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Conversion factor and other variables in the indoor retrospective radon activity concentration studies

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ABSTRACT

Estimation of indoor retrospective radon activity concentration can be performed by measuring the ^{210}Po surface activity concentration of the radon progeny on mirrors. The conversion factor between the surface activity concentration of ^{210}Po on mirrors and the retrospective activity concentration of ^{222}Rn in the air was determined. Several places and some variables have been considered. Exposures were performed in places with moderate and high concentrations of ^{222}Rn. The repeatability of results has been investigated. The dependence of concentration on exposure time has been checked. The effect of cleaning the mirror surfaces has been analyzed. The experimental results obtained for the possible ^{210}Pb-^{210}Po equilibrium inside the mirrors have also been studied and compared with the theoretically expected values.

1. Introduction

Continued inhalation of air with high radon content is the second cause of lung cancer, after tobacco smoking, as well known and recognized by the World Health Organization (Zeeb and Shannoun, 2009). Determination of indoor radon activity concentrations is therefore very important, and surveys for measurements have been designed not only for dwellings but also for working places (Martín Sánchez et al., 2012). To study the cause-effect relation, retrospective dosimetry can be a helpful tool. In this way, measuring methods requiring the determination of indoor radon activity concentration in a place in the past are necessary (Alavanja et al., 1999). One of these methods is the direct measurement of the ^{210}Po on the surface of mirrors that spend a very long time in a room. Decays of ^{222}Rn (half-life of 3.82 d) produce ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po, which are short-lived nuclides. They can be attached to aerosols and dust particles, with later deposition on the surface of the objects. Implantation of ^{210}Pb (half-life of 22.3 a) into a surface is produced by recoil of ^{214}Po. The ^{210}Pb decays to ^{210}Bi and ^{210}Po, and then, a direct measurement of ^{210}Po, in transient equilibrium with ^{210}Pb, can be performed (Martín Sánchez and de la Torre Pérez, 2012).

The ratio between the average ^{222}Rn activity concentration in air (A [Bq m^{-3}]) and the activity concentration of ^{210}Po measured on the mirror surface (S [Bq m^{-2}]) is the so-called K [m^{-1}] factor

$$K = A / S. \tag{1}$$

Some attempts to determine its value have been made in the past (Falk et al., 2001; Walsh and McLaughlin, 2001; Nikezic and Yu, 2006; Martín Sánchez and de la Torre PérezRuano Sánchez, 2014). The study of this factor in several situations and circumstances is one of the goals of this work.

Martín Sánchez et al. (2017) analyzed two cases of ^{210}Pb - ^{210}Po equilibrium on mirror surfaces: short-time exposure in a cave with a very high concentration of ^{222}Rn, and sequential measurements for long time exposure in a moderate-concentration room. Results reached in the second case were only preliminary, and so, exposure of the mirror in that situation has continued.

The implantation of nuclides on mirror surfaces and the corresponding K factor in two new places have also been studied: another cave and, the underground of a museum located in an old and historical building. Both sites present high radon concentrations. Additional variables have also been included in the studies, such as the variation of the implantation of nuclides with the exposure time, repeatability of measurements, and even the effect of cleaning the mirrors.

2. Materials and methods

Conventional 14 × 18 cm^2 mirrors (Fig. 1) were purchased in a normal store and exposed in several places. Measurements of the ^{210}Po surface activity concentration on mirrors were performed by alpha-

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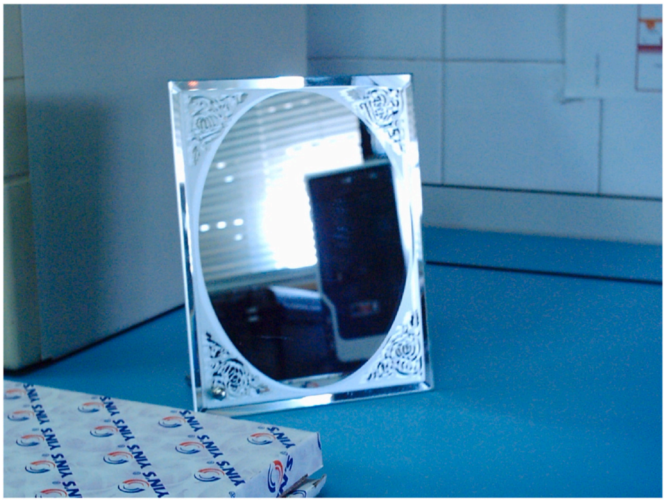

Fig. 1. One of the mirrors used in this work.

particle spectrometry with a silicon detector using the device described elsewhere (Martín Sánchez and de la Torre Pérez, 2012), registering the 5.3 MeV emissions from ^{210}Po (Martín Sánchez et al., 2017).

The mirrors were removed from the sampling places after the exposure and measured in the laboratory, except in the case of a moderate concentration, in which exposure has been continued, measuring from time to time, to study the evolution and the implantation of the nuclides on the surface. In another type of experience, several mirrors were exposed inside the same room and removed simultaneously. Even in another case, the exposure of several mirrors was started simultaneously in the same place, but they were taken out sequentially at different times. In all the studied places, in addition, the indoor radon activity concentration was measured with active or passive (or both) devices. Each situation will be described in the following. The results are quoted with the corresponding uncertainty. Completed details of the uncertainty components considered are shown in Table 1.

2.1. Previous studies

The first mirror (named CC1) was placed in a cave with a mean radon activity concentration of about 24 kBq m^{-3}, which can be considered very high (Martín Sánchez et al., 2013), for a relatively short time (119 d, from 28th June to October 25, 2011). After this exposure period, the mirror was sent to the laboratory to measure the ^{210}Po surface activity concentration and the evolution of its activity.

A second mirror (named B013) is being exposed in room B013 of the Department of Physics of the University of Extremadura since July 11, 2011 (and it continues at the time of writing this work: May 2022). The cumulative ^{210}Po concentration on the surface is also measured from time to time. Room B013 is a very well-characterized place with about 200 Bq m^{-3} constant indoor radon activity concentration level.

Table 1
Standard uncertainty budget for the ^{222}Rn activity concentration and ^{210}Po surface activity concentration.

Source	Assessment type	Standard uncertainty (%)	
		^{210}Po surf. act. conc. (2,5–118 Bq m^{-2})	^{222}Rn act. conc. (200 Bq/m^3)
Counting statistics	A	2,7 - 14	8,4
Background	A	0,2–7,8	3,0
Efficiency	B	0,7	–
Surface/Area	B	0,4	2,0
Geometric factor	A	0,4	–
Calibration factor	B	–	5,3
Combined uncertainty (k = 1)		3–16	10,6

More details of the above experimental sites are given by Martín Sánchez et al. (2014). Concentrations of radon in both locations were measured with the ionization chamber AlphaGuard PQ2000 PRO (Saphymo GmbH) measuring in the continuous mode in 1-h intervals.

2.2. Other places, other studies

Four mirrors (named PC1-4) were placed on 17th March 2017 in the underground of a museum, where the average concentration previously determined was 1.44 kBq m^{-3}. One of the mirrors (PC1) was removed on 17th April 2018, after 396 d of exposure. The other three mirrors (PC2-4) were simultaneously removed on 20th February 2019 (705 d) and measured several times in the following years. The goal of this part of the study was not only the continuation of the previous studies but also the investigation of the repeatability of results and the behaviour of the temporary variation of the radionuclide on the mirror surfaces. In this way, ^{210}Po surface activity is being determined and compared with the results expected by the theoretical predictions as explained below. In addition to these studies, the K factor and the activity variations have also been determined.

Five mirrors (named CM1-5) were placed at the same time (30th May 2019) inside a cave with 4.2 kBq m^{-3} average ^{222}Rn activity concentration. Each mirror was sequentially removed to study the dependence of implantation of ^{210}Pb and ^{210}Po vs radon exposure (kBq h m^{-3}). Mirrors of this experience have in common the exposure starting time and the conditions of the place of exposure. The exposure time was different for each mirror. The concentration of ^{222}Rn was determined using passive track detectors CR-39 for each exposure time.

2.3. Theoretical considerations

Expression for the ingrowing and decay of the activity can be known using the evolution equations (Bateman, 1910). Let A_n, and λ_n be the activity, and the decay constant of the component n, respectively, and N_1 the initial number of nuclei of the progenitor. Then, after the time t,

$$A_n = N_1\left(\prod_{i=1}^{n}\lambda_i\right)\left[\sum_{k=1}^{n}\frac{e^{-\lambda_k t}}{\prod_{j=1(j\neq k)}^{n}\left(\lambda_j-\lambda_k\right)}\right]. \quad (2)$$

This expression was programmed into a calculation sheet taking as variables the activities of ^{210}Pb (A_1), ^{210}Bi (A_2), and ^{210}Po (A_3), for fitting the ^{210}Po activity concentration measured on the mirror surface. A daily constant implantation of ^{210}Pb can be assumed covering all the exposure time. The initial number of nuclides N_1 should be then calculated. Values for the activities A_1, A_2, and A_3 were obtained by using repeatedly Eq. (2). This process was iterated for each (daily assumed) new implantation for the total time of the period. All the contributions for all the days were added. The activity was plotted versus time and compared with the values obtained experimentally. More details of this method are given elsewhere (Martín Sánchez et al., 2017).

2.4. Removing radionuclides by cleaning

An important subject concerning all the study described in this work is the level of fixation of the radionuclides on the surface mirrors. Cleaning the mirrors with conventional cellulose towels is a very usual task in any place. This action was suspected to interact with the nuclides on the mirror surfaces, perhaps removing them.

To study if the radionuclides on mirrors could be removed by cleaning, one-half of each mirror was treated by rubbing the surface with a cellulose towel. Measurements of ^{210}Po were performed on both, clean and not clean surfaces, and the results were compared for each case.

3. Results and discussion

Several experiments with different circumstances considered have been performed as explained in Sect. 2. Table 2 resumes each case.

3.1. Repeatability and ingrowth of the ^{210}Po surface activity concentration

To check the repeatability of the results, the ^{210}Po surface activity concentration was measured on the three mirrors PC2-4 starting with the end of their exposure. Nowadays, 22 determinations have been taken for each mirror. Results were 26.6 ± 4.1, 23.4 ± 1.9, and 25.5 ± 3.1 Bq m^{-2} (for each mirror, respectively), which gives a 25.2 ± 1.6 Bq m^{-2} average (SD/Average (%) = 6.4). If only the six last determinations are considered (corresponding to the time considered to reach ^{210}Pb-^{210}Po equilibrium), the values should be 20.5 ± 1.8, 21.2 ± 0.9, and 21.6 ± 1.6 Bq m^{-2}, respectively, with 21.1 ± 0.6 Bq m^{-2} average (SD/Average (%) = 2.8). These results show the repeatability of the measuring method.

Measurements were performed on the surfaces of the five mirrors CM1-5, removed sequentially from an environment with high radon concentration. More than ten ^{210}Po measurements have been made on each mirror, taking the maximum value as a reference for this part of the study. Results are plotted in Fig. 2. The fit to a straight line shows linear behaviour.

In all the cases described in this paragraph, cleaning the zone measured was applied using a cellulose towel before each measurement. This point is important and will be studied in Sect. 3.3.

3.2. Application of theoretical considerations

All the experimental results have been compared with those calculated by applying Eq. (2) to study the time evolution. An explanation to understand some discrepancies found between the experimental results and the calculated values on the mirror CC1 was proposed in a former work (Martín Sánchez et al., 2017). This mirror was placed in a cave with a mean radon activity concentration of about 24 kBq m^{-3}. The explanation assumes additional deposition of other ^{222}Rn daughters on the mirror surface. In this way, the addition to ^{210}Pb, the plate-out of ^{210}Bi or ^{210}Po, or both, was considered. In this case, the agreement with experimental results was good.

Measurements have been continued on the surface of mirrors CC1 (very high radon concentration) and B013 (moderate radon concentration). Fig. 3 shows the results. The starting time for ^{210}Pb decay in CC1 is considered when the mirror was removed from the cave, ending so its exposure. The coincidence between experimental results and those predicted by the evolution equations is very good in the first 2000 d. However, the last data (after 2000 d) do not follow the predicted decay. These points far away from the expected behaviour will be studied in the following section.

In B013 the activity is increasing due to the mirror being continuously exposed inside the room. The red lines accompanying experimental results (Fig. 3) are the minimum detectable activity concentration (MDC) for each case. Experimental results agree very well with the predictions in this case.

In the two new places studied now (mirrors PC1-4 and CM1-5) the results obtained show similar behaviour. As examples, the results obtained for PC3 and CM3 are plotted in Fig. 4, where the calculated values for the activity evolution are also depicted. Agreement between the first experimental and theoretical values could be good, but in general, the values do not fit correctly, especially in the last points. This effect was already noticed through the comments to the results for CC1, which had the same behaviour. A possible explanation is given in the following section.

3.3. Cleaning mirrors

When the effect of the application of cleaning with a cellulose towel was studied, and both halves of the mirror surface were measured, great differences were observed in all the results. As examples, two cases have been plotted in Fig. 5.

Two facts here must be remarked. In the first place, removing nuclides is very evident. This seems to indicate that, in the exposure, the implantation of nuclides on the surface mirrors is produced not only by recoil as assumed formerly but also for other slighter mechanisms, such as electrostatic attraction. The outer less fixed nuclides seem to be removed when the cleaning is applied. Secondly, the model for the variation of activity can be fitted better for the results without cleaning. This enhances the hypothesis of several mechanisms involved in the process of implantation-deposition of nuclides from the radon progeny onto the mirror surfaces.

The results obtained in the mirrors CC1 (see Fig. 3), PC1-4 and CM1-5 (see Fig. 4) can be now understood. An important decrease in the expected values for the last results was observed in all the cases. Mirrors were not cleaned or altered during the exposure time. The effect of cleaning should explain the differences found between the model and these results.

However, some points must be considered: the mirror B013 is continuously exposed, whereas, in all the other cases, the mirrors were removed after an exposure time. For B013, the implantation of nuclides on the mirror can be enhanced by cleaning the surface before each measurement, probably due to the dust can actuate as a barrier. Systematic cleaning of the mirrors with ended exposure (the other cases) should produce important elimination of nuclides before each measurement.

3.4. K factor

An important part of the study is the knowledge of the K factor. This

Table 2
Summary of the studies performed on the mirrors used in this work.

Places	Mirror name	Previous studies	Repeatability	Implantation vs Rn Exposure	Clean and not clean surfaces	Theoretical model comparison	K FACTOR
B013	B013	OK				OK	OK
Cave CC	CC1	OK				OK	OK
Museum	PC1					OK	OK
	PC2		OK		OK	OK	OK
	PC3		OK		OK	OK	OK
	PC4		OK		OK	OK	OK
Cave CM	CM01			OK	OK	OK	OK
	CM02			OK	OK	OK	OK
	CM03			OK	OK	OK	OK
	CM04			OK	OK	OK	OK
	CM05			OK	OK	OK	OK

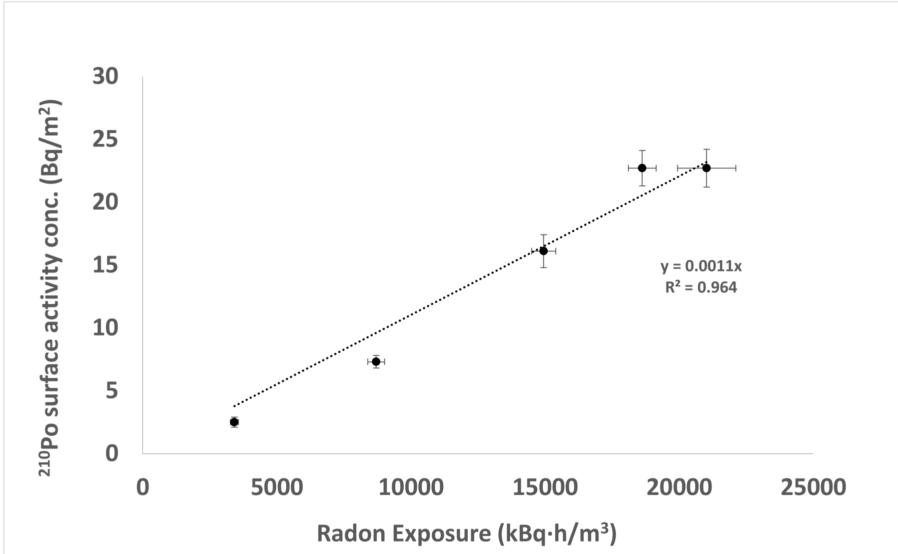

Fig. 2. Activity measured on the surface of mirrors CM1-5 each subjected to increased exposure in an environment with high radon concentration. Uncertainties are quoted with k = 1.

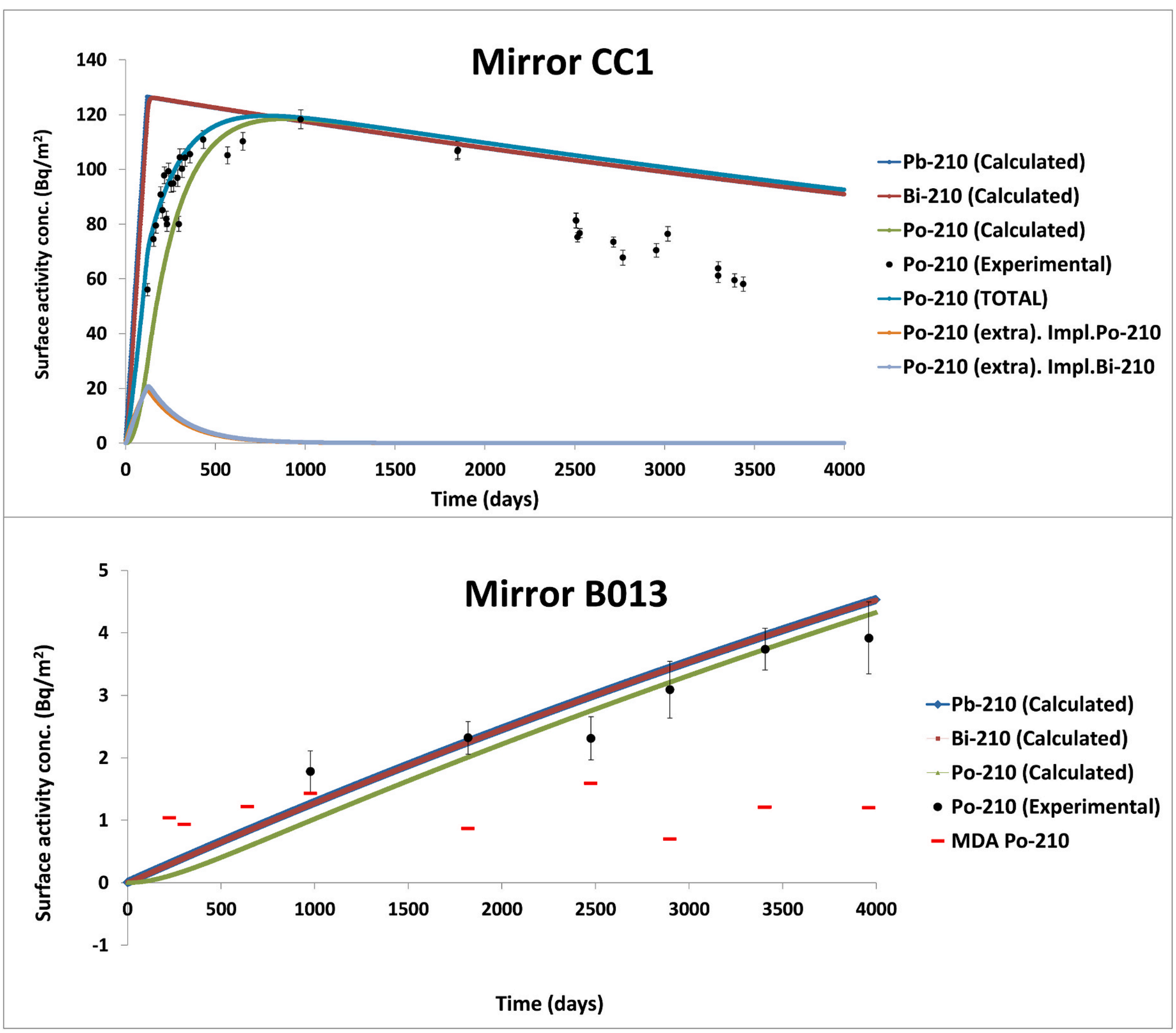

Fig. 3. Results obtained in the measurements of the mirrors CC1 and B013. The lines on the figures are the expected behaviour of the activity for the different components according to the evolution equations and theoretical considerations. Experimental data are plotted with uncertainties k = 1. Horizontal little red lines in B013 are the MDC from each measurement.

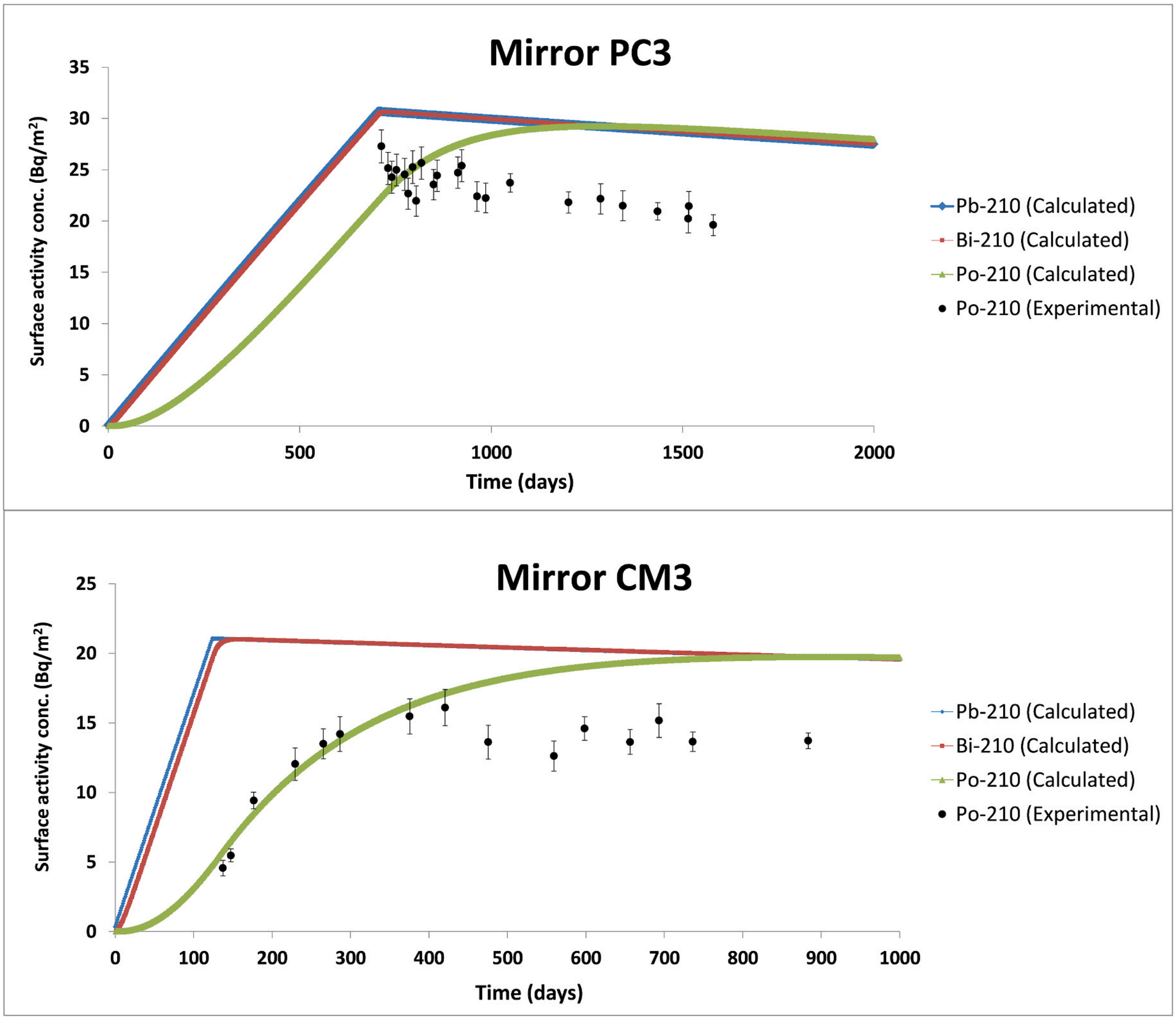

Fig. 4. As examples, plots show the surface activity concentration of ^{210}Po measured on two samples. Uncertainties are given with $k = 1$. The first experimental results in each graph fit correctly to the theoretical values, but the last results are not in agreement.

factor (for the estimation of retrospective indoor radon concentrations) is generally applied for glass objects that are 20 years old, or for objects of different ages but with the surface activity of ^{210}Po normalized to 20 years. Estimations of its value have been considered by taking the values for ^{210}Po surface activity concentrations obtained at the end of each exposure, except for B013 (because its exposure continues), in which the last value measured has been considered. Results for K (normalized to a period of 20 years) jointly with other data are given in Table 3.

Several comments are in order. The most important fact is the variation of the K factor with the concentration of radon. Although a decrease of the ^{210}Po activity in the surfaces seems to be accompanied by an increase in the radon activity concentration, this behaviour is nonlinear (otherwise K should be a constant).

All the values obtained for K are low compared with the factors given by other authors. Walsh and McLaughlin (2001) give 55,5 m^{-1}, and Nikezic and Yu (2006) 58.1 m^{-1}, but their approximations were only estimated through theoretical considerations. Falk et al. (2001), analysing results obtained in dwellings for more than 20 years, estimated an average value of 42 m^{-1} for K, but with very great dispersion, because their results ranged from 9 to more than 100 m^{-1} (even a single value of 1000 m^{-1} was communicated).

4. Summary and conclusions

Indoor retrospective radon activity concentration studies can be a useful tool helping in the knowledge of radon behaviour as an agent producing lung cancers. The techniques presented in this work are adequate. The results show that ^{210}Po can be measured quickly and directly on smooth surfaces such as mirrors.

The study to evaluate the repeatability of the method has been satisfactory. Mirrors exposed to identical conditions (same place and same radon concentration) give compatible results for the ^{210}Po (Bq m^{-2}). In the mirrors sampling the same place, with similar radon concentrations, but different radon exposures (by increasing the exposure time), the implantation of nuclides from the radon progeny in the surface mirrors was directly proportional to that radon exposure.

Behaviour of the ingrowing and decay of activity is well described by the evolution equations, but when compared with the experimental results, they are not always in agreement. Two scenarios must be distinguished: mirrors with permanent radon exposure and mirrors with ended exposure.

The comparative study of the effect of cleaning and not cleaning has only been carried out for mirrors with finished radon exposure. In this case, the results show a decrease in the radon progeny concentration on the mirror surface when cleaning is applied. Therefore, the ^{210}Po results measured on the not-clean surface of the mirror (and the cleaning was never applied) are in better agreement with the calculated values. The opposite behaviour happens in mirror B013, with permanent exposure and cleaning routinely applied before each measurement. In this case, the experimental results also agree with the calculated ones. Cleaning

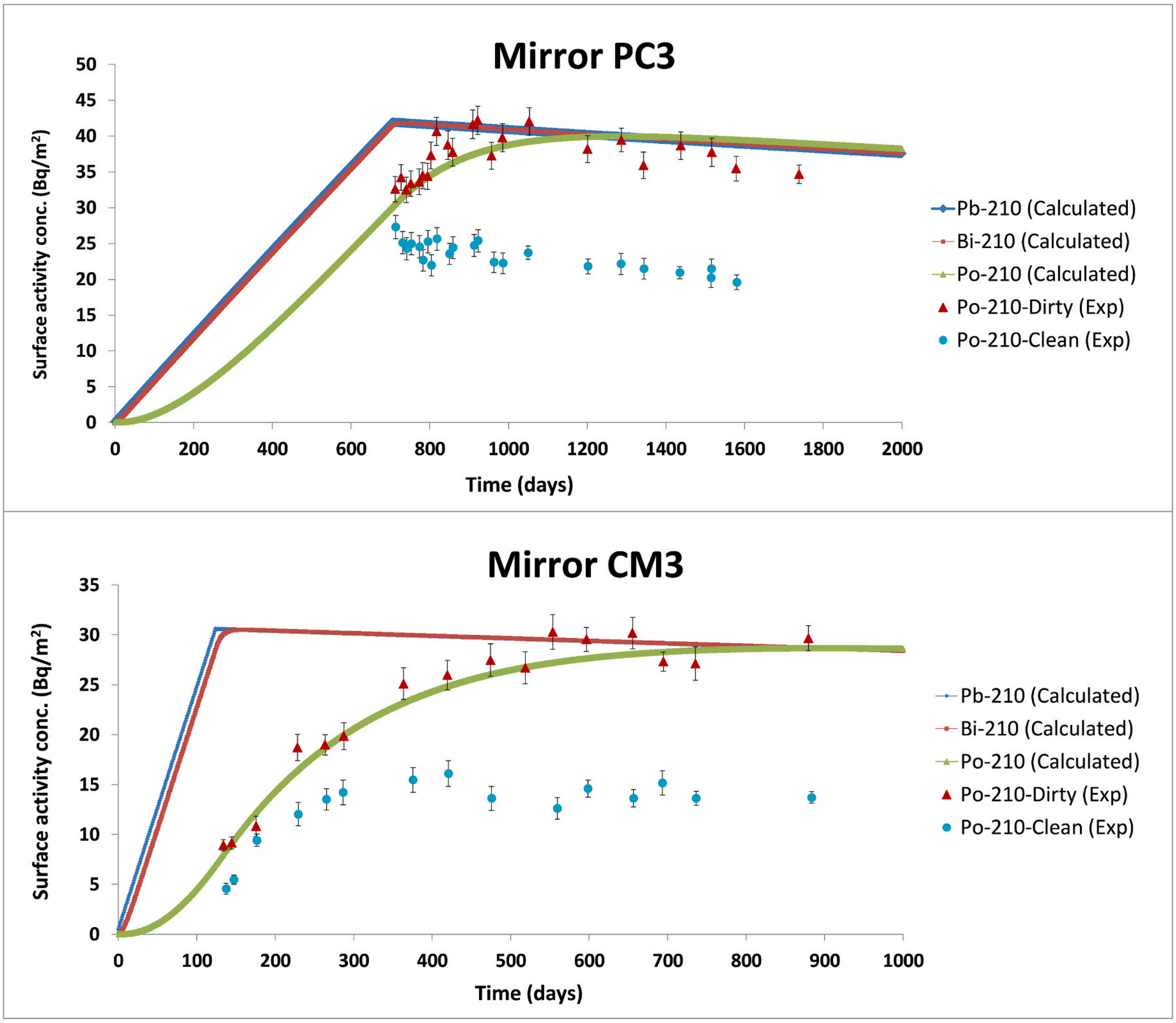

Fig. 5. As examples, plots show the surface activity concentration of ^{210}Po measured on two samples. Uncertainties are given with k = 1. Triangles are the results for the untreated surface half of each mirror, and circles are those obtained in the cleaned half part. Removing nuclides by cleaning is very evident. The expected activity-increasing behaviours, predicted by the evolution equations, are also plotted. Results obtained in the not-clean part of the mirrors agree with the predicted values given by the evolution equations.

Table 3
Global results for the exposure, radon activity concentration in the place, and ^{210}Po surface activity concentration in the mirror. Last column shows the values estimated for the K factor normalized to 20 a.

Places	Mirrors	Starting exposure	Ending exposure	Total exposure days	Total exposure (kBq·h/m^3)	Average ^{222}Rn act. conc. (Bq/m^3)	^{210}Po surf. act. conc. (Bq/m^2) clean part	K factor (m^{-1}) clean part
B013	B013	2011/07/14	2022/05/17[a]	3960	20436	215 ± 22	3,9 ± 0,4	29,8 ± 5,5
Museum	PC1	2017/03/17	2018/04/17	396	13671	1438 ± 144	13,7 ± 0,7	5,7 ± 1,1
	PC2	2017/03/17	2019/02/20	705	24339	1438 ± 144	31,6 ± 1,7	4,4 ± 1,4
	PC3	2017/03/17	2019/02/20	705	24339	1438 ± 144	27,3 ± 1,6	5,1 ± 1,8
	PC4	2017/03/17	2019/02/20	705	24339	1438 ± 144	30,5 ± 1,7	4,5 ± 1,5
Cave CM	CM1	2019/05/30	2019/07/03	34	3423	4194 ± 177	2,5 ± 0,4	7,9 ± 0,5
	CM2	2019/05/30	2019/08/13	75	8706	4782 ± 172	7,3 ± 0,5	6,7 ± 0,4
	CM3	2019/05/30	2019/10/02	125	14951	4923 ± 149	16,1 ± 1,3	5,2 ± 0,5
	CM4	2019/05/30	2019/11/27	181	18626	4376 ± 118	22,7 ± 1,4	4,8 ± 0,5
	CM5	2019/05/30	2020/02/14	260	21025	3754 ± 174	22,7 ± 1,5	5,9 ± 0,9
Cave CC	CC1	2011/06/28	2011/10/25	119	68544	24000 ± 2000	118,3 ± 3,4	3,4 ± 0,3

[a] Date of the last measurement.

the mirror after the exposure and before the measurement seems to improve the implantation of radionuclides in it. The dust on the mirror surface should act as a barrier. This effect is currently being studied by measuring the clean and not-clean surfaces of the B013 mirror. In real situations, all the mirrors are cleaned from time to time. To study correctly retrospective radon concentrations, soft cleaning with a cellulose towel should be recommended before the measurements. Cleaning the mirrors in this way gives results more reliable. In addition, laboratory determinations are closer to the usual behaviour in dwellings and workplaces.

An important question remains with the estimations about the K factor. Only the result for B013 is in the same order of magnitude as that found in the references. The other places studied presented unusually very high indoor radon concentrations. Exposures to a high level were initially necessary to correctly observe the mechanisms of implantation on the mirror surfaces. In addition, the mirrors were exposed for a short time (maximum of about two years), which does not seem adequate for the study of the K factor. We conclude that to conveniently study this factor, only prolonged and continuous exposures to radon should be considered. These studies are very tedious because a very long time is necessary to wait for each measurement in each situation. Many more places, and many more situations, should be studied. One must take into consideration that B013 is the only "normal" place (i.e. with not very high radon concentration) of all those studied in this work. In this room measurements and studies will be continued.

CRediT authorship contribution statement

J. de la Torre Pérez: Investigation. **A. Martín Sánchez:** Investigation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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