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. (2012). Radon in workplaces in Extremadura (Spain). *Journal of Environmental Radioactivity, 107,* 86-91, which has been published in final form at https://doi.org/10.1016/j.jenvrad.2012.01.009

Radon in workplaces in Extremadura (Spain)

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

Indoor radon measurements are usually associated with housing. However, a typical person spends about one-third of the day at their workplace. A survey was made of radon levels in workplaces in Extremadura (Spain). More than 200 measurements were performed in some 130 firms and organizations of different sectors (urban wellness centres, spas, caves, mines, water management facilities, underground carparks, wine cellars, museums, etc.). Activated charcoal canisters and track detectors were used for sampling. The results indicated the importance of performing this type of measurement because the exposure of workers can reach high values in some cases.

Keywords: Radon, exposure, workers.

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

1. Introduction

Human beings are constantly exposed to natural radiation sources. Radon (we shall use "radon" to mean ^{222}Rn for the sake of simplicity) is the main source of natural radiation, and the second leading cause of lung cancer after smoking (Zeeb and Shannoun, 2009). Indoor radon measurements are generally associated with dwellings. However, a typical person spends more than eight hours a day in their workplace, so that it is recognized as essential to monitor workers' exposure to radon to control their health risks (ICRP, 1993). There have been some surveys measuring indoor radon in dwellings and public buildings such as schools and offices (Llerena et al., 2010; Rafique et al., 2010), in workplaces (Clouvas et al., 2007; Papachristodoulou et al., 2010; Thinova et al., 2011; Vaupotič, 2008) and, for our region of Extremadura in particular, some studies of radon concentrations in buildings carried out by other workers (Baeza et al., 2003).

In Spain, radon concentration measurements are compulsory in workplaces according to the provisions of a recently issued regulation establishing recommendations and limits related to natural sources of radiation. This regulation requires the managers of professional activities involving such sources to carry out studies to determine whether there is any significant increase in the exposure of workers or of members of the public, and which may be considered as non-negligible from the point of view of radiological protection. The Spanish Nuclear Security Council has defined which places may be at risk of special exposure to radiation and to radon levels above the limits considered as safe (Sanz Alduán and Ramos Salvador, 2008). In particular, remedial action must be taken in places where the ^{222}Rn annual average activity concentration is above 400 Bq/m^3. In the case of workplaces frequented by the public, the limit level is 200 Bq/m^3 for recently constructed buildings, and 400 Bq/m^3 for previously existing buildings.

We have carried out a survey measuring radon in workplaces in the region of Extremadura (Spain) as part of a R&D project in nuclear safety and radiation protection. This paper presents the characteristics of the monitoring procedure and the results, showing the importance of the need to measure radon concentrations in workplaces.

2. Sampling and Methods

2.1. Detectors

Measurements were carried out using two types of probe: activated charcoal canisters (short-term exposure) and CR-39 nuclear track detectors (long-term exposure). The activated charcoal canisters allowed fast measurement (two-days exposure), which is used to quickly identify places with a significant concentration of radon, while the nuclear track detectors (three-month exposure) provided estimates for the annual average concentrations which are the values considered by the legislation.

The measurement method used with activated charcoal canisters was the EPA 520/5-87-005 standard procedure (Gray and Windham, 1987). Canisters were opened *in situ* and exposed for 48 h. This method has as its main advantage the use of very inexpensive devices, giving results quickly, and hence is especially useful for the location of “hot” points. The detectors are re-usable after heating for 24 hours at 120 °C. The standard for calibration was made by homogeneously spiking a blank canister with an aliquot of a ^{226}Ra solution with certified activity, closing the canister hermetically, and waiting for more than one month for secular equilibrium. Standard, collected samples, and blanks were measured by putting each one in the upper part of an inverted 3×3 NaI(Tl) detector integrated in 10 cm thick Pb shielding, inner-lined with Cu and Cd shells, and connected to the corresponding electronics. Once the spectrum had been recorded, the region of interest was chosen as that with energy ranging between 270 and 720 keV, corresponding to the emissions of the ^{222}Rn daughters, ^{214}Pb and ^{214}Bi. The measuring time for each canister in the NaI (Tl) detector was 20 minutes.

Nuclear track detectors were exposed for a period of 3 months in the workplaces. After this time, they were closed, stored, and sent to the University of Cantabria (Spain) for measurement. This laboratory has specialized in all types of radon measurements for many years, and is certified through international standards. The procedure used there was etching and direct reading of each CR-39 detector on a Radosys microscopy system. These detectors were therefore not re-usable.

2.2. Workplace selection

Spanish laws classify as places of special exposure concern (Sanz Aldúan and Ramos Salvador, 2008) spas and urban wellness centres, caves and galleries, mines (other than uranium mines, which have their specific regulation), installations where groundwaters are stored or treated, and underground or above ground workplaces where high levels of radon should be suspected. A first search for candidate workplaces with some of the above characteristics was made in the Region of Extremadura. In addition, some other measurements were carried out in places not specifically categorized as of special exposure, the aim being to characterize levels in non-systematically chosen places in such a way that the data were geographically representative of the entire region and all of its principal towns were covered.

About 130 companies were monitored. These belonged to various economic and cultural sectors in Extremadura (the sites described above, and other places such as winery cellars, underground carparks, museums, etc.). Figure 1 is a pie chart of the distribution of the type of place in the survey. The 31% of the measurements corresponding to places not classified in the above groups were made in shops, offices, schools, hotels, farmhouses, theatres, building-material firms, monasteries, cathedrals, etc. About 28% of all the determinations were made in museums, in view of the great number of these in Extremadura. However, caves, tunnels, and mines represented only 3% of the measurements, since there are only a few facilities of this type in the region.

Figure 1. Pie chart showing the type of workplace considered in the radon monitoring survey of Extremadura. About 130 companies and organizations were checked for indoor radon concentration

Both types of monitor (canisters and track detectors) were exposed in all the places except in some cases where a first determination showed very low radon concentrations, clearly indicating that long-time exposure was unnecessary. In large installations, or when special exposure was suspected, several rooms of the same building (or different buildings of the same company) were included in the monitoring. The locations selected for exposure of the detectors were chosen according to criteria of maximum time of exposure of workers, or of maximum risk (underground, rock flooring, old historical buildings, etc.).

3. Results and Discussion

More than 200 determinations were performed (in some companies several rooms were analysed). Only one canister was lost. However, only 67% of the track detectors were collected in good condition. The problem appeared to be the long time that this type of detector had to be left at the measuring site. An added difficulty is that the company can not detain its normal activities during the measurement period. In particular, cleaning, painting, improvement works, and other similar actions were performed in many workplaces during the time of exposure, with the consequent loss of some detectors.

3.1. Comparison of methods

Figure 2 shows a comparison of the results obtained by the two methods. However, the results obtained by the two types of detector could well be technically quite different. The canisters provide the radon concentration measured over a short time period (useful for locating cases of “hot spots”), allowing rapid estimation of the radon concentration at a point. Track detectors allow an estimate to be made of the annual average radon concentration since they are exposed for long periods, and seasonal corrections are applied (Bochicchio et al., 2005; Cortina et al., 2008; Kullab et al., 2001; Papaefthymiou et al., 2003; Singh et al., 2005). Although these corrections may under- or over-estimate the annual average concentration (Font, 2009; Moreno et. al., 2009), implementation of a great quantity of measurements is normally expensive and always very time consuming, so that only in the cases in which the average activity concentration was greater than 200 Bq/m^3 were seasonal studies made.

Figure 2. Comparison of the results obtained by measuring with canisters and with track detectors. Values greater than 1000 Bq/m^3 are not shown, for the sake of clarity.

A first perception is that the two methods give values in general of the same order of magnitude. However, the results obtained with canisters are normally lower than those obtained with track detectors. Two explanations can be given for this:

- The exposure time of the canisters was only a few hours; in this period, for the site's routine activity, the doors of the building are normally open. They are closed at night, so that the measured radon concentration is probably lower than for longer periods in which days and nights, and weekends and holidays are included (Kávási et. al., 2006),

- The results obtained with track detectors are annual averages and include seasonal variation corrections, whereas the final values for the canisters correspond only to the period of measurement; this effect has been studied by Miles (2001).

Only the track detector data (in the form of annual average radon concentrations) were taken into account for the results re- ported in this communication. Therefore, in the following, only these data will be considered in the discussion and conclusions.

3.2. Results by sector

Figure 3 shows the statistics of the track detector results in the survey performed over 15 months, during 2009 and 2010. The measured values varied over a wide range, the maximum value was about 40 kBq/m^3, which was measured in a touristic cave. Excluding this value, and taking into consideration the recommendations given by Font (2009), the results were binned into a histogram which was fitted to a log-normal distribution, obtaining an arithmetic mean of 229 Bq/m^3, a geometric mean of 130 Bq/m^3, geometric standard deviation of 3, for a range of 28 - 4337 Bq/m^3. Overall, 69% of the workplaces presented radon concentrations lower than 200 Bq/m^3, 18 % had radon concentrations between 200 and 400 Bq/m^3, and the other 13 % had values greater than 400 Bq/m^3.

The linked image cannot be displayed. The file may have been moved, renamed, or deleted. Verify that the link points to the correct file and location.

Figure 3. Distribution of the number of workplaces measured at different companies. One outlier value of 40 kBq/m^3 has been ignored in the plot and in the calculations.

Figure 4 shows the track detector results classified by sector. One observes in the figure that the distributions differed widely between sectors.

Figure 4. Distribution by sectors of the survey results obtained using track detectors.

Spas and urban wellness centres were considered together in the study because they are in the same category in the legislative norms, but (unsurprisingly) they present very different distributions of radon concentrations. While wellness centres use the normal urban water supply for their hydrotherapy and other health care treatments, spas are specifically located at the sites of mineral springs. In the urban wellness centres, all the results were lower than 200 Bq/m^3, with values ranging from 41 to 124 Bq/m^3, the mean value being 83 Bq/m^3. In the spas, 60% of the values were greater than 200 Bq/m^3, with 20% being above 400 Bq/m^3; the minimum value was 48 Bq/m^3 and the mean was 301 Bq/m^3. The greatest values were measured in two spas (905 and 696 Bq/m^3). Radiological protection actions are thus essential if workers spend much time in places which have such high concentrations.

The greatest values in the survey (about 40 kBq/m^3) were recorded in a cave in the north of Extremadura. This cave is a karst cavity with aragonite and calcite speleothems, probably isolated from the outside for millions of years. It was discovered by chance in 1967, and declared a natural monument in 1997. The main problem here is related to the employee acting as a guide, because in the past she had been spending up to 8 h a day in the cave, surpassing the maximum permissible exposure. Now, the time she spends in the cave has been limited to a few visits for scientific studies from time to time, taking care not to exceed the annual dose limit. Another cave of interest for tourism gave 1450 Bq/m^3. Subsequent measurements taken at the same place showed high variability in the results (between 1450 and 244 Bq/m^3), and this site is now being monitored to study why there are such marked temporal variations in the radon concentration.

The second-ranked place was a museum, located in an old historical building, with 4337 Bq/m^3 radon concentration. This value was obtained for a semi-underground room used as an exhibition room (it had probably been an ancient dungeon), whereas the level measured in the second floor library of this museum, where its employees are normally working, was below 200 Bq/m^3. Another museum also located in another old building in the same small town gave 1182 Bq/m^3, and other measurements in different workplaces in the same town also had high radon concentrations, showing that these high values are associated with characteristics of the soil and geology. Another museum in a different small town, but also located in an old historical building, had values of about 570 Bq/m^3, measured in the library. In this last case, improvement works are under way, and extraction conduits will be installed in order to diminish future radon concentrations.

Winery cellars and their associated facilities did not present any problems with their indoor radon concentrations. Eighteen wineries were measured, obtaining radon concentrations between 28 and 136 Bq/m^3, with an average of 68 Bq/m^3. In the case of the cellars (underground), the problem presented by the build-up of carbon dioxide minimizes that of radon, because these facilities are always adequately ventilated. Moreover, the workers spend little time inside the cellars, whose essential purpose is for the wines to repose. Other winery facilities consisted of open-air or ventilated ground floor installations in which the wine is fermented in large tanks. In all cases (whether underground or not), the concentrations were less than than 200 Bq/m^3.

Some other sectors, such as carparks, presented no problems of high radon concentrations in spite of being underground places. This is because they are very well ventilated due to the problem of carbon monoxide. Also, in this particular case, employees usually spend all their working hours inside a separated cubicle.

3. 3. Geographical characteristics

As radon emanates mainly from the ground, underground places would naturally be considered as prone to high radon concentrations (Moreno et al., 2009). However, the results of this survey showed no such differences between surface or underground sites (see Figure 5).

Figure 5. The distribution of the results of this survey showed no differences in the measured concentrations between surface and underground workplaces.

One of the most outstanding characteristics of the results of the survey was the strong dependence found of high concentrations of radon on the geographical situation of the site. This seems to indicate that one of the main causes of high radon concentrations is the characteristics of the ground on which the buildings have been constructed. Figure 6 shows the workplace radon concentration results plotted on two different maps of Extremadura: the environmental gamma radiation map (a), and the geological map (b).

Figure 6. Results for the indoor radon concentrations superimposed on the MARNA natural radiation map of Extremadura (a), and on the geological map (b).

The gamma radiation data were taken from the MARNA (or Map of Natural Radiation) Project (Quindós et al., 2004; Suárez Mahou et al., 2000), and the geological plot was adapted from that of Baeza et al. (2003). In general, the highest radon concentrations measured indoors in workplaces are directly related to those areas presenting the highest values of natural gamma radiation. This effect has been studied by Papachristodoulou et al. (2010). There is also a direct relationship between the indoor radon concentrations and the type of soil, as shown in Fig. 6(b). For this comparison, the region's soils were roughly classified into three general types: igneous rocks, detrital and alluvial materials, and metamorphic rocks and quartzite. The igneous rocks in the map include granitic areas, principally located in the northeast and centre of the region, where the radon concentrations were generally the highest. In order to emphasize these effects, the results for the measured radon concentration sites were classified according the type of soil, as plotted in Fig. 7.

Figure 7. Distribution of radon concentration plotted versus the different types of soil.

In sum therefore, the greatest exposure to natural gamma radiation and indoor radon concentrations seems to correspond to sites where buildings stand on granitic substrates (igneous rocks). The influence of the type of soil on environmental radioactive concentrations in this same region has been observed in a study of the radon concentrations in groundwaters in the same region, with the highest concentrations also being found in the groundwaters of granitic zones (Galán López and Martín Sánchez, 2008).

4. Summary and Conclusions

A survey was carried out to determine indoor radon concentrations in workplaces, with more than 200 measurements corresponding to about 130 companies in Extremadura (Spain). Two types of detector were used: canisters and track detectors. The canisters were used for the fast identification of “hot points”, because the exposure time needed was only two days. The track detectors allowed us to determine the average annual concentra- tion for each site, because the exposure time was three months. This long period caused some problems in that some detectors were lost.

The results show the importance of this type of study, because a far from negligible proportion of the workplaces exceeded the limits established as safe. In particular, about 34% of the monitored companies presented values which may be considered as indicative of the need to carry out a thorough check of the radon concentra- tion inside their installations, in order to diminish the radon exposure of the workers.

The high levels measured in one cave that is important in local tourism exceeded the values considered as safe by several orders of magnitude, indicating that other more precise and continuous measurements must be made. In this case, remedial actions, such as limiting the time workers spend inside the cave, are essential.

The values obtained in museums, spas, and hotels also call for special attention, particularly when their activities are carried out in old buildings. Although museums were at first considered places without any special risk, the results showed that, in general, their closed-in nature, usually needed to preserve artworks (paintings, sculpture, jewellery, etc.), leads to increased indoor radon concentrations.

No great differences in radon concentrations were found according to whether the place was situated at ground level or underground. For example, in carparks and winery cellars, which are usually underground workplaces, the levels of radon were not especially high. The existence of other noxious gases such as carbon dioxide or carbon monoxide present a major problem in these facilities, so that adequate ventilation and other mitigative actions are normally applied, thus decreasing the possible radon levels at the site.

It was also observed that the type of soil at a given site is a particularly important characteristic to be taken into consider- ation. In general, granitic zones presented the highest values of indoor radon concentration, so that for workplaces in these zones this relationship must be taken into account in the estimation of the dose received by workers (or by the general public).

It is important to note also that high levels were found at some sites chosen arbitrarily and initially not considered to be of any particular risk, such as shops, theatres, schools, offices, cathedrals, building-material firms, etc., (16% of these workplaces presented levels above 400 Bq/m^3). This indicates that surveys to measure and monitor indoor radon concentrations are advisable in all types of workplaces.

Acknowledgements

Thanks are due to the Spanish Consejo de Seguridad Nuclear (CSN) for financial support, and to the members of the University of Cantabria for collaborating with their readings of the track detectors. Special acknowledgement is due to the managers of the companies who collaborated in the study, for facilitating the conditions for the measurements.

References

Baeza, A., Navarro, E., Roldán, C., Ferrero, J.L., Juanes, D., Corbacho, J.A., Guillen, F.J., 2003. Indoor radon levels in buildings in the autonomous community of Extremadura (Spain). Rad. Prot. Dosim. 3, 263-268.

Bochicchio, F., Campos-Venuti, G., Piermattei, S., Nuccetelli, C., Risica, S., Tommasino, L., Torri, G., Magnoni, M., Agnesod, G., Sgorbati, G., Bonomi, M., Minach, L., Trotti, F., Malisan, M.R., Maggiolo, S., Gaidolfi, L., Giannardi, C., Rongoni, A., Lombardi, M., Cherubini, G., D'Ostilio, S., Cristofaro, C., Pugliese, M., Martucci, V., Crispino, A., Cuzzocrea, P., Sansone Santamaria, A., Cappai, M., 2005. Annual average and seasonal variations of residential radon concentration for all the Italian Regions. Rad. Meas. 40, 686-694.

Cortina, D., Durán, I., Llerena, J.J., 2008. Measurements of indoor radon concentrations in the Santiago de Compostela area. J. Environ. Radioact. 99, 1583-1588.

Clouvas, A., Xanthos, S., Antonopoulos-Domis, M., 2007. Pilot study of indoor radon in Greek workplaces. Rad. Prot. Dosim. 124, 68-74.

Font, Ll., 2009. On radon surveys: Design and data representation. Rad. Meas. 44, 964-968.Gray, D.J., Windham, S.T., 1987. EERF standard operating procedures for Radon-222 measurement using charcoal canisters. EPA 520/5-87-005. Environmental Protection Agency, Alabama.

Galán López, M., Martín Sánchez, A., 2008. Present status of ^{222}Rn in groundwater in Extremadura. J. Environ. Radioact. 99, 1539-43.

ICRP, 1993. Protection against Radon-222 at home and at work. ICRP Publication 65. Ann. ICRP 23 (2).

Kávási, N., Kovács, T., Németh, C., Szabó, T., Gorjánácz, Z., Várhegyi, A., Hakl, J., Somlai, J., 2006. Difficulties in radon measurements at workplaces. Rad. Meas. 41, 229-234.

Kullab, M.K., Al-Bataina, B.A., Ismail, A.M., Abumurad, K.M., 2001. Seasonal variation of radon-222 concentrations in specific locations in Jordan. Rad. Meas. 34, 361-364.

Llerena, J.J., Cortina, D., Durán, I., Sorribas, R., 2010. ^{222}Rn concentration in public secondary schools in Galicia (Spain). J. Environ. Radioact. 101, 931-936.

Miles, J., 2001.Temporal variation of radon levels in houses and implications for radon measurement strategies. Rad. Prot. Dosim. 93, 369-375.

Moreno, V., Bach, J., Baixeras, C., Font, Ll., 2009. Characterization of blowholes as radon and thoron sources in the volcanic region of La Garrotxa, Spain. Rad. Meas. 44, 929-933.

Papachristodoulou, C.A., Patiris, D.L., Ioannides, K.G., 2010. Exposure to indoor radon and natural gamma radiation in public workplaces in north-western Greece. Rad. Meas. 45, 865-871.

Papaefthymiou, H., Mavroudis, A., Kritidis, P., 2003. Indoor radon levels and influencing factors in houses of Patras, Greece. J. Environ. Radioact. 66, 247-260.

Quindós, L.S., Fernández, P.L., Gómez, J., Sainz, C., Fernández, J.A., Suarez, E., Matarranz, J.L., Cascón, M.C., 2004. Natural gamma radiation map (MARNA) and indoor radon levels in Spain. Environment International 29, 1091-1096.

Rafique, M., Rahman, S.U., Rahman, S., Matiullah, M., Shahzad, I., Ahmed, N., Iqbal, J., Ahmed B., Ahmed, T., Akhtar, N., 2010. Assessment of indoor radon doses received by the students in the Azad Kashmir schools, Pakistan. Radiat. Prot. Dosim. 142, 339-346.

Sanz Alduán, M.T., Ramos Salvador, L.M., 2008. CSN Criteria for radiological protection against exposure to natural radiation. Alfa 3, 46-53.

Singh, S., Mehra, R., Singh, K., 2005. Seasonal variation of indoor radon in dwellings of Malwa region, Punjab. Atmospheric Environ. 39, 7761-7767.

Suárez Mahou, E., Fernández Amigot, J.A., Baeza Espasa, A., Moro Benito, M.C., García Pomar, D., Moreno del Pozo, J., Lanaja del Busto, J.M., 2000. Proyecto Marna. Mapa de radiación gamma natural. Consejo de Seguridad Nuclear, Madrid.

Thinova, L., Rovenska, K., 2011. Radon dose calculation methodology for underground workers in the Czech Republic. Rad. Prot. Dosim. 145, 233-237.

Vaupotič, J., 2008. Comparison of various methods of estimation radon dose at underground workplaces in wineries. Rad. Environ. Biophys. 47, 527-534.

Zeeb, H., Shannoun, F., eds., 2009. *WHO handbook on indoor radon: public perspectives.* World Health Organization.