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Description of the behavior of an aquifer by using continuous radon monitoring in a thermal spa

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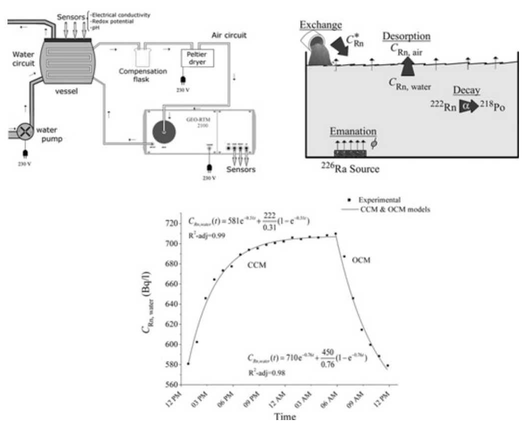
HIGHLIGHTS

- Radon in water is the major source of indoor air radon concentration in thermal facilities.
- Radon in water has been used to characterize the origin of water used for treatments in a spa.
- Preliminary dose assessment from radon exposure has been performed.

GRAPHICAL ABSTRACT



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ABSTRACT

Radon (^{222}Rn) levels in air and water have been analyzed continuously for almost a year in Las Caldas de Besaya thermal spa, north Spain. Radon is a naturally occurring noble gas from the decay of radium (^{226}Ra) both constituents of radioactive uranium 238 series. It has been recognized as a lung carcinogen by the World Health Organization (WHO) and International Agency for Research on Cancer (IARC). Furthermore the Royal Decree R.D 1439/2010 of November, 2010 establishes the obligation to study occupational activities where workers and, where appropriate, members of the public are exposed to inhalation of radon in workplaces such as spas. Together with radon measures several physico-chemical parameters were obtained such as pH, redox potential, electrical conductivity and air and water temperature. The devices used for the study of the temporal evolution of radon concentration have been the RTM 2100, the Radon Scout and gamma spectrometry was complementarily used to determine the transfer factor of the silicone tubes in the experimental device. Radon concentrations obtained in water and air of the spa are high, with an average of 660 Bq/l and 2900 Bq/m^3 respectively, where water is the main source of radon in the air. Radiation dose for workers and public was estimated from these levels of radon. The data showed that the thermal processes can control the behavior of radon which can be also influenced by various physical and chemical parameters such as pH and redox potential.

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1. Introduction

Radon (^{222}Rn) is a naturally occurring radioactive gas that has a half-life $T_{1/2}$ of 3.8 days. It is formed as the decay product of radium (^{226}Ra) with $T_{1/2} = 1600$ years, which is a member of the radioactive series of uranium (^{238}U) (Nazaroff and Nero, 1988). Uranium and radium found naturally in soil and rocks, provide a continuous source of radon. Because of its gaseous nature, radon is able to escape from the rock depending on the density and porosity thereof, being one of the most common radioactive elements in groundwater (Roba et al., 2012).

Under normal conditions, radon has a density of 9.73 kg/m^3, making it the densest gas of nature (International Agency for Research on Cancer, 1988). Of all noble gases, it is the most soluble in water. Owing to the nature of noble gas, its behavior is determined by physical processes. However its parent ^{226}Ra is highly reactive, it forms compounds as Ra^{2+}. Radium constitutes an efficient radon source when once dissolved, is absorbed by the surface of rocks and minerals of the aquifer (Surbeck, 2005), thus avoiding loss or diluting the concentration of radium in water during high flow processes or aquifer recharge.

Some hot springs have high concentrations of radium and radon. For example, studies in Spanish spas provide radon concentrations in water above 1800 Bq/l and 36 Bq/l for radium (Rodenas et al., 2008). The release of radon through water spas in the environment may be a risk to the health of workers of thermal installations as well as for patients (Welch and Mossman, 1994; International Agency for Research on Cancer, 1988).

When radon is inhaled, its disintegration products (^{218}Po and ^{214}Po) are deposited in the lungs, they emit alpha particles which could interact with biological tissues causing DNA damage (Anon., 2009). Due to its gaseous nature, which makes it possible to build up in enclosed spaces such as homes, spas, caves or mines, reaching high concentrations. In 1988 radon was classified as a human carcinogen by the International Agency for Research on Cancer (IARC), agency specializing in cancer research within WHO, from epidemiological studies of uranium miners (International Agency for Research on Cancer, 1988). Currently radon is recognized as the second cause of lung cancer in the population after tobacco (Anon., 2009). According to United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), the average dose received by the Spanish population is 3.7 mSv/year, which 2.4 mSv are due to natural radiation (the global value) where 1.3 mSv are relevant to radon (UNSCEAR, 2000).

This study focuses on two objectives. Both are based on continuous measurement of dissolved radon (^{222}Rn) in water and radon concentration in air of thermal facility, as well as of physical and chemical parameters.

The main objective is based on the characterization of the source of radon in the indoor air in the thermal spa. To do this, dissolved radon in water as natural tracer was monitored to evaluate transfer dynamics to air, to determine the aquifer dynamics and find the radon sources in the thermal installation. The second objective focuses on radiological protection of the workers and patients from the risks derived of radon inhalation. In addition, the concentrations found in air are compared with reference levels detailed in Spanish legislation (IS-33 instruction): if the annual average radon concentration is lower than 600 Bq m^{-3} no specific control is needed; if it is between 600 and 1000 Bq m^{-3} it must be applied a low level of control (follow up of annual average concentration), and if it is higher than 1000 Bq m^{-3}, a high level control must be implemented, which can be related with administrative/technical interventions in order to reduce the exposure of workers (Consejo de Seguridad Nuclear, 2012).

2. Materials and methods

2.1. Site description

Las Caldas de Besaya thermal spa is located beside the river Besaya in the village Los Corrales de Buelna (43°17′53″N, 4°04′23″O) about 30 km from Santander, the capital of the autonomous community of Cantabria, Spain (Fig. 1). Thermal water of this spa is characterized by a temperature between 34 and 37 °C, a sodium-chloride composition, bicarbonated and nitrogenous (Agencia de Evaluación de Tecnologías Sanitarias (AETS) Instituto de Salud Carlos III — Ministerio de Sanidad y Consumo et al., 2006).

The rock type in this area has sedimentary origin, the most part gray limestone and dolomite (Robador et al., 2008). Although the susceptibility of Cantabrian lithologies to release radon is very low (Quindós et al., 1991), the thermal facility is located over an inverted basin geological fault called "Frente Cabalgante del Escudo de Cabuérniga", which runs parallel to the coast. Several natural springs of thermal water are present along this fault and its relationship with the presence of high radon concentrations in water, as well as a more detailed description of the structural geology in the Cantabria region can be found in (González-Díez et al., 2009).

The period of annual opening of the resort is usually from March to December. It has many services and treatment techniques as baths, jets, circular showers, bubble baths, inhalations, sprays, sauna, underwater massage and manual massage.

In the thermal spa there are seven hot springs. The connection between them remains unknown and could be an interesting topic for future studies. To supply services, water is regularly pumped (from

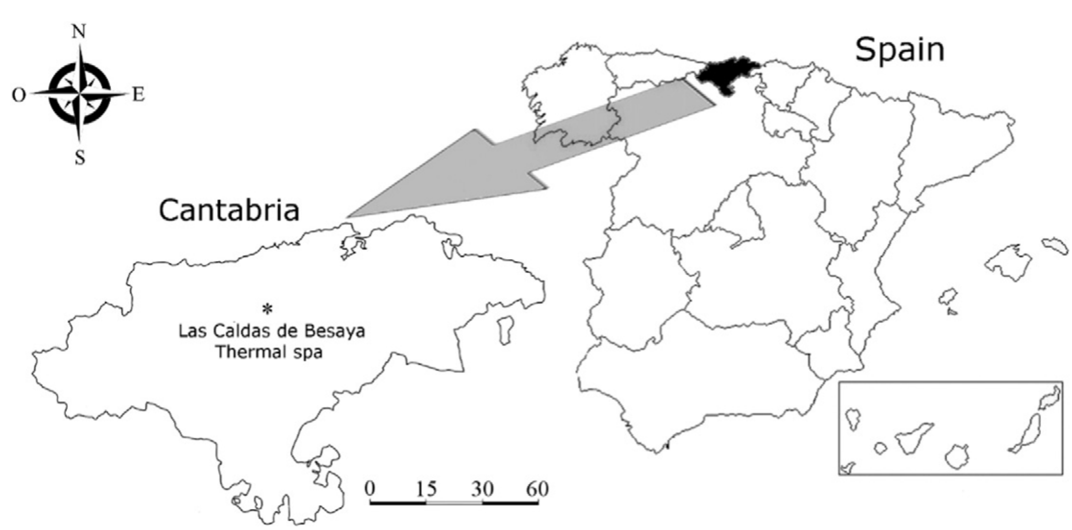

Fig. 1. Location of Las Caldas de Besaya thermal spa, Cantabria, Spain. 150 × 66 mm (300 × 300 dpi).

6:00 a.m. to 12:00 p.m.) from a well located outside of the installation, and part of this water is mixed with the water of the main well where measurements were performed.

2.2. Sampling and experimental device

Radon levels were determined inside thermal spa by measuring the gas concentration in water and air. Radon concentration in water, which outcrops at thermal spa, was measured every hour over five months, between March and August 2012. In the case of radon concentration in air the measures were extended until February 2013.

To determine the concentration of radon dissolved in water $C_{Rn,water}$ it has been used two methods. The first was performed continuously by the device RTM 2100 (Sarad GmbH, Germany) which scheme is shown in Fig. 2. The whole system consists in a closed air circuit by which air is continuously circulating through the semiconductor-based detection chamber of the RTM device by means of a low flow pump. The outside part of the air circuit is made of a silicone, which is highly permeable to radon. In addition, the measurement system of radon in water was studied in relation to diffusion tubes. The material and length of the tubes were changed in order to increase the efficiency of radon diffusion transfer.

When around 2 m of these tubes (3 mm of inner diameter and 1 mm of thickness) are submerged into the water contained in the vessel of 3 l volume, a given amount of radon dissolved in the liquid is passing by diffusion to the closed air circuit. Taking to account that the detection efficiency of RTM is highly sensitive to absolute humidity, and in order to avoid water condensation into the detector chamber, a Peltier dryer is placed nearby the entrance of air into the RTM device. Moreover, to prevent entrance of liquid water into the system in case of accident, a compensation bottle is also placed in front of the dryer.

The second method was based on grab sampling and determination by gamma spectrometry technique. Samples were collected every week, filling containers slowly in order to minimize water turbulences and subsequent radon losses by desorption. For the determination of ^{222}Rn concentration in water, hermetically sealed containers made from polyethylene were used for sample collection, and measured with a high-purity Ge coaxial detector, with a relative efficiency of 20%. Its resolution was 1.86 keV and it is logged inside a low activity iron casing. The samples were measured once the equilibrium between ^{222}Rn and its progeny was reached, 3 h after the sample collection, using the 609 keV photopeak of ^{214}Bi. The sampling and measurement procedures have been described in detail elsewhere (Rodenas et al., 2008).

Continuous measurement of radon concentration in air $C_{Rn,air}$ inside the spa were performed with the Radon Scout device (Sarad GmbH). Water samples were taken the main well while the Radon Scout device was located one meter away. Several physico-chemical parameters as electrical conductivity, redox potential and pH were taken together with radon concentration measurements by means of independent probes integrated in the measuring system. Water and air temperatures have also been monitored. The accuracy of radon measurement system was recently tested by participating on international intercomparison exercise (Gutiérrez et al., 2012).

2.3. Determination of transfer factor

Radon concentration provided by RTM 2100 is expressed in Bq/m^3 because this device measures radon concentration in the air circuit (see Fig. 2). In order to find a suitable transfer factor from radon in water to radon in air concentration, the above indicated gamma spectrometry measurements from grab samples were made. Although the transfer factor is dependent of water temperature (Cosma et al., 2008), it has been found that in case of the studied aquifer this parameter is practically constant, ranging less than 0.5 °C during the observed period. For this reason, the influence of temperature could be neglected in this study. Determinations by gamma spectrometry at the laboratory of environmental radioactivity of the University of Cantabria (LARUC) are continuously validated with ISO based quality assessment, as well as by participating successfully in international exercises of intercomparison (Gutiérrez et al., 2012). Therefore to obtain radon concentration in water expressed in Bq/l, the gamma spectrometry results have been compared with RTM 2100 ones. Additional low level radon concentration in water obtained from our laboratory where used to complete the relationship for a broader range of concentrations. Linear fit of Fig. 3 provides the transfer factor *tf*:

$$C_{Rn}(\text{gamma}) = tf \cdot C_{Rn}(\text{RTM}) \Rightarrow tf = (1,96 \pm 0,10)10^{-4}\ \frac{\text{Bq/l}}{\text{Bq/m}^3}. \tag{1}$$

This value of transfer factor is higher than the solubility coefficient of radon in water at around 36 °C (typical water temperature in the

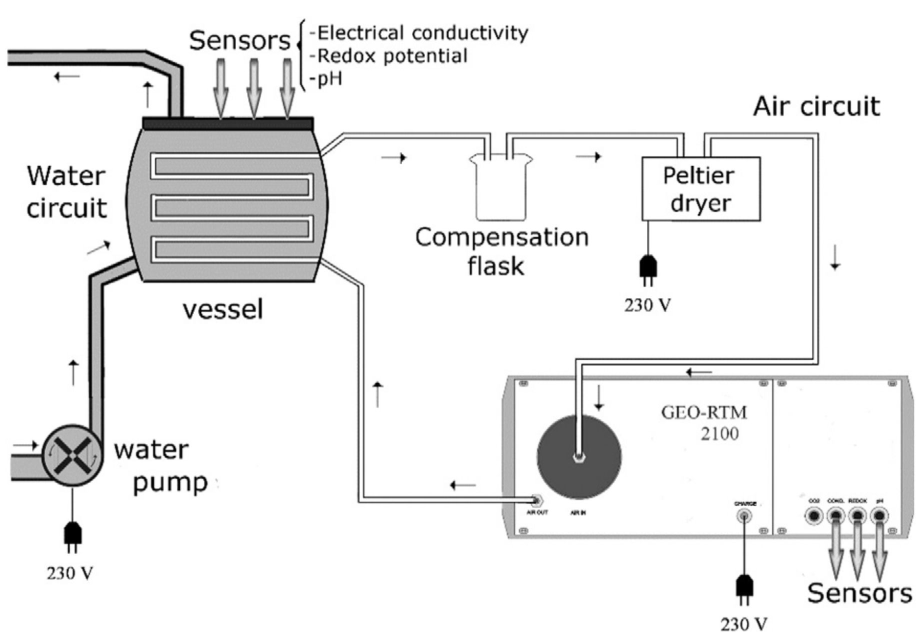

Fig. 2. Experimental device diagram used for continuous measurement of radon in water. 110 × 80 mm (600 × 600 dpi).

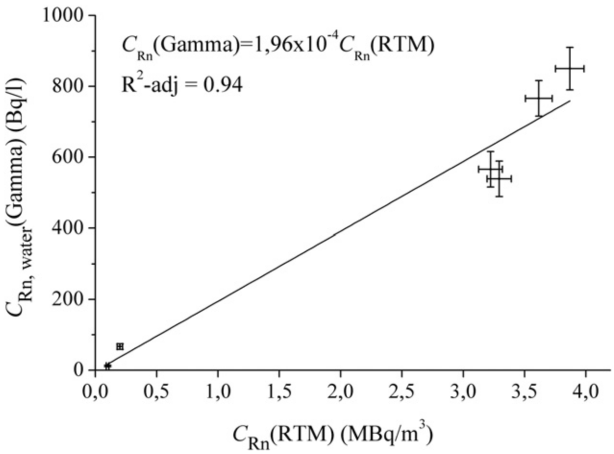

Fig. 3. Radon concentration obtained with gamma spectrometry $C_{Rn,water}$(gamma) versus obtained with RTM 2100 device C_{Rn}(RTM). 82.5 × 58 (1200 × 1200 dpi).

studied facility) (Cosma et al., 2008) because of the radon gradient inside the wall of the tube provoked by diffusive process.

2.4. Radon behavior models

Two different mathematical models were used to explain the variations of radon concentration observed continuously in the main well. The models were fed with the temporal series of radon concentrations measured in water, and then the fitting parameters used in these models have provided additional information, as the rate of water exchange between the main well and other secondary wells, or the degree of radon loss in the water-air interface per unit time.

2.4.1. Closed Compartment Model (CCM)

This model tries to explain the radon concentration variation with the time when the thermal spa is without activity (treatments, etc.…) (from 12 PM. to 6 AM). When external water is not pumped, main well doesn't received water from outside.

Closed Compartment Model supposes that it has a volume of water V with an initial radon concentration $C_{Rn,water}(t=0)$ named as C_0. Inside that water volume there is a radon source with a exhalation rate E and surface S. Radon concentration decrease is given by disintegration to and passage of radon dissolved in water to air. Therefore radon concentration dynamics (Kowalczk and Froelich, 2010) is set as:

$$\frac{dC_{Rn,water}}{dt} = \phi - \lambda C_{Rn,water} \qquad (2)$$

where $\lambda = \lambda(^{222}Rn) + \lambda_V\ [h^{-1}]$, $\lambda(^{222}Rn) \equiv {}^{222}Rn$ decay constant, $\lambda_V \equiv$ constant that reflects the radon loss in water-air interface per unit time, $\phi = ES/V \equiv$ radon emission rate [Bq/l h^{-1}].

The variation of radon concentration in water over time is given by the resolution of the Eq. (2):

$$C_{Rn,water}(t) = C_0 e^{-\lambda t} + \frac{\phi}{\lambda}\left(1 - e^{-\lambda t}\right). \qquad (3)$$

The asymptotic radon concentration in water C_{max} can be obtained with the next condition:

$$\frac{dC_{Rn,water}}{dt} = 0 \Rightarrow C_{max} = \frac{\phi}{\lambda}. \qquad (4)$$

2.4.2. Opened Compartment Model (OCM)

This model tries to explain the radon concentration variation with the time when the thermal spa is working (from 6 AM to 12 PM). In this case water with radon concentration C^* is pumped from outside, the main well receives part of this water. Model conditions are identical to CCM ones but it is necessary add an exchange term λ_I. Thus radon concentration dynamics in water (Kowalczk and Froelich, 2010) is given by:

$$\frac{dC_{Rn,water}}{dt} = \phi - \lambda C - \lambda_I\left(C_{Rn,water} - C*\right). \qquad (5)$$

The above equation resolution with the initial condition $C\ (t=0)$ named as C_1 is:

$$C_{Rn,water}(t) = C_1 e^{-\lambda^* t} + \frac{d}{\lambda^*}\left(1 - e^{-\lambda^* t}\right) \qquad (6)$$

where $d = \phi + \lambda_I C^*$ [Bq/l h^{-1}] and $\lambda^* = \lambda + \lambda_I$ [h^{-1}].

From adjustment parameters obtained in the CCM and OCM models shown in Table 1, exchange constants per unit time λ_V and λ_I have been calculated, and the estimation of radon concentration of outside well was used for testing the model. The result of fitting the experimental data on the radon concentration in water with the Closed and Opened Compartment Models referred to the first time series of Table 1, is shown in Fig. 4.See Table 2

2.5. Dose estimation

The effective dose rate expressed in mSv per period time was estimated from the following equation:

$$\dot{H}[\text{mSv/period}] = \text{WLM} \cdot f \qquad (7)$$

with f conversion factor whose value to the public and workers is 4 and 5 mSv/WLM respectively. The unit WLM (Working Level Month) is defined as exposure to 1 WL for a work period of one month (170 h):

$$\text{WLM} = \frac{\text{WL} \cdot t(\text{h/period})}{170} = \frac{C_{Rn,air}\left(\text{Bq/m}^3\right) \cdot F}{3700} \qquad (8)$$

where F is the equilibrium factor between radon and its progeny (International Commission on Radiological Protection, 1993). According to UNSCEAR for typical residential environments takes an average value of 0.4 (UNSCEAR, 2008).

3. Results and discussion

3.1. General results

As shown in Fig. 5, radon levels in water and air of Las Caldas de Besaya thermal spa are highly variable. Radon in water average is 660 Bq/l with maximum and minimum of 764 and 306 Bq/l respectively, while the radon in air average is 2900 Bq/m^3 with 10,400 and 890 Bq/m^3 maximum and minimum values respectively in the months studied. One of the highest levels of Spain according to the article by Soto et al. (1995) which shows the results achieved from measurements performed in 54 spas.

Dose estimation was obtained from Eqs. (7), (8) and the average value of radon concentration in air, 2900 Bq/m^3 whose maximum and minimum are 10,400 and 890 Bq/m^3 respectively. Dose calculate was done annual and monthly for workers with 2000 and 170 h spent on the installation respectively. In the case of patients, dose was calculated for a week with 2 h of permanence per day. However, as these dose estimations have been made under the assumption of standard working conditions, which do not fit perfect with the specific conditions of this workplaces, it would be necessary to monitor radon in air in each area and include personal dosimeters to workers in order to obtain more accurate and reliable values (Quindós et al., 2014).

Table 1
Results of the parameters which characterize the equations given by CCM and OCM models obtained through fitting the experimental data on the dates indicated.

	CCM				OCM			
Date	C_0[a] (Bq/l)	ϕ (Bq/l h^{-1})	λ (h^{-1})	R^2[b]	C_1[a] (Bq/l)	d (Bq/l h^{-1})	λ* (h^{-1})	R^2[b]
March 14–15	581	222 ± 11	0.31 ± 0.02	0.99	710	450 ± 20	0.76 ± 0.03	0.98
April 25–26	601	170 ± 10	0.24 ± 0.02	0.97	660	310 ± 20	0.52 ± 0.03	0.99
May 13–14	557	270 ± 30	0.40 ± 0.04	0.94	657	420 ± 30	0.72 ± 0.05	0.99
June 19–20	585	280 ± 30	0.40 ± 0.04	0.96	650	470 ± 30	0.80 ± 0.05	0.99
July 7–8	534	230 ± 20	0.33 ± 0.03	0.96	619	370 ± 50	0.70 ± 0.09	0.99
August 2–3	533	320 ± 30	0.48 ± 0.04	0.97	703	420 ± 20	0.67 ± 0.04	0.96

[a] Initial concentration taken as fixed parameter, which uncertainty is 3%.
[b] Adjusted coefficient of determination R^2-adj.

3.2. CCM and OCM models

Radon concentration in water usually has a periodic behavior with minimum values reached usually in the beginning of the afternoon during working days, and the highest ones around 7–8 AM in the same days. The models described in Section 2.4 try to explain this periodicity.

Radon behavior in water seems to be well explained by the proposed models CCM and OCM. Experimental data are adjusted accurately by equations models. In all cases the adjusted coefficient of determination R^2-adj is not less than 0.94.

Several conclusions can be drawn from the CCM model results. Losses of radon in water are mainly controlled by the desorption air through the water surface ($\lambda \approx \lambda_v$), making water the main source of radon at spa. Emission rate ϕ takes a mean value of 260 ± 60 Bq/l h^{-1}, value that could be taken as reference to characterize the main well. However it has high variability and might not be constant over year, can be controlled by other factors such as the flow of water coming out the spring that supplies the main well.

From OCM model radon concentration of outside well C^* have been obtained (see Table) being most of results consistent with 560 ± 80 Bq/l the article of Soto et al. (1991, 1995). This model has also allowed characterizing the exchange of water volume per unit time through constant λ_I between the main well and the outside one, and λ_v, which indicates the radon loss in water-air interface per unit time.

3.3. Anomalies in water radon concentration

It have been found several anomalies, which means, from the point of view of radon concentration in water, a behavior which CCM and OCM models are unable to explain. Fig. 5 shows radon concentration in water and a number of physico-chemical parameters, periodic behavior is observed except in shaded area (on April 8 and 14 to 17). Concentration in both periods was higher, in the second range periodicity given by the OCM model did not appear. Further water temperature and conductivity were significantly decreased this period.

In period where anomalies were observed, thermal spa was closed, i.e., water from outside well was not pumped. Therefore, OCM model does not determine radon dynamics in this case. It is remarkable that radon desorption to air through the water surface depends on the temperature thereof (Hunyadi et al., 1999).

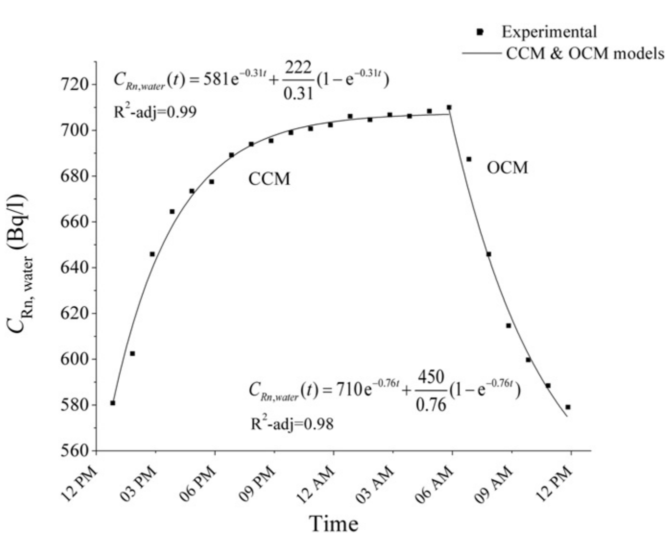

Fig. 4. Fit made according to the CCM and OCM models to experimental data on the radon concentration in water from 14th (12:50 PM.) to 15th (11:50 AM.) March. 120 × 90 mm (1200 × 1200 dpi).

3.4. Relationship between radon in water and radon in air

To determine the relationship between radon in water and air concentrations, a correlations study has been carried out. For April, radon concentration is inversely correlated, for the other months there is no correlation. Fig. 6 shows anticorrelation between water and air radon, furthermore shaded area indicates the period where anomalies in water radon concentration were found. As mentioned in previous sections, the daily maximum radon concentration in water appears around 7–8 AM when activity of the spa begins, and pump bringing fresh water into the main well starts working. At the same time, services such as jets, showers or inhalations also begin increasing the concentration of radon in air gradually by desorption from the water. The high variability of radon concentration in air corresponding to the seasonal end, could be attributable to storms and flood events in the thermal spa. Moreover, that variability indicates the adequacy of continuous monitoring when precise radiological protection have to be made. However, it would be necessary a larger number of measuring points given the large volume and spaces within the spa to know more accurately radon in air dynamics. The influence of this kind of events on radon in water and radon in air concentrations should be afforded in more detail in further studies.

3.5. Correlations with physico-chemical parameters

Correlations study was made between the radon concentration in water and physico-chemical parameters, as well as radon concentration in air with air temperature. Some inverse correlations were found between radon in water and pH and air temperature. On the other hand, positive correlation was observed between radon in water and

Table 2
Parameter results of λ_v, λ_I and C^* obtained from Eqs. (3) and (6) and data shown in Table 2. Uncertainty in values was achieved through propagation of errors.

Series	λ_v (h^{-1})	λ_I (h^{-1})	C^* (Bq/l)
March	0.43 ± 0.01	0.32 ± 0.03	430 ± 80
April	0.23 ± 0.02	0.28 ± 0.04	500 ± 100
May	0.39 ± 0.04	0.32 ± 0.06	470 ± 160
June	0.39 ± 0.04	0.40 ± 0.06	480 ± 130
July	0.32 ± 0.03	0.37 ± 0.09	380 ± 170
August	0.47 ± 0.04	0.19 ± 0.06	500 ± 200

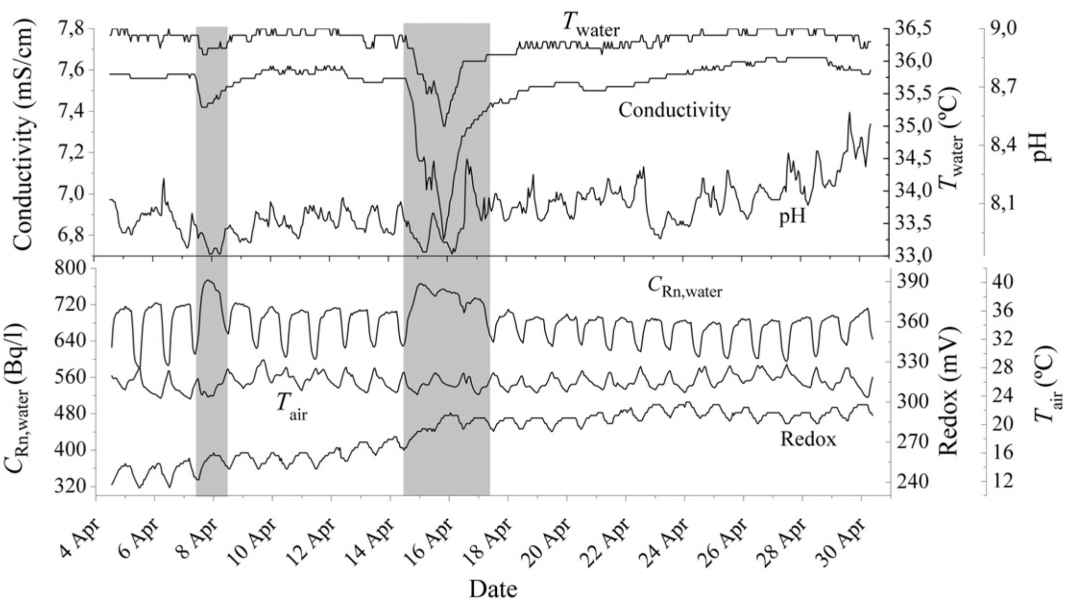

Fig. 5. Experimental results of radon concentration in water, conductivity, redox potential, pH and water and air temperatures measured in April with RTM 2100 device. 155 × 85 mm (1200 × 1200 dpi).

redox potential and, between radon in air and air temperature. Correlations with conductivity and water temperature were not found.

Theoretical models explain quite well the radon behavior in water, it is basically controlled by the operation of the spa. However conductivity and water temperature decrease when the spa is closed (see Fig. 5). These parameters are related with the origin of water, showing the main well lower values than those corresponding to the outside well. Leaks from rain or river would decrease greatly conductivity and water temperature. Radon solubility decreases with temperature, accordingly outside well has less radon than the main, assuming the same radium content in rocks of both.

One of the conclusions that have been mentioned is that the radon source in air is water. Nevertheless radon behavior in air has high variability and it can be controlled by many factors such as ventilation, number of patients using the facilities and atmospheric pressure. There is direct correlation between radon in air and air temperature, this may be due to the operation of the spa. And a low inverse correlation between radon in water and air on April, being insignificant for the other months, which does not lead to a reliable result.

Analyzing the correlations of the radon concentration in water with the other parameters measured, it has been found that the pH and redox potential have moderately and high direct correlation respectively. Those conditions are suitable for the radio solution (Ra^{2+}) is absorbed by the aquifer surface, thus constituting an excellent source of radon. There is a high inverse correlation with air temperature that as in the previous case may be due to operation of the spa. With water temperature would expect an inverse correlation, but as variations of it are the order of the thermometer uncertainty, no significant results were found.

3.6. Dose estimation

IS-33 instruction establishes 600 Bq/m^3 as reference level of radon concentration in air. Levels at Las Caldas de Besaya thermal spa far exceed this level, establishing above 1000 Bq/m^3, which would lead applying a high level of control. In this situation, the general principles of operational radiation protection under Title IV of RPSRI (Anon., 2001) must be applied. In practice, this implementation will take place gradually, considering the level of exposure, the number of workers affected and existing protection alternatives. It should be studied the radon concentration in air in all places where workers are commonly found, in order to know more precisely the characteristic dose of each working activity. Dynamics knowledge of radon concentration in indoor air would apply radiation protection systems based on changes in work schedule.

According to the dose estimation to workers and public, the first could receive in a year of work up to 18 mSv, over 7 times the annual global average dose associated with natural radiation. They will receive the global average doses associated with radon in a month working at spa. In case of patients the dose received in a week would be the

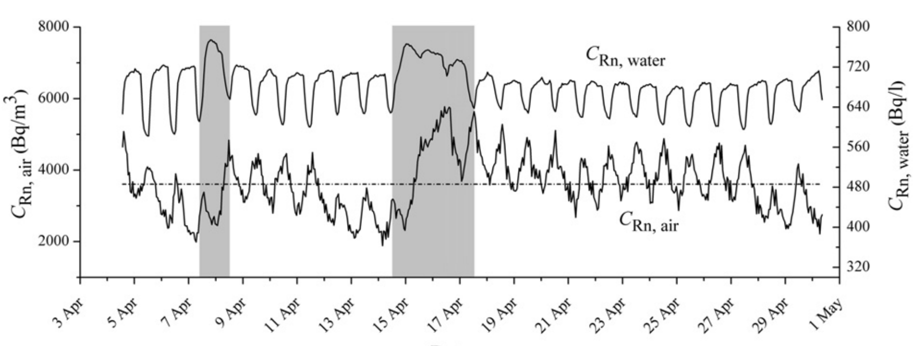

Fig. 6. Radon concentration in water and air time series for April, 2012 measured with RTM 2100 and Radon Scout devices respectively. Dashed line indicates monthly average for radon in air which value is 3600 Bq/m^3. 155 × 72 mm (1200 × 1200 dpi).

equivalent of a chest radiograph, ranging around 0.1 mSv per week, a contribution to the annual dose of little relevance.

4. Conclusions

Continuous radon monitoring have been performed continuously for almost a year in the indoor air and water of a thermal spa located in the North of Spain. With an integrated measuring system, also other parameters like temperature, pH or conductivity were monitored in water as well. The system was set up for the specific measuring conditions by means of validated gamma spectrometry technique for radon in water analysis. The system can be used in a wide variety of scenarios were radon in water monitoring is required, but a good determination of the transfer factor of the diffusive tubes corresponding to the specific water temperatures of each case should be carried out.

It was observed that radon concentration in water usually showed a periodic behavior, related with the daily operation of the thermal facility. In order to explain it, two simple theoretical models where successfully applied. The equations given by these models are adjusted to the experimental data quite accurately. From CCM (closed compartment model) it can be concluded that the loss of radon in water are controlled mainly by radon desorption to the indoor air through the water surface. At the same time, this fact points out the free water surface inside the facility as a major source of indoor air radon concentration. However, more detailed measurements of radon in air should be performed in different rooms for treatment in order to refine this conclusion, as the concentration of radon in air can be significantly influenced by a number of other factors such as ventilation, the work of showers, jets, and other techniques used in the spa.

On the other hand, the results obtained from the application of OCM (open compartment model) provided an approximation of the radon concentration in the outer well which supplies the facilities. Most values obtained were consistent with previous results present in literature. This model also allowed characterizing the exchange of volume per unit of time with constant water between the main well and the outer one.

The measurement of additional parameters like conductivity and water temperature have provided some relevant information about the origin of water. For example, the water from the river or rain usually can present lower conductivity and very low temperature compared to thermal water, causing decreases in these two parameters when it is pumped to the main well. Analyzing the correlations of the radon concentration in the water with the other parameters measured, it was found that the pH and redox potential have a moderate to high direct correlation respectively. However, the correlations found have to be studied more in depth in order to obtain a conclusion with applicability to other projects or applications.

Finally, and according to Spanish regulation related with exposure to radon in workplaces, a preliminary dose assessment was done for workers and public. The annual average indoor radon concentration indicate that it would be necessary to apply the principles of operational radiological protection established in Title IV of RPSRI (Soto et al., 1995). In practice, this implementation would take place gradually, considering the level of exposure, the number of workers affected and the existing alternatives protection.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2015.11.052.

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