16ENV10 MetroRADON

Report on Activity A.4.3.2

SUBG Results for Radon in Air, Radon in Soil-gas and Radon exhalation from soil, obtained in the frames of "METRORADON: Intercomparison on indoor radon at LNR Saelices el Chico (Salamanca, Spain)", organized by LaRUC

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1. Measurements of radon in air

A novel type of detector is used - a DVD (used as a solid state track detector) and two thin Makrofol N foils (used as radon absorber) facing the DVD surface. Another DVD half is used to keep the foils close to the disk. The detector is designed in such way that radon progeny in the air doesn't have an influence on the signal. The background exposure of the detectors was not estimated based on the transit detectors, but with an alternative approach! That is because the transit detectors showed very high exposure – 79 kBq.h/m3. It is possible that significant part of this exposure was accumulated in the LNR lab while the other sets of detectors were exposed and it shouldn't be taken into account.

The detectors were calibrated by exposure of an identical set of detectors to radon atmosphere at the calibration facility at Sofia University. The exposure was carried out at the same temperature as the exposure in the intercomparison at LNR. The obtained results are shown in the Table below.

	Overall Results (kBq h m^{-3})	
	Value	Uncertainty
1st Exposure	**317**	**32**
2nd Exposure	**796**	**62**

Table 1. Results from the exposures of radon in air carried out in the frames of the intercomparsion at LNR in November 2018. The presented uncertainties are at the level of 1 standard deviation and include the calibration uncertainty and the standard deviation of the results of the 10 detectors in each group.

2. Measurements of radon in soil gas and radon exhalation

The method based on liquid scintillation counting of polymers is proposed in [1]. For the measurements of radon in soil-gas and radon exhalation from soil the metal rod (stainless steel rod with holes along its length with Marofol N foils packed in thin kitchen polyethylene sheet inside) was used. The foils were placed 5 cm apart and the last one reached a depth of 75 cm. The rod with the foils was hammered (see Fig. 1) at 15:30 on 05 Nov. 2018 and pulled-out at 10:30 on 07 Nov. 2018. The temperatures measured at the beginning and at the end of the exposure at about 20 cm below the ground were in the range 10 – 11^{o}C.

Figure 1. Hammering the rod in the "Green"

The foils were placed in glass LS-vials with THM-cocktail shortly after the exposure and measured at the "Triathler" LS-counter provided by the LaRUC-team. For the activity estimation, the signal in the alpha-channel obtained more than 6 hours after the end of exposure was used. The beta-channel signal was not used because of its high and variable background counting rate. Based on the observed count rates in the alpha- and beta-channel it seems that about 10% of the alphas are counted as betas. Despite of the loss of alphas, this separation seems good as no betas are counted in the alpha-channel (Fig.2 (middle)).

The activity concentration in the soil was determined as:

$$C_A = \frac{n_0 k_{des}}{\varepsilon_c \varepsilon_s V}$$

where n_0 is the net counting rate of the foil corrected to the moment of its placement in the vial, k_{des} is a correction factor for the radon desorbing from the foils in the minutes before the foil is placed in the vial, ε_c is the counting efficiency in the alpha-channel of the Triathler, ε_s is the sampling efficiency for the exposure conditions and V is the volume of the foil. The estimation of each of the above values is briefly described below.

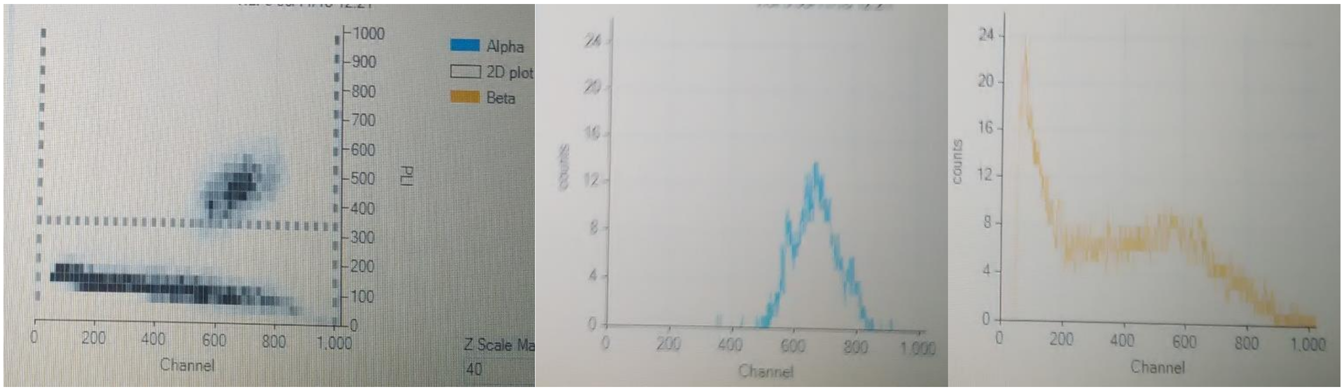


Figure 2. (left) 2D-plot at Z=40 – visualization of the alpha/beta separation of the Triathler and the PSA-threshold. (middle) Alpha- and (right) beta-spectra obtained with this PSA.

In order to calibrate the Triathler two of the samples (with the foils buried at the two highest depths) measured at the Triathler were brought in Sofia and measured at the RackBeta. The foils were followed at the RackBeta for a few days and the net signal was extrapolated to the moment of the preparation of the samples (see Fig. 3). The net count-rates of in the alpha-channel of the Triathler were also decay-corrected to that moment. The ratio between the count-rates of the Triathler and the RackBeta and the known counting efficiency of the RackBeta in the two samples were used to estimate the counting efficiency of the Triathler in the alpha-channel.

The obtained estimate is ε_c =2.615(39) for the PSA-threshold shown in Fig. 2, which is close to the value used in the preliminary estimation ε_c=2.73(27). Additionally, we worked on the 2D plots of the samples in order to improve the alpha/beta separation by moving the PSA-threshold and adding Energy-threshold (see Fig.4). That resulted in about 10% increase in the counting rate in the alpha-channel and the counting efficiency for this PSA-value, estimated using equation (2), is ε_c =2.863(42).

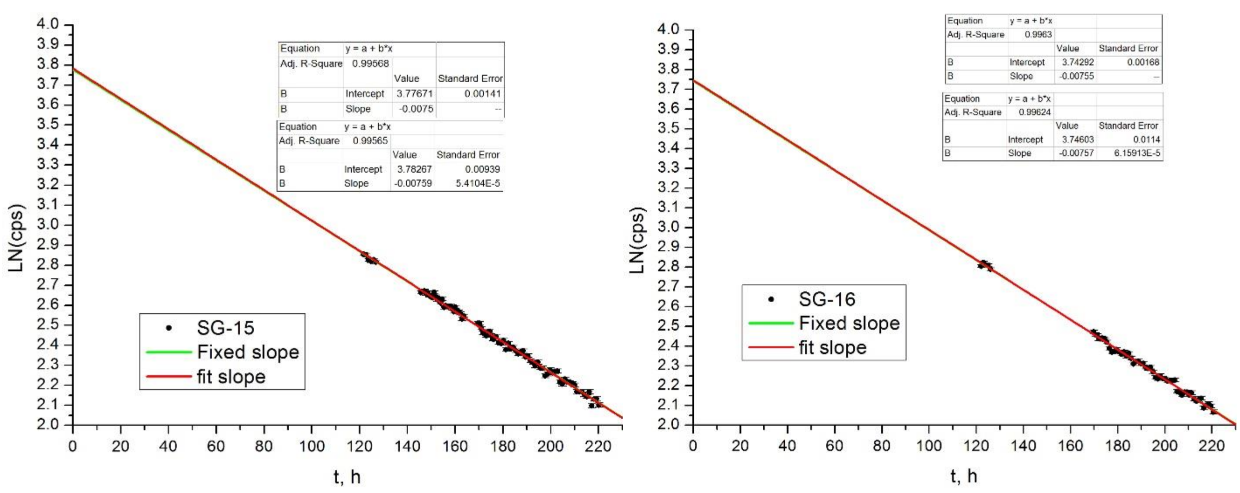


Figure 3. Logarithm of the net signal as a function of time for the two samples. Two fits are applied – one with fixed slope-parameter equal to the Rn-decay constant and the other with free slope-parameter. As it is seen, for each sample the fit-parameters of the two fits coincide within the uncertainties.

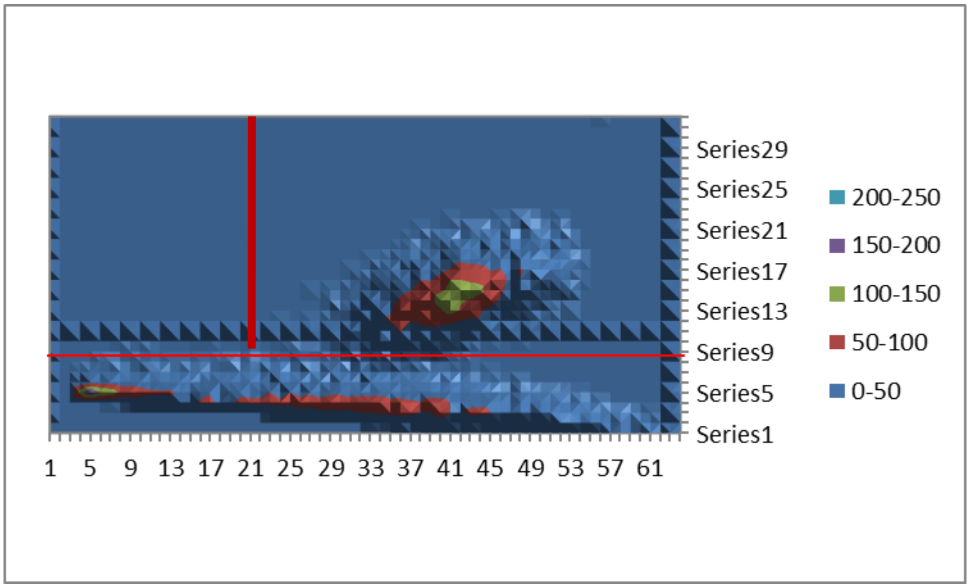


Figure 4. 2D-plot at Z=0 – visualization of the alpha/beta separation of the Triathler – the black triangles mark the original PSA-threshold, the horizontal red line marks the new PSA-threshold and the vertical red line the Energy threshold.

In order to determine the sampling efficiency (ε_s) we conducted an experiment to determine the partition coefficient K and L_D of Makrofol N at t=10^o C using an approach presented in [2]. The obtained values are K=183(12) and L_D=23.9(10) um. A sorption/desorption model [3] was used to estimate the sampling efficiency of the foils for the given exposure conditions assuming constant activity concentration and temperature in the soil. The estimated value for the sampling efficiency multiplied by the volume of the foil is $\varepsilon_s V$= 1.49x10^{-5}(14) m^3 (actually, that product is needed to estimate the ^{222}Rn concentration). As the foils desorbed from the moment the rod was pulled-out to the moment of their placement in the LSC-vials, the desorption correction for each foil C_{des} was also estimated.

To estimate the activity concentration of ^{222}Rn at infinity ($C_{A,\infty}$) by the gradient method, the depth profile of C_A(d) should be fitted with the function:

$$C_A(d) = C_{A,\infty}\left(1 - e^{-\frac{d}{L_{D,soil}}}\right)$$

where $L_{D,soil}$ is the diffusion length of radon in the soil and d is the depth. The exhalation rate J_0 can then be determined with the parameters determined by the fit:

$$J_0 = \lambda L_{D,soil} C_{A,\infty}$$

As the uncertainty of $C_A(d)$ is dominated by systematic contribution mainly due to the estimate of the ε_s, the fit can be directly applied to the counting rate (after the decay and desorption correction is applied) then $C_{A,\infty}$ can be calculated. The fit was applied to the decay and desorption corrected count rates obtained directly by the Triathler and that obtained with the modified PSA threshold(see

Fig. 5). No qualitative difference between the two fits is found. Moreover, the difference between the obtained values for $C_{A,}$ and J_0 by the two approaches are smaller than their respective uncertainties. The values of the fit with the higher *R*-squared value were chosen for the estimates. For visualization purposes the fit curve and the count rates are converted in activity concentration and plotted in Fig. 6.

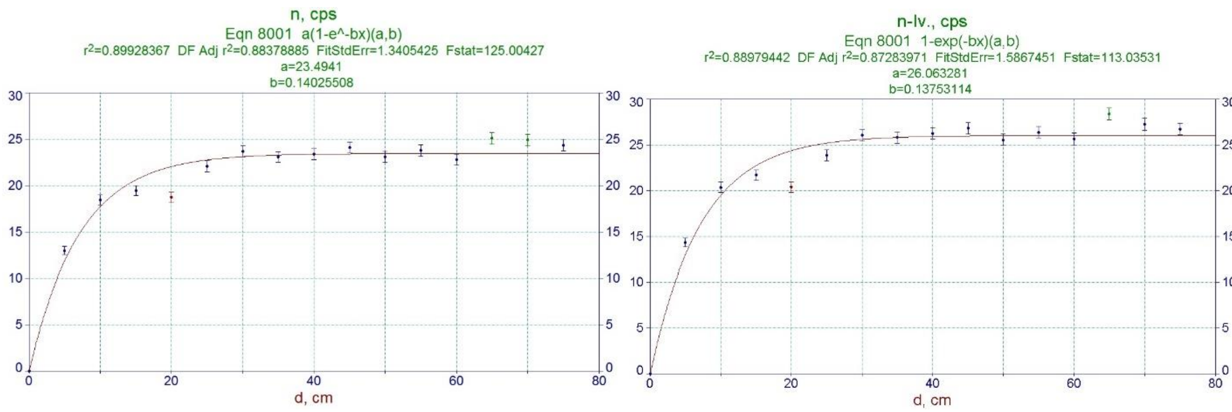


Figure 5. Applying the gradient method (left) on the count rates of the Triathler and (right) on the count rates with the modified PSA-threshold.

The exhalation rate was additionally estimated through the “missing” activity near the surface with the following equation:

$$J_o = \lambda \sum_i \frac{C_{A,\infty}+C_{A,i}}{2} \Delta d ,$$

where $C_{A,i}$ is the the i-th data point and Δd is the distance between each measurement point. This estimate is 10% greater than the one obtained by the fit due to the “drop” of the point at d = 20 cm. There could be a physical reason for that drop. Therefore, this was the estimate of J_0 reported as a final results.

Results:

	Radon in soil concentration (kBq m^{-3})	
Location	Value	Uncertainty
"Green Ballesteros" (next to the LNR) Offices site	602	57

	Radon Exhalation (Bq m^{-2} h^{-1})	
Location	Value	Uncertainty
"Green Ballesteros" (next to the LNR)	361	33

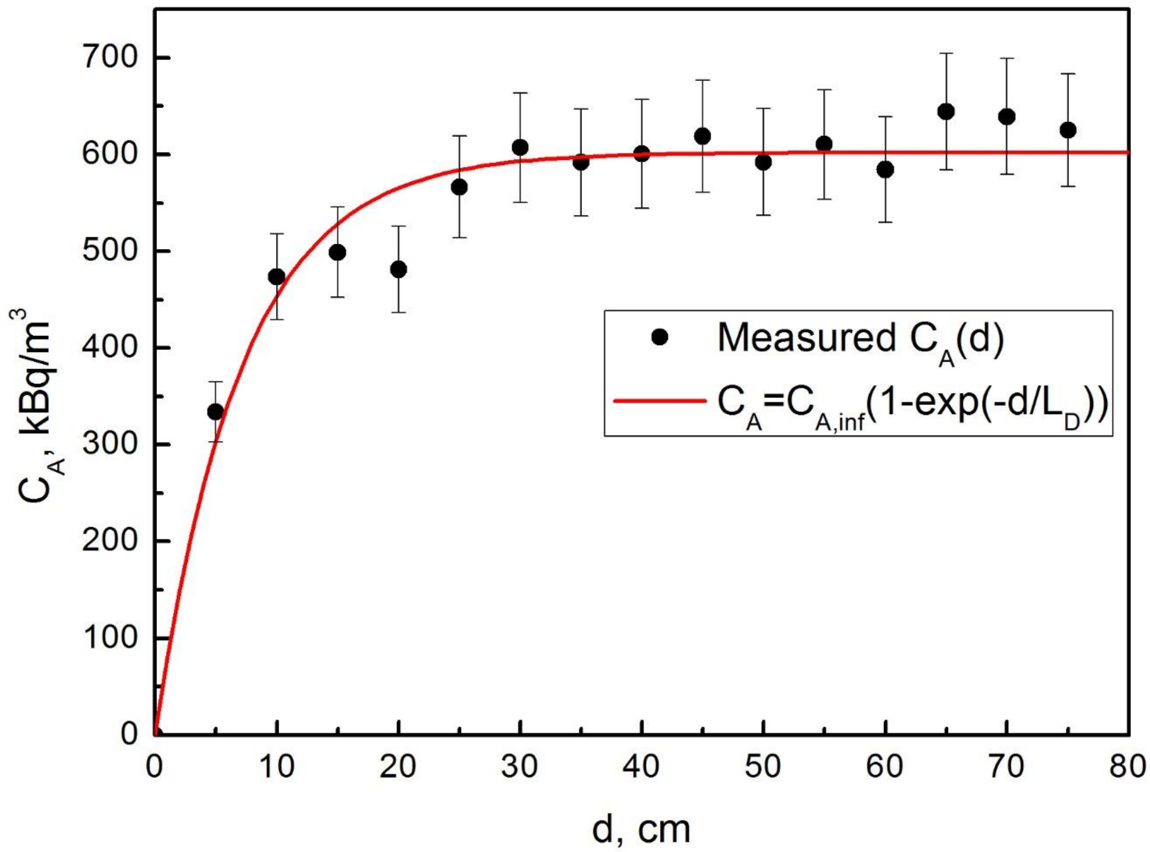


Figure 6. Depth profile of the radon concentration.

3. Comment on the exhalation measurement results and comparison of the results to the other participants

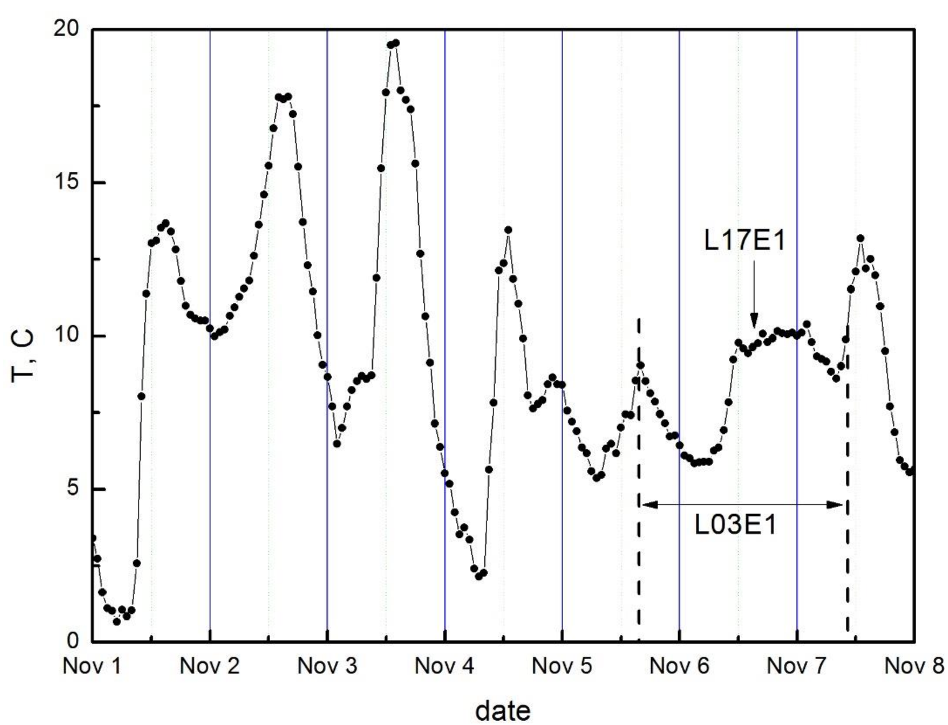
The large differences in the results of the radon exhalation measurements require some explanation. Although the measurements are very few to draw definitive conclusions, still some consideration can be made. As could be seen in Fig. 7 (Table 10 from the Metroradon report, WP.3.3.3.), measurements L03E1 and L17E1 overlap in time, but they differ in two orders of magnitude. However, the first method is cumulative, while the second is discrete. During the period of these measurements, the weather was mostly rainy and windy and the soil was soaked, which would impede the exhalation. That is seen in the radon depth profile shown in Fig.6 (the data is obtained by L03 and used for the gradient method). The diffusion length of radon in soil corresponding to that profile is L_D=7.1(7)cm. Due to the windy weather, there were some cloudless and sunny time windows and the discrete measurement L17E1 was carried out in such a window (see the weather data in Fig. 8). The sun would dry the soil and lead to increase in the radon exhalation, which could be a possible explanation for the observed difference between the results of the two measurements.

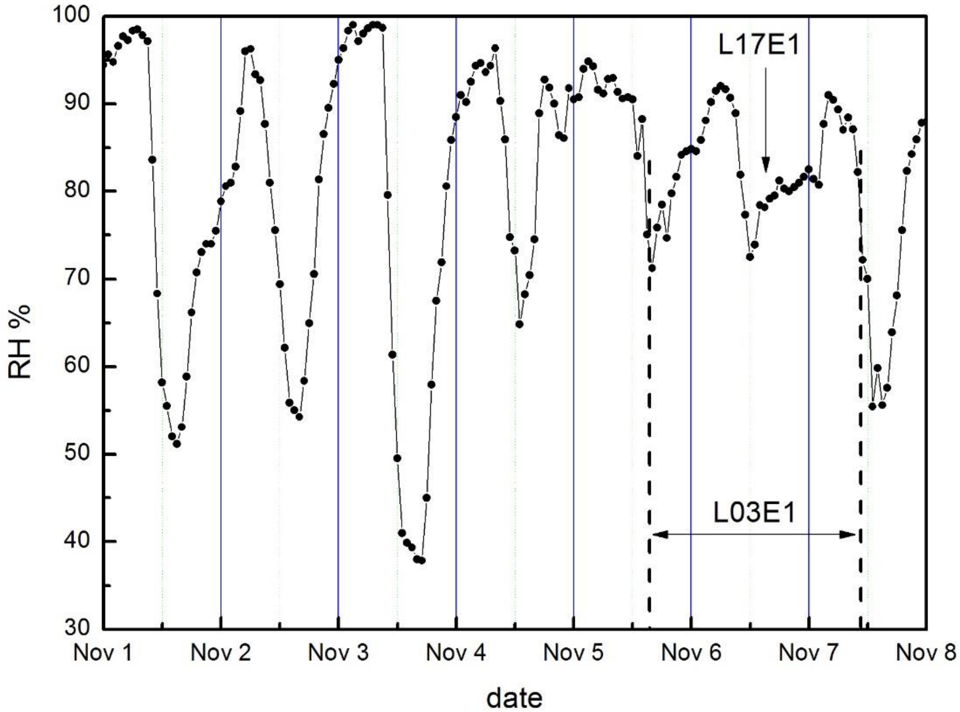
On the other hand, the two cumulative measurements L03E1 and L20E1 could not be compared directly, as they were carried out at different times. The weather was dry and sunny in the week of the L20E1 measurement, in contrast to the weather during the L03E1 measurement. Although no definitive conclusions can be drawn, these results indicate the significant effect of the weather on the radon exhalation rate, which deserves more thorough study. More details about the intecomparison performed at Saelices el Chico are given in [4].

Table 10. Radon exhalation J and its uncertainty $u(J)$ performed in the indicated date with the methodology used by each participant to conduct the test in the "Green Ballesteros".

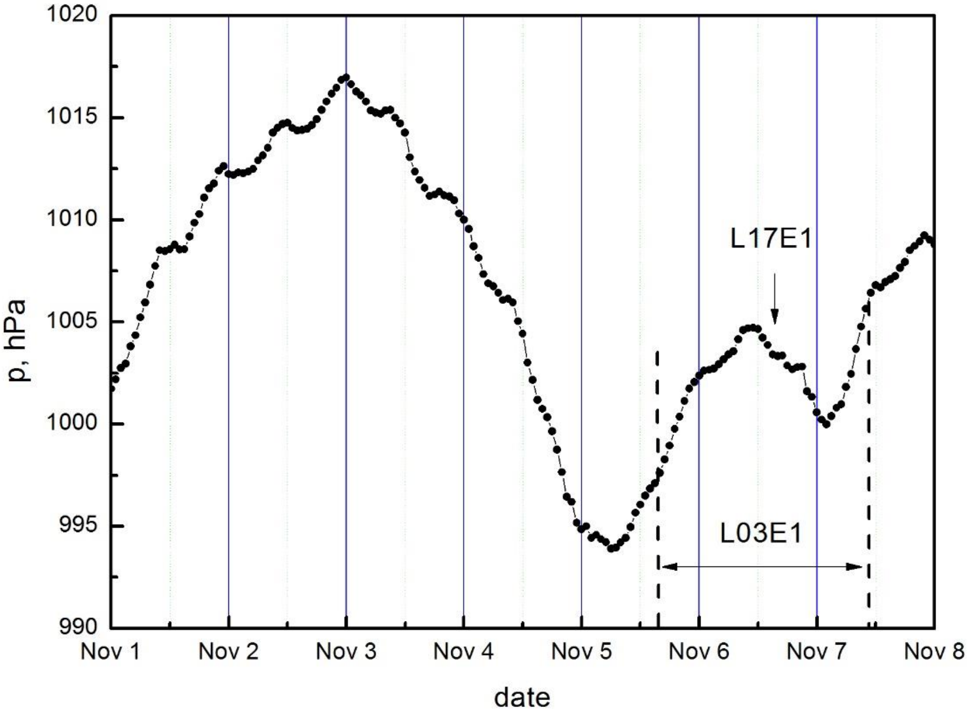
Code	J (Bq m^{-2} h^{-1})	$u(J)$ (Bq m^{-2} h^{-1})	Date (2018)	Methodology
L03E1	361	33	5 Nov 15:30 to 7 Nov 13:30	Gradient method with polycarbonate foils
L17E1	14719	1939	6 Nov 15:30 (approx. 1 hour.)	Accumulation method
L20E1	35100	8200	15 Nov 10:00 to 16 Nov 10:00	Absorption in activated charcoal collector

Fig. 7. Table 10 from the UC report.





RH %
date
L17E1
L03E1
100
90
80
70
60
50
40
30
Nov 1
Nov 2
Nov 3
Nov 4
Nov 5
Nov 6
Nov 7
Nov 8



p, hPa
date
L17E1
L03E1
1020
1015
1010
1005
1000
995
990
Nov 1
Nov 2
Nov 3
Nov 4
Nov 5
Nov 6
Nov 7
Nov 8

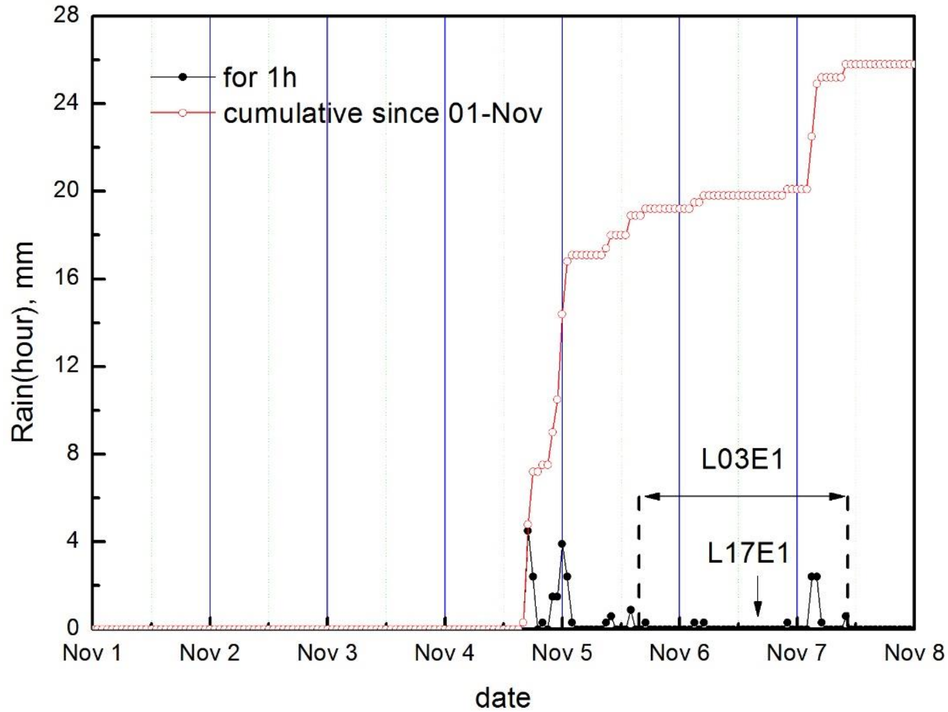


Fig.8. Meteorological data for the period of the intercomparison. The times (or intervals) of the radon exhalation measurements are marked. The temperature increase and the RH decrease indicate sunshine before and during measurement L17E1, which could facilitate radon exhalation. The transient drop in the atmospheric pressure could also facilitate the exhalation.

References:

[1]. K. Mitev, Ch. Dutsov, S. Georgiev, T. Boshkova, D. Pressyanov, "Unperturbed, high spatial resolution measurement of Radon-222 in soil-gas depth profile", Journal of environmental radioactivity, 196 (2019) 253-258.

[2]. K. Mitev, P. Cassette, S. Georgiev, I. Dimitrova, B. Sabot, T. Boshkova, I. Tartès, D. Pressyanov, "Determination of ^{222}Rn absorption properties of polycarbonate foils by liquid scintillation counting. Application to ^{222}Rn measurements", Applied Radiation and Isotopes, 109 (2016) 270-275.

[3]. D. Pressyanov, K. Mitev, S. Georgiev, I. Dimitrova. "Sorption and desorption of radioactive noble gases in polycarbonates", Nuclear Instruments and Methods in Physics Research Section A, 598/2 (2009) 620-627.

[4]. D. Rabago, L.S. Quindos, J. Quindos, E. Fernandez, A. Fernandez, L. Quindos, S. Celaya, I. Fuente, C. Sainz, Intercomparison of indoor radon and geogenic radon measurements under field conditions, Metroradon report, WP.3.3.3., February, 2019.