

Modelling seasonal variations of natural radionuclides in agricultural soils

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Abstract. Estimating activity of natural radionuclides in agricultural soil is very important for the protection of public health because the released radioactivity can enter the food chain. Radioactivity measurements were carried out in two different dates (winter and summer) in agricultural soil using a GRM-260 gamma-ray spectrometer. The study area (100 m x 100 m) was an olive orchard in southern Italy. Measurements were carried out at 361 locations in January and July 2011. At the same locations, soil water content was measured to take into account the effect of soil moisture on radioactivity. A multi-Gaussian approach was used to explore and map the activity of naturally occurring radionuclides and soil water content for both seasons of measurements. The minimum radioactivity values were recorded in winter and the maximum values in summer, probably as a consequence of changes in weather and soil conditions (rainfall, soil moisture, temperature).

Key words: Soil, radionuclides, seasonality, olive orchard

Introduction

Radioactivity is omnipresent in the earth's crust in different amounts for both natural and man-made origin (Akhtar et al., 2005). Soil on the earth's crust is a source of radioactive risk, not only for continuous exposure to human being, but also, in case of agricultural soil, for the entry of radionuclides into the food chain and consequently into humans through the crops. At this regard soil-to-plant transfer constitutes one of the key processes, since in the long term the entry of radioelements into human food chains is controlled by the uptake of plant roots.

The amount of radioactivity in soil depends upon soil type and its uses, but also upon climate and particularly on thermal and rainfall regime of a region.

Quantifying temporal variability of gamma rays in soil is crucial for monitoring the available amount of radioactivity for crops.

Calabria Region has a Mediterranean climate, which is characterized by dry summers and relatively intense rains during spring and autumn. Soil properties in Calabria are intermediate between those of cold and subtropical climates (Llauradó et al., 1994). These characteristics produce an evident seasonality in

radionuclides caused by the sharp contrasts in temperature and relative humidity (Baeza et al., 2001).

The objective of the study was to analyze the seasonal variations of activity for ⁴⁰K, ²³⁸U and ²³²Th in an agricultural soil of southern Italy. In addition, seasonal variations of ⁴⁰K and soil water content were interpolated and mapped to take into account the effect of soil moisture on radioactivity.

Materials and Methods

The study area (10000 m²) was located in an olive orchard in the southern Italy. In two different dates (January and July), called from now on winter and summer, measurements of radioactivity for three natural nuclides ⁴⁰K, ²³⁸U, ²³²Th in soils were carried out *in situ* at 361 locations (Fig. 1) by means of the portable gamma-ray spectrometer GRM-260 (GF Instruments®). Each measurement included the full spectrum of the natural gamma-radiation (counts per 4 minutes) and registered in 256 channels, each of which equal to 12 keV. The counts were then transformed into activity of the corresponding radioactive elements. The spectrum is divided into four parts, groups of channels called ROI (Region Of Interest), in relation to the peak positions of

studied radionuclides and due to the resolution of the scintillation detector. This instrument was designed for spectral measurements of natural and artificial radionuclides in field and laboratory conditions, giving information on the type and the concentration of radionuclides in rocks, soil and water.

Soil water content (SWC) was also measured using the reflectometer TRASE TDR System (Soilmoisture Equipment Corp.).

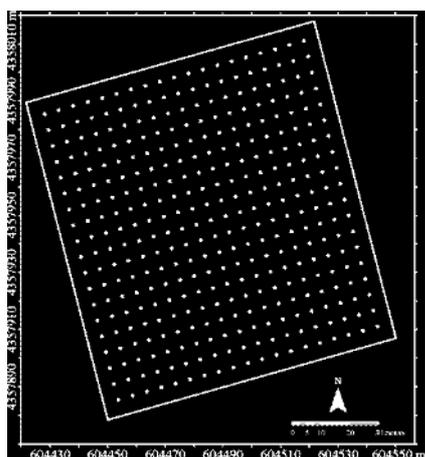


Fig. 1. Sample point locations

Gamma-ray measurements and soil water content can be regarded as regionalized variables (Matheron, 1971) and values at unsampled locations were predicted using the geostatistical methods. They provide a valuable tool for studying the spatial structure of radioactivity and SWC by taking into account the spatial autocorrelation in data to create mathematical models of spatial correlation structures commonly expressed by variograms. The Geostatistical methods are most efficient when carried out on variables that have Gaussian distributions, because a few exceptionally large or small values may contribute to several squared differences and inflate the average variance (Webster and Oliver, 2007). Gaussian anamorphosis (Wackernagel, 2003) allows one to transform a variable with a skewed distribution into a Gaussian variable regardless of the shape of the sample histogram. The kriging of such normalized data is referred to as multi-Gaussian kriging (Verly, 1983; Goovaerts, 1997; Wackernagel, 2003). It is based on a multi-Gaussian model and requires a prior Gaussian transformation of the initial attribute $\{Z(\mathbf{x}), \mathbf{x} \in R^2\}$ into a Gaussian-shaped variable $\{Y(\mathbf{x}), \mathbf{x} \in R^2\}$ with zero mean and unit variance. Ordinary multi-Gaussian kriging was used to predict radionuclides at unsampled locations. Ordinary multi-Gaussian kriging leads to a weakening of the stationary assumption and makes it robust to a departure of the data from the ideal stationary model (Emery, 2005).

Finally, the values of the Gaussian variables were estimated at the nodes of a 1 m x 1 m interpolation grid and back-transformed using the inverse Gaussian

anamorphosis.

Results and Discussion

Descriptive statistics of radioactivity and soil water content data for the two seasons are presented in Table 1. Radioactivity data ranged from 3.13 to 65.73 and from 3.13 to 115.81 Bq kg⁻¹ for potassium, from 0.12 to 4.08 and from 0.05 to 7.41 Bq kg⁻¹ for uranium, from 0 to 0.37 and from 0 to 0.45 Bq kg⁻¹ for thorium in winter and summer respectively. Soil water content data ranged from 0.174 to 0.491 and from 0.960 to 0.287 m³ m⁻³ in winter and summer respectively. All

Table 1. Basic statistics of winter and summer activity for ⁴⁰K (Bq kg⁻¹), ²³⁸U (Bq kg⁻¹), ²³²Th (Bq kg⁻¹). Soil water content (SWC, m³ m⁻³) basic statistics are also reported.

	⁴⁰ K	²³⁸ U	²³² Th	SWC
Winter				
Min	3.13	0.12	0	0.174
Max	65.73	4.08	0.37	0.491
Mean	13.59	0.76	0.17	0.350
Median	12.52	0.74	0.16	0.358
Dev. St.	8.60	0.39	0.06	0.473
Kurtosis	9.99	20.90	0.31	0.72
Skewness	2.66	3.49	0.46	-0.10
Summer				
Min	3.13	0.05	0	0.096
Max	115.81	7.41	0.45	0.287
Mean	18.95	0.83	0.16	0.164
Median	15.65	0.74	0.16	0.152
Dev. St.	15.26	0.51	0.06	0.045
Kurtosis	9.21	77.28	1.80	0.02
Skewness	2.64	6.73	0.56	0.95

The radionuclides activity shows a minimum in January and, as it was expected, precipitations increased soil water content and affected the changes in the apparent activities of ⁴⁰K, ²³⁸U and ²³²Th in soil, even if the seasonal fluctuations of activity were minimal and they were more marked for ⁴⁰K and less for ²³⁸U and ²³²Th (see maximum values in Table 1). That was due to the low values of background radioactivity and consequently appreciable variations between the two seasons were minimal. However, lower radionuclides activities occurred during the winter when the radioactive gases are trapped near the surface, while in summer, the reduced soil water content allowed a greater level of radio-emission. In summer gases will move upward carrying radionuclides with them and thereby increasing ionization near to soil surface.

When soil water content was high for longer time periods, the gamma-radiation levels decreased, as a result

of reduced rates of gas diffusion. The exhalation rate of the radioactive gases is dictated by the soil water content and is influenced by the soil permeability. Also the influence of air temperature seems to be important. Similar conclusions on the seasonal variations in the environmental gamma-radiation were reported in literature (Baciu, 2006; Lebedyte et al., 2003; Prasad et al., 2005; Yamazaki et al., 2002).

Hereafter only the results for ⁴⁰K activity and soil water content are reported.

Data of winter and summer activity for ⁴⁰K and soil water content (SWC) were transformed through the Gaussian Anamorphosis. For each variate, a variogram map (not shown) was calculated and they did not show any significant difference as a function of direction. Results of variographic analysis for all variates are reported in Table 2. The experimental variogram for the Gaussian winter activity for ⁴⁰K was modelled by a nested variogram that combines two basic structures (Table 2) including a nugget effect and a spherical model (Webster and Oliver, 2007) with a range of 6.5 m. The nugget effect implies a positive intercept of the variogram. It arises from errors of measurement and spatial variation within the shortest sampling interval (Webster and Oliver, 2007). The range is a distance over which pairs of values are spatially correlated. The very short range value indicates that winter measured data of ⁴⁰K activity are weakly structured.

For the Gaussian summer activity of ⁴⁰K, the experimental variogram was modelled by a nested variogram that combines three basic structures (Table 2): a nugget effect; a spherical model with a range of 12 m, and a spherical model with a range of 58.80 m and. The two ranges indicate the presence of processes acting at different spatial scale (short and long range).

The experimental variogram for Gaussian winter data of soil water content was modelled by a nested variogram that combines three basic structures (Table 2) including a nugget effect, a short range exponential model with a range of 20.60 m and a long range spherical model with a range of 100 m. Finally, the experimental variogram for Gaussian summer data of soil water content was modelled by a nested variogram that combines two basic structures (Table 2) including a nugget effect and a spherical model with a range of 18.60 m.

Using the multi-Gaussian ordinary kriging, the Gaussian data of winter and summer activity for ⁴⁰K and soil water content (SWC) were interpolated and mapped (Figs 2 and 3). In winter values of specific activity for ⁴⁰K were very low, while in summer the high values of activity for ⁴⁰K occurred in the right part of the field, which corresponds to the wettest areas. In winter soil water content was more homogeneous than summer and it would seem to determine the specific activity for ⁴⁰K and its spatial pattern.

Conclusion

Monitoring an olive orchard using gamma-ray spectrometry allowed to analyze the seasonal variations of activity for ⁴⁰K, ²³⁸U and ²³²Th. Moreover, the effects

of soil water content on radioactivity has been taken in account. Mapping soil water content and the activity for ⁴⁰K allowed their spatial relationship.

Table 2. Variogram model parameters for Gaussian data of winter and summer activity for ⁴⁰K and soil water content (SWC).

Variate	Model	Range (m)	Sill (-)
G ⁴⁰ K _{winter}	Nugget	-	0.2780
	Spherical	6.50	0.4301
G ⁴⁰ K _{summer}	Nugget	-	0.4931
	Spherical	12.00	0.0795
	Spherical	58.80	0.3089
G SWC _{winter}	Nugget	-	0.2619
	Spherical	20.60	0.5440
	Spherical	100.00	0.2226
G SWC _{summer}	Nugget	-	0.3054
	Spherical	18.60	0.7597

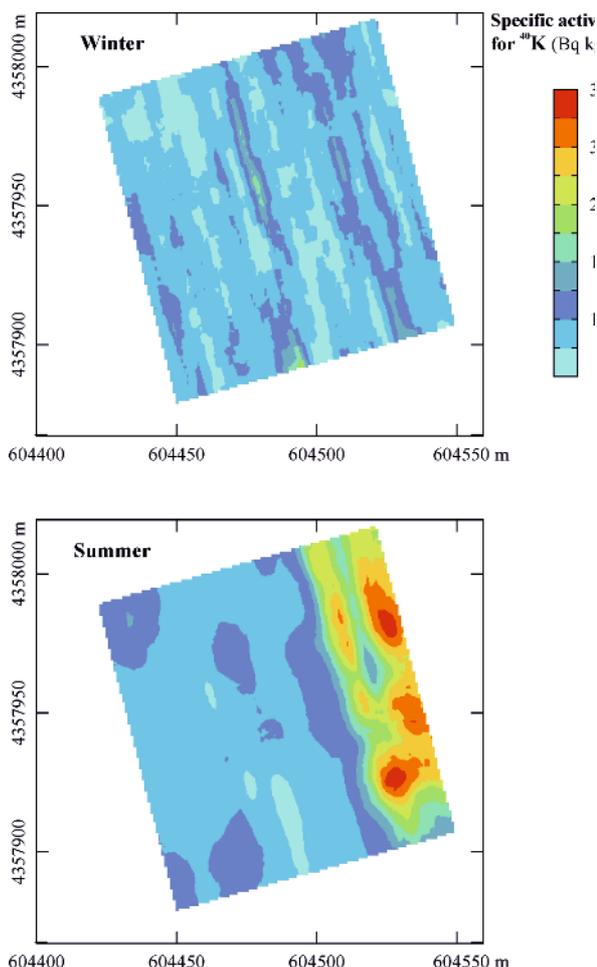


Fig. 2. Maps of specific activity for ⁴⁰K.

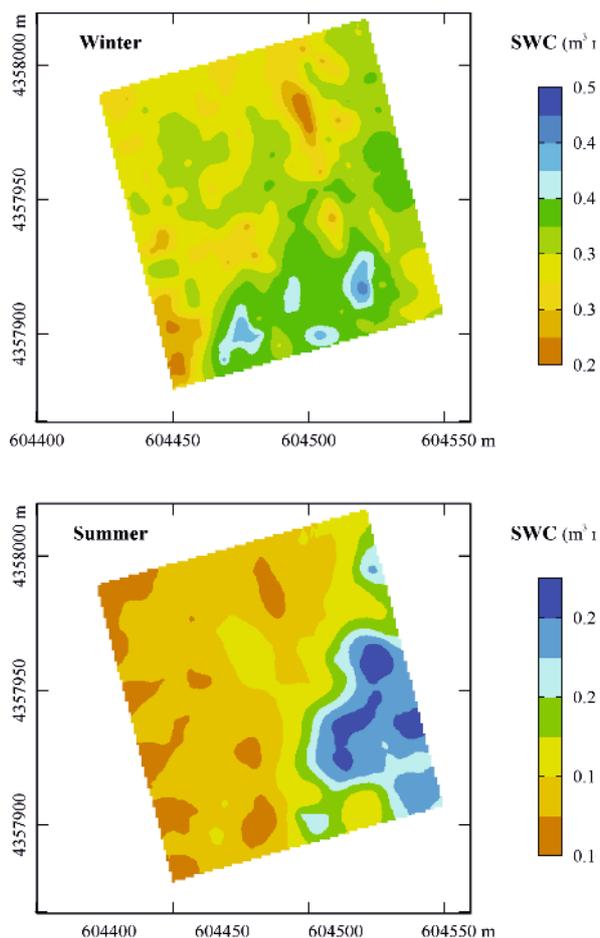


Fig. 2. Maps of soil water content.

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