COMPARISON OF PASSIVE AND ACTIVE RADON MEASUREMENT METHODS FOR PERSONAL OCCUPATIONAL DOSE ASSESSMENT

by

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To compare the performance of the active short-term and passive long-term radon measurement methods, a study was carried out in several closed spaces, including a uranium mine in Iran. For the passive method, solid-state nuclear track detectors based on Lexan polycarbonate were utilized, for the active method, AlphaGUARD. The study focused on the correlation between the results obtained for estimating the average indoor radon concentrations and consequent personal occupational doses in various working places. The repeatability of each method was investigated, too. In addition, it was shown that the radon concentrations in different stations of the continually ventilated uranium mine were comparable to the ground floor laboratories or storage rooms (without continual ventilation) and lower than underground laboratories.

Key words: active measurement, AlphaGUARD, passive measurement, radon, solid-state nuclear track detector

INTRODUCTION

Radon (^{222}Rn) and its short-lived decay products are the most important contributors of human exposure to ionizing radiation from natural sources. Until the 1970, radon and its progenies were regarded as radiation health hazards encountered only in the mining and processing of uranium ore. As a result of the scientific reports issued by globally trusted organizations and commissions like WHO and ICRP, that notion has significantly changed [1, 2]. Epidemiological surveys have shown that indoor radon is responsible for a substantial number of lung cancers in the general population. The majority of indoor radon concentrations occur in the lower range of a lognormal distribution frequency curve, therefore, the vast majority of radon-induced lung cancers are thought to be a result of exposure to low and moderate radon concentrations [1, 3].

Regarding the considerable cancer risk of low and moderate radon concentrations, accurate estimation of the mean indoor radon concentration is of great importance. Passive long-term techniques such as the use of solid-state nuclear track detectors (SSNTD) are expected to provide proximate estimations of average radon concentrations. This is due to the fact that SSNDT integrates radon activity over a long time (often one to several months). These detectors possess dominant capabilities for spectrometry and for defining the incidence angle of particles (alpha, proton, *etc*.), too [4]. The disadvantages of passive long-term methods are that they are time-consuming and that the majority of them are not real-time. On the other hand, short-term sampling methods such as active radon grab devices are fast and almost real-time. However, they only provide radon information for short sampling times. Despite the variations of radon release rates over time, active techniques are widely used for estimation of average radon concentration levels [1]. The objective of this work is to define the correlation of the active short-term radon concentration measurement method with the passive long-term method (as the preferable approach). To this end, both active and passive methods were utilized to evaluate radon concentration in various closed spaces, including a uranium mine, and the results of the two methods were compared. The repeatability of each method was also investigated.

MATERIALS AND METHODS

The main difference between the passive and the active method is that the former is an integrative

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method which provides no details about the variation of radon concentration over the measuring time, whereas the latter is often appropriate for defining the fluctuations of radon concentration during the measurement period [5].

Despite this major property difference between the two methods, both are commonly used for indoor radon measurement. In order to compare them, a series of measurements was carried out in different stations of a uranium mine, as well as in several other closed spaces over a month-long period in October and November 2015. The devices used in both methods were calibrated at the National Radon Calibration Center, prior to the experiments. The measurements were performed in stations A, B, C, D, and E of the uranium mine. Station A was a ground level office room located in the mine area, station B the main entrance of the mine well. C, D, and E were stations in the underground mine pathways (E was deeper than C and D). Station A was irregularly ventilated through its door or windows (no mechanical ventilation). The underground mine stations were continuously ventilated. Apart from this, measurements were carried out in four other spaces, including a ground floor storage room without any windows or active ventilation (station F), a ground floor office room (G), an underground gamma calibration room (station H), and a ground floor uranium tailing room (station I). The gamma calibration room was equipped with a ventilation fan in operation only during exposure time. None of the normal working conditions of the rooms were changed during the experiments. The repeatability of each method, standard deviation of the measurements, as well as the correlation of the mean concentration obtained by the two methods and compliance of personal doses estimated by the said methods were evaluated and compared.

Long-term measurements by the passive method

In the passive method, the detector used included a small piece of an alpha track detector (film) enclosed in a diffusion chamber covered by a high efficiency particulate air (HEPA) filter. The main films used as alpha track detectors are poly allyl diglycol carbonate (CR-39), cellulose nitrate (LR-115), polycarbonate (Makrofol) and Lexan polycarbonate. In this study, Lexan polycarbonate ($C_{16}H_{14}O_3$) was used for alpha track detection. After subtracting the background counts, the number of tracks per unit surface area (made observable through electrochemical etching) is directly proportional to the integrated radon concentration in Bqh/m^3. Depending on the expected radon concentration, alpha-track detectors are deployed for an exposure period ranging from one month to a year. These detectors are insensitive to humidity, temperature, and background beta and gamma radiations [5]. Nonetheless, very high humidity may form a very thin film of water on the detector surface that stops the alpha particles.

The radon diffusing chamber used in this study is shown in fig. 1. The chamber is a cylindrical plastic container. The diameter, height and internal volume of the diffusion chamber were 66.4 mm, 86.3 mm, and 249 cm^3, respectively. Radon progeny products and atomized aerosols were prevented from entering the cup by the glass fiber filter. The detector film was held at the bottom of the chamber via a plastic holder (adjustable up to 4 cm above the bottom). The thickness, density, and diameter of the used Lexan polycarbonate film were, respectively, 250 μ, 1.29 g/cm^3, and 3 cm.

Frothy two Lexan films techniques were used in this study. Six films were used for background measurements, the others were placed in stations A to I for one month. In each sampling station, four detectors were installed 1 m above the floor, at a distance of 15 cm from the wall, next to each other, in order to investigate the repeatability of the method. All detectors were collected at the same time (after a month) and transferred to the laboratory for processing. The electrochemical etching solution used was a mixture of 15 % KOH, 40 % C_2H_5OH, and 45 % H_2O. The etching was performed at 25 ºC for 3 hours. The etching solution was applied to the irradiated side of the films, while the conductive solution, HCl 3 %, was applied to the opposite side. Track density was converted into radon concentration in Bq/m^3 using the calibration factor. The Lexan film was calibrated at the National Radon Calibration Laboratory of Iran equipped with a 107.285 kBq ^{226}Ra source. To evaluate the background effect of the response of alpha-track detectors, six unirradiated films from the same Lexan sheet were developed under the same etching conditions. The mean track count on the films was 140 track per cm^2,

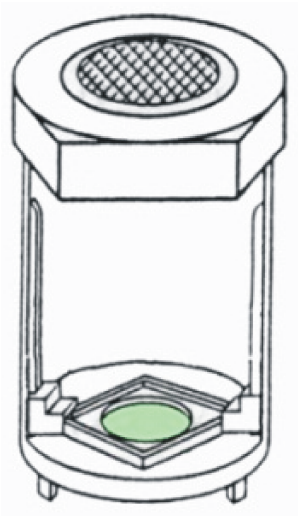

Figure 1. Radon diffusion chamber with the detector

with the standard deviation of 20.6 track per cm^2. The sensitivity of the used chamber to radon gas was 13.55 track per $cm^2/kBq\,d/m^3$ and the minimum detection limit (MDL) of the method for month-long measurements was defined (0.127 kBq/m^3). The overall error (uncertainty) caused by different factors for single passive measurements was 9 %.

The results below the MDL were considered to amount to 0.5 MDL, according to the method proposed by the defense threat reduction agency (DTRA) for personal dosimetry [6]. This is a rational presumption in statistical operations when calculating the mean or cumulative dose values, due to the fact that the measured values below the MDL can neither be considered zero nor altogether omitted from the data.

Short-term measurements by the active method

A radon gas analyzer type AlphaGUARD PQ2000 instrument was used in the diffusion mode to measure radon concentrations. Radon progeny products and aerosol particles were prevented from entering the ionization chamber of the AlphaGUARD by a HEPA filter (only radon-222 passes through). The system also registered air temperature, pressure and relative humidity. Measurements lasting 30 minutes were performed by the AlphaGUARD at installation points of the track detectors. The measurements were repeated daily at all of the stations between 1 p. m. to 4 p. m. for a week. The overall error caused by different factors for the single active measurements was 20 %.

RESULTS AND DISCUSSION

The results of passive measurements in stations A to I are presented in tab. 1. As is shown, all the measured concentrations by the passive method in A and B are below the MDL. Thus, these low radon concentration stations needed longer measuring times (longer measurement time = lower MDL). According to the DTRA method for calculating mean radon concentrations, the values below the MDL are considered to equal 0.5 MDL. Although, when all of the detectors are below the MDL, the standard error of their mean (SEM) is not definable (stations A and B).

The daily readings by the active method in each station were averaged over the weeklong measuring period. The mean ^{222}Rn activity concentrations estimated by the active and passive methods with the related SEM are compared in tab. 1. As can be seen, ignoring A and B for which the passive results < MDL, the mean concentrations measured by the passive method in spaces other than the mine are much higher than those of the active method. This could be due to the fact that ventilation (natural or mechanical) in those spaces was often operative only at active measuring times.

The correlation coefficient between the results obtained by the active (AlphaGUARD) and passive (radon diffusion chamber with Lexan) methods for ^{222}Rn mean concentration is 0.428. This value of the correlation coefficient for the two independent data series represents a medium linear relationship between the two indoor radon measurement methods. A similar study in Saudi Arabia for comparing the short-term and long-term radon measurement methods reported a correlation coefficient of 0.38 between the two methods, with the long-term method showing higher average concentrations [7]. In another study, using weekly active and annually passive measurements, the correlation coefficient between the active and passive method was reported as 0.86 [8]. The high correlation reported in the latter case is, likely, due to the very long sampling period that is not practically feasible in many surveys, particularly not when the active method is concerned.

The mean radon concentrations estimated by the active and passive methods in stations A, B, and E are remarkably close to each other. In station C, on the other hand, the result obtained through the passive method is surprisingly lower than the active method (almost down to a third). Our study revealed that the station had been inactive during the measuring period (no operation) and without effective ventilation at the time. The high humidity in this station probably led to formation of a thin layer of water on both of the chambers' filters and films surfaces which, in turn, stopped the alpha particles and affected the measurement results. The interesting finding of this study is the higher radon concentration in the ground floor storage room (F) and the underground gamma calibration room (H) than in all of the underground uranium mine stations and the uranium tailing room (I) through the passive method. This can be explained by the presence of continuous ventilation in the mine, as opposed to the almost sealed indoor atmosphere of station F and occasional ventilation of station H. The significant differences between the active and passive measurement results for station H can also be related to its occasional ventilation that causes considerable radon concentration variations. For stations F and I, the much lower mean concentrations estimated through the active method than the passive one are explainable based on fresh air entering the room along with the examiners.

Except for station C for which the high humidity is deemed to be responsible for the difference between the passive and active measurement results, the two methods have produced close results wherever the ventilation of the space was almost constant (B, D, and E). In contrast, in spaces with fluctuating radon concentrations due to the frequent opening of doors or windows (G) or due to irregular mechanical ventila-

Table 1. ^{222}Rn activity concentrations measured by the passive and active methods

Station	Passive method			Active method		
	Detectors' readings [Bqm^{-3}]	Average concentrations and standard errors [Bqm^{-3}]	CV	Average concentrations and standard errors [Bqm^{-3}]	Measurements for repeatability check [Bqm^{-3}]	CV
A	<MDL	63.5 (the standard error is not definable)	–	59.0 4.59	–	–
	<MDL					
	<MDL					
	<MDL					
B	<MDL	63.5 (the standard error is not definable)	–	66.5 8.86	–	–
	<MDL					
	<MDL					
	<MDL					
C	145.1	146.3 6.69	0.06	448.2 34.33	350	0.09
	157.4				320	
	154.9				300	
	127.9				323	
D	206.6	300.1 53.77	0.06	407.0 88.89	325	0.36
	228.7				341	
	322.2				338	
	442.8				372	
E	238.6	194.2 52.68	0.07	199.1 34.80	336	0.54
	<MDL				354	
	309.9				310	
	164.8				370	
F	542	764 98.12	0.21	314.42 62.80	460	0.26
	947				333	
	657				435	
	910				293	
G	163	244.4 91.7	0.93	117.57 18.94	128	0.75
	<MDL				40	
	259				20	
	492				28	
H	276	474.7 143.55	0.45	196.28 55.31	447	0.6
	200				299	
	611				162	
	812				207	
I	<MDL	87.6 24.1	0.55	21.67 1.90	30.63	0.2
	<MDL				27.13	
	<MDL				20.5	
	159.9				20.63	

tion (H), as well as in isolated but relatively small spaces F and I where the entry of the examiner (for active measurement) considerably affected indoor air quality, the results of the two methods are very different. In comparison with other mines, the one under study shows a low radon concentration when compared with the reported concentration of about 1800 to above 5000 Bq/m^3 at some points of a phosphate mine [9] and may be compared to a low-grade ore underground mine (Jaduguda mine in India) with radon concentration of a few hundred Bq/m^3 [10].

Repeatability of active and passive methods

For comparison of the repeatability of the two methods, measurements with AlphaGUARD were done four consecutive times (in 1 hour cycles) at the same points in stations C to I (the second column of tab. 1, from the right). The readings of the set of the four radon diffusion chambers in each station were used to calculate the repeatability of the passive method. The standard deviation to the mean ratio – coefficient of variation (CV) – of the methods was calculated using the said data. Stations A and B were excluded from this comparison, owing to their < MDL radon concentrations in the passive approach. The CV of the active method for C, D, and E stations were in the range of 0.06 to 0.93, whereas for the passive method, the CV were between 0.09 and 0.75. Therefore, the repeatability of the passive method was better than that of the active method. This is owing to the fact that the passive long-term measurement is not sensitive to the variation of radon concentration over the

measuring period, while the result of the active short-term measurement is highly dependent on the concentration of radon at the time of grabbing.

Calculation of the effective dose based on the results obtained by active and passive methods

The effective dose was calculated according to eq. (1) [11].

$$E_D \quad (C_{Rn} \quad F)/3700 \quad (t/170) \quad DCF \qquad (1)$$

where E_D is the effective dose [mSvy^{-1}], C_{Rn} – the radon concentration in Bq/m^3, t – the expected annual working time (2000 h per year), F – the equilibrium factor (0.4 for indoor), and DCF – the working level month (WLM) to dose conversion factor (5 mSv per WLM). One working level (WL) is any combination of short-lived decay products of radon in a liter of air that ultimately emits 1.3 10^5 MeV of alpha energy. Based on the ICRP1994, the dose conversion factor expresses the relationship between the effective dose and potential alpha energy concentration of inhaled short-lived radon decay products [12].

The results are depicted in tab. 2. According to the calculations, the average annual radon-induced effective dose of the mine workers (stations A to E) based on the active method is in the range of 0.14-2.85 mSvy^{-1}, while the one based on the passive method is between 0.4-4.58 mSvy^{-1}. Although stations C (mine), F (the ground floor storage room), and I (the ground floor uranium tailing room) were of a very low occupancy factor during the measurement period, their annual effective doses were calculated for full occupation (2000 h/y) for better comparison with the other spaces. Indeed, the actual effective doses of workers in those stations depend on the fraction of total annual working time spent at each station.

The action level proposed by the ICRP is in the range of 3-10 mSv per year and the values obtained by both active and passive methods are less than the lower limit of this threshold for the mine. The calculated annual effective doses are also lower than the annual occupational dose limit (20 mSv averaged over a 5 year period). Of course, these values are related only to the internal effective doses that a worker receives in the mine, albeit the external dose should also be considered.

Table 2. Annual effective dose calculated, based on the results of the active and passive methods

Station	Annual effective dose based on the active method [mSv]	Annual effective dose based on the passive method [mSv]
A	0.37	0.40
B	0.42	0.40
C	2.85	0.87
D	2.59	1.80
E	1.26	1.23
F	1.99	4.58
G	0.74	1.17
H	1.25	2.84
I	0.14	0.55

CONCLUSIONS

In this research paper, indoor radon concentrations were measured in various closed working spaces, including a uranium mine in autumn. Both active and passive methods were employed and their results compared. The results showed that the mean radon concentrations estimated by the passive method using a diffusion chamber (Lexan polycarbonate films) has a relatively low correlation (0.428) with the approximations made by the active method using AlphaGUARD. The background variation of polycarbonate films is not ignorable because it affects the repeatability of the passive method, especially when low and medium radon concentrations are concerned. Our study has shown that the coefficient of variation of the passive method is generally lower than the active method. Moreover, it suggests reliance on the active method only for spaces with minimum fluctuation in their indoor air quality, especially at the time of the active measurements. Therefore, performing measurements by the active method for estimating the average indoor radon concentration is not recommended. The results also confirm the need for making a proper balance between the expected radon concentration and the length of the measuring period for the passive method. Generally, long measuring periods are highly recommended if the expected radon concentration is low or medium.

The estimated projected annual effective doses received by the miners, as well as the workers in other spaces, were calculated. It was shown that radon concentrations in some underground facilities were higher than in low-grade uranium ore mines. The estimated annual doses were lower than the recommended occupational dose limits, however, seasonal variations cannot be ignored. Radon concentrations in the studied closed spaces may be considerably different in summer or winter, due to changes in humidity, natural ventilation and air conditioning.

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AUTHORS' CONTRIBUTIONS

The design is to be attributed to F. Mianji and E. Hasanzadeh. E. Hasanzadeh, carried out the measure-

ments and recorded the data. All authors were involved in analyzing and discussing the results and the manuscript was written by F. Mianji, E. Hasanzadeh, and Y. Sadighzadeh.

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ПОРЕЂЕЊЕ ПАСИВНИХ И АКТИВНИХ МЕТОДА МЕРЕЊА РАДОНА РАДИ ПРОЦЕНЕ ДОЗЕ ПРОФЕСИОНАЛНО ИЗЛОЖЕНИХ ЛИЦА

Како би се извршило поређење активних краткотрајних и пасивних дуготрајних метода за мерење радона, спроведена је студија у неколико затворених објеката, укључујући и рудник уранијума у Ирану. Код пасивне методе коришћени су чврсти траг детектори на бази лексан поликарбоната, док је за активне методе коришћен AlphaGUARD детектор. Студија је усмерена на корелацију процењене средње концентрације радона у затвореном простору са личним дозама радника на различитим радним местима. Такође је проверена и поновљивост сваке методе. Додатно је показано да је концентрација радона у различитим деловима рудника уранијума са сталном вентилацијом упоредива са концентрацијом у приземним лабораторијама или магацинима (без сталне вентилације) и нижа од концентрације у лабораторијама испод површине земље.

Кључне речи: активно мерење, AlphaGUARD, пасивно мерење, радон, чврсти траг детектор