NATURAL RADIOACTIVITY OF BUILDING MATERIALS COMING FROM A VOLCANIC REGION

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ABSTRACT

The results of a program for the characterization of radioactivity of various soils and building materials of the Campania region have been here updated and discussed. This characterization has carried out by performing gamma ray spectrometry for the determination of the total activity concentration in the analysed samples, and alpha ray spectrometry of ionised descendants of radon, collected in a electrostatic cell, to measure the emanated fraction of this radioactive gas. Materials of different characteristic and origin have bean studied and results, confirming the relative high radioactivity of he materials coming from this region, underline many differences respect their radioactivity content and their capacity to generate radon, as well.

INTRODUCTION

It is well known that building materials contribute to the indoor radon levels, mainly at the upper floors of the houses. In the Campania region, the presence of uranium and thorium traces in the pyroclastic materials, largely used in the building construction, makes this contribute particularly meaningful (Gialanella, 1988; Sabbarese, 1993). To better understand and quantify such a contribute, we began, some years ago, the study of the radioactivity of local materials. In this work more recent measurements on a larger number of materials are reported; in particular the activity concentration for ^{238}U, ^{232}Th and ^{40}K and radon emanation coefficients have been measured. In the study also some commercial materials have been included. Moreover, the radon emanation coefficients have been measured on samples of soil, which in any case is the main source of the indoor radon, responsible of more than one half of the total exposition to the ionising radiations. The long living parents of this radionuclide are worldwide present and its characteristic makes possible its migration in the soil and from soil to the enclosed places, where it can reach concentrations depending on the source strength and on the frequency of the air exchange. It is well known that not the full amount of radon produced in the soil or inside a material can reach the external air: in fact only a fraction of radon atoms acquires enouigh kinetic energy to leave the grain of the material where it has been generated and to reach the empty space in the porous materials. This process is said *emanation* and the emanated radon fraction is the *emanation coefficient*. The determination of this parameter is one of the most important elements for the study of the radon flux toward the indoor air (Nazzaroff, 1988).

In particular, the subsoil of the territory of the Campania is rich in materials coming from the eruptions of several volcanic systems of the region. Many of these materials have been largely used in the past as building materials, and now they are still used and can be considered responsible not only for the radon release but also for the external gamma ray exposition. Consequently the knowledge of the characteristics of the building materials is a good starting point for the evaluation of the expected radon concentrations and hence for the planning of a survey.

With this aim, in the past years we started to make measurements on samples of such kind of materials coming from the local quarries (Roca, 2000). Now the preliminary results of a more systematic study are presented, where also some commercial materials and soils have been included.

MATERIALS SELECTION

After the analysis of the most interesting matrices to study, a final choice has been made of four groups of materials: soil samples, man-made and natural building materials and various material coming from local quarries. Soil samples from the most superficial layer have been took in tree different pedologic settings:

Arciano (Arciano 1, Arciano 2): from vesuvian volcanic soils, labelled as *Typic Hapludand*.

Castelvolturno (Castelvolturno 1, Castelvolturno 2, Castelvolturno 3): generally coming from typical environment: coast line and floodplain : in the last one two samples from a *Cromic Udic Haplustert* (Castelvolturno 1) e from an *Oxyaquic Xerofluvent* (Castelvolturno 2) have been took; in the first one has been took the sample Castelvolturno 3, from a *Typic Xeropsamment*.

Somma-Vesuvio (Somma Vesuviana) from a volcanic soil labelled as *Vitrandic Xerorthent*.

The man-made materials are the most used for the walls building. The natural materials are used in the houses structures building (tuff) and in the decoration work (lava).

. Regarding tuff, samples originating from different eruptions and different crystallisation ways have been selected. Finally, since in Campania a large number of quarries has been used in the past years (350) and many of these are today still running, a selection has been made to sampling the most widely used materials.

METHODS

Samples have been dried in two steps: at 60°C, for 36 hours, and at 110°C, for 12 hours, to eliminate the humidity gradually, without loss of any radioisotope. All samples have been minced to less than 1 mm and their density has been made close to that of the reference sources. After this treatment, the samples have been sealed in metallic boxes, whose cover has been soldered to avoid leak of radon during the waiting for reaching the radioactive equilibrium. This waiting time, has been of 30 days. After that, the elemental analysis has been carried out by counting the emitted gamma-rays by a 70% efficiency and 1.8 keV at 1,3 MeV resolution HP Ge. detector. Each measurement has been at least 2 hours long, to reach a statistical error less than 5% on the peak area. The efficiency has been measured in the used geometries and for different densities.

RESULTS I: RADIOACTIVITY IN SOLIDS

The considered natural radionuclides have been: ^{232}Th, ^{226}Ra, in addition to ^{40}K , which is also present practically in all minerals coming from the crust of the earth. For calculating the ^{232}Th activity, we measured activity of its daughters ^{212}Bi, ^{208}Tl and ^{228}Ac, from the γ lines at 727 keV, 860 keV e 911 keV respectively . The ^{226}Ra activity was calculated measuring the activity of the daughters ^{214}Pb and ^{214}Bi from the lines at 295 keV, 352 keV and 609 keV. All γ lines where selected on the basis of their intensity. Final values of the activities of ^{232}Th and of ^{226}Ra where calculated with a weighted average of the values obtained from the respective descendants.

Specific activities of the analysed radionuclides are shown in tab 1, 2, 3 e 4, where the values corresponding respectively to soils, man-made and volcanic building materials and various materials coming from quarries are considered

A first comment about these results is the correspondence between the soil characteristics and natural radioisotopes distribution. In fact the samples coming from Arciano's site shown the main content of radioactivity, according with the volcanic origin of the pedogenetic substrate and with their content of organic substance, which is higher respect to other analysed samples. On the opposite side, we find the sample of a sandy soil, Castelvolturno 3, characterised by a low content of organic substance.

Moreover we can see a marked difference between the activity concentrations of the natural respect to the man-made materials. The first ones result also of a factor then higher respect to second ones. In any case the radioisotope present with higher activity in any kind of material is ^{40}K, with an activity reaching more than 10 kBq/kg in the case of the lavic stone.

This is an interesting result, mainly in the case of the building materials, being directly connected to gamma exposition in homes and work places.

Soils	Specific activity (Bq/kg)					
	Th232	***Err Th***	***Ra226***	***Err Ra***	***K40***	***Err K***
Arciano 1	119	14	262	3	1553	60
Arciano2	83	7	319	8	1358	54
Castelvolturno 1	75	4	55	3	636	25
Castelvolturno 2	56	4	53	3	739	30
Castelvolturno 3	17	2	13	1	353	14
Somma Vesuviana	40	4	142	4	768	35

Table1. Specific activity of natural radionuclides measured in soils.

Man-made building naterials	Specific activity (Bq/kg)					
	Th232	***Err Th***	***Ra226***	***Err Ra***	***K40***	***Err K***
Sand	10	2	15	1	344	21
Bricks	20	2	14	1	282	15
Gypsum brick	5	1	7	1	56	8
Tuff brick	4	2	26	2	55	9
Concrete brick	3	1	17	1	58	8
Sand	4	1	16	1	34	6
Calce	12	3	17	2	84	14
Siporex	10	2	7	1	192	16
Cement	28	3	24	2	445	26

Table 2. Specific activity of natural radionuclides measured in man-made building materials.

Volcanic origin building materials	Specific activity (Bq/kg)					
	Th232	***Err Th***	***Ra226***	***Err Ra***	***K40***	***Err K***
Grey-tuff - Avellino	106	6	79	3	1387	52
Grey-tuff (Caserta)	102	6	90	4	1945	70
Neapolitan yellow tuff	86	6	73	3	2031	71
Yellow tuff – volcano La pietra	99	5	68	3	1589	58
Yellow tuff (PSerra c)	17	2	12	1	453	23
Yellow tuff (PSerra f)	80	5	67	9	1414	54
Grey tuff (Tuoro)	121	6	107	4	1740	61
Green tuff - Ischia	93	5	61	3	1625	61
Pumices Avellino 1	229	8	172	5	1880	67
Pumices Avellino 2	170	7	141	5	2074	71
Vesuviana lava	93	8	438	11	2163	83

Table 3. Specific activity of natural radionuclides measured in natural building materials

Quarry materials	Specific activity (Bq/kg)					
	Th232	*Err Th*	*Ra226*	*Err Ra*	*K40*	*Err K*
CARS3TG	111	4	97	2	1964	33
POZZOL03	1048	11	713	6	6846	57
MARMOA15	33	4	79	2	634	19
LAVIC STONE	852	9	510	4	11286	62
SABCAMP	310	7	231	3	4926	48
CALCDRAG	27	2	28	1	160	9
CEMENMAD	402	9	380	5	6593	60
LIMOCC02	190	7	284	4	4355	50
BAUXITE	181	4	83	2	129	8
SAND C	10	1	15	1	255	6
IGNIMBR	44	3	38	2	640	19

Table 4. Specific activity of natural radionuclides measured in various quarry materials.

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RESULTS II: RADON EMANATION MEASUREMENTS

The emanation coefficient of ^{222}Rn in samples of soil and in different kinds of tuff has been obtained by the ratio between the activity of emanated radon fraction measured in a collection chamber, after that the equilibrium of the radon respect to ^{226}Ra has been reached, and the activity of ^{226}Ra in the sample, obtained by γ-ray spectrometry, as already said. The emanated radon activity has been measured by α ray spectrometry of the ionised daughters of the ^{222}Rn. The α-line at 7687 keV of the ^{214}Po has been used, because the interference of the line of the direct daughter of ^{222}Rn, the ^{218}Po, at 6003 keV, with the line of ^{212}Bi at 6090 keV. The values obtained for each sample are reported in tab. 5 and in tab.6 for the soils and for the volcanic origin materials, respectively. The more evident result is that yellow tuff and pumices release a quantity of radon which is 5-6 times higher respect to grey tuff, characteristic of the inner countries of the Campania and respect to green tuff of the Ischia isle.

Sample	Emanation coefficient
Arciano 1	0,43 $\pm$ 0,06
Arciano 2	0,32 $\pm$ 0,05
Castelvolturno 1	0,13 $\pm$ 0,04
Castelvolturno 2	0,16 $\pm$ 0,05
Castelvolturno 3	0,25 $\pm$ 0,09

Somma Vesuviana	0,05 ± 0,01

Tanle 5 Radon emanation coefficients in soils samples.

Volcanic	Emanation coefficient
Neapolitan Yellow tuff	0.73 ± 0.11
Yellow tuff - Vulcano La Pietra	0.57 ± 0.09
Yellow tuff - Monte Epomeo, Ischia	0.16 ± 0.02
Grey tuff – Caserta	0.12 ± 0.02
Grey tuff – Avellino	0.08 ± 0.01
Grey tuff – Tuoro (Caserta)	0.14 ± 0.02
Pumices	0,63 ± 0,11

Tabella 6. Radon emanation coefficients in volcanic materials

CONCLUSIONS

At present the program is still in progress, since the lot of materials under study and the long waiting time necessary to reach the equilibrium between ^{226}Ra and daughters both for activity and for emanation measurements; in particular the last ones have been carried out only on soils and on volcanic materials. However, also these temporary results are very interesting, showing a wide dispersion among the characteristic of the various materials and the occurrence of high value of radium activity and emanation coefficients. Observing the data, it is easy to see that non always to high radium contents corresponds high radon emanation. For instance, the two samples of yellow Neapolitan tuff shown a emanation coefficient more than 3 times higher than others tuff samples and the same behaviour is shown from the two Arciano's soil samples. Moreover, the high value of the emanation coefficient for the pumice sample, suggest that the porosity is the parameter which can explain these results, whose complete interpretation shall come after the careful analysis of the other physics properties of the considered materials.

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