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A Statistical Analysis of the Follow-Up Study of Radon Measures in Private Homes and Public Buildings Carried out in Four European Towns with an Esteem of Lung Cancer Risk

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ABSTRACT

Buildings can artificially concentrate radioactive radon gas of geologic origin, exposing occupants to harmful α particle radiation emissions that damage DNA and increase lung cancer risk. We investigated how radon exposure varies in public and residential buildings by floor, geological soil, by occupant behaviour and season in two year-long measurement campaigns in four European towns (Torino twice, Bergen, Lund and Reykjavik) based upon CR-39 detectors. Radon concentration data were analyzed using multiple approaches coupling geologic, geographic, architectural, seasonal data with advanced statistical tests. Soils investigation about the concentration amount of the two main radon ancestors, uranium and thorium, were carried out in sample collected near the measurements points. Measurements of radon concentration by a gamma spectrometer were done in water samples taken around Torino too, in order to give an evaluation of another possible radon source. Uncertainty on average radon concentration has a large impact to define an efficient and integrated strategy for national radon control in order to reduce human exposure. Radon as well as its progenies account for around 50% of the total effective dose received from all sources of natural radiation (around 2.4 mSv annual dose) and is responsible for a sensible fraction of lung cancer and leukemia. All the results are shown and discussed and an evaluation of the cancer and leukemia risk related to radon exposure data has been computed.

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Introduction

We are living surrounded by radiation emitters, which are part of our environment and the flux of natural radiation is the primary source of human non-medical exposure to ionizing radiation. The main component of the population dose is due to the exposure to radon as well as its progenies, which account for more than fifty percent of the total effective dose received from all sources of natural radiation (around 2.4 mSv annual dose) [1]. Epidemiological studies suggest that exposure to radon leads to an increased risk of lung cancer. Radon is a radioactive, odorless and colorless noble gas, Uranium and Thorium are the first ancestor elements in a long series of decays that produce all the radon isotopes. Radium and radon also form other daughter elements as they decay, such as ^{238}U that has a half-life of 4.4 billion years: ^{222}Rn comes from its decay chain, arising directly from ^{226}Ra. ^{222}Rn, the longest half-life of radon isotopes, has a half-life of 3.8 days and is an alpha emitter. The second most important radon isotope, ^{220}Rn, that has a half-life of 55 seconds, is the progeny of ^{232}Th that, like uranium isotopes, is distributed throughout the environment. It also has a very long half-life (1.41 x 10^{10} yr) and decays by emission of an alpha particle generating a series of radioactive daughters, many of which emit alpha radiations too. There are three known natural isotopes of radon, ^{222}Rn and ^{220}Rn are both alpha emitters and are the most significant isotopes to health, because of the much shorter half-life of all the others (see Table 1).

Table 1: Radioactive Properties of Natural Radon Isotopes

Isotope	From:	To:	Half life	α radiation
^{219}Rn	^{223}Ra	^{215}Po (Actinium series)	3.9 s	6.82 MeV (82%), 6.55 MeV (13%)
^{220}Rn	^{224}Ra	^{216}Po (Thorium series)	54 s	6.28 MeV
^{222}Rn	^{226}Ra	^{218}Po (Uranium series)	3.8 d	5.49 MeV

Among radon decay products there are alpha and beta radioactive isotopes that combined with dust and aerosol particles present in the air can be aspired and deposited in the human respiratory system where they decay and become a source of significant radiological exposure [2,3]. Alpha particles (α) could ionize and damage the DNA found in the cells of living organisms. Alpha radiation has a very small range and is blocked by human skin, but it can be a carcinogen if it enters the body throughout the respiratory tract and radon can be easily inhaled. Beta (β) and gamma (γ) radiation penetrate deeper into the body and can also cause damage to genetic material, leading to the development of a malignant tumor , but with a lower coefficient of risk [4,5]. Radon, as a noble gas, is generally chemically inactive: it is a heavier atom that can be found in monoatomic state only, so its size is much smaller than any molecule and it has a very high penetrability, it is quite mobile and can easily migrate both in the Earth's crust and in the air. The inhalated radon comes primarily from the soil (which contains the radon ancestors U and Th), building materials, water and natural gas. Radon is transported to the Earth's surface by diffusion and convection. The amount of radon exhalation strongly depends upon the location (soil type, soil geology) and atmospheric conditions (pressure, wind strength and direction, humidity, snow cover, etc.). The extent of exhalation is also closely correlated with the presence of tectonic faults. Such faults are an excellent path for radon migration to the surface even from very deep geological layers. Granite soils show high uranium content, while the content in sedimentary rocks is usually lower. Active seismic zones, tectonic movement areas, volcanic zones and geothermal fields are significant sources of radon. In the four European areas where we carried out the year-long survey (Bergen area, Lund area, Reykjavik area and Torino area twice), radon mainly occurs where granitoid massifs and metamorphic rocks with higher uranium and thorium contents are found. The U and Th concentration in soil have been evaluated and discussed (see Table 3 in the following). In Torino important radon concentration was also measured in buildings over tuff stone soil. The average value of ^{222}Rn concentration in tuff samples collected in the Torino hillside were found significantly higher (up to 60%) than the other rock sample averages collected in Torino. In general rocks with a mosaic geological structure, with a lot of cracks, brittle rocks and tectonic dislocations make it easy for the radon gas to move upwards and this movement is also made easier by flowing groundwater and carbon dioxide. In Piedmont for instance, the region where Torino is located, a very high radon concentration is found in low-mineralized waters with a groundwater flow period of several years (Lurisia sources). The link between groundwater, radon concentration and radon emanation was studied in Torino area measuring radon concentration in wells and groundwater using a gamma ray spectrometry based on a HpGe detector, 60% efficiency, shielded by 10 cm lead against cosmic rays and natural background. The radon concentration was evaluated from the γ peak emission of its ^{214}Bi progeny at 609.3 keV (45.5% of the total gamma emission of ^{214}Bi), 1120 keV (14.9%) and 1764 keV (15.3%). ^{214}Bi has a half-life of 19.7 min and is always in equilibrium, because of its short half-life, with ^{222}Rn. From the evaluated amount of ^{222}Rn a correction to take into account the contribution to the total concentration given by ^{220}Rn has been applied. In water sources around Torino and in Torino suburbs (Italy) we measured within 160 ÷ 190 Bq/L; in well waters (10 wells were analyzed) the radon concentration ranges from 72 up to 163 Bq/L, the maximum values was found in wells dig in tuff placed at Torino hillside. In surface waters (natural ponds and reservoirs) of the Torino area we measured from 1 to 9 Bq/L and in drinking water of municipal aqueduct < 2 Bq/L. The reasons of this big spread about radon values in waters lies primarily in the geology of the soils, i.e the amount of uranium and thorium, the movement of radon throughout soil gas and ground water. Highlights that the concentration of radon in spring and hand pump water is influenced by the concentration of ^{226}Ra in rocks and the abundance of ^{238}U minerals, despite the relatively low solubility of radon in water, that is strongly related to the temperature of water too [6]. In Table 2 the radon coefficents of solubility in water and other liquids are shown, the concentration of radon in atmospheric air outdoor is usually low.

Table 2: Chemical-Physical Properties of Radon

Properties	Value
Melting point	-71.0 °C
Boiling point (1 atm pressure)	-61.8 °C
Density (1 atm, 20°C)	9.96 kg/m^3
Specific heat (at 20°C J/g mol)	0.094
Evaporation heat (kJ/mol)	18.1
Coefficent of solubility in water at 1 atm pressure	
T=0°C	0.57
20° C	0.250
37°C	0.167
100°C Coefficent of solubility at 1 atm and T=18°18	
Hexane	16.56
Liquid paraffin	9.20
Toluene	Toluene

Radon Distribution in Europe

If we compare the indoor radon concentration measured in the four European towns where the survey was carried out, we notice that it varies from a few to several dozen Bq/m^3. In houses, this concentration can be much higher up to several thousand Bq/m^3 [7]. Data covering the 27 European countries seem to show that radon is responsible up to around 8% of lung cancer deaths, a new evaluation of lung cancer risk related to indoor radon concentration is shown from our measurements [8]. In the UK only, the proportion is estimated at 3.3% (0.2% of all deaths). The US Environmental Protection Agency (EPA) estimates that

1 in 15 homes in the U.S. (about seven million) has radon levels over safety threshold [9]. Since 2006, environmental and indoor measurements have been promoted in all Europe to create an atlas with the average indoor radon concentrations in rooms at ground level. In the Torino area, during the second yearlong campaign we tried to find a correlation with the help of the statistical data analysis between indoor radon concentrations and the floor of the room where the measures was done. The distribution of results strongly correlates with the geological conditioning of the site. In our measurement campaigns we observe the highest values in the Bergen area and in some local area of Torino, associated with granitic soil and, for Torino, tuff stone. A special consideration need Reykjavick, built on a vulcanic soil, placed not far from active vulcanoes, but with a very low radon concentration, both indoor and outdoor. The Iceland rocks, mostly igneous, show a very low uranium and thorium concentration (see Table 3, our data fully confirm this statement). In spite of its vulcanic origin, the Iceland values are the lowest of Europe and one of the lowest in all the world. The arithmetic mean for all Europe (from the available data, that nowadays are really a lot, even if not distributed in a homogeneous way on all the European territory: in fact there are still area with almost no measures) was 98 Bq/m^3, and the median was 63 Bq/m^3, but there were large variations between different countries: for example, in the Czech Republic over 90% of the country's area had an average indoor radon concentration above 100 Bq/m^3, while in Lithuania this percentage did not exceed 10% and in Reykjavick the concentration is so low that is hard to measure it. Because of the close link between the geology of the soils and radon concentration, during the measure campaigns, estimation of the amount of uranium and thorium in rock samples collected nearby the measurements points was done using the gamma ray spectrometer before described for water measurements. U concentration in different rock types is measured from 1.001 MeV gamma peak emitted from ^{234}Pa which is a second daughter nuclide of ^{238}U series and is always in equilibrium with the parent U. This peak is well resolved by HpGe detector and gives accurate enough concentration of U in the samples. Since the yield of 1.001 MeV γ energy is low (0.6%), the sensitivity obtained by using IAEA standard RGU-1 is 0.4 count/ppm/5000s for 50g of sample weight. Thorium concentration is evaluated from ^{228}Th isotope decay chain, exploiting its β decay branch (half-life of 1.911 y) by detecting 911.6 keV and 969.1 keV γ peaks from ^{228}Ac decay (26.6% and 16.6% respectively). In Table 3 it is shown the results of uranium and thorium concentration in the rock sample collected near the radon measurement points, all figures have an estimated uncertainties of ± 5 ppm.

Table 3: Uranium and Thorium Concentration in ppm by γ Detection Method using a 60% Efficency HpGe Spectrometer. Each measure has an estimate uncertainty of ± 5 ppm (from [7]).

Area	U	Th
Torino Area		
sedimentary rock	12	21
limestone	19	23
tuff stone 1	23	34
tuff stone 2	39	47
Reykjavik area		
basic igneous stone	< 5	< 5
basaltic rock	< 5	< 5
tholeiite	< 5	< 5
Bergen area		
sedimentary rock	26	68
granite 1	63	90
granite 2	69	97
Lund area		
granite 1	32	48
limestone	51	83
sandstone	29	42

The results for the igneous rocks of volcanic origin from Reykjavik is amazing, even if well known. Comparing these results with the U and Th amount present in the igneous rocks collected from all the Mediterranean Volcanos (Stromboli and Vulcano in the Eolian Islands, Etna in Sicily and Mount Vesuvio near Naples), the Reykjavick rocks show one or two order of magnitude lower [10,11]. With the same experimental method, we carried out a measurement on three lava rocks from Mount Vesuvio to have a direct comparison: we found U concentration ranging from 29 up to 92 ppm, from 36 up to 107 ppm for Th, with an uncertainty evaluated of around 18%.

Indoor Radon Concentration

Radon escaping from the Earth's crust and into atmospheric air can penetrate buildings. The share of radon in the air inside a statistically representative building, assuming full air exchange every hour, is as follows: the soil accounts for nearly 80% of the radon source, the second source is in building materials responsible for 12% of radon, and the third is atmospheric air accounting for about 9.3% [12]. Water and natural gas together account for less than 1% [13]. Radon is much heavier than air (7.6 times) and should remain in the basement layer but the foundation of the building requires "penetrating" the soil surface and reaching deeper layers, where radon concentrations can be supposed to be much higher. The basic mechanism for the entry of this gas into houses is always the pressure difference between the inside and the outside: usually the pressure inside is a few pascals lower than outside the building. This phenomenon is caused by devices, such as those for air conditioning or ventilation, which work in a house and "pump out" the air. Another reason is the fact that the house has in autumn and winter a higher temperature than outside: warmer air is lighter and thinner, it produces less pressure and causes radon to escape from the soil (or wall) and rise. Increasing the indoor pressure, making it around 1% higher than outside pressure or even less, is the most efficient method to tackle down radon entering the houses. Radon could also move from the lower rooms and building walls up. The main ways in which radon penetrates inside buildings are leaks in the building, such as cracks and crevices of the concrete screed, structural gaps and cracks in the walls. Another source of radon in houses is its escape directly from walls and ceilings made of materials which contain some amount of radium, uranum or thorium. Generally, it can be stated that radon from the ground dominates on the lower floors of the building, and the higher the floor, the greater the share of radon coming from building materials used in the construction. Higher levels of radioactivity are found in industrial raw materials: fly ash, slag and concrete but building materials are in general a less important source of radiation than the soil on which the building is erected [14]. Some energy conservation interventions can influence the indoor radon concentration: an example of such a phenomenon is thermomodernisation of buildings which results in worsening ventilation, leading in the end to an increase in radon concentration. In France, Czech Republic and Russia the

concentration of radon after sealing the buildings increased by 1.6 times, and in England, 1.7 times [15]. Between the source of radon we mention also water because radon is released from it (in small amount) when domestic sanitary facilities (e.g., showers, baths) are used (see also Table 2 and the variation of the radon solubility coefficient in water with temperature). The concentration of radon inside buildings shows high daily and seasonal variability. In autumn and winter, when temperatures are low outside, doors and windows remain closed most of the time and radon concentrations in rooms reach much higher values.

The content of radon inside a building occurs by diffusion and convection, the latter, like advection, depends upon temperature and increases with its gradient [16,17]. It is estimated that airing the room can reduce radon levels indoor by up to 70% [18]. In recent years cellar conversion and new basement creation is widespread in Italy where basement living and working is frequent. In the Torino University, building basements are often used as classrooms, laboratories, archives or other working activities. All basements are potentially at risk of elevated radon levels. During the second survey campaign we investigated the radon concentration in ground floor, in some basements and underground floors used by the activities of Torino University as required by Italian law Dlgs101/20 [26]. An integrated strategy or action plan for national radon control is needed to effectively reduce human exposure.

Experimental

The measures were done by CR-39 out in four European towns: Bergen, Lund, Reykjavik and Torino, budgeted under the European contract EURA [19]. CR-39 detectors were placed for six months mostly in private houses and some schools at the room ground [7]. In the Torino area a second yearlong campaign was done from February 2022 up to March 2023. The second campaign was dedicated to measure radon concentration inside buildings of University of Torino used for the University activities. This time each CR-39 detector was placed between 1 m up to 2 m from the room ground of University buildings, according to ISO-1665 rules and to a direct request from the University Dosimetry Service, with sample time of four months to have more details about seasonal variation with respect to the first campaign [7]. Along the second measurement campaign the minimum possible measurable concentration is around 7 Bq/m^3 in four months exposure, lower down to 1÷2 Bq/m^3 in the first one [20]. In total 19 buildings of University of Torino spread around the city were monitored, the CR-39 detectors were placed in one room at the ground floor, in 104 rooms in the building basement, in 45 rooms at first underground floor, in 16 rooms at -2 floor and 11 rooms at -3 floor, for a total of 177 rooms monitored during all the second survey campaign. According to ISO 1665-8 rules, we established at each monitored room of the second annual survey the average value for radon concentration equal to the mean value measured by at least two CR-39 detectors when the result of the single measure shows a difference less than the computed uncertainty. As a general rule, a CR-39 detector was placed every 50 m^2 room surface, for very big local like laboratories or archives a detector was placed each 100 m^2. On the other hand, when the difference was higher or much higher than the uncertainties, we take the bigger concentration value. Being x_{max} and x_{min} the highest and smallest concentration value recorded in the same room, we have:

$$(x_{max} - x_{min}) \leq \varepsilon_{min} \Rightarrow \frac{\sum_{k=1}^{n} x_k}{n} \pm \frac{\sqrt{\sum_{k=1}^{n} \epsilon_k^2}}{n},$$

$$(x_{max} - x_{min}) \leq \varepsilon_{min} \Rightarrow Max(x_1, x_2, \ldots, x_n) \pm \varepsilon_{x_{max}}$$

Where ε_{min} is the lowest uncertainty between two single measures, x_k is the arithmetic mean concentration computed by the values of each CR-39 detector in a four-month exposition time in the room. We consider a time interval of four months exposure for each CR-39 in order to study the seasonal fluctuations. In the first campaign each CR-39 detector was exposed for six months, and we take the average of every detector placed in the same room as radon concentration value. In the second survey, to compare the results of the measures in the different buildings and rooms, placed at different level with respect to the ground, we decide to normalized the single room data at the average annual concentration for this same measurement point. The data normalization for any rooms with the computing of their uncertainty has been carried out by the following formulas:

$$\frac{C_1}{C} \pm \sqrt{\left(\frac{\varepsilon_1}{C}\right)^2 + \left(\frac{\varepsilon_C \cdot C_1}{C^2}\right)^2}$$

$$\frac{C_2}{C} \pm \sqrt{\left(\frac{\varepsilon_2}{C}\right)^2 + \left(\frac{\varepsilon_C \cdot C_2}{C^2}\right)^2}$$

$$\frac{C_3}{C} \pm \sqrt{\left(\frac{\varepsilon_3}{C}\right)^2 + \left(\frac{\varepsilon_C \cdot C_3}{C^2}\right)^2}$$

where C_1, C_2, C_3 are respectively the arithmetic mean concentration value recorded at the first, second and third-year quarter (four-months period) for each room and ε_1, ε_2, ε_3 are their uncertainty, C is the arithmetic mean annual concentration of each single room (computed from all the CR-39 exposure values) and ε_C its standard deviation. In Table 4 the normalized values over the three four-months periods taken into account are shown.

Table 4: Results of the Data Normalization Over the Three Four-Months Periods Taken into Account in the Second Torino Measurement Campaign

Period	C_P / C_{year}	min.	Max.	Median
Feb - Jun	0.80±0.28	0.29	1.98	0.78
Jun - Oct	0.96±0.34	0.26	2.22	0.95
Oct - Mar	1.23±0.36	0.09	2.11	1.24

First Measurement Campaign

For each town we had different values of radon concentration, and even in the same town the spread is ranging from 4 up to 1835 Bq/m^3 for Bergen, from 9 to 1059 Bq/m^3 for Lund and from 6.7 to 63 Bq/m^3 for Torino [7]. A peculiar situation could be observed for Reykjavik, where in spite of the soil of igneous volcanic nature the level of radon is very low, ranging from 1.6 to 48 Bq/m^3: most likely Iceland has the lowest level of radon in Europe if not in the world. In the two towns where the radon peak shows problematic figures (Bergen and Lund) the uranium and thorium concentration in soil and rocks has values between two up to three times higher than what has been measured in Torino (see Table 3): if the case of Bergen could be explained because the town is built over granitic rocks with the highest concentration of U anf Th, Lund instead is mainly built on a sedimentary soils (limestone and sandstone) that nevertheless show good U and Th presence. As far as we know only a restricted portion of the Lund soil is granite rock. We found higher values of radon concentration in sandstone samples and lower values, around 10% less, in limestone samples. Soils and rocks with higher permeability such as sandstone, limestone and tuff have larger pores and fissures allowing radon to move more easily. On the other hand, soils and rocks with low permeability such as clay and shales (that is the composition of the Torino soil near the banks of the river Po and in the plane zone, while in the

Torino hillside we have soils rich of tuff stone) tend to retain radon and limit its mobility. The permeability of the soil and rocks plays a crucial role in the movement of radon through the environment and have a significant impact on the indoor radon concentration. Granitic rocks, despite showing low porosity, may display high permeability if highly fractured. On the other hand, volcanic rocks can have high porosity due to the presence of vesicles, yet their permeability is often constrained by a lack of connectivity among these pores. Limestones can exhibit a wide range in permeability, from very low in microcrystalline limestones to very high in fractured limestones or those with substantial intergranular porosity: this is the main reason of variability of radon values in sedimentary rocks. Since the typical organization of these rocks in layers, the dip of these ones in the soils could also contribute to increase the variability of indoor radon concentration. For Reykjavik the very low radon levels is in full agreement with such a low concentration of uranium and thorium in soil and rocks (even if they are volcanic rocks) and we were able to give only an upper limit: the uranium/thorium concentration was under the sensibility of our instrumentation. Even considering samples of hot and cold water for radon analysis collected from boreholes and springs in Iceland in 2014 and 2015, radon activity is generally rather low, in most cases less than 5 Bq/L [13]. Only 8% of water samples had a measured radon activity higher than 5 Bq/L, with a maximum activity of 10.8 Bq/L [13]. If we compare the radon concentration in surface (cold) waters of Torino area (between 1 to 9 Bq/L) that is not at all volcanic, we can see the strong peculiarity of Iceland. The geographical distribution of the samples (rocks and waters) shows that in Iceland radon activity is generally lower within the active rift zones. This is most likely due to the very low uranium content of the tholeiites typically erupted within the rift zones. Higher radon values (> 5 Bq/L) are in most cases close to extinct central volcanoes and thus, in agreement with our basalt rock measures (< 2.0 mg of U and Th for each kg of sample, translating the unit of measures reported in Table 3 with an uncertainty of more than 100%). It seems plausible that the water sampled has been in contact with felsic rocks. For Reykjavik urban area we measured an indoor radon levels with an arithmetic mean of 11 Bq/m^3 with a standard deviation of 8.5 and a geometric mean of about 8.1 Bq/m^3 with a median absolute deviation (MAD) of 5.9 Bq/m^3 (see Table 5). The close values obtained for arithmetic mean and geometric mean can be seen in Rejkyavick and Torino data only. Measures done in the other two towns show a very different situation indeed. In Torino during the first campaign in private houses we measured an arithmetic mean of 26.6 Bq/m^3 with a standard deviation of 11.2 and a geometric mean of 24.59 Bq/m^3 with a MAD of 7.41 Bq/m^3. The large spread of values in the radon data set suggests to evaluate the statistical dispersion by MAD that is a better statistic estimator, being more resilient to outliers in the data set than the more common standard deviation. In fact, when using standard deviation, the distances from the arithmetic mean are squared and then large deviation are weighted more heavily (even overestimated) and thus outliers could heavily influence it. In MAD the deviations of a small number of outliers become irrelevant. Only 5% of the Reykjavik measurements show an indoor radon concentration of more than 30 Bq/m^3 out of a total of 450 measurement points. The cities of Bergen and Lund have a mutual comparable situation: measures were done in more than 240 private houses and public building for each town and the geometric mean of indoor radon concentration is 31.16, 53.5 Bq/m^3 respectively, with 37.06 and 60.79 Bq/m^3 for the associated MAD. The arithmetic mean is 87.7 Bq/m^3 with a standard deviation of 207.7 Bq/m^3 for Bergen and 102.6 Bq/m^3 with a standard deviation of 154.8 Bq/m^3 for Lund (see Table 5) [7]. A very big concentration spread indeed and this is the proof of how different could be the geophysical conditions related to radon, even in a limited area. The results from uranium and thorium concentration in the samples rocks collected in the two towns show a variation up to 100% (see Table 3).

Table 5: Arithmetic Mean, Standard Deviation, Geometric Mean and Statistical Dispersion by Median Absolute Deviation (MAD) of Measured Radon Concentration (from [7])

Town	Arithmetic mean Bq/m^3	Standard deviation Bq/m^3	Geometric mean Bq/m^3	MAD Bq/m^3
Bergen	87.7	207.7	31.16	37.06
Lund	102.6	154.8	53.5	60.79
Reykiavik	11.0	8.5	8.1	5.9
Torino	26.6	11.2	24.59	7.41

The distribution of indoor radon concentrations follows more or less closely a log-normal distribution, but, at least for our data, not a standard log-normal distribution. We had to work out a specific "ad hoc" version. The standard log- normal distribution gives poor results of the χ^2 test applied to the histogram fit. For buildings situated in soils or geological locations with a consistent source of radon in the ground, conformance with the log-normal distribution can be understood in terms of the multiplicative factors affecting the relationship between uranium-thorium concentration in the ground. The fact that local distributions of indoor radon usually conform to the log-normal can be explained because the mixture of a number of different log-normal distributions will often result again in a log-normal distribution. But in some cases (that are not so rare), the distributions of indoor radon could deviate from a log-normal distribution and this is more likely to happen, according to our observations, for the radon concentrations above 300 Bq/m^3. The fits using our developed log-normal distribution of all the measured annual average radon concentrations in the four town are shown in Figure 1 [7]. The probability density function (pdf) of the log-normal distribution that we used is log-norm.pdf (x, σ, f.loc, S) , where x are the measured radon concentration data (x > 0), σ is the shape parameter and it is also the standard deviation of the log of the distribution (σ > 0), f.loc is the location parameter evaluated by each fit (see Figure 1) and S is the median of the distribution, known also as scale parameter (S > 0). μ = log(S), the μ parameter is the mean of the log of each distribution.

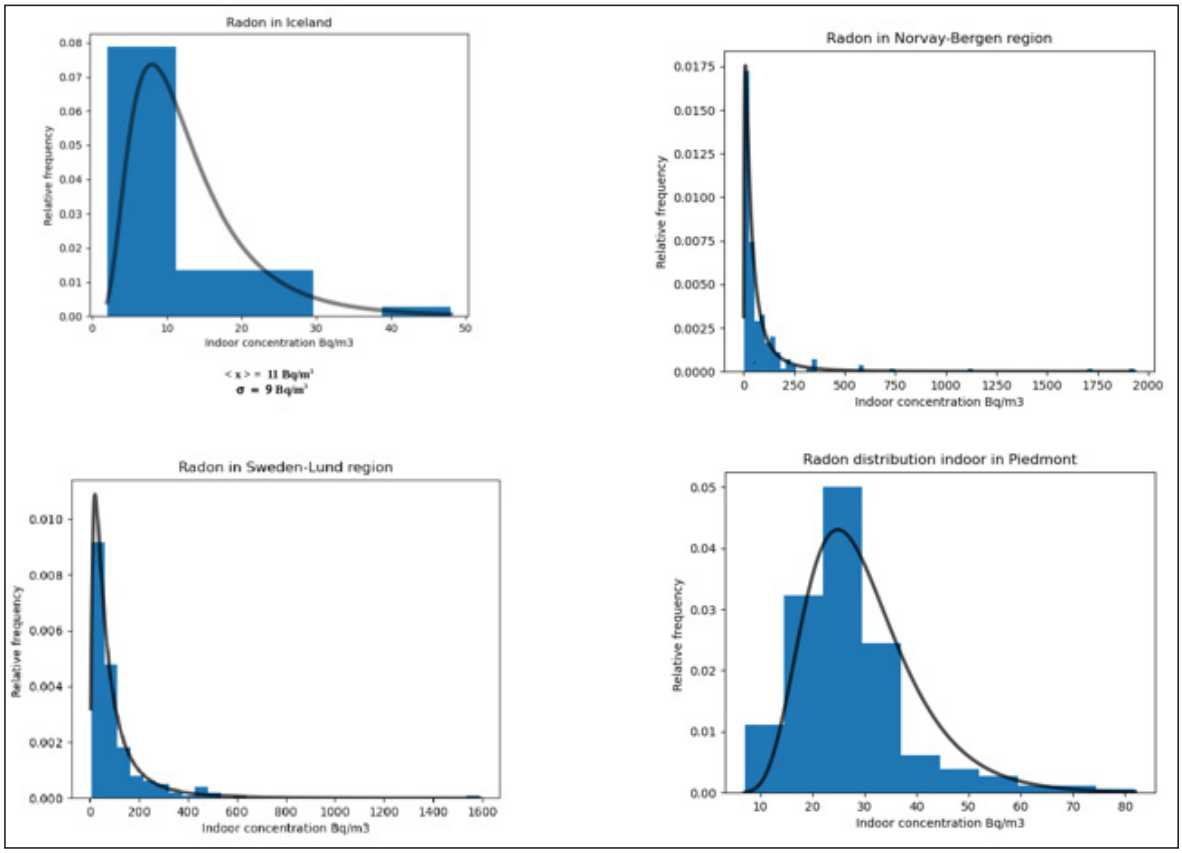


Figure 1: Indoor Radon Distribution in the Four European Towns Involved in the EURA Project

The full black line in each graph shows the log-normal distribution of the experimental data (see text for more details about the log-normal parameters and [7]).

It is apparent as the Torino situation is in the middle between the Reykjavik and Bergen – Lund. Torino (Piedmont) soil is mainly composed by sedimentary rocks and limestone, with a high percent of clay and shales near the river Po banks. Tuff stone is present in the hillside, in general there is few granite rocks around. Many Torino University buildings are placed nearby the river Po banks. The uranium/thorium percentage in the limestone rock of Torino area shows a clear lower concentration with respect to similar rocks collected in Lund and the same is for the sedimentary rocks collected in Bergen: this has a direct effect on the overall radon concentration. The highest values of radon concentration measured in Torino is in the tuff stone soil zone. The radon concentration measures for the Bergen area and Lund area show a similar situation, with the peak of highest radon concentration found in the Bergen area, in full agreement with the uranium/thorium concentration measurements. In fact, the Bergen granite where the highest uranium/thorium concentration values are found is by far the most common type of soil/rock in the Bergen area. In Table 6 the χ^2 tests applied to all the log-normal fits are shown and it is apparent that a good degree of confidence has been reached: the adopted log-normal distribution seems to work very well in describing the collected radon data in spite of the big spread of the values. Looking at the case of Bergen and Lund, we can see a sort of small bump at radon concentration over 300 Bq/m^3. In these points, local distributions might have very different means and standard deviation from the other measurement points. The log-normal fit is passing outside and just touching the lower part of the error bars of these points. If we consider this effect as an outlier ignoring it, we may underestimate its contribution to the total number of homes/buildings with very high radon values. We might conclude that the log-normal distribution at high radon concentration is different from the log-normal distribution below 300 Bq/m^3: the local distribution from high radon areas has different parameters to most of the data presented elsewhere. Therefore, the numbers of homes above a few hundred Bq/m^3 should be different to and greater than was predicted from national survey methods, based upon standard log-normal distribution. From the log-normal distribution (that according to χ^2 tests is within 95% agreement with all the collected data set) we evaluate the percentage of houses/buildings in each town with radon concentration levels higher than 100, 200 and 300 Bq/m^3, with the warning of a possible underestimation for concentration higher than 300 Bq/m^3. At the moment we do not have enough information to solve this puzzle, more geological and radon measures are needed. All the results are summarized in Table 7. In the town of Reykjavik and Torino is most unlikely to find buildings with more than 300 Bq/m^3 radon concentrations, but for Lund and Bergen we observe a situation with a sensible risk.

Table 6: χ^2 Test for Log-Normal Distribution fit. The α=1-p Value of 0.05 Correspond to a Confidence Level of 95% between the fit and the Experimental Data. The Evaluated End-Points Show the Upper Limit of each χ^2 test where ends the Agreement of at Least 95% between the fit and the Histogram, Taking into Account the Error Bars, the Degrees of Freedom, the Number of Bins and the Bin Length

Town	χ^2 Test	Degrees of freedom (ν)	Estimate end point (α=0.005)
Bergen	4.48	17	8.67
Lund	2.62	9	3.33
Reykjavik	0.14	4	0.71
Torino	2.35	9	3.33

Table 7: Percentage of Houses/Buildings in each Town with an Expected Radon Concentration Higher than the Reference Level. The Percentage is Computed from the Log-Normal Distribution Fitting the Radon Data set Collected in the First Measure Campaign

Town	>100 Bq/m³	>200 Bq/m³	>300 Bq/m³
Bergen	20%	8%	4%
Lund	31%	12.6%	6%
Reykjavik	0.1%	0.0001%	-
Torino	0.15%	0.0001%	-

Second Measurements Campaign at Torino

The second measurement campaign at Torino was dedicated to monitor for one yearlong workplace located in 19 buildings spread in the city center and used by Torino University activities. In all the monitored room, the concentration results are always less than 300 Bq/m³ and 92% of the monitored rooms show a concentration value less than 100 Bq/m³. We never found concentration values around zero, that means a background presence of radon inside the ground and the surrounding rocks. Even in this case we use an "ad hoc" log-normal distribution for radon concentration C, to explain the experimental results [21-23]:

$$f(C) = \frac{1}{\sigma \cdot \sqrt{2\pi}} \cdot \frac{e^{-\frac{(lnC-\mu)^2}{2\sigma^2}}}{C}$$

where:

$$\mu = \frac{1}{n}\sum_{i=1}^{n} ln(C_i) = ln(MG),$$

$$\sigma = \sqrt{\frac{1}{n-1}\sum_{i=1}^{n}[ln(C_i) - \mu]^2} = ln(DSG),$$

C_i are the year arithmetic mean radon concentration measured in each room over all the three quarters, MG and DSG are the geometric mean and geometric standard deviation of the overall measures. The median is equal to its multiplicative mean, i.e. MED(C) = e^{μ}=μ*. The geometric or multiplicative mean of the used log-normal distribution is:

$$MG(C) = e^{\mu} = \mu^*$$

in this case * is equal to 1, as it is apparent from the previous formulas, and it equals the distribution median. The geometric (or multiplicative) standard deviation DSG is:

$$DSG(C) = e^{\sigma} = \sigma^*$$

with * = 1. The mode or point of global maximum of the probability density function is:

$$Mode(C) = e^{\mu - \sigma^2}$$

The very big spread of values in the data set could affect the arithmetic mean and the standard deviation: in order to better describe our data, we chose as statistical parameters the geometric mean (more sensitive to the smallest values that are too often underestimated by the arithmetic mean ruled by the highest values) and the median absolute deviation (MAD) that is a robust measure of the variability of a sample of quantitative data and less sensitive to outliers values too. In Table 8 are shown the statistical parameters of the second survey data set with the absolute maximum value (Max) and the absolute minimum value (min). In Figure 2 the year average radon concentration in all the 177 rooms of Torino University monitored are shown. The log-normal distribution seen in Figure 2 is produced using μ = 3,17 ±0,08, σ = 0,63 ± 0,10 and f.loc = 1,13±0,38: this time f.loc is quite different from what has been computed from the previous measurement campaign (f.loc = 3.38) where mainly private house were monitored. Also, in this case the reduced χ^2 = 0,51, shows a good agreement from experimental results and the statistical model. Because the two set of data were collected in the same town (Torino), in buildings widespread around the town surface more or less homogeneously, we do not think the main reason of this variation is due only to geological reasons. The University buildings are in general older than private houses involved in the first survey, and between University buildings same are historical ones, but we think that the main reason of the big f.loc parameter difference in the two data set is due to the different destination of the monitored rooms (open to public activities in the second case) and, above all, in the second survey rooms in the basements and underground floors were involved: could f.loc parameter be sensitive by the room level (see Table 9)? At the moment we could not give any answer.

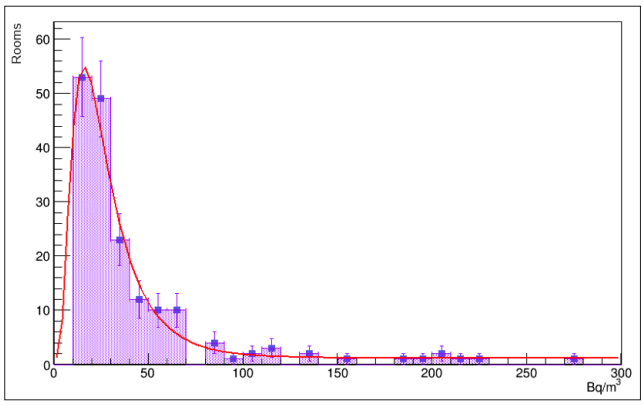


Figure 2: Average Year Concentration of ^{222}Rn Inside the 177 Rooms Monitored from 2022 up to 2023

Table 8: Statistical Resumé of the Annual Radon Concentration Distribution measured Inside Torino University Buildings

Arithmetic Mean	42.1 Bq/m^3
Standard Deviation	43.7 Bq/m^3
Geometric mean	31.1 Bq/m^3
MAD	9.7 Bq/m^3
Max	279 Bq/m^3
min	11 Bq/m^3

Table 9: Statistical parameters of year long radon concentration measures organized by floor

Floor	Number of rooms	Arith. mean	min.	Max.	Median	Geom. mean
Ground floor	1	35 ± 5	-	-	-	35
Basement	104	39 ± 43	11	279	25	29
Floor -1	45	58 ± 53	13	225	39	42
Floor -2	16	26 ± 8	16	45	25	25
Floor -3	11	29 ± 11	19	61	24	27

In the Torino University buildings, radon is found above all in the basement rooms or in the first underground rooms. In the previous campaign indication of the room floor was not recorded, in the second one the data are organized taking into account of the room level (see Table 9).

In Figure 3 the data of Table 9 are shown with a linear fit. The annual arithmetic average of radon concentration of each room has been used to normalized the values of every four-month exposure period, according with the method described in Paragraph 4, in order to enhance the seasonal effect. Piedmont has a continental climate, with hot and sultry summers and cold and humid winters, the rain is mainly concentrated in falls and springs. But in the last years we noticed a sensible variation from this climate model, with longer periods of drought (drought was very unusual since few years ago) and violent storms with increasing hail phenomena in late spring and along summer: most likely a change in the seasonal variation could be expected, new surveys are needed. Surprisingly the data show that radon is accumulated above all on the basement and in -1 floor, with a small decrease in the -2 level but in the inferior levels the radon concentration is kept almost constant, contrary to what we were expected. We can advance two hypotheses: a) this is purely a statistical effect due to the small number of monitored room in the deeper underground levels or b) radon is more likely migrating towards the surface when it has the possibility to reach the rooms placed on or just near the ground surface. Another effect that we have to consider is the presence of conditioning set to force the air changing, but we have these equipments in the deepest levels too: it could be only a matter of conditioning efficiency? In the Torino University buildings forced ventilation and climatization system are usually present in order to improve air quality in underground work places. This is not the case for work places over the ground. It is true that we measure the highest values of radon concentration where the conditioning set was for some period out of order, independently from the room level, but these are peculiar situations that have been taken into account during the analysis of data, together with the correction due to seasonal variation. In Figure 4 it is shown the year average radon concentration vs the level of the monitored room Even in this second campaign where public buildings were concerned, the deviations of the distribution of indoor radon data from the log-normal trend seen previously at high concentration values are apparent and are examined. According to simulation done over data taken in Belgium inside private houses these deviations could have origin from a local disequilibrium from the three main components of indoor radon concentration, that is radon from subsoil, outdoor air and building materials [24]. We have some doubts about the fraction of the outdoor air component in the total budget of the radon concentration (we think is almost irrelevant), on the other hand we think that the building material component deserves special attention. The conclusion of this disagreement between experimental indoor data and log-normal distribution could be ascribed to background variation during the time and then to an intrinsic difficulty in evaluation the background level, above all when long time exposure (like in our case) has been done [24]. A solution could be, as suggested by [24], organizing the data in homogeneous geological group (that we suspect it is one of main causes of local radon variation) and in buildings homogenous by building material, building technique and use [24]. In this paper we group our data from public buildings and private houses, from the geological side and building material we could not operate.

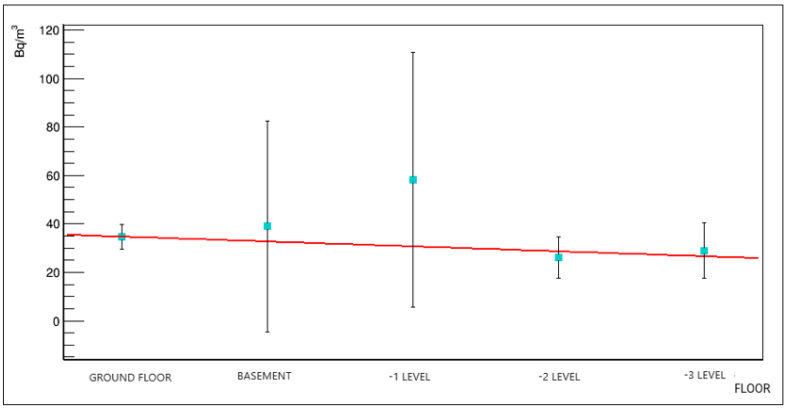


Figure 3: Arithmetic Mean of Year Long Radon Concentration as a Function of the Floor. The fit linear parameters are: f (x) = ax + b. with a= 2.03 ± 2.51, b=36.68 ± 6.98

Citation: Visca L, Calabria R, Sagona M, Amoroso A, Colombetti P, et al. (2024) A Statistical Analysis of the Follow-Up Study of Radon Measures in Private Homes and Public Buildings Carried out in Four European Towns with an Esteem of Lung Cancer Risk. Journal of Earth and Environmental Science Research. SRC/JEESR-304. DOI: doi.org/10.47363/JEESR/2024(6)208

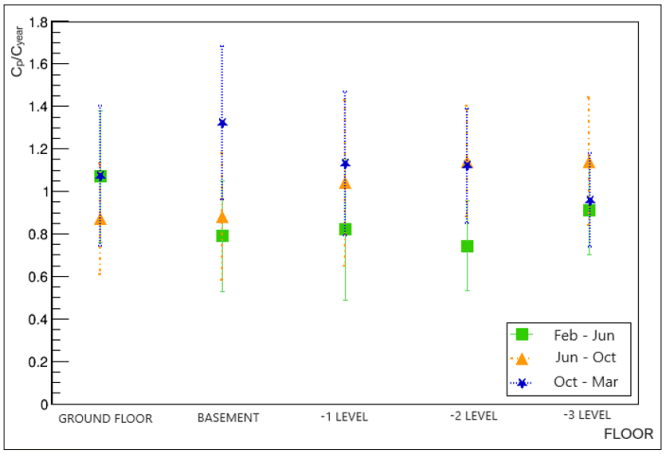
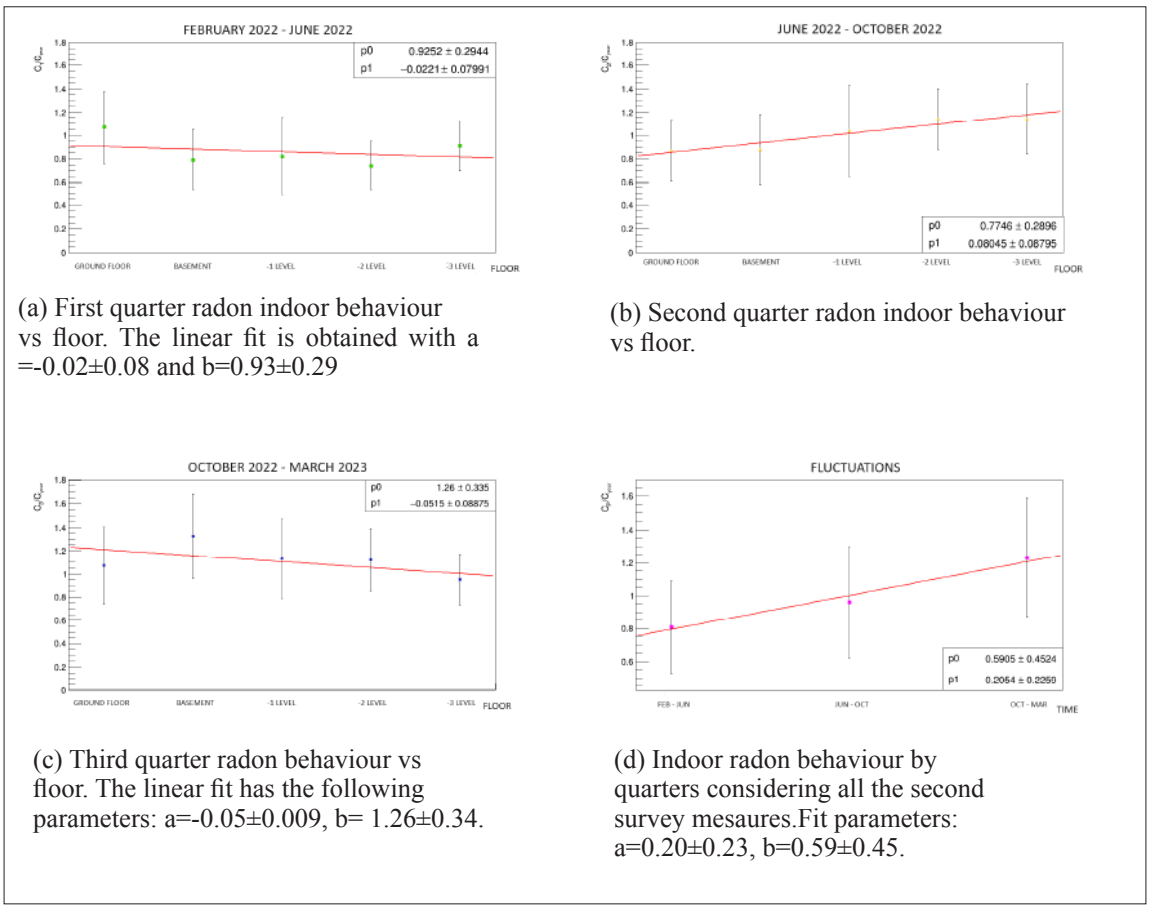


Figure 4: Annual ^{222}Radon Concentration vs the Building Floor, C_P/C_{year} is the Arithmetic Mean Concentration Value Normalized to the Year Average



(a) First quarter radon indoor behaviour vs floor. The linear fit is obtained with a =-0.02±0.08 and b=0.93±0.29

(b) Second quarter radon indoor behaviour vs floor.

(c) Third quarter radon behaviour vs floor. The linear fit has the following parameters: a=-0.05±0.009, b= 1.26±0.34.

(d) Indoor radon behaviour by quarters considering all the second survey mesaures.Fit parameters: a=0.20±0.23, b=0.59±0.45.

Figure 5: Indoor Radon Data Organized by Quarters, in Order to see the Seasonal Variation, and by Floor

In order improving our knowledge of the radon seasonal variation as a function of the floor in the second survey data, in Figure 5 the data are shown organized by quarters starting from February 2022 up to March 2023. The arithmetic means of all data collected at the same room and in the same quarter C_p is normalized with the overall arithmetic mean from all the data taken during the yearlong, as previously explained in Paragraph 4 (C_p/C, with P = quarter considered, C = total average). In Figure 5(a) a constant and linear behavior is shown: a decrease with respect to the annual average is observed in the first quarter apart by the ground floor. In the second quarter (Figure 5(b)) it is apparent an opposite behaviour: the indoor radon concentration is increasing in this time period with the depth of the floor. It is worth to notice that is quarter over the summer holiday when only few people or no one is inside the rooms, at least surely no students (that are the highest percentage) are around. The third quarter (Figure 5(c)) covers mostly the coldest months of the year and we see again a clear decrease of radon concentration vs the underground depth of the rooms. Taking the arithmetic mean of each quarter, normalized at the general average from our second survey data set, we obtain the linear behaviour shown in Figure 5(d): the indoor concentration is increasing in the cold months, as many authors have already pointed out. The Figure 5(d) data are summarized in Table 4 where are reported the data of each quarter.

Health Risk Evaluation

Data covering the 27 European countries seem to show that radon is responsible up to around 8% of lung cancer deaths, in this work we show a new evaluation of lung cancer due to radon starting from our fresh data [8]. The risk evaluation was done using two different methods, one for each data set: this because in the first campaign, where four European towns were involved, mainly private houses were concerned, while in the second survey only buildings of the University of Torino were involved and the final goal was to evaluate the risk for the Torino University workers (since the students spend only a very short fraction of their lifelong time in these rooms). Starting from the first campaign data, estimating the risk due to the exposure of a population to indoor radon concentration is a two steps process. In the first step the absorbed dose to the lung is calculated through very complex models, which take into account both physical parameters and physiological parameters. The absorbed dose however is strongly model-dependent and it is very difficult to get a clear idea about the goodness of the adopted model. As far as the first survey is concerned, according to [12], we evaluate the value of 0.05 mSv/y per 1 Bq/m^3 of radon concentration measured, with an indoor occupancy factor equal to 0.8 [12], [25]. Therefore, we reach the effective dose threshold value of 200 mSv/y for 400 Bq/m^3 of indoor radon. In the second step the risk connected with the effective dose to the lung is computed using the statistical results of the epidemiological studies on individuals professionally exposed to ionizing radiation. According to [27], using the threshold value of 20 mSv/y would mean an annual risk of about 1x10^{-3} equivalent to a 7% lifetime risk for a constant exposure along 70 years in the conditions before discussed but this figure could highly change with the accepted conditions: anyway, no matter which set of data we decide to use, for 400 Bq/m^3 indoor radon, the results are within the 5 – 15% interval for the lifetime risk [26]. In the second campaign we use a method more suitable for workers. In order to evaluate the human health risk related to radon exposition (lung cancer and leukemia are considered) we put the factor f = 6.7 10^{-9} (Sv m^3)/(Bq h) We used the formula [27]:

$$(AeD) = C_{Rn}\left[\frac{Bq}{m^3}\right] \cdot t\left[\frac{h}{year}\right] \cdot 6.7 \cdot 10^{-9}\left[\frac{Sv \cdot m^3}{Bq \cdot h}\right] \quad (2)$$

where AeD is the year effective dose, C_{Rn} is the overall geometric mean annual concentration computed from the second campaign data, and t is the time spent inside the room. For people working in the room we take an average time of about 2000 hour/year, while for private houses we consider an occupancy factor equal to 0.8 [25]. Starting from the year effective dose computed from (2), we evaluated the extra risk for lung cancer and leukemia of Torino University workers due to radon exposition using the coefficent C = 4.2 · 10^{-2} Sv^{-1}. Integrating over a working period of 50 year we get:

$$(IoR) = (AeD)\left[\frac{Sv}{year}\right] \cdot 4.2 \cdot 10^{-2}\ Sv^{-1} \cdot 50[year] \quad (3)$$

with IoR the increment of risk. According with the previous described working hypothesis we compute the year effective dose for people working in all the Torino University buildings equal to 0.42 mSv/year: this is the dose related to time spent in the rooms of Torino University only. Taking into account that 2000 hours/y of exposure means an occupancy of the University rooms by the workers of about 22% of the year total time and considering a mean overall indoor radon concentration computed by all the second campaign data set of C_{Rn} = 31.1 Bq/m^3, we can convert the 0.05 mSv/y per Bq/m^3 of the first method to 1.6 mSv/y. Because with the first method an occupancy of 0.80 has been considered, that is almost four time higher than the 0.22 in the working place, we get an effective dose of 0.44 mSv/y with the first method to compare with the 0.42 mSv/y of the second one: the two figures are surprisingly in good agreement, taking into account all the approximations, variations and so on that we have to face. On the other hand the first method has been applied to the concentration data coming from four European towns (that show great differences), it was related to private houses, the 7% risk is referred to 70 year of life and computed for a dose of 200 mSv/y equivalent to 400 Bq/m^3 indoor radon concentration, a figure that takes into account only situation clearly outside the legal threshold [7]. The extra statistical risk for workers of Torino University to develop lung cancer and leukemia because radon exposure on the working place, according to the data of our second measurements campaign, is equal to 1.75 x 10^{-5} that means in 50 years' time an increase of 0.09%, if the present radon concentration will stay constant, with an evaluate overall uncertainty of 28%.

Conclusion

In the first measurement campaign we report the indoor radon concentration in four European urban area (Reykjavik, Bergen, Lund and Torino), all measures were done using CR-39 detectors, each CR-39 detector was exposed for about 6 months and two measures were carried out for each room with the detector placed at the room ground. Radon activity concentration shows seasonal variation, due to hot or cold weather as well as during wet and dry season periods. The longer the duration of a measurement the lower in general is its variability and during the yearlong first survey six-month exposure time for each CR-39 detector can be the best compromise to estimate the average values. The data show the strong correlation between indoor radon concentration and the surrounding soil geology but in the data set there are not indications of the floor where the measure was carried out. The obtained results shown a very peculiar situation for Reykjavik, an area where the indoor radon concentration reaches most likely one of the minima in all the world in spite to the high volcanic activity and seismicity. Torino in Italy shows a good spread of values: the minimum values are monitored in the area near the river Po banks, where the soil, mainly made of clay and shales, has a low permeability and tends to limit radon mobility. The only Torino area where the radon values reach concentration above 200 Bq/m^3 is in the hillside where there is a sensible presence of tuff stone. Bergen and Lund show high radon values. The soil of Bergen is mainly granites much likely fractured with a good permeability and with a relatively high concentration of uranium and thorium that are the radon ancestor elements: this could explain, even if not in an exhaustive way, the high indoor radon concentration in the Bergen houses. Lund shows a limestone ad sandstone soil with a sensible higher uranium/thorium concentration with respect to Torino and with a good percent of its soil occupied by granite: Lund has higher values of radon with respect to Torino and lower level with respect to Bergen. Apart from the building construction techniques, that are not investigated in this study, the soil component could explain the radon values and the fractured granite soil is clearly the most efficient radon source. About the other radon source under attention, the tuff stone, little could be said because it only occupies a small portion of Torino area concentrated in the hillside. It is worth to stress the strong correlation between radon ancestors' presence, Uranium and Thorium, in the soil and the level of indoor radon: Reykjavik situation is a strong proof. In the second survey, carried out only in the town of Torino and only inside buildings of University of Torino with the goal to evaluate the increase risk for the Torino University workers to develop lung cancer and leukemia in the working place, the CR-39 detectors were exposed for four months in order to study the seasonal effects, and information about the floor of each measurement point was registered. We noticed that

rooms in the basement and at the ground floor have an increase of radon values in the fall and winter season, likely due to the cold weather and the related conditioning effect. At -2 level the minimum values have been measured between February and June and also in this case we think the conditioning set played an important role, but at -3 floor we observe an unexpected seasonal variation with an increase of radon concentration in summer and a decrease during the cold months. As far as the basement is concerned, the highest values are reached during winter and spring months. We have only one room for sure monitored in the ground floor. Analyzing the second campaign data set we could claim that not only the floor is affecting the indoor radon concentration values but also the conditioning set and the way it is used. The arithmetic mean MA of all the second survey measurements is MA = 42.1 Bq/m^3, the geometric mean MG is equal to 31.1 Bq/m^3: these results are less than the overall indoor radon concentration of all the Piedmont territory and considering all types of buildings, that is equal to 71.4 Bq/m^3 a good result for Torino University indeed [23]. Taking into account also the first campaign results in Torino area, we get that 92% of Torino buildings have an indoor radon concentration less than 100 Bq/m^3, in agreement with the official data in [23]. With an indoor radon concentration of around 400 Bq/m^3 we computed a risk increase to contract lung cancer in 70 year lifetime, with an evaluated indoor time occupancy of about 80% of the total, equal to 7%, no matter which method or set of data we decide to use. The results are within the 5 ÷ 15% interval for the lifetime risk, over the attention threshold established by European rules: the high values of indoor radon concentration must be tackled down to protect the population health. The effective dose for people working in Torino University building has been evaluated at 0.42 mSv/y with an extra risk to contract lung cancer and leukemia at the working place in 50 years' time equal to 0.09%. If we apply the same method and conditions to the data taken in the first campaign, where private houses were concerned, we get an effective dose of 0.44 mSv/y: surprisingly the two values are in good agreement taking into account all the possible variations between the two cases.

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