

Origin and transport of radon in a dry and in a wet mofette of COVASNA, Romania

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ABSTRACT

The appearances of significant efflux of carbon dioxide through soil surfaces is a frequent natural phenomena in the wider vicinity of relatively young volcanic fields. These mofettes are often utilised as dry carbon dioxide spas to treat patients suffering from cardiovascular diseases. The carbon dioxide always carries some naturally occurring radioactive radon gas with it. On the one hand this radon gas might pose some radiation risk for the patients and staff of such carbon dioxide spas, on the other hand it may be used as a natural tracer for the study of transport of its carrier gas in the subsurface and in the pools. In this work we have measured the spatial variation of radon activity concentration in the gas phase of the carbon dioxide gas pool of the Cardiology Hospital and in the gas and water phases of the Hell-Mud, which is a wet open pit mofette in the city of Covasna. We have developed a hydro- and gas-geological conceptual and mathematical model with which we were able to describe the measured spatial and temporal behaviour of radon gas concentration. We have found that the relatively low radon concentrations at the bottom of these pools can be explained by removing the radon content of water by intense bubbling degassing of the oversaturated carbonated waters.

1. Introduction

The emission of low temperature (<100 °C), carbon dioxide rich gases from soils, which are related to post-volcanic magma degassing activity or thermal decarbonisation, are called mofettes. This phenomenon is common in Transylvania, especially in the surroundings of Ciomadul Mountains, which is the youngest volcano of the Carpathian-Pannonian region. Radiocarbon dating of charcoal fragments showed that the last eruption was about 30.000 years ago (Harangi et al., 2010). In a recent study Harangi et al. (2020) confirmed this age using (U–Th)/He zircon dates combined with U–Th in situ rim dates along with luminescence and radiocarbon dating. Since then the volcano has been in dormant period. In spite of the relatively long quiescence, geophysical studies (Popa et al., 2012; Szakács and Seghedi, 2013), as well as intense “post-volcanic activity” (dry CO₂ emanations, CO₂-rich mineral springs, bubbling swamps and lakes, and carbonate spring deposits) (Szakács, 2010) suggest that there could still be a melt-bearing magma body in the crust. Seismic tomography data of Popa et al. (2012) show that earthquakes as well as a low-velocity lithosphere column at crustal and subcrustal depths have been detected beneath the

Ciomadul area. This can be viewed as a hotter, hence more ductile portion of the crust and lithospheric mantle, thus related to a thermal anomaly generated by magma ascent path during the last volcanic activity, or a crustal magma chamber beneath Ciomadul. Such data and evidences lead Szakács and Seghedi (2013) to the conclusion that Ciomadul might still be able to produce more eruptions in the future.

Kis et al. (2017) estimated the CO₂ gas release on focused gas emissions and diffused soil degassing in selected parts of the seemingly inactive Ciomadul. The estimate of the total CO₂ output in the area was 8.70•10³ tons/year. They presumed a tectonic control of gas emission, since fractures can enhance gas transport via creating main migration pathways towards the surface. Most of the focused emissions (mofettes and bubbling pools) with high fluxes are localized at the periphery of the youngest volcanic complex. Most probably because the oldest part of Ciomadul volcanic area (Puturosul and Bálványos lava domes) are more fractured than the youngest volcanic edifices (explosive craters of St. Ana and Mohoš).

Based on natural carbon dioxide emissions pools that are filled up with gas are often used for therapeutic treatments in the form of dry CO₂ spas. These dry “saunas” where patients sit or stand in the pools, while

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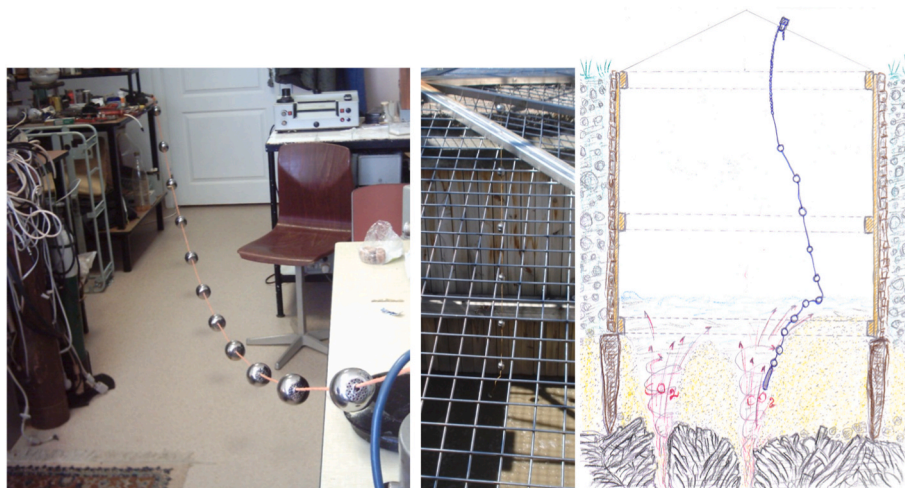


Fig. 1. The prepared Radamons in the lab and as they are attached to the grille of Hell-Mud.

deep origin gases swirl around their lower bodies healing several medical complaints, are also called mofettes. The curative gas is applicable for treating cardiac and peripheral artery diseases (arteriosclerosis in the lower limbs, coronary diseases), high blood pressure, arterial complications in diabetics, postoperative treatment following arterial surgeries (Ballagi, 1995). Examples of mofettes are the Stinky (Torjai-Büdös) Cave in Transylvania, Romania (Szabó, 2005) or the CO₂ seepage in Mátraderecske, Hungary (Vásárhelyi et al., 1997).

The deep origin mofette gas, along its path to the surface, collects different amount of radon from rocks and soils. Radon is a naturally occurring colourless, odourless, tasteless radioactive gas that is a decay product of radium and part of the uranium decay chain. Its most stable isotope, ²²²Rn, has a half-life of 3.8 days. Radon can be an excellent natural tracer of some subsurface flow processes. Despite the rather small concentration, it is well measurable due to its radioactivity. In some cases radon, as a tracer, can be used for the study of transport of pool-filling mofette gas. By analysing the spatial and temporal variations of radon gas, we can obtain useful information on the flow and stability conditions of gases in the pools of mofettes, and on the mixing of mofette gases with surface air.

The aim of this work was to find the origin and the main transport processes that carry radon gas into the pools of a dry and wet mofette of Covasna town, Romania. In order to identify the sources and transport of radon we have developed a relatively simple hydro- and gas-geological conceptual and mathematical model with which we were able to describe the various spatial and temporal behaviour of radon gas concentrations measured in Cardiology Hospital and in Hell-Mud.

2. Materials and methods

2.1. Site description

Covasna is a town, situated in the inner part of the Carpathians bend, just 50 km from the so called Vrancea region, which is one of the most active seismic zones in Europe. The town is referred to as the land of mofettes, table waters and spas. It is well-known for its mineral waters and richness in carbon dioxide seepages (Gyila et al., 2017). Although the distance between Covasna and Ciomadul is 50 km, there is a connection via fault lines and fractures. According to Péter (2017) here is the Eastern edge of the mofetic aureole of the Călimani-Gurghiu-Harghita volcanic chain.

Geologically the Covasna area is made up of Paleocene and Cretaceous sedimentary deposits (Cretaceous Flysch) (Néda et al., 2008 a). Vaselli et al. (2002) derive the geogenic CO₂ from both mantle derived processes and thermometamorphism of recently subducted limestones.

The investigated mofettes have different geological background. The mofette of Cardiology Hospital is an artificial one, where the gas is introduced at the bottom of the pool through a pipe of a 168 m deep borehole named F10. Carbon isotope ratio study was done by Szántó (2002) from the samples of F10 borehole. The analysis indicated that the emerging gas contains both mantle and carbonate derived components.

Hell-mud is located in the main square of Covasna. It is situated in a 4.2 m deep pit, surrounded by a stone wall. The water level fluctuates more than a few meters depending on the meteorological conditions, compared to the outer surface. As a result, a detector placed near the water level may have been situated even below or above the water level.

2.2. Measurement method

The ²²²Rn activity concentration in air was measured with Radamon type etched track radon detectors developed by the Radon Group of the Institute for Nuclear Research (Csige and Csegzi, 2001). We have used TASTRAK type Poly-Allyl-Diglycol-Carbonate (PADC) polymer as track detector, which polymer is better known with its trade name CR-39 (Columbia Resin, 1939). After exposure, the CR-39 detectors were chemically etched in a 6.25 N NaOH solution at a temperature of 70 °C for 5 h. Etched tracks were counted manually under an optical microscope. Scanned area was 1 cm² for background and low track densities determination. At high track densities, a total of at least 1000 alpha particle etched tracks were counted, and the scanned area was measured. The ²²²Rn activity concentration, C_{Rn} (Bq/m³) was calculated from track density: ρ (number of tracks/cm²), the exposure time: t_{exp} (days), and the calibration factor of the radon monitor: $\beta = 27$ (Bq/m³)/((number of tracks/cm²)nday). The Radamon was originally experimentally calibrated in radon chambers in Sweden and in United Kingdom in the 1990s and recently tested in 2019 in the Hungarian National Radon Chamber (Budapest) in normal air conditions.

Parallel measurements of ²²²Rn activity concentration with Radamon and an ionisation chamber type AlphaGUARD radon monitor in an atmosphere containing varying carbon dioxide concentration resulted in the same results within uncertainty of the detectors. In an experimental study Shahrokhi et al. (2015) compared the response of different type of active radon monitors in atmospheres containing carbon dioxide up to 89%. They have found that the sensitivity of the AlphaGUARD did not change with carbon dioxide concentration. Therefore, in the current work we have used the very same calibration factor as obtained from calibrations in normal air conditions for the measurement of ²²²Rn activity concentration in the gas phase of mofettes containing carbon dioxide in different concentrations up to about 90%.

This Radamon etched track detector was also used for measuring the

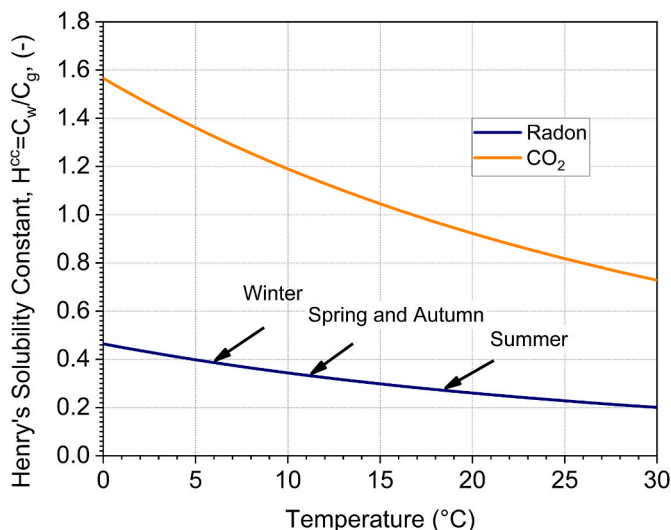


Fig. 2. Henry's law solubility constant of radon and carbon dioxide gas in water as a function of temperature (Sander, 2015).

activity concentration of ^{222}Rn dissolved in the muddy water of the pool of the Hell-Mud wet mofette. To do this, the Radamon radon detector was packed into a thin (15 μm) radon permeable rubber foil (masculan® condom) (Hakl et al., 1995). For aesthetic reasons, the Radamons thus packed were also placed into stainless steel tea leaf holder. The tea leaf holders (with the radon detectors inside them) then were attached to a rope at a certain distance from each other. The upper end of the rope was fixed to a bar of the stainless steel railing that covers the pool of the Hell-Mud for safety reasons. A counterweight was fixed at the lower end of the rope, which kept the whole rope stretched out. In this way, each radon detector was located at the same height relative to the external ground level (Fig. 1).

The ^{222}Rn concentration of water was measured in-situ, no water samples were taken. For Radamon radon detectors, immersed in water, the ^{222}Rn activity concentration in the sensitive volume of the detector is equal to the equilibrium gas phase concentration, which is related to the ^{222}Rn activity concentration dissolved in water by using the temperature dependent Henry's law solubility constant (H^{cc} , Fig. 2).

2.3. Conceptual model

We have several, independent observations on how radon activity concentration varies in a dry mofette, like the one of Cardiology Hospital, or in a wet one, like Hell-Mud. We have managed to develop a model, which can explain the spatial and temporal variations of radon concentrations measured at the above mentioned sites. The conception is that the radon content of deep origin carbon dioxide at higher depths is relatively low, roughly the same (3 kBq/m^3) as of the gas taken from a 168 m deep borehole, which feeds the Great Mofette Pool of The Cardiology Hospital. These upward migrating deep origin gases interact with groundwater. In the very close vicinity of the Hell-Mud mofette runs the Covasna Creek. We have observed a number of gaseous and non-gaseous groundwater leakages into the Covasna Creek both on the side walls and on the bottom of the creek bed. This indicates that the area of the Hell-Mud is a discharge area having an upward directed vertical seepage velocity component.

We assume that the relatively high radon content of the water of the Hell-Mud pool originates from the accumulation of radon from a few metres thick near-surface sedimentary rock layer, characterized by high radon emanation rate, as the gaseous groundwater under the Hell-Mud pool penetrates it. Usually the water velocity of groundwater is low which means actually no influence of the water velocity to the ^{222}Rn uptake from the rock material. However, at the site of the Hell-Mud, the

seepage velocity is greatly increased by the pressure gradient enhancement due to the high oversaturation of groundwater with carbon dioxide. Therefore, the dissolved radon content of groundwater that discharges to the Hell-Mud pool may depend on the seepage velocity. Water, due to a faster leakage through sedimentary rocks with high radon emissions, can accumulate less radon. At the same time the water entering in the sludge of Hell-Mud is substantially oversaturated with carbon dioxide. As a result of the pressure drop, gaseous carbon dioxide is released in the form of bubbles, which leaves into the airspace of Hell-Mud. During outgassing process the carbon dioxide also carries away a significant part of the dissolved radon. Therefore, the concentration of dissolved radon in aqueous phase of the pool space is significantly lower than that of the groundwater feeding Hell-Mud. In addition, the amount of leaving radon also depends on the temperature of sludge, due to the temperature dependence of radon to water/gas phase distribution.

We can also describe all of this with mathematical formalism, which gives us the ability to compare our hydro- and gas dynamics model, which originated from the conceptual model, against our measurement results. During modelling, we used the "trial and error" method in order to find the most appropriate fit of model calculation results to measurement data.

2.4. Model calculations

The transport of ^{222}Rn gas in the gas phase of the pools of both the wet (Hell-Mud) and dry (Cardiology Hospital) mofettes is described by an advection-(eddy)diffusion equation:

$$D_a \frac{d^2 C(z)}{dz^2} - v \frac{dC(z)}{dz} - \lambda C(z) = 0 \quad (1)$$

where $C(z)$ is the ^{222}Rn activity concentration at depth z , measured from ground surface, directed upwards; $\lambda = 2.09838 \cdot 10^{-6}/\text{s}$ is the decay constant of ^{222}Rn ; D_a is the (molecular + eddy) diffusion coefficient of ^{222}Rn and v is the average vertical gas flow velocity.

For underground radon transport, the geological environment under the wet mofette is basically divided into two parts: one is a deep rock (below $z_3 = -7 \text{ m}$) characterized by a relatively low (G_d) radon source term, on which there is an order of magnitude higher (G_s) radon-emitting sedimentary rock layer. The groundwater level, which is also the upper level of water in the pool of the Hell-Mud mofette, can be found in the sedimentary rock, the distance from the surface of which is $z_1 = -2.45 \text{ m}$. The bottom level of the muddy water in the pool of the Hell-Mud mofette is at $z_2 = -4.2 \text{ m}$. The porosity of deep and sedimentary rocks was equally set to $\varphi = 0.5$. A 1 dimensional model was created, in which highly carbonated groundwater seeps upwards in the deep and sedimentary rock layers with an average intrinsic velocity of v . This groundwater seeps into the pool of the Hell-Mud mofette through the bottom surface of the pool. The water level in the pool is approximately the same as in the adjacent sedimentary rock. In the pool volume the water is very well mixed and leaks to the adjacent sedimentary rock through the sidewalls of the pool. The gas extraction takes place entirely in the water phase of the pool of the Hell-Mud. Although it is possible that the degassing of the water by bubble formation starts at greater depth than the bottom of the pool, in this simple model calculation this was not considered.

In stationary case the spatial variation of ^{222}Rn activity concentration in the rock layers can be described as:

$$D \frac{d^2 C}{dz^2} - v \frac{dC}{dz} + G - \lambda C = 0, \quad (2)$$

where $C(z)$ is the ^{222}Rn activity concentration in liquid phase that completely fills the pore space. D is the dispersion coefficient. Molecular diffusion can be omitted since the molecular diffusion length of radon in water is approx. 1 cm. However, dispersion, due to the relatively high flow-velocity of groundwater below the Hell-Mud pool justifies a

Table 1

Fitted parameters from experiments.

	α , [K]	H_0^{cp} , [mol/m ³ Pa]	Reference
Radon	2600	$9.2n10^{-5}$	Lide and Frederikse (1995)
Carbon dioxide	2400	$3.3n10^{-4}$	

diffusion coefficient corresponding to a few meters of diffusion length. Longitudinal dispersion length was considered proportional to the size of the domain and the dispersion coefficient was calculated as the product of the longitudinal dispersion length and subsurface flow velocity. Thus the differential equation above can be referred to as an advection - dispersion equation rather than an advection - diffusion one. The same dispersion coefficient was used both for deep and sedimentary rock. G is the ²²²Rn source term expressing the activity of radon that is emanated into a unit volume of liquid phase per unit time (Bq/ (m³s)).

The solution of the above differential equation in the deep rock can be written as:

$$C_d(z) = C_{d0}e^{\gamma_1 z} + \frac{G_d}{\lambda}, \quad z < z_3,$$

and in the sedimentary rock layer as:

$$C_s(z) = C_{s0}e^{\gamma_2 z} + \frac{G_s}{\lambda}, \quad z_3 \leq z \leq z_2,$$

where

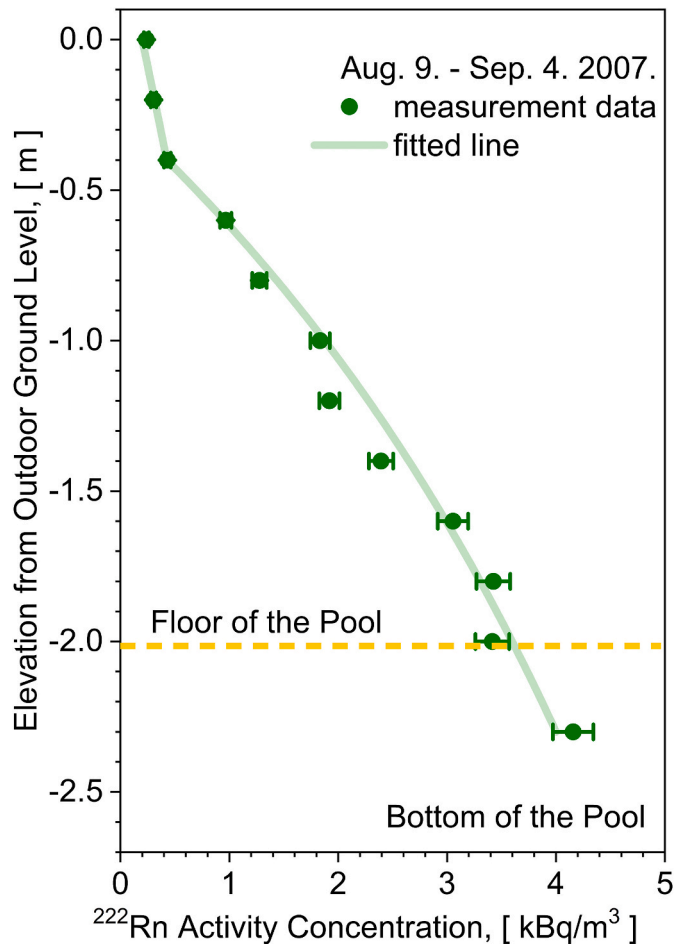


Fig. 3. Model calculations fitted to the vertical profile of average ²²²Rn activity concentration in the mofette of Cardiology Hospital in the summer of 2007.

$$\gamma_1 = \frac{v + \sqrt{v^2 + 4D\lambda}}{2D}, \quad \gamma_2 = \frac{v - \sqrt{v^2 + 4D\lambda}}{2D}.$$

The coefficients C_{d0} and C_{s0} can be obtained from the boundary conditions at the interface of the deep and sedimentary rocks, that is from equality of ²²²Rn activity concentrations and of ²²²Rn activity flux densities at $z = z_3$. The results are:

$$C_{d0} = \frac{\gamma_2}{\gamma_2 - \gamma_1} \frac{G_s - G_d}{\lambda} e^{-\gamma_1 z_3}, \quad C_{s0} = \frac{\gamma_1}{\gamma_2 - \gamma_1} \frac{G_s - G_d}{\lambda} e^{-\gamma_2 z_3}.$$

The radon balance equation for the water in the pool of the Hell-mud is:

$$\frac{dC_w}{dt} = \frac{1}{z_2 - z_1} (j_2(z_2) - j_b - v\varphi C_w) - \lambda C_w. \quad (3)$$

Here $j_2(z_2)$ is the bulk ²²²Rn activity flux density at the bottom of the pool, which can be specified as:

$$j_2(z_2) = (v - D\gamma_2) e^{\gamma_2 z_2} \varphi C_{s0} + v\varphi \frac{G_s}{\lambda}.$$

j_b is the ²²²Rn activity flux density transported from the water to the gas phase of the pool by bubbles. This term is obtained by multiplying the volume flux density Q_{bv} of carbon dioxide gas in the bubbles (m³/m²s) and the ²²²Rn activity concentration in bubbles ($C_b = C_w/H^{cc}$).

$$j_b = Q_{bv} C_b.$$

The ²²²Rn activity concentration in bubbles is related to the ²²²Rn activity concentration in the water (C_w) through the Henry's partition coefficient (H^{cc}), the temperature dependence of which is given as

$$H^{cc}(T) = \frac{C_w}{C_b} = H^{cp}(T) R T = H_0^{cp} R T e^{\alpha \left(\frac{1}{T} - \frac{1}{T_0} \right)} \quad (4)$$

$R = 8.314 \text{ J/(mol K)}$ is the ideal gas constant, T , [K] is the absolute temperature and $T_0 = 298.15 \text{ K}$ is a reference temperature. H_0^{cp} and α are constants that can be determined experimentally.

There is a similar relationship between carbon dioxide dissolved in molecular form in water and the concentration of carbon dioxide in gas phase. The parameters matched to measurement data are shown in the following (Table I):

Volumetric flux density (Darcy velocity) of water entering the pool at the bottom is:

$$q = v\varphi, \quad [m^3 / m^2 s].$$

Flux density of dissolved carbon dioxide transported by water is:

$$Q = q^* [CO_2]_w, \quad [mol / m^2 s].$$

We assume that the flux density of carbon dioxide gas that separates from water in the form of gas bubbles is

$$Q_b = q([CO_2]_w - [CO_2]_t), \quad [mol / m^2 s],$$

where $[CO_2]_t = H_{CO_2}^{cp}(T) p_{CO_2}$ is the equilibrium concentration of carbon dioxide dissolved in water at partial pressure of carbon dioxide in bubbles of $p_{CO_2} = 10^5 \text{ Pa}$ and at temperature T .

Finally we need to convert the material flux density into a volume flux density. This requires the use of molar concentration of carbon dioxide in the bubbles. According to the ideal gas equation: $p_{CO_2} = [CO_2]_b RT$, from which $[CO_2]_b = p_{CO_2} / (RT)$.

The volume flux density of carbon dioxide leaving through bubbles is:

$$Q_{bv} = \frac{Q_b}{[CO_2]_b}, \quad [m^3 / m^2 s].$$

The radon flux density leaving through bubbles is:

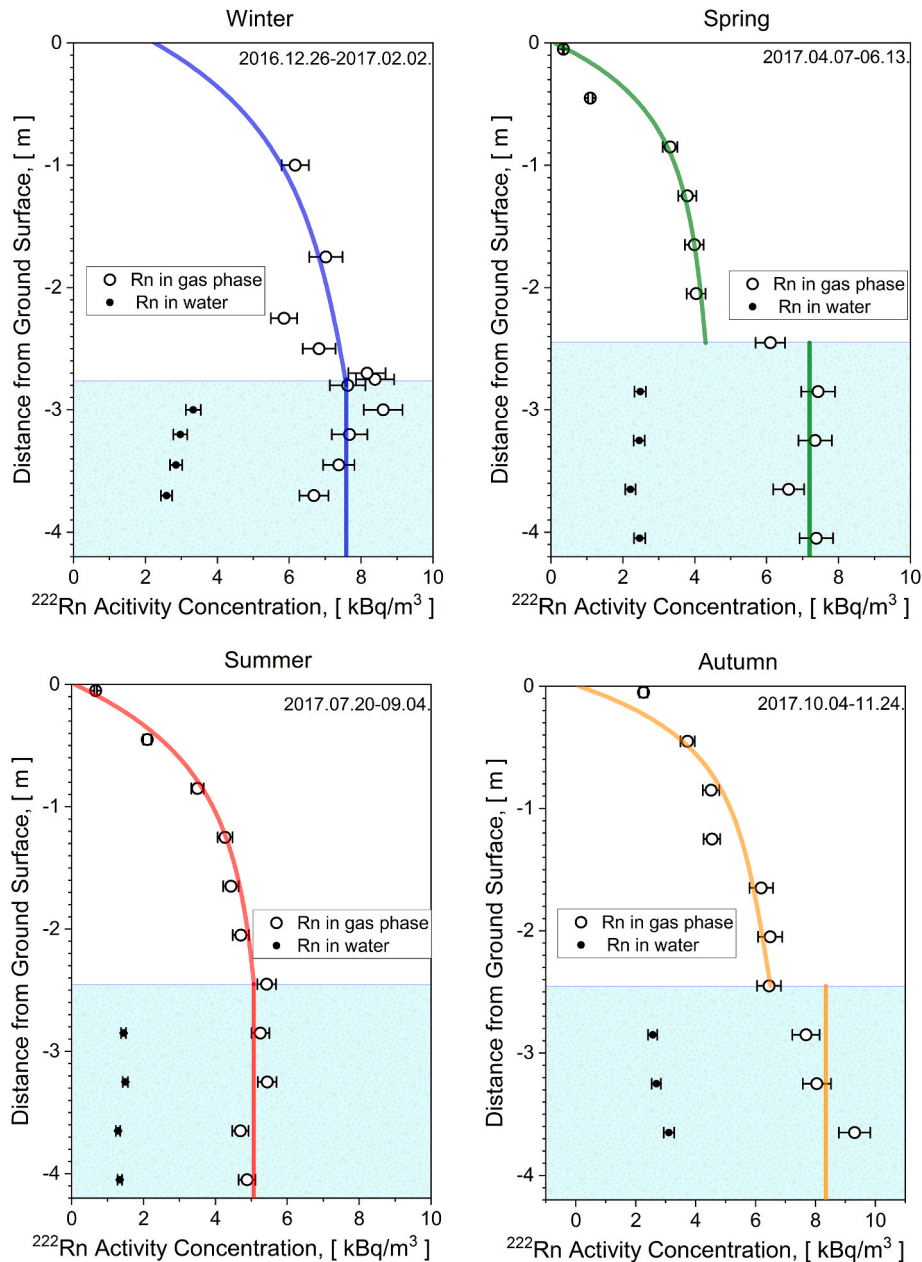


Fig. 4. Seasonal variation of radon concentration at Hell-Mud. Best fits were obtained assuming leakage velocities resulting in the same gas yield of $1.2 \text{ m}^3/\text{h}$ for winter and spring; $2.4 \text{ m}^3/\text{h}$ gas yield for summer and $0.7 \text{ m}^3/\text{h}$ gas yield for autumn.

$$j_b = Q_{bv} C_b = Q_{bv} \frac{C_w}{H_{Rn}^{cc}(T)}$$

Substituting all of these in equation (3) of radon balance of pool space in stationary case:

$$(v - D\gamma_2) e^{\gamma_2 z_2} \varphi C_{s0} + v\varphi \frac{C_w}{\lambda} - Q_{bv} \frac{C_w}{H_{Rn}^{cc}(T)} - v\varphi C_w - \lambda(z_1 - z_2) C_w = 0.$$

From which

$$C_w = \frac{(v - D\gamma_2) e^{\gamma_2 z_2} \varphi C_{s0} + v\varphi \frac{C_s}{\lambda}}{\frac{Q_{bv}}{H_{Rn}^{cc}(T)} + v\varphi + \lambda(z_1 - z_2)}.$$

where C_w is the radon activity concentration of the water in the pool.

By substituting C_{s0} in the former equation the following expression is obtained:

$$C_w = \frac{\frac{\varphi}{\lambda} \left\{ \frac{(v - D\gamma_2)\gamma_1}{\gamma_2 - \gamma_1} (G_s - G_d) e^{\gamma_2 h_s} + vG_s \right\}}{\frac{Q_{bv}}{H_{Rn}^{cc}(T)} + v\varphi + \lambda h_w}$$

where $h_s = (z_2 - z_3)$ is the thickness of the sediment under the bottom of the pool and $h_w = (z_1 - z_2)$ is the thickness of the water layer in the pool.

3. Results and discussion

3.1. Cardiology Hospital

This artificial mofette is genetically related to the Neogene Volcanic range of the Eastern Carpathians (Néda et al., 2008 a). While in the mofette of Mátradereske (Hungary), located on the same Neogene Volcanic range, a radon activity concentration of 125 kBq/m^3 (Tóth et al., 1996) has been measured in the emanated gas, according to Sonoc

Table 2
Data for calculation of radon content in water.

Measuring period	Average air temperature, [°C]	Average wind speed, [m/s]	Water temperature, [°C]	H^{ec} , [-]
Winter	-7.0	0.04	6.0	0.387
Spring	12.1	0.15	11.0	0.334
Summer	19.9	0.09	18.0	0.275
Autumn	6.4	0.09	11.0	0.334

et al. (1984) and the measurements done by Szabó (1992) shows that there is relatively low radon activity concentration in the mofette of Cardiology Hospital. These results are consistent with our measurements (Fig. 3), and show that the ^{222}Rn content on the floor of mofette pool (at -2.0 m from outdoor ground surface) of Covasna Cardiology Hospital, is stably around 3 kBq/m³.

In the mofette of Cardiology Hospital patients stand at different levels of the pool depending on the level of carbon dioxide gas present in the pool. As the pool is 2 m deep and most of the time the gas fills it up almost completely, patients usually stand at medium levels and practically never go to the floor of the pool.

As can be seen on Fig. 3, our advection-eddy diffusion model fits measurement data relatively well. During fitting the diffusion coefficient, the ^{222}Rn activity concentrations at the boundaries and the average gas flow velocity were changed. Furthermore the vertical domain was divided into a lower and upper part at around $z = -0.4$ m, which were characterised by different (eddy) diffusion constants in order to obtain a reasonable fit to experimental data. Physical reasoning of this division is based on the assumption that the mixing of the gas in the upper part of the pool is much higher than in the lower part where the gas sits more stably in the pool.

Fig. 3 shows that the average ^{222}Rn activity concentration decreases from the bottom up. Similar spatial distribution can be observed in most of the mofettes (Néda et al., 2008a,b; Cuoş et al., 2014; Incze et al., 2016; Sóki and Csige, 2016). The higher radon concentration value at the bottom of the mofette is in connection with the geological feature of the mofette area. Towards the ground level the mixing of mofette gases with ambient air reduces the concentration of CO₂ and radon too in the pool space. Carrier gases such as CO₂ (or CH₄) assume a dominant role in controlling transport and redistribution of radon (or He and other trace gases) towards the surface (Etioppe and Martinelli, 2002). Therefore, in most cases the source of radon is related to mofettes. Furthermore, the vertical distribution of radon is similar to its carrier.

3.2. Hell-Mud

At Hell-Mud the variation of radon concentration was measured as a function of distance from the ground surface. Four profiles were measured each representing a season during 2016–2017. The results and the model calculations fitted to the measurement data are shown in Fig. 4. Best fits were obtained assuming leakage velocities resulting in the same gas yield of 1.2 m³/h for winter and spring; 2.4 m³/h gas yield for summer and 0.7 m³/h gas yield for autumn. The assumed reason for these differences is a seasonal variation in intensity of stratified water flow. It is a general experience that the intensity is characterized by a late autumn - early winter minimum and a spring - early summer maximum (Nyizsalovszki és Szabó, 2003).

On these figures we also show the ^{222}Rn activity concentration dissolved in water. For this we used the results of detectors underwater and the temperature dependent Henry's law solubility constant. Table II Summarises the data used for calculation of radon content in water.

We have found that radon concentrations show different pattern depending on seasons (Fig. 4). In the spring period, the lower ^{222}Rn activity concentration in the airspace may be related to the more frequent windy and stormy periods that can sweep out the mofette gases

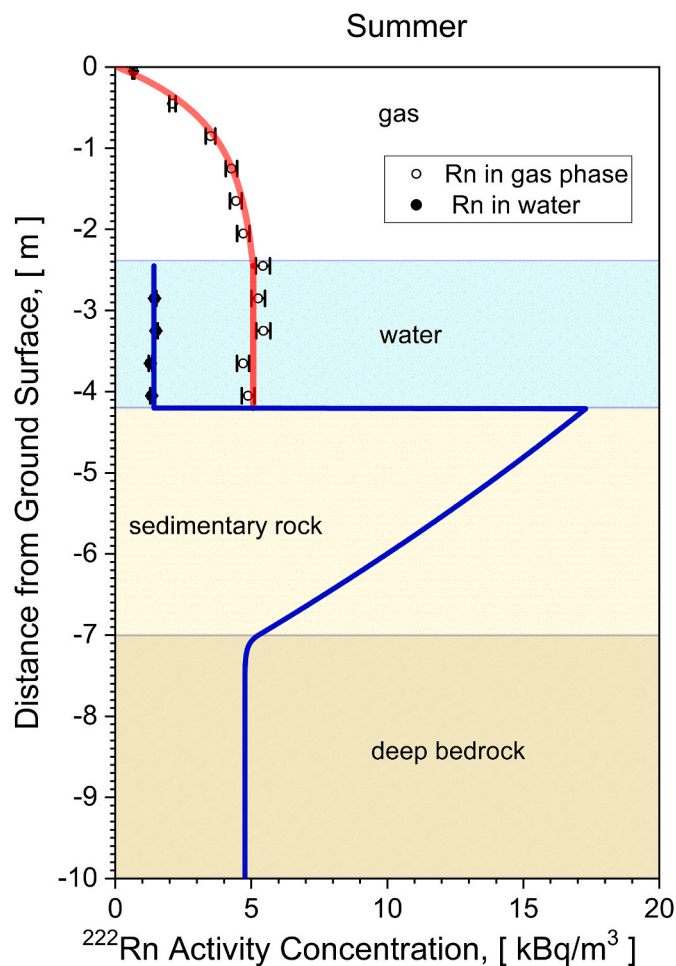


Fig. 5. Model calculations for spatial variation of ^{222}Rn activity concentration in deep bedrock and sedimentary rocks in Hell-Mud at summer period.

from the airspace of Hell-Mud. It can be seen that the calculated ^{222}Rn activity concentrations in the water phase relative to the gas phase are smaller in summer than in winter due to the less solubility of radon at higher temperatures. In this model the water in the pool is very well mixed. The apparent systematic decrease (winter) and increase (autumn) of ^{222}Rn concentration with depth in the water may either due to measurement uncertainty or some unknown effect not considered here.

We have identified groundwater and wind speed as main driving factors in determining radon levels in water and air, respectively. The radon content of the water in the pool is the result of uptake of radon from the surrounding sedimentary rock layer and the degassing phenomena in the pool. While in the airspace of the mofette the origin of radon is the gas bubbling out from water.

Assuming that the geological source of mofette gas in Hell-Mud is the same as in Cardiology Hospital, the higher ^{222}Rn activity concentration can be explained with a few metres thick radium rich sedimentary rock layer lying near to surface (Fig. 5), through which more radon can be accumulated. The scale of enrichment depends on the leakage velocity of groundwater. There is an inverse proportion between leakage velocity and radon content. In this model the leakage velocity was a fitting parameter, the value of which was determined by the trial and error method, and its value was found to be in the order of 1–2 m/d. The faster the leakage the less radon can be accumulated.

Fig. 5 contains model calculations of Hell-Mud that describe transport of radon in different geological media. The radon content of groundwater oversaturated with deep origin CO₂ is relatively low in the

deep bedrock (~5 kBq/m³). The upward leaking groundwater accumulates more radon from the radium rich sedimentary rock layer. Carbon dioxide is outgassing in the water of wet mofette, and carrying away much of the dissolved radon content, too.

4. Conclusion

Based on the results obtained in this study, we can conclude that the chosen advection – eddy diffusion model describes relatively well the measurement results in the airspace of mofettes. With our conceptual model we can also interpret the vertical radon profiles measured in different seasons in Hell-Mud. From the results above we may conclude that the source of radon in Hell-Mud is the same gas as in Cardiology Hospital, however it collects more radon from near surface soil layers.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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