

LONG-TERM AVERAGE RADON LEVELS MEASURED IN A FEW DAYS: IMPLICATIONS TO RISK COMMUNICATION AND MITIGATION STRATEGY

Dobromir S. Pressyanov
Sofia University “St. Kliment Ohridski”, Bulgaria
E-mail: pressyan@phys.uni-sofia.bg

Abstract

In the European Council directive (2013/59/EURATOM) the reference levels for radon are based on the annual average concentration (max. 300 Bq/m³). Waiting a year or longer for detector exposure and analysis completion demotivates many stakeholders to give serious consideration to the radon problem and its consequent mitigation. We propose an approach for a speedy problem identification, yet that is still relying on a direct measurement of the annual average radon concentration. The CD/DVD method is used, where one or more compact disks (CDs) or digital versatile disks (DVDs) that were present for a duration of more than one year inside the house are analyzed. A radon problem in the house can then be identified quickly (in days) after the disks are collected and, if necessary, the mitigation can also be initiated shortly thereafter. The whole process is best managed by a radon expert. To perform prompt mitigation, the expert can collaborate with small licensed construction companies. Using this approach, shortly after a radon problem has been identified twenty buildings in Bulgaria (mostly kindergartens and schools) were successfully mitigated.

Introduction

After the radon issue was re-addressed by the World Health Organization (WHO, 2009) and the risk coefficient was nearly doubled by the International Commission on Radiological Protection (ICRP, 2010), the efforts to include protection against indoor radon in the radiation protection legislation were reinforced in many countries. Due to the large temporal variations the averaged measurements over long durations are determining the conclusions for the radon risk, specifically – not over exposure durations of less than 3 months, but instead over longer exposure durations (WHO, 2009). The recent directive of the Council of the European Union (2014) recommends the member states to establish reference levels based on the annual-average radon concentration (these levels should not exceed 300 Bq/m³). The methods scheduled for practical use and decision making based on such reference level should be able to assess the annual average radon concentration, preferably by a direct measurement method.

Traditionally, long-term measurements of indoor ²²²Rn are made by passive radon detectors left indoors for exposure durations that can be as long as one year. The measurements are prospective and the results become available with a delay of months or a year after the detectors are placed in the studied dwelling. This time delay between the decision to test and the eventual problem identification affects the stakeholders' attitude and can reduce the concern about the reality of the radon hazard and the need for measures to reduce it. Our experience indicates that the recommendation “the average radon concentration significantly exceeds the reference level and protective measures are recommended” delivered after months or a year has lost most of its motivating power.

Reporting the annual-average radon concentrations speedily with a sound conclusion of whether the reference level is significantly exceeded (in terms of statistical significance, e.g.,

with larger than 95% probability) would keep the attention of the concerned stakeholders on the problem and would facilitate a prompt decision to mitigate. This is especially valid for public buildings (*e.g.*, kindergartens and schools) where decision makers have a public responsibility and are looking for confidence in a determination whether the reference level is exceeded or not.

The CD/DVD method for radon measurements was first proposed as a tool for retrospective radon dosimetry (Pressyanov et al., 1999; 2001) that is mostly needed for radon epidemiology. The method employs the remarkably high radon absorption ability of the polycarbonate material of which CDs/DVDs are made and its track-etch properties. The track-density at depth greater than 80 μm beneath the front surface of the disk is almost perfectly correlated with the integrated radon concentration (Pressyanov et al., 2001; 2003). In the last 15 years the method has been extensively studied and employed in practice (Pressyanov, 2007; 2009, Pressyanov et al., 2003; 2010). These studies demonstrated sufficient sensitivity for quantitative retrospective measurements within the whole range of concentrations that can be found in the indoor environment (Pressyanov et al., 2003). The comparison between results obtained by CDs/DVDs and those by diffusion chambers (the conventional and widely used method for long-term integrated radon measurements) showed a very good agreement both at high and low levels of indoor radon (Pressyanov et al., 2010). In this report we explore the ability of the CD/DVD method to be employed for the purpose of making a quick identification whether buildings exceed the annual average radon concentration reference level.

Materials and methods

The methodology consists of the following steps:

- **Obtaining proper CD/DVD.** The disks selected for analysis should be at least one year old. Only disks that have been stored (no matter whether bare, in envelopes, cases etc.) in the building of interest are suitable. An important step is the disk dating and the dating uncertainty assessment. In some case the disks can be exactly dated using the disk labels or records, or when the disk is obtained with a magazine that has a date on it, or if bought at the same time as a computer or software which purchase date is known. Otherwise, the disk is dated by an interview with its owner and using labels or records on the disks as controls. In such a case the following approach was followed: the disk owner was asked to specify the time window (t_1 , t_2) within which the disk was purchased or otherwise obtained new and taken indoors. With $\delta = t_2 - t_1$ the width of this time interval, and by assuming a uniform probability distribution within this time interval we use the standard deviation, 0.289δ with which the start time t_m of the exposure duration is known. The most likely start time is taken as the middle of the interval, expressed as $t_m = (t_1 + t_2)/2$. The uncertainty in the subsequent exposure duration, t , between the most likely start time t_m and the disk pick-up time t_3 (which is known exactly) for our analysis is then estimated by the same standard deviation of the start time, 0.289δ .
- **Disk processing.** First, a thin layer (usually about 80 μm) is removed from the front surface of the disk specimen (usually $\frac{1}{4}$ piece of the disk) by pre-etching with aqueous solution of 52% KOH (m/v) and 40% methanol (m/v) at 30⁰ C. This step takes about 90 min. After that the tracks at the studied depth are revealed by electrochemical etching (ECE). The ECE process is performed at effective electric field of 3.0 kV/mm at a frequency of 6 kHz and a

temperature of 25⁰ C. The etching solution is a mixture of ethanol with 6M KOH solution in a 1:4 volume ratio. For the first 30 min the etched surface is in contact with the solution without electric field. After that the electric field is applied for 3 hours. For the processing we use laboratory equipment that includes thermostatic metal plates where disk specimens are placed and a programmable HV/HF generator of 6 kHz frequency and variable effective voltage up to 4 kV¹. A picture of one generator is shown in Figure (1). The etched tracks are counted automatically – by a computer scanner and using dedicated software (Mitev et al., 2010).

The entire analysis can be completed within one day and up to 50 disk specimens can be processed in parallel in the author's laboratory.

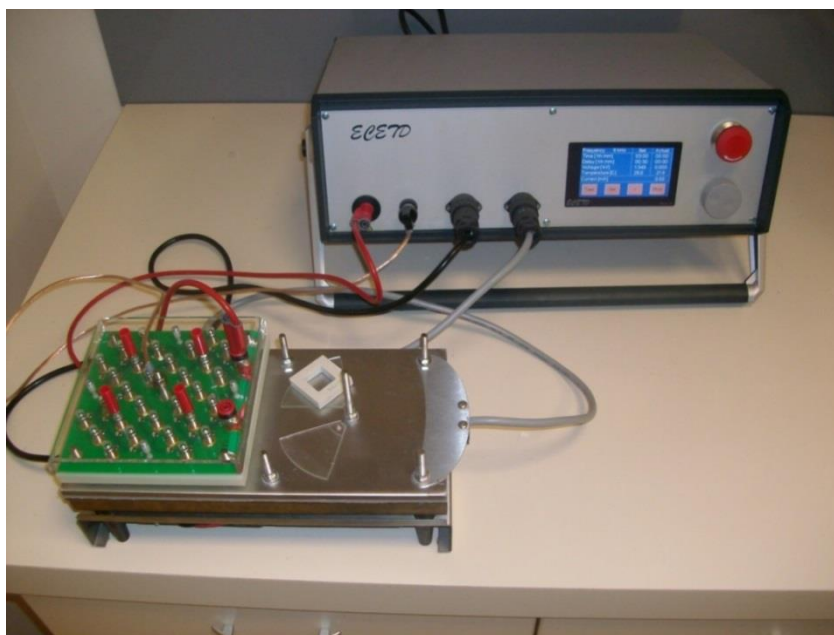


Figure (1): A photo of the thermostatic plate and the programmable HF/HV generator. The generator can be programmed by touch-screen or by an external computer.

Consider the “signal” of the detectors to be the net track density (n_0) at the studied depth. The net track density ($n_0 = \text{total track density} - \text{background track density}$) is determined for each analysed CD/DVD. The integrated ²²²Rn activity concentration (I) is calculated as $I = n_0/CF$, where CF is the calibration factor (Pressyanov, 2009). The CF is determined by exposing the disks to a reference ²²²Rn integrated activity concentration. One feature of the CD/DVD method is that two modes of calibration are possible: standard (*a priori*) and individual *a posteriori* calibration of detectors. The standard CF is the average CF determined for a group of new disks. The individual *a posteriori* calibration consists of additional exposure of a piece of the analyzed disk to a reference ²²²Rn integrated activity concentration in the laboratory. The individual CF of each analyzed disk is determined by the increment of the signal, as described by Pressyanov et al. (2010).

(1) model ECETD, Micon K Ltd., Varna, Bulgaria, <http://www.micon.bg>

The average ^{222}Rn activity concentration C over an exposure duration t of the disk is calculated according to the following formula:

$$C = \frac{I}{t} = \frac{n_0}{CF \cdot t}. \quad (1)$$

The uncertainty in C is calculated by the propagation of the uncertainties in n_0 , CF and t (Taylor and Kuyatt, 1994) resulting in the following relative uncertainty:

$$\frac{u(C)}{C} = \sqrt{\frac{u^2(n_0)}{n_0^2} + \frac{u^2(CF)}{CF^2} + \frac{u^2(t)}{t^2}}, \quad (2)$$

where u denotes the uncertainty in the corresponding quantity. The uncertainty of n_0 ($n_0 = \frac{N}{S} - n_b$) was determined by track-counting statistics (Poisson statistics assumed) and the uncertainty in the background track density (n_b) by the following expression:

$$u\left(\frac{N}{S} - n_b\right) = \sqrt{\frac{N}{S^2} + u^2(n_b)}, \quad (3)$$

where $u(n_b)$ is the uncertainty in the background track density, and N is the total number of etched tracks counted over the etched area S .

Results

The potential of the method to identify dwellings with radon above the reference levels was studied by modelling. First, the sensitivity of the method was analyzed following the approach of Currie (1968). According to Currie (ibid.), a quantitative result should be reported if the signal (net track density in the present case) exceeds a certain “critical” level that depends on the background. This analysis was performed using the experimentally determined average background track density of 6.3 ± 2.4 tracks/cm² (Dimitrova, 2011). The average ^{222}Rn concentrations that correspond to the “critical” level, according to Currie (1968) are shown in Figure (2) for disks of ages in the interval 1 to 10 years.

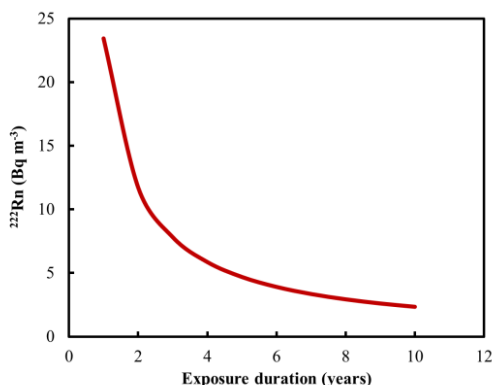


Figure (2): The average ^{222}Rn activity concentrations that correspond to the “critical level for detection” (Currie, 1968) for disks of different age (exposure duration).

The figure shows that measuring concentrations well below 100 Bq/m³ is achievable with any disk exposed more than one year to the building air. This radon concentration is also the

lowest value that WHO recommends as a possible reference level. With disks older than three years, average radon concentrations of 10 Bq/m^3 and below would be measurable. Therefore, we conclude that any disk that is exposed more than one year in the house can serve as a useful detector able to determine the existence of a radon problem in that building, based on a measurement of the average radon concentration over the time it was exposed.

To study the uncertainty (see Eqns. 2 and 3), a calculation was done, assuming a disk with an etched area on which tracks were counted over $S = 4 \text{ cm}^2$. The number of tracks was assumed to follow a Poisson distribution. The average background track-density mentioned before was also assumed. Both standard and individual *a posteriori* calibration were considered. For the standard calibration the numerical value for the relative uncertainty $\frac{u(CF)}{CF} = 0.17$ was used, which was based on our extensive experience with uncertainties in individual *CF*s and individual variations between different brands of CDs/DVDs. For the case of individual *a posteriori* calibration $\frac{u(CF)}{CF} = 0.10$ was used as a conservative choice. The third component in the uncertainty equation is the exposure duration uncertainty. In modelling we have considered two scenarios with different time window δ of the exposure duration. In the first, $\delta = 1$ year was assumed to be constant for all disks. In the second scenario δ was smaller for younger and longer for older disks. In the illustrated cases the ratio δ/t was considered constant and assumed to be 0.25. The second scenario accounts for the fact that the exposure duration of the disks that are exposed shorter can usually be determined with higher accuracy than for the disks that are exposed over a longer time period. For instance, the exposure duration of a CD purchased one year ago can typically be determined to be better than “between 0.5 and 1.5 years ago” ($t = 1$ year, $\delta = 1$ year), while it can be more difficult to determine the exposure duration to within one year interval for a disk that we know is roughly exposed 10 years or more.

With the stated assumptions, the uncertainty of the radon concentration using CD/DVD measurement uncertainty was modelled for different radon levels, disk exposure durations and calibration scenarios. In Figure 3 uncertainties that correspond to the average ^{222}Rn concentrations of 100 Bq/m^3 , 150 Bq/m^3 and 300 Bq/m^3 are shown for standard and individual calibration. For relevance these radon levels were chosen to be existing well known for mitigation decision making: 100 Bq/m^3 is the WHO (2009) lowest guidance level above which to mitigate, 150 Bq/m^3 is the rounded value of the EPA-USA action level (4 pCi/L), and 300 Bq/m^3 is the maximum possible value for a reference level, recommended by the WHO and the Council of the European Union. As the figure indicates, the relative uncertainty decreases with the exposure duration of the disk, but the improvement in the uncertainty with the disk exposure duration is small for disks exposed longer than 4 years and tends to be 18% with standard and 12% with individual calibrations.

Using the uncertainty results, the values beyond which a conclusion can be drawn at 95% statistical confidence can be modelled. This is illustrated in Figure (4) for the reference level of $C_R = 150 \text{ Bq/m}^3$. If the measured value is above the upper lines (that is $C_R + 1.645u(C_R)$, where 1.645 approximates the one-sided 95% confidence interval for random, normally distributed values) there is larger than 95% probability that the mean ^{222}Rn concentration is at or larger than C_R . Alternatively, for values less than $C_R - 1.645u(C_R)$ (the lower lines) the probability that the mean is below C_R is greater than 95%. For the case and under the assumptions illustrated by Figure (4) with a disk with an exposure duration of four years or more and standard calibration used, and for measured concentrations higher than 200 Bq/m^3 one can be confident (with more than 95% confidence level) that the average radon concentration exceeds 150 Bq/m^3 . Similarly, for measured radon concentration smaller than

100 Bq/m³ one can be confident (with more than 95% confidence level) that the average concentration is below 150 Bq/m³. With individual *a posteriori* calibration these confidence borders are closer, namely at 180 Bq/m³ and 120 Bq/m³, respectively.

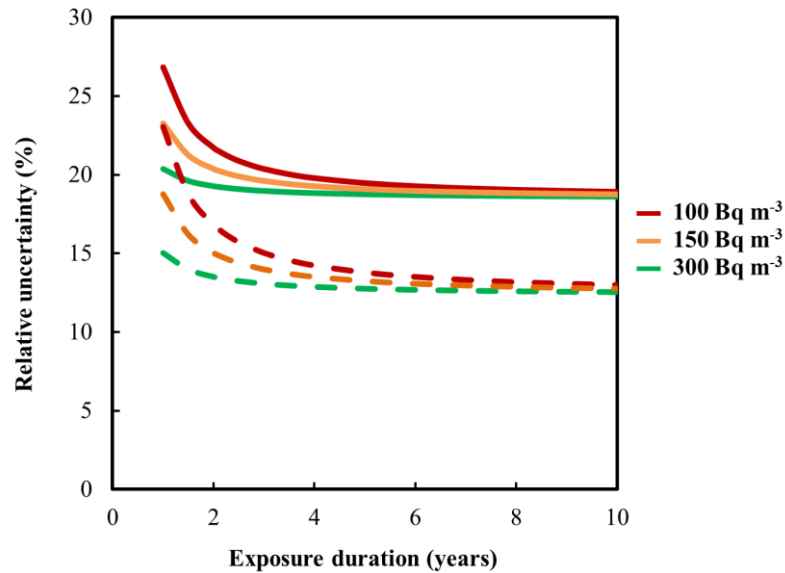


Figure (3): Relative uncertainty under standard, *a priori*, calibration (solid line) and individual, *a posteriori*, calibration (dashed line) using $\delta/t = 0.25$ exposure scenario. The relative uncertainty at levels 100, 150 and 300 Bq/m³ are shown depending on the disk exposure duration.

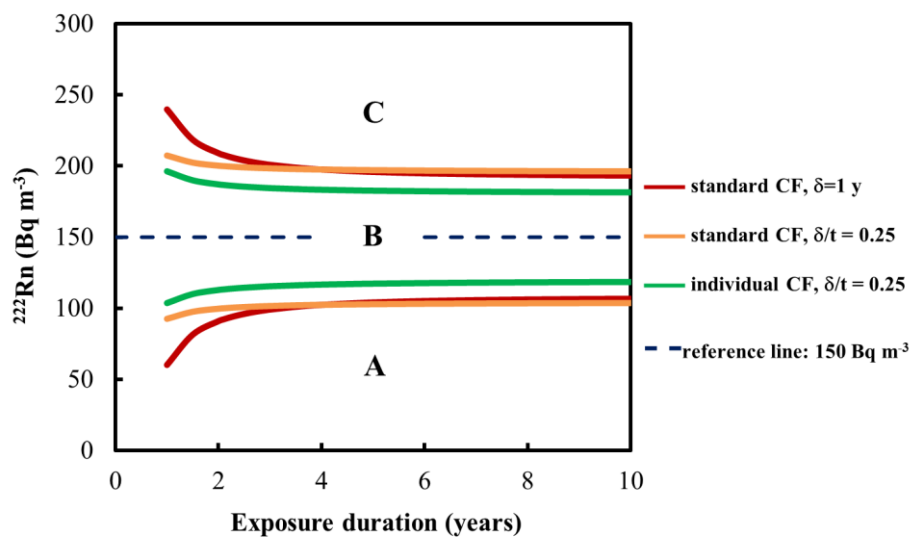


Figure (4): Levels that allow conclusions at the 95% statistical significance for the reference ²²²Rn concentration of 150 Bq/m³. In zone A the conclusion can be drawn that “the average ²²²Rn concentration is significantly less than 150 Bq/m³”. In zone C the conclusion can be drawn that “the level 150 Bq/m³ is significantly exceeded”. In zone B the conclusion “exceeds” or “is significantly less than” cannot be drawn at 95% significance and the result is undetermined in this sense.

Discussion

The results demonstrate that by analysis of any CD/DVD exposed for more than one year the average ^{222}Rn concentration can be quantitatively estimated starting from levels that are sufficiently less than any reference or action level within the interval 100-300 Bq/m³. When the studied building is large and inhomogeneity in radon distribution between different parts or rooms can be expected it is a good practice to analyze several disks from different parts or rooms.

As potential problems we considered the possibility that the results can be biased by erroneous labeling or due to a disk peculiarity - e.g., anomalous individual *CF*, or background that is much higher than the average (if the background is lower than the average, the bias is much lower than the reference values and cannot corrupt the conclusions). Determining the disk exposure time as well as possible is important for the reliability of results and this step needs special care. The critical point here is the confidence in the time interval in the past when the disk was obtained. Our experience indicates that among the typically large number of home CDs/DVDs there are always those that can be reliably dated, especially among the disks that are max. 5-6 years old. Analysis of two (or more) disks stored at the same place can reduce the probability for individual bias due to erroneous labels.

Regarding outliers of the calibration factors, *CFs*, our more than fifteen years of experience indicates that individual variations among different brands of CDs/DVDs are usually within 15% larger or smaller with few exceptions. Having applied this method for more than fifteen years we have observed only one CD-outlier with a calibration factor that was a factor of four smaller (Pressyanov, 2007). This case was easily identified at the processing stage, due to the anti-solvent film coating on the surface and the great retardation of the chemical pre-etching. No other similar case was identified within thousands of analyzed disks so far. Therefore, we consider this case a rare exception and even if it will happen in the future, it will be easily identified during the disk processing. In any case the case of a “*CF*-outlier” will be identified by individual *a posteriori* calibration.

Another potential bias can occur when the analyzed disk has been purchased or is obtained with a very high background. Among more than 40 CDs/DVDs we have analyzed for background only one remarkable outlier was found with a background track density of 49.6 tracks/cm² (Dimitrova, 2011). The analysis revealed, that a disk with such background will provide a result that overestimates the true radon concentration by 245 Bqm³ if the disk is 1 year exposed, 123 Bq/m³ when 2 years exposed, 82 Bq/m³ when 3 years exposed, and about 25 Bq/m³ when the disk is 10 years exposed. Such bias can lead to a false positive result, but only if disks with relatively short exposure times (1-3 years) are analyzed. With older disks the result will be biased upward, but not sufficiently to jump the conclusion from “no problem” to a “confident problem” category. Despite the sufficiently low probability (estimated to be less than 3%) to generate a false positive because of high background, this probability can be further reduced by following the practice to prefer disks that are at least four years exposed, and to analyze at least a second disk from each location. This would also reduce the potential bias incurred from incorrect disk labeling.

The present approach to identify dwellings with high radon uses “detectors” that are available in almost each dwelling and can be applied even when resources needed for large radon detection campaigns are scarce or missing. The prompt reporting and the fact that the results can be directly compared with the reference level can keep the stakeholder’s interest and

motivation on their radon issue relevant. However, the radon measurement alone cannot solve the problem of overexposure to radon and the real protection to humans requires technical measures to reduce the radon levels. To organize this when the stakeholder is determined to mitigate, but there is still no radon mitigation industry on in the regional area, can be challenging. To avoid postponing or cancellation of the mitigation, ad hoc cooperation was organized (with the agreement of the stakeholder) between a radon expert that has the knowledge and experience regarding mitigation, and employing small companies licensed for construction work but not experienced in specific radon mitigation techniques. The process was managed and supervised by the radon expert who organized diagnostic measurements (radon distribution, determined routes of entry, measured pressure field extension, etc.) and provided technical concept for the mitigation project, supervised its execution and organized and conducted the post-mitigation measurements. Within this approach 20 buildings in Bulgaria were successfully mitigated, all of them by active depressurization and ventilation systems. Sixteen of them were large buildings containing kindergartens and schools in the area of Sofia, the capital of the country. The effect of their mitigations is illustrated in Figure (5).

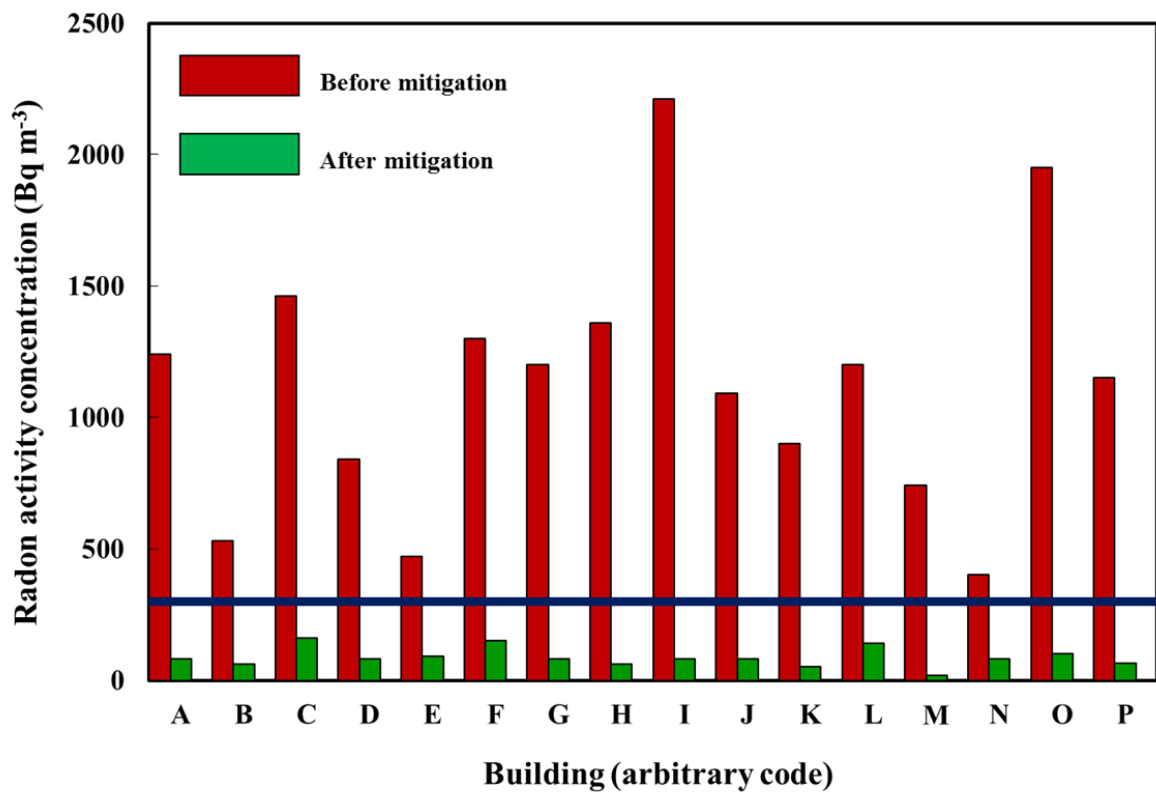


Figure (5): Effect of mitigation (by active sub-slab depressurization) in 16 kindergartens and schools. The effect was studied by grab sampling measurements made with closed room conditions before and after mitigation. The methods and equipment for these measurements are described by Pressyanov et al. (2014). The measurements before were made just prior to the mitigation and the measurements after were made at least one week after the mitigation system was activated. Results for rooms with the highest ²²²Rn levels before mitigation are shown. The horizontal line represents the level of 300 Bq/m³.

Conclusions

We conclude by stating that the CD/DVD method is a useful and fast decision tool for identification if buildings have an annual average radon concentration exceeding a chosen reference level. Both the sensitivity and the uncertainty calculations show that the method is usable for the purpose defined. The calculations show that any CD/DVD exposed for more than one year can serve for this purpose for the current reference levels used. The calculations suggest also that the optimum exposure time interval for disks is four to six years in order to reveal whether the reference level is exceeded when analyzed as discussed. With the CD/DVD method the result for the annual average radon concentration can be available within one day after providing the disk(s) for analysis. The probability for outliers is sufficiently low. In case elevated radon levels are identified we believe it is important the affected stakeholder has the option for immediate consultancy on the next steps towards mitigation. In our experience it is helpful to have a radon expert who manages the work and provides continuity, albeit the mitigation work itself was executed by construction companies. This way the CD/DVD method can take its place in the chain of prompt operative actions, from detection to mitigation, that aim to safeguard human lives from the dangers of indoor radon.

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