QUALITY STUDY OF ELECTRET RADON FLUX MONITORS BY AN "IN SITU" INTERCOMPARISON CAMPAIGN IN SPAIN

C. GROSSI, A. VARGAS, D. ARNOLD
Institute of Energy Technologies (INTE),
Technical University of Catalonia Barcelona,
Spain

Abstract

Radon is nowadays used as a tracer for atmospheric transport model validation because of its physical and chemical characteristics. ^{222}Rn source term characterization becomes necessary for more accurate validations into the current non-stopping progression of atmospheric dispersion model development. Direct measurements of radon exhalation flux density are carried out in order to obtain a reliable map with required spatial and temporal resolutions. To measure radon exhalation flux density, the INTE (UPC) institute uses an integrated electret ionization chamber (EIC) system, also known as electret radon flux monitors. Different configurations of this device have been developed by the manufacturer and used in a comparison campaign held in the summer of 2008. In the comparison campaign, of radon exhalation flux density, measurement was carried out at four locations of the eastern Spain. The electret radon flux monitors response in different soils and with different environmental conditions was analyzed. The different electret radon flux monitor chambers show coherent results in comparison to the other integrated and continuous radon flux systems which also participated in the campaign.

Introduction

^{222}Rn is one of the radioactive daughters of ^{238}U, which is always present in soil and rocks with different concentrations. Being a noble gas, ^{222}Rn can migrate through soil pores and be finally released into the atmosphere without interacting with other chemical species during its pathway. It is liberated from the soil with variable rates, depending on soil characteristics and on external environmental parameters.

Radon gas is nowadays used as a tracer for atmospheric transport model validations because, as it is an inert gas, its concentration in air is uninfluenced neither by wet or dry depositions nor by any other chemical processes. Radioactive decay is the only removal process. Another advantage of using radon as a tracer is that it is natural occurring and it is continuously exhaled into the atmosphere. Thus, its application avoids the use of artificial releases for model validation purposes.

Besides its usefulness in atmospheric sciences, radon exhalation rate estimates also provide basic information to characterize radon-prone areas for radioprotection aims.

Thus, it is clear that the ^{222}Rn source term characterization from the land surface becomes necessary for more accurate validations into the current non-stopping progression of atmospheric dispersion model development.

Many studies have been done and are still continuing to improve ^{222}Rn source estimation according to spatial and temporal variations [1]. The scientific community is now working on developing a radon source term map which would supply input data for atmospheric transport modelling at global and local scales. Simultaneously, research for improving both integrated and continuous methods, to directly measure radon flux, is being pursued.

In this study the quality of different integrated Electret Ion Chamber (EIC) monitor geometries has been analysed. The EIC are integrated devices which are able to measure radon exhalation from the sampled soil directly "in situ". These are commercial E-Perm instruments by the Rad. Elec. Inc. Company.

In order to compare EIC performance with other direct radon exhalation flux density measurement methods, a joint comparison campaign has been carried out in Spain [2] by the Institute of Energy Technology (INTE) of the Technical University of Catalonia (UPC), Basel University and Huelva University (UHU).

Direct methods for radon flux estimations

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 [1]. In spite of sufficient understanding of the theoretical processes controlling the release of ^{222}Rn from soil to the atmosphere [3], quantification of radon exhalation flux density and its distribution over the earth is still lacking because of a lack of direct and extensive ^{222}Rn exhalation flux density measurements in many regions.

Direct measurements of ^{222}Rn exhalation flux density are made using the accumulation method which allows radon gas to accumulate and to be measured in a chamber placed over the soil. Likewise, radon exhalation flux density can be directly measured both by integrated and continuous instruments which are based on the well-known accumulation method by Morawska [4]. This method is based on the accumulation of the ^{222}Rn emitted from the sampled soil surface in a known volume monitor during a time period (T). Temporal variation of the ^{222}Rn concentration in the chamber is expressed as follows (Eq. (1)):

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

where C(t=0) = 0 is the initial Rn concentration (Bq m^{-3}); E_{Rn} is the exhalation velocity, defined as the ^{222}Rn gas leaving the soil per 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 constant (λ^*). λ^* quantifies the possible changes between the air inside the monitor with external air due to volume leaks. Another physical factor which should be taken into account in ^{222}Rn accumulation is the so-called back-diffusion, which accounts for the possibility of ^{222}Rn being adsorbed back from the soil surface. This last factor is not significant for short-time measurements as applied in our analysis [10]. The solution of Eq. (1) is Eq. (2):

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

The 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 20 days. In the case of short-time measurements and negligible leakages in the chamber, $\lambda^0 t \ll 1$ can be assumed.

Continuous monitor for radon exhalation flux density measurements are based on the simplification of Eq. (2) by developing the exponential, leading to Eq. (3):

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

This gives a linear relation between the ^{222}Rn concentration in the chamber and time. F (Bq m^{-2} s^{-1}), is the ^{222}Rn exhalation flux density, A, the surface area covered by the accumulation chamber and V the available volume for the gas diffusion inside the monitor.

With integrated monitors for radon exhalation flux density measurement, F is calculated using the average ^{222}Rn concentration (Eq. (3)) inside the chamber over a given time T calculated by Eq. (4):

$$C(Rn)Av = \frac{1}{T}\int_0^T C(t)dt = \frac{E_{Rn}}{\lambda^0 V_u}\left[1 - \left(\frac{1 - e^{-\lambda^0 T}}{\lambda^0 T}\right)\right] = \frac{FxA}{\lambda^0 V_u}\left[1 - \left(\frac{1 - e^{-\lambda^0 T}}{\lambda^0 T}\right)\right] \tag{4}$$

The integrated electret ion chamber

^{222}Rn concentration in the chamber can be estimated after a given time period by integrated and passive EIC monitors. These are commercial monitors which allow measurement of the radon concentration inside a closed chamber. The monitor works as an integrating ionization chamber. The radon gas exhaled from the soil enters directly into the chamber through a Tyvek window. Inside the chamber, radon decays and the resulting ionized air discharges the positive electret voltage (700 V), which is located on the top of the monitor. The voltage drop during a measurement time (T) is proportional, by a calibration factor [7], to the radon concentration inside the chamber.

During the radon exhalation flux density measurement, completely sealed EIC monitors are used to obtain the background contribution. The sealing of the device prevents air infiltration inside the chamber, and the discharging of the electret on the top is then just due to the environmental gamma radiation, which is considered to be the background contribution to the measurements. The background contribution is subtracted to calculate net flux result.

The sensitivity and the dynamic range of the radon exhalation flux density monitors depend on the thickness of the electret and on the chamber volume. Several combinations of electret and chambers are available. A single device scheme is illustrated in Figure 1, with a short-term ST, electret and a hemispherical chamber of 960 ml volume, H, in the so-called HST configuration.

This EIC monitors are quite inexpensive and different monitors simultaneously can be used during a measurement campaign at each site in order to get more accurate measurements. Furthermore, the voltage drop is easily measured by a portable voltage reader. The commercial availability of the stable electrets of different thicknesses, suitable chambers and a low-cost electret voltage reader has made it practical to have a viable EIC system.

Electret ion chambers (EIC) have been used for the measurement of gamma radiation since 1978 [7]. This method has not been widely used, however, because of the possible errors introduced by environmental factors such as temperature and humidity on the electret stability. These problems have been solved [8].

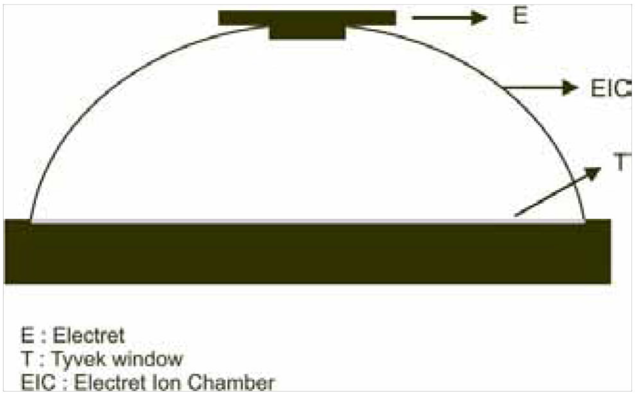

FIG. 1. Scheme of an Electret Ion Chamber on the soil surface.

EIC monitors are available with two different geometries. The scheme illustrated in Figure 1 needs to be placed directly on the top of the soil surface. In this case a paper towel has to be located under the monitor to prevent the Tyvek filter being contaminated by the soil. Another prototype monitor has been equipped with an aluminium collar, as is shown in Figure 2, which should, in principle, facilitate the installation of the monitor within the soil and avoid filter contamination by some stops.

The radon concentration after a time T is measured by the electret ion monitors using equation (4), where the exponential concentration increase is compensated by the radon decay and possible chamber leaks. This last factor has a non-negligible contribution to the device reliability. The ventilation constant for the EIC monitors has been certified to be equal to zero by the manufacturer [7].

FIG. 2. Radon Flux monitors with aluminium collars for in-soil installation [7].

Intercomparison campaign for EIC monitor quality study

Electret ion chamber consistency was observed under different environmental conditions and for different soil types. Additionally, the EIC monitor results have been compared with other direct methods within an intercomparison campaign. This measurement campaign has been carried out at four Spanish sites: Teruel, Los Pedrones, Quintanar de la Orden and Madrid in summer 2008 [2].

Continuous measurement devices such as the AlphaGUARD (Genitron Instruments GmbH, Frankfurt, Germany) and the Sun Nuclear model 1027 have been used for radon exhalation flux density measurements by Basel and the Huelva University, respectively. In these monitoring devices, the ^{222}Rn activity concentration is accumulated inside a closed volume, placed on the sampled soil surface, and it is then continuously measured by alpha spectrometry.

The AlphaGUARD monitor is placed near the accumulation volume chamber. The sampling air is pumped inside the monitor and concentration values are provided for each 10-min interval. A small 1-litre plastic bottle is used to prevent aerosols and ^{220}Rn from entering the AlphaGUARD [9] and [13].

The Sun Nuclear monitor is located directly inside the accumulation volume and the radon flux is measured by ^{218}Po (6 MeV) alpha spectrometry [12].

Integrated activated charcoal detectors have also been used for radon exhalation flux density measurements in the campaign study. These monitors are based on ^{222}Rn adsorption on activated charcoal [6]. The charcoal is placed on the soil surface to accumulate ^{222}Rn over a time period T. ^{222}Rn is then determined through γ spectrometry [6] of its progeny, ^{214}Pb (295 keV and 352 keV) and ^{214}Bi (609 keV) which are in secular equilibrium with radon.

Finally, the two different EIC monitor geometries described in the previous section were used in the present work. Particularly, three monitors in the HST configuration and with aluminium collars, two monitors in the HST configuration and without aluminium collars and three background monitors were used at each campaign site.

During the campaign, the atmospheric temperature and relative humidity conditions ranged from 20ºC to 35ºC and from 35% to 60%, respectively.

Results for the EIC detectors in the intercomparison campaign in eastern Spain

Figure 3 shows the radon exhalation flux density result, measured at each site by the five different EIC monitors. Results at each of the sites are in agreement and they fall into one-sigma. Radon exhalation flux density measurements done by EIC monitors with aluminium collars (C1, C2 and C3) seem to be slightly less stable than others made by monitors without collars (1, 2), maybe due to some volume definition error at the soil installation.

The weighted average value of EIC monitor results compared with radon exhalation flux density results from other integrated and continuous direct methods is presented in Figure 4. EIC monitors (vertical and green bar) show good agreement with the charcoal integrated system (pointed and red bar) and with the continuous monitors Sun Nuclear (pointed and blue bar) and AlphaGUARD (diagonal and white bar). A coefficient of variation between 10% and 23% was calculated between the two monitors. This value is in accordance with the 34% found in previous studies [10].

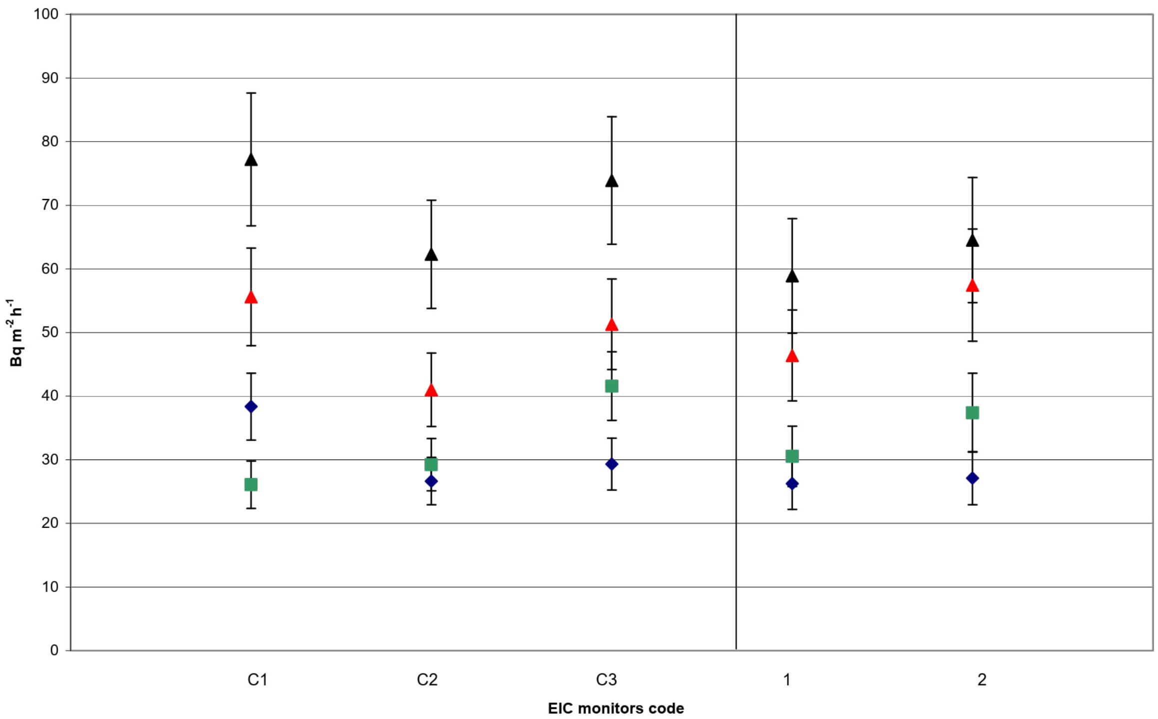

FIG. 3. Radon exhalation flux density results at Teruel (blue), Quintanar de la Orden (red), Los Pedrones (green) and Madrid (black) by EIC monitors with aluminium collars (C1, C2 and C3) and without collars (1, 2).

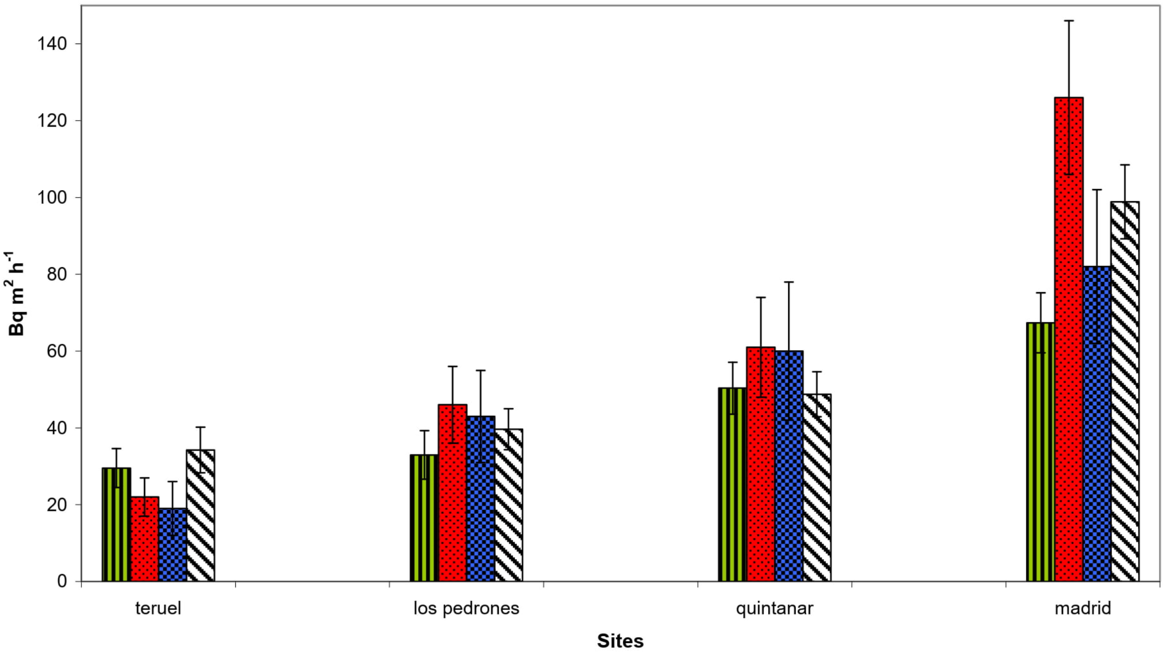

FIG. 4. ^{222}Rn exhalation flux density results at each campaign site measured by EIC (vertical and green bar), charcoal (pointed and red bar), Sun Nuclear (square and blue bar) and Alpha GUARD (diagonal and white bar) monitors.

Preliminary EIC monitor calibration

The EIC monitors, in the HST configuration, seem to have the most appropriate technical and economical features for their utilization during radon exhalation flux density site characterization. Furthermore, the EIC chambers without aluminium collars are easier to install on soil and their utilization does not lead to volume definition errors (Figure 3).

Although these monitors have shown quite good results, they are still prototype devices and more accurate calibration is needed in order to define the ventilation factor contribution during a measurement. A first monitor calibration has been performed at the Huelva University laboratory through the bed exhalation method using a well-characterized terrain with a known ^{226}Ra content of 50 Bq Kg^{-1}. The reference terrain exhalation has been calculated by the potential exhalation method [13].

After three different measurements performed by the two EIC monitors in the HST configuration and without collars, a $\lambda^0 = \lambda+\lambda^*$ of 0.20 h^{-1} has been found.

In Figure 5 the results of EIC monitor in the HST configuration, without aluminium collars are reported taking a ventilation factor of 0.20 h^{-1}.

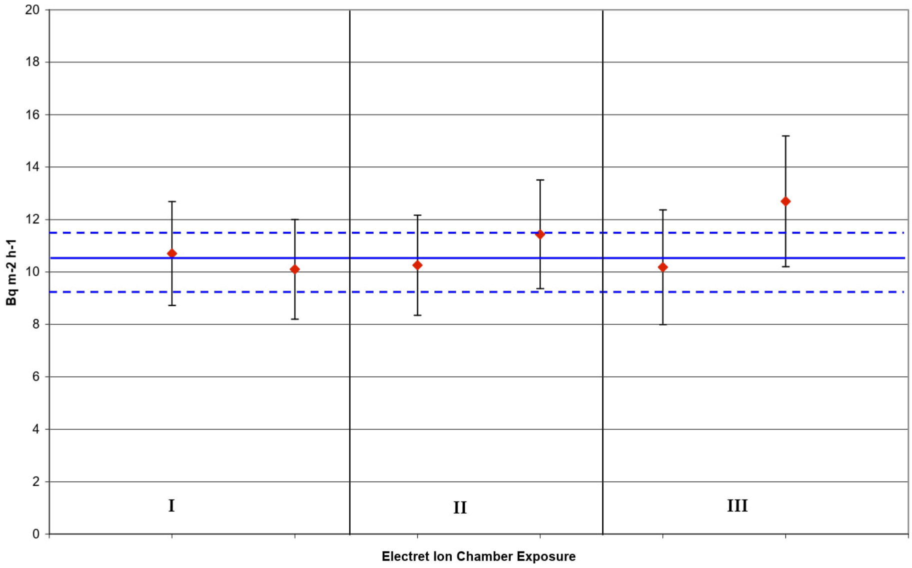

FIG. 5. Radon exhalation flux density results by EIC monitors, with a ventilation lambda of 0.2 h^{-1}, during a preliminary calibration study. Three different measurements have been performed and compared with the reference exhalation terrain.

Figure 6 presents the exhalation campaign results for the EIC monitors with and without ventilation correction at each site, only for the monitors without aluminium collars. These results are compared with radon flux values measured by AlphaGUARD monitors, which has been taken as a reference monitor. Radon exhalation flux density values by EIC monitors taking into account the ventilation factor of 0.20 h^{-1}, are also in agreement with the values measured with the AlphaGUARD. Therefore, research into this field should continue.

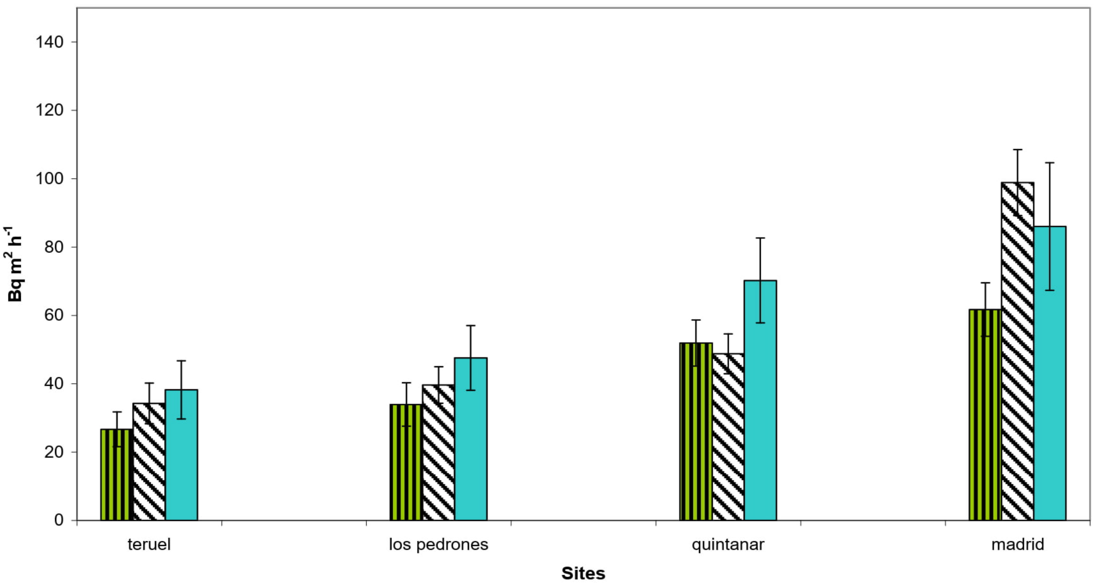

FIG. 6. The radon exhalation flux density results by EIC monitors with ventilation lambda equal to zero (vertical green bar) are compared with values taking into account the ventilation lambda (0.20 h^{-1}) (sky blue bar). Radon flux values by the AlphaGUARD method are also reported as a reference (diagonal white bar).

Conclusion

The intercomparison campaign and study shows that the EIC work properly and are in agreement with other direct methods, both integrated and continuous, for ^{222}Rn exhalation flux density measurements. In order to evaluate the quality of the EIC system, the results of an intercomparison campaign carried out at four eastern Spanish sites has been used.

Under standard environmental conditions the electret system works properly. However, the electret system could be strongly influenced by humidity condensation in the EIC chamber, which leads to electret passive discharge. Therefore, measurements with this device should not be advised when water condensation is likely. The intercomparison campaign was carried out during solar hours to avoid dew.

Mainly, it can be concluded that EIC monitors are appropriate in field campaigns due to their performance, mobility and price. The EIC monitors without aluminium collars are more practical to install because they do not have associated definition volume mistakes.

It has also been found that a crucial point for future performance analysis is the definition of a reference exhalation source in order to calibrate and carry out radon exhalation flux density detectors. Further work is planned with Huelva University to get a better quality control procedure.

Acknowledgement

Thanks to Cristina Parages and José Carlos Saez-Vergara, from the CSN and from the CIEMAT respectively, who helped us at the REA stations during the measurement campaign. Thanks to Israel Lopez-Coto, from Huelva University, for the preliminary radon flux monitor calibration. This study was partially supported by the Swiss National Science Foundation (grant no. 200020–117622/1) to FC.

REFERENCES

[1] WMO - 1st International expert meeting on sources and measurements of natural radionuclides applied to climate and air quality studies. Technical report No 155, World Meteorological Organization Global Atmosphere Watch, (2004).

[2] GROSSI, C., VARGAS, A., CAMACHO, A., et al., Inter-comparison of different direct and indirect methods to determine ^{222}Rn flux from soil, submitted to Appl. Radiat. (July 2009).

[3] PORSTENDORFER, J., Properties and behaviour of radon and thoron and their decay products in air, J. Aerosol Sci. **25** (1994) 219–263.

[4] MORAWSKA, L., PHILLIPS, C.R., Determination of the radon surface emanation rate from laboratory emanation data. Sci. of The Total Environ. **106** (1980) 253–262.

[5] LEHMANN, B.E., IHLY, B., SALZMANN, S. et al., An automatic static chamber for continuous ^{222}Rn and ^{222}Rn flux measurements from soil. Radiat. Meas. **38** (2003) 43–50.

[6] DUEÑAS, C., LIGER, E., CANETE, S., et al., Exhalation of ^{222}Rn from phosphogypsum piles located at Southwest of Spain, J. Environ. Radioact. **95** (2007) 63–74.

[7] KOTRAPPA, P., DEMPSEY, J.C., STIEFF, L.R., Recent advances in electret ion chamber technology for radiation measurements. Radiat. Prot. Dosim. **47** (4) (1993) 461–464.
[8] KOTRAPPA, P., STIEFF, L.R., Application of NIST ^{222}Rn emanation standards for calibrating ^{222}Rn monitors, Radiat. Prot. Dosim. **55** (1994) 211–218.
[9] SZEGVARY, T., CONEN, F., STOHLKER, U., et al., Mapping terrestrial gamma dose rate in Europe based on routine monitoring data, Radiat. Meas. **42** (2007) 1561–1572.
[10] DE MARTINO, S., SABBARESE, C., A Method for Emanation Coefficient Measurement of ^{222}Rn and ^{220}Rn from Soils, Phys. Chem. Earth **22** (1–2) (1997) 19–23.
[11] HUTTER, A.R., KNUTSON, E.O., An international inter-comparison of soil gas radon and radon exhalation measurements, Health Physics **74** 1 (1998) 108–114.
[12] KAPLAN, I., Nuclear physics II, Addison-Wesley, Japan (1963).
[13] KELLER, G., FOLKERTS, K.H., MUTH, H., Method for the determination of ^{222}Rn radon and ^{220}Rn Thoron, exhalation rates using alpha spectrometry, Radiat. Prot. Dosim. **3** 2 (1982) 83–89.
[14] LOPEZ-COTO, I., MAS, J.L., BOLIVAR, J.P., GARCIA-TENORIO R., A short-time method to measure the radon potential of porous materials, Applied Radiation and Isotopes **67** (2009) 133–138.