Modelling seasonal variations of natural radioactivity in soils: A case study in southern Italy

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The activity of natural radionuclides in soil has become an environmental concern for local public and national authorities because of the harmful effects of radiation exposure on human health. In this context, modelling and mapping the activity of natural radionuclides in soil is an important research topic. The study was aimed to model, in a spatial sense, the soil radioactivity in an urban and peri-urban soils area in southern Italy to analyse the seasonal influence on soil radioactivity. Measures of gamma radiation naturally emitted through the decay of radioactive isotopes (potassium, uranium and thorium) were analysed using a geostatistical approach to map the spatial distribution of soil radioactivity. The activity of three radionuclides was measured at 181 locations using a high-resolution γ -ray spectrometry. To take into account the influence of season, the measurements were carried out in summer and in winter. Activity data were analysed by using a geostatistical approach and zones of relatively high or low radioactivity were delineated. Among the main processes which influence natural radioactivity such as geology, geochemical, pedological, and ecological processes, results of this study showed a prominent control of radio-emission measurements by seasonal changes. Low natural radioactivity levels were measured in December associated with winter weather and moist soil conditions (due to high rainfall and low temperature), and higher activity values in July, when the soil was dry and no precipitations occurred.

1. Introduction

Knowing the natural radioactivity in a region is essential to provide a reference baseline map for studying the concentration and behaviour of radioactive isotopes, their transfer through ecosystems, and modelling their spatial distribution and transport (Askouri *et al.* 2011) in view of the annual effective dose received by the general population (Munazza *et al.* 2008; Singh *et al.* 2009; Agbalagba and Onoja 2011).

The environment in which man lives contains a natural background ionizing radiation level that consists of contributions from cosmic rays and terrestrial radioactivity (from both natural and man-made sources). The contributions from these components vary with local geology, altitude and geomagnetic latitudes (Ajayi and Ibikunle 2013).

The most abundant natural terrestrial radioactive isotopes are members of decay series of ^{238}U and ^{232}Th and the non-series ^{40}K . The radioactive decay of these elements results in the emission of

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gamma-rays of sufficient intensity that can be found and measured in most rocks and soils (Guagliardi *et al.* 2013a). Gamma rays emitted from the ground surface relate to the primary mineralogy and geochemistry of the bedrock (Priori *et al.* 2014; Beamish 2015; Ramasamy *et al.* 2015) and the secondary products of weathered materials and soil features (Beamish 2013; Guagliardi *et al.* 2013b; Chandrasekaran *et al.* 2015). Much of the uranium and thorium in igneous rocks is concentrated in a few accessory minerals such as zircon, sphene, and apatite (Johnson 1979). The minerals which bear ^{238}U and ^{232}Th are generally associated with felsic intrusions, particularly with younger ones. The uranium and thorium content of rocks generally increases with acidity, with the highest concentrations found in pegmatites. The highest concentrations of uranium and thorium in sedimentary rocks usually occur in shales. The potassium content of rocks also increases with acidity. In general, potassium is concentrated in micas and feldspars; so, all mafic and ultramafic rocks, which do not host these minerals, have very low K-activity. The potassium content of sedimentary rocks is highly variable, but tends to be higher in shales than in carbonates or sandstones.

In addition, weathering and soil-forming processes modify the distribution and emission of radioelements from the original bedrock source (Buccianti *et al.* 2009). Therefore, geochemical, physical and natural atmospheric interactions influence the distribution patterns of radionuclides in soil (Maharana *et al.* 2010). Radioactivity in soil depends on its formation and the transport processes that were inbuilt since soil formation. Additionally, the type and thickness of the soil cover, porosity, bulk density, moisture, and temperature play a significant role in the amount of radio-ions and γ -ray attenuation (de Groot *et al.* 2009; Dragović *et al.* 2012; Dent *et al.* 2013; Beamish 2015). With regard to the last two variables, it is demonstrated that the gamma-natural activity is intimately related to seasonal events that are responsible for major or minor radio emissions, which can be monitored at the ground surface. Hence, the seasonality is a feature which has to be considered in radioactivity surveys, since radionuclides exhibit air concentration time series with characteristic variations (Guagliardi *et al.* 2013b) according to temperatures and humidity.

Using gamma-ray spectrometry as a mapping tool requires an understanding of radioelements distribution and mobility (Buccianti *et al.* 2009; Guagliardi *et al.* 2013a, c; Chandrasekaran *et al.* 2015).

Geostatistics (Matheron 1971) is particularly suited to the study of the activity of thorium, uranium and potassium in soil because it provides

valuable tools to study their spatial structure and interpolation techniques to estimate the activity values at unsampled locations, known generically as kriging (Chilès and Delfiner 2012). Interested readers are referred to the many texts in which geostatistical methods have been described (Journel and Huijbregts 1978; Goovaerts 1997; Wackernagel 2003; Webster and Oliver 2007; Chiles and Delfiner 2012, among many others).

The main objective of this study was to model, in a spatial sense, the soil radioactivity in an urban and peri-urban area in southern Italy (Cosenza-Rende, Calabria), whose peculiar assemblage of geological, geomorphological, pedological, and climatic features make it representative of larger areas in the central Mediterranean and in the peri-Mediterranean belt. The interest of the international community in urban areas and their surroundings is recently growing because of a number of potential threats to the environment and the man's health, because of the urbanization and human pressure increase. In the study area, measures of gamma radiation naturally emitted through the decay of radioactive isotopes (potassium, uranium and thorium) were analysed using a geostatistical approach to map the spatial distribution of soil radioactivity, with a special focus on the potential influence of seasonal changes on it. This issue represents a crucial point for (i) a better understanding of the major factors that control the activities of natural radionuclides, such as source lithologies, pedogenetic features and associated processes (Buccianti *et al.* 2009; Milenkovic *et al.* 2015; Guagliardi *et al.* 2016), and (ii) a correct assessment of the radioactivity spatial distribution, due to the intrinsic heterogeneity and high spatial variability of soil properties through space (and time) at different hierarchic levels, i.e., from the landscape or catchment scale to the pedon, its constitutive horizons and any distinct pedological features, up to the micro- and sub-microscopic scales (Wilding and Drees 1983; McBratney *et al.* 2000; Lesch and Corwin 2008; Scarciglia *et al.* 2009, 2011; Argyraki and Kelepertzis 2014; Korobova *et al.* 2014). These changing features of the soil body and parent rocks may also affect drainage conditions and moisture availability, which in turn may affect field measurements of radionuclide activities during different seasons. Therefore, a correct knowledge of this seasonal behaviour may represent an essential basis for inspiring appropriate strategies for the acquisition of soil radioactivity.

2. Materials and methods

The study area includes the administrative district of two cities in southern Italy (Calabria): Cosenza

and Rende. It covers 92 km², is densely populated with about 106,000 inhabitants and poorly industrialized. It is representative of Mediterranean areas for common geological, geomorphological, pedological, climatic, and urbanization characteristics, as well as for the widespread presence of rocks hosting radionuclides (Buttafuoco *et al.* 2011; Bruno *et al.* 2012; Guagliardi *et al.* 2013d, 2015; Ricca and Guagliardi 2015).

Geologically, the study area lies in Pliocene succession of sands and gravels, silty clays with silt interlayers, Pleistocene to Holocene alluvial sands and gravels and very small outcrops of Miocene carbonate rocks (Guagliardi *et al.* 2012; Fabbriatore *et al.* 2014). Sediments overlap the Palaeozoic intrusive–metamorphic complex of Sila Massif, formed by paragneiss, biotite schists, grey phyllitic schists, which are often affected by weathering processes (Le Pera *et al.* 2001).

The climate regime of Calabria is typically Mediterranean with some marked seasonal differences (Buttafuoco *et al.* 2010).

Measurements were carried out at 181 locations (figure 1) using portable gamma-ray spectrometer (GF Instruments 2001). Each measurement included the full spectrum of the natural gamma-radiations.

⁴⁰K, in natural conditions, contains a fraction of a radionuclide that can be detected directly. Conversely, the determination of ²³⁸U and ²³²Th was indirect because it is based on the detection of the ²¹⁴Bi radionuclide, which is a member of ²³⁸U decay series in the case of uranium, and on the measurement of the radionuclide ²⁰⁸Tl in the case of ²³²Th. Interested readers can be referred to Guagliardi *et al.* (2013a) for more technical details.

The survey was carried out in summer and winter to take into account two different meteorological conditions.

Geostatistical theory is based on the observation that the variability of measurable quantities, called regionalized variables, have a particular spatial structure. The activities of each isotope (thorium, uranium, and potassium), $z(\mathbf{x}_\alpha)$, at points $\mathbf{x}_\alpha$ ($\mathbf{x}$ is the vector of coordinates x , y and α the sampling points = 1, ..., N) and $\mathbf{x}_\alpha + \mathbf{h}$ are autocorrelated, and autocorrelation depends on both the vectors, $\mathbf{h}$ (module and direction), which separates the two points, and the activity of each isotope considered. Each activity value can be seen as a particular realization, or outcome, of a random variable $Z(\mathbf{x}_\alpha)$ and the set of dependent random variables $\{Z(\mathbf{x}_\alpha), \alpha = 1, \dots, n\}$, constitutes a random

Figure 1. Radioactivity measurement location map of the study area.

function $Z(\mathbf{x})$ (Webster and Oliver 2007; Chilès and Delfiner 2012).

The experimental variogram $\gamma(\mathbf{h})$ is a quantitative measure of the spatial correlation of the regionalized variable $z(\mathbf{x}_\alpha)$ and describes the major spatial features of the attribute under study. It is a function of the vector $\mathbf{h}$ (distance and direction) of data pairs values $[z(\mathbf{x}_\alpha), z(\mathbf{x}_\alpha + \mathbf{h})]$. The variogram is the expected value of the squared differences:

$$\gamma(\mathbf{h}) = \frac{1}{2N(\mathbf{h})} \sum_{\alpha=1}^{N(\mathbf{h})} [z(\mathbf{x}_\alpha) - z(\mathbf{x}_\alpha + \mathbf{h})]^2, \quad (1)$$

$N(\mathbf{h})$ is the number of data pairs for a given class of distance and direction. To estimate the variogram analytically for any distance $\mathbf{h}$, a theoretical variogram model is fitted to the experimental variogram. The variogram models can represent bounded or unbounded variation. In the bounded models, the variance has a maximum (sill) at a finite lag distance (range) over which pairs of values are spatially correlated. A cross-validation test is used to check the best fitting and consists in taking each data point, in turn, removing it temporarily from the data set and using its neighbouring information to predict the value of the variable at its location. The mean error (ME) and the mean squared deviation ratio (MSDR) were used to evaluate the goodness of fit. The estimate is unbiased, if the ME value is close to 0, while the model for the variogram is accurate, if the MSDR (ratio between the squared errors and the kriging variance) value is 1.

Geostatistical methods are most efficient when carried out on variables that have Gaussian distributions because few exceptionally large or small values may contribute to several squared differences and inflate the average variance (Webster and Oliver 2007). Gaussian anamorphosis (Chilès and Delfiner 2012) is a flexible approach to transform a variable with a skewed distribution, into a Gaussian variable regardless of the shape of the

sample histogram. The initial attribute $\{Z(\mathbf{x}), x \in R^2\}$ is transformed into a Gaussian-shaped variable $\{Y(\mathbf{x}), x \in R^2\}$ with zero mean and unit variance, such that

$$Z(\mathbf{x}) = \phi[Y(\mathbf{x})]. \quad (2)$$

To transform the raw variable into a Gaussian one, this function has to be inverted as follows:

$$Y(\mathbf{x}) = \phi^{-1}[Z(\mathbf{x})]. \quad (3)$$

The Gaussian anamorphosis can be achieved by using an expansion into Hermite polynomials $H_i(Y)$ (Wackernagel 2003; Bleines *et al.* 2008) restricted to a finite number of terms:

$$\phi(Y) = \sum_{i=0}^n \psi_i H_i(Y). \quad (4)$$

Multi-Gaussian kriging (Goovaerts 1997; Wackernagel 2003) is a kind of kriging, which uses such normalized data. In this study, ordinary multi-Gaussian kriging (Goovaerts 1997) was used to estimate, at the nodes of a 1 m $\times$ 1 m interpolation grid, and map thorium, uranium, and potassium activities. Finally, the predicted values were back-transformed using the inverse Gaussian anamorphosis (equation 2).

All geostatistical analyses were performed using the software Isatis®, release 2015 (<http://www.geovariances.com>).

3. Results and discussion

The results of the gamma-ray spectrometry survey of the study area soils showed a gamma activity, which ranged from 0.02 to 1.21 Bq kg⁻¹ (in winter) and from 0.62 to 7.86 Bq kg⁻¹ (in summer) for ⁴⁰K, from 0.02 to 1.22 Bq kg⁻¹ (winter) and from 0.36 to 5.32 Bq kg⁻¹ (summer) for ²³⁸U, from 0.01 to 0.12 Bq kg⁻¹ (winter) and from 0.05 to 2.96 Bq kg⁻¹ (summer) for ²³²Th, and from 2.42 to 290.34 Bq kg⁻¹ (winter) and from 328.05 to 476.15 Bq kg⁻¹ (summer) for TC.

Table 1. Variogram model parameters for Gaussian data activity of ⁴⁰K, ²³⁸U, ²³²Th and total radioactivity.

Variable	Winter			Summer		
	Model	Range (m)	Sill (–)	Model	Range (m)	Sill (–)
G ⁴⁰ K	Nugget		0.5864	Nugget		0.5493
	Spherical	1894.98	0.4319	Spherical	1178.77	0.3774
G ²³⁸ U	Nugget		0.7604	Nugget		0.5434
	Spherical	1518.86	0.2885	Spherical	812.84	0.2934
G ²³² Th	Nugget		0.4513	Nugget		0.4961
	Exponential	1532.04	0.4994	Exponential	1007.98	0.4434
G total radioactivity	Nugget		0.5014	Nugget		0.4466
	Exponential	2357.35	0.4380	Spherical	2013.47	0.2217

Note. Practical range equal to 3r.

Data related to winter and summer activities for ^{40}K , ^{238}U , ^{232}Th and TC were transformed through the Gaussian anamorphosis. In the variographic analysis of all isotopes, no anisotropy was evident in the maps of the 2-D variograms (not shown) and a bounded isotropic variogram model was fitted for each experimental variogram. In the fitted models, except for winter and summer thorium data and for winter data of total radioactivity, two basic structures (table 1) were combined including a nugget effect and a spherical model. For winter and

summer thorium data and for winter data of total radioactivity, the fitted models (table 1) included a nugget effect and an exponential model.

The nugget effect arises from errors of measurement and spatial variation within the shortest sampling interval. The spherical model (Webster and Oliver 2007) is given by

$$\gamma(h) = \begin{cases} c \left[\frac{3}{2} \frac{h}{a} - \frac{1}{2} \left(\frac{h}{a} \right)^3 \right] & \text{if } h \leq a \\ c & \text{if } h > a \end{cases}, \quad (5)$$

where c is the sill and a is the range.

Figure 2. Maps of ^{40}K and ^{232}Th activities in summer and winter.

The exponential model (Webster and Oliver 2007) is

$$\gamma(h) = c \left[1 - \exp \left(-\frac{h}{r} \right) \right], \quad (6)$$

where c is the sill and r is a distance parameter that defines the spatial extent of the model. The function does not show a finite sill and approaches its sill asymptotically. Therefore, it does not have a finite range and, for practical purposes, an effective range is usually taken as the distance at

which the variogram value equals 95% of the sill (approximately $3r$).

The results of cross-validation were satisfactory because the statistics used, i.e., the mean of the estimation error and variance of the mean-squared deviation ratio, were quite close to 0 and 1, respectively.

The fitted variogram models were used with the multi-Gaussian ordinary kriging to produce the maps of winter and summer activities for ^{40}K and ^{232}Th (figure 2), and ^{238}U and TC (figure 3).

Figure 3. Maps of ^{238}U activity and total count in summer and winter.

Figures 4 and 5 show the maps of kriging standard deviation for ^{40}K and ^{232}Th , and ^{238}U and TC.

The two-periods studied spatial patterns of radionuclide concentrations for the investigated surface soils highlight, along with the variability of geological and pedological units, the influence of seasonality.

The soil radioactivity concentrations reveal a spatial distribution related to differences between winter and summer measurements, although in some areas it is quite similar, especially where the

lowest radioactivity values were detected. Major seasonal changes occur for TC and ^{238}U .

The radionuclides activity shows a minimum in December, which corresponds to the winter measurements when the radioactive gases are trapped near the surface, while in summer, when rainfall decreased the soil water content, it shows a greater level.

In summer, gases move upward carrying radionuclides with them and thereby increasing ionization near to soil surface (Guagliardi *et al.* 2013d,

Figure 4. Maps of kriging standard deviation for ^{40}K and ^{232}Th .

2016). Therefore, the decrease in gamma-radiation levels may be probably due to high soil water content in winter, as a result of reduced rates of gas diffusion. Also, the influence of air temperature seems to be important. Hence, the activities of ^{238}U , ^{232}Th , and ^{40}K exhibit a positive correlation with air temperature and are negatively correlated with precipitations. This feature influences the spread of radiation in soils, where water molecules probably form a thin physical shield to gamma ray detection in winter, which conversely disappears in summer. Similar conclusions on the seasonal

variations in the environmental gamma-radiation were reported in the literature (Yamazaki *et al.* 2002; Lebedyte *et al.* 2003; Prasad *et al.* 2005; Baciú 2006). The novelty of the present study is to point out more clearly the issue showing how seasonal climatic variations can affect the radionuclides activities through the application of geostatistical methods. The exhibition of these variations, with radioactivity peak values in summer, is well evident in the study area, which, despite being quite locally dimensioned, can be considered a typical Mediterranean scenario because embodies its main features.

Figure 5. Maps of kriging standard deviation for ^{238}U and total count.

4. Conclusions

Because of harmful effects of radiation exposure on human health, radioactivity has become an environmental concern for local public and national authorities. Modelling and mapping the seasonal variations of radioactivity in soil have allowed the analysis of the activity of natural radionuclides. The amount of radioactivity in soil depends not only on soil type and its uses, but also on climate, particularly on thermal and rainfall regimes, of a region.

The study area provided an excellent opportunity for quantifying the temporal variability of gamma rays in soil and for monitoring the available amount of radioactivity for its geological, geomorphological, pedological and climatic features, which are common to other similar areas in the Mediterranean Basin.

In this analysis, the maps of radioactivity have clearly shown that the values of TC and the activities of ^{40}K , ^{238}U and ^{232}Th are higher in summer than in winter, when they are often less than 1 Bq kg^{-1} . The differences between the two seasons may probably be a consequence of changes in meteorological conditions and soil settings (due to rainfall, soil moisture and drainage conditions, and temperature).

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