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Article

Indoor radon concentration and gamma dose rate in dwellings of the Province of Naples, South Italy, and estimation of the effective dose to the inhabitants

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Abstract – The indoor radon concentration was measured in 471 dwellings in the Province of Naples. Radon concentration measurements were carried out using LR-115 passive alpha track detectors (SSNTDs) that were exposed for two consecutive semesters. The annual average radon concentration in the dwellings was found to vary from 21 to 722 Bq m^{-3} with an average value of 107 ± 75 Bq m^{-3}. In about 93% of the dwellings the radon concentrations were found to be below 200 Bq m^{-3}, which is the recommended level by Italian law for new buildings. Simultaneously with the radon concentration measurements, indoor gamma dose rates were measured in 388 dwellings. Indoor gamma dose rates were measured using Thermoluminescent Dosimeters (TLDs) exposed in each dwelling for six months. The arithmetic mean value was found to be 327 ± 102 nGy h^{-1}, after the subtraction of the cosmic ray contribution. A weak correlation between the indoor radon concentration and gamma dose rate was found. The correlation between indoor radon measurements and gamma dose rates and the same building characteristics was also studied.

Keywords: radon / gamma dose rate / LR-115 detectors / TLD dosimeters

1 Introduction

Major exposure to ionizing radiation for the general population arises from terrestrial radiation and cosmic rays (UNSCEAR, 2000; Al-Saleh, 2007; Almgren *et al.*, 2008). In particular, internal exposure due to radon and its progeny is responsible for more than 50% of the natural background dose to human beings, as reported by UNSCEAR (2000). Today, it is well known that exposure to indoor radon and its short-lived decay products contributes significantly to an increasing risk of lung cancer (Lubin *et al.*, 2004; Bochicchio *et al.*, 2005b; Darby *et al.*, 2005; Krewski *et al.*, 2005, 2006). After inhalation, radon is almost completely exhaled due to its long half-life (3.82 d) and being an inert gas, while its progenies, specially the two radon daughters with short half-lives, ^{218}Po and ^{214}Po, being electrically charged, can be attached to dust or smoke particles (aerosol) in indoor air. When these particles are inhaled, a fraction of them is deposited in the lungs, where they emit alpha particles that are absorbed in the nearby lung tissue, damaging the pulmonary epithelium and thereby increasing the probability of having lung cancer. Indoor exposure to radon daughters is considered responsible for 10–15% of the total cancer deaths in the USA (NRC, 1993). In Italy, the estimate of lung cancer deaths attributable to radon exposure was found to be about 10% (Bochicchio *et al.*, 2013). Worldwide, many investigations have been conducted to evaluate this kind of exposure in homes and to develop useful remediation. On the basis of recent epidemiological studies, the recent Council Directive (Euratom, 2013) reports a statistically significant increase in lung cancer risk from prolonged exposure to indoor radon at levels of the order of 100 Bq m^{-3} and it states that national action plans are needed to address long-term risks from radon exposure. The International Commission on Radiological Protection (ICRP, 1993) has established an action level between 200 Bq m^{-3} and 600 Bq m^{-3} . Based on the review of the epidemiological studies on cohorts of miners and population-based case-control studies, ICRP Publication 115 (ICRP, 2010) determined a new nominal risk coefficient of 8 × 10^{-10} Bq h m^{-3} and with this value, an annual dose of about 20 mSv per year can be calculated by 600 Bq m^{-3}. So, the Commission, in the associated Statement on Radon (2009), revised the reference level for the radon concentration in dwellings from 600 Bq m^{-3} to 300 Bq m^{-3}. ICRP Publication 126 (ICRP, 2014) questioned the protection strategy against radon based on the distinction between protection approaches for dwellings and workplaces (ICRP, 1993, 2007) and now recommends an integrated approach for protection against radon exposure in all buildings, whatever their purpose and the status of their occupants. This strategy is based on an optimization principle below a derived reference level set in terms of the concentration in

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air. The national authorities are encouraged to set a reference level in the range 100−300 Bq m^{-3}, taking the prevailing economic and societal circumstances into account. In Italy there is no legislation for protection against exposure to radon in dwellings; currently, only the elaboration of a legislative proposal to that effect is part of the National Radon Plan (PNR) (Ministry of Health, 2002). Today, it is assessed that the majority of inhabitants in urban areas spends about 80% of their time indoors, so their exposure to ionizing radiation from building materials could be relevant (UNSCEAR, 2000). In Italy, only one national survey has been carried out that investigated the radon concentrations and gamma dose rate in dwellings in the early '90s. Subsequently, in the Province of Naples, other studies were performed to determine the radon concentration in homes and workplaces (Pugliese *et al.*, 2013, 2014; Quarto *et al.* 2013, 2014). In this study, simultaneous measurements of the radon activity concentration and gamma dose rate in some dwellings of the Province of Naples are shown. In this region, in particular in the coastal area, where some active volcanoes are present, the concentrations of undifferentiated pyroclastic rock are elevated. These materials, very rich in uranium and thorium traces, widely used in house construction, make the natural radiation exposure particularly significant. Moreover, the statistical relationship between the radon concentration and gamma dose rate and some building characteristics such as the age of the dwelling, main building materials and floor level were analyzed.

2 Materials and methods

2.1 Radon specific activity measurements

Radon measurements were carried out with LR-115 passive alpha track detectors (SSNTDs). The choice of dwellings to be monitored was not carried out with a random sampling, but it was based on our convenience and the availability of the inhabitants. However, the monitored houses were very different in building materials, age of construction and the floor of the monitored room. In each dwelling the detector was exposed in the room where the inhabitants spent most of their time, generally the living room and bedroom, for two consecutive semesters in order to obtain a concentration averaged over the whole year. Homeowners were asked to fill out a questionnaire containing details about their lifestyle and dwelling characteristics. The questionnaires, returned with the track detectors and TLDs, provided information on the type of dwelling, age of dwelling, floor of the room where the detectors were placed, type of major building materials, and type of materials lining the walls and the floor. After exposure all detectors were chemically etched using a solution of 2.5 N NaOH at 60 °C for 110 min. For LR-115 detectors, the number of tracks increases linearly as the residual thickness decreases. So, it is necessary to determine the residual thickness to normalize the observed track density to the nominal final thickness (6.5 μm). The residual thickness was determined by acquiring the image of the detector by means of a scanner with double lighting and its mean brightness in the gray scale was determined using suitable software for image processing[1]. Using a previously determined calibration curve, the brightness was then converted into residual thickness The automatic counting of tracks was also performed using the ImageJ software. The radon activity concentration was calculated using the formula:

$$C_{Rn} = \frac{N}{E \times T} \quad (1)$$

where N is the track density corrected by the background and normalized to the nominal thickness of 6.5 μm, E the calibration factor and T the exposure time. The Lower Limit of Detection (LLD) of the method was estimated to be 4 Bq m^{-3}. The quality assurance of the whole method was performed by participating in the intercomparison exercise organized by the German Federal Office for Radiation Protection (BfS). The participation in the intercomparison showed that the results were included in the good category with an uncertainty of 12% with respect to the references (data not published). Moreover, the Laboratory of Radioactivity (LaRa) has accreditation according to ISO (2008) and the European Standard (ISO/IEC, 2005) for an "Integrated measurement method for determining the average activity concentration of radon 222 in the environmental air using passive sampling and delayed analysis" (ISO, 2012).

2.2 Gamma dose rate measurements

Measurements of the indoor gamma dose rate were carried out simultaneously with the radon measurements in 388 houses. Measurements were performed using TLDs exposed for six months in the same position where the LR-115 detectors were placed. In this survey, LiF:Mg,Ti (TLD 100) chips were used due to their low cost, high sensitivity and low fading. When ionizing radiation hits a TL material, electrons are freed from some atoms and moved to other parts of the material, leaving behind "holes" of positive charge. Subsequently, when the TL material is heated, the electrons and the "holes" re-combine, and release the extra energy in the form of light. The light intensity can be measured, and related to the amount of energy initially absorbed. Calibration of the TLD 100 was carried out using a photon beam with an average energy of 3 MeV at the LINAC of the Istituto Tumori "Fondazione Pascale" of Naples. The TLDs were exposed to doses ranging between 0.2−1.2 Gy, for which the thermoluminescence (TL) glow curves were recorded and the areas of the main peak were determined. A calibration curve between the dose rate and TL intensity was built and the calibration factor of 4.3 ± 0.4 nC mGy^{-1} was determined. After exposure, all TLDs were read between 100 °C and 300 °C using a heating rate of 10 °C s^{-1} with a TL reader (Harshaw 3500). For the gamma dose rate measurements, the LaRa has accreditation according to ISO (2008).

2.3 Statistical analysis

For each dwelling, in order to obtain annual values, the time-weighted average radon concentration from two consecutive semesters was calculated using the exposure time, expressed in days, as a weight. The normality of the log-transformed data was tested by the Shapiro-Wilk test and the

[1] ImageJ software: Image Processing and Analysis in Java, version 1.46r, National Institutes of Health, USA.

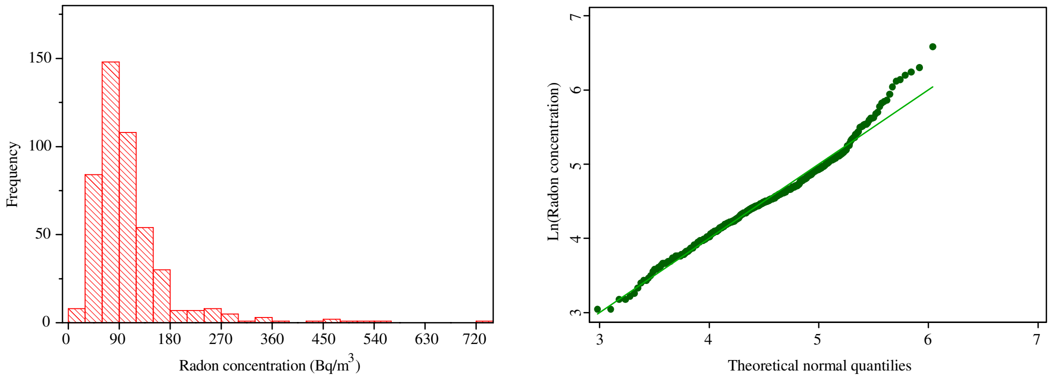

Figure 1. (a) Frequency distribution of radon concentrations in dwellings of province of Naples; (b) Q-norm plot of natural log-transformed radon concentration.

homogeneity of variance was tested by Bartlett's test. The comparison between pairs of groups was performed by means of Student's t-test, while the comparisons among multiple groups were performed by Analysis of Variance (ANOVA). The significance of difference between groups was evaluated with Bonferroni's post-hoc test. The relationship between the indoor gamma dose rate and radon concentration was analyzed using Pearson's correlation. All statistical analyses were performed using Stata software (Stata Corp, College Station, TX, USA).

3 Results and discussion

3.1 Indoor radon activity concentrations

The frequency distribution of the annual radon concentrations measured in the 471 dwellings is reported in Figure 1a. The experimental data show an approximately log-normal distribution, although the Shapiro-Wilk test failed to assess the normality (p-value < 0.001) because of the departure from normality of the data distribution's upper tail (Figure 1b). The radon concentration measured in the entire investigated area ranged from 21 to 722 Bq m^{-3}, with an arithmetic mean of 107 ± 75 Bq m^{-3} and a geometric mean of 91 Bq m^{-3}. The average radon activity concentration measured in the present survey is higher than the Italian national average, which is 75 Bq m^{-3} and Campanian average, which is 95 Bq m^{-3} (Bochicchio *et al.*, 2005a). This result is attributable to the fact that the soil of the Province of Naples is very rich in pozzolan and tuff with respect to other places in the Campania region and other regions of Italy. Eighty-six percent of the monitored dwellings were single-family houses, 9% were in multistorey buildings with less than 10 dwellings and only 5% were in multistorey buildings with more than 10 dwellings. Forty-four percent of the dwellings were built mainly of bricks and stone, 23% of concrete and 30% of tuff. About 26% of the monitored rooms were placed on the ground floor/in the basement, while 35% were on the first floor, and 38% on a floor higher than the first floor.

The results show that in 93% of houses, the radon concentrations are below 200 Bq m^{-3}, which is the recommended level of Italian legislation for new buildings, and only 1% are higher than 400 Bq m^{-3} which is the recommended level of Italian legislation for old buildings. Currently in Italy there is no mandatory program for the implementation of remediation actions of homes where the radon concentration exceeds recommended levels. Consequently, in the absence of a law, only individual interventions are made by public agencies or individuals sensitive to the issue.

It is well known that indoor radon concentrations are generally affected by several factors such as the age of the dwelling, kind of floor, building materials, etc. and so the effects of these building characteristics on the radon levels were examined. In Table 1, descriptive statistics of radon activity concentrations and their analysis of variance (ANOVA) by major factor are reported. The analysis of variance was applied to the log-transformed data after checking in all cases the homogeneity of the variances within the groups to be compared with the Barlett test.

As shown in Table 1, the one-way analysis of variance on the log-transformed data presents a statistically significant

Table 1. Analysis of variance of the indoor radon activity concentration by major factor.

Floor level	***N***	**Range (Bq m^{-3})**	**GM (Bq m^{-3})**	**GSD**	***p*-value**
Basement and ground floor	121	36–546	111	1.6	
First floor	163	21–513	91	1.6	<0.001
Second or higher floor	176	21–722	80	1.7	
Year of construction					
Before 1919	81	21–463	116	1.8	
1919–1960	104	31–546	109	1.6	<0.001
After 1960	274	21–722	79	1.6	
Main building materials					
Concrete	101	28–722	90	1.8	
Stone, bricks	196	21–463	83	1.7	<0.001
Tuff	131	21–420	107	1.7	

GM = geometric mean; GSD = geometric standard deviation; p-value = significance level observed for analysis of variance for comparing among groups.

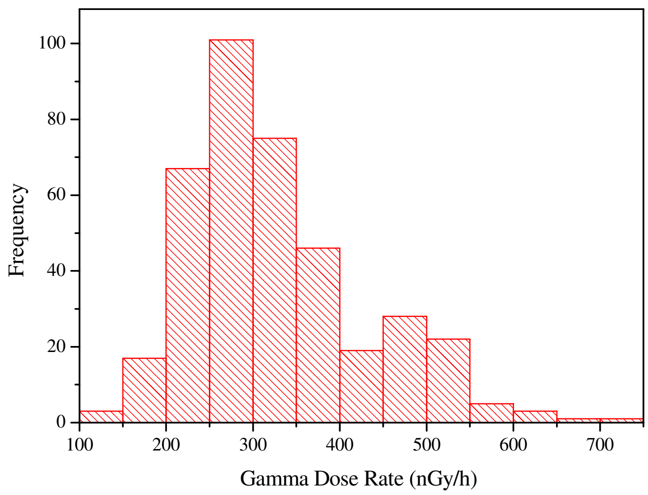

Figure 2. Frequency distribution of indoor gamma dose rate in dwellings of province of Naples.

dependence on the floor ($p < 0.001$). The Bonferroni post-hoc test indicated that the radon concentration of basements and ground floors were higher than those measured in the first and upper floors. Building materials are the second source for indoor radon due to ^{226}Ra content and the exhalation rate of radon from materials. For this reason, an analysis of the influence of different building materials on radon concentration was performed. Data were grouped into three categories based on the different materials: i) concrete, ii) stone and brick, and iii) tuff. The ANOVA analysis showed that the highest radon concentrations correspond to dwellings made of tuff and the difference between houses built with different materials is statistically significant ($p < 0.001$). Also, the influence of the age of the building on radon specific activity was estimated by grouping the houses into three categories i) built before 1919, ii) built between 1919−1960, and iii) built after 1960. The ANOVA analysis demonstrated that the annual radon concentrations are significantly higher for older houses compared with those built after 1960 ($p < 0.001$). The higher values reported for older houses could be explained by the fact that the ventilation rate in older houses is less than the new ones because they generally have smaller or fewer windows. Moreover, in the old buildings there is not good air tightness between the soil and first floor.

3.2 Indoor gamma dose rate

The frequency distribution of the gamma dose rate in air from terrestrial sources measured in the 388 houses is shown in Figure 2. The gamma dose rate indicates a normal-like distribution although the Shapiro-Wilks test failed to assess normality ($p < 0.001$). The fraction related to terrestrial sources was estimated by subtracting from the total value the contribution of the cosmic component, using Sakellariou's model (Sakellariou *et al.*, 1995). The arithmetic mean was found to be 327 ± 102 nGy h^{-1}, while the lowest value measured was 136 nGy h^{-1} and the highest 702 nGy h^{-1}. The mean value of the indoor gamma dose rate obtained is higher than the national average, which is 105 ± 10 nGy h^{-1} (Bochicchio *et al.*, 1996a).

Table 2. Statistical analysis of influence of major factors on the gamma dose rate.

Main building materials	*N*	Range (nGy h^{-1})	AM± SD (nGy h^{-1})	*p*-value
Concrete	59	152–593	299 ± 80	
Stone, bricks	37	216–524	322 ± 71	<0.05
Tuff	24	258–506	354 ± 65	
Materials lining the floor				
Tile	215	136–622	304 ± 88	
Wood	10	192–466	359 ± 79	0.2030
Granite	13	267–388	321 ± 36	
Marble	46	158–515	315 ± 88	
Materials lining the walls				
Wallpaper	88	136–587	355 ± 104	
Plaster	269	136–701	316 ± 701	<0.05

AM = arithmetic mean; SD = standard deviation; *p*-value = significance level observed in the analysis of variance for comparing groups and Student's t-test.

This result could be due to the presence of active volcanoes in the coastal area of the Province of Naples, with the consequence of an elevated presence of undifferentiated pyroclastic rocks (Ortolani and Pagliuca, 1987), widely used in building construction.

The main source of indoor gamma radiation is the building materials and the materials lining the walls and floors of the rooms. To assess the effect of these factors on the gamma dose rate measured, the dwellings were categorized into three groups according to the main building materials: cement, brick and stone, and tuff. The ANOVA analysis shows that the mean values of the indoor gamma rate were significantly different for dwellings with different main building materials. The results of the analysis of factors affecting the gamma dose rate are reported in Table 2.

The Bonferroni post-hoc test shows that the mean values of the indoor gamma dose rate are significantly higher for dwellings built of tuff compared with those constructed of concrete ($p < 0.001$), while no statistically significant difference was found between the gamma dose rate measured in houses built of tuff and those built of brick and stone. The materials lining the walls are plaster and wallpaper, while for the floors the materials used are tiles, wood, granite and marble. The statistical analysis (ANOVA) revealed no significant differences among the various coating materials of floors ($p = 0.2030$), while Student's t-test shows that houses with walls covered in wallpaper have an average level of gamma dose rates greater than those with walls made of plaster.

In order to find a relationship between the indoor radon concentrations and indoor gamma dose rates, correlation analysis between these two variables measured in the same room was analyzed and is reported in Figure 3. A correlation coefficient of 0.1921 was found, which indicates a very weak

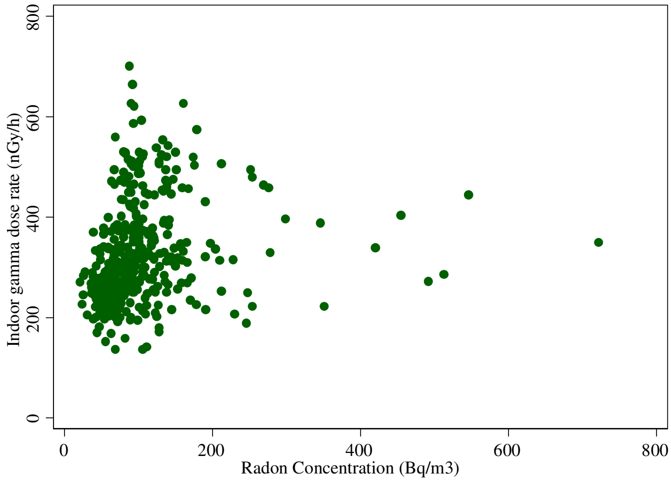

Figure 3. Scatter plot of indoor gamma dose rate versus indoor radon activity concentration for 388 dwellings where simultaneous measurements of radon concentrations and gamma dose rate were performed in the Province of Naples.

positive correlation. Because the indoor radon concentration and gamma dose rate have different distribution, a log conversion was applied to the radon concentration: in this case, the correlation coefficient becomes 0.052. Applying the statistical Fisher test to the linear regression model between the two variables, a p-value <0.01 was obtained, which shows that the model used may explain a significant proportion of the variance of the phenomenon.

The results show that the highest values of the gamma dose rate were measured in correspondence with values of radon that were relatively not high (<200 Bq m^{-3}). This finding might suggest that radon and its progeny contribute little to the gamma dose in air. Some authors (Iimoto *et al.*, 2001; Sundal and Strand, 2004; Pilkyte and Buktus, 2005; Kurnaz *et al.*, 2011) report that the indoor radon concentration has a coarse correlation with the gamma dose rate; the reason for the difference could be attributed to the high ventilation rate in the dwellings, but unfortunately, this information was not collected during the survey.

3.3 Radiological effect estimates of the indoor gamma dose rate and radon

To evaluate the effective dose received by the adult population, the UNSCEAR model (2000) was used, that assumes an indoor occupancy factor of 0.8 and a conversion coefficient of 0.7 Sv Gy^{-1}. The annual effective dose was calculated by the following equation:

$$H_{\text{gamma}} = D \times T \times C \times O \quad (2)$$

where H_{gamma} is the annual effective dose from the indoor gamma dose rate (mSv y^{-1}), D the indoor gamma dose rate in nGy h^{-1}, T the time in hours (8760 h y^{-1}), C the conversion coefficient (0.7 Sv Gy^{-1}) and O the occupancy factor (0.8).

The estimated annual effective dose by the gamma dose rate in air was found to be 1.6 mSv y^{-1}, which is higher than the Italian national average (0.4) mSv y^{-1} (Bochicchio *et al.*, 1996a). Moreover, taking into account the UNSCEAR model, the annual effective dose to the general population due to radon was calculated using the following formula:

$$H_{\text{radon}} = C_{\text{Rn}} \times F \times O \times T \times C \quad (3)$$

where H_{radon} is the annual effective dose from indoor radon, C_{Rn} the indoor radon activity concentration in Bq m^{-3}, F the equilibrium factor (0.4), O the occupancy factor (0.8), C the dose conversion coefficient (1.4×10^{-8} mSv per Bq m^{-3} h) and T the time in hours (8760 h y^{-1}).

The dose conversion coefficient reported in formula (3) is obtained by equating the lung cancer risk of 8×10^{-10} per Bq m^{-3} h with the total detriment for the general population reported in ICRP Publication 103 (2007), which is 5.7×10^{-2} Sv^{-1}. The dose coefficient obtained using the lung cancer risk reported in ICRP Publication 103 is approximately double that obtained with the value reported in ICRP Publication 65 (1993) and about 1.5 times that reported in UNSCEAR (2000). In this study, the annual effective dose due to indoor radon was found to change from 0.8 to 18.2 mSv y^{-1} with an average of 4.2 mSv y^{-1}. These values are higher than the Italian national average value, which is 1.2 mSv y^{-1} (Bochicchio *et al.*, 1996b).

4 Conclusion

In this study, the indoor radon concentration and indoor gamma dose rate were investigated in some dwellings of the Province of Naples, South Italy. For radon, values of 107 Bq m^{-3} and 91 Bq m^{-3} for the arithmetic and geometric mean, respectively, were found. For the gamma dose rate, a mean value of 327 nGy h^{-1} was found. A weak correlation was found between the indoor radon activity concentration and indoor gamma dose rate. The results show a relationship between radon concentrations and some building characteristics such as the age of dwellings, floor of the room monitored and building materials. Moreover, the indoor gamma dose rate was influenced by building materials, but no correlation was found with the coating materials of floors. The average value for the annual effective dose due to radon was found to be 4.2 mSv y^{-1}, based on the estimated annual radon activity concentration, while the annual effective dose due to indoor gamma exposure was found to be 1.6 mSv y^{-1}.

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