Radiation Measurements 46 (2011) 112–118

Contents lists available at ScienceDirect

Radiation Measurements

journal homepage: www.elsevier.com/locate/radmeas

Inter-comparison of different direct and indirect methods to determine radon flux from soil

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ARTICLE INFO

Article history:
Received 8 June 2009
Received in revised form
8 April 2010
Accepted 31 July 2010

Keywords:
Radon flux
Direct and indirect methods
Accumulation chamber
Radium
γ Dose rate
Inter-comparison

ABSTRACT

The physical and chemical characteristics of radon gas make it a good tracer for use in the application of atmospheric transport models. For this purpose the radon source needs to be known on a global scale and this is difficult to achieve by only direct experimental methods. However, indirect methods can provide radon flux maps on larger scales, but their reliability has to be carefully checked. It is the aim of this work to compare radon flux values obtained by direct and indirect methods in a measurement campaign performed in the summer of 2008. Different systems to directly measure radon flux from the soil surface and to measure the related parameters terrestrial γ dose and ^{226}Ra activity in soil, for indirect estimation of radon flux, were tested. Four eastern Spanish sites with different geological and soil characteristics were selected: Teruel, Los Pedrones, Quintanar de la Orden and Madrid. The study shows the usefulness of both direct and indirect methods for obtaining radon flux data. Direct radon flux measurements by continuous and integrated monitors showed a coefficient of variation between 10% and 23%. At the same time, indirect methods based on correlations between ^{222}Rn and terrestrial γ dose rate, or ^{226}Ra activity in soil, provided results similar to the direct measurements, when these proxies were directly measured at the site. Larger discrepancies were found when proxy values were extracted from existing data bases. The participating members involved in the campaign study were the Institute of Energy Technology (INTE) of the Technical University of Catalonia (UPC), Huelva University (UHU), and Basel University (BASEL).

1. Introduction

Radon characterization from the soil surface allows estimating the external contribution to indoor ^{222}Rn concentrations and provides a necessary input term to the study of atmospheric transport processes. In the first case, generating radon flux maps can be useful for regulatory national organisations in order to classify radon prone areas (Bochicchio, 2008; Kreuzer et al., 2003). In the second case, the ^{222}Rn source term, in combination with observations of atmospheric ^{222}Rn concentration, facilitates the evaluation and calibration of atmospheric transport models (Zahorowski et al., 2004; Arnold et al., 2010). The ^{222}Rn source is mainly located on the land surfaces. Its only sink in the atmosphere is by radioactive decay with a half-life of 3.8 days. The large contrast between terrestrial and oceanic radon flux makes ^{222}Rn an ideal atmospheric tracer at a regional scale (Lupu and Cuculeanu, 2001; Whittlestone et al., 1992). Development of atmospheric dispersion models has progressed to a point where improved knowledge of the ^{222}Rn source term becomes necessary for more accurate validation (WMO, 2004). The ^{222}Rn concentration in soil and its emission rate depend on the geology of the area, the porosity and permeability of the soil, the terrain structure and the associated ^{238}U mineralization. Diffusion is the process which allows ^{222}Rn to escape from soil pores to the atmosphere after its formation by ^{226}Ra decay. In spite of sufficient understanding of the theoretical processes controlling release of ^{222}Rn from soil to the atmosphere (Nazaroff and Nero, 1988; Porstendorfer, 1994), comprehension of the radon flux and its distribution over the earth is still under question because of a lack of direct radon flux measurements in many regions. Direct measurements of radon flux are ideally made using the accumulation method (Keller et al., 1982; Keller and Schutz, 1988) which allows ^{222}Rn gas to accumulate and to be

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doi:10.1016/j.radmeas.2010.07.021

measured in a chamber placed over the soil. Nevertheless, this approach is not practical on a worldwide scale. A more realistic alternative might be to obtain at least a modest number of accumulator measurements, but at carefully chosen locations, representative of different geological regions and to extrapolate these measurements to similar regions.

Nowadays, other approaches are also used to overcome the need for labour-intensive direct observations. Indirect mapping of radon flux is possible thanks to existing knowledge about parameters related to radon flux and for which more large-scale information is available than for radon flux itself. Different approaches have been applied in order to indirectly map the radon flux. The simplest assumption is that of a radon flux value from the land surface of 1 atom cm^{-2} s^{-1} between 60°S and 60°N, of 0.5 atom cm^{-2} s^{-1} between 60°N and 70°N and of 0 atom cm^{-2} s^{-1} for the sea (Rash et al., 2000). A modification with a linear decrease from 1 atom-cm^{-2} s^{-1} at 30°N to 0.2 atom cm^{-2} s^{-1} at 70°N was proposed (Conen and Robertson, 2002). These estimates are based on atmospheric ^{222}Rn inventories. Another approach has been recently developed within the Swiss project "European radon flux map for atmospheric tracer applications" by Basel University. It is based on the determination of an empirical linear relation between the radon flux and the terrestrial γ dose on a European scale. These last values were extracted during the project from routinely reported emergency monitoring data. These data are available in quasi real time at the Joint Research Centre of the European Commission of ISPRA (Italy) from each of the 3600 stations of the national emergency monitoring network in Europe, EURDEP (Szegvary et al., 2007a).

Knowledge about the used detectors from each contributing country, the detector elevation above sea level and the possible contribution from artificial radionuclides, mainly ^{137}Cs, enables the extraction and the spatial modelling of the terrestrial γ dose (Szegvary et al., 2007a). The dose-rate values for the EURDEP network regarding Spain are provided by the REA, the Spanish Automatic Surveillance Network of the Spanish Nuclear Safety Council (CSN). Another option to obtain the terrestrial γ dose rates is by radiometric data generated through the Spanish National Uranium Exploration and Investigation Plan. This work has developed into the Spanish MARNA project (Quindós Poncela et al., 2004).

The present study aims to compare different methods for direct and indirect radon flux estimation in specific Spanish areas and under their most typical climatologic conditions. An inter-comparison campaign was carried out at four selected Spanish sites in the summer of 2008. Direct radon flux measurement systems required to be compared and previous indirect methods (Szegvary et al., 2007a; Quindós Poncela et al., 2004) wanted to be validated. Continuous and integrated techniques were applied for direct measurements of radon flux form soil. Terrestrial γ dose rate and ^{226}Ra activity soil measurements were done and used to indirectly estimate radon flux. The members involved in the campaign were Huelva University (UHU), Basel University and the Institute of Energy Technology (INTE) of the Technical University of Catalonia (UPC).

2. Material and methods

The measurement campaign across Eastern Spain was performed during the summer of 2008, in the middle of July.. In order to avoid rainy episodes that may influence soil humidity, terrestrial γ dose rate or radon flux, the campaign was carried out during the most typical eastern Spanish climatologic conditions. The radon flux (Bq m^{-2} h^{-1}) and γ dose rates (nSv h^{-1}) were measured at each site by several monitors and in different points in order to get average values. The ^{226}Ra activity (Bq kg^{-1}) was determined in the laboratory on soil samples collected during this time. Each parameter was determined with different equipment by the campaign members with the aim of comparing the results.

2.1. Sites

Four Eastern Spanish sites were selected in this measurement campaign. The chosen locations were Teruel, Los Pedrones, Quintanar de la Orden and Madrid. The location characteristics, the average humidity and the temperature conditions during the measurement campaign are reported in Table 1. These locations are included within the Spanish Automatic Surveillance Network (REA) which allows an evaluation between our measurements of terrestrial γ dose rates with the routine REA data. Furthermore, this selection of sites promised a wide range of radon flux values to be obtained as suggested by the European radon map (Szegvary et al., 2009). Finally, the direct measurements were considered a validation of the anomalously high radon flux values indicated by this map in comparison to the rest of Europe.

2.2. Terrestrial γ-dose rate

Total γ dose measurements were performed at each station at 1 m above the ground with the following portable monitors. A High Pressure Ionization Chamber RS-112 (HPIC, GE Reuter-Stokes, Inc.) was used by the INTE-UPC. This model has a volume of 4.2 l. It is filled with Argon gas at a pressure of 25 atm. Its measurement range is between 10 nGy h^{-1} and 5 μGy h^{-1} with a precision of 5%. A GammaTRACER (Genitron Instruments GmbH, Frankfurt, Germany) with a dual Geiger-Müller-tube was used by Basel University. This device has one tube with 110 pulses per min at 100 nSv h^{-1} and another tube with 0.2 pulses per min at 100 nSv h^{-1}. The measurement range is between 10 nSv h^{-1} and 10 Sv h^{-1}. A proportional counter probe LB 123D by the Berthold company was used by the UHU. Its sensitivity is 0.20 μSv h^{-1} per 1 cps. The energy range is from 30 keV to 2 MeV. The dose rate range covers 6 decades from 50 nSv h^{-1} to 10 mSv h^{-1}. All devices were located within 5 m from each other. The measurements represent the total γ dose rate. They include a terrestrial component, mainly from ^{40}K, ^{238}U, ^{232}Th and their progeny, a cosmic component, principally muons, an anthropogenic component, largely from ^{137}Cs, and an inherent background of the measurement devices. The terrestrial natural γ dose rate was obtained by subtraction of all the other components.

Terrestrial dose-rate data have also been reported from the REA monitoring network, the MARNA map (Quindós Poncela et al., 2004), which has a spatial resolution of 1 km, and the European radon map (Szegvary et al., 2007a). The REA Network has an automatic radiological station (ERA) at each site which consists of dual Geiger-Müller tubes, model BAI9305 from the Berthold company, to observe both low and high γ doses. Two data series were extracted from the REA Network. One is directly provided by the Spanish Nuclear Safety Council (CSN) and the other has been corrected according to a past optimization quality study carried out by Sáez Vergara et al. (2002). This last correction was effectively realized by a comparison between the reference CIEMAT dose-rate

Table 1
Overview of the site characteristics. Latitude and longitude are reported in decimal degree together with site height. Present average humidity and temperature conditions during the measurement campaign were also recorded.

Site	Latitude	Longitude	Height (m)	R.H. (%) min–max	T (°C) min–max
Teruel	40.4505	−1.0587	1082	33–36	30–33
Los Pedrones	39.4714	−0.9705	678	50–60	22–25
Q. de la Orden	39.7870	−3.5377	693	48–53	22–26
Madrid	40.5799	−3.6232	682	37–50	25–31

monitors, located at standard height of 1 m above the ground, and the REA dose-rate monitors.

2.3. ^{226}Ra activity

Soil samples were collected at each campaign station at different depths between 0 and 70 cm by rock sampling technique. The soil samples of 100 g in weight were then put into air-tight Petri dishes of 100 ml in volume. The Petri dishes were kept for 30 days to allow the ^{226}Ra to reach secular equilibrium with its short-lived progeny. The soil samples were then analyzed at the INTE-UPC and at the UHU laboratories by ^{214}Pb and ^{214}Bi radium progeny γ spectrometry. The ^{226}Ra activity was also reported from the MARNA map. This method, as opposed to spectrometry, uses acquired terrestrial γ dose from all the natural radionuclides (D_{tot}) to obtain the ^{226}Ra contribution to the dose (D_{Ra}) by the empirical equation, $D_{Ra} = 0.24 * D_{tot}^{1.01}$. This result is then employed to derive the radium activity in the soil using the hypothesis that ^{226}Ra activity is closely related to ^{238}U activity (Quindós Poncela et al., 2004).

2.4. Radon flux

The radon flux at the selected Spanish locations was measured during this campaign by four different systems; two integrating systems and two continuous monitors. The first were the electret radon flux monitor and the activated charcoal method. The continuous devices were an AlphaGUARD (Genitron Instruments GmbH, Frankfurt, Germany) and a Sun Nuclear model 1027. All these integrated and continuous systems are based on the well known accumulation method (Morawska and Philips, 1980). The evolution ^{222}Rn concentration in the chamber can be measured by continuous monitoring or estimated after a given time period by the integrated systems.

A commercial version of a radon flux monitor (E-PERM) has been used in this study with short-term, ST, electret and H chamber of 960 ml volume. This is a hemispherical dome with a 15 cm diameter Tyvek window (Kotrappa et al., 1993, 2004). It works as an integrating ionization chamber with an electrically charged electret on its internal top. The Tyvek window allows the radon gas exhaled from the soil to enter the electret ion chamber over a time period *T*. The electret voltage drops in relation to the total ionized air due to the radon decay inside the chamber. The electret system response is not influenced by the environmental temperature and humidity conditions as has been tested in different works such as in Vargas and Ortega, 2006.

The single device scheme is illustrated in Fig. 1 as representative of all accumulation methods. A number of 8 H chambers were used in this campaign at each site, integrating over 6 h measurement time. Three of them were used to subtract the radioactive background. The chambers were located at 50 cm distance from each other, covering a total area of 1 m^2.

The other integrating system used to determine the radon flux, involved ^{222}Rn adsorption on activated charcoal, has been explained in detail elsewhere (Countess, 1976; Fremman and Hartley, 1986; Dueñas et al., 2007). The charcoal response is influenced by the environmental conditions as previous studies have shown (Ronca-Battista and Gray, 1988; Vargas and Ortega, 2006). The ^{222}Rn collector is placed on the soil surface to be accumulate ^{222}Rn over a time period *T*. ^{222}Rn is then determined through its progeny ^{214}Pb (295 KeV and 352 KeV) and ^{214}Bi (609 KeV) in secular equilibrium conditions by gamma spectrometry (Kaplan, 1963). A number of 3 charcoal canisters were exposed at each site during 6 h and covering a total surface area of around 0.5 m^2. Each charcoal canister was made of a cylindrical box with a circular surface of 10 cm diameter and 1 cm high. The canisters were located directly on soil surface, avoiding void volume between this and absorbent material. The ^{222}Rn activity concentration accumulates inside a closed volume which is placed on the soil surface. The ^{222}Rn gas, in the chamber volume was measured continuously by the AlphaGUARD monitor (Genitron) and the Sun Nuclear model 1027 monitors. The Sun Nuclear monitor is a patented electronic detecting device using a diffused-junction photodiode sensor to measure ^{222}Rn gas concentration by radon progeny electrodeposition (López-Coto et al., 2009). The monitor was located directly inside the accumulation volume and the radon flux was measured by ^{218}Po (6 MeV) alpha spectrometry during 6 h at one point in each measurement location. The AlphaGUARD monitor was placed near the accumulation volume chamber. The sampling air was pumped inside the monitor at a flow rate of 0.3 l/min and concentration values were provided for each 10 min interval. A small 1-l plastic bottle was used to prevent aerosols and ^{220}Rn from entering into the AlphaGUARD (Lehmann et al., 2003). The AlphaGUARD have been acquiring during 2 h at 3 different points at each measurement location.

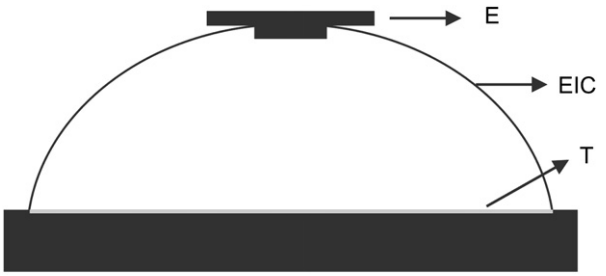
The four different systems were located close to each others, covering a circular surface area with a radius of about 5 m. Table 2 summarises the device characteristics and their geometrical position during the campaign measurements.

The ^{222}Rn emitted from the soil surface is accumulated during a time period (T) in each integrated and continuous known volume monitor (De Martino and Sabbarese, 1997). The temporal variation of the ^{222}Rn concentration in the chamber is expressed by the follow equation:

$$\frac{dC(t)}{dt} = \frac{E_{Rn}}{V_u} - \lambda^0 C(t) \quad (1)$$

where $C(t=0) = 0$ is the initial concentration (Bq m^{-3}); E_{Rn} is the exhalation velocity which is defined as the ^{222}Rn gas quantity leaving the soil in the time unit (Bq h^{-1}); V_u is the available chamber volume (m^3) and the constant $\lambda^0 = \lambda + \lambda^*$ (h^{-1}) is given by the sum between the ^{222}Rn decay (λ) and the ventilation constants (λ^*). The λ^* quantifies the possible changes of ^{222}Rn with external air because of leaks in the chamber. Another physical factor which may be taken into account in the ^{222}Rn accumulation is called back-diffusion and it means the possibility of ^{222}Rn being adsorbed back from the soil surface. This last factor is not significant for short-time measurements (Morawska, 1989) as applied in our analysis. The solution of the Eq. (1) is:

$$C(t) = \frac{E_{Rn}}{\lambda^0 V_u}\left(1 - e^{-\lambda^0 t}\right) \quad (2)$$



E : Electret
T : Tyvek window
EIC : Electret Ion Chamber

Fig. 1. Scheme of the ionization chamber type H (E-PERM) used for radon flux measurement. The ionization chamber (EIC) is located on the ground (black space) and the exhaled radon can enter through the Tyvek window (T). The radon concentration is proportional to the potential drop due to radon decay on the positively charged electret (E) located on the chamber top in the air chamber.

Table 2
Overview on characteristics of direct and indirect measurements methods to determine ^{222}Rn flux from soil.

Method	Principle	Technique	N. of device/ measurement replicates	N. of location for each measurement site	Single measurement period (h)	Reference
Direct methods						
Charcoal	Accumulation of ^{222}Rn in charcoal canister;	γ spectrometry	3/1	1 on ground surface	6	Dueñas et al., 2007
E-Perm	Accumulation of ^{222}Rn in chamber	Voltage discharge in Teflon	8/1	1 on ground surface	6	Kotrappa et al., 1993
AlphaGUARD	Accumulation of ^{222}Rn close volume	α spectrometry	1/3	1 on ground surface	1	Lehmann et al., 2003
Sun Nuclear	Accumulation of ^{222}Rn in closed volume	α spectrometry	1/1	1 on ground surface	4	–
Indirect methods						
GDR-1 Szegvary-RS112	Empirical relation with measured γ dose rate	dual Geiger-Müller	1/1	1 at 1m above ground	5	Szegvary et al., 2009
GDR-2 Szegvary-GammaTRACER	Empirical relation with measured γ dose rate	Ionization chamber	1/2	2 at 1m above ground	2	Szegvary et al., 2007b
^{226}Ra (INTE/UHU)	Empirical relation with ^{226}Ra concentration in soil	Laboratory γ spectrometry	1/1	1 at depth of 0–30 cm	1	Quindós Poncela et al., 2004; Nazaroff and Nero, 1988;
Indirect reference methods						
GDR Radon Map	Empirical relation with γ dose rate extracted from EURDEP database	–	–	–	–	Szegvary et al., 2007a
^{226}Ra MARNA map	Empirical relation with ^{226}Ra concentration in soil derived from MARNA project	–	–	–	–	Quindós Poncela et al., 2004; Nazaroff and Nero, 1988;

where value $E_{Rn}\lambda^{0^{-1}}V_u^{-1}$ (Bq m^{-3}) is the saturation concentration value exhaled in the air-tight chamber after almost 30 days. In the case of short-time measurements and negligible leakages in the chamber, $\lambda^0 t << 1$ can be assumed.

The previous Eq. (2) can be simplified by developing the exponential into Eq. (3):

$$C(t) = \frac{E_{Rn}}{V_u}t = \frac{F \cdot A}{V_u}t \quad (3)$$

Equation (3) describes a linear relation between the ^{222}Rn concentration in the chamber and time. F (Bq m^{-2} s^{-1}) is the radon flux and A is the surface area covered by the accumulation chamber. The linear method (Eq. (3)) was used for both continuous methods in the present study.

The radon fluxes by integrated systems have been measured using the average ^{222}Rn concentration inside the chamber above a given time T calculated by equation:

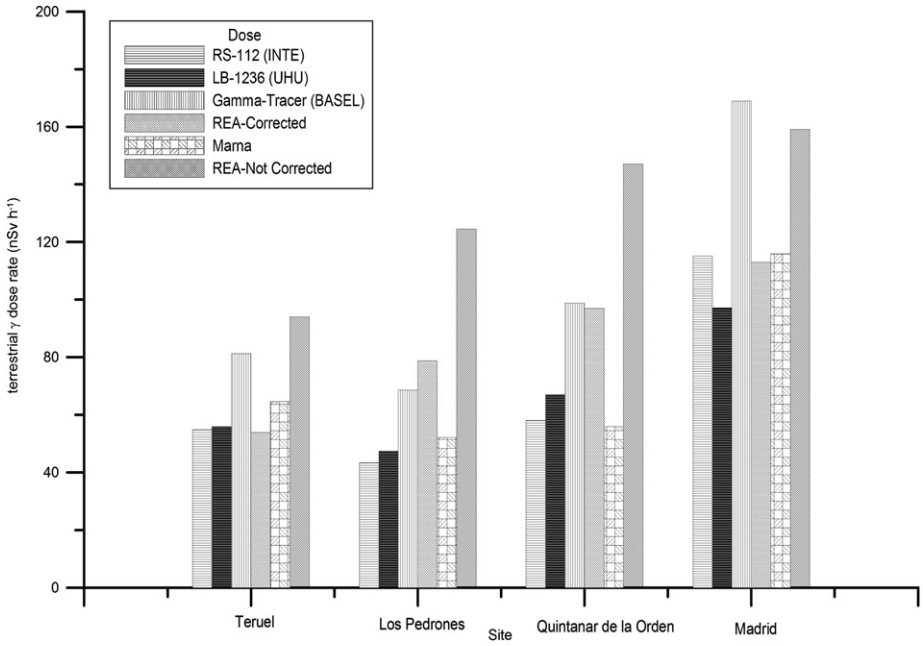


Fig. 2. The dose rate measured at each site by RS-112 (INTE), LB-1236 (UHU) and GammaTracer (BASEL) monitors are shown. The dose rate for each REA station and their corrected values are reported in according to Sáez Vergara et al. (2002). Furthermore, the values obtained from the MARNA map are also included.

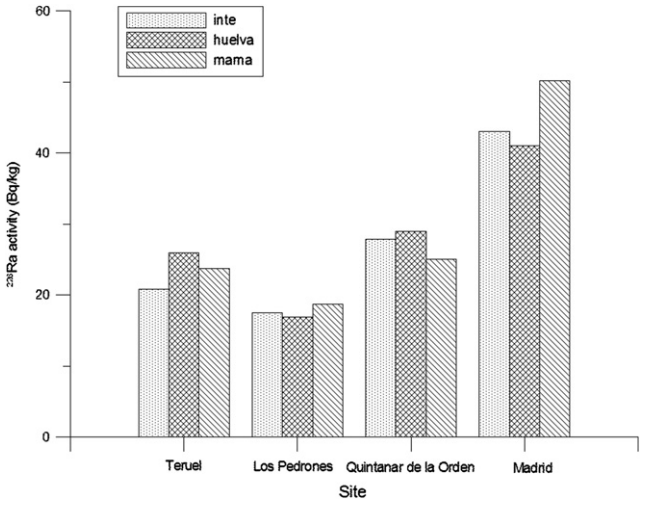


Fig. 3. Radium activity concentration measured by the INTE-UPC laboratory (pointed bar), UHU laboratory (vertical line bar) and that obtained by using the MARNA equation $D_{Ra} = 0.24^{*}\ D_{tot}^{1.01}$ (diagonal line bar) are shown.

$$\overline{C}(Rn) = \frac{1}{T}\int_0^T C(t)\mathrm{d}t = \frac{E_{Rn}}{\lambda^0 V_u}\left[1 - \left(\frac{1 - e^{-\lambda^0 T}}{\lambda^0 T}\right)\right] = \frac{F \cdot A}{\lambda^0 V_u}\left[1 - \left(\frac{1 - e^{-\lambda^0 T}}{\lambda^0 T}\right)\right] \quad (4)$$

From Eq. (3) and Eq. (4) the radon flux F can be easily derived, for the continuous and the integrated methods, respectively.

Radon fluxes have also been estimated using two indirect methods. The first method (GDR in Table 2) consists of the empirical equation $y = 0.89x - 11.01$, derived by Szegvary et al., 2007b. The previous relationship has been extrapoled using local γ-dose rate data, that have a resolution depending on European Radiological Data Exchange Platform, EURDEP (http://eurdep.jrc.ec.europa.eu/) data (Szegvary et al., 2007a). The y, in the empirical equation, represent the estimated radon flux (Bq m^{-2} h^{-1}) and x is the available terrestrial γ dose in nSv h^{-1}. The γ dose-rates measured by RS-112 (GDR-1) and by the GammaTRACER (GDR-2) have been used as input data. Furthermore, the reference Radon map method, GDR, was analyzed.

The other indirect method (^{226}Ra in Table 2) is based on the diffusion equation $F = C_{Ra}\,\lambda f \rho\,[Dc/(\lambda\epsilon)]^{1/2}$ (Nazaroff and Nero, 1988), where C_{Ra} is the ^{226}Ra activity (Bq kg^{-1}), F is the radon flux, f is the emanation material coefficient, ρ is the soil density, D_c is the bulk material diffusion coefficient, which is influenced by soil characteristic and conditions (T and HR), and ϵ is the material porosity. Typical geological parameters, for soil characteristics similar to the soil sites campaign, have been chosen for f (0.23), ρ (1.5 10^3 kg m^{-3}), D_c (2.0 10^{-7} m^2 s^{-1}) and ϵ (0.25) (Nazaroff and Nero, 1988). ^{226}Ra activity was obtained from the MARNA dose rate data and its conversion to activity concentration using the equation described in Section 2.3. The indirect methods and their comparative applications within this study are summarised in Table 2.

3. Results and discussion

In the following section the obtained results for the terrestrial γ dose rate, ^{226}Ra activity and the radon flux are presented and discussed.

3.1. Terrestrial γ dose rate

Fig. 2 shows the terrestrial dose rate results obtained by the different equipment and methods. The measured data from the RS-112 (INTE-UPC) and the LB-1236 (UHU) monitoring were in good agreement with the MARNA map for each campaign site. The measured value from the uncorrected REA is too high in comparison with the other measured values. However, the same REA are significantly closer to the other measured values after the correction developed by Sáez Vergara et al. (2002). The values measured by GammaTRACER tended to be higher than the other data.

3.2. ^{226}Ra soil activity mass

^{226}Ra soil activity mass in soil samples from each campaign site was analyzed at the INTE-UPC and the UHU laboratories. The ^{226}Ra

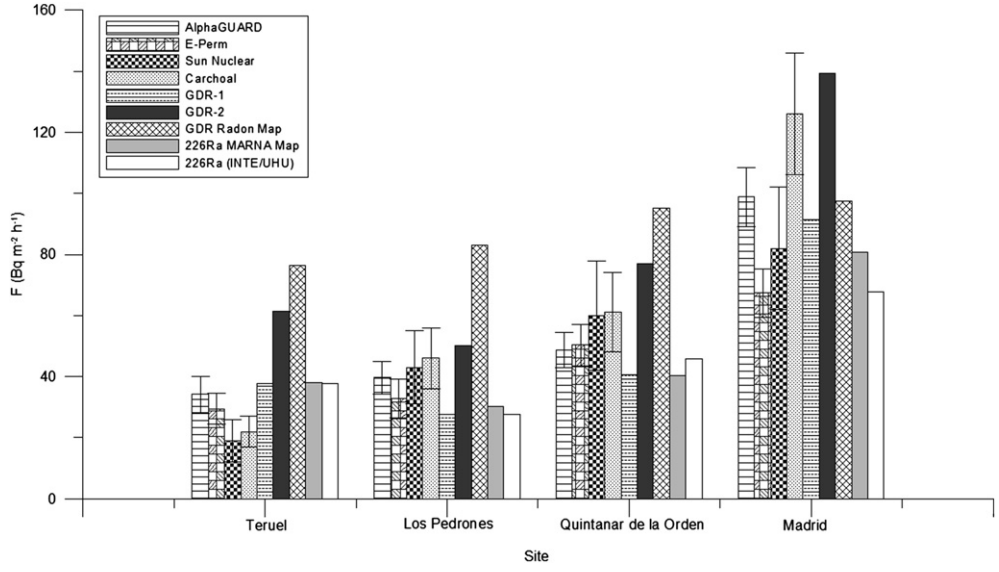


Fig. 4. Radon flux levels measured at each site by direct and indirect methods are presented. Values measured by the continuous Sun Nuclear (UHU) and AlphaGUARD (BASEL) monitors, and by the integrated detectors E-PERM (INTE) and charcoal (UHU), are compared. Radon flux (y) estimated in order to apply the equation proposed in Szegvary et al. (2007a),$y = 0.89x - 11.01$, with dose rate (x) data from measurements by EURDEP data (GDR), RS-112 (GDR-1) and GammaTracer (GDR-2) are also reported. Furthermore, radon flux is also estimated using the equation $F = C_{Ra}\ \lambda\ f\ \rho\ [Dc/(\lambda\ \epsilon)]^{1/2}$ at each site with radium concentration obtained from MARNA project (^{226}-MARNA map) and by laboratories measurement (^{226}Ra INTE/UHU).

activity results in the soil samples from the first terrain layer, which ranges between 0 and 30 cm, were compared as shown in Fig. 3. ^{226}Ra activities were also measured for deeper soil sampled, until 70 cm, showing a quite homogeneous distribution. The radioactivity concentrations measured at aforementioned laboratories are in agreement within a confidence level of 95%. An evaluation was also done between previous data and the ^{226}Ra activity calculated by the MARNA empirical equation (Quindós Poncela et al., 2004). These last values were within a range of ±5% of the directly measured ^{226}Ra activity average for Teruel, Q. de la Orden and Los Pedrones soil samples. The results show less agreement for the Madrid soil sample, where the MARNA ^{226}Ra activity was within a range of ±15% of the directly measured average concentration. In order to explain this difference, the gamma spectrum obtained form the soil samples in Madrid was analyzed. The analysis showed a high activity concentration of ^{40}K (1200 Bq kg^{-1}) and of ^{228}Ra (50 Bq kg^{-1}) at the Madrid site, which could influence the reliability of the empirical equation presented in Section 2.3. The ^{40}K and the ^{226}Ra activity at the other sites have a mean value, respectively, of 300 Bq kg^{-1} and 23 Bq kg^{-1}, which are on average expected in Spain (UNSCEAR, 2000). Therefore, the high activity found in Madrid undermines the hypothesis to obtain at least 65% of contribution from ^{226}Ra. It could explain the difference between MARNA derived values and campaign results observed in Madrid.

3.3. Radon flux

The results of radon flux data by direct measurements and indirect methods are shown in Fig. 4. The plot shows the observations by different campaign partners. Radon flux data calculated by the equation obtained from Szegvary et al. (2007a), and by the diffusion equation methods, both described in Section 2.4, are also reported. The data measured by continuous monitors, AlphaGUARD and Sun Nuclear, and by integrated methods, EPERM and charcoal, show that the agreement among the participants was about 10% (SD/mean) in Los Pedrones and Q. de la Orden sites, and about 23% in Teruel and Madrid sites. The activated charcoal gives some differences, compared to the other systems, in Madrid and in Teruel sites. Nevertheless, these values fall within 2σ and they have not been rejected. Indirect radon flux estimation, obtained by applying the empirical equation by Szegvary et al. (2007a) in Section 2.4, were within a range of ±30% of the directly measured radon flux average when they were based on the terrestrial dose directly measured by RS-112 (GDR-1). These indirect radon flux estimates were larger than the direct radon flux measurements when we used dose-rates from the EURDEP (GDR) database or from GammaTRACER (GDR-2) measurements. This is due to the high dose-rate values shown in Fig. 2. Indirect radon flux values, obtained by applying the MARNA equation in Section 2.4 were within 20% and 40% of the directly measured radon flux averages when ^{226}Ra activity in the soils were obtained from literature (^{226}Ra MARNA map) and by laboratories measurements (^{226}Ra INTE/UHU), respectively.

4. Conclusion

Direct and indirect methods, for the determination of radon flux from soil have been used in an inter-comparison campaign carried out at four selected Spanish sites in summer 2008 under the most typical climatologic conditions for the eastern Spain. The geological characteristics of the object sites allowed observing a wide range of radon flux values ranging from 40 Bq m^{-2} h^{-1} and 90 Bq m^{-2} h^{-1}. Direct methods to determine radon flux, including both continuous and integrated systems, showed a good agreement with a coefficient of variation between 10 and 23%. This value is in accordance with the 34% found out in the study by Hutter and Knutson (1998). Indirect methods based on the measurement of terrestrial γ dose rate, or ^{226}Ra soil activity, and their empirical correlation with radon flux, have been applied and evaluated. Results show that these correlations give radon flux values within ±20–40% of the directly measured radon flux average when the ^{226}Ra activity and the terrestrial γ dose-rate measurements are accurately measured. This was observed for ^{226}Ra activity obtained either by MARNA map or by direct measurement at INTE and at UHU laboratories and when terrestrial dose rate was measured by RS-112 and by LB-1236 monitors. However, it was found that the terrestrial γ dose-rate values from the Automatic Spanish Surveillance network (REA) and those obtained by GammaTRACER resulted in an overestimation of radon flux. Finally, the influence of seasonal and daily environmental conditions fluctuations on radon flux values are going to be studied in a new campaign which will be carried out during 2010.

Acknowledgement

This study was supported by the Swiss National Science Foundation (grant no. 200020-117622/1) to FC.

Thank to Cristina Parages and José Carlos Saez-Vergara, from the CSN and from the CIEMAT respectively, who helped us to enter into the REA stations during the measurements campaign.

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