was subtracted, although in most cases it was found to be negligible. In the case of multiplets, a deconvolution was carried out, and if more than one gamma-line was present, the activity was determined combining the different contributions by a weighted mean.

Uncertainties of analyses

The pure statistical uncertainty in each gamma-line was in most cases less than 10 %, apart from the special cases of ^{234}Th, ^{234m}Pa and ^{235}U where either interfering gamma-lines were present, or where the counting statistics was low due to the small efficiency or branching ratio. In a few cases, the same sample was measured twice, with a time interval of a few weeks, without finding any significant difference between the first and the second result.

The FEP efficiency has an uncertainty of 10 % due to the fact that the Monte Carlo simulation of the density was not exactly the one of the sample, affecting a contribution of inhomogeneous matter distribution on the determination of the radionuclide concentration. The uncertainty of the calculated massic activity was a combined standard uncertainty [32] with a coverage factor k equal to one, corresponding to about 68 % confidence level. The uncertainty budget for the activity detected in the soil samples showing the relative contribution of each component to the total uncertainty is given in Table 1. Regular analysis of reference materials [33, 34] helped to attain good quality of results.

Results and discussion

Table 2 lists the massic activities of the detected radionuclides in 205 soil samples, as well as the total dose rate in the air, giving the range and geometric mean values. The frequency distribution of the measured massic activities of all analyzed radionuclides followed a log-normal distribution. The goodness of the fit for the given distribution was tested by the Chi-squared test. The geometric mean values and their geometric standard deviations were calculated accordingly.

The geometric mean of the massic activity of ^{40}K was 468 Bq kg^{-1}, somewhat higher than the median value for soil in the world of 400 Bq kg^{-1} [31]. Over all, potassium is distributed rather homogeneously (Fig. 1).

The U-Ra massic activities had a geometric mean value of 31 Bq kg^{-1}, which is equal to the worldwide average abundance in soil (32 Bq kg^{-1} [31]). The highest values were found at the south-western border towards the region Campania (Fig. 1).

The Th_{eq} massic activities had a geometric mean of 41 Bq kg^{-1}, which is a little higher than the world's average abundance in soil of 30 Bq kg^{-1} [31]. The highest values (seven times higher than the geometric mean value) were found in a small region at the south-western border to the regions Lazio and Campania (Fig. 1).

The external gamma-dose rate in the air (Fig. 1) due to naturally occurring radionuclides has a geometric mean value of 62 nGy h^{-1}, slightly lower than the average value for Italy (74 nGy h^{-1}). The calculated value of the dose rate due to cosmic rays shows a mean value of 36 nGy h^{-1}. The geometric mean of the total gamma-dose rate in the Molise region is 99 nGy h^{-1}. The highest values (215 nGy h^{-1}) can be found in the south-western part of the Molise region, where the uranium and thorium levels and the average altitude are high, as well as in the northern part of the Molise region, where the highest mountains are situated (142 nGy h^{-1}). Compared with previously published data for the Mediterranean part of Spain [35], Greece [36], and Turkey [37] and references therein, the values observed for the Molise region are in good agreement. Compared to the countries in the northern Europe [7], the Molise values are somewhat higher due to different geological and pedological conditions. The same holds for the central Europe, e.g. for Slovakia [38], and for Slovenia [39]. The geometric mean of the Molise region is well within the world average values [31], which indicates clearly that there is no particular radiological impact on the population.

Cesium-137 levels with large variability were found in almost all soil samples. The highest values resulted from samples collected at the northern border towards the Abruzzo region. The ^{137}Cs origin was certainly from global fallout of atmospheric nuclear weapons tests (mostly carried

Table 1 Uncertainty budget for the activity detected in soil samples showing the relative contribution of each component to the total uncertainty

Source of uncertainty	Maximal relative contribution to the uncertainty (%)
Counting statistics	46
Efficiency (Monte Carlo simulation)	46
Source-detector geometry	4.6
Mass determination	3.2
Intrinsic background	0.2
Total	**100**

Table 2 Summary of results of analysis of 205 soil samples from Molise region showing the range and geometric mean

Parameter	U_{eq} (Bq kg^{-1})	Th_{eq} (Bq kg^{-1})	^{40}K (Bq kg^{-1})	D_{tot} (nGy h^{-1})
Minimum	12.4	9.3	93.0	57
Maximum	98.8	220	867	215
Geometric mean	31	41	468	99
Geometric standard deviation	1.5	1.6	1.4	1.2

out in late 50's and early 60's [40], and the Chernobyl accident, e.g. [41]. Moreover, erosion effects could have impact on the re-distribution of the formerly deposited ^{137}Cs on the soil.

In order to explain the special features in the distribution of uranium, thorium and potassium, a correlation between the geological characteristics and the radiometric results has been carried out. The areas with the highest Th_{eq} massic activities observed in the south-western border of Molise correlated with the calcareous sediments and flysch/marl deposits in the Sannio-Molise unit close to Isernia. The observed highest U massic activities were connected with the carbonatic and calcareous-dolomitic formations of the Lazio-Abruzzi and Campania-Lucania units (Le Mainarde in the south-west and Monti del Matese in the south).

Pedologically the highest Th_{eq} massic activities coincide with the presence of the Calcaric Regosol, whereas the Dystric Cambisol mainly covers the rest. The areas with high U levels are also present at the mountainous areas covered with the Luvisols.

The distribution of ^{40}K is rather homogeneous over the whole territory. The "hot spots" with activities close to 900 Bq kg^{-1} cannot be correlated clearly at the present stage of the survey with any geological formation. One possible explanation could be the presence of micaceous material, which is rich in potassium. The only part, which could be related to a geological formation is the coastal area (pre-adriatic foredeep), where the ^{40}K levels are consistently higher in areas mostly formed by sand, claystones and conglomerates. In the coastal areas with the highest ^{40}K levels we also find the Vertic Cambisol and Chromic Vertisol, which are both rich in clay minerals. Similar observations and conclusions with respect to the correlation of natural radionuclide levels to geological and soil formations have been drawn in the past on both local scales (Piemonte [11], Abruzzo [12, 13], and in Turkey [37]), as well as on larger scales (in USA [1], Slovakia [35], and Slovenia [38]).

The observed variability in ^{137}Cs contamination of soil is related to the original deposition of global fallout, expected to be very uneven due to the orography of the region and the amount of rainfall in various areas [42]. Unfortunately, no information was available for the rainfall amount at the sampling sites. It can only be said that concerning the orography, the sites with the highest activity were all situated above 1,000 m a.s.l., on top of the mountains.

Conclusions and outlook

The first radionuclide distribution maps of the whole Molise region have been presented. It can be concluded that the Molise region shows a quite limited content of naturally occurring radioactive elements, all lying within the world average values for soils. A correlation was found between the highest ^{232}Th levels and the areas in the south-west of Molise, containing calcareous sediments and flysch/marl deposits, as well as the Calcaric Regosol. The highest ^{238}U (^{226}Ra) levels were correlated with the calcareous and dolomite formations in the southern and western parts of Molise, mostly covered with the Luvisol. The ^{238}U and ^{232}Th decay chains secular equilibrium in collected soil samples was confirmed within the experimental uncertainty. Finally, ^{40}K was homogeneously distributed with some "hot spots" that could be linked to rock formations containing mica minerals, particularly in coastal areas, mostly made of sand, claystones and conglomerates, but also covered with the Vertic Cambisol and Chromic Vertisol, both rich in clay minerals. The contamination from ^{137}Cs (global fallout and the Chernobyl accident) has also been found, predominantly in the areas located above 1,000 m a.s.l.

The measured gamma-dose rate in the air due to naturally occurring radionuclides in the soil is comparable to those measured in other Mediterranean areas. Together with the dose rate due to the cosmic rays, the total value is well within the world average, and of no particular radiological impact for the population.

In the near future a more detail investigation in the areas where the Th and U contents were significantly higher than in the rest of the Molise region is under consideration with the aim to gain a better understanding of correlation with the geological and pedological formations. The identification of

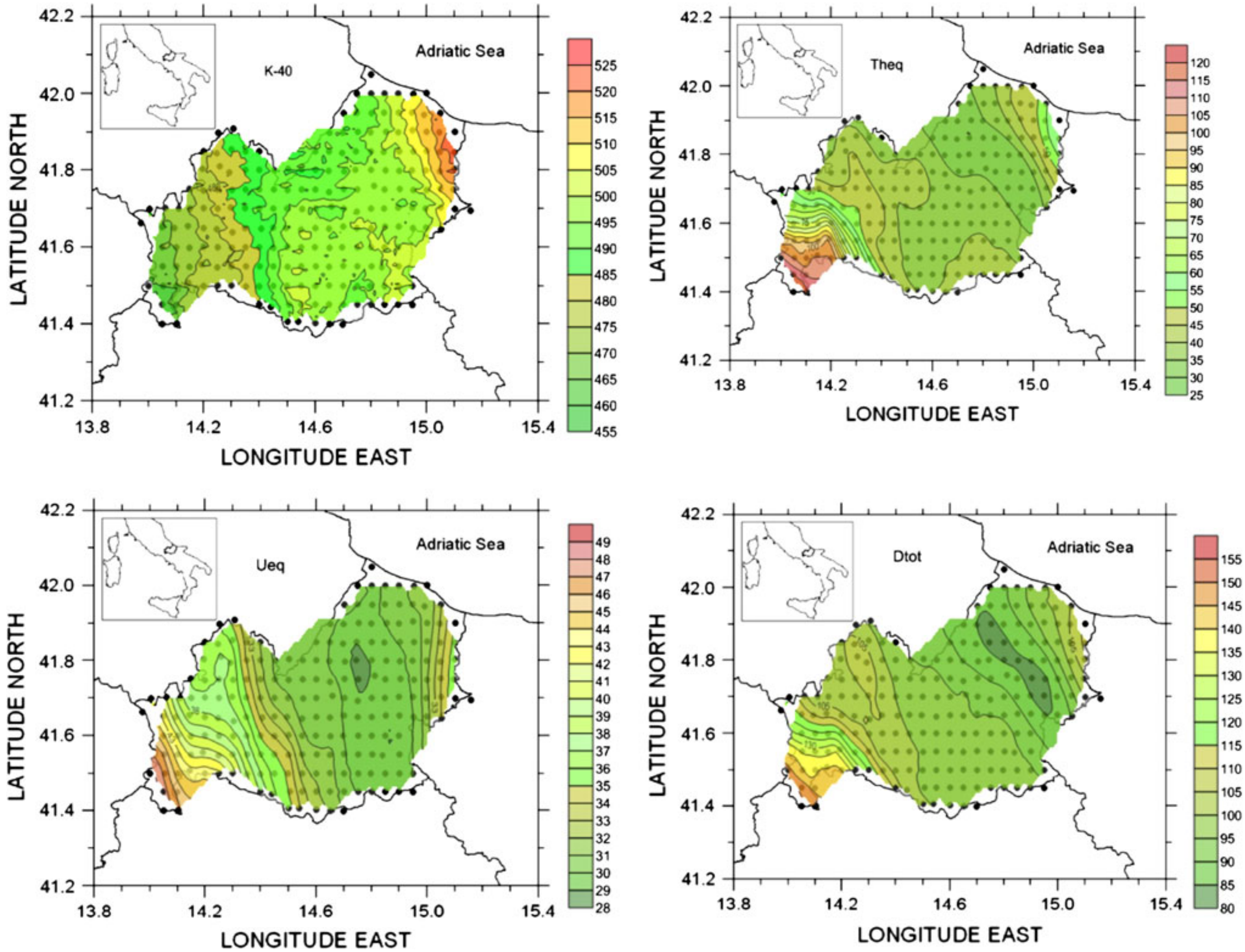

Fig. 1 Radioactivity maps of the Molise region showing massic activities (in Bq kg^{-1}) of ^{40}K, U$_{eq}$ (represented by ^{226}Ra), Th$_{eq,}$ and the total dose rate due to natural radioactivity and cosmic radiation (in nGy h^{-1})

nature of the "hot spots" in the ^{40}K distribution would also be of interest. What is clearly missing up to now in the radionuclide mapping of soils is a more detail and specific investigation of the role of tectonics on the distribution of radionuclides. This could be very interesting as the territory of the Molise region contains some important NW–SE dislocation lines.

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