

# Comparison of two $^{222}\text{Rn}$ mass exhalation rate measurement methods by study of Hungarian adobe building materials

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**Abstract.** Adobe samples were taken from settlements in three distinct areas (SE-, NE-, and S-Hungary). The  $^{222}\text{Rn}$  mass exhalation rates of the samples were estimated by two different techniques for aim of a comparison exercise, both using Al-accumulation chambers and RAD7 detectors. One technique (growth curve method) is based on measuring the growth curve of  $^{222}\text{Rn}$  concentration in the chamber for ten days, and the other one (equilibrium method) on measuring for four hours the  $^{222}\text{Rn}$  equilibrium concentration after three weeks (about  $5(T_{1/2})$ ) in the sealed chamber. The latter one is faster for high number of samples but cannot take into account the leakage from the measurement circle. In case of  $^{220}\text{Rn}$ , only the equilibrium concentrations can be used ( $5(T_{1/2}) \approx 5$  min). In addition,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  activity concentrations were determined by gamma-ray spectrometry, and grain size distributions were measured by wet sieving and with a laser grain size analyzer. Our results show that below a determined and controlled leakage rate, the tested equilibrium method provides an acceptable estimate of  $^{222}\text{Rn}$  mass exhalation rate on a time consuming way, and that adobe building material potentially can cause elevated indoor  $^{220}\text{Rn}$  concentrations in all of the studied areas. It is also clear that the grain size distribution is an important property of adobe building materials which has a different influence on  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  exhalation.

**KEYWORDS:**  $^{222}\text{Rn}$  mass exhalation rate,  $^{220}\text{Rn}$ , growth curve, equilibrium method, adobe building material

## INTRODUCTION

The only gaseous isotope of the  $^{232}\text{Th}$  decay chain is  $^{220}\text{Rn}$  (thoron), which, similarly to  $^{222}\text{Rn}$  (radon) in the  $^{238}\text{U}$  decay chain, increases radiation dose obtained by humans from natural sources. Risk from  $^{220}\text{Rn}$  is considered low in many cases (e.g. UNSCEAR 2000) due to its short half-life (55.6 s) compared to  $^{222}\text{Rn}$  (3.8235 d). However, several studies (e.g. Németh et al. 2005, Sciocchetti et al. 1992, Shang et al. 2005, Yamada et al. 2005, Yonehara et al. 2005) show higher concentrations in dwellings made mostly by soil. Adobe is a natural building material made from clay, sand (soil), water and organic material. After mixing the raw materials and pressing into shapes, the adobe bricks are dried in sunshine, without any burning procedure which could close the pores. Hence, adobe has a high potential to exhale both  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  originating from the naturally occurring  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  in the soil.

Mass exhalation rate is a measurable property of each building material which strongly affects indoor  $^{222}\text{Rn}$  or  $^{220}\text{Rn}$  concentrations. It is given in the activity or the number of  $^{222}\text{Rn}$  or  $^{220}\text{Rn}$  escaping from the sample per mass per time interval. Reliable  $^{222}\text{Rn}$  mass exhalation rate measurements can be performed by long term measurements which take up to ten days for one sample. Hence, there are efforts to reduce the needed measurement time. The usage of a time consuming method started recently at Eötvös University (Budapest, Hungary) and a comparison with a widely used and reliable method (e.g. Jonassen 1983, Stranden 1988, Petropoulos et al. 2001, Sakoda et al. 2008) is needed. This comparison is a part of a larger study on Hungarian adobe dwellings and the higher significance of  $^{220}\text{Rn}$  in them.

## SAMPLES

Three distinct areas were selected for collecting adobe samples made of the local soil. These selected areas are Békés County (SE-Hungary), E-Mecsek Mts. (S-Hungary) and Sajó and Hernád Rivers Valleys (NE-Hungary) (Fig. 1.), where alluvial sediments of Körös and Berettyó Rivers, loess and redeposited loess are prevailing, respectively (Gyalog 2005). Adobe houses are highly typical and common on these areas. The above mentioned alluvial and terrestrial sediments are the main raw materials of adobe building material samples depending on the local soil of each settlement. To date, 11 samples have been studied from Békés County and 8 samples from E-Mecsek Mts. and 8 from Sajó and Hernád Rivers Valleys.

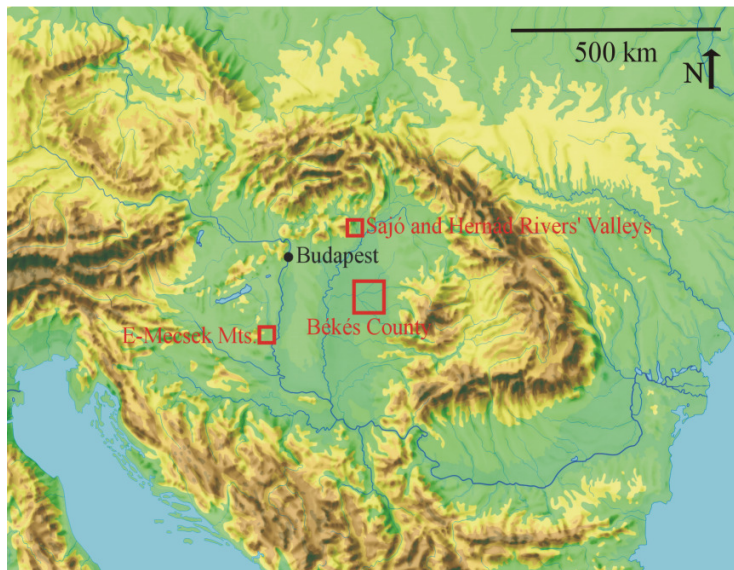


Fig. 1. Map of Carpathian-Pannonian Region showing the adobe sampling areas in Hungary: Békés County (SE-Hungary), E-Mecsek Mts. (S-Hungary) and Sajó and Hernád Rivers Valleys (NE-Hungary) marked by red squares.

## METHODS

### Mass exhalation rates of $^{222}\text{Rn}$

The  $^{222}\text{Rn}$  mass exhalation rates of 27 adobe samples cut to about 200 g cubic blocks were estimated by two different techniques, both using the same Al-accumulation chamber connected to the same RAD7 radon detector via the same plastic tubing, gas drying unit and aerosol filter for each sample. The samples were dried in the air of the laboratory to ensure they had similar moisture content.

The *growth curve method* is based on measuring the growth curve of increasing  $^{222}\text{Rn}$  concentration in the closed Al-chamber for 10 days (e.g. Jonassen 1983, Stranden 1988, Petropoulos et al. 2001, Sakoda et al. 2008). We apply 30 minutes cycles and the growth curve is described by the following equation (Eq. 1.).

$$C(t) = (C_{max} - C_{bg})(1 - e^{-(\lambda+\alpha)t}) + C_{bg} \quad (1)$$

where  $C(t)$  is the increasing  $^{222}\text{Rn}$  concentration above the sample in the sealed measurement circle, measured in every 30 minutes ( $\text{Bq m}^{-3}$ ),  $C_{max}$  is the maximum of  $^{222}\text{Rn}$  concentration reached by the growth curve ( $\text{Bq m}^{-3}$ ),  $C_{bg}$  is the  $^{222}\text{Rn}$  concentration of the background, i.e. the value of Y axis at the end

point of the growth curve ( $\text{Bq m}^{-3}$ ),  $\lambda$  is the decay constant of  $^{222}\text{Rn}$  ( $\text{h}^{-1}$ ),  $\alpha$  (alpha) is the degree of  $^{222}\text{Rn}$  loss mainly due to the leakage ( $\text{h}^{-1}$ ) and  $t$  is the elapsed time from the start of the measurement (h). After carrying out the measurements we fitted the growth curves by Origin 6.0 software and obtained the two needed parameters,  $C_{max}$  and  $\alpha$ , which are used in the  $^{222}\text{Rn}$  mass exhalation rate calculations. All of the other needed parameters can be determined easily and the mass exhalation rate of the sample can be calculated by the Eq. 2.

$$E_{mass} = \frac{\lambda + \alpha}{M} C_{max} (V_{net} + V_{det}) \quad (2)$$

where  $E_{mass}$  is the  $^{222}\text{Rn}$  mass exhalation rate ( $\text{Bq kg}^{-1} \text{h}^{-1}$ ),  $M$  is the weight of the sample in the Al-accumulation chamber (kg),  $V_{net}$  is the difference between the volume of the Al-accumulation chamber and the volume of the sample ( $\text{m}^3$ ) and  $V_{det}$  is the sum of internal volume of the RAD7 radon detector, the drying tube and the plastic tubes ( $\text{m}^3$ ).

The *equilibrium method* was recently taken into use at Eötvös University, Budapest, Hungary by Horváth, Á. (Department of Atomic Physics). It is based on measuring the  $^{222}\text{Rn}$  equilibrium concentration by 15 minutes cycles for 4 hours above the sample after 3 weeks (about  $5 T_{1/2}$ ) keeping it in the sealed chamber. The sum of activities in the chamber above the sample and the background activity is equal to the activity of the mixed volume of the measurement circle. Adjusting this equality, the mass exhalation rates can be calculated according to the Eq. 3.

$$E_{mass} = \frac{\lambda}{M} (C_{meas} V_{net} + (C_{meas} - C_{bg}) V_{det}) \quad (3)$$

where  $C_{meas}$  is the average of measured activity concentrations ( $\text{Bq m}^{-3}$ ). In this latter calculation no leakage is assumed which can lead to lower estimations of  $^{222}\text{Rn}$  mass exhalation rates but it is faster for high number of samples.

### Mass exhalation rates of $^{220}\text{Rn}$

In case of  $^{220}\text{Rn}$ , the same samples during the same measurements were analyzed. However, only the equilibrium method (Eq. 3.) can be used because of the short half-life of  $^{220}\text{Rn}$  ( $5 T_{1/2} \approx 5 \text{ min}$ ) which does not allow for the detection of the increasing period of  $^{220}\text{Rn}$  concentration. However, this short half-life also avoids the importance of taking into account the leakage, since  $^{220}\text{Rn}$  has not enough time to leave the measurement circle. The difference in the calculations is that the decay constant ( $\lambda$ ) of  $^{220}\text{Rn}$  is used, the average measured  $^{220}\text{Rn}$  activity concentrations ( $C_{meas}$ ) is corrected by the decay of this isotope during its flow in plastic tubes from the chamber to the RAD7 (DurrIDGE Co. 2000) and that the background concentration can be neglected.

Note that both  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  mass exhalation rates ( $E_{mass}$ ) can be given in  $\text{s}^{-1} \text{kg}^{-1}$  units instead of  $\text{Bq kg}^{-1} \text{h}^{-1}$ , dividing the values by the decay constants ( $\lambda$ ). In the followings, we use this unit for presenting the  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  mass exhalation rates.

### Activity concentrations of $^{226}\text{Ra}$ and $^{232}\text{Th}$

The activity concentrations of  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  of 20 samples powdered after the mass exhalation rate measurements were analyzed by gamma-ray spectrometry using a GC1520 - 7500SL HPGe detector. Gamma rays from powdered samples placed in Al-containers were measured for 16 hours. The detection efficiencies were determined by Monte-Carlo simulation. Absolute transition probabilities come from NuDat2.5 online database. Analysis of  $^{226}\text{Ra}$  was done by its own 186.1 keV peak taking into account that it is overlapping with  $^{235}\text{U}$ 's 185.7 keV peak, and assuming the existence of natural appearance rate of

$^{238}\text{U}$  (99.3%) and  $^{235}\text{U}$  (0.7%) and secular equilibrium between  $^{238}\text{U}$  and  $^{226}\text{Ra}$  (Ebaid et al. 2005). For  $^{232}\text{Th}$  activity concentration determination the  $^{228}\text{Ac}$  peak at 911 keV was used.

### Grain size distributions

Till now grain size distributions of 19 samples were determined by wet sieving and laser grain size analysis. After soaking each of them with 200-500 g weight in distilled water, wet sieving was carried out by Fritsch sieves with diameters of 2, 1, 0.5, 0.25, 0.125 mm and 63  $\mu\text{m}$  coupled with Fritsch Analysette3 sieve shaker. The smallest grain size fraction dispersed in distilled water was collected into a plastic vessel. After sampling this liquid the numbers of different sized grains with diameters below 63  $\mu\text{m}$  were measured by Fritsch Analysette22 laser grain size analyzer.

## RESULTS AND DISCUSSION

### Comparison of the two $^{222}\text{Rn}$ mass exhalation rate measurement methods

Since the time consuming equilibrium method does not allow taking into account the leakage, which produce  $^{222}\text{Rn}$  loss from the measurement circle, it is obvious why it shows less  $^{222}\text{Rn}$  mass exhalation rates (with  $39 \pm 26\%$  on the average) than the growth curve method (Fig. 2.).

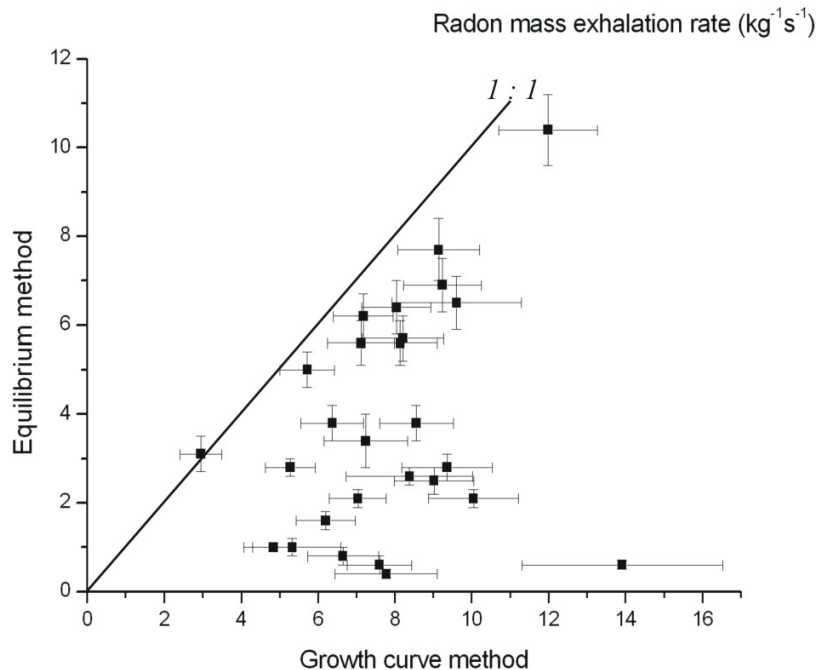


Fig. 2. The  $^{222}\text{Rn}$  mass exhalation rate results given by equilibrium method vs. given by growth curve method: 1:1 line presents the perfect fitting of results. Our data below this line show that equilibrium method measures lower values than growth curve method. (n = 27)

The  $^{222}\text{Rn}$  loss processes are described by the value of  $\alpha$  (Eq. 1. and 2.), which is calculated in growth curve method. Plotting the ratio of equilibrium and growth curve methods' results, which is 1 (unit) in the ideal case, with  $\alpha$  of each experimental setup, we observed that neither of the growth curve and equilibrium  $^{222}\text{Rn}$  mass exhalation rate measurement methods can provide reliable results when the leakage is too revealing. In the followings we ignore data points with  $\alpha$  higher than  $0.02\text{ h}^{-1}$  and also those when one of the results of the methods, in the calculation of the ratio, have high statistical uncertainty (Fig. 3.).

The ratio of equilibrium and growth curve methods' results shows a connection with  $\alpha$  of each experimental setup and approaches 1 (unit) when  $\alpha$  is around 0 (zero) (Fig. 3., red line). In this case the  $^{222}\text{Rn}$  mass exhalation rate results of different methods are the same. This indicates that the time consuming equilibrium method also can provide a good estimation of  $^{222}\text{Rn}$  mass exhalation rate below a certain, controlled degree of  $^{222}\text{Rn}$  loss. We consider the results of the equilibrium method being acceptable when  $\alpha$  is below  $0.005\text{ h}^{-1}$ , in this case the two methods give higher than 75% fitting results (Fig. 3., blue line).

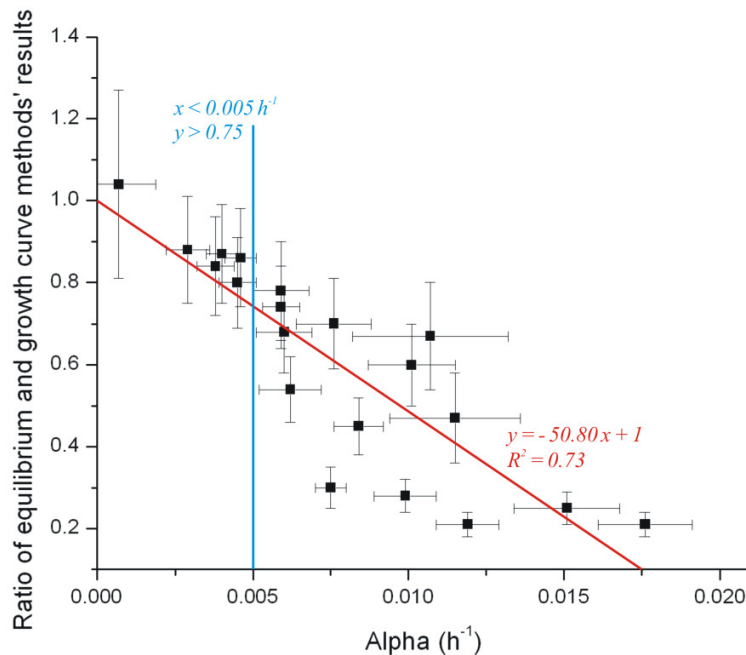


Fig. 3. The ratio of equilibrium and growth curve methods' results vs. alpha ( $\alpha$ ) presenting the  $^{222}\text{Rn}$  loss (leakage) of each experimental setup: the red line shows the connection between the plotted values, i.e. as the leakage decreases, the ratio reaches 1 (unit), when the  $^{222}\text{Rn}$  mass exhalation rate results of different methods are the same. Using the equation of this fitted line, the methods give 75% consistent results when  $\alpha$  is below  $0.005\text{ h}^{-1}$  presented by the blue line. ( $n = 20$ )

These results allow providing a way to carry out higher number of reliable measurements in a shorter period of time via getting know the limits of equilibrium method and the degree of needed leakage control of this experimental setup.

### Results of adobe samples – an overview

The determined and accepted as correct  $^{222}\text{Rn}$  (21 samples) and  $^{220}\text{Rn}$  (27 samples) mass exhalation rates do not show correlation with each other but the averages and standard deviations are similar ( $7.6 \pm 2$ ,  $6.6 \pm 2\text{ s}^{-1}\text{ kg}^{-1}$ , respectively). We compare these results to  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  mass exhalation rates of other different types of building materials (Ujic et al. 2010). In the cited reference most of the samples showed  $^{222}\text{Rn}$  mass exhalation rates below  $10\text{ s}^{-1}\text{ kg}^{-1}$  and all of the samples  $^{220}\text{Rn}$  mass exhalation rates below  $3.3\text{ s}^{-1}\text{ kg}^{-1}$ , Hungarian adobe building material samples show similar  $^{222}\text{Rn}$  but significantly higher  $^{220}\text{Rn}$  exhalation potential.

All of the measured  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  activity concentrations of 20 adobe samples ( $27 \pm 3$ ,  $30 \pm 5\text{ Bq kg}^{-1}$ , respectively) are close to the average values of Hungarian soils (UNSCEAR 2000). Adobe building

materials from E-Mecsek Mts. contain a slightly higher amount of  $^{232}\text{Th}$  ( $34\pm 3 \text{ Bq kg}^{-1}$ ) than the rest of the samples ( $27\pm 4 \text{ Bq kg}^{-1}$ ).

The 19 determined adobe grain size distributions show two characteristic peaks, a smaller one between 1-5  $\mu\text{m}$  and a more dominant one between 10-40  $\mu\text{m}$  grain sizes. Only small differences were found between adobe samples.

The  $28\pm 7 \%$  of  $^{222}\text{Rn}$  and  $22\pm 7 \%$  of  $^{220}\text{Rn}$  – originating from the  $^{226}\text{Ra}$ - and  $^{232}\text{Th}$ -content – can be exhaled by the studied adobe samples. The values fall between 10 and 40 %. These are in the range of normal values 5 to 70 % for rocks and soils (Nazaroff et al. 1988).

Very weak correlation factors were observed for  $^{226}\text{Ra}$  or  $^{232}\text{Th}$  activity concentrations and  $^{222}\text{Rn}$  or  $^{220}\text{Rn}$  mass exhalation rates, respectively. At the same time, it is observed that the 10-40  $\mu\text{m}$  peak of grain size distribution is connected to radon exhalation and the 1-5  $\mu\text{m}$  peak to thoron exhalation, mainly, what confirms that short half-lived  $^{220}\text{Rn}$  can escape mainly from the finest sized grains.

## CONCLUSIONS

- Below a determined and controlled leakage rate ( $\alpha$ ), the tested equilibrium method provides an acceptable estimation of  $^{222}\text{Rn}$  mass exhalation rate on a time consuming way.
- Examined volumes of adobe samples can exhale almost the same average number of  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  and more  $^{220}\text{Rn}$  than other building materials in the cited reference, hence adobe building material potentially can cause elevated indoor  $^{220}\text{Rn}$  concentrations in all of the studied areas.
- Grain size distribution is an important property of adobe building materials which has a different influence on  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  exhalation.

## ACKNOWLEDGEMENT

The authors gratefully thank the help of local people, Angéla Barossné Szőnyi, Ottó Csorba, Győző Jordán, Hedvig Éva Nagy, Botond Papp and Péter Völgyesi, the support of Lithosphere Fluid Research Lab (Eötvös University, Budapest), Doctoral School of Environmental Sciences (Eötvös University, Budapest) and the Organizers of the NSFS-2011 Conference.

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