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RADON MEASUREMENTS WITH CHARCOAL CANISTERS
Temperature and Humidity Considerations

by

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Radon testing by using open-faced charcoal canisters is a cheap and fast screening method. Many laboratories perform the sampling and measurements according to the United States Environmental Protection Agency method – EPA 520. According to this method, no corrections for temperature are applied and corrections for humidity are based on canister mass gain.
The EPA method is practiced in the Vinča Institute of Nuclear Sciences with recycled canisters. In the course of measurements, it was established that the mass gain of the recycled canisters differs from mass gain measured by Environmental Protection Agency in an active atmosphere. In order to quantify and correct these discrepancies, in the laboratory, canisters were exposed for periods of 3 and 4 days between February 2015 and December 2015. Temperature and humidity were monitored continuously and mass gain measured. No significant correlation between mass gain and temperature was found. Based on Environmental Protection Agency calibration data, functional dependence of mass gain on humidity was determined, yielding Environmental Protection Agency mass gain curves. The results of mass gain measurements of recycled canisters were plotted against these curves and a discrepancy confirmed. After correcting the independent variable in the curve equation and calculating the corrected mass gain for recycled canisters, the agreement between measured mass gain and Environmental Protection Agency mass gain curves was attained.

Key words: radon, charcoal canister, mass gain, humidity, calibration factor

INTRODUCTION

Radon testing by open-faced charcoal canisters has been practiced for several decades, but the basic ideas were introduced as early as 1906 by Ernest Rutherford and applied by Arthur Eve in 1907 [1].Today, many laboratories (including 3 from Serbia) perform measurements according to the Environmental Protection Agency (EPA) 520 method (in the following text: EPA method or Method) using open-faced canisters [2]. The method was established in 1980s, based mostly on the work of George [3], as well as contributions from other authors [4-6]. The advantages of the method involving passive charcoal canisters are several: it does not require a power source or trained staff at the measurement site, it is cheap and simple, its exposure times are 2 to 6 days, and it is fast compared to other passive methods [3]. On the other hand, the relative humidity (*RH*) in air, temperature and other factors can have a significant influence on the efficiency of radon collection of charcoal canisters

and it is often not possible to estimate the influence and apply necessary corrections. Additionally, radon concentrations on the measurement site can vary by a factor of 10 or more, in which case the activated charcoal canisters do not have integrating properties, especially for longer sampling times [7, 8]. Because of the mentioned disadvantages, the EPA method has a large uncertainty which is not always easy to estimate and is, therefore, mainly used as a screening method.

The EPA method is used to measure ^{222}Rn, but not other radon isotopes. This isotope with its short-lived progeny is a significant health hazard. It is naturally occurring and a member of the uranium series (a ^{226}Rn daughter). Typically, indoor concentrations are much larger than outdoor concentrations and indoor radon originates from soil, water and building materials [9].

In the case of commercially available canisters, calibration curves are often provided with the canisters. However, manufacturers often provide EPA calibration curves, more specifically, curves from the EPA addendum for an active atmosphere [10]. Some authors have already expressed concern as to whether

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EPA calibration curves are appropriate for commercial canisters [11]. Even if the canisters are made of the same charcoal type, with the same mass, design and dimensions as EPA canisters, they may be used differently. The most obvious difference is using recycled canisters as opposed to using canisters as received from the manufacturer. This distinction is important because the correction for humidity is based on canister mass gain during sampling and recycled canisters typically contain a smaller quantity of adsorbed water than the new canisters, which introduces bias.

The EPA method is practiced in Vinca Institute of Nuclear Sciences (VINS) with recycled canisters. In the course of measurements, it was established that the mass gain of the recycled canisters differs from the mass gain measured by EPA in active atmosphere [12]. In order to quantify and correct for these discrepancies, a year-long measurement campaign was organized in 2015 in a laboratory building with monitored humidity and temperature. Mass gains were predicted based on EPA calibration data [10, 11] and compared with measured mass gains and a correction suggested. Additionally, the influence of temperature on mass gain was investigated.

EPA METHOD, CALIBRATION CURVES AND ASSOCIATED PROBLEMS

The EPA method is based on radon adsorption of activated charcoal and subsequent measurement of radon progeny gamma energies on a gamma spectrometer. Low activity activated charcoal with good adsorption characteristics is packed in cylindrical tin canisters and covered with stainless steel screens. The canister is closed prior to and after radon sampling with a padded tin lid, to prevent radon exchange between the activated charcoal and the atmosphere. The suggested sampling time is between 48 h and 144 h [2].

After the sampling, at least 3 hours should elapse before the measurement on the gamma spectrometer for the secular equilibrium to be achieved between radon and its progeny. The measurements are performed in the region of the gamma spectrum that contains 295 keV and 352 keV gamma energies of ^{214}Pb and ^{214}Bi gamma energy at 609 kV. Usually, one continuous region of interest (ROI) that contains all three gamma energies is selected [2, 13].

After the measurement, radon concentration is determined from eq. (1), where A_{Rn} is the radon concentration in the air, N – the net count in the selected ROI, C_f – the calibration factor, D_f – the decay factor, E – detector efficiency and t – the sampling time [2, 3]

$$A_{Rn} \quad \frac{N}{C_f D_f Et} \qquad (1)$$

In order to obtain the net count, background spectrum must be subtracted from the canister spectrum. Usually, a canister that is never exposed to the atmosphere is used as the "background" canister and its spectrum is collected in the same geometry as for the exposed ones. The decay factor allows for radon radioactive decay and is calculated for the time between the sampling midpoint and the start of measurement. Detector efficiency is determined by measuring the "standard" radon canister with the known concentration of ^{226}Ra in equilibrium with its progeny. Standard canisters should be in all other aspects identical with regular radon canisters. The calibration factor is expressed in volume per time and is obtained empirically. It is used to relate radon concentration in air to radon activity adsorbed in the charcoal bed [2, 3].

For indoor measurements, the most important influence quantities are humidity and radon sampling time and the corrections for these quantities are performed according to the EPA method. They are based on calibrations for a batch of canisters exposed at 3 different humidity levels and for 6 sampling times. The calibration factor for every single measurement is determined from eq. (2) which is obtained simply by rearranging eq. (1)

$$C_f \quad \frac{N}{A_{Rn} D_f Et} \qquad (2)$$

Humidity is usually not known during the sampling so, instead, the difference between the canister mass after sampling and the mass before sampling is used (canister mass gain) to estimate humidity. Four calibration curves are generated based on this data. The first curve is the calibration factor *vs.* mass gain for 48 hour exposures only (in the following text: calibration factor curve, C_f curve). The remaining three calibration curves, adjustment-factor curves or A_f curves, provide adjustment factors used to "adjust" for sampling times other than 48 hours.

Adjustment factors have the same units as calibration factors and the same values for 48 hours sampling. However, C_f values for times other than 48 hours were not published and the way in which A_f values were determined is not completely clear from the EPA method. The A_f curves are generated for low (20 %), medium (50 %) and high (80 %) humidity [2].

With the calibration data available, C_f is calculated for any exposure, based on mass gain and sampling time, according to eq. (3)

$$C_f \quad C_{f,48} \frac{A_{f,t}}{A_{f,48}} \qquad (3)$$

First, $C_{f,48}$ is determined from the C_f curve, based on mass gain. Then the appropriate A_f curve is selected, based on following criteria: if mass gain is lower than 1 g, the low humidity curve is used; if mass gain is between 1 g and 4 g, the medium humidity curve is used; if mass gain is higher than 4 g, the high humidity curve is used. A_f values for 48 hours ($A_{f,48}$) and sampling time ($A_{f,t}$) are determined from the selected curve [2].

The EPA calibration method [2] was performed in a static atmosphere. However, field studies had shown that, even if the canisters were deployed according to EPA 520, measured values were larger than the true average. This discrepancy was attributed to air movement in the vicinity of the canisters during field measurements. Therefore, new calibrations were performed in an active atmosphere and the results were published in the EPA addendum [10]. There are significant differences between calibration and adjustment factors, but also between the mass gain and sampling time for the same humidity, which underscores the importance of the EPA method airflow. EPA calibrations were performed with canisters as received from the manufacturer [11], but canisters that are recycled by heating have also been used [3].

THE INFLUENCE OF TEMPERATURE AND HUMIDITY ON CALIBRATION FACTOR

Ambient temperature and calibration factor

According to the EPA method, no corrections are performed for ambient temperature during radon sampling [2] which causes the method to be much simpler and cheaper. Temperature corrections would introduce an additional parameter to the calibration procedure, causing a drastic increase in the number of calibration measurements. Also, it would be necessary to monitor temperature during every field exposure which would, in turn, require the use of additional equipment and personnel. However, according to the published findings, a significant influence of temperature on the calibration factor has to be taken into account.

The EPA method is mostly founded on the work of George who found "no discernible differences in the response of the device" for temperatures between 18 °C and 27 °C [3]. Therefore, it seems valid that the correction for temperature is not included in the EPA method. However, other authors found a significant dependence of charcoal adsorptive properties on temperature. One group of authors found a 68 % decline in radon adsorption when temperature increased from 15 °C to 30 °C [14]. Another group, however, found a decline of 28 % when temperature increased from 10 °C to 27 °C for 50 % humidity, and a 40 % decline for 80 % humidity. If temperature increases from 16 °C to 27 °C, the decline is lower: 21 % and 24 % [8].

It is evident that each group of authors chose a different temperature range. The question is what temperature range is appropriate, having in mind that radon sampling is performed indoor and extreme temperatures are highly improbable. The answer is different for different applications and climates, but in most cases the range between 16 °C and 27 °C should be sufficient. According to data from [8], if calibration

is performed at 21 °C (70 °F), as is the case with EPA calibration curves, the uncertainty introduced by an unknown temperature during radon sampling should not surpass 12 % (assuming rectangular distribution, $k = 1.73$).

Humidity and calibration factor

As already mentioned, the influence of humidity is corrected for by estimating humidity based on canister mass gain. There are two problems in this approach, that of application and the one of design.

The application of the correction method is questionable because, even if the humidity is the same during calibration and field exposure, mass gain might be different. This might be due to airflow or dissimilar canister preparation [10, 11], but also due to ambient temperature and, possibly, an unalike charcoal mass in the canisters of the same batch. The last problem might especially occur with recycled canisters that were repeatedly mailed to and used by the end users, during which some of the charcoal might be lost.

The design problem is constituted by the fact that there are only 3 A_f curves for the 3 levels of humidity. Therefore, for any sampling time other than 48 hours, the dependence of A_f on humidity is a step function. In eq. (3), $C_{f,48}$ is modified by the ratio of $A_{f,t}$ and $A_{f,48}$. This ratio is calculated for the tabulated data published in the EPA addendum for different sampling times (tab. 1) [10]. The difference between the low humidity and medium humidity curve is of the order of magnitude of several percents for any sampling time. However, the difference between medium humidity and high humidity curves is significant, possibly because the breakpoint was achieved for high humidity exposures [15]. The difference becomes larger with longer sampling time, the largest difference being 62.5 % for 144 h. Therefore, it is of critical importance to use the appropriate Af curve, especially for long sampling time and medium/high humidity.

Dependence of mass gain on humidity, sampling time and temperature

In the EPA addendum, mass gain *m* data are published only for 48 h sampling time. However, data for all sampling times in an active atmosphere are published in [16], presumably received from EPA in pri-

Table 1. Comparison of $A_{f,t}/A_{f,48}$ for different sampling times for all three humidity levels

RH \ t	24 h	48 h	72 h	96 h	120 h	144 h
Low	1.31	1.00	0.81	0.69	0.61	0.56
Medium	1.36	1.00	0.80	0.68	0.58	0.52
High	1.51	1.00	0.70	0.55	0.43	0.32

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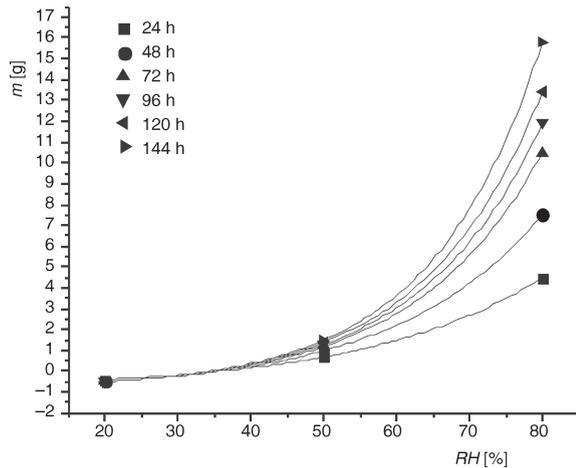Nuclear Technology & Radiation Protection: Year 2016, Vol. 31, No. 1, pp. 65-72

Figure 1. Mass gain dependence on time and humidity, based on the data published by Blue and Jarzemba [16]; solid lines represent fitting by eq. (4)

vate communication [11]. This data is graphically presented in fig. 1. The dependence of mass gain on humidity is different for different sampling times, so 6 curves in total (mass gain curves) should be generated. All of the curves that were generated can be represented by eq. (4)

$$\Delta m_c \quad A e^{RH/b} \quad C \qquad (4)$$

where, Δm_c is the canister mass change during calibration exposure and A, b, and C are fitting parameters. It is important to note that canisters used during calibration are not necessarily prepared in the same way as canisters used for on-site measurements and, hence, the distinction.

To our knowledge, there are no published data on the dependence of mass gain on temperature, presumably, because this influence was negligible. In this paper, this assumption was tested.

EXPERIMENTAL PROCEDURE

The Radiation and Environmental Protection Laboratory of VINS uses commercially available radon canisters. With the canisters, the manufacturer provided EPA-generated curves for the active atmosphere. Since no radon calibration chamber is available in Serbia, radon canisters are used with the received curves. Canisters are recycled after the use by heating for 3 hours at 105 °C and then reused. All data presented in this paper are obtained via recycled canisters.

In the VINS laboratory, the canisters were exposed for periods of 3 and 4 days between February 2015 and December 2015, so as to cover all four seasons. Temperature and humidity were monitored continuously, since it is not possible to adjust humidity in the Laboratory.

As all of the four seasons are covered, the temperature and humidity ranges included in the experiment are typical of indoor conditions in the type of climate involved.

During every exposure, at least two canisters were exposed in two different rooms (up to 10 canisters at the same time). However, only the results obtained in the room where the thermo-hygrometer is located are reported in this paper.

The thermo-hygrometer was duly re-calibrated and all reported results corrected according to the calibration certificate. Thermo-hygrometer records evaluate every minute, according to which the average value was calculated.

RESULTS AND DISCUSSION

Canister mass gains, along with canister exposure time, average humidity and temperature are presented in tab. 2. Variations of temperature and humidity are presented with the standard deviation. The uncertainty of determining mass gain was 0.02 g and is not shown in tab. 2.

Correlation between temperature and mass gain

The assumption that the influence of temperature on mass gain can be neglected for typical exposure conditions was tested. In order to minimize the influence of humidity, the correlation between temperature and mass gain was calculated for canister exposures in a narrow humidity interval. Most canisters were exposed in a humidity interval between 55 % and 65 %. This interval was chosen because it provided the most data points.

The calculated value of Pearson correlation coefficients is 0.250, while the Spearman correlation coefficient is 0.289. Based on VINS experimental data, there is no significant correlation between temperature and mass gain in the covered temperature interval. The data are graphically represented in fig. 2.

Comparison of experimental data with EPA mass gain: humidity curves and proposed correction

Experimental data from tab. 2 are graphically presented in fig. 3. EPA mass gain curves for 3 and 4 days are plotted in the same graph for comparison. From fig. 3(a), it is evident that there is a significant difference between the two datasets. However, the question is whether a suitable correction could be found so that EPA data can be used for recycled canisters.

Table 2. Canister mass gain and average temperature and humidity with standard deviations

3 day exposures			4 day exposures		
Mass gain [g]	Average humidity [%]	Average temperature [°C]	Mass gain [g]	Average humidity [%]	Average temperature [°C]
1.81	43.2 1.1	16.2 1.3	2.37	49.1 2.7	17.03 0.57
1.86	43.0 1.7	17.4 1.0	2.47	46.4 1.7	18.0 1.2
2.07	47.51 0.76	15.7 1.4	2.52	47.4 1.3	18.7 1.1
2.15	45.5 1.4	17.58 0.56	2.78	48.0 2.4	16.53 0.63
2.30	49.0 2.3	16.8 1.2	2.87	47.8 1.2	18.29 0.90
2.44	47.8 1.0	16.6 1.3	3.04	49.2 2.3	17.92 0.77
2.45	50.3 2.6	18.35 0.75	3.05	49.2 1.4	18.3 1.0
2.51	45.65 0.46	17.1 1.7	3.10	47.8 1.3	17.7 1.4
2.55	51.5 2.1	17.5 1.2	3.61	50.7 4.5	22.88 0.74
2.80	49.3 1.1	18.2 1.4	3.78	48.4 3.8	19.18 0.79
2.91	49.7 2.9	16.41 0.98	4.01	54.2 1.4	24.68 0.62
3.02	49.22 0.39	17.9 1.5	4.11	50.8 3.1	19.2 1.4
3.15	55.6 1.4	22.76 0.27	4.31	55.4 2.0	18.33 0.17
3.16	49.0 1.8	18.28 0.96	4.40	54.5 1.7	18.07 0.93
3.20	53.2 2.0	24.70 0.24	5.35	56.9 1.3	17.92 0.33
3.22	51.82 0.87	17.10 0.89	5.46	56.2 1.9	22.10 0.43
3.44	54.8 1.3	17.6 1.0	5.46	62.4 3.2	19.61 0.52
3.87	50.8 1.1	18.94 0.41	5.47	59.4 3.5	24.1 1.0
3.91	53.2 1.5	17.3 1.1	5.50	57.2 3.2	19.29 0.71
3.93	57.9 3.8	25.44 0.38	5.53	56.2 1.9	25.23 0.47
4.09	53.8 1.5	19.08 0.31	5.74	57.6 3.3	20.32 0.29
4.35	56.2 1.6	20.29 0.22	6.15	59.6 1.6	19.33 0.81
4.51	58.9 1.6	21.39 0.32	6.32	60.5 1.7	19.24 0.55
4.76	61.6 2.4	20.46 0.11	6.37	61.4 2.6	23.54 0.25
5.04	59.85 0.69	20.37 0.65	6.81	61.6 4.7	24.86 0.52
5.17	60.5 1.2	18.54 0.67	6.86	58.7 2.2	25.28 0.54
5.35	62.29 0.53	18.55 0.45	7.05	62.0 2.7	21.58 0.54
5.41	56.9 1.3	25.97 0.50	7.24	63.2 1.7	23.33 0.34
5.58	57.4 1.7	25.27 0.65	7.42	59.9 3.1	22.70 0.22
5.63	62.51 0.63	23.52 0.24	8.39	64.1 1.4	22.66 0.39
5.69	62.0 1.1	24.59 0.34	8.52	60.1 1.3	25.37 0.55
5.75	61.43 0.91	21.76 0.31	9.05	65.7 3.4	22.35 0.66
5.93	62.9 1.8	23.66 0.46			
5.99	64.0 1.8	21.52 0.30			
7.35	64.1 4.4	21.39 0.16			
8.66	66.6 2.5	23.67 0.20			

The correction was introduced and explained in the following way: if new canisters contain the amount of water vapor that corresponds to the equilibrium at a certain humidity, recycled canisters contain a smaller amount of water corresponding to the equilibrium at the humidity in question. The difference between the two humidity levels is denoted by RH_0. Since the manufacturer claims that EPA calibration curves are appropriate for the canisters used by VINS, curves modified by taking into account RH_0 should be able to explain the experimental data.

If RH_0 is included in eq. (4), eq. (5) is obtained as

$$\Delta m_r = A e^{(RH - RH_0)/b} + C \qquad (5)$$

where Δm_r is the mass gain measured when using recycled canisters. RH_0 values for 3 and 4 days were determined by fitting experimental data by eq. (5), while parameters A, b, and C were fixed at values obtained by fitting EPA data by eq. (4). The only parameter varied was RH_0. The obtained values were 9.1 % and 10.5 % for 3 and 4 days, respectively.

In order to check if the correction by RH_0 is appropriate, the adjusted R^2 value was determined. Its values are 0.873 for 3 day sampling and 0.864 for 4 day sampling. The strong correlation suggests that the
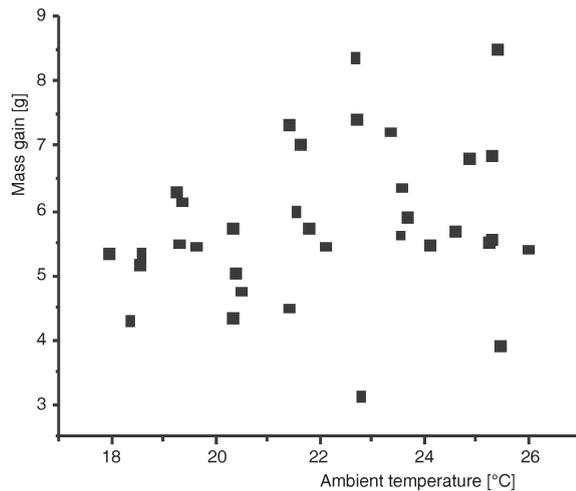
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Figure 2. Dependence of mass gain on ambient temperature: *RH* = 60 %   5 %

modified EPA mass gain curves can, thus, be used to predict mass gains for recycled canisters. Modified curves are presented in fig. 3(b).

Application of the suggested correction

EPA calibration curves are based on the mass gains of new canisters. In order to use EPA calibration curves, it is necessary to find the dependence of Δm_r on Δm_c. If eq. (5) is divided by eq. (4) and solved for Δm_c, eq. (6) is obtained

$$\Delta m_{rcorr} \quad (\Delta m_r \quad C)e^{RH_0/b} \quad C \qquad (6)$$

In eq. (6), Δm_c is denoted Δm_{rcorr} to indicate that it represents the corrected mass gain for recycled canisters and not the mass gain measured by EPA. The correction is applied in the following way: parameters C and b are determined from calibration data for new canisters and parameter RH_0 from the differences between curves for new canisters and recycled canisters. Mass gain, Δm_r is measured for recycled canisters and, finally, Δm_{rcorr} is calculated. The corrected mass gain is used to obtain C_{f48} from the calibration factor curve and to select the appropriate adjustment factor curve.

CONCLUSIONS

The most important influence quantities for radon measurements by using open-faced charcoal canisters are humidity and radon sampling time and the corrections for these quantities are available in the EPA method. However, even though the commercial canisters used by VINS were received from the manufacturer along with the EPA addendum calibration curves, some discrepancies between mass gains obtained by EPA and by VINS were observed. The dis-
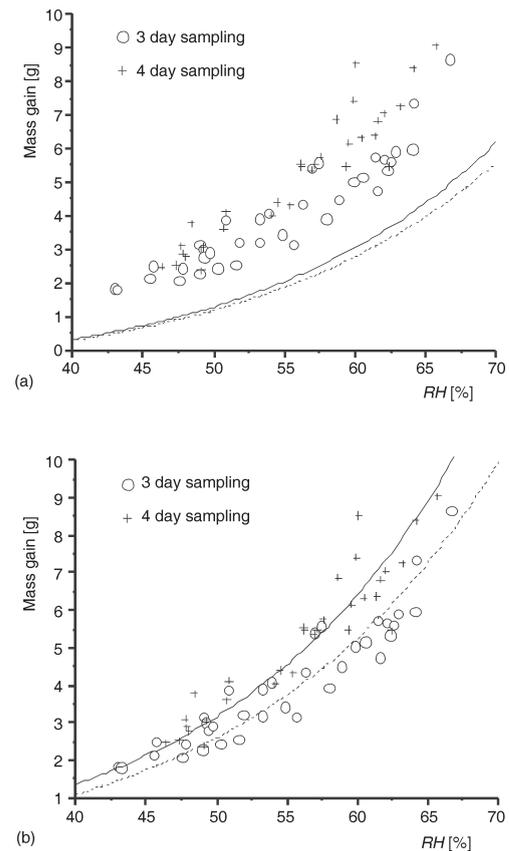

Figure 3. Experimentaly obtained mass gain and curves generated from EPA data; the dashed and solid lines are for 3 and 4 day exposures, respectively;
(a) original curves obtained by fitting EPA data by eq. (4),
(b) corrected curves obtained by fitting VINS data by eq. (5)

crepancies originated from using recycled canisters resulting in larger mass gains.

It has been shown that EPA mass gain curves are in good agreement with VINS experimental data if the humidity offset, RH_0, is introduced. The calculated values of RH_0 were 9.1 % and 10.5 % for the 3 and 4 days sampling time. These data were used to calculate m_{rcorr}, corrected mass gains for recycled canisters, by combining modified and unmodified mass gain curves. It is necessary to estimate m_c, because EPA addendum calibration curves are based on mass gains of new canisters. The proposed correction will be tested in the future interlaboratory comparisons for radon concentration measurements.

The correlation between temperature and mass gain was also investigated. An analysis of the results has shown that no significant correlation exists.

AUTHORS' CONTRIBUTIONS

The manuscript was written by M. Z. Živanović and J. D. Nikolić, and figures were prepared by M. Z. Živanović. Experimental data were provided by all authors. The theoretical analysis was carried out by G. K. Pantelić and M. Z. Živanović. Mass gain curve correction was proposed and explained by J. D. Nikolić and M. Z. Živanović. All authors analyzed and discussed the results and reviewed the manuscript.

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**МЕРЕЊЕ РАДОНА ПОМОЋУ УГЉЕНИХ КАНИСТЕРА
Разматрање утицаја температуре и влажности**

Испитивање концентрације радона помоћу угљених канистера је брза и јевтина скрининг метода. Многе лабораторије спроводе узорковање и мерење у складу са методом Агенције за заштиту животне средине Сједињених Америчких држава (ЕПА) – ЕПА 520. Према овој методи, корекције на температуру се не примењују, док су корекције на влажност засноване на промени масе канистера.

У примени ЕПА методе у Институту за нуклеарне науке "Винча", користе се рециклирани канистери. У току мерења утврђено је да промена масе рециклираних канистера не одговара променама масе измереним од стране ЕПА у активној атмосфери. Да би се утврдила разлика и извршиле потребне корекције, у Институту су спровођена тродневна и четвородневна излагања канистера од фебруара до децембра 2015. године. Температура и притисак су континуално мерени у лабораторији и мерена је промена масе. Није утврђена значајна корелација између промене масе и температуре. На основу ЕПА калибрационих података конструисане су криве зависности промене масе од влажности. Резултати промене масе за рециклиране канистере упоређивани су са наведеним кривама и разлике су потврђене. Пошто је извршена корекција фунцкије која описује зависност промене масе од влажности, а вредности промене масе за рециклиране канистере кориговане, постигнуто је слагање са ЕПА кривама.

Кључне речи: радон, угљени канистер, разлика маса, влажности, калибрациони фактор