



# A new fast screening method for estimating building materials hazard indices with correlated inputs

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## Abstract

In this work we investigate a new fast screening method for estimation of chosen hazard indices (*HI*) using correlated inputs dedicated for small 3.00 g samples using a novel  $\mu$ DOSE. The system detects  $\alpha$  and  $\beta$  particles separately, along with  $^{220}\text{Rn}/^{216}\text{Po}$ ,  $^{219}\text{Rn}/^{215}\text{Po}$ ,  $^{212}\text{Bi}/^{212}\text{Po}$  and  $^{214}\text{Bi}/^{214}\text{Po}$  decay pairs. Four separate decay pairs along with  $\alpha$  and  $\beta$  particle count rates are used to quantify decay chains. The excess  $\beta$  count rates is used to quantify the  $^{40}\text{K}$  radioactivity. This provides radionuclide estimates that are correlated—and this correlation is taken into account in calculating hazard indices with their corresponding uncertainties. Calculated hazard indices are verified against state-of-the-art High Resolution Gamma Spectrometry (HRGS) equipped with a High Purity Germanium (HPGe) detector manufactured by Canberra. This research shows that results obtained with the  $\mu$ DOSE system correspond to the results obtained with HRGS and when the activity correlation is taken into account the *HI* uncertainties are similar in value for both methods.

**Keywords** Building materials · Natural radioactivity · Correlated uncertainties ·  $\alpha$  and  $\beta$  counting · Building materials hazard indices

## Introduction

Building materials are derived from Earth's resources, such as soil or rocks, and thus they contain naturally occurring radionuclides from the  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay series, as well as  $^{40}\text{K}$  and, as a result, their use carries a risk of radiation exposure. This exposure can be external from direct gamma radiation exposure or internal from inhalation of radioactive radon/thoron [1]. The latter can be especially harmful in an indoor environment and can contribute to development of lung cancer, if there is no appropriate ventilation [2]. For this reason a number of regulations have been introduced providing so-called hazard indices. For example, to ensure safety regarding the dose acquired exclusively from building materials, activity concentration index value of 1 can be used as a conservative screening tool [3]. Similar

mathematical formulation can be seen in other hazard indices, namely: radium equivalent activity, representative level index, absorbed and annual gamma dose rates, respectively, gamma effective indices, as well as external and internal radiation hazard indices.

The issue of building materials radioactivity is well documented in various works, as listed in Table 1. However, because the  $\mu$ DOSE system is a relatively new setup for estimating  $^{40}\text{K}$  as well as  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{232}\text{Th}$  decay chain members activities [4], no research on hazard indices has been done so far. A 2022 study [5] on the accuracy of the  $\mu$ DOSE system shows good agreement with well-established methods of dosimetry, such as HRGS or thick source alpha counting. The study also mentions that the correlation of results provided by the  $\mu$ DOSE system improves precision of dose rate estimation; however, this correlation has not been investigated on its own and nor has its significance in comparison to the HRGS method.

The aim of this work is to test how uncertainties of hazard indices are influenced, if radionuclide estimates are correlated. This is tested with two independent setups of HRGS and the  $\mu$ DOSE system that provide uncorrelated and correlated estimates respectively. In addition, advantages of reducing hazard indices uncertainties are investigated by

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using correlated radionuclide estimates and drawbacks as errors are induced by disequilibrium in radioactive decay chains.

## Experimental

### Materials

All 7 samples that were investigated in this work are frequently used building materials of known origin: clay bricks ( $\text{SiO}_2$ , feldspars), beach rock (sand and/or gravel,  $\text{CaCO}_3$ ) and sand ( $\text{SiO}_2$ ). Samples were dried in a drying chamber at an elevated temperature of 80 °C for several days. During drying sample masses were monitored to ensure water was removed. Every sample material was divided into two subsamples for  $\mu\text{DOSE}$  and HRGS measurement. Detailed descriptions of sample preparation for these systems is provided below.

Subsamples and reference materials for  $\mu\text{DOSE}$  system measurements were ground in a Fritsch Pulverisette 6 planetary mill for 45 min at 200 rpm each to a fine powder, then 3.00 g of prepared material was placed onto 70 mm diameter sample discs and measured using the  $\mu\text{DOSE}$  system.

Subsamples and reference materials for HRGS measurements were put and sealed in  $\gamma\text{BEAKERS}$  [21, 22] (ca. 100 g samples) and then stored for a period of about 30 days to obtain secular equilibrium to avoid bias that arises from  $^{222}\text{Rn}$  emanation [23–27]. Subsamples were measured for 24–48 h, times varying depending on the activities of each individual sample for obtaining optimal count rate statistics.

### $\mu\text{DOSE}$ system— $\alpha/\beta$ and delayed coincidence counting

For  $\alpha/\beta$  particle measurements  $\mu\text{DOSE}$  system was used, described in detail in [4, 5]. It is designed for detecting  $\alpha$ - $\alpha$  ( $^{220}\text{Rn}/^{216}\text{Po}$ ,  $^{219}\text{Rn}/^{215}\text{Po}$ ) and  $\beta$ - $\alpha$  ( $^{212}\text{Bi}/^{212}\text{Po}$ ,  $^{214}\text{Bi}/^{214}\text{Po}$ ) decay pairs and identifying the isotope pair the particles come from, based on the characteristic time intervals between the subsequent particle emissions. These pairs can later be used to determine  $^{238}\text{U}$ ,  $^{235}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  content. This is done with assumption of secular equilibrium, where the activities of the decay pairs are equal to the activities of corresponding decay chains parent radioisotopes:  $^{238}\text{U}$ ,  $^{235}\text{U}$  or  $^{232}\text{Th}$ . The remainder of the emitted  $\beta$  particles are assigned to  $^{40}\text{K}$ . The system was calibrated using IAEA-RGU-1, IAEA-RGTh-1 and IAEA-RGK-1 [28] reference materials obtained from the International Atomic Energy Agency. In this work a system that was calibrated for 3.00 g was used.

Background activity was obtained for a 3.00 g background plastic disc placed accordingly in the sample holder. The background measurement lasted ca. 48 h and was performed in the same laboratory conditions as were the samples.

### High resolution gamma spectrometry

Gamma spectrometry was performed with a Canberra HPGe detector with FWHM of 1.8 keV and relative efficiency 40% at 1332 keV. The HRGS system was calibrated with IAEA-RGU-1, IAEA-RGTh-1 and IAEA-RGK-1 [28] reference materials. Activities were calculated for selected energy lines: 295.2 keV, 351.9 keV ( $^{214}\text{Pb}$ ) and 609.3 keV ( $^{214}\text{Bi}$ ) for uranium series; 338.3 keV, 911.2 keV ( $^{228}\text{Ac}$ ) and 583.2 keV ( $^{208}\text{Tl}$ ) for thorium series; and 1460.8 keV ( $^{40}\text{K}$ ) (data obtained from NuDat 3.0).

Background activity was obtained through a 167 h measurement of an empty  $\gamma\text{BEAKER}$  in an identical laboratory setting in which samples were measured.

## Theoretical

### Radiation hazard indices

To determine whether building materials meet set standards, are within established norms and are safe to use, a number of hazard indices were devised. In many cases hazard indices are estimated from  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  concentrations and provide a simplified information for a given risk factor. Frequently used indices are calculated as a linear combination of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  radionuclides contents in the following way:

$$HI = S_{\text{Ra}}A_{\text{Ra}} + S_{\text{Th}}A_{\text{Th}} + S_{\text{K}}A_{\text{K}} \quad (1)$$

where  $HI$  is the hazard index,  $A_{\text{Ra}}$ ,  $A_{\text{Th}}$  and  $A_{\text{K}}$  are  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  concentrations expressed in  $\text{Bq}\cdot\text{kg}^{-1}$  respectively,  $S_{\text{Ra}}$ ,  $S_{\text{Th}}$  and  $S_{\text{K}}$  are parameters for a given hazard index. List of hazard indices,  $S_{\text{Ra}}$ ,  $S_{\text{Th}}$ ,  $S_{\text{K}}$  parameters and a brief summary is provided in Table 1.

### Uncertainty propagation for uncorrelated and correlated inputs

For uncorrelated inputs given hazard index uncertainty ( $\sigma_{\text{Th}}$ ) can be is calculated as:

$$\sigma_{HI}^2 = (S_{\text{Ra}}\sigma_{\text{Ra}})^2 + (S_{\text{Th}}\sigma_{\text{Th}})^2 + (S_{\text{K}}\sigma_{\text{K}})^2 \quad (2)$$

where  $\sigma_{Ra}$ ,  $\sigma_{Th}$  and  $\sigma_K$  are uncertainties of  $^{226}Ra$ ,  $^{232}Th$  and  $^{40}K$  activities. Usually HRGS measurements are considered to be uncorrelated. However for correlated inputs hazard index uncertainty should be calculated using the formula that takes into account correlations:

$$\sigma_{HI}^2 = [S_{Ra} \ S_{Th} \ S_K] \begin{bmatrix} \sigma_{Ra}^2 & \sigma_{Ra-Th} & \sigma_{Ra-K} \\ \sigma_{Th-Ra} & \sigma_{Th}^2 & \sigma_{Th-K} \\ \sigma_{K-Ra} & \sigma_{K-Th} & \sigma_K^2 \end{bmatrix} \begin{bmatrix} S_{Ra} \\ S_{Th} \\ S_K \end{bmatrix} \quad (3)$$

where non-diagonal  $\sigma$  elements are covariances of elements given in subscripts. Depending on sign and value of non-diagonal elements the final  $HI$  uncertainty will give different values. For uncorrelated inputs non diagonal  $\sigma$  are zeros and Eqs. 2 and 3 provide the same values.

## Results and discussion

### Correlated $^{226}Ra$ , $^{232}Th$ and $^{40}K$ estimates

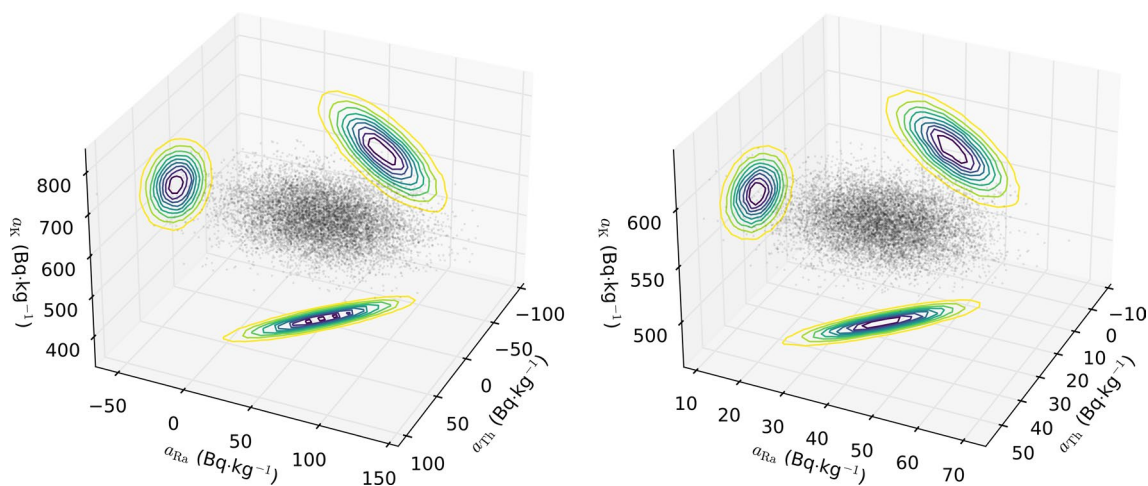
The  $\mu$ DOSE system provides correlated  $^{226}Ra$ ,  $^{232}Th$  and  $^{40}K$  estimates. This arises because the system detects  $\alpha$  particles that can be emitted from decay chains while  $\beta$  particles are emitted from decay chains and  $^{40}K$ . Four decay pairs  $^{220}Rn/^{216}Po$ ,  $^{219}Rn/^{215}Po$ ,  $^{212}Bi/^{212}Po$ ,  $^{214}Bi/^{214}Po$  detected by the  $\mu$ DOSE system, provide information on relative  $^{226}Ra$ ,  $^{232}Th$  and  $^{40}K$  content. Detailed calculation procedure is provided in [29]. Correlations of  $^{226}Ra$ ,  $^{232}Th$  and  $^{40}K$  contents are visualised in Fig. 1 where 100 k points were drawn with respect to correlations determined by  $\alpha$ ,  $\beta$  and four decay pairs counting statistics.

**Table 1** Radiation hazard indices; names, parameters and descriptions of 7 hazard indices, along with references to other works where the indices were studied

$HI$ —Hazard index	$S_{Ra}$	$S_{Th}$	$S_K$	Short description	References
Radium equivalent activity ( $Ra_{eq}$ )	1	1.43	0.077	Radium equivalent activity $Ra_{eq}$ helps determine the purpose a given building material can be used for, e.g. homes, industries, roads/bridges, foundations of non-residential constructions or whether it is not suitable for any type of construction use at all	[6–17]
Representative level index ( $RLI$ )	$150^{-1}$	$100^{-1}$	$1500^{-1}$	Representative level index $RLI$ allows for estimating gamma radiation levels associated with concentrations of specific nuclides	[7–10, 12, 16]
Absorbed gamma dose ( $D_r$ in $nGy \cdot h^{-1}$ )	0.92	1.1	0.08	Absorbed gamma dose rate $D_r$ is a value dependent on “average” room parameters ( $4 \times 5 \times 2.8$ m) with wall and ceiling thickness at 20 cm and their density of $2.350 \text{ kg} \cdot \text{m}^{-3}$ (for concrete)	[8, 10, 12, 14, 17, 18]
Annual effective dose rate* ( $H_R$ in $mSv \cdot a^{-1}$ )	$1.13 \cdot 10^{-3}$	$1.13 \cdot 10^{-3}$	$9.82 \cdot 10^{-5}$	Annual effective dose rate $H_R$ is a parameter dependent on the absorbed gamma dose rate multiplied by a conversion factor ( $0.7 \text{ Sv} \cdot \text{Gy}^{-1}$ ) and outdoor occupancy factor (0.2)	[8, 10, 12]
Activity concentration index	$300^{-1}$	$200^{-1}$	$3000^{-1}$	Presented in the form proposed by the European Commission, the value of $I_\gamma$ is an indicator if material in use exceeds the established safety levels that depend on the dose criterion and the type of use as well as the amount of material	[6, 8, 10, 12, 14–20]
External radiation hazard index	$370^{-1}$	$258^{-1}$	$4810^{-1}$	Both internal ( $H_{in}$ ) and external ( $H_{ex}$ ) radiation hazard index has a function similar to the activity concentration index, but they take into account the way radiation interacts with the human body (inhalation or external influence)	[7–10, 12, 13, 15, 16, 18]
Internal radiation hazard index	$185^{-1}$	$259^{-1}$	$4810^{-1}$		

Parameters are applicable for radionuclides concentrations expressed in  $\text{Bq} \cdot \text{kg}^{-1}$

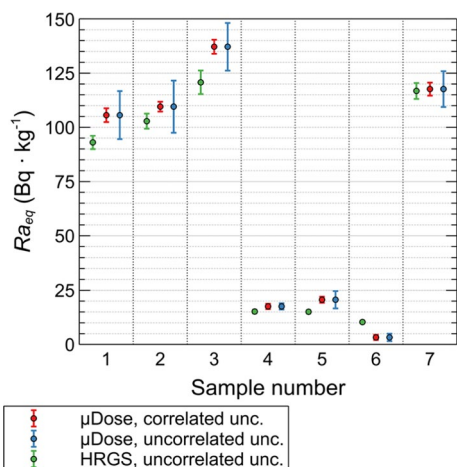
\*Usually presented as:  $H_R = D_r \cdot 8.766 \cdot 0.2 \cdot 0.7 \cdot 10^{-6}$



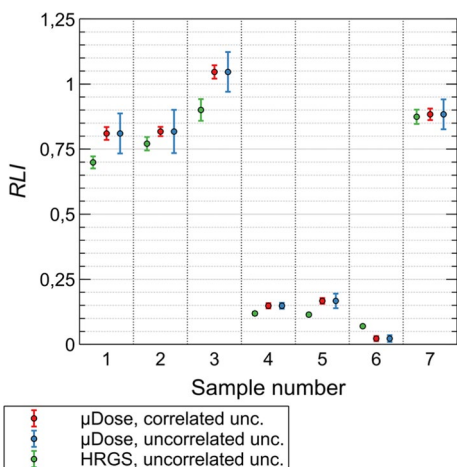
**Fig. 1** Two  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  compositions. Isolines are showing projected 2-D PDF contour-plots drawn from a multivariate normal distribution for the  $\mu\text{DOSE}$  system for 1 h (a) and 10 h (b) measurements

**Table 2** Numerical data of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  radionuclides contents from  $\mu\text{DOSE}$  and HRGS

Sample number	$\mu\text{DOSE}$	$\begin{bmatrix} \sigma_{\text{Ra-226}}^2 & \sigma_{\text{Ra-226, Th-232}} & \sigma_{\text{Ra-226, K-40}} \\ \sigma_{\text{Th-232, Ra-226}} & \sigma_{\text{Th-232}}^2 & \sigma_{\text{Th-232, K-40}} \\ \sigma_{\text{K-40, Ra-226}} & \sigma_{\text{K-40, Th-232}} & \sigma_{\text{K-40}}^2 \end{bmatrix}$ terms in $\text{Bq}^2\cdot\text{kg}^{-2}$ each	HRGS		
	$[\mu_{\text{Ra-226}} \ \mu_{\text{Th-232}} \ \mu_{\text{K-40}}]$ terms in $\text{Bq}\cdot\text{kg}^{-1}$ each		$^{226}\text{Ra}$ , $\text{Bq}\cdot\text{kg}^{-1}$	$^{232}\text{Th}$ , $\text{Bq}\cdot\text{kg}^{-1}$	$^{40}\text{K}$ , $\text{Bq}\cdot\text{kg}^{-1}$
1	[29.83 18.56 639.35]	$\begin{bmatrix} 47.15 & -38.82 & 35.77 \\ -38.82 & 34.92 & -38.56 \\ 35.77 & -38.56 & 724.39 \end{bmatrix}$	$25.21 \pm 0.89$	$23.4 \pm 1.7$	$446 \pm 22$
2	[36.42 24.17 500.63]	$\begin{bmatrix} 55.81 & -46.63 & 44.42 \\ -46.63 & 41.69 & -45.52 \\ 44.42 & -45.52 & 601.58 \end{bmatrix}$	$25.55 \pm 0.89$	$28.8 \pm 2.0$	$469.0 \pm 22.7$
3	[33.20 31.11 772.12]	$\begin{bmatrix} 45.74 & -37.99 & 35.73 \\ -37.99 & 34.40 & -38.42 \\ 35.73 & -38.42 & 804.74 \end{bmatrix}$	$28.4 \pm 1.6$	$36.9 \pm 2.8$	$513 \pm 44$
4	[1.39 0.28 204.49]	$\begin{bmatrix} 0.59 & -0.12 & -0.85 \\ -0.12 & 0.16 & -0.31 \\ -0.85 & -0.31 & 194.19 \end{bmatrix}$	$2.66 \pm 0.17$	$3.00 \pm 0.29$	$106.9 \pm 8.4$
5	[0.0 4.77 179.01]	$\begin{bmatrix} 6.65 & -4.75 & 2.90 \\ -4.75 & 3.94 & -4.11 \\ 2.90 & -4.11 & 196.24 \end{bmatrix}$	$3.64 \pm 0.24$	$3.68 \pm 0.35$	$80.0 \pm 6.7$
6	[0.59 1.88 0.0]	$\begin{bmatrix} 0.83 & -0.44 & -0.20 \\ -0.44 & 0.56 & -1.10 \\ -0.20 & -1.10 & 160.78 \end{bmatrix}$	$4.93 \pm 0.26$	$4.06 \pm 0.41$	$-5.1 \pm 2.3$
7	[33.80 28.06 567.43]	$\begin{bmatrix} 20.30 & -19.73 & 24.85 \\ -19.73 & 22.78 & -33.20 \\ 24.85 & -33.20 & 228.52 \end{bmatrix}$	$27.90 \pm 0.86$	$33.9 \pm 2.1$	$525 \pm 24$



**Fig. 2** Radium equivalent activity ( $Ra_{eq}$ ) with uncertainties calculated for all samples

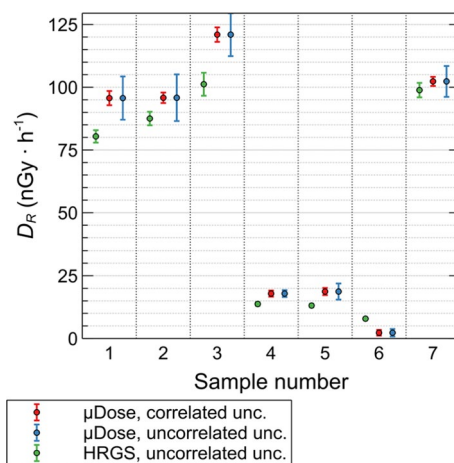


**Fig. 3** Representative level index ( $RLI$ ) with uncertainties calculated for all samples

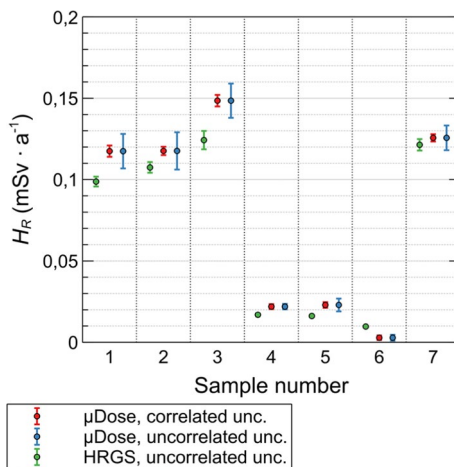
### Hazard indices and their uncertainties

Hazard indices and their uncertainties were estimated for the  $\mu$ DOSE system and HRGS using radionuclide content. Numerical values are provided in Table 2. In the case of the  $\mu$ DOSE system uncertainties are calculated using two approaches: first neglects correlations (Eq. 2) and the second approach takes correlations into account (Eq. 3). This is illustrated on Figs. 2, 3, 4, 5, 6, 7 and 8 which provide comparison of hazard indices values and their uncertainties.

Data presented in Figs. 2, 3, 4, 5, 6, 7 and 8 shows that the uncorrelated uncertainties for  $\mu$ DOSE measurements are greater than the correlated uncertainties. This shows that by including the correlation between the activities of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the estimation of hazard indices



**Fig. 4** Absorbed gamma dose ( $D_R$ ) with uncertainties calculated for all samples

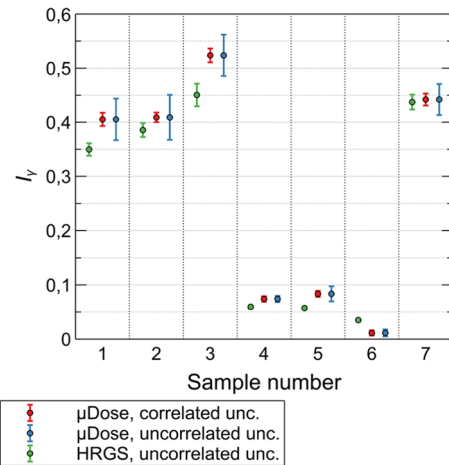


**Fig. 5** Annual effective dose rate ( $H_R$ ) with uncertainties calculated for all samples

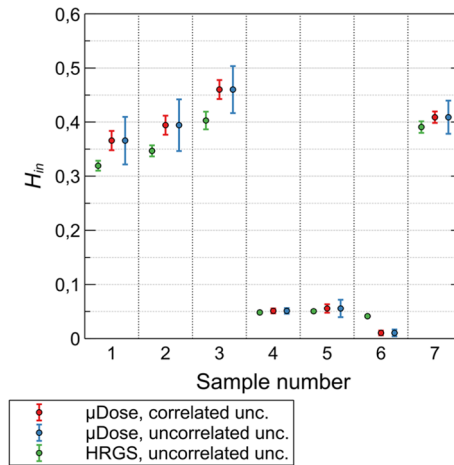
uncertainties, the provided results are more precise. This is due to the fact that several elements within the covariance matrices (Eq. 3) are negative which contributes to the lessening of the uncertainties. The correlated uncertainties from  $\mu$ DOSE measurements are also similar in value to the ones obtained from HRGS, thus proving that the measurement accuracy is maintained regardless of the chosen method.

### Screening—measurement time, uncertainty and type II error

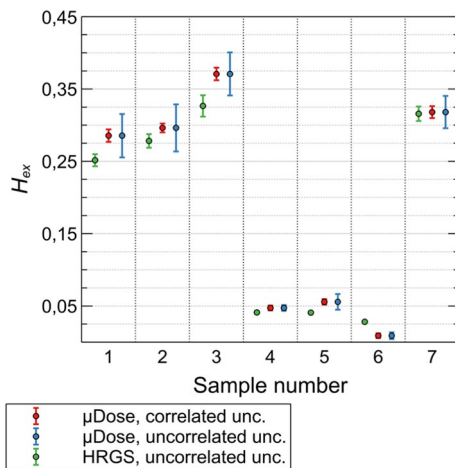
In some cases sample mass and sample throughput can be a limiting factor; therefore, an investigation on how uncertainty changes as a function of time was performed. The  $\mu$ DOSE system offers the possibility of measuring small samples and in this work the system was calibrated



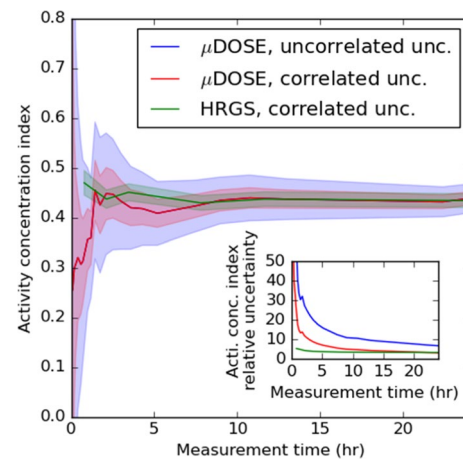
**Fig. 6** Activity concentration index  $I_\gamma$  with uncertainties calculated for all samples



**Fig. 8** Internal radiation hazard index ( $H_{in}$ ) with uncertainties calculated for all samples



**Fig. 7** External radiation hazard index ( $H_{ex}$ ) with uncertainties calculated for all samples



**Fig. 9** Activity concentration index value uncertainty as a function of measurement time for HRGS (100 g samples) and  $\mu$ DOSE system (3.00 g samples)

for 3.00 g samples, whereas HRGS was calibrated for 100 g. Figure 9 shows activity concentration index value and uncertainty as a function of measurement time for HRGS and  $\mu$ DOSE system. In both cases uncertainties encompass counting statistics, sample mass and reference materials uncertainties which are the main uncertainty contributors for prolonged measurements. Despite large mass differences,  $\mu$ DOSE uncertainties that take into account correlations are comparable with HRGS (Fig. 9). For the first few hours of the measurement activity concentration index uncertainty is relatively large for  $\mu$ DOSE. After 10 h (Fig. 9) uncertainties from the  $\mu$ DOSE and HRGS systems are at the same level, which further proves that the  $\mu$ DOSE system is a reliable screening tool and can be an alternative to conventional HRGS. Nevertheless, algorithms that

are used for measuring net peak area do not operate well on poorly defined baseline and therefore first peak quantification, used for activity estimation, was available after 46 min of measurements and required manually adjusting regions for peak detection.

In case of screening building materials for activity concentration index (Fig. 9) it can be observed that after ca. 1 h of measurement its value is well below recommended value 1 [3]. For shorter measurements HRGS counting statistics (discussed in the paragraph above) and  $\mu$ DOSE counting statistics do not provide definitive material classification. This finding has substantial implications for future studies, especially when sample mass or measurement time is a limiting factor. What is more, understanding the differences between

correlated and uncorrelated uncertainties allows researchers to take advantage of correlated ones.

While screening *HI* there are two classification errors: type I error “false positive” and type II error “false negative”. If *HI* value is above threshold, additional and prolonged measurement should be made to resolve this. In case of type II error, this can be difficult to detect and can have more significant consequences. For example, given *HI* can falsely be assumed to be within limits. This can happen because there are several factors, not related to counting statistics, that contribute to this. For example  $^{238}\text{U}$  decay chain can be in disequilibrium, [25, 30, 31] or sample chemical composition can be unknown. This can cause issues in both HRGS as well as in the  $\mu\text{DOSE}$  system and unfortunately those factors are not controlled routinely due to cost of additional measurements. Nevertheless, *HI* screening limits can be set to reduce the risk of type II errors at the expense of type I error.

The proposed methodology and obtained findings allow to bridge a gap in the existing literature, especially in the context of improving the throughput with measurement systems like  $\mu\text{DOSE}$ . The precision of conducted measurements ensures that safety thresholds are reliably met, reducing risks associated with type I and II errors.

## Conclusions

In this study, values of selected hazard indices of building materials were compared using the  $\mu\text{DOSE}$  system for 3.00 g samples against the HRGS calibrated for 100 g samples. The primary objective was to understand the significance of the correlation of activities of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  provided uniquely by the  $\mu\text{DOSE}$  system. The findings indicated that the uncertainties in hazard indices from both the HRGS and  $\mu\text{DOSE}$  systems, when correlations are considered, are comparable in value. Furthermore, accounting for these correlations offers a substantial enhancement in the precision of results compared to when they are disregarded.

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**Author contributions** JR: Conceptualization, Methodology, Formal analysis, Investigation, Writing—Original Draft; JP: Conceptualization, Methodology, Formal analysis, Investigation, Writing—Original Draft; KT: Conceptualization, Methodology, Formal analysis, Investigation, Writing—Original Draft, Supervision; GP: Conceptualization, Methodology, Formal analysis, Investigation, Writing—Original Draft, Supervision; AS: Investigation.

## Declarations

**Conflict of 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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