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Investigation of the influence of high humidity and exposure duration on the measurement results of radon concentration by means of PicoRad system in the CLOR calibration chamber

Olga Stawarz,
Maria Karpińska,
Kalina Mamont-Ciesla

Abstract PicoRad system based on activated charcoal collectors and liquid scintillation counter is one of the most popular passive methods of radon measurements which may be used both indoors and in other environments such as caves, tunnels, mines etc. It is well known that charcoal is also a very good adsorber of water vapor and it can reduce adsorption efficiency for radon. In PicoRad collectors, the charcoal is mixed with a dessicant to lower this effect. A series of expositions of PicoRad detectors was performed in a Central Laboratory for Radiological Protection (CLOR) radon calibration/climatic chamber to study the effect of high relative humidity on the airborne radon concentration measurements and the dependence of the results on the duration of exposure. The results obtained from the PicoRad system were referred to the mean concentrations delivered by an AlphaGUARD monitor, Genitron (GmbH), Germany. The main conclusions are the following: 1) the PicoRad system results are not affected by high relative humidity, including extreme values of 90–96%, for the duration of exposure up to ca. 24 h; 2) in the relative humidity range of 75–96% the correction coefficient linearly depends on the duration of exposure reaching a value of ca. 16 for 96 h exposure; 3) the PicoRad system delivers results corresponding better to the mean value of radon concentration in the last 6 h interval of exposure than that in the whole exposure duration. This finding is particularly important for the calibration of PicoRad collectors in the conditions of decreasing radon concentration due to radon decay.

Key words radon • activated charcoal • relative humidity • duration of exposure • PicoRad system

O. Stawarz, K. Mamont-Ciesla✉
Dosimetry Department,
Central Laboratory for Radiological Protection,
7 Konwaliowa Str., 03-194 Warsaw, Poland,
Tel.: +48 22 814 01 59, Fax: +48 22 811 16 16,
E-mail: kalinam@clor.waw.pl

M. Karpińska
Department of Biophysics,
Medical University of Białystok,
2A Mickiewicza Str., 15-089 Białystok, Poland

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Introduction

Although radon dose to lungs comes mainly from short-lived radon daughters, most of its estimations for general population is based on the measurements of radon gas concentration instead of radon progeny and acceptance of a constant equilibrium factor between radon and its short-lived decay products. It is so because integrating passive measurements of radon gas concentration are simpler and cheaper, more convenient and practical from the point of view of people who are the subject of radon exposure. The most common passive methods of radon measurements in large-scale surveys are solid-state nuclear alpha track detectors [1] and activated charcoal collectors [4]. While the track detector method is fully integrating over usually a couple of months or longer, the activated charcoal collectors are used over exposure periods of a few days, up to 6 and the mean radon concentration can be obtained as early as 4 h after the end of exposure [9]. Thus, they are applied when the results are required quickly. The short duration of useful exposure of the charcoal canisters arises from the fact that the charcoal is a collector of radon, but not detector, and the half-life of ^{222}Rn is only

3.8 days. As the charcoal based method utilizes the ability of the activated charcoal to adsorb gases and vapors on its surface, radon, various airborne contaminants and water vapor are competing with each other [2]. Thus, environmental conditions – relative humidity and temperature – may change the efficiency of radon adsorption. Many authors [3, 11] investigated theoretically and experimentally temperature and relative humidity effects for various kinds of charcoal collectors and they obtained incoherent and sometimes even contradictory findings [10]. Charcoal collectors are not perfect integrators because radon adsorption is always accompanied by radon desorption when collectors are left open. George [5] and Cohen [4] showed that in the conditions of large variation of radon concentration the last several hours of exposure have a dominant meaning for the delivered result. After exposure, the collectors must be tightly closed and returned to the laboratory, where they are immediately analyzed either by gamma spectroscopy [5, 12] or by alpha scintillation counting following desorption of radon in the liquid scintillator [6]. The latter technique is utilized in a very popular, commercially available system known as PicoRad, supplied by AccuStar Labs (USA). The PicoRad collectors were designed mainly to be used in indoor climatic conditions. A question arises if they can be used in extreme relative humidity in such places as underground area, spas, bathrooms, cellars.

The aim of this work was to study the effect of high relative humidity and exposure duration on the measurements of airborne radon concentration using PicoRad collectors. The present authors also investigated optimal time interval before the end of exposure to determine calibration factor in the conditions of lowering radon concentration due to its decay in a radon standard chamber.

Experimental

The PicoRad system

The PicoRad system is described in detail by Iimoto *et al.* [7]. It comprises four components: liquid scintillation vials, Insta-Fluor Plus liquid scintillator, liquid scintillation counter (LSC) Canberra Packard TRI CARB 1900TR and NITON Corporation software PicoRad 5.9. The vials of the shape of cylinders 60 mm high and 25 mm in diameter, contain, in the upper part, a small canister filled with a 3 g mixture of activated charcoal and silica-gel, closed with perforated bottom and top. Both things: the vial and canister are made of high density polyethylene not allowing to let radon through. The vial is open during exposure period which can last 12 h to 96 h, with recommended 48 h, according to the Manual of NITON Corporation, PicoRad 5.9, Program for Radon Analysis, Appendix 3. At the laboratory, the vial is eluted with a scintillation cocktail at least 8 h before analysis in the LSC. The cocktail desorbs the radon from the activated charcoal and after 8 h the state of radioactive equivalence of radon with its progeny is formed. Scintillations produced by the alpha and beta particles are counted in the TRI CARB 1900TR analyzer. The analysis is controlled by NITON software program PicoRad 5.9. The program calculates the average radon concentration in the air C (in Bq/m^3) for inserted data on the counts per min, counting time, start and stop time of exposure interval, elution time, average temperature T during exposure. In the software, the temperature dependence described by the exponential function $Ae^{0.036T}$ is applied.

The radon calibration chamber

A series of PicoRad collectors exposures was performed in the walk-in radon calibration chamber installed at the CLOR (Fig. 1). The chamber body, constructed by NEMA Company, Germany, is an air-tight air-conditioned room made of 100 mm PUR sandwich elements covered outside with zinc coated steel and plastic, and inside with stainless steel with DVP (pressure distributing plate). Its inner overall dimensions are: 2.75 m × 2.25 m × 2.00 m and a volume of ca. 12.375 m^3. The chamber has an anteroom of 3.12 m^3 (1.50 m × 1.04 m × 2.00 m), a viewing heated window, manipulating gloves and a number of input ports which permit injecting radon gas and aerosols, sampling inside air and connecting instruments (e.g. aerosol counter) outside. The chamber is equipped with a ventilation system to remove the radon outdoors after experiments.

Climatic conditions – temperature and relative humidity – can be steered manually or automatically. Temperature may be set up from – 30°C to +60°C with stability of ± 1°C and relative humidity from 10% to 95% with a long time margin of ± 5% (for the temperature range from +10°C to +60°C).

To create radon atmosphere in the chamber, an external radon generator, manufactured by Pylon Electronic Development Co., is connected to two ports on the opposite sides of the chamber and the collected radon being pumped into the chamber for half an hour. The activity of the dry flow-through Ra-226 source in the generator is 137.27 kBq (± 4% at a 95% confidence level). Thus, the maximum achievable radon concentration in the chamber at the beginning of exposure interval can be ca. 11 kBq/m^3 and is decreasing with the decay constant rate of radon.

Fig. 1. Radon calibration chamber at the CLOR.

Radon concentration in the chamber is measured continuously by means of an AlphaGUARD monitor manufactured by Genitron Instruments (GmbH), Germany. According to the AlphaGUARD PQ2000/MC50 – Multiparameter Radon Monitor – Characterization of its physical properties, the monitor readings on radon concentration are not sensitive to the humidity up to 100% rH (with non condensing). This was confirmed by checking the correctness of the AlphaGUARD monitor readings against the Pylon radon standard generator in the normal and extremely high relative humidity in the CLOR radon chamber. Such tests are performed periodically in the frame of the quality system. In 2003, this monitor participated in a national interlaboratory comparison which took place in the CLOR radon chamber along with five other AlphaGUARD monitors [8]. The conformity of the results delivered by all six monitors was perfect in the condition of both normal and very high relative humidity. Therefore, the AlphaGUARD monitor could serve as a reference device in this study.

The continuous output record of AlphaGUARD delivered the hourly averaged radon concentrations.

Results and discussion

There were performed thirty three exposures of three PicoRad collectors each at the same temperature of ca. 25 ± 2°C in various conditions of relative humidity in the range from 20% to 96% and for various exposure duration in the acceptable by PicoRad software range from 12 h to 96 h. In order to compare the PicoRad results with the reference radon concentrations obtained from AlphaGUARD monitor, the correction coefficients $CC(t)$ were calculated according to the following formula:

$$CC(t) = AG(t)/\text{PicoR(texp.)}$$

where: PicoR(texp.) is the arithmetic mean for three PicoRad collectors exposed during the exposure duration texp.; texp. is the exposure duration of PicoRad collectors; $AG(t)$ is the average radon concentration calculated from the AlphaGUARD monitor for period t before the end of exposure; t is equal to either the whole exposure duration or the last 24 h or the last 12 h or the last 6 h of the exposure duration; $CC(t)$ is a correction coefficient corresponding to the reference value of $AG(t)$.

In Tables 1, 2, 3 and 4, there are given arithmetic means and their standard deviations of the correction coefficients for:

- three groups of the relative humidity ranges: 20–96%, 20% – ca. 75% and ca. 76–96% and
- four groups of the PicoRad exposure intervals: 16–24 h, 46–48 h, 72 h, 90–96 h.

The tables differ from each other by the parameter t which is the time interval before the end of exposure. The average value of radon concentration for period t calculated from the AlphaGUARD continuous record was considered as reference value for the PicoRad results. In Table 1, the parameter t is equal to the whole exposure period, in Table 2 – to the last 24 h, in Table 3 – to the last 12 h and in Table 4 – to the last 6 h of the exposure period.

The correction coefficients decrease with decreasing value of the parameter t and it reaches the lowest value of 1.22 for t = 6 h which is the average value for all results in the whole range of the relative humidity from 20% to 96%. This value of $CC(t = 6\text{ h})$ probably results from the difference in the calibration factors between the tested PicoRad system and the AlphaGUARD monitor.

The correction coefficient reaches the highest value of 15.7 for high relative humidity, the longest time of exposure texp. and the parameter t, both being equal to 96 h.

In Figs. 2 and 3, there are shown relationships between the correction coefficients $CC(t)$ and the

Table 1. Mean values of correction coefficients and their standard deviations calculated basing on the AlphaGUARD results for t equal to the whole exposure period

Relative humidity	texp. – Exposure period of PicoRad collectors			
[%]	16–24 h	46–48 h	72 h	90–96 h
20–96	**1.32 ± 0.04**			
20–75	1.33 ± 0.05	1.69 ± 0.09	2.41 ± 0.67	2.29 ± 0.03
76–96	1.30 ± 0.07	5.27 ± 0.04	8.40* ± 1.41	15.7 ± 0.9

* Two results.

Table 2. Mean values of correction coefficients and their standard deviations calculated basing on the AlphaGUARD results for t equal to last 16–24 h of exposure time

Relative humidity	texp. – Exposure period of PicoRad collectors			
[%]	16–24 h	46–48 h	72 h	90–96 h
20–96	**1.32 ± 0.04**			
20–75	1.33 ± 0.05	1.52 ± 0.06	1.94 ± 0.56	1.60 ± 0.001
76–96	1.30 ± 0.07	4.66 ± 0.04	6.93* ± 1.17	10.94 ± 0.16

* Two results.

Table 3. Mean values of correction coefficients and their standard deviations calculated basing on the AlphaGUARD results for *t* equal to last 12 h of exposure time

Relative humidity	texp. – Exposure period of PicoRad collectors			
[%]	16–24 h	46–48 h	72 h	90–96 h
20–96	**1.27 ± 0.03**			
20–75	1.29 ± 0.04	1.44 ± 0.05	1.87 ± 0.55	1.52 ± 0.01
76–96	1.23 ± 0.05	4.36 ± 0.04	6.62* ± 1.12	9.91 ± 0.08

* Two results.

Table 4. Mean values of correction coefficients and their standard deviations calculated basing on the AlphaGUARD results for *t* equal to the last 6 h of exposure time

Relative humidity	texp. – Exposure period of PicoRad collectors			
[%]	16–24 h	46–48 h	72 h	90–96 h
20–96	**1.22 ± 0.03**			
20–75	1.24 ± 0.04	1.41 ± 0.04	1.84 ± 0.55	1.46 ± 0.003
76–96	1.18 ± 0.06	4.17 ± 0.04	6.47* ± 1.10	9.68 ± 0.05

* Two results.

exposure time of PicoRad collectors for four series corresponding to the parameter *t* equal to the total

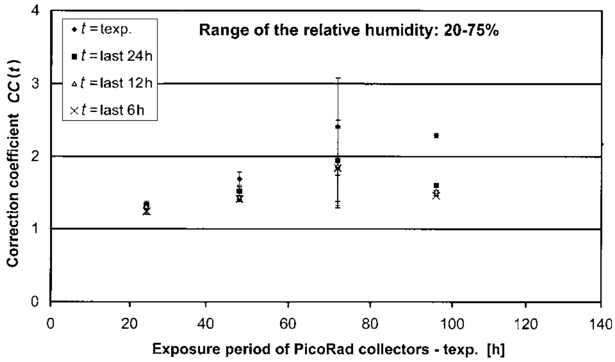

Fig. 2. Correction coefficient *CC*(*t*) vs. time of exposure of PicoRad collectors in the range of relative humidity 20–75%.

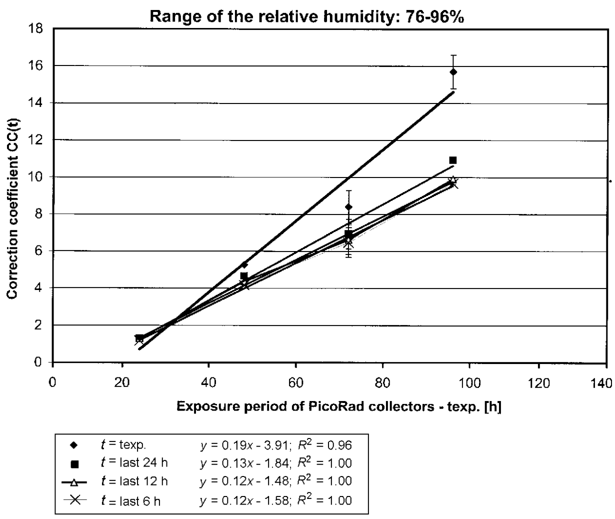

Fig. 3. Correction coefficient *CC*(*t*) vs. time of exposure of PicoRad collectors in the range of relative humidity 76–96%.

exposure time (96 h, 72 h or 48 h) and to the last 24 h, 12 h and 6 h. Figure 2 illustrates the results in the relative humidity range of 20–75% and Fig. 3 – those above 75% rH.

In the range of 76–96% rH, the correction coefficients strongly depends on the period of exposure (texp.). This can be well described by a linear dependence for all four values of the parameter *t*.

In the range of the relative humidity below 75%, this dependence is rather weak and is noticeable only for the series *t* = texp. The weighted average of the correction coefficients calculated for all results from three other series (for parameter *t* equal to 24 h, 12 h and 6 h) is 1.51 ± 0.08. It should be pointed out that the values of correction coefficients for texp. equal to 72 h are burdened with relatively big standard deviations due to the just two measurements taken into account of the mean value.

Conclusions

Results of radon concentration in the CLOR calibration chamber measured by means of the PicoRad system in various conditions of the relative humidity at constant temperature and for various duration of exposure were compared with the reference values obtained from the AlphaGUARD monitor. This comparison indicates that the lowest correction coefficient *CC*(*t*) of 1.22 is constant for the exposure duration of 16 h to 24 h in the long range of the relative humidity from 20% to 96%. This means that for the exposure shorter than ca. 24 h the desiccant applied in the PicoRad collectors efficiently prevent adsorption of water vapor in the charcoal.

A comparison of *CC*(*t*) for different values of the parameter *t*, which is the lenth of the end of exposure period of the AlphaGUARD continuous record considered as reference, shows that for all groups of results the comparison coefficients have the lowest values for t equal to the last 6 h of the exposure period. This suggests that the PicoRad collectors "remember" the concentration from the last hours rather than the mean for a longer period. This can be important in the conditions of changing radon concentration particularly in the calibration of PicoRad collectors in atmosphere of decreasing radon concentration due to radon decay.

In the range of high relative humidity, above 75%, the correction coefficients increase linearly with increasing exposure duration in the range from 16 h to 96 h for all the investigated values of t and the maximum value reaches 15.7 for t = texp. = 96 h.

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