

# METHODOLOGY AND RESULTS OF A NATURAL RADIOACTIVITY ASSESSMENT IN A REGION OF CENTRAL ITALY

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## INTRODUCTION

The natural radioactivity survey being conducted in Umbria (800 000 inhabitants) is designed not only to evaluate mean doses associated with indoor exposure but above all to identify those areas and building features with the highest radiation exposure risk factor. Consequently, the sampling of houses must be particularly exact and the experimental measuring techniques particularly reliable at low exposure levels, too.

The study began by identifying five main geological areas within the region. A town which would be representative from an architectural point of view was then selected from within each area, the building materials used therein were analyzed (also by gamma spectrometry) and building material origins were identified. A certain number of houses built with these materials were then selected. Indoor and outdoor radiation measurements were taken with an ionization chamber and suitable data cards were compiled including information on the houses, inhabitants and respective home-utilization patterns. A gamma radiation exposure measurement campaign was conducted quarterly over an entire year using thermoluminescence detectors whereas a similar campaign to monitor radon with passive diffusion detectors was also begun and conducted according to the same schedule.

## METHODS FOR MEASURING EXTERNAL EXPOSURE AND RESULTS

In order to evaluate the exposure levels expected to be found in the houses, spectrometry measurements of building material specific activity were taken with a GeLi detector, after it had been suitably calibrated with radiation sources with energies ranging from 88 to 1836 keV in various-density matrices (from 1000 kg/m<sup>3</sup> to 1333 kg/m<sup>3</sup>). Analysis of the 35 samples enabled the range of building material specific activity levels to be identified. Table 1 summarizes measurement results. It should be noted that the specific activity values reported are generally higher for radionuclides of the Th-232 families than for those of the U-238 families. A rough computer program was designed for planning the passive gamma exposure measurements. This program, starting from the spectrometry results, calculates expected exposure. Calculation hypotheses were fairly simple, i.e., that a source be formed by two walls of the materials identified, a wall in hollow brick, the ceiling and the floor in hollow brick and cement components, the missing wall being equated to the openings due to windows and doors. Furthermore, each surface was approximated to an infinite half space, in which radionuclides are distributed uniformly and in secular equilibrium. Air attenuation was assumed to be negligible and the build-up factor in air less than 5% for the energy range studied.

Table 1 - Specific Activity Range for Building Materials

Material	Specific Activity ( $\text{Bq}\cdot\text{kg}^{-1}$ )		
	K-40	Bi-214	Pb-212
TUFF	(1468 $\pm$ 48) $\div$ (1952 $\pm$ 50)	(136 $\pm$ 8) $\div$ (243 $\pm$ 8)	(468 $\pm$ 8) $\div$ (541 $\pm$ 9)
CEMENT PZ	(607 $\pm$ 24) $\div$ (667 $\pm$ 26)	(72 $\pm$ 4) $\div$ (81 $\pm$ 4)	(164 $\pm$ 4) $\div$ (172 $\pm$ 4)
CEMENT PTL	(253 $\pm$ 17) $\div$ (270 $\pm$ 18)	(17 $\pm$ 2)	(27 $\pm$ 2) $\div$ (29 $\pm$ 2)
GRAVEL	(100 $\pm$ 13) $\div$ (134 $\pm$ 12)	(11 $\pm$ 2) $\div$ (14 $\pm$ 2)	(14 $\pm$ 2) $\div$ (16 $\pm$ 2)
SAND	(393 $\pm$ 19) $\div$ (491 $\pm$ 18)	(8 $\pm$ 2) $\div$ (11 $\pm$ 2)	(13 $\pm$ 2) $\div$ (20 $\pm$ 2)
BRICKS	(550 $\pm$ 21) $\div$ (883 $\pm$ 31)	(29 $\pm$ 3) $\div$ (81 $\pm$ 4)	(49 $\pm$ 3) $\div$ (148 $\pm$ 4)
POZZOLANA	1888 $\pm$ 46	238 $\pm$ 8	481 $\pm$ 8

The characteristic of  $\text{LiF}$ ,  $\text{CaF}_2:\text{Dy}$  and  $\text{MgB}_4\text{O}_7$  thermoluminescence detectors were then studied. The minimum exposure detectable was seen to vary from 0.1 mR to 5.0 mR depending on the type of detector used. Linearity, for X and  $\gamma$  energy from 27 keV to 1250 keV, was checked in the exposure range between a few mR and 3 R for  $\text{LiF}$  and  $\text{CaF}_2:\text{Dy}$ . The coefficient of correlation was seen to be good. Reproducibility, within the exposure range studied, was seen to be  $\pm 4\%$  for commercial TL dosimeters (TLD-100, TLD-200 and TLD-700) but considerably less for  $\text{MgB}_4\text{O}_7$  dosimeters. Fading was studied in TLD-100s and TLD-200s suitably shielded for different periods of time and at mean ambient temperatures of 20 °C and 30 °C and mean relative humidities of 65% and 50%, respectively. Suitable statistic check tests showed no significant fading in  $\text{LiF}$  detectors over the entire period considered whereas  $\text{CaF}_2:\text{Dy}$  detectors began to show significant variations (20%) from initial values after 30 days. Lastly, detector energy dependence was studied with different types of packaging.

To avoid the necessity of coupling a filter system to detectors, with the consequential anisotropy and related calibration difficulties, it was finally decided to select the least energy-dependent detector, i.e., the  $\text{LiF}$  (TLD-100) detector packaged in a black paper envelope 67 mg/cm<sup>2</sup> thick. Differences between ambient temperatures inside and outside the package were evaluated between 26 °C and 51 °C with a digital thermometer having copper and constantan probes. The maximum difference was found to be  $(1.6 \pm 0.1)$  °C. Beta contribution to exposure, considering source spectrum and package characteristics, was seen to be approximately 2% of the total.

Dosimeters thus selected were paired and placed both in one bedroom and in the most lived-in room of each home selected. Table 2 reports the mean exposure rates measured over the year in some sampling homes grouped according to building materials, without subtracting cosmic ray contribution (that has been measured at the centre of Lake Trasimeno).

Table 2 - Indoor Exposure Rates in Three Different Towns

Towns	Building Material	Exposure Rates ( $\mu\text{R}\cdot\text{h}^{-1}$ )
Foligno	Stone	10.7 $\pm$ 1.1
	Hollow bricks and Cement	20.9 $\pm$ 2.1
	Red Tuff with Black Scoriae	30.1 $\pm$ 3.1
Orvieto	Local Tuff and Pozzolana	61.3 $\pm$ 6.1
	Local Tuff and Cement	55.3 $\pm$ 5.5
	Local Tuff	60.0 $\pm$ 6.0
Todi	Stone	14.3 $\pm$ 1.4
	Stone and Red Tuff with	
	Black Scoriae	18.1 $\pm$ 1.8

## METHODS FOR MEASURING INTERNAL EXPOSURE AND RESULTS

In order to evaluate internal exposure for inhabitants, radon and radon daughter concentrations must be measured in the houses. This may be done with integrated and/or prompt measurements taken with an active or passive device. Integrated measurements are essential to assess the mean concentrations because of the well-known radon dependence on ventilation and atmospheric conditions. Conversely, prompt measurements give an idea of radon and radon daughter concentration variations.

As integrating and passive devices, Solid State Nuclear Track Detectors were chosen for their practicality, economy, and specific qualities: sensitivity to alpha particles without being influenced by gamma radiation or electrons, and for their nonfading and stability of recorded information even after reading<sup>(1)</sup>.

Initially, considerable laboratory-based experimental work was done with CR-39 and LR-115(Kodak-Pathé). For the indoor survey, the first type was preferred because it has no energy threshold. After some tests, CR-39 about 600  $\mu\text{m}$  thick from American Acrylics was chosen. Etching conditions were optimized paying particular attention to etching times, weight losses and thickness reductions being studied with both NaOH (at 70 °C) and KOH (at 60 °C) at various times. The resultant data were compared with the theoretical curves of alpha penetration in CR-39 with varying energy.

Particular CR-39 diffusion dosimeters were designed and built in the laboratory after an analysis of specific literature and some preliminary experiments performed fastening small cups containing CR-39 to some walls of the laboratory rooms. At this stage, tracks are still being counted with an optical microscope, however, automation of this step has also been studied<sup>(2)</sup>.

To test the detection method, some diffusion dosimeters were distributed in some of the same survey homes in the rooms with TLDs. Preliminary calibration of these diffusion dosimeters was performed with a radon chamber at the ENEA Centre of Casaccia. Table 3 shows the mean radon concentrations for some types of building materials

Table 3 - Mean Radon Concentration in Homes Built with Different Materials

Building Material	Mean radon concentration ( $\text{Bq} \cdot \text{m}^{-3}$ )
Stone	$46 \pm 12$
Tuff	$73 \pm 19$
Stone + Tuff	$70 \pm 29$
Tuff + Cement	$90 \pm 30$

for the spring-summer survey (90 days). Background has been subtracted. Final calibration at the National Radiological Protection Board Laboratories is planned for the near future.

Lastly, the Harshaw Radon Daughter Analyzer was tested as a prompt and active device in some selected houses, under different ventilation conditions, and within this framework satisfactory agreement was found with passive detector results. Systematic measurements with this equipment would provide estimates of radon and radon daughter concentrations in terms of Working Levels thereby enabling internal exposure variations connected with season, ventilation, atmospheric and living conditions to be evaluated.

#### CONCLUSIONS

It is most certainly too early to draw any definite conclusions from the measurements taken so far. However, a correlation between building materials used and indoor external exposure levels does seem feasible at this stage.

Annual doses of radiation absorbed by the inhabitants living in tuff rock homes in southern Umbria are in the region of 4.2 mGy, cosmic radiation included, assuming a home occupancy factor of 0.8. Adding to this value the mean individual absorbed dose rate of approximately 0.7 mGy/y due to outdoor gamma exposure from cosmic rays and terrestrial radiation in the same area, provides<sup>(3)</sup> an annual effective dose equivalent from gamma radiation of approximately 3.4 mSv.

Conversely, preliminary results concerning internal exposure based on only one three-month campaign and on the measurements of radon concentration alone, do not enable forecasts of expectable internal radiation doses to be made.

#### REFERENCES

- 1) i.e., see Proceedings of the 11<sup>th</sup> International Conference on Solid State Nuclear Track Detectors. Bristol (U.K.), September 1981.
- 2) See G.Campos Venuti et al. in Proceedings of the 12<sup>th</sup> International Conference on Solid State Nuclear Track Detectors, Acapulco (Mexico). September 1983.
- 3) Ionizing Radiations: Sources and Biological Effects. United Nations Scientific Committee, 1982 Report.