Radon and thoron exhalation rate, emanation factor and radioactivity risks of building materials of the Iberian Peninsula

S. Frutos-Puerto[1], E. Pinilla-Gil [1], E. Andrade[2,3], M. Reis[2,3], M.J. Madruga[2,3], C. Miró [4]

[1] Department of Analytical Chemistry, University of Extremadura, Av. de Elvas, s/n, 06006 Badajoz, Spain

[2] Centro de Ciências e Tecnologias Nucleares, Estrada Nacional 10, ao km 139,7, 2695-066 Bobadela LRS, Portugal

[3] Laboratório de Proteção e Segurança Radiológica, Instituto Superior Técnico, Universidade de Lisboa, Estrada Nacional 10, ao km 139,7, 2695-066 Bobadela LRS, Portugal

[4] Department of Applied Physics, University of Extremadura, Av. de la Universidad, s/n, 10005 Cáceres, Spain

Corresponding Author:

S. Frutos-Puerto[1]

Av. de Elvas, s/n, 06006 Badajoz, Spain

samfrutosp@unex.es

Abstract

Radon (^{222}Rn) and thoron (^{220}Rn) are radioactive gases emanating from geological materials. Inhalation of these gases is closely related to an increase in the probability of lung cancer if the levels are high. The majority of studies focus on radon, and the thoron is normally ignored by its short half-life (55.6 s). However, also the thoron decay products can cause a significant increase in dose. In buildings with high radon levels, the main mechanism for entry of radon is the pressure-driven flow of soil gas through cracks in the floor. Both radon and thoron can also be released from building materials to the indoor atmosphere.

In this study, the ^{222}Rn and ^{220}Rn exhalation rates (mass and surface) and the emanation coefficinets an extended variety of common building materials manufactured in the Iberian Peninsula (Portugal and Spain) but exported and used in all countries of the world were determined by using active measuring system. Radon and thoron emission from samples collected in the closed chamber was measured by an active method that uses a continuous radon/thoron monitor. The correlations between exhalation rates of these gases and their parent nuclide exhalation (radium/thorium) concentrations were examined. Finally, indoor radon and thoron and the annual effective dose were calculated from radon/thoron concentrations in the closed chamber. Zircon is the material with the highest concentration values of ^{226}Ra and ^{232}Th and the exhalation and emanation rates. Also in the case of zircon and some granite, the annual effective dose was higher than the annual exposure limit for the general public of 1 mSv y^{-1}, recommended by the European regulations.

Introduction

Radon and thoron are significant contributors to the average dose from natural background sources of radiation. They represent approximately half of the estimated dose from exposure to all natural sources of ionizing radiation (*UNSCEAR, 2008)*.

Inhalation of these radioactive gases and their decay products can cause health risks, especially in poorly ventilated areas. Long-term exposure to high levels of radon/thoron in home and working area increases the risk of developing lung cancer. Radon is the second leading cause of the increase in the probability of lung cancer after tobacco smoke (Torres-Durán et al., 2014).

After its formation, these two radioisotopes are susceptible to escape, firstly from the grains constituting the material (known as emanation), and secondly, from the surface of the material (known as exhalation). These parameters depend, among other factors, on the half-life, consequently affecting the accumulation rate of these gaseous radioisotopes in indoor environments, and therefore, to the exposure of the human body to radiation. For radon, the half-life is 3.8 days while for thoron, just 55.6 s so, due to this difference, the equivalent radiation dose from thoron and its progeny (^{212}Pb and ^{212}Bi) is estimated around of 10 % of that due to radon and its progeny (^{214}Pb and ^{214}Bi) in indoor environments (UNSCEAR, 2016)*g*.

These factors lead to a complicated thoron measurement technique that, together with the fact that its inhalation dose is not relevant in most cases, has made that the majority of the existing studies focus on the radon to the detriment of the thoron. Many of them also include measures of ^{40}K, ^{226}Ra and ^{232}Th along with risk indexes definitions trying to evaluate the radiological health hazards of these radionuclides (Turhan & Gündüz, 2008; De With, De Jong & Röttger, 2014; Kumar et al., 2015; Kayakökü, Karatepe & Doğru, 2016; Madruga et al., 2018) or the effective dose due to radon and its progeny (Sabiha-Javied, Tufail & Asghar, 2010).

Nevertheless, despite thoron indoor concentration is generally lower than for the radon, the ^{212}Pb thoron progeny (half-life of 10.6 h) can accumulate to significant levels in breathable air, aggravating its inhalation risk (World Health Organization, 2009). Some studies have demonstrated that thoron concentrations can be comparable to radon and its progenies in some areas of elevated radiological risk. Furthermore, computational studies (de With & de Jong, 2011) taking into account factors such as the ventilation and air exchange, the building dimensions, dispersion and deposition, mitigation measures, and material properties indicates that thoron effective doses can reach the 35 % of the total contribution.

Therefore, these studies demonstrate the recent and growing interest that has emerged in recent decades by the study of thoron in building materials [16, 19, 36, 37] although no further studies have been reported yet focusing in the assessment of the thoron risk index in the building materials used in buildings.

Among the methods to measure both exhalation rate and emanation factor of radon and thoron isotopes in building materials, passive methods, that use solid-state nuclear track detector, accumulation chamber methods and active methods with radon/thoron monitors, can be found (Zhang et al., 2012).

In previous work, the gamma radiations emitted from ^{226}Ra, ^{232}Th and ^{40}K for some of these materials were studied, as well as the radiological health hazards associated with the external gamma radiation (Madruga et al., 2018). In another study (Frutos-Puerto et al., 2018), a technique of measurement of thoron had been developed and applied to the analysis of exhalation of 5 materials. In the present work, expanded with more materials, we study the radon and thoron exhalation and emanation properties of an extended variety of common building materials used in the Iberian Peninsula (Portugal and Spain). The correlations between exhalation rates of these gases and their parent nuclide exhalation (radium/thorium) concentrations were examined. Furthermore, indoor radon/thoron and the annual effective dose were calculated from radon/thoron concentrations in the closed chamber. Measurements were carried out by an active method that uses a continuous radon/thoron monitor RTM1688-2 (SARAD GmbH, Dresden, Germany).

Materials and methods

Materials and sample preparation

Forty-one samples from quarries and suppliers of the most commonly used building materials manufactured in the Iberian Peninsula were collected. The mass of each sample ranged between 1 and 5 kg. Fig. 1 shows the geographical origin of the materials. The materials were divided into two classes: materials coming from natural sources, NM, naturally occurring radioactive materials (NORM) incorporating waste after industrial processing, PM ("Directive. 2013/59/Euratom of 5 December 2013," 2014). Within each classification of materials are found:

Materials type NM:
- Concretes. Used in bulk amounts:
    - Conventional.
    - 100% of the natural aggregate becomes electrical furnace slags.
    - 100% of the natural aggregate becomes blast furnace slags.
    - Self-compacting.
    - High-resistance.
    - Mortars of resistance 5 and 7.5, respectively.
- Cements. Used in bulk amounts and superficial applications:
    - Type I Portland cement with less than 3% fly ash.

111   - White cement.

112   - Cement glue.

113   - Rapid cement.

114 • Natural stones. Used as bulk and superficial products:

115   - Marble.

116   - Granite.

117   - Slate.

118 • Ceramic tiles as refractory and ceramic products to cover floors and walls, mainly:

119   - Tiles

120 • Raw materials of very different types and composition:

121   - Wood collected from Eucalyptus and Castahea Sativa trees.

122   - Aggregates as sand or clay bricks.

123   - Zircon.

124   Materials type PM:

125 • Industrial products resulting from the sulphate industry of the North of Spain:

126   - Gypsum

127   - Plastic cement

128   Sample preparation consisted into crushing and drying building materials in an oven for 48 h

129 at 105ºC, before its grounding and sieving (2 mm particle size).

130 *Gamma spectroscopic analysis*

131   To carry out the γ-emissions measurements, the milled samples were dried and placed in 160

132 cm^3 cylindrical containers made of plastic or in 1000 cm^3 Marinelli beakers, both, hermetically

133 sealed for 28 or more days. This period is sufficient for equilibrium to occur between the

134 radioisotopes of ^{226}Ra and ^{232}Th initially contained in the material and their decay products.

135   To obtain the ^{232}Th and ^{226}Ra content an HPGe semiconductor detector was employed

136 according to the methodology followed by Madruga et al. (Madruga et al., 2018)

137   In summary, the activity concentration for ^{232}Th and ^{226}Ra (*A*) was calculated by the

138 following expression:

$$C = \frac{N}{t \, P \, \epsilon_f} \tag{1}$$

139 where N stands for net counts, t for data collection time, P for emission probability and ϵ_f for the

140 efficiency of the detector for the corresponding peak. Besides, uncertainty in the yield is also

141 included since several γ-ray peaks were used for the calculation of ^{232}Th and ^{226}Ra activity.

142

143 *Determination of massic exhalation rate and emanation factor*

144 Exhalation is the amount of radon (radon activity) as obtained from a given layer (geological

145 material on the surface/surface exposure) mainly the outer thinner part of the crust and it is given

146 in Bq h^{-1}, according to the Netherlands Standardization Institute ("Netherlands Standardization

147 Institute. Dutch Standard: Radioactivity measurement. Determination method of the rate of the

148 radon exhalation of dense building materials," 2001). Exhalation can be related to the mass of the

149 samples (massic radon/thoron exhalation, and its value is expressed Bq/kg h). The method

150 already referred and similar to that of other authors (Hassan et al., 2011) was employed to assess

151 the massic exhalation of ^{222}Rn and ^{220}Rn and it is schematized in Fig. 2.

152 The calculation of ^{222}Rn and ^{220}Rn exhalation was carried out according to the expressions

153 presented in the Miró et al. (Miro et al., 2014) from the formula of the temporal variation of the

154 radon concentration C(t), in Bq/m^{3}:

$$\frac{dc}{dt} = \frac{EM}{V} - \lambda C - \alpha C \qquad (2)$$

155 where E (Bq/kg^1 h^1) is the radon-specific exhalation rate, M (kg) the mass of the sample, V (m^3)

156 the air volume of the container, λ (1/h^1) the ^{222}Rn or ^{220}Rn decay constant and α (h^{-1}) the leakage

157 rate from the container.

158 By solving Equation (2), the radon concentration growth as a function of time is given by:

$$C(t) = \frac{EM\left[1 - e^{-(\lambda+\alpha)t}\right]}{(\lambda + \alpha)V} + C_0 e^{-(\lambda+\alpha)t} \qquad (3)$$

159 being C_0 (Bq m^{-3}) the radon concentration at $t = 0$.

160 The ^{222}Rn exhalation (E_{Rn222}) and α numeric calculation are made by adjusting by least-

161 squares of the C vs t experimental data to the mathematical function given by equation (3).

162 However, due to its short half-life, after the first cycle (2 hours) of measurements, the

163 concentration of thoron in the container will reach its maximum value, remaining constant until

164 the end of the measurements. So, from equation (3) the massic thoron exhalation, E_{Rn220}, can be

165 calculated from the expression (4), which do not consider α value because it is much smaller than

166 the thoron decay constant, λ_{Rn220}:

$$E_{Rn220} = \frac{C_{Rn220}\ \lambda_{Rn220}\ V}{M} \tag{4}$$

167 where C_{Rn220} (Bq m^{-3}) is the average concentration of thoron in the container during the interval

168 of measurement from the first cycle of 2 hours.

169   The emanation factor (amount of radon and thoron atoms that escape from the grains

170 constituting the material into the interstitial space between the grains), ε_{Rn}, was calculated by the

171 following equation for both radioisotopes (Stoulos, Manolopoulou & Papastefanou, 2003):

$$\varepsilon_{Rn} = \frac{E_{Rn}}{C_i \lambda_d} \tag{5}$$

172 where C_i is the ^{226}Ra or ^{232}Th content (Bq kg^{-1}) of the sample for radon and thoron, respectively,

173 λ_d, the decay constant and E_{Rn} the exhalation

174   Equation (5) is applicable for all measured building materials because the dimensions of the

175 samples were chosen to be equal to the diffusion length of these gases for these materials, around

176 4 cm (Stoulos, Manolopoulou & Papastefanou, 2003).

177

178 *Determination of annual effective dose*

179   The ^{222}Rn/^{220}Rn content accumulates in the surrounding air in a dwelling room, from

180 building materials, depends on factors such as the room dimension, the parent element

181 concentration, the subsequent exhalation directly from the soil and building materials in walls or

182 soil (radon gain), the air exchange and the isotope radioactive decay. Therefore, attending to

183 these factors, building materials may cause an excess in the indoor ^{222}Rn or ^{220}Rn activity

184 concentrations, which is described by the following equation (Amin, 2015):

$$A_{Rn} = \frac{E_A\ S}{V_r\ \lambda_v} \tag{6}$$

185 where, A_{Rn}, is the ^{222}Rn or ^{220}Rn activity concentration (Bq m^{-3}) in the air of the room; E_A is the

186 surface exhalation rate (Bq m^{-2} h^{-1}); S is the exhalation area (m^2); V_r is the volume of the room

187 (m^3) and λ_v is the ventilation rate of the room (h^{-1}). Ratio S/V is taken to be 2 and λ_v, 0.5 h^{-1}

188 (*United Nations Scientific Committee on the Effects of Ionisin Radiation (UNSCEAR), Sources,*

189 *Effects and Risks of Ionizing Radiation, Report to the General Assembly*, 2016). Considering the

190 value of the sample emanation surface in the container (0.0078 m^2; circumference of 5 cm^2), and

191 the mass of the sample (M), the surface exhalation rate (E_A) for the building materials can be

192 calculated, using the following equation:

$$E_A = E_{Rn} \frac{M}{0.0078} \tag{7}$$

193   This radon concentration model can then be used to determinate the annual effective doses of

194 ^{222}Rn by equation (8), recommended by the United Nations Scientific Committee on the Effects

195 of Atomic Radiation (*United Nations Scientific Committee on the Effects of Ionisin Radiation*

196 *(UNSCEAR), Sources, Effects and Risks of Ionizing Radiation, Report to the General Assembly*,

197 2016):

$$D_{Rn222} = A_{Rn222} \, F_e \, T_a \, CF_{Rn222} \tag{8}$$

198 where D_{Rn222} is the annual effective dose of ^{222}Rn (Sv y^{-1}); A_{Rn222} is the activity concentration for

199 ^{222}Rn (Bq m^{-3}); CF_{Rn222} is the dose conversion factor for ^{222}Rn progeny (Sv per Bq h m^{-3}); F_e is

200 the equilibrium factor for ^{222}Rn and its progeny, and T_a is the annual work time. The standard

201 parameters were estimated using the RP 122 publication of EC 2002 (European Commission,

202 2002). The values of CF_{Rn222} were assumed to be 9×10^{-9} Sv per Bq h m^{-3} and the T_a, 7000 h y^{-1}.

203 The value of F_e was assumed to be 0.4 as reported in (*United Nations Scientific Committee on the*

204 *Effects of Ionisin Radiation (UNSCEAR), Sources, Effects and Risks of Ionizing Radiation,*

205 *Report to the General Assembly*, 2008).

206 Similarly, for ^{220}Rn:

$$D_{Rn220} = A_{Rn220} \, F_e \, T_a \, CF_{Rn220} \tag{9}$$

207 where, D_{Rn220} is the annual effective dose of ^{220}Rn (Sv y^{-1}); A_{Rn220} is the activity concentration for

208 ^{220}Rn (Bq m^{-3}); CF_{Rn220} is the dose conversion factor for ^{220}Rn progeny (40×10^{-9} Sv per Bq h m^{-3})

209 and T_a is the annual work time, 7000 h y^{-1} (European Commission, 2002). F_e is the equilibrium

210 factor for ^{220}Rn and its progeny, 0.1 (*United Nations Scientific Committee on the Effects of*

211 *Ionisin Radiation (UNSCEAR), Sources, Effects and Risks of Ionizing Radiation, Report to the*

212 *General Assembly*, 2008).

213   However, since the diffusion length of ^{220}Rn is very short it is complex and ambiguous to

214 calculate the internal exposure due to ^{220}Rn exhaling from the building material. The indoor

thoron concentration in air depends on the distance from the wall [21, 23] as presented in the following equation:

$$A_{Rn220}(X) = \frac{E_{ARn220}}{\sqrt{\lambda_{Rn220}D_{eff}}}\exp(-\sqrt{\frac{\lambda_{Rn220}}{D_{eff}}}X) \tag{10}$$

where, $A_{Rn220}(X)$ is the [220]Rn concentration at a distance, X, from de wall. E_{ARn220} is the [220]Rn estimated surface exhalation rate by equation (7), D_{ef} is the effective diffusion coefficient herein taken as 1.8 m^{2}h^{-1} [21], λ_{Rn220} is the decay constant of [220]Rn, 45 h^{-1}.

It is reasonable to assume that the human respiratory organs are not more than 40 cm distance from the wall. Therefore, the [220]Rn concentration at the distance of 40 cm calculated by equation (10), A_{Rn220}, is used to determinate the annual effective doses of [220]Rn with equation (9).

Results and discussion

The results of activity concentration for [226]Ra, C_{Ra}, massic exhalation, E_{Rn222}, and emanation factor, ε_{Rn222}, for [222]Rn are summarised in Table 1.

In all samples, activity concentration for radium was above the detection limit (DL) except for the wood sample. In general, these results are comparable to those measured on a worldwide scale [2, 45-48]. Thus, the values for radium content in building materials are less than the permissible value (370 Bq kg^{-1}), which is acceptable as a safe limit (Group Experts of the OECD Nuclear Energy Agency, 1979). The only exception was in the radium concentration in zircon, the highest value for the mean concentration was 2070 Bq kg^{-1}.

In many samples, the exhalation rate was lower than the DL (because of E_{Rn222} < DL) with exception of all samples of slate, granite and zircon. The maximum value on average was obtained for zircon, 429 mBq kg^{-1} h^{-1}, which is much higher than that found for the aggregate and the granites. The values of exhalation rates reported in Table 1 correspond well with the values reported by other authors [15, 50-53]. The variation in radon exhalation rates (one order of magnitude, in some cases) can be attributed to variations in radium concentrations, porosity, and surface crystallography. The emanation factor range from 0.2 % to 22.0 % for ceramic and aggregates respectively. These values are similar to the measured in worldwide scales [2, 15, 46, 49].

243 The results of activity concentration for ^{232}Th, C_{Th}, massic exhalation, E_{Rn220}, and emanation

244 factor, ε_{Rn220}, for ^{220}Rn are summarised in Table 2.

245 The highest mean value for ^{232}Th activity concentration is shown by zircon (340 Bq kg^{-1}),

246 and the lowest mean value is obtained for wood (0.6 Bq kg^{-1}). The mean values of the ^{220}Rn

247 massic exhalation rate range from 2.2 of the ceramic to 169 Bq kg^{-1} h^{-1} for zircon, respectively.

248 The results show that the thoron exhalation rate is higher in zircon samples and lower in ceramic

249 samples. This can presumably be explained by the different distributions of ^{224}Ra parent element

250 in the different types of samples. It should be noted how the difference between the values of

251 exhalation rate in granites (range from 2.6 to 144 Bq kg^{-1} h^{-1}) reveal their different mineralogical

252 composition. The emanation factor range from 0.3 % to 29 % for ceramic and wood, respectively.

253 The ranges of results of all these parameters are in good agreement with the values reports by

254 other authors [29, 54].

255 A correlation study of ^{222}Rn mass exhalation rate concerning ^{226}Ra content, as shown in Fig.

256 3a, showed a good linear correlation coefficient (R^2 = 0.9961). These results show that the ^{222}Rn

257 mass exhalation rate increases as the ^{226}Ra content is higher in the samples. This good linear

258 correlation has already been observed by other authors, some of them with values very close to 1

259 (Amin, 2015). As could be expected (Fig. 3b), no correlation (R^2 = 0.0109) was found between

260 the ^{222}Rn emanation factor and the ^{226}Ra content.

261 A similar correlation of ^{220}Rn mass exhalation rate with ^{232}Th content is shown in Fig. 4a,

262 which shows a more weak correlation between the two quantities (R^2 = 0.8336). These results

263 show that the ^{220}Rn mass exhalation rate increases for samples with higher ^{232}Th contents, as

264 observed before for the ^{222}Rn exhalation rate and ^{226}Ra contents.

265 Moreover, as could be expected (Fig. 4b), no correlation (R^2 = 0.0115) was found between

266 the ^{220}Rn emanation factor and the ^{232}Th content. Finally, no correlation (R^2 = 0.118) was found

267 between the ^{222}Rn emanation factor and the ^{220}Rn emanation factor as shown in Fig. 4c.

268 The results obtained for indoor contribution, surface exhalation rate, activity concentration in

269 the air of the room, and annual effective dose, for the different building materials had been shown

270 in Tables 3 and 4 for ^{222}Rn and ^{220}Rn, respectively. Therefore, Table 3 shows that the mean

271 values of ^{222}Rn surface exhalation rates varied from 9.2 to 3206 mBq m^{-2} h^{-1} for ceramic and

272 zircon, respectively. The ^{222}Rn contribution of building materials to indoor ^{222}Rn considering the

273 model room mentioned above, range from 0.04 for ceramic samples to 13 Bq m^{-3} for zircon. As a

274   result of this, the annual effective dose ranged from 0.9 µSv y^{-1} for ceramic to 323 µSv y^{-1} for

275   zircon. These values are in agreement with the worldwide range [2, 55].

276   In the case of ^{220}Rn (see Table 4), the surface exhalation rate average varied from 22 to 1264

277   Bq m^{-2} h^{-1} for cement and zircon respectively. Its contribution of building materials to indoor

278   ^{220}Rn at 40 cm of the wall considering the model mentioned above, range from 2.0 for the cement

279   to 112 Bq m^{-3} for zircon. Mean values of the annual effective dose ranged from 16 µSv y^{-1} for

280   gypsum to 1300 µSv y^{-1} for zircon. These values are similar to those found by other authors for

281   building materials (Ujić et al., 2010). However, estimation of annual effective dose from indoor

282   thoron indicated the mean value of zircon and some values of granites had been higher than the

283   annual exposure limit for the general public of 1 mSv y^{-1}, recommended by European Directive

284   2013/59/Euratom [40].

285

Conclusions

287   In this study, the radon and thoron exhalation and emanation properties of building materials

288   commonly used in the Iberian Peninsula (Portugal and Spain) were measured by using an active

289   method with a continuous radon/thoron monitor. The correlations between exhalation rates of

290   these gases and their parent nuclide exhalation (radium/thorium) concentrations were examined.

291   Finally, on the estimation of the indoor radon/thoron, the annual effective dose was calculated.

292   In general, ^{226}Ra content in building materials is less than the permissible value, 370 Bq kg^{-1},

293   except for zircon, which means the value was 2100 Bq kg^{-1}. For this material, the maximum

294   value on average of ^{222}Rn massic exhalation rate (429 mBq kg^{-1} h^{-1}) was also obtained. The

295   emanation factor ^{222}Rn/^{226}Ra range from 0.2 % to 22.0 % for ceramic and aggregates,

296   respectively. On average, the highest value for activity concentration of ^{232}Th and massic ^{220}Rn

297   exhalation rate were showed by zircon, 340 Bq kg^{-1} and 169 Bq kg^{-1} h^{-1}, respectively. The

298   emanation factor of ^{220}Rn/^{232}Th range from 0.3 % to 29 % for ceramic and wood, respectively.

299   The correlation between the radon mass exhalation rate and the ^{226}Ra contents as well as the

300   correlation between the thoron mass exhalation rate and ^{232}Th contents are in good agreement.

301   The mean values of ^{222}Rn surface exhalation rates varied from 9.2 to 3206 mBq m^{-2} h^{-1} for

302   ceramic and zircon, respectively. The ^{222}Rn contribution of building materials to indoor ^{222}Rn

303   considering the model room mentioned above, range from 0.04 for ceramic samples to 13 Bq m^{-3}

for zircon. So, the annual effective dose ranged from 0.9 μSv y^{-1} for ceramic to 323 μSv y^{-1} for zircon.

In the case of ^{220}Rn, the surface exhalation rate average varied from 22 to 1264 Bq m^{-2} h^{-1} for cement and zircon respectively. Its contribution of building materials to indoor ^{220}Rn at 40 cm of the wall, range from 2.0 for cement samples to 112 Bq m^{-3} for zircon. Mean values of the annual effective dose ranged from 16 μSv y^{-1} for gypsum to 1300 μSv y^{-1} for zircon. Therefore, in the case of zircon and some granites, the annual effective dose was higher than the annual exposure limit for the general public of 1 mSv y^{-1}, recommended by the ICRP.

Acknowledgements

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