Natural radioactivity distribution and soil properties: a case study in southern Italy [1]

Ilaria Guagliardi, Nicola Ricca, Maria Grazia Cipriani, Donatella Civitelli, Raffaele Froio, Anna Lia Gabriele, Gabriele Buttafuoco

National Research Council of Italy - Institute for Agricultural and Forest Systems in the Mediterranean (ISAFOM), Via Cavour 4-6, Rende - Cosenza (Italy) ilaria.guagliardi@isafom-cnr.it

Rosanna De Rosa

Department of Earth Sciences, University of Calabria, Ponte Pietro Bucci, 87036 Arcavacata di Rende - Cosenza (Italy)

Abstract: Mapping environmental radioactivity from field gamma-ray spectrometry is a valuable tool for understanding and interpreting pedological control of naturally occurring radioactivity. Soil properties and water content affect the behaviour of natural radioactivity. The main aim of the study were to explore and map the activity of three naturally occurring radionuclides (^{232}Th, ^{238}U, ^{40}K) in an olive orchard and investigate the relationship between some soil properties and the activity of the three radionuclides.

Keywords: natural radioactivity, soil, water content, grain size

1. Introduction

Environmental natural radioactivity in the soil is due to the decay of radionuclides derived from minerals in the earth’s crust. Many naturally occurring elements have radioactive isotopes, but only potassium, and the uranium and thorium decay series have radioisotopes producing gamma rays of sufficient energy and intensity to be measured by gamma ray spectroscopy (IAEA, 2003). The radioactive isotope of potassium ^{40}K occurs as a fixed proportion of K in the natural environment and these gamma rays can be used to estimate the total amount of K present. Uranium and thorium occurs naturally as the radioisotopes ^{238}U, ^{235}U and ^{232}Th. Neither ^{238}U nor ^{232}Th emit gamma rays and their concentrations are estimate from their radioactive daughter products and reported as equivalent uranium (eU) and equivalent thorium (eTh).

The mineral composition of the parent material controls the natural radioactivity of soils (Navas et al., 2011) and the processes of weathering, sedimentation, leaching and sorption, and the movement of groundwater may influence activity levels of natural radionuclides (Dowdall and O’Dea, 2002). Soils play a major role in the cycling of radionuclides and their physico-chemical properties influence the mobility and bioavailability of these radionuclides in terrestrial ecosystems (Kabata-Pendias and Pendias, 2001). A fundamental characteristic of the soil, which greatly influences the environmental transport of radioactivity, is the distribution by grain size.

Approximately 95% of the measurable gamma radiation is emitted from the upper 0.5 meters of the profile (Gregory and Horwood, 1961) and the value of gamma spectroscopy lies principally in the amount of radioisotopes of K, U and Th contained in rocks and soil profiles (Dickson and Scott, 1997). Signal attenuation of radioactivity increases by approximately 1% for each 1% increase in volumetric soil water content (Cook et al., 1996).

---

[1] This research was funded by the Action 2 – Public research laboratory mission oriented. APQ – Scientific Research and technological Innovation in Calabria Region. Laboratory for Food Quality, Safety, and Origin (QUASIORA).

Geostatistical methods provide us a valuable tool to study spatial structure of the activity of radionuclides. They take into account spatial autocorrelation of data to create mathematical models of spatial correlation structures commonly expressed by semivariograms. The interpolation technique of the variable at unsampled locations, known as kriging, provides the ‘best’, unbiased, linear estimate of a regionalized variable in an unsampled location, where ‘best’ is defined in a least-square sense (Webster and Oliver, 2007).

The main objectives of the study were: (a) to explore and map the activity of three naturally occurring radionuclides (^{232}Th, ^{238}U, ^{40}K) in an olive orchard, (b) to investigate the relationship between some soil properties and the activity of the three radionuclides.

2. Materials and methods

The experimental area (100 m x 100 m) is an olive orchard located in southern Italy (Calabria). Ground measurements of gamma rays were carried out using the portable gamma-ray spectrometer GRM-260 of the 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 conventional approach to the acquisition and processing of gamma-ray data is to monitor four broad spectral regions of interest (ROI) corresponding to potassium-40 (ROI_K), uranium-238 (ROI_U), thorium-232 (ROI_{Th}) and the total count (ROI_{TC}).

The gamma ray measurements were made at 361 points at the nodes of a regular 5 x 5 m grid. Volumetric soil water measurements were made at the same locations with 45-cm long rods of a two-probe Trase System TDR (time domain reflectometry) (Topp and Davis, 1985).

Soil samples were collected at only 100 points at the nodes of a regular 10 x 10 m grid, then they were air-dried, ground, passed through a 2 mm sieve and analysed for particle size fractions by the pipette method.

The gamma ray and soil measurements $z(\mathbf{x}_\alpha)$ at different locations $\mathbf{x}_\alpha$ (x is the location coordinates vector and α the sampling points) were interpreted as a particular realization of a random variable $Z(\mathbf{x}_\alpha)$ and analysed using the theory of random functions (Chilès and Delfiner, 1999; Webster and Oliver, 2007; Wackernagel, 2003, among others). As quantitative measure of spatial correlation of the observations $z(\mathbf{x}_\alpha)$, was used the variogram $\gamma(\mathbf{h})$ which is a two-point statistics used to quantify the variability between two random variables separated by a lag vector $\mathbf{h}$. Multi-Gaussian ordinary kriging (Verly, 1983) was used to predict and map the gamma rays and soil particle size fractions values at unsampled locations. It allows spatial prediction of soil properties regardless of the shape of the sample histogram. The multi-Gaussian approach is based on a multiGaussian model and requires a prior Gaussian transformation of the initial attribute $\left\{Z(\mathbf{x}),\ \mathbf{x}\in R^2\right\}$ into a Gaussian-shaped variable $\left\{Y(\mathbf{x}),\ \mathbf{x}\in R^2\right\}$ with zero mean and unit variance. Such a procedure is known as Gaussian anamorphosis (Wackernagel, 2003).

3. Results and conclusions

In opposition to what was expected (Bihari and Dezső, 2008, among many others), no significant correlation was found between soil particle size and gamma ray measurements.

Experimental variograms (Fig. 1) were computed along four directions of azimuth (0, 45, 90, and 135 in degrees clockwise from N-S axis) for gamma-ray data, particle size fractions and soil water content. Then, a variogram model was fitted to the experimental values of semivariance. Figure 1 shows the variograms only for the spectral regions of interest (ROI) corresponding to potassium-40 (ROI_K) and the total count (ROI_{TC}), the soil water content and the soil silt content.

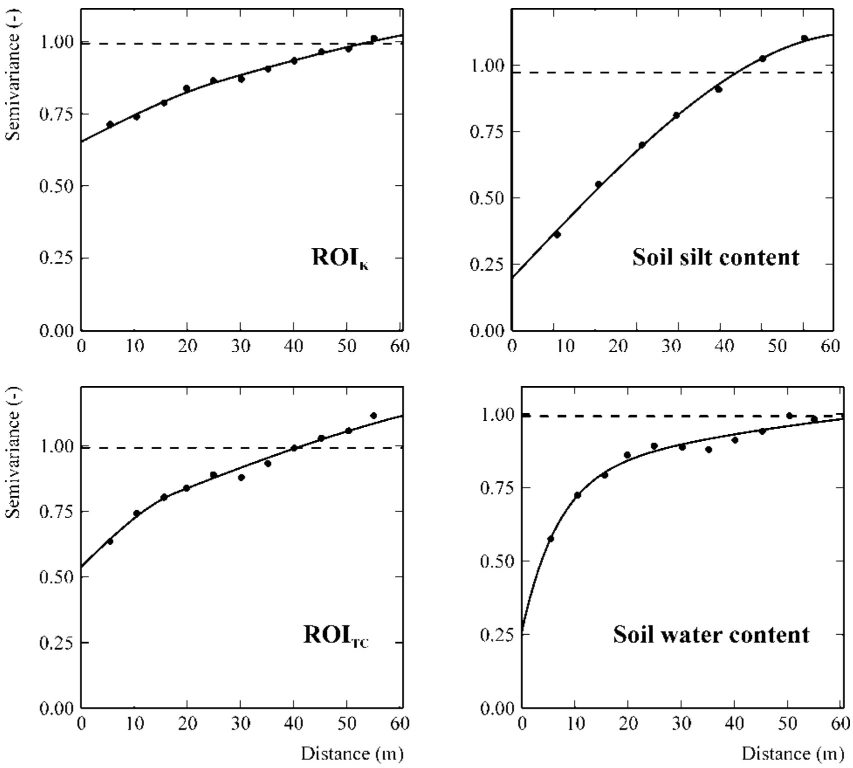

Figure 1: Variograms for the spectral regions of interest (ROI) corresponding to potassium-40 (ROI$_K$) and the total count (ROI$_{TC}$), the soil water content and the soil silt content. The experimental values are the filled points and the solid lines are of the model of variograms. The dashed lines are the experimental variances.

For ^{40}K (ROI$_K$) a nested variogram model was used including three basic structures (Fig. 1): (1) a nugget effect of 0.6529; (2) a spherical model (Webster and Oliver, 2007) with a range of 26 m and a sill of 0.0624; (3) a spherical model with a range of 100 m and a sill of 0.3846. The nugget effect is a discontinuity at the origin of the variogram and relates to measurement errors and to spatial sources of variations at distances smaller than shortest sampling interval (Journel and Huijbregts, 1978). The variogram model for the total count (ROI$_{TC}$) included three basic structures (Fig. 1): (1) a nugget effect of 0.5387, (2) a spherical model with a range of 18 m and a sill of 0.1363; (3) a spherical model with a range of 100 m and a sill of 0.5516.. The variogram model for the soil silt content included two basic structures (Fig. 1): a nugget effect of 0.1964 and a spherical model with a range of 83 m and a sill of 0.9252. The nested variogram model used for the soil water content included three basic structures (Fig. 1): (1) a nugget effect of 0.2619; (2) an exponential model with a practical range of 21 m and a sill of 0.5440; (3) a spherical model with a range of 100 m and a sill of 0.2226. The goodness of fitting was verified by cross validation and the results were quite satisfactory because the statistics used, i.e. mean of the estimation error and variance of the standardised error, were quite close to 0 and 1, respectively.

Finally, using the multi-Gaussian kriging, the spectral regions of interest (ROI) corresponding to potassium-40 (ROI$_K$) and the total count (ROI$_{TC}$), the soil water content and the soil silt content, were interpolated and mapped (Fig. 2).

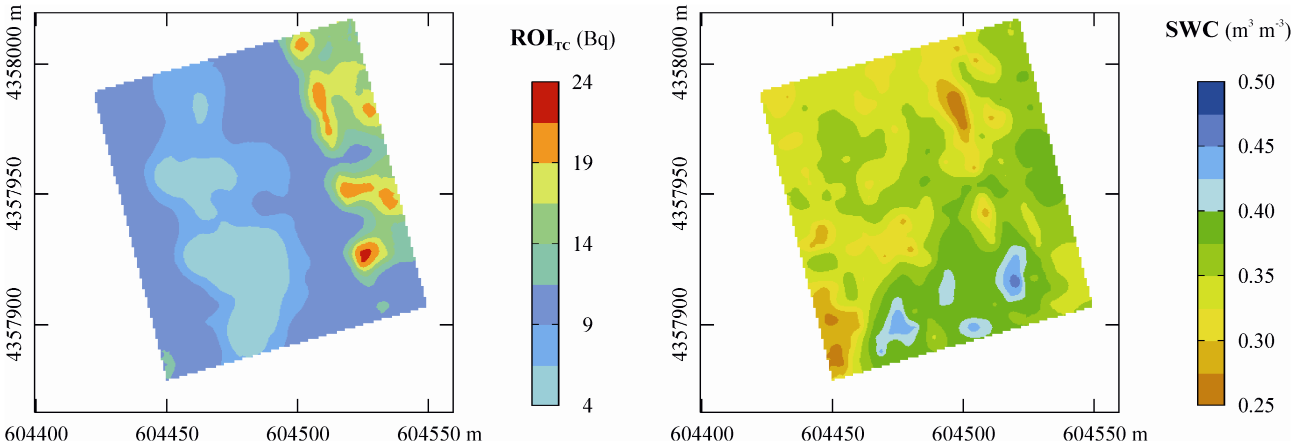

Figure 2: Maps obtained using multi-Gaussian kriging for the spectral regions of interest (ROI) corresponding to the total count (ROI$_{TC}$) and for the soil water content.

Contrarily to what was expected, there was no clear relation between ROITC and soil water content (Fig. 2). Taylor et al. (2002) reported that the attenuation of gamma-rays through the soil varied with bulk density and water content and the signal attenuation increased by approximately 1% for each 1% increase in volumetric water content (Cook et al., 1996). Probably, the lack of relation was due to the rather homogeneous soil water content (mean content = 35%, standard deviation of 4.7%). A new soil survey with low soil water content will confirm the relation between the activity of radioisotopes and water content.

References

Bihari Á., Dezső Z. 2008. Examination of the effect of particle size on the radionuclide content of soils. Journal of Environmental Radioactivity 99, 1083-1089.

Cook S.E., Corner R.J., Groves P.R., Grealish G.J. 1996. Use of airborne gamma radiometric data for soil mapping. Australian Journal of Soil Research, 34, 183–194.

Dickson B.L., Scott, K.M. 1997. Interpretation of aerial gamma-ray surveys: adding the geochemical factors. AGSO. Journal of Australian Geology and Geophysics, 17, 187–200.

Dowdall, M., O'Dea, J., 2002. Ra-226/U-238 disequilibrium in an upland organic soil exhibiting elevated natural radioactivity. J. Environ. Radioact. 59, 91-104.

Gregory A.F., Horwood, J.L. 1961. A Laboratory Study of Gamma-Ray Spectra at the Surface of Rocks. Mines Branch Research Report R.85. Department of Mines and Technical Surveys, Ottawa.

IAEA, 2003. Guidelines for radioelement mapping using gamma ray spectrometry data. IAEA-TECDOC-1363, International Atomic Energy Agency, Vienna, pp. 173.

Kabata-Pendias, A., Pendias, H., 2001. Trace Elements in Soils and Plants, pp. 413, third ed.. CRC, Boca Raton, Fl.

Navas A., Gaspar L., López-Vicente M., Machín J., 2011. Spatial distribution of natural and artificial radionuclides at the catchment scale (South Central Pyrenees). Radiation Measurements 46, 261-269.

Topp G.C; Davis J.L. 1985. Measurement of soil water content using time-domain reflectometry (TDR): a field evaluation. Soil Science Society of America Journal, 49, 19–24

Verly G. 1983. The multigaussian approach and its application to the estimation of local reserves. Mathematical Geology, 15, 259–286.

Wackernagel H. 2003. Multivariate geostatistics: An introduction with applications. Berlin: Springer-Verlag, p. 387.

Webster, R., Oliver, M. A., 2007. Geostatistics for Environmental Scientists. 2nd Ed. Wiley, Chichester.