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Preliminary Results for the Characterization of the Radiological Levels of Rocks in Tuscany Region

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INTRODUCTION

The environmental background levels of natural radiation due to the nuclides in rocks vary in significant amounts that depend on the geological and geomorphological features of a territory. The main source of terrestrial gamma-ray radiation exposure to humans is from ^{238}U, ^{232}Th decay chains and ^{40}K decay.

This paper reports a part of the results of the Research Project "Measurement of natural radioactivity and mapping of the radioisotope abundances of Tuscany Region", which started at August 2008 and it is supported by founds of Tuscany Region. The aim of this project is to realize the thematic maps of radioactivity content and in particular of the abundances of eU[1], eTh[1] and ^{40}K. These goals will be achieved by integrating the information from measurements on samples in laboratory with in-situ investigations and airborne surveys. The Legnaro National Laboratory (LNL) is the national leader for the design and realization of high-resolution gamma-ray spectrometers, portable and massive NaI(Tl) detectors.

The MCA_Rad gamma-ray spectrometry system [1] was designed and built up at LNL for measuring large amount of samples with a minimum attendance: these features fit perfectly with the requirements of this project. This system is able to measure any type of materials (solid, liquid, gas), and due to the high efficiency and its geometric symmetry, absolute activity measurements are possible with systematic errors below 5%.

MATERIALS AND METHODS

The Tuscany Region occupies an area of about 23×10^3 km^2 and it includes 15 paleogeographic domains (figure 1). The sedimentary rocks constitute the main reservoir covering the large part of the Region, the igneous rocks are mainly present in the south part, while in the north-west part of the Region (i.e. Apuane succession) we meet metamorphic rocks: in this region are located the main marble caves of Italy.

The sampling strategy was planed on geological arguments: the homogeneous units are recognized by low density of samples, permitting to focus a high density of samples on the heterogeneous areas. Excluding the quaternary deposits when one sample per 25 km^2 was collected.

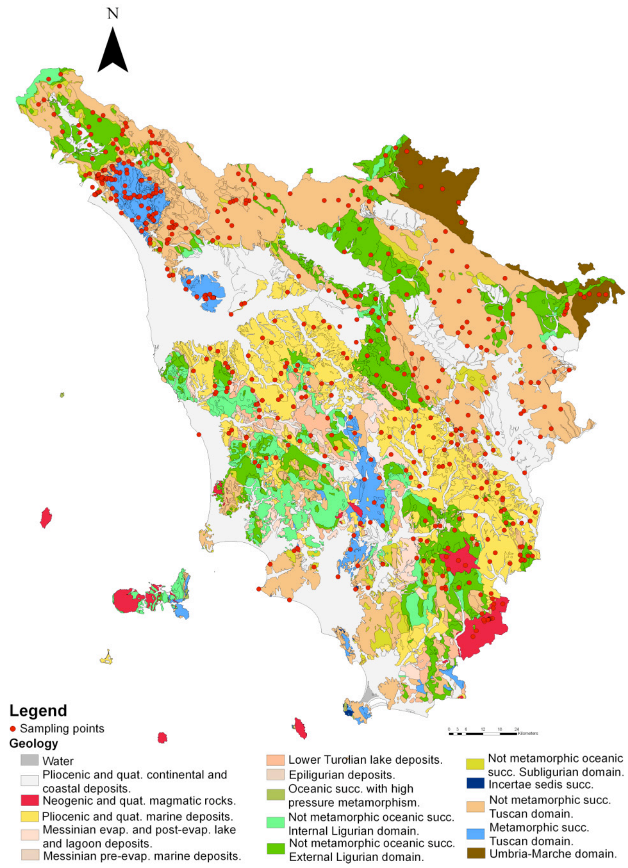

Fig. 1. The paleographic domains of Tuscany Region and the sampling sites.

The information collected during the sampling was organized in a geo-database (GeoDB). The two operators working on field fill the GeoDB with GPS coordinates and the main information about the state of outcropping, the weather conditions and the geological features (unit, formation, lithology).

[1] These concentrations by weight are determined indirectly from ^{238}U and ^{232}Th daughter products (^{214}Bi and ^{208}Tl respectively), that are assumed to be in *equilibrium* with their parent isotope.

The 529 samples collected were crushed, sieved and then placed in a drying oven at temperature 60°C in order to remove the moisture. The 200 cc cylindrical polycarbonate containers filled with the ground rock were weighted and labeled with a barcode. Finally they were stored and kept sealed for 38 days in order to reach the radioactive secular equilibrium between ^{226}Ra and ^{222}Rn (10 half- lives of ^{222}Rn).

The MCA_Rad output (counts, specific activity and abundance) it is adapted in order to easily fill the GeoDB permitting an user-friendly management of the data. This procedure is designed to minimized the human errors and optimize the manpower.

The specific activity of ^{238}U and ^{232}Th was calculated under the assumption of secular equilibrium, using the gamma transitions of energy, 609.3 keV for ^{214}Bi (eU) and 583.2 keV for ^{208}Tl (eTh), while for ^{40}K was calculated through the gamma transition of energy 1460.8 keV. The achieved minimum detection activity (MDA) calculated following [2] were 0.2 Bq/kg for ^{214}Bi, 0.3 Bq/kg for ^{208}Tl and 1.8 Bq/kg for ^{40}K. A preliminary screening for 3600 s of spectrum acquisition reported only 10% of samples below the MDA value, which were further measured for 14400 s.

RESULTS AND CONCLUSIONS

In the current report it was successfully achieved the measurement of 529 sample of rocks by using the MCA_Rad system.

The Falterona-Cervarola unit (FCU) and the Macigno formation (MF) revealed an homogeneous radioactive content, 767.4 ± 187.4 Bq/kg and 829.5 ± 181.5 Bq/kg respectively. Covering about 20% of the territory, mainly in the north and east part of the region, FCU and MF are characterized by sandstone flysch: the homogeneous content of radioisotope is supported by the presence of an homogeneous lithology.

In Tuscany Region we find the main types of rocks characterizing the Earth's crust: sedimentary, igneous and metamorphic rocks.

Concerning the sedimentary rock, the carbonate rocks (lime, dolomite, travertine) are characterized by a radioactivity content lower than the clastic rocks (clay, sandstone, sand): 124.5 ± 133.7 Bq/kg and 724.6 ± 239.5 Bq/kg respectively. In some neogenic deposits (marine and evaporitic) we find an alternation of sandstone, conglomerate, limestone, marl, mudstone, clay and silty-marly clay: the large spread of specific radioactivity values (317.3 ± 230.0 Bq/kg) reveals this alternation.

The igneous rocks coming from Tuscan Magmatic Province show elevated levels of radioactivity (1738.2 ± 886.4 Bq/kg). In more details the acid igneous rocks (volcanic ash, tuff, lapillus) and basic igneous rocks (basalts) are characterized by two opposite levels of radioactivity: 2014.6 ± 611.0 Bq/kg and 80.0 ± 85.1 Bq/kg respectively.

The metamorphic rocks are the result of alteration of existing rocks by either excessive heat and pressure, or through the chemical action of fluids. Although in general this alteration can cause chemical changes and/or structural modification to the minerals making up the rock, the radioactivity content in the samples of the metamorphic rocks can be related to those of the lithology of origin. On the other hand the perturbation from metamorphic processes increase the spread of the radioactivity values.

In Table 1 we summarize the ^{40}K, $_{eq}$U and $_{eq}$Th abundances organized in the classes of rocks described above. This classification is a key of the interpretation of the natural radioactivity content of the region.

The environmental and geological information collected in the GeoDB will permit to refine this framework and to build thematic maps by using geostatistical methods. Finally the GeoDB will be updated to the server database (MySQL) and then will be published on line by using Web-GIS service based on Google Earth® platform.

Table 1. Average of the 40K, eU and eTh abundance for the main lithologies. The standard deviation is calculated on N samples.

Lithology	N.	^{40}K (%)	eU (ppm)	eTh (ppm)
Sedimentary Rocks				
Clastic	198	2.1 ± 0.7	2.6 ± 1.1	10.6 ± 4.6
Carbonatic	80	0.3 ± 0.4	1.3 ± 1.4	2.0 ± 1.9
Lime-Clay	49	0.9 ± 0.7	1.5 ± 0.9	4.6 ± 3.2
Igneous Rocks				
Basic	9	0.2 ± 0.2	0.9 ± 1.6	3.0 ± 7.2
Acid	54	5.3 ± 1.6	11.3 ± 5.8	56.6 ± 26.3
Metamorphic Rocks				
MetaClastic	63	2.5 ± 1.1	2.5 ± 1.1	11.5 ± 4.4
MetaCarbonatic	34	0.5 ± 0.6	0.8 ± 0.8	2.4 ± 3.2
MetaMagmatic	4	3.3 ± 1.7	2.6 ± 1.3	14.3 ± 8.3

[1] G.P. Bezzon et al, LNL Annual Report, 2008, 154.
[2] L.A. Curie, Anal. Chem., 40 (1968) 586.