Building Materials as a Source of Gamma Radiation and Radon Concentration: Tests and Improvement of Experimental Methods

C. Nuccetelli[1], C. Bolzan[2], G. Grisanti[1] and S. Risica[1]

[1] Physics Laboratory, National Institute of Health,
Viale Regina Elena 299, 00161 Rome, Italy
[2] Fellow of the Physics Laboratory, National Institute of Health

INTRODUCTION

The Italian national survey on natural radioactivity in dwellings showed that in the central and southern part of Italy the widespread use of building materials of volcanic origin (tuff and pozzolana) has two important consequences: high levels of radon concentration indoors and the presence of elevated γ dose rates (1,2,3). The significant role of some building materials as a source of natural radiation has led the authors to develop a method to characterise them in buildings. In fact, preventive assessment of building materials to decide whether or not they are appropriate for use in construction can be relatively simple in the laboratory with suitable procedures of gamma spectroscopy. Indeed, some computational models (4,5,6,7) have been formulated to calculate the indoor gamma dose rate from the activity concentration of the building materials. Taking a sample from an existing building for an accurate laboratory analysis, however, is clearly not practical.

In order to overcome these problems, the authors developed a new method to evaluate the characteristics of building materials as gamma ray and radon sources in the field (8,9). This was done combining the results of *in situ* measurements (gamma spectrometry (10) and gamma dose rate assessment) and the computational estimates of the Markkanen room model (4). The method makes it possible to assess activity concentration in the walls of a room without having to measure a sample in the laboratory by gamma spectrometry. Shown to work well in a real situation, other experimental tests and a sensitivity analysis were performed to assess the uncertainties in the estimates. In this paper, the results of these tests are presented and an evaluation of the uncertainties discussed. The method also estimates the radon exhalation from walls.

MATERIALS AND METHODS

The method developed by the authors consists of a combination of experimental results and an elaboration with a mathematical model using suitable hypotheses. The logical scheme underlying the method is shown in fig.1.

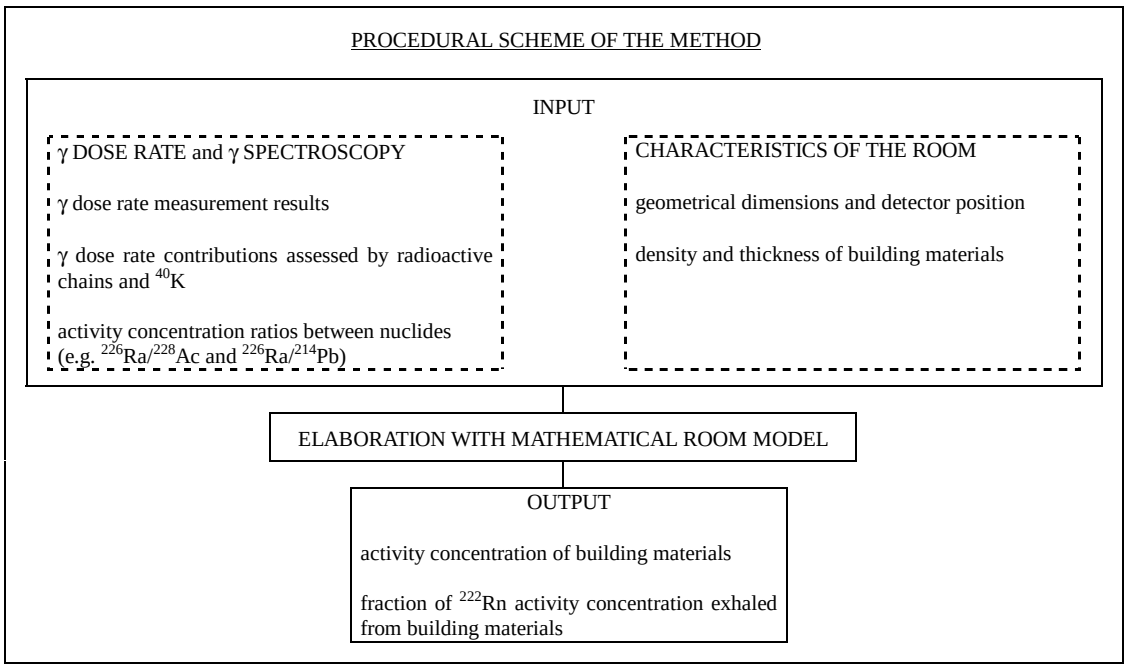

Fig.1 Logical scheme of the integrated method

The experimental measurements necessary for this integrated approach are: absorbed dose rate in air due to gamma irradiation (*gamma dose rate*) and gamma spectra. Gamma dose rate was measured with a Reuter Stokes high pressure ionisation chamber (RS) or a plastic 3" x 3" scintillator (Automess 6150ADb with reading device 6150AD6). Gamma spectra were performed with an EG&G coaxial high purity germanium spectrometer (HPGe), 26% efficiency and 1.73 keV resolution. The latter technique was applied in indoor environments, using the methodology proposed by Miller and Beck (8,9,10), which makes it possible to determine the contributions of U (by means of ^{226}Ra contribution), Th (by means of ^{228}Ac contribution) and ^{40}K to the gamma dose rate. A good agreement between the gamma dose rate assessed by means of the spectroscopic peak analysis and the one measured as described above proves the applicability of the method in an indoor environment. The method has already been applied in 7 dwellings and 1 office in Rome and its applicability can be seen in columns 3 and 4 of Table 1 (8). Table 1 also reports the results obtained in two different test rooms; more details about these will be given in the following.

dwelling	building material	dose rate HPGe nGy h^{-1}	dose rate RS* nGy h^{-1}	dose rate % contribution U	Th	K	activity concentration ratio ^{226}Ra/^{228}Ac	activity concentration ratio ^{226}Ra/^{214}Pb
B	tuff	401 ± 12	398 ± 13	22.8	62.0	15.2	0.67 ± 0.08	1.31 ± 0.15
D	tuff	237 ± 10	258 ± 11	25.2	57.4	17.4	0.76 ± 0.08	1.28 ± 0.14
F	tuff	355 ± 10	340 ± 12	24.0	62.0	14.0	0.66 ± 0.07	1.23 ± 0.15
G	tuff	335 ± 12	323 ± 12	23.3	64.5	12.2	0.65 ± 0.10	1.31 ± 0.17
office	tuff	288 ± 11	294 ± 12	25.3	58.4	16.4	0.78 ± 0.08	1.31 ± 0.14
test room 1	tuff	246 ± 10	255 ± 11	19.7	61.0	19.3	0.60 ± 0.09	1.31 ± 0.18
test room 2	tuff	310 ± 12	330 ± 12	22.9	55.9	21.1	0.73 ± 0.10	1.13 ± 0.13
A	cement	161 ± 10	155 ± 10	25.4	52.6	21.8	0.62 ± 0.10	1.00 ± 0.13
C	cement	278 ± 11	253 ± 11	25.7	57.6	16.7	0.65 ± 0.08	1.07 ± 0.12
E	cement	300 ± 12	271 ± 11	24.5	60.0	15.5	0.62 ± 0.07	1.06 ± 0.12

* indoor cosmic ray contribution, estimated as 25 nGy h^{-1}, is subtracted.

Table 1 Results of the elaboration from γ spectra measurements. The fourth column shows the γ dose rates measured with Reuter Stokes (RS) chamber.

The method devised by the authors makes use, first of all, of a further application of indoor γ spectroscopy, aimed at estimating the ratios of activity concentrations between natural radionuclides, whether or not they pertain to the same radioactive chain. This is done by applying a *normalising* method to full absorption peak counts (8,9). This methodology can be applied in specific situations in which the outdoor γ flux dependence on activity concentration for uniform distribution of radioactivity (11) can be used in indoor environment. This condition is fulfilled in dwellings with thick walls (0.3 m - 0.8 m). The estimate of activity concentration ratios is very useful given that *in field* gamma spectroscopy, particularly indoors, cannot give quantitative information on activity concentration, in this case of the building material, as it is impossible to carry out an efficiency calibration in any particular indoor geometry.

The second step of the proposed method uses a room model (4) in an inverted sense. Indeed, room models are set up to assess specific gamma dose rates (Gy h^{-1} per Bq kg^{-1}) of the main natural radionuclides in a point of a room using, as input parameters, the dimensions of the room, the thickness and density of the walls, floor and ceilings, and the co-ordinates of the considered point. In this way, indoor gamma dose rate at that point can be assessed in all situations. In our case, input data are: structural and geometrical parameters of the room considered, indoor gamma dose rate measured with a gamma dose rate detector, percentage contribution of natural radionuclides to dose rate, and the activity concentration ratio of ^{226}Ra to ^{228}Ac, evaluated by means of gamma spectroscopy as detailed above. With the *inverted* use of the model, output results are activity concentrations of ^{222}Rn decay products (^{222}Rndp), ^{228}Ac chain and ^{40}K in building materials. Moreover, by means

of the ^{226}Ra to ^{228}Ac ratio, the activity concentration of ^{226}Ra can be calculated, as can that of exhaled ^{222}Rn (in Bq kg^{-1}), once the activity concentration of radon decay products is subtracted.

Concerning the mathematical code, different room models make different choices concerning the radionuclides to be considered in the natural chains and their gamma photo-peaks. In principle, the absorbed dose rate in air due to building material should be calculated by means of the chosen room model, considering the gamma line of ^{40}K and every gamma line of the ^{238}U and ^{232}Th natural chains. However, for practical assessment, different room models consider only some averaged gamma photo-peaks of the natural chains and the ^{40}K. The average gamma energy is generally calculated by using the emission probability as a weighting factor and, therefore, effective emission probability is the sum of the single ones.

In particular, the Markkanen model uses one averaged gamma line for the ^{238}U chain (averaged energy = 810 keV and emission probability = 2.12%, based on the 23 most important lines of ^{214}Pb and ^{214}Bi) and two gamma lines for the ^{232}Th chain (587 keV with 2.05% based on the 25 most important lines of ^{228}Ac, ^{224}Ra, ^{212}Bi, ^{212}Pb and ^{208}Tl; 2615 keV with 0.356%). This is possible because the energy absorption and the attenuation coefficients are rather smooth functions of energy in the range of (240-1800) keV. Only the intensive 2615 keV gamma line of the ^{232}Th chain is treated separately because this single line causes over 40% of the thorium chain dose rate.

In the previous studies (8,9) the authors had already slightly modified the Markkanen model to make the list of energy peaks in the ^{232}Th chain ñ fundamental in the considered building materials ñ more complete. The new averaged gamma line is 593 keV with 2.19%.

For this study, the absorbed dose rate in air was calculated for each photo-peak of the ^{238}U and ^{232}Th chain nuclides, with energy higher than 70 keV and absolute emission probability more than 1%. In particular the 186 keV line of ^{226}Ra was taken into account. For the photo-peaks with emission probability less than 1%, two effective gamma lines were calculated for energy below and above 1 MeV. Then the overall absorbed dose rate in air, considering also the gamma line of ^{40}K, was calculated. The differences among the specific absorbed dose rates (nGy h^{-1} per Bq kg^{-1}) calculated with the different methods mentioned above in the Markkanen model reference room (5m x 4m x 2.8m; wall, floor and ceiling thickness = 0.2 m and density = 2320 kg m^{-3}) are shown in Table 2. The more accurate method, with respect to the original Markkanen method, produces specific dose rates that are slightly different (-3% for ^{222}Rn decay products and +7% for the ^{232}Th chain). For routine in field assessment, the much longer computational time required for this accurate procedure does not seem justified; research activities, however, may need high accuracy in the final outcomes.

Mathematical code	Specific dose rate (nGy per Bq kg^{-1})		
	^{238}U	^{232}Th	^{40}K
4 gamma lines (4)	0.905	1.056	0.076
modified 4 gamma lines (8,9)	0.905	1.132	0.076
this work method	0.014 (^{226}Ra) 0.876 (^{222}Rndp)	1.130	0.076

Table 2. Comparison of the specific dose rates calculated with three different methods.

EXPERIMENTAL TESTS AND UNCERTAINTIES ANALYSIS

This combined approach had already been experimentally verified, as regards the estimate of activity concentrations in the walls, in one test house of which the building material was available for measurement by gamma spectroscopy in the laboratory using a coaxial HPGe gamma spectrometer (38% efficiency, 1.95 keV resolution) in Marinelli geometry (8,9). A direct test of ^{222}Rn exhalation was not feasible. However, the values obtained with the method in different environments were considered coherent with the values of radon concentration in air measured in the same environments by means of passive detectors (8). Concerning activity concentration, the agreement between laboratory results and those of the new method was excellent for the ^{232}Th and ^{238}U chains, whereas a difference of less than 10% was obtained for ^{40}K, as shown in the first line of Table 2. The agreement was judged acceptable, due also to the fact that the uncertainty of the method had not yet been evaluated. In any case, at least one more test was deemed necessary to validate the method. For this purpose, another experimental test of the method was carried out in a second test house of which the building material was available for measurement by gamma spectroscopy in laboratory.

The room (6 x 5.1 x 2.3 m^3) was built with tuff of density 1300 kg m^{-3}. Wall thickness was 0.3 m and floor and ceilings were made of cement. *In situ* measurements were made in one corner, 1 meter from the corner

at 0.50 m height, with the devices described earlier. A sample of the tuff was measured in laboratory by means of gamma spectra. Results of activity concentrations of ^{228}Ac (equivalent to ^{232}Th), ^{226}Ra and ^{40}K measured and calculated with the proposed method are compared in Table 3. The agreement of the results is surprisingly good also in the second test room for the ^{232}Th and ^{238}U chains, and once again the value of ^{40}K was found to be underestimated by less than 10%. This outcome could indicate a systematic methodology error and must be investigated. Concerning the activity concentration ratio ^{226}Ra/^{214}Pb, which estimates in field the exhalation of ^{222}Rn from building materials, the agreement between the partially independent outcomes obtained with *in situ* γ spectroscopy (see last column in Table 1) and those obtained with the integrated method (in Table 3 $C_{Ra\text{-}226}/C_{Rn\text{-}222dp}$) is within the uncertainty range, as discussed in the following.

Concerning the sources of uncertainties of the proposed method, they can be divided into two categories: experimental uncertainties and uncertainties originating from the hypotheses concerning the applicability of the indoor spectroscopy method.

The first category could include: statistical error of photo-peak counting, uncertainty of the efficiency calibration for point sources of the HPGe detector (11), uncertainty in the measurement of total γ dose rate with dose rate meter and error relating to characteristics of the measured room. Usually the spectra were cumulated for a long enough time to assure statistical error less than 1% for the most representative peaks of the radioactive chains and of ^{40}K; on the other hand, the uncertainty in the efficiency calibration can be estimated at around 3%. The error in the results of the dose rate meter (Reuter Stokes or Automess plastic scintillator) was always less than 3 % in the intensive fields of γ radiation determined by the studied radioactive building materials. Finally, the measurement errors in room geometrical information can be considered negligible. Density ρ of the building materials is also an important parameter for computation. The variations due to an erroneous estimate of density can be important (9) but can be made negligible when realistic figures of ρ are taken into account for known materials. On the whole, the experimental contributions to the overall uncertainty of the method should not exceed 5%.

Method of evaluation	Test room 1				Test room 2			
	$C_{Th\text{-}232}$ Bq kg^{-1}	$C_{Ra\text{-}226}$ Bq kg^{-1}	$C_{Rn\text{-}222dp}$ Bq kg^{-1}	$C_{K\text{-}40}$ Bq kg^{-1}	$C_{Th\text{-}232}$ Bq kg^{-1}	$C_{Ra\text{-}226}$ Bq kg^{-1}	$C_{Rn\text{-}222dp}$ Bq kg^{-1}	$C_{K\text{-}40}$ Bq kg^{-1}
integrated method	190	110	80	900	350	260	190	2040
gamma spectrometry on sample*	187 ± 4	109 ± 2		956 ± 13	339 ± 8	254 ± 14		2166 ± 35

* concentration with total uncertainty (1 σ).

Table 3. Activity concentration of ^{232}Th, ^{226}Ra, ^{222}Rndp and ^{40}K in the walls of a room measured by means of gamma spectrometry in laboratory and the proposed method.

A much more subtle procedure is needed to estimate to what extent the hypotheses concerning applicability have repercussions on the overall uncertainty of the final results. First of all, one has to estimate when the agreement between the γ dose rate build-up with spectroscopy and the γ dose rate measured with the detector can be considered good enough to calculate confidently the dose rate contribution of the radioactive chains and ^{40}K (10). The authors decided to accept a spectroscopy value within ± 10% of the detector value. Another source of methodological uncertainty is the use of outdoor expressions for attenuated photon flux to estimate the ratios between the indoor activity concentrations of the radionuclides. In this case, the check on secular equilibrium of some nuclides of the same chain (e.g. ^{214}Pb and ^{214}Bi, ^{212}Pb and ^{212}Bi and ^{208}Tl) assures a sort of *self-consistency* for the method. Normally, the errors in activity concentration ratios are around 10-15 % and this value is considered the limit for the accuracy of the equilibrium estimates. Analysis of the most relevant sources of uncertainty, experimental and non, makes it possible to estimate the overall uncertainty in the activity concentration values and the fraction of exhaled ^{222}Rn as between 15 and 20%. This accuracy can be considered very good for an in field assessment of the radioactive characteristic of building materials.

CONCLUSIONS

The integrated method presented, a combination of some experimental results and elaboration with a mathematical model, makes it possible to assess the radioactivity content of building materials in field, i.e. without having to draw a sample to be measured in laboratory. Therefore the method, successfully tested in two rooms, can be used routinely to estimate the activity concentration and the fraction of exhaled ^{222}Rn with an overall uncertainty between 15 and 20%. The ^{232}Th activity content and the ^{222}Rn exhalation characteristic, considered together, can also provide an indirect but very useful indication of the presence of ^{220}Rn (8).

Future activities will involve verifying this method in rooms built with materials other than tuff (cement

and bricks, concrete, ...); moreover, analysis of the effect of net of rooms around the investigated room will be undertaken. The study of the observed systematic underestimate of the ^{40}K activity concentration will also be probed.

ACKNOWLEDGEMENTS

This study was carried out within the framework of Research Program PR-22/IS, Subproject 1, jointly financed by the Ministry of the Environment (Rome, Italy) and the Italian National Institute of Health.

REFERENCES

1. F.Bochicchio, G.Campos Venuti, C.Nuccetelli, S.Piermattei, S.Risica, L.Tommasino and G.Torri, *Results of the Representative Italian National Survey on Radon Indoors*. Health Phys. 71(5), 741-748 (1996).
2. F.Bochicchio, G.Campos Venuti, C.Nuccetelli, S.Risica and F.Tancredi, *Indoor measurements of ^{220}Rn and ^{222}Rn and their decay products in a Mediterranean climate area*. Environ. Int. 22(S1), S633-S639 (1996).
3. F.Bochicchio, G.Campos Venuti, F.Felici, A.Grisanti, G.Grisanti, S.Kalita, G.Moroni, C.Nuccetelli, S.Risica and F.Tancredi, *Characterisation of some parameters affecting the radon exposure of the population*. Radiat. Prot. Dosim. 56(1-4), 137-140 (1994).
4. M.Markkanen, *Radiation Dose Assessment for Materials with Elevated Natural Radioactivity*. Report STOK-STO 32, Finnish Centre for Radiation and Nuclear Safety, Helsinki, Finland (1995).
5. R.Mustonen, *Methods for Evaluation of Radiation from Building Materials*. Radiat. Prot. Dosim. 7(1-4), 235-238 (1984)
6. E.Stranden, *Radioactivity of building materials and the gamma radiation in dwellings*. Phys. Med. Biol. 24, 921-930 (1979).
7. L.Koblinger, *Calculation of exposure rate from gamma sources in walls of buildings*. Health Phys. 34, 459-463 (1978).
8. C.Nuccetelli and C.Bolzan, *In situ gamma spectroscopy to characterise building materials as radon and thoron sources*. In Proceedings of: Radon in the living environment, 19-23 April 1999, Athens, Greece (1999), in press.
9. S.Risica, C.Bolzan and C.Nuccetelli, *Radioactivity in building materials: experimental methods, calculations and an overview of the Italian situation*. In Proceeding of: Radon in the Living Environment, 19-23 April 1999, Athens, Greece (1999), in press.
10. K.M.Miller and H.L.Beck, *Indoor gamma and cosmic ray exposure rate measurements using a Ge spectrometer and pressurised ionisation chamber*. Radiat. Prot. Dosim. 7 (1-4), 185-189 (1984).
11. H.L.Beck, J.De Campo and C.Gogolak, *In situ Ge (Li) and NaI (Tl) gamma-ray spectrometry*. U.S. Atomic Energy Commission Report HASL-258, New York, USA (1972).