Mapping of Natural Radioelements Using Gamma-Ray Spectrometry: Tuscany Region Case of Study

G.P. Bezzon¹, G.P. Buso¹, C. Broggini⁵, A. Caciolli^{2,5}, I. Callegari², T. Colonna², E. Guastaldi², F. Mantovani^{3,4}, S. Mariani^{2,4}, G. Massa², R. Menegazzo⁵, L. Mou², C. Rossi Alvarez⁵, M. Shyti^{3,4}, G. Xhixha^{3,4,6}.

¹ INFN, Laboratori Nazionali di Legnaro, Legnaro (Padova), Italy. ² Center for GeoTechnologies, San Giovanni Valdarno, Italy. ³ Physics Department, University of Ferrara, Ferrara, Italy. ⁴ INFN, Ferrara Section, Ferrara, Italy. ⁵ INFN, Padova Section, Padova, Italy. ⁶ Faculty of Forestry Science, Agricultural University of Tirana, Tirana, Albania.

INTRODUCTION

The main source of terrestrial gamma-ray radiation exposure to humans comes from ²³⁸U, ²³²Th decay chains and ⁴⁰K decay. Gamma-ray spectrometry is a consolidated methodology extensively utilized by many nations for investigating and mapping of natural radioelements [1].

Tuscany has become the first Italian region which joint the list of nations which have a detailed knowledge of their territory regarding natural radioactivity distribution. The territory where investigated through gamma-ray spectrometry implying measurements utilizing laboratory, portable and airborne instruments in order to realize the thematic maps of radioactivity content and in particular of the abundances of $eU(^a)$, $eTh(^a)$ and ^{40}K .

The Tuscany Region occupies an area of about $23 \times 10^3 \text{ km}^2$ and it includes 15 paleogeographic domains subdivided in about 120 different litotypes (figure 1). It have an extension for mountain and hills of about 25% and 67% respectively and only 8% of the territory has a plain geomorphology. Therefore, the survey program was planed taking into consideration the geological, geographical and morphological variability of the territory under investigation and divided in three approaching scales undertaking laboratory, in-situ and airborne measurements following the IAEA guidelines [1]. In order to finalize the necessities of such an extensive study dedicated instruments were designed, realized and calibrated at the Legnaro National Laboratory (LNL). The aim of this paper is to emphasize the utilization of such instruments and the results obtained.

MATERIALS AND METHODS

The strategy adopted in this study was by merging the information gathered in laboratory, in-situ and airborne through self designed instruments in order to accomplish specific tasks relative to the study objectives.

The first approach was designated in contact with the territory where a sampling strategy was planed on geological arguments increasing the sampling frequency with area non-homogeneities. In order to be able to manage high frequency sampling rates an HPGe highresolution gamma-ray spectrometry called MCA-Rad was designed [2,3]. MCA-Rad is a fully automated instrument able to work and process a high number of samples with minimum attendance. This instrument was utilized to measure the natural radioactivity concentration of about 677 rock samples and 1236 soil samples collected in about 120 days over 2 years distributed as in figure 1.



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

The second approach was oriented in order to have onfield data of natural radioactivity concentration by using

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

portable gamma ray spectrometers. The instruments were configured to be easily transportable and give a quick, few minutes, and accurate result. In order to enhance the potentialities of the NaI(Tl) detector working on field, a new calibration and analysis method was developed by using full spectrum analysis with non-negative least square FSA-NNLS [4]. This method permits to perform static (placed on ground) and dynamic (mounted in a back-bag at 1m of height) measurements, averaging the signal coming for about 2 m^2 and 200 m^2 respectively, showing accuracy similar to the site geochemical variability for a plain geometrical configuration. In order to cross-validate the FSA-NNLS analysis method, a comparison between measurements on samples and measurements in-situ with portable instruments was performed. For this reason 80 sites were chosen over an area of about 3500 km² on the Ombrone river valley: for each site the results obtained in situ, during a static measurement lasting 5 min, were compared with the log-normal average of five samples collected in the corresponding site and measured in laboratory with the MCA-Rad system.

Knowing the distribution of abundances for each isotope $(\delta K, \delta eU, \delta eTh)$ in an area we found a correction factor within 1 sigma for in-situ measurements (Eq. 1).

$$\begin{split} K_{Nal(Tl)} & [\%] = (0.97 \pm 0.06) \ \delta K & [\%] \\ eU_{Nal(Tl)} & [ppm] = (1.11 \pm 0.15) \ \delta eU & [ppm] \\ eTh_{Nal(Tl)} & [ppm] = (0.92 \pm 0.08) \ \delta eTh & [ppm] \end{split}$$
(1)

The third approach was to survey extensive areas by utilizing massive gamma-ray spectrometry detectors mounted in aircrafts. For this reason, the so-called AGRS 16L system was constructed; it is composed by 4 NaI(Tl) detectors for a total volume of about 16L and configured for aircraft transportation. The system is also equipped with a GPS and Pressure and Temperature sensors necessary for global positioning and for altimetry measurements. The combination of the calibration and analysis methodology using FSA-NNLS, and the rigorous choice of the aircraft, permits us to analyze sufficient statistics of data in bins of 2 seconds, having a spatial resolution of about 0.2 km² during airborne surveys. The airborne survey program was extended in 100 h distributed in few test flights and 33 effective flights covering a total area of about 20% of the Tuscany region. The explored area was for a large fraction of plain morphology.

RESULTS AND CONCLUSIONS

During this work were successfully designed, realised and utilized instruments and methodologies used in gamma-ray spectrometry for mapping natural radioelements, which results are very satisfactory.

The MCA-Rad system was utilized to measure over 1913 samples of rock and soil. In figure 2 (a,b,c) are shown the distributions of K, eU, eTh in rocks and soils normalized for data measured by taking the natural logarithm of the respective abundances knowing that the

trace-elements diffusion exhibits an log-normal distribution [5]. It is obvious that radioelements in soil show a more homogeneous distribution relative to the one in rocks which spread is more broaden. Therefore, in order to have a detailed study one must consider also the distribution of radioelement in rocks.



Fig. 2. Distribution of K, U, Th normalized for 677 rock samples and 1236 soil samples measured by MCA-Rad system.

The portable gamma-ray spectrometer gave an excellent answer in the determination of eU, eTh and K

The design and realisation of massive detectors mounted in aircraft was successfully finalized constructing the first airborne gamma-ray spectrometry system operating in Italy. In figure 3 is shown an example of the potentiality of such instrument: the map of total activity concentration deduced from AGRS_16L during a flight of about 2h covering an area of about 225 km².



Fig. 3. Distribution of total activity concentration in Elba Island deduced from AGRS 16L gamma-ray spectrometry.

ACKNOWLEDGEMENTS

The authors thanks Antongiovanni Mauro, Fabbri Andrea, Fabbri Paola, Morini Domenico, Pagotto Claudio and all the people which support us during this work.

- [2] C.R. Alvarez et al, LNL Annual Report, 2010.
- [3] G.P. Bezzon et al, LNL Annual Report, 2009.
- [4] A. Caciolli et al., to be published.
- [5] L.H. Ahrens, Geochimica et Cosmochimica Acts, 5 (1995) 49-73.

^[1] IAEA TECDOC No.1363 (2003) - Guidelines for radioelement mapping using gamma ray spectrometry data.