This is the peer reviewed version of the following article: Martín Sánchez, A., de la Torre Pérez, J., Ruano Sánchez, A. B., & Naranjo Correa, F. L. (2013). Additional contamination when radon is in excess. *Applied Radiation and Isotopes*, *81*(2013), 212-215, which has been published in final form at https://doi.org/10.1016/j.apradiso.2013.03.004

Additional contamination when radon is in excess

A. Martín Sánchez*, J. de la Torre Pérez, A. B. Ruano Sánchez, F.L. Naranjo Correa

Department of Physics, University of Extremadura, 06006 Badajoz, Spain.

Abstract

A study of the behavior of the ^{222}Rn progeny on clothes, skin and hair has been performed in a place with very high radon concentration. In the past, radon concentration was established to be about 32 kBq/m^3 in a very high humidity environment inside a tourist cave in Extremadura (Spain). The results show that ^{222}Rn daughters are adhered on clothes, skin and hair, adding some radioactive concentration to that due to radon and its progeny existing in the breathable air.

Keywords: radon, radon progeny, external additional radioactive contamination

*Corresponding author. Phone: +34 924 289 526. Fax: +34 924 289 651. E-mail: ams@unex.es

1. Introduction

Radon is a radioactive noble gas that naturally emanates from the Earth's crust, being the main source of natural radiation which we are subjected to (Baeza et al., 2003; UNSCEAR, 2000; Zeeb and Shannoun, 2009). Radon has three natural isotopes, being the most abundant and potentially dangerous ^{222}Rn ("radon" will be used henceforth instead of ^{222}Rn, for the sake of simplicity). This gas is usually found indoor mixed with air. The descendants of radon emit γ rays, and alpha and beta particles, being ^{214}Pb and ^{214}Bi the most important radionuclides in terms of external exposure. Radon descendants formed in the air tend to settle on surfaces, because their daughters act as metal ions (Aceña Barrenechea and Crespo, 1989).

In a survey on radon concentration performed in Extremadura (Spain), an underground cave was found presenting an annual concentration of about 40 kBq/m^3 (Lario et al., 2006; Martín Sánchez et al., 2012). This cave is a karts cavity with aragonite and calcite speleothems, probably isolated from the outside world for millions of years. Discovered by chance in 1967, it was declared natural monument in 1997. The environmental conditions inside the cave are very stable, with temperature always about 17 ºC, and relative humidity near to saturation (about 95 %). To correctly determine possible seasonal variations in radon concentration, several surveys have been performed in this cave (Lario et al., 2006).

Several rooms interconnected by labyrinth galleries form the cave. Due to its characteristics, and to preserve its natural conditions, no artificial actions have been undertaken inside the cave. Some galleries are specially low and narrow, so all people visiting the cave must compulsory wear special clothes (speleologist's gear) composed by a work overall, rubber boots and a helmet (with light). A net covering the hair of the head under the helmet is also necessary for hygienic reasons, in order that one helmet can be used by other people in different visits.

The goal of this work was to study the radiation contamination to workers in this place. In the past, the guide went into the cave for hours in the same day, accompanying several groups of tourists. This means that this and other workers might have surpassed the legally established safety limits from the radiological point of view.

While performing the surveys, the staff members of our laboratory were checked for contamination after each visit to the cave. Radioactive contamination of clothes, skin and hair was observed on the people visiting this cave, and it was decided to perform a systematic study to know the origin and development of this additional radioactive contamination.

2. Experimental methods and sampling

In this study, measurements of radon concentration in air and radioactive contamination in clothes, skin and hair were made with several devices. The measurements of radon concentration in air were performed inside the cave with the continuous monitoring equipment Alphaguard, model PQ 2000 PRO (Saphymo GmbH). This radon detector is based on a design-optimized pulse ionization chamber with an active volume of 0.56 L working with a potential of 750 V. This design allows the determination of radon concentration and its variations "in situ". Furthermore, temperature and humidity are also registered simultaneously. When the equipment is switched on, a very complete internal quality assurance system is activated checking control routines and repeating modes. The equipment is very stable over several years and originally traceable to standards from NIST, NPL, and PTB. Additionally, the device was recently tried in an international intercomparison exercise (Gutiérrez-Villanueva et. al., 2012). Although the uncertainty assigned to a result is estimated by the internal microprocessor of the device taking into consideration several factors, when several measurements were performed in a given place, the uncertainty was estimated as the standard deviation. In normal conditions the minimum detectable activity is typically about 2 Bq/m^3.

To determinate the possible adherence of the descendants of radon in clothes, skin and hair, activity was measured on the worker clothes and directly on parts of their body with a contamination meter MiniTRACEβ. This device is a Geiger-Müller-Pancake with an active counter tube surface of 15.55 cm^2 protected by a grid. A standard ^{60}Co plane source was positioned exactly 3 mm away the grid to calibrate the instrument in the factory. In our laboratory the calibration was checked using several standard sources. The activities on clothes, skin and hair were determined with the detector parallel to the surface being measured, with 1 cm gap. Additional geometry corrections and uncertainties were taken into consideration (Cornejo Díaz et al., 2011). Total uncertainty was estimated considering several components (Table 1).

Table 1

Standard uncertainty budget for the measurements taken with the contamination meter.

Uncertainty component	Relative standard uncertainty contribution (%) (k = 1)	Assessment type
Counting	Standard deviation (5 - 20)	A
Dead time	2	B
Background	20	A
Efficiency calibration	5	A
Geometry	15	A
Homogeneity of samples	5	B
Other	2	B
Combined uncertainty	Variable (26 - 32)	

Other measurements were made on the worker's overalls with a portable alpha spectrometer designed to rapidly identify alpha-emitting radionuclides. The device is formed by an aluminium canister with one of the flat faces open and directed towards the sample to be measured. A silicon detector of the type CAM PIPS from CANBERRA, 450 mm^2 of active area, was placed inside. A triple standard source of ^{233}U, $^{239/240}Pu$, and ^{241}Am, with known activity for each radionuclide, made at CIEMAT, was used to calibrate the spectrometer, and geometrical corrections were considered (Martín Sánchez and de la Torre Pérez, 2012).

The study was carried out after a visit to the cave in June 2011. Specifically for this study, two researchers and the tour guide spent one and half hour inside the cave monitoring for radon concentration. To study the behavior of radon daughters on clothes, one of the researchers (from now on, Man 1) attached a handkerchief on his right arm, under the work overall, and other similar handkerchief over the clothes, directly exposed to the environmental conditions of the cave. To study possible skin contaminations, the guide was wearing a latex glove on one of her hands, maintaining the other hand without glove. A third researcher was helping in the measurements (Man 2). Measurements for radioactive contaminations were carried out on skin, clothes and hair of the two researchers and the guide just before the access to the cave and right after the exit.

3. Results and Discussion

Indoor radon concentration on the cave has been measured several times, observing seasonal variations, as already noticed by Lario et al. (2006). Radon concentration detected the day of sampling was 27 ± 2 kBq/m^3, being the minimum value measured inside the cave, which has a yearly average of 40 kBq/m^3 (Martín Sánchez et al., 2012). The temperature was 18 ºC and the relative humidity was 90%.

The first type of measurement was performed on the handkerchiefs. Before the access to the cave, the activity level on both handkerchiefs was 5 ± 2 Bq. After the visit, the measurement on the internal handkerchief gave 30 ± 9 Bq, while 400 ± 106 Bq were registered on the external handkerchief. These results show that some radioactive contaminants were mainly adhered externally to the clothes.

To study the subsequent behavior of these radioactive emitting nuclei, a continuous measurement on the external handkerchief was performed. The activity was decreasing over the time with the behaviour shown in Figure 1. The half-life of the decay was calculated from the value of the coefficient obtained from a fit of the logarithmic values of the activity to a straight line, obtaining 34 ± 4 min. These results show that the radioactive particles adhered on clothes should come mostly from ^{214}Pb, a radon descendant with 27 min half-life. The presence of other radon daughters with lower half-life such as ^{218}Po ($T_{1/2}$ = 3 min) and ^{214}Bi ($T_{1/2}$ =20 min) should be not discarded.

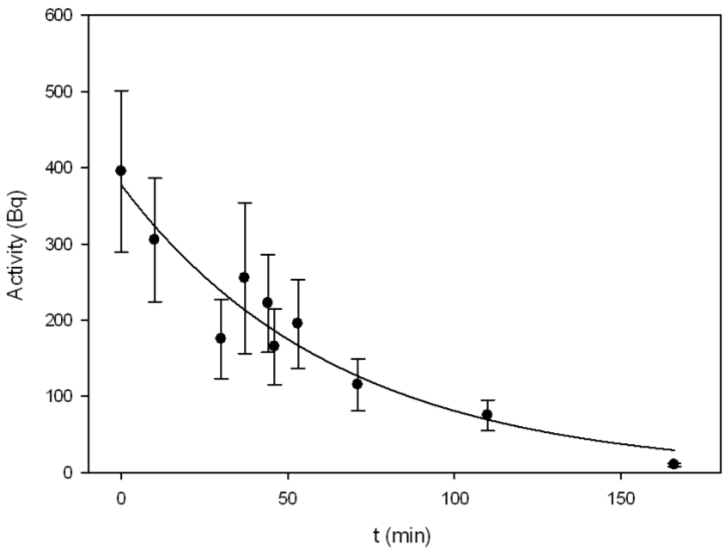

Figure 1. Activity measured on the external handkerchief. The decay of the activity is evident. Solid line is the fit of experimental results to an exponential function.

The same type of measurement was performed on different parts of the researchers work overalls. The results showed a great variability, indicating that the particles did not adhere homogeneously throughout the suit, but depending on the sweat, moisture and body area. The average radioactivity levels found on the work overalls were about 300 ± 90 Bq after the visit, showing a noticeable radioactive contamination on clothes induced by adherence of radon daughters.

In order to try to determine more precisely the radioactive elements adhered on the overalls, a measurement was performed with the portable alpha spectrometer. Due to the thickness, roughness and inhomogeneity of the sample, the alpha-particle spectrometric analysis was difficult because self-absorption or energy attenuation effects are normally present. For this type of samples, the measured alpha-particle peaks in general degenerate to shapes with low slopes on the low-energy side of the spectrum, broadening significantly the peak shape (Aceña Barrenechea et al., 1977; Aceña Barrenechea and Tormo Ferrero, 1977; Martín Sánchez and de la Torre Pérez, 2012).

Figure 2 shows the spectrum obtained measuring for 1874 s immediately after the exit of the cave. A single broad peak with energy about 7770 ± 170 keV was clearly observed, which was assigned to the alpha-particle emission from the radon daughter ^{214}Po (7687 keV).

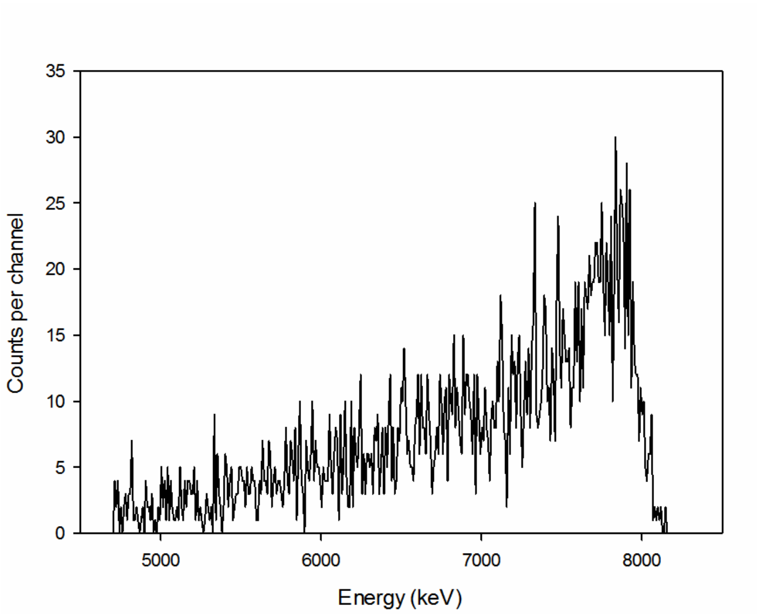

Figure 2. Alpha-particle spectrum obtained from the work overall measured with the portable spectrometer.

Other measurements were performed on the skin of the hands of the three people exposed to radon before the access to the cave, right after the exit, after washing with plain water, and after a second washing with abundant water and soap. As the guide wore a latex glove on one of her hands by the sampling time, leaving the other hand without glove, additional measurements were performed on her hands. Table 2 shows the results of all these determinations. From these results one can conclude that radioactive contaminants were clearly deposited on the skin, but when the glove was used the effect was noticeably reduced. It seems then advisable to wear gloves in these conditions for skin protection. Washing with a great quantity of water and soap after an external radioactive contamination continues being a practice highly recommended.

Table 2

Activity (Bq) directly measured with the contamination meter on the skin of the hands of the people monitoring inside the cave. In the fifth column the results obtained measuring directly the protective glove have been added for comparison.

Measurements	Man 1	Man 2	Guide		
			Hand without glove	Hand with glove	On the glove
Before the visit	5 ± 2	4 ± 2	5 ± 2	5 ± 2	5 ± 2
After the visit	215 ± 65	370 ± 111	500 ± 133	50 ± 15	360 ± 108
After washing with water	140 ± 42	130 ± 40	200 ± 60	---	---
After washing with soap and water	40 ± 12	90 ± 28	85 ± 26	---	---

One of the researchers (Man 1) who entered the cave has a beard, and the activity on it was measured after the exit, giving about 650 ± 172 Bq. This value was higher than the value measured on his hands or on his clothes (215 and 300 Bq, respectively). This result can be easily explained due to the effect of electrostatic attraction between the radioactive contaminants and the hairs of the beard. Therefore, entering the cave wearing a beard is not recommended.

Measurements were also taken on the hair of the head of Man 1, showing not significant growth of the radioactive content after the visit. The principal reason of this must be the use of the protective net over his head, avoiding direct contact of the hair with the external air. Radioactive contaminants did not adhere to this covered area of the body, showing that they are really deposited on the exposed surfaces mainly by electrostatic attraction.

4. Summary and conclusions

An important conclusion of this work is that in environments with very high radon concentrations, this gas presents additional problems to those related to breathable air. This study shows the importance of taking into account other effects of radon daughters on the skin of people when the radon concentration is very high. The measurements made on the handkerchief and overalls show that radon daughters are attached by electrostatic attraction to clothing. This same effect occurs on the skin. For this reason, the use of proper clothing, helmet or hat and gloves is highly recommended when the radon concentrations are very high.

Specifically, the main action to be taken is to avoid any type of contact of any part of the body with the air, in the same way that especial clothing is used when some danger due to high natural or man-made radioactive contamination are present. In case of body surface contamination, a quick shower with abundant water and soap after being exposed is the commonly followed method for promptly decontamination.

Acknowledgements

Financial support from the Spanish Consejo de Seguridad Nuclear and from the Government of Extremadura (Ref. GR10160) is acknowledged. Thanks are also due to the Government of Extremadura for facilitating the monitoring in the cave. Special thanks are given to the guide for her participation and collaboration in the study.

References

Aceña Barrenechea, M.L., Crespo, M.T., 1989. Medida de los descendientes del radón en aire por espectrometría alfa. CIEMAT, Madrid.

Aceña Barrenechea, M.L., García-Toraño, E., Rivero Núñez, M.C., 1977. Espectrometría alfa de fuentes radiactivas gruesas I. Aplicación a la determinación de emisores alfa. Report JEN 384, CIEMAT, Madrid.

Aceña Barrenechea, M.L., Tormo Ferrero, M.J., 1977. Espectrometría alfa de fuentes radiactivas gruesas II. Posibilidades de aplicación al estudio de equilibrios radiactivos en minerales uraníferos. Report JEN 393, CIEMAT, Madrid.

Baeza, A., Navarro, E., Roldán, C., Ferrero, J.L., Juanes, D., Corbacho, J.A., Guillén, F.J., 2003. Indoor radon levels in buildings in the Autonomous Community of Extremadura (Spain). Radiat. Prot. Dosim. 103, 263-268.

Delacroix, D., Guerre, J.P., Leblanc, P., Hickman, C., 2002. Radionuclide and Radiation Protection Data Handbook. Radiat. Prot. Dosim. 98 (1), 2002.

Eckerman F.K., Ryman J.C., 1993. External exposure to radionuclides in air, water and soil. EPA-402-R-93-081. Environmental Protection Agency, Washington.

EURATOM, 1980. Council Directive 80/836, of 15 July 1980 amending the Directives laying down the basic safety standard for the health protection of the general public and workers against the dangers of ionizing radiation. O.J. L. 246, 1-72.

Gutiérrez-Villanueva, J.L., Sainz Fernández, C., Fuente Merino, I., Quindós López, L., Quindós López, J., Fernández Villar, A., Casal Ordas, S.E., López Abascal, D., Arteche Laso, D., Fernández López, E., Quindós Poncela, L.S., 2012.

International Intercomparison Exercise on Natural Radiation Measurements under Field Conditions. University of Cantabria, Santander.

ICRP, 1991. 1990 Recommendations of the International Commission on Radiological Protection, ICRP Publication 60. Ann. ICRP 21 (1-3).

Lario, J., Sánchez-Moral, S., Cuezva, S., Taborda, M., Soler, V., 2006. High ^{222}Rn levels in a show cave (Castañar de Ibor, Spain): Proposal and application of management measure to minimize the effects on guides and visitors. Atmos. Environ. 40, 7395-7400.

Martín Sánchez, A., de la Torre Pérez, J., Ruano Sánchez, A.B., Naranjo Correa, F.L., 2012. Radon in workplaces in Extremadura (Spain). J. Environ. Radioact. 107, 86-91.

Martín Sánchez, A., de la Torre Pérez, J., 2012. Portable alpha spectrometer. Appl. Radiat. Isot. 70, 2267-2269.

UNSCEAR, 2000. United Nations Scientific Committee on the Effects of Atomic Radiations. Sources and effects of ionizing radiations. Report to the General Assembly, with Scientific Annexes. Vol. I: Sources. United Nations, New York.

Zeeb, H., Shannoun, F., 2009. WHO handbook on indoor radon: public perspective. World Health Organization, Geneva.