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Indoor Radon Concentration and Risk Estimation: the EURA PROJECT

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Abstract

The indoor radon concentration level has been monitored in selected locations in four European countries (Iceland, Italy, Norway, and Sweden) during a yearlong measurement campaign using time-integrated passive radon dosimeters containing CR-39 track detectors. The measurements were carried out in private houses and public buildings like schools, for an exposure time of up to 6 months for each detector. Experimental data shows a great variation between different geographic areas, and often the average levels are over the acceptable radon limit of 300 Bq/m³ recommended by the International Commission on Radiological Protection (ICRP). To investigate a relationship between indoor radon exposure and lung cancer, estimating the cumulative levels of exposure to indoor radon for an individual or population is necessary. We analyse the data sets and investigate the factors influencing indoor radon concentrations in order to determine the best use of the experimental information. The results show that the variables associated with indoor radon levels are strictly linked to the soil geology. Analyzing the data sets enables improved assessment of radon exposure in a given area. The average absorption effective dose equivalent for a person is computed, and the risk of lung cancer per year is evaluated.

Keywords: Indoor Radon-Emission Measures; Radon Background Concentration; Environment-Protection; Uranium; Thoron.

1. Introduction

Radon is a gas produced by the radioactive decay of radium. Radioactive decay is a natural, spontaneous process in which the atomic nucleus of one element decays or breaks down to form another element by losing or emitting out nuclear particles (protons, neutrons, electrons, etc.). When radium decays to form radon gas, its nucleus emits two protons and two neutrons tied up together to form an alpha particle (or ⁴He nucleus). Radon itself is radioactive because it also decays, losing an alpha particle and forming polonium. Uranium and thorium are the first elements in a long series of decays that produce radon. Uranium and thorium are referred to as the parent elements, and radium and radon are called daughters. Radium and radon also form other daughter elements as they decay.

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Uranium has a half-life of 4.4 billion years and forms the longest half-life of radon isotopes (^{222}Rn) that is only 3.8 days. ^{232}Th is an element that, like uranium isotopes, is distributed throughout the environment. It has a very long half-life (1.41×10^{10} yr) and decays by emission of an alpha particle creating a series of radioactive daughters, many of which emit alpha radiations too. One of these daughters is the second most important radon isotope, ^{220}Rn that has a half-life of 55 seconds. If, for instance, a jar was filled with ^{222}Rn , in 3.8 days only half of the radon would be left. But the newly made daughter products of radon would also be in the jar, including polonium, bismuth, and lead. Also polonium has radiative isotopes: from ^{222}Rn decay it is produced ^{218}Po that decays with a half-life of 3.04 min emitting an alpha particle of 6.11 MeV. It is mainly ^{218}Po and ^{216}Po (from ^{220}Rn decay, ^{216}Po emits alpha of 6.7 MeV), both produced by radon in the air, that by breathing enter the people's lungs, stick to the lungs and/or bronchial tissue where the emitted energetic alpha particles could hurt the cells and causes cancer illness. The radon's daughters are the main responsible for the cancer risk: many studies have been done and the World Health Organization – International Agency for Research on Cancer classified radon and its decay products as carcinogenic elements of group 1, that means the group of the most dangerous ones [1]. Breathing radon is nowadays acknowledged the second source of lung cancer after smoking. Radon levels in outdoor air, indoor air, air soil and ground water can be very different. Outdoor air ranges from less than 10^{-3} Bq/L to about 4 Bq/L, but it probably averages about 10^{-2} Bq/L (with the usual conversion of $1 \text{ Bq/m}^3 \sim 0.001 \text{ Bq/L}$, i.e. $1 \text{ m}^3 = 1000 \text{ L}$: it is a common use to refer to liter unit instead the m^3 when dealing with radon concentration in the air to avoid too small figures).

Radon in indoor air ranges from less than $4 \cdot 10^{-3}$ Bq/L to about 1000 Bq/L, but it probably averages between 10^{-3} and 10^{-2} Bq/L. Radon in soil air (the air that occupies the pores in soil) ranges within (10 - 30) 10^{-2} Bq/L up to more than 1000 Bq/L; most soils in Piemonte (Italy) contain between 5 and 20 Bq of radon per liter of soil air. The amount of radon dissolved in ground water ranges from about 1 to nearly 1000 Bq/L. In water sources originated from granitic rocks around Torino and in Torino suburbs (Italy) we measured around 160 – 190 Bq/L, in well waters (10 wells were analyzed) the radon concentration ranges from 72 up to 163 Bq/L (the highest values are associated to wells dig in tuft); in surface waters 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 lie 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. The first scientific investigations about radon exposure were targeted on workers that have to spend a lot of time underground like miners but nowadays studies of the radon effect on the public health have been carried on considering the radon level inside private houses. Because levels of radon vary so much from place to place, and because houses differ in their vulnerability to radon, it is important that all houses be measured for radon. The evaluation of the health risk is affected by big uncertainties [2, 3], but it has been proved the interaction between radon and smoking causes a sensible increase of the lung cancer risk.

According to EPA (Environmental Protection Agency - USA) the percent of lung cancer related to radon exposure is about 9% of all the cases. In Italy around 1% of homes and buildings can reach a radon concentration over 400 Bq/m³ and 4% have concentration over 200 Bq/m³, therefore, according to a preliminary analysis, the average risk to develop a lung cancer related to radon exposure during the all life is evaluated between 0.5 up to 5-15% of the total amount of lung cancer diagnosis done in Italy per year. Recent epidemiological studies [4] carried out new analysis of old data of radon concentration coming from different measurement campaigns done with different radon monitors, reached a much better statistical confidence levels: the conclusions seem to strongly support the estimated value within 5 – 15% risk to develop a lung cancer during the all life related to radon exposure and the threshold of safety for radon concentration seems that must be correct towards lower values with respect to present law prescriptions in the European countries. Even with the threshold of 200 Bq/m³ accepted by European rules the risk is statistically sensible. But the most striking evidence is the strong correlation between radon and smoking: smokers exposed to even low radon level get a probability up to 3 times higher to develop a lung cancer during their life, in a sort of multiplicative effect raised when both causes are present [5]. The indoor radon concentration level was monitored during a yearlong measurement campaign in 250 selected points of each of the following European towns: Torino in Italy, Bergen in Norway, Lund in Sweden and Reykjavik in Iceland under the framework of European ERASMUS+ plan, in this work we present, analyze and discuss the obtained results.

2. The Geology of Radon

Studies of the geology of radon include measuring the amount of uranium and thorium distributed in rocks and soils: uranium and thorium are the radon sources. It is also an important part of the geology of radon investigating how radon moves and enters buildings from soil and through water system. All rocks contain some uranium and thorium although most contain just a very small amount between 1 and 3 parts per million (ppm): this is what we measured in rock samples collected in the Reykjavik area. In general, the uranium and thorium content of a soil will be around the same as their content of the rock from which the soil was derived but some types of rocks show higher than average concentration of these two elements. These include light-colored volcanic rocks, dark shales, granites (that we found in the areas around Bergen), sedimentary rocks that contain phosphate, tuft stone and metamorphic rocks derived from these rocks (the last ones describe the soil situation around Lund).

These rocks and their soils may contain up to 200 ppm uranium. The higher the uranium level is in an area, the greater the chances are the houses in that area have high levels of indoor radon, but some houses in areas with lots of uranium in the soil show low levels of indoor radon, and other houses on uranium-poor soils have high levels of indoor radon. From these observations we could deduce that the amount of radon in a house is affected by other factors in addition to the presence of uranium and thorium in the underlying soil. Just as these two elements are present in all rocks and soils, so are radon and radium because they are daughter products formed by the radioactive decay of them. When a nucleus of radium decays by ejecting an alpha particle, the newly formed radon atom recoils in the opposite direction. This recoil is the source of kinetic energy that is the most important factor affecting the release of radon from mineral grains. The location of the radium in the mineral grain (how close it is to the surface of the grain) and the direction of the recoil (whether it is toward the surface or the interior of the grain) determine the probability of the newly formed radon atom to enter the pore space between mineral grains: the recoil of the radon atom is quite strong. If water is present in the pore space, the moving radon atom slows very quickly and is more likely to be trapped in the pore space.

For most soils, only 10 to 50 percent of the radon produced could escape from the mineral grains and enters the pores. Because radon is a noble gas, it is monoatomic and occupies a volume that is much smaller than the smallest molecule, as hydrogen, and this helps a lot in moving throughout the rock pores. Radon has a very high mobility, much greater than uranium and thorium and does not react with other elements to be fixed in the solid matter in rocks and soils. Therefore radon can leave the rocks and soils, by escaping into fractures and openings in rocks and into the pore spaces between grains of soil and then it can travel a great distance before it decays. The ease and efficiency with which radon moves in the pore space or fracture affects how much radon could enter the building. The method and speed of radon movement through soils is controlled by the amount of water present in the pore space (the soil moisture content), the percentage of pore space in the soil (the porosity), and the "interconnectedness" of the pore spaces that determines the soil's ability to transmit water and air (the permeability).

Radon moves more readily through permeable soils, such as coarse sand and gravel, than through impermeable soils, such as clays. Fractures in any soil or rock like geological faults allow radon to move more quickly. Radon in water moves slower than radon in air. The distance that radon moves before most of it decays is less than 4 cm in water-saturated rocks or soils, but it is much more (meters) throughout dry rocks or soils and reaches hundreds of meters in faults. For these reasons, buildings in areas with drier, highly permeable soils and bedrock, such as hill slopes and bottoms of valleys, coarse glacial deposits, fractured or cavernous bedrock or built over or near a geological fault may have high levels of indoor radon. Even if the radon content of the air in the soil or fracture is under the outdoor risk threshold (~ 10 Bq/L), the permeability of these areas permits radon-bearing air to move greater distances before it decays and then to reach high indoor concentration. Radon moving through soil pore spaces and rock fractures near the surface of the earth usually has a great probability to escape into the atmosphere. Where a house is present, however, soil air often flows toward its foundation for three reasons: differences in air pressure between the soil and the house, the presence of openings in the house's foundation, and increases in permeability around the basement (if one is present). In constructing a house with a basement, a hole is dug, footings are set, and coarse gravel is usually laid down as a base for the basement slab. Then, once the basement walls have been built, the gap between the basement walls and the ground outside is filled with material that often is more permeable than the original ground. This filled gap is called a disturbed zone. Radon moves into the disturbed zone and the gravel bed underneath from the surrounding soil.

The backfill material in the disturbed zone is commonly rocks and soil from the foundation site, which also generate and release radon. The amount of radon in the disturbed zone and gravel bed depends on the amount of uranium and thorium present in the rock at the site, the type and permeability of soil surrounding the disturbed zone and underneath the gravel bed, and the soil's moisture content. The air pressure in the ground around most houses is often greater than the air pressure inside the house. Thus, air tends to move from the disturbed zone and gravel bed into the house through openings in the house's foundation. All house foundations have openings such as cracks, utility entries, seams between foundation walls and slabs, sumps, permeable foundation materials, and uncovered soil in crawl spaces and basements. Most houses draw less than one percent of their indoor air from the soil; the remainder comes from outdoor air, which is generally quite low in radon. Houses with low indoor air pressures, poorly sealed foundations, and several entry points for soil air, however, may draw as much as 20 percent of their indoor air from the soil. Even if the soil air has only moderate levels of radon, levels inside the house may be very high. During the present radon measure campaigns, estimation of uranium and thorium concentration in natural rock samples collected nearby the measurements points was done by a gamma ray spectrometry based on a HpGe detector, 60% efficiency with respect to $2'' \times 2''$ NaI reference crystal, shielded by 10 cm lead against cosmic rays and natural background. General criteria for the selection of a peak in the estimation of concentration of an element is that it should have good gamma yield and also it should have as low as possible contributions from any other gamma emissions.

Based on this criteria, U concentration in different rock types is measured using 1.001 MeV gamma energy 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 gamma 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. The concentration is mainly estimate by detecting 911.6 keV and 969.1 keV gamma lines from ^{228}Ac decay (26.6 and 16.6% respectively). In Tab 1 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.

The results for the volcanic rocks of Reykjavik is amazing, even if well known. If we compare these results with the uranium and thorium concentration that shows the rocks from the Mediterranean Volcanos like Stromboli, Vulcano (in the Eolian Islands), Etna in Sicily and Mount Vesuvio near Naples, then in all these situations the uranium and thorion concentration is between one or two order of magnitude higher than what measured for Reykjavik rocks and so the radon concentration too. We could not find a satisfactory explanation for this striking difference in the scientific literature.

3. Indoor Radon Measures

Indoor radon has been measured in many houses, schools, and commercial buildings across four European towns: Bergen (NOR), Lund (SW), Reykjavik (IC) and Torino (I). Around 2000 measure points have been considered during a year-long campaign. For the most part, these measurements have been made in private homeowners using passive detection devices CR-39 provided by the Italian national agency ENEA, all measures were done at the building ground floor. This method was originally developed for detecting heavy charged particles [6] and subsequently used for radon measurements. It is based upon the fact that an alpha-particle leaves a clear track of damage in dielectric media. Later the tracks can be etched with a suitable etchant (e.g. NaOH), by enlarging them sufficiently to become visible, if needed, under an optical microscope. The number of tracks is obviously proportional to the concentration of radon and/or radon progeny and the time of exposure. The tracks were counted by an automatic system based upon a spark counter [7].

The film with the etched-through holes is placed in a capacitor. The top electrode is a metallized film (typically aluminized mylar) and the other electrode is made of metal. When a dc voltage is applied to the capacitor, the etched tracks begin to breakdown sequentially and each spark vaporizes a hole in the aluminum, effectively removing the conductor layer on one side and is easily counted. The advantage of etched track detectors is apparent, since the dielectric media respond to only alpha particles and even a threshold alpha energy could be set up: the main radon isotopes emit quite energetic alpha, therefore these detectors are quite selective for radon and its progeny. The used CR-39 are made of polyallyl di-glycol carbonate (PADC). The measure of radon concentration for radiation protection purposes involves some problems because radon has a short-lived progeny with totally different physical-chemical properties. In fact radon is a noble gas while its decay products are solid (metallic ions) which could be attached to any small particle normally present in air as well as any other surface. For this reason the equilibrium, i.e. when the activity of each decay product equals that of the parent radon, is rarely attained. The processes influencing the concentration of radon progeny in the air are related to atmospheric aerosols, plate-out (surfaces), recoil from aerosols and surfaces, and decay. Ionization, electrostatic forces, gravity, airflow, and steady- state molecular diffusion influence the deposition on surfaces. If an average diffusion coefficient in air is taken equal to $0.06 \text{ cm}^2/\text{s}$ for ^{222}Rn , ^{218}Po and ^{214}Po , a concentration of radon decay products near the surfaces can be calculated. Their concentrations significantly decrease at distances less than about 10 cm from the walls and at a distance above 50 cm the influence of plate-out can be neglected [8].

Each CR-39 detector was exposed for up to 6 months and at least two measures were carried out for each houses, the radon concentration was computed and certified by ENEA of Italy, which also calibrated the CR-39. For a correct approach to radon concentration investigation, a great spread of values must be taken into account. These variations occur on many time scales, from hourly to annually, and depends on seasonal factors and weather conditions, building characteristics, operation of dwelling heating and refrigerating systems, living habits, source relative strength, etc. The longer the duration of a measurement the lower in general is its variability: individual short-time measurements (minutes or few hours) can show variations higher than a factor of 10. Measurements lasting a few days generally show variations less than a factor of 10 (see for instance [9]), while the seasonal variations found in measurements of six-months duration are usually well within a factor of 5 [10], while annual variations are well within a factor of 2, e.g. measurements carried out for a 5 year period in 40 residences in Grand Junction (Colorado, USA) show a mean coefficient of variation approximately around 22% [11]. Because of the magnitude of these variations, one-year measurements are considered the best compromise to estimate the average values. For all the four urban areas the measures were done within the following approaches: invitation of volunteers from the communities of residents of buildings, offering participation in this activity to students and staff members of the local high schools and/or educational institutions organizing the research project EURA.

To disseminate information about the study and attract volunteers to participate, a seminars of experts under the supervision of research team members were organized in each towns and a website was created to provide more detailed information about this project. 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 A peculiar situation could be observed for Reykjavik, where in spite of the volcanic rocks around the town 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 1), while for Reykjavik the very low radon levels come with such a low concentration of uranium and thorium in soil and rocks (even if they are volcanic rocks) that we were able to give only an upper limit. In Reykjavik 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 [12], radon activity is generally rather low, in most cases less than 5 Bq/L. Only 8% of water samples had a measured radon activity higher than 5 Bq/L, with a maximum activity of 10.8 Bq/L [12].

Usually the hot water samples generally have a higher radon activity than cold water samples, but samples from boiling boreholes have a lower radon activity as radon partially moves into the vapor phase. The geographical distribution of the samples indicates 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/kg translating the unit of measures reported in Table 1 with an uncertainties 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/m³ with a standard deviation of 8.5 and a geometric mean of about 8.1 Bq/m³ with a median absolute deviation (MAD) of 5.9 Bq/m³ (see Table 2). Note the close values obtained for arithmetic mean and geometric mean, a case that we see in the Torino data too. Measures done in the other two towns show a very different situation indeed. In Torino we measured an arithmetic mean of 26.6 Bq/m³ with a standard deviation of 11.2 and a geometric mean of 24.59 Bq/m³ with a MAD of 7.41 Bq/m³. The large spread of values in the radon data set suggests to evaluate the statistical dispersion by MAD that in the present measure conditions is a robust 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/m³ 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/m³ respectively, with 37.06 and 60.79 Bq/m³ for the associated MAD. The arithmetic mean is 87.7 Bq/m³ with a standard deviation of 207.7 Bq/m³ for Bergen and 102.6 Bq/m³ with a standard deviation of 154.8 Bq/m³ for Lund (see Table 2). 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 variations up to 100%. (Table 1).

Table 1. Uranium and Thorium concentration in ppm measured by gamma detection in a 60% efficiency shielded HpGe spectrometer. Each measure have an estimate uncertainties of +/- 5 ppm

Area	U	Th
Torino area		
sedimentary rock	12	21
limestone	19	23
tuft stone 1	24	34
tuft stone 2	38	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

Table 2. Arithmetic mean, standard deviation, geometric mean and the statistical dispersion by median absolute deviation (MAD) of measured radon concentration

Town	Arithmetic mean Bq/m ³	Standard deviation Bq/m ³	Geometric mean Bq/m ³	MAD (Median Absolute Deviation) Bq/m ³
Bergen	87.7	207.7	31,16	37,06
Lund	102.6	154.8	53,5	60,79
Reykjavik	11.0	8.5	8,1	5,9
Torino	26.6	11.2	24,59	7,41

It has been observed by many measure campaigns that the distributions of indoor radon concentrations follow more or less closely to 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 histograms 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 and radon in indoor air. 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 happens, according to our observations, for the radon concentrations above 300 Bq/m³. We study this effect in order to better estimate the number of homes with very high radon level. The fits using our developed log-normal distribution of all the measured annual average radon concentrations in the four town are shown in Figures 1 to 4. The probability density function (pdf) of the log-normal distribution that we used is $\text{log-norm.pdf}(x, \sigma, f.loc, S)$, where x are the measured radon concentration data ($x > 0$), σ is the shape parameter and is also the standard deviation of the log of the distribution ($\sigma > 0$), $f.loc$ is the location parameter evaluated by each fit (see Figures 1 to 4) and S is the median of the distribution, known also as scale parameter ($S > 0$). $\mu = \log(S)$, where the μ parameter is the mean of the log of the each distribution.

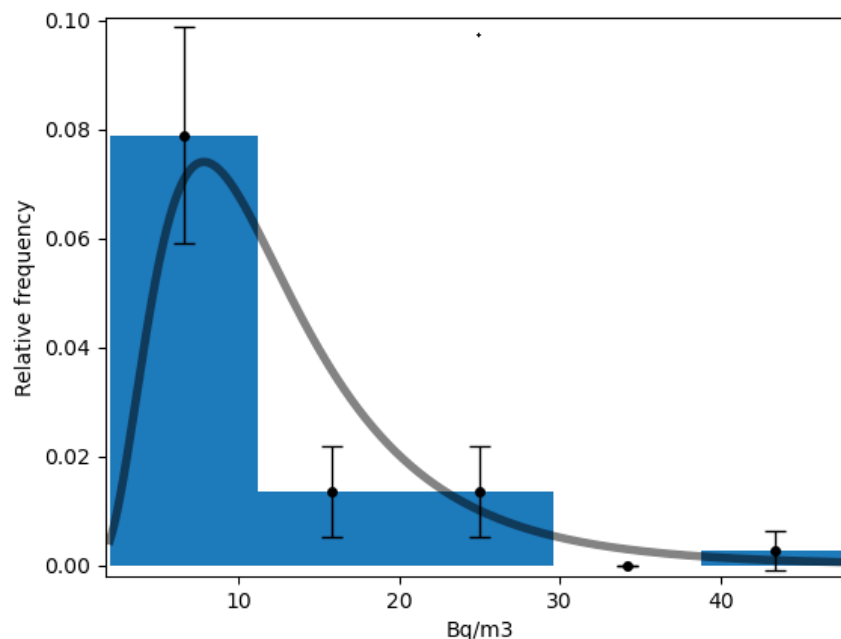


Figure 1. Indoor radon concentration in Reykjavik. The full black line shows the log-normal distribution fit with a $f.loc = 2.40$. The histogram error bars are purely statistical

In Figure 1 the radon data set collected in Reykjavik are shown with the results of the log-normal fit. In Figure 2 data from Torino and surrounding area are shown. It is apparent as the Torino situation is in the middle between the Reykjavik and Bergen – Lundt (see Figures 3 and 4). Torino soil is mainly compose by sedimentary rocks and limestone, there is almost not granite around. 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 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 near tuft stone soil. The radon concentration measures for the Bergen area is shown in Figure 3, for Lund area see Figure 4: both graphic shows a similar situation, with the peak of highest radon concentration found in the Bergen area,

in full agreement with the uranium/thorium rock 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 3 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/m³. In these points, local distributions might have very different means and standard deviation from the other measurement points. The log-normal fit pass 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/m³: the local distribution from high radon areas have different parameters to most of the data presented elsewhere. Therefore the numbers of homes above a few hundred Bq/m³ should be different to and greater than was predicted from national survey methods, based upon log-normal distribution.

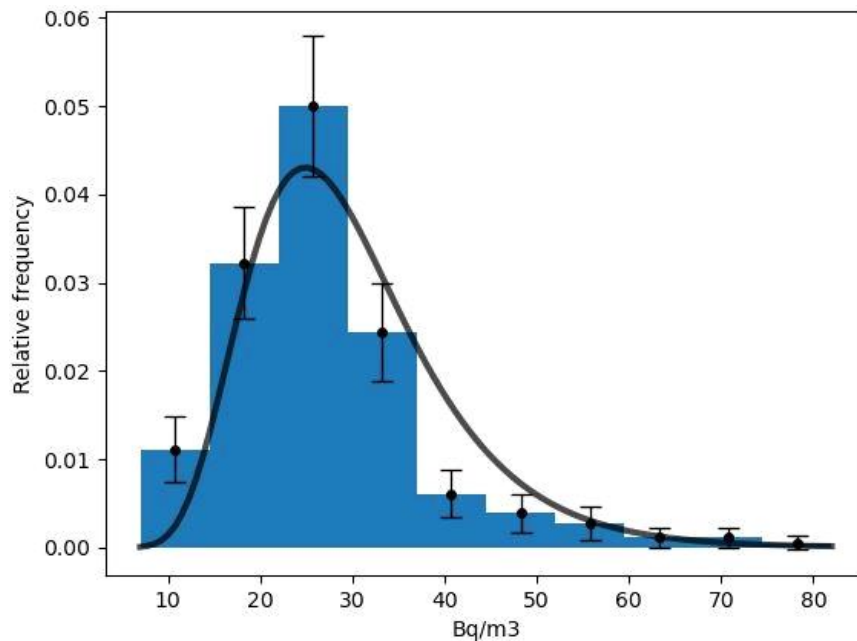


Figure 2. Indoor radon distribution in Torino and surroundings area. The full black line shows the log-normal distribution fit of the experimental data with a $f_{loc} = 3.34$. The histogram error bars are purely statistical

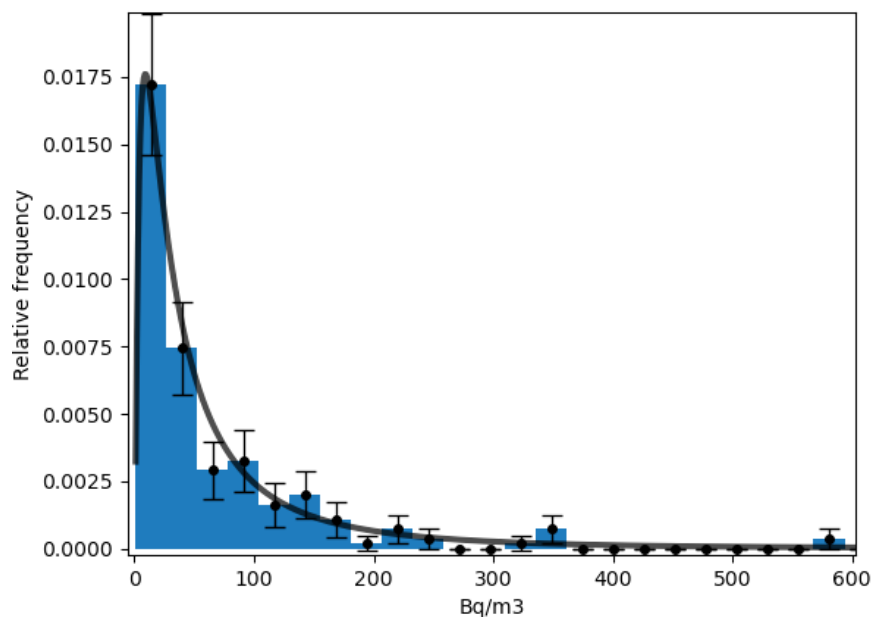


Figure 3. Indoor radon distribution in Bergen and surroundings area. The full black line shows the log-normal distribution fit of the experimental data with a $f_{loc} = 3.60$. The histogram error bars are purely statistical. Note the little bump between 300 and 400

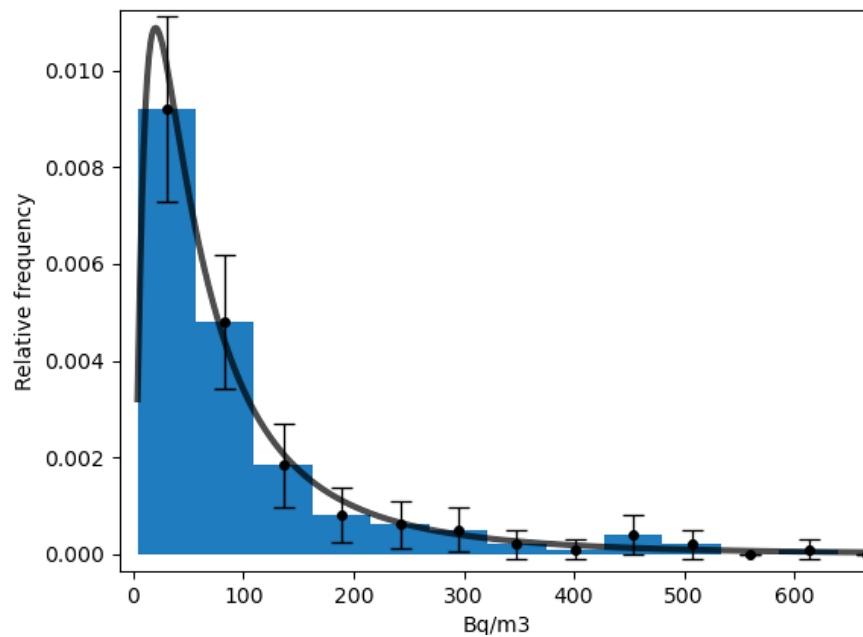


Figure 4. Indoor radon distribution in Lund and surroundings area. The full black line shows the log-normal distribution fit of the experimental data with a $f.loc = 4.09$. The histogram error bars are purely statistical. Note the small bump between 400 and 500

Table 3. χ^2 tests for log-normal distribution fit. The $\alpha = 1-p$ 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 tests where ends the agreement

Town	χ^2 Test	Degrees of freedom ($\nu=3$)	Estimated end point ($\alpha = 0.05$)
Bergen	4.48	17	8.67
Lund	2.62	9	3.33
Reykjavik	0.14	4	0.71
Torino	2.35	9	3.33

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/m³, with the warning of a possible underestimation for concentration higher than 300 Bq/m³. 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 4. In the town of Reykjavik and Torino is most unlikely to find buildings with more than 300 Bq/m³ radon concentrations, but for Lund and Bergen we observe a situation with a sensible risk.

Table 4. Percentage of houses/buildings in each town with an expected radon concentrations higher than the shown level. The percentage is computed from the log-normal distribution fitting the radon data set

Town	>100 Bq/m ³	>200 Bq/m ³	>300 Bq/m ³
Bergen	20%	8%	4%
Lund	31%	12.6%	6%
Reykjavik	0.1%	0.0001%	-
Torino	0.15%	0.0001%	-

4. Radon Risk Estimation

One way to estimate the risk due to the exposure of a population to radon concentration in indoor air is based on two step procedure. In the first step the absorbed dose to the lung is calculated through very complex models, which take into account both physical parameters (radon and radon progeny concentration, fraction of progeny attached to aerosols, unattached fraction, aerosol size distribution, etc.) and physiological parameters (characteristics of the respiratory tract, respiratory rate, thickness of bronchial epithelium, location of target cells, etc.). These models (e.g. [13]) are continuously evolving. An improved model of the respiratory tract to be used in a wide range of circumstances has been adopted by ICRP [14].

It has to be emphasized that although the dose is mostly due to inhaled radon progeny (and its characteristics) rather than to radon, some studies [15-17] show that, in domestic ambient air, changes in ventilation rate produce opposite variations in the equilibrium factor F (i.e. the ratio between radon progeny and radon gas concentration) and in the unattached fraction, so that the absorbed dose to the lung remains relatively constant at a given radon concentration. We define as absorbed dose by $D = dE/dm$ where dE is the mean energy released inside a mass dm of matter by incident ionizing radiation. For radiation protection, a tissue - or organ - average absorbed dose DT is defined by $DT = E_T/m_T$, where E_T is the total energy given to a tissue - or organ - of mass m_T . The absorbed dose is calculated by models and is strongly model-dependent, weighting factors (related to the specific effects of alpha particles and to the lung sensitivity to radiation) have to be applied in order to obtain the "effective dose", which is the quantity considered to be proportional to the health effects [18]. Different values of the effective-dose/exposure factor have been proposed (see [19]).

Here, it has been used, according with [19], the value of 0.05 mSv/y per Bq/m³ of radon concentration measured, with an indoor occupancy factor equal to 0.8 (see [20]). The occupancy factor could vary in the four towns under investigation, because of the different lifestyle but the weather conditions of Bergen, Lund and even Torino, an industrial town of North West of Italy are not so different and we adopt the same coefficient for all. Therefore, for 400 Bq/m³ the effective dose is 20 mSv/y, 15 mSv/y for 300 Bq/m³ and 5 mSv/y for 100 Bq/m³. In the second step the risk connected with the effective dose to the lung is evaluated. This evaluation is made using the results of the epidemiological studies on persons exposed to ionizing radiation, mainly studies on survivors of Hiroshima and Nagasaki, although in this case the exposure conditions were very different from those due to residential radon. In particular these survivors were exposed to gamma and neutron radiation, while the main radiation emitted by radon progeny is alpha radiation. In [18] a review is made, adopting a risk/dose factor for general public of 5×10^{-5} probability of fatal cancer per mSv of effective dose.

Using this figure, 20 mSv/y (associated to 400 Bq/m³ exposure) would imply a 1×10^{-3} annual risk, corresponding to a 7% lifetime risk for an exposure of 70 years in the conditions before discussed. The annual risk drops down to 0.25×10^{-3} , corresponding to 1.75% lifetime risk for 70 year exposure at 100 Bq/m³. The main problems of the dosimetry approach are the appropriate choice of the weighting factors to obtain the effective dose [21] and the use risk/dose factors derived from epidemiological data for persons (such as the Japanese survivors) exposed to radiation other than that from radon progeny. For these and other reasons at present the most popular approach is to use only the miner epidemiology data (see [14]) to estimate the risk. The evaluated lifetime risk for a constant exposure during 70 years to 1 Bq/m³ radon concentration is of 0.87×10^{-4} (against 5×10^{-5} as before), from the data of miners, with an indoor occupancy factor equal to 0.8, as before. Then if we consider an exposure equal to 20 mSv/y we have an annual risk of 1.6×10^{-3} , corresponding to around 11% lifetime risk for 70 years exposure. Anyway no matter which set of data we decide to use, the results are within the 5 – 15% interval for the lifetime risk. Which is a reasonable risk that each person could bear during his lifetime? This is the question that the European Government has to answer considering the upper limits, defined by law, for the level of radon in the buildings.

5. Conclusions

Inhalation of radon and its progeny is the most important component of public exposure to natural radiation. Many epidemiological studies supported by experimental evidence of mutagenesis studies in cell culture and laboratory animals have established the fact that radon and radon progeny are established human lung carcinogens. Extrapolation from extensive studies on miners suggests that radon is the second leading cause of lung cancer death after tobacco smoking. In vitro cytogenetic studies demonstrated that radon induces different types of genetic and cytogenetic damage that is likely to play a role in radon lung carcinogenesis. The majority of studies on the relationship between radon and other types of cancer showed weak or no association. Besides health concerns, radon is also important in other fields, e.g. hydrological research, detection of geological fault lines, prediction of earthquakes, uranium deposits, and oil exploration. The importance of the subject suggested too many scientists develop methods for the measurement and mitigation of radon. In most studies, the techniques used involve the use of solid-state nuclear track detectors.

In this study, we report the indoor radon concentration measures taken in four different urban areas (Reykjavik, Bergen, Lund, and Torino) located in four European countries. All measures were taken at ground-floor residences using CR-39 detectors, each CR-39 detector was exposed for a maximum of up to 6 months, and at least two measures were carried out for each house. Research measurements pointed out that radon activity concentration changes during the winter and summer months, as well as during wet and dry season periods. The longer the duration of a measurement, the lower in general its variability, and taking into account the magnitude of these variations, one-year measurements with up to a month of exposure time for each CR-39 detector can be the best compromise to estimate the average values. The obtained results show an idyllic situation for Reykjavik, an area where the indoor radon concentration reaches most likely one of the minima in all the world, in spite of the high volcanic activity and seismicity. Looking at the radon concentration measured in the Mediterranean volcanic areas such as the region around Vesuvio, Etna, and Stromboli in Italy, where very high levels of indoor radon concentration, sometimes going up to 1000 Bq/m³, are measured, all of

Iceland's territory behaves just the opposite way. Even the uranium/thorium concentration found in the Iceland basaltic rocks is much lower compared to the same type of rock collected near the Italian volcanoes.

The Torino area in Italy shows values for indoor radon concentration that are in agreement with what could be expected in an area where the soil is mainly made of limestone and sedimentary rocks. According to these measurements, the only areas where radon concentrations exceed 200 Bq/m^3 are those with a detectable presence of tuft stone. Both Bergen and Lund show high radon values. The soil of Bergen is mainly granite with a relatively high concentration of uranium and thorium, which are the radon parent elements, and this could explain, even if not in an exhaustive way, the high indoor radon concentration in the Bergen buildings and houses. Lund shows a limestone soil with a sensible higher uranium/thorium concentration with respect to Torino, and a good percent of its soil is occupied by granite. Lund has higher values of radon with respect to Torino and has a comparable situation with respect to Bergen. Apart from the building construction techniques, which were not investigated in this study, the soil component could only partially explain the radon values, and the granite soil is clearly one efficient radon source.

Other radon sources under attention: we could mention the tuft stone in the Torino area, but little could be said because the tuft stone only occupies a small portion of the Torino soil. One conclusion of this work is that a mixture of log-normal distributions more accurately captures the real underlying distributions of measured radon concentrations than the standard single log-normal model. But it must be taken into account that most likely the log-normal model deviates from underestimating the percentage of houses/buildings with high radon levels, i.e., over 300 Bq/m^3 , with a non-negligible effect on the evaluation of radon risk. Estimating the risk due to the exposure of a population to indoor radon concentration is a two-step process. In the first step, the absorbed dose to the lung is calculated through very complex models that take into account both physical and physiological parameters. The absorbed dose, however, is strongly model-dependent and it is very difficult to establish the effectiveness of the adopted model. Here, according to [19], we adopted 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 (see [20]). Therefore, we reach the effective dose threshold value of 20 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 considered using the results of epidemiological studies on persons professionally exposed to ionising radiation. According to [18], using the threshold value of 20 mSv/y would mean an annual risk of about 1×10^{-3} , equivalent to a 7% lifetime risk for a constant exposure of 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, the results are within the 5–15% interval for the lifetime risk.

6. Declarations

6.1. Author Contributions

Conceptualization, D.B., L.L.B. and G.E.; methodology, D.B. and G.G.; experimental data collection & analysis: D.B., L.L.B., G.E., U.H.E., C.P., and B.E.D.S.; writing—original draft preparation, D.B., G.G.; writing—review and editing, G.G. All authors have read and agreed to the published version of the manuscript.

6.2. Data Availability Statement

Data available are collected in publicly accessible repository. The data presented in this study are openly available by sending an e-mail to: lucia.bjorsvik@hfk.no or d.barca.db@gmail.com.

6.3. Funding

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6.4. Ethical Approval

Not applicable.

6.5. Declaration of Competing Interest

The authors declare that there is no conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/or falsification, double publication and/or submission, and redundancies have been completely observed by the authors.

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