RADON CONCENTRATION ACTIVITY IN A NORM CONTAMINATED HOUSE. EVALUATION AND RISK ASSESSMENT

F. Nicolace*, S. Procopio**, S. Coppola**, P. Barbuto**,S. Ferro**, G. Ranieri**, S. Pascuzzi**, M. Bonanno***

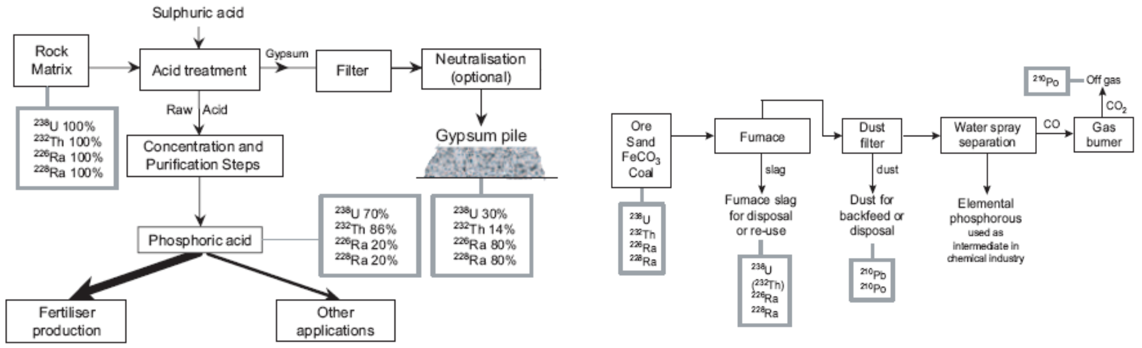
*ARPACAL, *Centro Epidemiologia, Dipartimento di Catanzaro (CZ), Italia*
***ARPACal, Laboratorio Fisico Ettore Majorana Dipartimento di Catanzaro (CZ), Italia*
****STS, Solar Technology System, info@sts-energy.it*

ABSTRACT

The NORM[1] may be the raw material, the product or the residue in the processing of many industrial activities where the radiation risk is generally undervalued compared to the chemical risk. Each working activity involving the use, the storage or the production of materials and/or residues containing NORM, may cause an increase of radiation risk coming from the level of natural radioactivity that is higher than earth's crust level. Legislative Decree n.241/00, requires the risk exposure radiometric control of the workers and the population in case of work activities that deal with NORM and are included in Annex Ia. Crotone, the city of the Achaeans, a Greek colony known Italian chemical industry and power of the last century, located on its territory residues and materials containing NORM which resulted in an increase of radioactive environmental fund. Some properties of these materials are, however, similar to those of sedimentary rocks of phosphate type. A large quantity of phosphorous silicate resulting from the industrial production cycle, was used for filling roads, squares, schools, public offices and private homes. Through the determination of radon concentration this study aims to estimate the amount of radon originated by the presence of NORM within in the crawl space of a dwelling scale model made using the same material and the same construction techniques used for Croton City. A further object of this study is assessing the radiation transparency of the concrete used for the coverage of the crawl space itself. The main objective, therefore, is to verify the hypothesis the processing residues containing NORM, used for the construction of living environments, increase the effective dose to the population and to workers, not only for the external radiation but also for the contribution due to inhalation of radon generated by phosphorus residues. In the recent past, in this area the industrial wastes were considered inert and, therefore, suitable for filling volumes for building construction.

INTRODUCTION

The first industrial production in the city of Crotone dates back to 1928 with the construction of a plant for the production of sulfuric acid obtained from the pyrite through an oxidation of the iron disulfide. The sulfuric acid was used to attack a yellowcake mineral called phosphorite from Algeria and containing predominantly calcium phosphates. The phosphorite was used for the production of calcium superphosphate with low content of phosphorus pentoxide (Figure 1). Processing of phosphorite (diphosphate) represented a solid basis for the marketing of phosphorous products: phosphate fertilizers, detergents, animal feed, elementary additives and pesticides. The radioprotection interest is considerable especially for the production processes involving a precipitation of residues containing NORM, such as phosphogypsum. Among the most important cycles you may include: phosphoric acid production by the wet process, attack phosphate rock with nitric acid and process producing elemental phosphorus thermally. In the installations for the production of fertilizers of the city of Croton it was produced about 5 10^3 tons of processing residues between phosphogypsum and, silicates of blue[2] gray color containing NORM in quantity not defined. Only a marginal part of processing residues was disposed in a landfill for inert overlooking the Ionian coast to the border with the mouth of Esaro's river. A large quantity of these residues, not easily quantifiable however, thanks to its mechanical properties, has been used as filling material for roads, ports, yards of schools, public places, and in the crawl spaces of civilian homes.



Flow diagram of the sulphuric acid phosphorus extraction process.

Flow diagram of thermal production of elemental phosphorus.

Fig. 1: *main production processes that characterized the Crotone area*

The study is really timely for the discovery of phosphorous compounds in a former vocational school in the city and for a radon gas measurement campaign in the workplace. It emerged from the information sheets, an almost certain probability that the waste material, with a high content of natural radioactivity, was systematically used in civil rooms in place of more expensive and not readily available inert material. Waste material was used for filling crawl spaces and foundations of many buildings. After an initial screening tests aimed at determining the activity concentration of radon produced by the material concerned, and housed in a known volume and sealed, it proceeded with the experiment design, based on a single purpose. It wanted to play civil dwelling in scale with the same construction techniques used since the seventies to the early nineties, in which the filling of the hornet's nest was made with an adequate mass of residual phosphorous, from the radiometric characteristics notes and also following signs of local workers. The body of the studied factory (Figure 2) is a floor above ground from the plan dimensions of 1.00 mx 1.00 m, proportionate to a typical building envelope, and with accurate construction details as the thickness of the walls, their inner tubes, internal and external plaster of the house, we named in the loyalty to the most popular name of this city, Mary's House (Figure 2). The parts that make it up are:

- foundations;
- the crawl space and the screed flooring;
- the closing wall;
- slab’s roof;
- the roof;

The foundation beams are made of reinforced concrete with a specific weight $\gamma = 23 \div 23{,}5$ kN/m^3 on a raft of the same material. The crawl space is filled with a mass of 61 ± 1 kg of residual phosphorous ($\rho = 1220$ kg/m^3) and on the latter has been realized a screed in cement mortar with a thickness of 3 cm. The closing wall , hollow brick ($\Gamma = 8$ kN/m^3) it was placed with the axis of the holes in the vertical such as to increase the volume of air present on the walls of the house. The roof slab with brick flooring blocks and a screed mortar complete the work. Everything was plastered inside and out trying to reduce the thickness to a minimum and painted with washable paint. The roof, made of two layers of corrugated sheet metal with closing gables of the same features of closing wall, has the task to brighten the work seen. Finally, there is a wooden door useful to facilitate operations within the Mary's House suitably sealed when taking measurements.

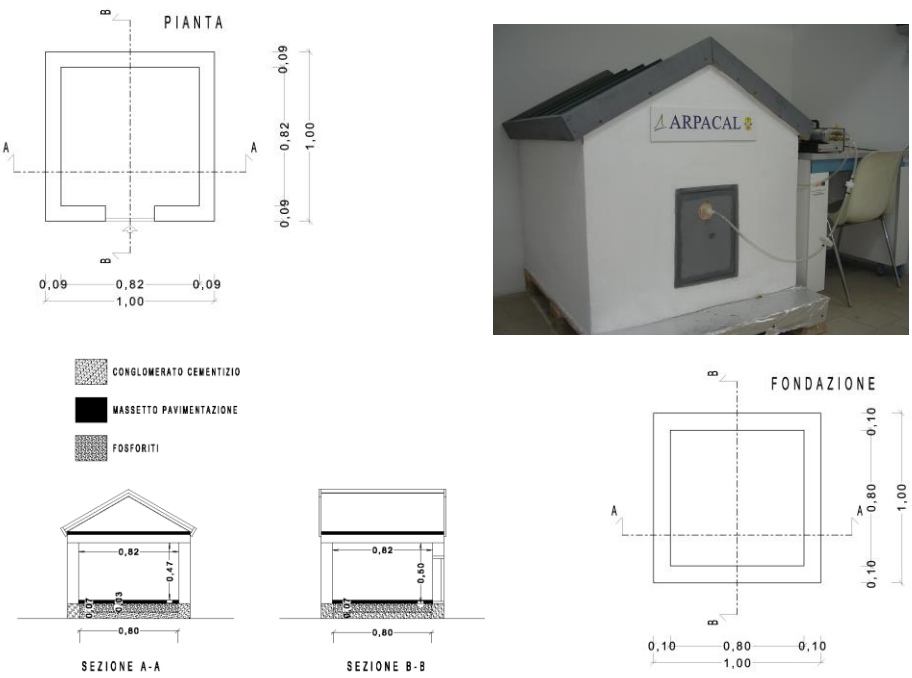


Fig.2: *construction details of Mary's House*

Below in Table 1 shows the characteristics of radiometric phosphorous compounds used for filling the crawl space. The material comes from a forecourt of a former professional school of the city, initially covered with a layer of bitumen and easily recover.

Campioni (61): silicati fosforosi prelevati Scuola ex Ciapi - Crotone	**ICP- Massa** [Bq/kg]	**Spettrometria Gamma** [Bq/kg]
Concentrazione di attività ^{238}U	1253±182	940±45
Concentrazione di attività ^{232}Th	30±4	10±2

Tab.1: *radiometric characteristics of phosphorous silicates*

MATERIALS AND METHODS

The concentration of radon gas activity in Mary's House was observed with two technical measures:

1) the continuous measurement with a monitor MR1 (Lucas cell) active mode;
2) four passive exposure meters electret short terms in configuration SST[3];
3) two passive exposure meters to long-term electret LLT configuration;
4) two passive exposure meters in SST configuration packed in waterproof radon envelopes, for the measurement of gamma dose in the dwelling.

The measurement of range in the air dose was also obtained from continuous observation with a monitor proportional (Berthold LB123) to the housing center, in measurement periods different and with a counting time of 600 seconds. The continuous monitoring of the concentration of radon is made using a measuring chain formed by: a monitor type MR1 with a scintillation detector, a Lucas cell with a sensitivity of 0,0341 [cpm/(Bq/m^3)] coupled to a photomultiplier. The measurement was made with an active execution mode, with forced circulation within the Lucas cell by an artificial flow of 0,25l/min. On the entrance door of Mary's House, also useful to load the filling material, it has been practiced with a plug filter to draw the air in the internal volume and bring it to the monitor for the continuous determination of the concentration of radon activity. Depending on the measurement configurations, before the start of counting the door it has been suitably siliconed to contain any leaks of gas. All of the monitoring phases, both in continuous and with passive devices, involved in the different configurations, the measuring periods of at least seven days. As is known in the electret long (long-term) or short duration (short term) are disk of Teflon electrically loads, mounted on a conductive plastic counting chamber type L or S depending on the counting solutions. The measurement technique is called E-Perm system and is based on the detection of α radiation emitted during radioactive decay. The gas penetrates in these devices by diffusion and effect of its decay ionizes the volume of the counting chamber by downloading the surface of the disk potential. The difference between the initial and final electrical potential is possible to determine the concentration of radon volumetric activity in Bq / m3presente at a particular site using the relationship [1]:

$$[^{222}Rn] = \left\{ \frac{V_i - V_f}{C_F \cdot t_e} - C_\gamma \right\} \cdot H \qquad [1]$$

where [^{222}Rn] in Bq/m^3 is the concentration of radon in the air activities; There Vf in Volts, the initial and final surface potential of the elettrete; CF [Volts m3 / Bq day], the calibration coefficient; you in days is the exposure time, which for this survey had a minimum duration of 7 days; C_γ in Bq/m^3 is the equivalent concentration of radon due to gamma radiation; H is the dimensionless correction factor for the altitude. The potential reader used for reading the electret is a RadElec E-Perm[4]. Having built the Mary's House[5] six days, was respected a period of rest before placing it on the ground floor of the laboratory, the experiment site is known and where the annual average radon concentration to be 35±12 Bq/m^3. It is the continuous measurement and with passive exposure meters by sealing the volume was initiated for a week with the intent to know the initial radon levels (Fig.3) and verify the seal of the built volume. Subsequently, it was carried out the filling of the crawl space loading in conditions of safety the residual phosphor, in three successive phases with the aim of determining in a known volume, a relationship between the mass of the residual phosphor and the concentration of radon activity, over to determine the radon component generated by the phosphorous materials. The measures in the different phases have covered the same time counting and the basic climate parameters pressure, temperature and humidity in the media have not undergone significant oscillations. The physical quantities listed below describe the initial provision of Mary's House:

- average concentration of radon activity in a continuous 61±13 Bq/m^3 and with passive measuring 47±15 Bq/m^3;
- average internal pressure: 101 ± 2 kPa;
- average internal temperature of 31 ± 8 ° C;
- relative humidity 41%.

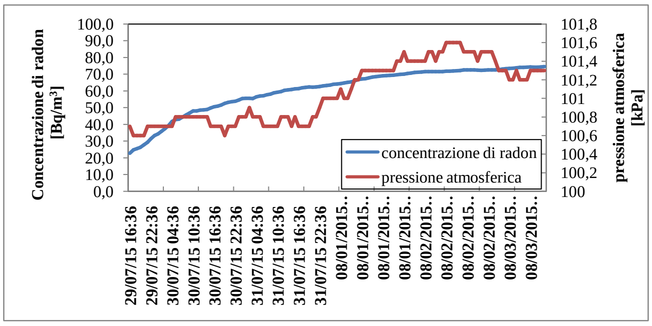


Fig.3: *initial level of radon in Mary's House*

The level of radon in Mary's House is made from a primordial component and a phosphorous due to the presence of residues used for the filling of the crawl space [2]:

$$[^{222}\boldsymbol{Rn}] = [^{222}\boldsymbol{Rn}]_{\mathbf{0}} + [^{222}\boldsymbol{Rn}]_{\boldsymbol{Rf}} \quad [\mathbf{2}]$$

having understood with: $[^{222}Rn]$, the concentration of total volume radon in Bq/m^3; $[^{222}Rn]_0$ in Bq/m^3 the combination of the initial radon concentration present in the environment in which the confined Mary's House was hosted and that produced by the construction in the volume of interest; $[^{222}Rn]_{Rf}$ in Bq/m^3 is the component of radon concentration generated by the processing of phosphorous residues accommodated in a volume of 0,05 m^3, which is the spatial characteristic of the crawl space. It is observed that 3 cm of concrete used for the surface coverage of the crawl space and to realize the screed of the flooring have increased by some Becquerel per cubic meter, the concentration of radon. We have checked that the transparency of the concrete to radon, no screen factor as is known, only a reduction by a factor of 0,5 of the irradiation generated range from phosphor material and a complete reduction of the beta contamination, from 2 to 43 cps at contact cps. With the relation [2] it was possible to estimate the radon component generated from phosphorous compounds used and with the UNSCEAR 2000 model was made an effective dose calculation due to inhaled radon and only generated from this component [3]:

$$[\boldsymbol{H_E}] = [^{222}\boldsymbol{Rn}]_{\boldsymbol{Rf}} \cdot \boldsymbol{E_q} \cdot \boldsymbol{T} \cdot \mathbf{9}\boldsymbol{nSv}(\boldsymbol{Bqhm}^{-\mathbf{3}})^{-\mathbf{1}} \qquad [\mathbf{3}]$$

having intense:$[^{222}Rn]_{Rf}$ in Bq/m^3, the component of the average concentration of radon generated by phosphorus processing residues; *Eq* the equilibrium factor between radon and its progeny of 0,4; T is the exposure time in hours, taken to be equal to 7000 h in a year; 9 is the conversion factor for the effective dose[6] .

RESULTS

Table 2 below presents the levels of radon measured continuously in different configurations and volumes: the initial volume 0,4 m^3, 0,33 m^3 after filling the crawl space with phosphorous and 0,32 m^3 residues after realizing the paving with 3 cm of concrete. The last column of the table shows the gamma dose rate measured in the air inside the house in a central point of interest volume

Configurazione	**Concentrazione di attività di radon [Bq/m^3]**	**Rateo di dose gamma [nGy/h]**
Mary's House con vespaio libero	61±13	91±8
Mary's House: vespaio con 61±1 kg di residui uraniferi - silicati fosforosi	102±17	421±50
Mary's House: vespaio con 61±1 kg di residui uraniferi e una copertura con 3 cm di calcestruzzo	116±17	210±24

Tab.2: *natural radioactivity and sets of Mary's House*

During the loading phase of the phosphor material, useful to the filling of the crawl space, it proceeded with a dosage of mass introduced to determine a relationship between the uraniferous residues laid inside the dwelling and radon levels produced in a known volume. Figure 4 indicates that substantially by a certain amount of mass in then there is a "saturation" of the volume that houses the radioactive material.

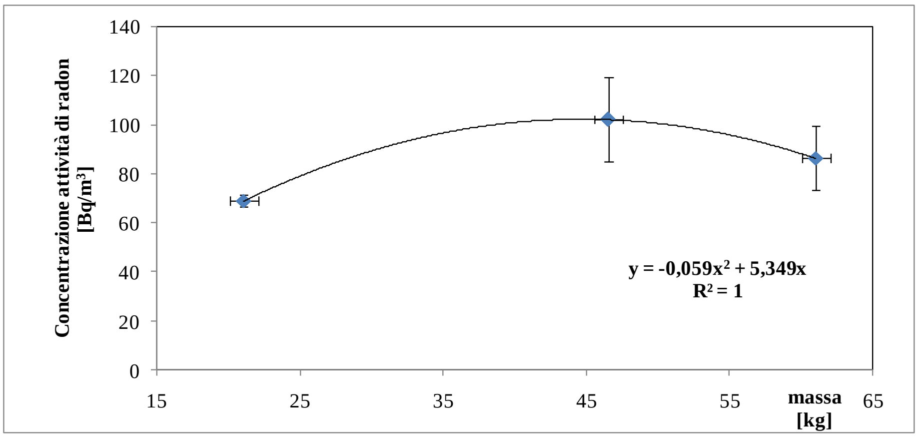


Fig. 4: *the concentration of radon and the mass of phosphoric residues*

As little as 41 kg of phosphorous residues to determine the higher level of radon concentration recorded during monitoring. If you evaluate uncertainties about the level of concentration it can be said that 41 kg of waste are sufficient to reach the plate - out of the concentration. Other considerations related to gas exhalation and concentration distributions were not considered. Figure 5 presents the radon levels of the house in three configurations and with two modes of measurements used. It can be appreciated, in general, the agreement of the two active and passive techniques, and especially the level of concentration measured with passive monitors in short period configuration and the entire situation, in which you should find a normal house. The radon level, in Mary's House, which is home to the crawl space NORM residues from chemical processing industry of the past, has a natural radioactivity nearly three times the environmental fund that is registered in a normal building, if you refer to the values more conservative.

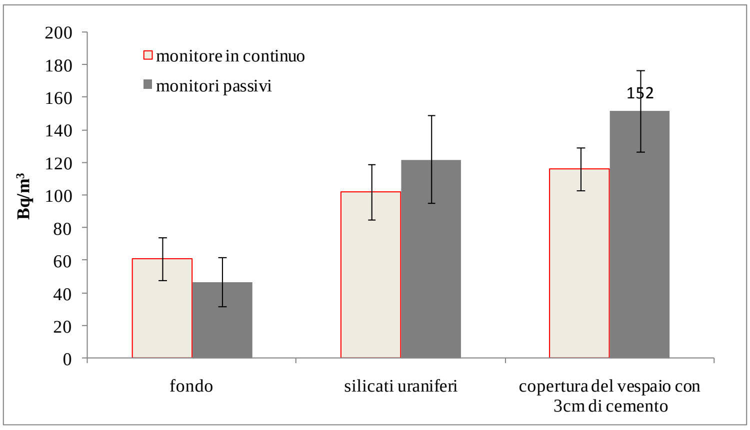


Fig. 5: *the concentration of radon activity measured with the techniques passive and active measurement*

In figure 6 are reported the continuous recordings of the concentration of radon activity in measuring periods of at least 7/10 days, in the three configurations of the house. You can appreciate the graphical differences. The last configuration, the one with the concrete pavement was observed for a longer measurement period (15 days).

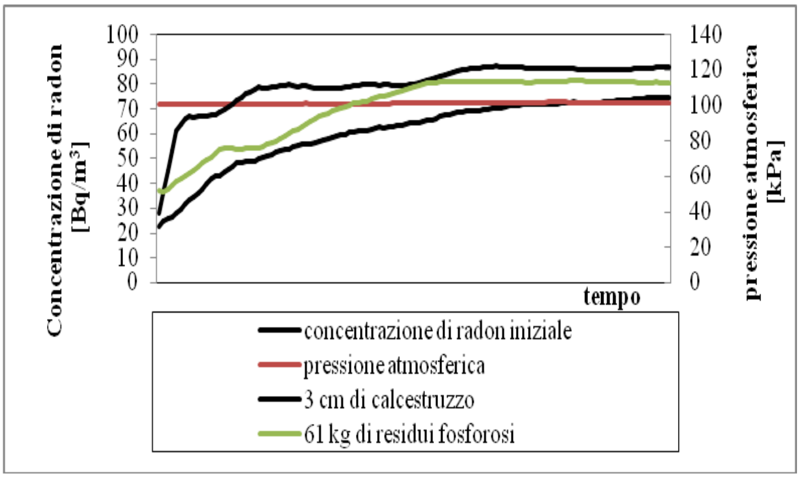


Fig. 6:*la concentrazione di attività di radon misurata con il monitore in continuo*

If you apply the relation [2] and is considered a more conservative situation you will find that the radon component generated by the phosphorous material is equal to 111 Bqm^{-3}. If this data is normalized for the volume of interest and the mass of the radioactive source used for the filling of the formwork, one can derive an activity factor relative to radon generated by each unit mass of phosphor material. In particular by performed calculations show that σ, factor activity for radon due to the phosphorous compounds used in indoor environments is of 0,58 $Bqkg^{-1}$. The result, when applied to a real house of 100 m^2 where it is assumed a volume of crawl space of 20 m^3 and you are using the volumetric mass of the residues of the experiment, 1,220 kgm^{-3}, one can estimate the contribution of concentration of radon activity due to the use of materials containing NORM which is equal to 76 Bq m^{-3}. Obviously in a real situation this concentration will not necessarily add up in algebraic terms, the initial concentration, typical of the geology of the place of construction of the dwelling. If we focus our attention only on the component of natural radioactivity but sets, and using the model UNSCEAR 2000 for the calculation of the effective dose [3] which corresponds to the concentration value of radon activity of 76 Bq m^{-3}, is It has a dose value of 1,9 mSv; if using the conversion factor of the Italian legislation Legislative Decree n. 241/00 contribution to the phosphorous dose of 0,64 mSv in a year.

The total dose for the population, including radon and required by this standard is 1 mSv/year.

DISCUSSIONS

The study, with all the scientific reserves appropriate, returns some important reflections as well as results that can be examined, discussed or analyzed. What is certain is that where residues have been employed phosphorous in indoor environments as substitutes for the more expensive inert material, in addition to a dose increase natural range, it is determined a considerable increase of the radon gas levels in the living environment. The scale model built is not a scientific exercise but rather a response to the radiation protection of the territory and for those who are committed to reduce the risk of lung cancer for the population and for the workers, based on observed data. The Mary's House is a project that is still developing through the evaluation of remedial actions to be applied to reduce the radon concentration.

REFERENCES

[1] *Extent of Environmental contamination by Naturally Occurring Radioactive Material (NORM) and technological options for mitigation*. Technical Reports series n. 419. International Atomic Energy Agency, Vienna, 2003

[2] Doc. XXIII n. 7 *Commissione parlamentare d'inchiesta sulle attività illecite connesse al ciclo dei rifiuti*

[3] Caresana M . et al, *Uncertainties evaluation for electrets based devices used in radon detection*, Radiation Protection Dosimetry Advance Access published November 30, 2004

[4] *Intercomparison Radon Passive*, BFS-Berlino 2015

[5] I materiali per la costruzione della Mary's House sono stati forniti dalla ditta STS, *Solar Technology System*, info@sts-energy.it

[6] A Kinsara et al, *Distribution on ^{222}Rn Concentration in an inhabited area adjacent to the AJA Granitic Heights of Hail Province, Saudi Arabia*, Health Physics, vol.108 N.1 January 2015 pag.59