222Rn concentration in public secondary schools in Galicia (Spain)

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A B S T R A C T

In the framework of a 222Rn screening campaign that was carried out in 58 public secondary schools in Galicia (NW Spain), the largest radon-prone area in the Iberian Peninsula, a positive correlation between indoor 222Rn concentration and outdoor gamma exposure rate was obtained. A new approach to the data acquisition in screening surveys was tested, improving the performances of this type of study and gathering useful data for future remedial actions. Using short-period detectors (charcoal canisters) firstly, in order to detect places showing 222Rn concentrations over 400 Bq m−2, the number of locations to be measured with long-period detectors (etched track detectors) is reduced. In this screening campaign, 34% of the schools surveyed presented at least one site exceeding the 400 Bq m−2 recommended action level established by the EU, and 15% had at least one site with 222Rn values over 800 Bq m−2. The maximum value recorded was 2084 ± 63 Bq m−2. These results are discussed and compared with data obtained in schools of several countries with similar geology. Seven schools were also studied for seasonal variations of 222Rn activity concentration. The results were not conclusive, and no significant correlation between season and 222Rn concentration was established. Finally, a continuous 222Rn concentration monitor was placed in the secondary school exhibiting a mean value of the 222Rn concentration very close to 400 Bq m−2. Maximum 222Rn concentration values were found to occur at times when the school was unoccupied.

1. Introduction

Galicia (in Northwest Spain) belongs to the Iberian Massif (or Hesperic Massif), and is dominated, almost entirely (apart from a little sedimentary tertiary basin) by outcrops of igneous fractured rocks and metamorphic rocks included on the “crystalline rocks” category of (Banks and Robis, 2002). These rocks have a high variety of petrologic types: igneous acidified, medium and basic, schists, migmatises, granulites, quartzites, marbles, and amphibolites, some of them rich in 238U. The Galician Autonomous Community was identified in the MARNA Project as the most extensive radon-prone area in Spain (Quindós et al., 2004). This nationwide survey of natural gamma emission reported exposure rates in this area of 0.36–2.15 pGy kg−1 s−1 (5–30 μR h−1) (from 2006 onward, the SI recommends to use C kg−1 s−1 instead of R h−1; 1R = 2.58E-04 C kg−1). Quindós et al. (2004) related the outdoor gamma exposure values with indoor 222Rn concentrations assigning values ranging from 200 to “more than 400” Bq m−2 for this region, in agreement with their scarce experimental data. A subsequent work in the same area (Quindós et al., 2008) concluded that external gamma radiation can only be used as a qualitative indicator of high indoor radon concentrations when extensive areas are studied and no quantitative indoor radon predictions can be inferred from it. The soil content of 222Rn is the best indicator. Other international work (e.g., Limoto et al., 2001) obtained weak positive correlations between gamma dose rate and radon concentration indoors. One of the aims of this work was to verify if there exists a positive correlation between the indoor radon concentration and the outdoor environmental gamma dose.

At present (2010), there is no specific legislation in Spain regarding 222Rn protection and the reference values used are those provided by the EU (The Council of the European Union, 1990, 1996). Unlike other countries such as Ireland (Synnott et al., 2006), the Slovak Republic (Dúrcik et al., 1997) or Belgium (Poffin et al., 1992), neither systematic surveys nor any kind of organized measurements have been implemented in the Galician Autonomous Community to characterize the radiological quality of the air in public schools.

Between 2006 and 2009, the LAR-USC (Laboratorio de Análisis de Radiaciones of the Universidade de Santiago de Compostela) carried out a screening campaign of 58 public secondary schools throughout the Galician Community. These data have been crossed

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with data of outdoor gamma exposure rates obtained from MARNA project, in order to determine the possible correlation between both magnitudes.

Mainstream concepts often consider the use of short-term measurements as ill-advised because of the seasonal variation in radon. However, short-term measurements can be used, if they are correctly interpreted, to identify locations with elevated radon concentrations (WHO, 2009). For this survey, we used a new protocol, mixing different radon detection methods to make this type of study cheaper and giving useful information for subsequent remedial actions, without loss of scientific strictness.

Simultaneously, a seasonal study was carried out in seven schools in the city of Santiago de Compostela (Galicia’s capital) in order to confirm previous studies of indoor $^{222}$Rn concentration by Cortina et al. (2008), who reported a low seasonal variation in the radon concentration. The radon concentration was found to be a little higher in autumn and winter than it was in the warm period, which is in agreement with other findings (Bochicchio et al., 2005; Kullab et al., 2001; Papaefthymiou et al., 2003; Singh et al., 2005). Moreover, in the same direction, Finland and Sweden recommend measurements during the season from October to April as during this period the heating produces higher radon concentrations.

Finally, a $^{222}$Rn continuum monitor was used in one school in Santiago de Compostela having very high radon concentration levels, in order to determine a possible synchronization between the maximum daily $^{222}$Rn levels and the school activity.

### 2. Experimental method

The study began with systematic measurements using short-exposure-time detectors (charcoal canisters) in all the selected schools. In those places with $^{222}$Rn concentration values over 400 Bq m$^{-3}$, we used long-exposure-time detectors (etched track films) to confirm the values obtained.

The charcoal canisters (Model RA40V from F&J Specialty) were used following a modified EPA 520/5-87-05 protocol (Gray and Windham, 1987), in which an HPGe detector (Ortec, model CMX-50) with digital electronics (Ortec, model DSPEC) was used instead of a NaI (Tl) scintillator. Each charcoal canister was exposed for a period of 48 h. Once secular equilibrium between $^{222}$Rn and its short-period daughters ($^{214}$Pb and $^{214}$Bi) was reached, $^{222}$Rn concentration was measured by comparing the activity of the sample in the energy spectrum region between 295 keV and 609 keV with the equivalent activity of a standard source made of a charcoal canister like the ones used in the measurements, activated with a calibrated source of $^{226}$Ra traceable to the Spanish standard (CIEMAT certificate Ref. P4/100/1MB04RN042). The average sample analysis time was 10 min. The MDA for this measurement method was 8 Bq m$^{-3}$ typically (calculated following the EPA 520/5-87-05 protocol).

Long-exposure-time measurements were carried out using Kodalp ha etched track detectors (with LR-115 film as active volume) exposed for 90 days, typically. The detectors were then analyzed by the provider: GT-Analytic (Austria).

A traceability system was set up both for sampling and measuring, in order to guarantee the quality of the results. Inter-comparison exercises with other laboratories and double exposures at control sites, using two detectors at the same time and place, were performed throughout the measurement campaign and allowed us to estimate the statistical uncertainty.

The charcoal canisters showed a 2% mean deviation between pairs (standard deviation of 2.2 Bq m$^{-3}$), while track detector reproducibility demonstrated a 4% mean deviation between pairs (standard deviation of 58 Bq m$^{-3}$). Kodalp ha is

### Table 1

Geometric mean by floors with geometric standard deviation (GSD in brackets). Semi-basements and basements are all together in the basements category, and mezzanines in the upper level category.

<table>
<thead>
<tr>
<th></th>
<th>Basements (Bq m$^{-3}$)</th>
<th>Ground floors (Bq m$^{-3}$)</th>
<th>Upper levels (Bq m$^{-3}$)</th>
<th>Total (Bq m$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Capital (Santiago de Compostela)</td>
<td>177 (3)</td>
<td>290 (3)</td>
<td>119 (2)</td>
<td>189 (3)</td>
</tr>
<tr>
<td>Rest of Galicia</td>
<td>209 (3)</td>
<td>169 (3)</td>
<td>149 ± 2</td>
<td>171 ± 3</td>
</tr>
<tr>
<td>Total</td>
<td>202 (3)</td>
<td>179 (3)</td>
<td>137 (2)</td>
<td>174 (3)</td>
</tr>
</tbody>
</table>
guaranteed by the provider to be accurate within 10%, based on comparative tests conducted by the Federal Office for Calibration and Measurement (BEV), in Vienna.

Continuous measurements of $^{222}\text{Rn}$ concentration, together with temperature, pressure and humidity, were carried out using an AlphaGuard 2000 Pro monitor (Genitron Instruments, Germany); calibration was certified by Genitron Instrument’s professional calibration system (Frankfurt, Germany), and traceable to international standards. The $^{222}\text{Rn}$ concentration measurement accuracy is better than 0.1% within the rank of our measurements. The MDA estimated by this measurement method is 20 Bq m$^{-3}$. The study included 51 public secondary schools uniformly distributed over the Galician Community territory, based on a selection criterion of two schools every 1 km$^2$ (Compostela area (1 school per 1 km$^2$)) to have additional sites were included in the schools selected for the seasonal study.

To plan the measurements, we visited all the selected schools and selected four sites in each, corresponding to the most inhabited places (by amount of time occupied or number of people using the room). Classrooms or offices located primarily in basements and on ground floors were selected for the study. Two additional sites were included in the schools selected for the seasonal study.

In the first stage, we took short-term measurements using charcoal canisters in each of the four sites selected on every school. Measurements were performed at weekends in order to minimize interference with school activities. Sites with $^{222}\text{Rn}$ concentrations above 400 Bq m$^{-3}$ were cross-checked with long-term measurements (Kodapha). In the seasonal study, short-term measurements were repeated quarterly. Sites with $^{222}\text{Rn}$ concentrations exceeding 400 Bq m$^{-3}$ were again measured with long-term detectors.

The charcoal canisters were given to a contact person in each school. This person was responsible for placing the detectors at the selected sites and completing the statistical forms. After exposure, the canisters were returned to our laboratory by express mail and measured. The Kodalpha detectors were sent directly to the schools with precise instructions for use.

Local meteorological parameters used in the analysis of the continuous $^{222}\text{Rn}$ monitor data were taken from the on-line records of the weather station nearest to the school selected for the continuous measurement study: the Santiago-EOAS station, operated by the public institution Meteogalicia (www.meteogalicia.com).

### 3. Results and discussion

We took a total of 374 short-term measurements with charcoal canisters placed at 246 locations in 58 different schools. The results followed log-normal distributions, with a maximum value of $2259 \pm 38$ Bq m$^{-3}$. Table 1 shows the geometric mean (GM) and geometric standard deviation (GSD) classified by height (compared to ground floor level) and geographical reference. The GM concentration obtained in this study for ground floors in Santiago de Compostela was 285 Bq m$^{-3}$, with a GSD of 2.5 (68% of data between 114 Bq m$^{-3}$ and 710 Bq m$^{-3}$) being consistent with a previous work in the same area using charcoal canisters (Cortina et al., 2008), which provided a GM of 253 Bq m$^{-3}$ and GSD of 2.5 (with a 68% of data between 86 Bq m$^{-3}$ and 743 Bq m$^{-3}$).

Table 2 shows the number and percentage of secondary schools with at least one room (classroom and office) measured within a specific $^{222}\text{Rn}$ concentration interval. It must be noted that some schools can contribute with more than one room to each range and, then, numbers and percentages in this table are not addable (between them). The first survey with short-term measurements (left column in Table 2) revealed that 41% of the schools studied had at least one room with $^{222}\text{Rn}$ activity concentrations exceeding the EU recommend level of 400 Bq m$^{-3}$.

Sites exceeding 400 Bq m$^{-3}$ were re-measured using long-term detectors. Of the 63 detectors exposed, the maximum concentration registered was $2084 \pm 63$ Bq m$^{-3}$ (compared with 2002 $\pm 34$ Bq m$^{-3}$ measured with charcoal canisters). These measurements were used to refine the results obtained from the short-term measurements (right column in Table 2). The long-term measurements revealed that more than one-third of the schools surveyed had rooms with $^{222}\text{Rn}$ concentrations over 400 Bq m$^{-3}$ (left column Table 2). Of the 58 centres studied, 15% had classrooms or offices with concentrations exceeding 800 Bq m$^{-3}$.

### Table 3

Mean (±standard deviation) seasonal variation of the $^{222}\text{Rn}$ concentration with respect to the annual mean value.

<table>
<thead>
<tr>
<th></th>
<th>Summer</th>
<th>Autumn</th>
<th>Winter</th>
<th>Spring</th>
</tr>
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<tbody>
<tr>
<td>Mean deviation (%)</td>
<td>$-12 \pm 34$</td>
<td>$18 \pm 37$</td>
<td>$-12 \pm 36$</td>
<td>$-3 \pm 28$</td>
</tr>
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</table>
in a radon-prone region as Galicia, this measurement strategy is and long integration times) are compatible (4% deviation on we obtained exceed those reported in other European countries. In our work, the GPS positions of each school were recorded in of 234,137 gamma exposure rates measured (MARNA-Galicia). In locations that follows a normal distribution. This correlation was activity concentration and the natural logarithm of the gamma mean 222Rn activity concentration measured in each of the sites in the secondary monitor, where \( \rho \) is the standard deviation.

The percentages obtained using the different methods (short and long integration times) are compatible (4% deviation on average with an SD of 3%), leading to the conclusion that, at least in a radon-prone region as Galicia, this measurement strategy is useful. This work validates short-term measurements with charcoal canisters, being well suited both for detecting “hot spots” and for simultaneous testing of many sites and locations, as well as for providing quick (~4 days) highly reliable results. In a second phase of study long-term measurements can be used to focus only on targets of interest, saving time and money.

MARNA was a nationwide project conducted by the Consejo de Seguridad Nuclear (CSN) to determine the background gamma exposure rate in Spain, with a typical grid-cell of 6 km \( \times \) 4 km. For Galicia, this resolution was improved to 1 km \( \times \) 1 km with a total of 234,137 gamma exposure rates measured (MARNA-Galicia). In our work, the GPS positions of each school were recorded in order to look for the correlation between the gamma exposure rate outdoors and radon concentration indoors. Figs. 2 and 3 show the relation between the natural logarithm of the 222Rn activity concentration and the natural logarithm of the gamma exposure rate given by the MARNA-Galicia grid-cell at the school locations that follows a normal distribution. This correlation was studied separately for basements + ground floors (Fig. 2) and upper floors (Fig. 3). A linear correlation (Pearson coefficient of 0.216 with a statistical significance of 0.002) between both magnitudes was observed for basements and ground floors, giving a slope of 0.7 with an SD of 0.2. The high dispersion of data in Fig. 2 is typical of radon measurements that show a large variability because of both the weather conditions and the location of the detector (at the level of a few meters). The slope of the relationship between gamma dose rate and radon concentration for basements and ground floor is higher than the 0.00324 reported by limoto et al. (2001), for work in Taiwan. However, this correlation was not observed for the upper floor as shown in Fig. 3, which, in this case, shows a slope compatible with zero, of ~0.32 with an SD of 0.52.

The measurement campaign included a seasonal study performed in seven schools in the city of Santiago de Compostela. It revealed small deviations from the annual mean that were not statistically significant (Table 3). They were consistent with previous studies in the same area (Cortina et al., 2008), which reported similar seasonal variations, but had winter and summer maximums with high relative uncertainties.

A study of the time relation of the daily 222Rn activity cycle with the school open time (when personnel and students were in the centre) was done at the school in Santiago de Compostela presenting the second highest 222Rn concentration of the centres surveyed. This school is located in an historic building, 400 years old, with a wood ceiling and a stone structure that is poorly insulated from the soil. It was monitored quarterly using charcoal canisters for one year and, in a second stage, etched track detectors were installed at six different sites exhibiting high 222Rn concentrations (Table 4). To complement these measurements, a continuous 222Rn monitor was used for nine days in two classrooms, identified as P1 and P2, and one office, P3. On the ground floor, P1 is situated directly opposite to P2, whereas P3 is on the upper floor directly over P1. Indoor 222Rn activity concentration, temperature, pressure and relative humidity were recorded every 10 min. Outdoor temperature was also recorded, with the same sampling frequency, from the public data of the Santiago-EOAS weather station located approximately 500 m away. During data processing this information was averaged to 1 h intervals, which were in turn averaged for the nine days studied. In the analysis, school days and weekends were differentiated for the three sites. Fig. 4a and b shows the average values for P1.

Fig. 4a shows the daily 222Rn concentration cycle during school days; school hours are in grey. The difference between indoor and outdoor temperature seems to be what increases the 222Rn concentration during the night. Central heating is working from 7:00 to 10:30 TU and from 16:00 to 19:30 TU, increasing the difference between indoor and outdoor temperature in the first part of the night and favouring the rising of the 222Rn concentration. An inertia phenomenon could explain the 222Rn maximum hours after the central heating stopped (Cortina et al., 2008). Fig. 4b shows data collected at the weekend, when the temperature gradient does not show any clear influence on the 222Rn concentration activity observed. The 222Rn activity concentration on the weekend was constantly high (~1300 Bq m\(^{-3}\) as compared with the oscillation between ~1173 Bq m\(^{-3}\) during school hours and ~1400 Bq m\(^{-3}\) the rest of the time on school days), and did not show the natural daily cycle that was previously observed.

### Table 4
Mean 222Rn activity concentration measured in each of the sites in the secondary school selected for studying the possible synchronization between 222Rn concentration and school hours. Values obtained by different measurement methods are shown. Alphaguard: average of the measurements taken with the continuous 222Rn monitor, where \( \pm \) is the standard deviation.

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<tr>
<td>P1</td>
<td>1929 ± 21</td>
<td>1582 ± 34</td>
<td>1220 ± 32</td>
<td>1323 ± 12</td>
</tr>
<tr>
<td>P2</td>
<td>841 ± 15</td>
<td>587 ± 29</td>
<td>321 ± 16</td>
<td>839 ± 7</td>
</tr>
<tr>
<td>P3</td>
<td>911 ± 16</td>
<td>564 ± 28</td>
<td>398 ± 15</td>
<td>657 ± 5</td>
</tr>
<tr>
<td>P4</td>
<td>455 ± 12</td>
<td>517 ± 31</td>
<td>–</td>
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</tr>
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<td>P5</td>
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<td>301 ± 21</td>
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<tr>
<td>P6</td>
<td>483 ± 12</td>
<td>434 ± 26</td>
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Even though the statistics of the present study are rather limited, they are instructive to compare our findings with more general studies performed in similar geological regions such as Ireland, with 8.6% of schools registering over 400 Bq m\(^{-3}\) (Synnott et al., 2006), or Luxembourg, with 1.4% of schools exceeding this limit (Poffijn et al., 1992). In the Slovak Republic, 2.5% of kindergartens and schools found to have concentrations greater than 200 Bq m\(^{-3}\) (Durčík et al., 1997). We can conclude that the values we obtained exceed those reported in other European countries.

Outdoor temperature was also recorded, with the same sampling frequency, from the public data of the Santiago-EOAS weather station located approximately 500 m away. During data processing this information was averaged to 1 h intervals, which were in turn averaged for the nine days studied. In the analysis, school days and weekends were differentiated for the three sites. Fig. 4a and b shows the average values for P1.

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Last but not least, we also searched for correlations between 222Rn concentration and other parameters. An analysis based on Pearson coefficients indicated a positive linear correlation (Pearson coefficient average 0.755), between the indoor 222Rn concentration and temperature difference between indoor and outdoor temperature for all rooms studied and for school days. This is very probably the consequence of the central heating of the building, being clearly not the dominant mechanism at the weekend, when 222Rn concentration was higher and constant.

Even though the highest 222Rn concentration values were recorded outside school hours (8:00–13:25 UT and 15:00–17:30 UT), the continuous average 222Rn concentrations were above the EU action level (~400 Bq m\(^{-3}\)) in P1 and P2. P3 (located on the upper of the building) presented 222Rn concentration values very near the EU action level.
4. Conclusions

We observed a positive linear correlation between the gamma exposure rate in the area where the schools are located, and the $^{222}\text{Rn}$ activity, when measured in basements and ground floors. Further, this work validates short-term measurements with charcoal canisters, being well suited for detecting “hot spots” and for simultaneous testing of many sites and locations, as well as for providing quick highly reliable results. Short- and long-integration-time measurements are compatible, but etched track detectors generally gave lower $^{222}\text{Rn}$ concentration values than charcoal canisters. Nevertheless, they gave values closer to the continuous data when averaging school days and weekends. The largest discrepancies were found in places with the highest $^{222}\text{Rn}$ concentration.

The seasonal study did not show a statistically significant correlation with $^{222}\text{Rn}$ concentration.

We found that 34% of the schools surveyed have at least one site (classroom or office) with $^{222}\text{Rn}$ concentration greater than the EU recommended action level of 400 Bq m$^{-3}$. A total of 15% were found to have sites with $^{222}\text{Rn}$ activity concentrations above 800 Bq m$^{-3}$. These results are higher than the reported by other European countries with similar geological characteristics, and clearly excessive in light of the new 100 Bq m$^{-3}$ reference level recommended by the World Health Organization (WHO, 2009).

It is necessary to increase the number of surveyed locations covering more densely the geographic extension of the Galician radon-prone area in order to study more in depth the relation between $^{222}\text{Rn}$ concentration and gamma exposure rate. Indeed, from the point of view of the population radiation protection, we
think that it is necessary to extend the scope of the survey, not only to secondary schools, but to kindergartens and primary schools, in order to establish guidelines for a prevention and mitigation strategy.

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References


