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Natural radioactivity in soils and materials of the Campania region (Italy)

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Summary. — The results of radioactivity content of samples of different types of soils and building materials characteristic of the Campania volcanic region (South Italy) have been reported and discussed. This characterization has been carried out by performing: i) γ-ray spectrometry measurement, using a hyperpure germanium detector in very low background conditions, for the determination of the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K; ii) α-spectrometry of ionized descendants of ^{222}Rn, collected in an electrostatic cell, to measure the emanated fraction of this radioactive gas. Materials of different characteristic and origin have been studied. Results prove the high radioactivity of the samples coming from this volcanic region, underlining the differences among their radioactivity content and their capacity to generate radon.

1. – Introduction

The subsoil of the territory of the Campania region, located in the south-western part of Italy, is characterized by different geological settings [1-4]: sedimentary rocks (karst limestones, carbonate and siliciclastic rocks), having low/very low permeability and porosity, mainly represent the lithological outcrops of the area; some basins (Campanian Plain, Sele River Plain) are mainly formed by lacustrine-marine deposits, having medium-low permeability and porosity; volcanic areas are rich in pyroclastic materials and volcanic rocks, having medium/medium-high permeability and porosity, coming from the eruptions of several volcanic systems of the region (mainly Campi Flegrei and Vesuvius). Many of these latter materials have been largely used in the past and are still

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used as building materials, as well as being the base of the building construction [3, 5]. It is well known that materials and soils of such nature are rich in uranium and thorium traces, and, therefore, are considered responsible for the γ-ray exposition and for the radon release [6, 7]. Radon is a natural radioactive gas present in the environment and the most abundant isotopes are the ^{222}Rn and ^{220}Rn belonging to the natural decay chains of ^{238}U and ^{232}Th, respectively, which are the constituent elements of the Earth's crust [8, 9]. The radon gas migrate in the soil and from soil (or building materials) to the enclosed places, where it can reach concentrations depending on the source strength and on the frequency of the air exchange [8, 10].

The knowledge of the characteristics of the building materials and soils and their radiological characterization is a key point for the evaluation of the expected indoor radon activity concentrations and exposure risk from uranium and thorium [5, 7, 9]. To understand and quantify such a contribution, the present study investigates the natural radioactivity on several samples of building materials and soils of different settings and origin from the Campania region, typical in the architectural constructions for both public and private buildings [3,5]. In the study also some commercial materials have been included. In particular, the work studies the activity concentration of the ^{226}Ra (progeny of ^{238}U and direct parent of ^{222}Rn) and the ^{232}Th (^{220}Rn progenitor) by means of γ-ray spectroscopy; and the ^{222}Rn emanation coefficients through α-spectrometry [11-13]. Also, the ^{40}K radionuclide is investigated, being widely distributed on Earth's crust and being a significant contributor of the γ-radiation exposure [7]. The ^{222}Rn is the main source of the indoor radon, responsible for more than half of the total exposition to the ionising radiations [14, 15]. In fact, not the full amount of radon produced in the soil or inside a building material can reach the external air: only a fraction of radon atoms acquires enough kinetic energy to leave the grain of the material where it has been generated, and to reach the empty space in the porous materials for, finally, being released in the atmosphere [11, 16]. This process is called emanation and the emanated radon fraction is called emanation coefficient [15, 17]. The determination of this parameter is one of the most important elements for the study of the radon flux in the indoor air [5, 11, 15]. A previous paper focusing on a similar radiological characterization of natural building material from the same region was published [5]. Here different building materials samples, together with non-natural material and soils samples are considered.

2. – Soils and building materials selection

After the analysis of the most interesting matrices to study, different from those in the paper [5], the following three groups of investigated samples from the Campania region were selected: soil samples, non-natural (man-made) and natural building materials. All the samples are collected across the entire Campania region and are representative of different geological and geophysical settings of the region [7]; while the man-made building materials are chosen as commonly available in the market.

The soil samples were taken from the most superficial layer, and sorted by different pedologic settings, according to the current system of soils classification [18]: i) two samples from Arciano (Arciano 1, Arciano 2), Vesuvian volcanic soils, group of Typic Hapludand; ii) three samples from Castelvolturno (Castelvolturno 1, Castelvolturno 2, Castelvolturno 3), typical coast line and floodplain, group of Cromic Udic Haplustert, Oxyaquic Xerofluvent and Typic Xeropsamment, respectively; iii) one sample from Somma Vesuviana (Somma-Vesuviana 1), volcanic soil, group of Vitrandic Xerorthent.

The man-made natural building materials samples are chosen among the most used for the buildings structures and walls: loam, bricks, gypsum brick, tuff brick, concrete brick, lime, aerated concrete (siporex), cement. One sample for each type of material is presented.

The natural-origin building materials samples are selected since heavily used in the Campania region in the past and currently [3]. There are materials of volcanic origin: grey tuff from the Caserta province, yellow tuff from the Naples province, green tuff from Ischia island, pumices from the Avellino province, Vesuvius lavic stone. Regarding tuff, the collected samples are originated from different past eruptions and different crystallisation processes. Finally, since in the Campania region a large number of quarries has been used in the past years (350 years) and many of these are today still running, a selection has been made to sample the most widely used building materials [19]: karst stone, pozzolan, ignimbrite, marble, limestone, silt, bauxite.

3. – Methods of analysis

The 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 [20]. Then, they were ground to obtain fine grains of about 1 mm. After this treatment, the samples have been sealed in metallic boxes, whose cover has been soldered to avoid leak of radon during the time for reaching the radioactive secular equilibrium [5]. After that time, the elemental analysis has been carried out by counting the emitted γ-rays by a coaxial High-Purity Germanium (HPGe) detector (spectrometer), contained within a lead shield that ensures a very low background (fig. 1(a)). The detector is equipped with a standard liquid nitrogen cooling dewar [21]. The measurement time was such that a sufficiently high counting statistic was obtained, to reach a statistical error $\leq$5% on the γ-ray peaks area. A specific software for the γ-peak identification, fitting, area and activity calculation has been used [21]. The investigated natural radionuclides have been: ^{232}Th and ^{226}Ra. These are the radioisotopes representing the main external source of irradiation to the human body, and measured commonly during radon surveys [13, 22]. For calculating the ^{232}Th activity concentration, its daughters ^{212}Bi, ^{208}Tl and ^{228}Ac, from the γ-lines at 727 keV, 860 keV e 911 keV, respectively, have been measured. The ^{226}Ra activity concentration has been calculated measuring the daughters ^{214}Pb and ^{214}Bi from the γ-lines at 295 keV and 352 keV plus 609 keV, respectively. Final values of the activities concentrations of ^{232}Th and of ^{226}Ra were calculated with a weighted average of the values obtained from the respective descendants [5, 21]. The γ-line at 1460 keV is used for ^{40}K. The uncertainties have been calculated by propagation error considering the errors associated to counting, peaks analysis, γ-emission probability, energy and efficiency calibration, and sample mass [5].

The emanation coefficient of ^{222}Rn in the investigated samples has been obtained by the ratio between: the activity concentration of emanated radon fraction measured in a collection chamber after the equilibrium of ^{222}Rn with respect to ^{226}Ra has been reached, and the activity concentration of ^{226}Ra in the sample from γ-ray spectrometry [5]. This ratio represents the emanated radon part divided by the total radon part of the measured sample [23]. The unknown numerator of the ratio, *i.e.*, the emanated radon activity concentration, has been measured by α-spectrometry of the ionized progeny of the ^{222}Rn, electrostatically collected on a silicon detector inside a metallic chamber [24, 25]. The chamber has a bottom part where the minced samples are placed, divided by a grid from the electrostatic chamber part where the radon emanates from the sample (fig. 1(b)).

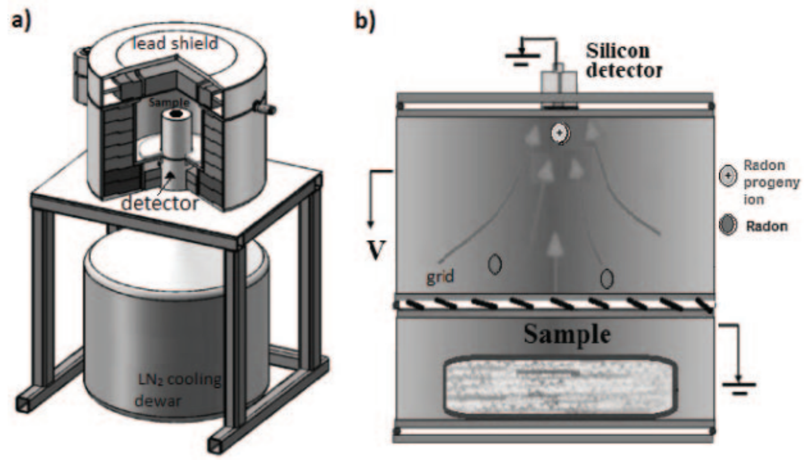

Fig. 1. – Instrumentations used for the radiological characterization of soils and materials samples from the Campania region: (a) the HPGe γ-ray spectrometer for the ^{232}Th, ^{226}Ra and ^{40}K content determination; (b) the collection chamber for the α-spectrometry of ionized radon progeny. The spectrometer specifications of both commercial devices are reported in [21, 26].

The α-lines at 7687 keV of the ^{214}Po, at 6003 keV of the ^{218}Po have been used for the spectrometry. A homemade software for the α-peak identification, fitting, area and activity calculation (using a specific calibration factor of the detector) has been used [17].

4. – Results and discussion

The activity concentrations, in terms of Bq/kg, of the analyzed radionuclides ^{232}Th, ^{226}Ra and ^{40}K; and the ^{222}Rn emanation coefficients obtained for each investigated sample are reported in table I.

The radium, thorium and potassium levels from γ-ray spectrometry highlight the effect of genetic nature, evolution degree and mineralogical composition of all the investigated samples [13, 27]. The results on soil samples prove the correspondence between the soil characteristics and their natural radioisotope distribution. In fact, the samples coming from Arciano and Somma Vesuviana sites show the greatest content of radioactivity, in accordance with the volcanic origin of the pedogenetic substrate, which is higher with respect to the other soil samples analyzed. On the opposite side, sand soil samples of the Castelvolturno site are characterised by a low content of radioactivity [7, 28]. As for the results on building materials: a marked difference between the activity concentrations of the man-made materials and the natural materials has been found. The first ones present values considerably lower ($<$30 Bq/kg for ^{232}Th-^{226}Ra, and $<$450 Bq/kg for ^{40}K) than the second ones, so they are preferred as building materials. This is an interesting result because building materials are directly connected to gamma exposition in homes and work places [13, 29]. Among the selected natural materials (also from quarries), the higher activity concentrations ($>$400 Bq/kg for ^{232}Th-^{226}Ra, and $>$6000 Bq/kg for ^{40}K) emerge from pozzolan and lavic stone, materials consisting mainly of volcanic rocks and slags, mainly from the areas of the Campi Flegrei caldera [30]. The second group of medium activity concentrations materials comes from pyroclastic sediments,

TABLE I. – *Results, with uncertainty (Δ), of the radiological characterization of soils and materials samples from the Campania region: the activity concentration of ^{232}Th, ^{226}Ra and ^{40}K measured by HPGe γ-spectrometer, and the ^{222}Rn emanation coefficients using α-spectrometry.*

	Sample	Activity concentration (Bq/kg)						Emanation coefficient	
		^{232}Th	Δ	^{226}Ra	Δ	^{40}K	Δ	^{222}Rn	Δ
Soil	Arciano 1	119	14	262	3	1553	60	0.43	0.06
Soil	Arciano 2	83	7	319	8	1358	54	0.32	0.05
Soil	Castelvolturno 1	75	4	55	3	636	25	0.13	0.04
Soil	Castelvolturno 2	56	4	53	3	739	30	0.16	0.05
Soil	Castelvolturno 3	17	2	13	1	353	14	0.05	0.01
Soil	Somma-Vesuviana	40	4	142	4	768	35	0.25	0.09
Man-made material	Loam	10	2	15	3	344	21	0.04	0.02
Man-made material	Bricks	20	2	14	2	282	15	0.05	0.01
Man-made material	Gypsum brick	5	1	7	1	56	8	0.02	0.01
Man-made material	Tuff brick	4	2	26	2	445	16	0.21	0.09
Man-made material	Concrete brick	3	1	17	1	101	8	0.05	0.03
Man-made material	Lime	12	3	17	2	192	14	0.06	0.02
Man-made material	Siporex	10	2	7	1	84	9	0.02	0.01
Man-made material	Cement	28	3	24	3	214	26	0.15	0.04
Natural material	Grey tuff	106	6	85	3	1747	61	0.21	0.02
Natural material	Yellow tuff	104	7	82	4	1859	67	0.62	0.12
Natural material	Green tuff	89	6	82	4	1625	54	0.23	0.03
Natural material	Pumices	197	7	151	5	1933	62	0.39	0.08
Natural material	Lavic stone	512	18	488	11	9221	83	0.16	0.03
Natural material	Karst stone	111	4	97	2	1347	33	0.31	0.02
Natural material	Pozzolan	741	17	422	6	6412	97	0.07	0.02
Natural material	Ignimbrite	144	7	100	2	640	19	0.02	0.01
Natural material	Marble	33	4	47	2	634	17	0.09	0.02
Natural material	Limestone	27	2	28	1	161	9	0.08	0.02
Natural material	Siltstone	11	7	14	4	129	8	0.12	0.04
Natural material	Bauxite	181	10	83	2	954	50	0.03	0.01

ranging from 80 to 200 Bq/kg for ^{232}Th-^{226}Ra and from 900 to 2000 Bq/kg for ^{40}K. The remaining natural materials are siliciclastic and carbonate rocks, which present very low radioactivity levels (<50 Bq/kg for ^{232}Th-^{226}Ra and <650 Bq/kg for ^{40}K) [5, 31, 32]. The radioisotope present with higher activity concentration in any kind of sample is the ^{40}K, which is present practically in all minerals coming from the crust of the Earth and contributes significantly to the γ-radiation exposure [7].

The results of the ^{222}Rn emanation coefficient underline the differences among the radioactivity content of the samples and the capacity of the same to emanate radon. Soil samples and man-made building materials show a correspondence between radon emanation coefficient and their natural radioactivity [11, 33, 34]. On the contrary, natural materials do not always respect this correspondence: the occurrence of high value of activity concentration of ^{226}Ra (direct parent of ^{222}Rn) in the samples building materials does not always correspond to high ^{222}Rn emanation coefficient [5, 35]. The more evident result is that pozzolan and lavic stone, with high radioactivity content, have low emanation power. Yellow tuff releases a quantity of radon higher than all studied materials, even compared to those of volcanic origin [36]. This effect is mainly to be attributed to the different porosity of the materials, and in particular of tuffs, which strongly increases the probability for a high number of radon atoms to escape from the grain into the intergranular spaces and then into surface air [36, 37].

The results of radioactivity content and radon emanation coefficients on the investigated soils and building materials characteristic of the Campania region nearly confirm what was found in previous works, which analyzed samples of similar nature [5, 28, 29, 31-33, 35-39].

5. – Conclusions

A wide study of the radioactivity content and the radon emanation coefficient in several samples of different pedologic settings soils and building materials (natural and non-natural) characteristic of the Campania region (Italy) is presented. Most of them have been and are widely used in building construction. The analyses have been performed by γ-ray spectrometry for the determination of the total activity concentration, and by α-spectrometry to measure the emanated fraction of radon gas. The obtained results show radioactivity values reflecting the characteristic of the various materials and soils considered; higher values in volcanic-origin samples are reported. Moreover, the occurrence of high value of radium activity concentration does not always correspond to high radon emission. The parameters computed in this study can contribute to the estimation of radon entry rates in living environments (according to the equations governing the radon transport through diffusion and advection) to assess the human health hazards of indoor radon accumulation [38]. This study can be useful as input for the creation of an identification model of risk areas in Campania.

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