µDose: a compact system for environmental radioactivity and dose rate measurement

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Abstract

µDose is a novel compact analytical instrument for assessing low level $^{238}$U, $^{235}$U, $^{232}$Th decay chains and $^{40}$K radioactivity. The system is equipped with a dual $\alpha/\beta$ scintillator allowing to discriminate between $\alpha$ and $\beta$ particles. The unique build-in pulse analyzer measures the amplitude of each individual pulse, its shape and the time interval between subsequent pulses. This allows the detection of pulse pairs arising from subsequent decays of $^{214}$Bi/$^{214}$Po, $^{220}$Rn/$^{216}$Po, $^{212}$Bi/$^{212}$Po and $^{219}$Rn/$^{215}$Po. The obtained $\alpha$ and $\beta$ counts and four separate decay pair counts are used to assess the $^{238}$U, $^{235}$U, $^{232}$Th and $^{40}$K specific radioactivities in the measured samples through the use of radioactivity standards. The µDose system may be equipped with various photomultipliers and counting containers to assess radionuclide concentrations of samples of masses ranging between 0.4 g and 4 g. As a result, the user can customize the system to their needs and maximize the instrument's performance. The system is controlled by a
dedicated software with a graphical user interface and modules for system calibration, data visualization, specific radioactivity calculations and dose rate determination using the infinite matrix assumption.

**Introduction**

Thick source alpha counting (TSAC) has been developed\(^1\) to provide a relatively easy and inexpensive way of assessing the low level content of alpha emitters in biological tissues. Later this technique was adapted to determining the uranium and thorium content in samples of fired ceramics for assessing the annual dose for luminescence dating.\(^2\) In TSAC, the powdered sample is placed on a plastic sheet coated with a very thin layer of the scintillator ZnS:Ag attached to its surface. Alpha particles, emitted by U and Th series members, upon reaching the screen produce scintillations with practically 100% efficiency. In order to estimate the contribution from the U and Th series, additionally the so-called slow and sometimes fast pairs are counted. Such pairs are fast successions of counts due to the short lived \(^{216}\)Po \((t_{1/2} = 0.145 \text{ s})\) in the \(^{232}\)Th series and \(^{215}\)Po \((t_{1/2} = 1.78 \text{ ms})\) in the \(^{235}\)U series. Today TSAC technique is widely used\(^3\)--\(^10\) for assessing the annual dose in trapped charge dating techniques (optically stimulated luminescence, thermoluminescence, electron spin resonance). However, there are some significant limitations in the conventional TSAC technique. For example, the activity of \(^{40}\)K which is a major dose contributor to the dose rate in environmental samples, cannot be determined using this technique and an independent determination by different means is usually performed.\(^11\)--\(^14\) Additionally, the \(^{238}\)U decay chain may cause problem due to possible disequilibrium.\(^15\)--\(^17\) Another drawback is the influence of sample's reflectance on the TSAC efficiency which can cause up to 6% error in an apparatus setting proposed in ref \(^2\).

In addition to the TSAC technique an alternative method of \(\beta\) counting was proposed.\(^18\) This is a much more rapid technique of annual dose rate determination, however it cannot assess the specific radioactivities of \(^{238}\)U, \(^{235}\)U, \(^{232}\)Th decay chains or \(^{40}\)K.\(^18\)--\(^19\)
In the current work, we describe a novel system - μDose - that opens up new possibilities and removes limitations described in the previous paragraph. This is done through the employment of a dual α/β scintillator module, a new measurement setup, a new pulse analyzer unit, and advanced data processing. The system allows detection of two additional β/α decay pairs in addition to the above mentioned α/α pairs. The first pair arises in the $^{232}$Th series from subsequent decays of $^{212}$Bi and $^{212}$Po where $^{212}$Po has a half-life of 299 ns. The second β/α pair arises in the $^{238}$U series from subsequent decays of $^{214}$Bi and $^{214}$Po where $^{214}$Po has a half-life of 164 µs. Therefore, four decay pairs can be used to assess the specific activity of thorium and uranium decay chains as well as potassium activity.

The μDose system is designed with emphasis on natural radioactivity measurement and the software is equipped with modules for dose rate measurement that is dedicated to trapped charge dating. With some modifications system may be adapted for other purposes as well.

In the following sections we provide a detailed description of the μDose system, its performance, decay chain activity measurement method and $^{40}$K assessment.

**Experimental Section**

**System construction**

μDose is a very compact system as it takes just over 20 cm × 20 cm of desk space. The entire electronics, including a stable high voltage power supply, a photomultiplier and a pulse analyzer are built into the system and no additional components except a PC (which can control several such systems) are required for system operation. In the μDose system, the sample is placed in a dedicated sample container which is placed below the photomultiplier (Figure 1). The container is equipped with a dual reusable α and β scintillator module covered by a 0.2 µm replaceable silver foil. The silver foil is easily penetrated by β and the vast majority of α particles emitted by the sample. The silver layer increases the number of photons that reach the photomultiplier (PMT) and it also removes the measurement
variability that arises from the sample’s reflectance. The sample container is gas-tight to prevent radon migration from and into it. The sample itself has a geometry of a thin disk which matches the diameter of the photocathode. Depending on the expected sample mass, the system can accommodate PMT's which have photocathode diameter from 30 up to 70 mm.

![Block diagram of the µDose system.](image)

**Figure 1:** Block diagram of the µDose system.

**Electronics**

The pulse analyzer has been described in detail before,\textsuperscript{20} therefore here only a brief description is given. The $\alpha$ and $\beta$ particles produce scintillations in two different scintillator layers. The generated pulse shapes are different for each of the two scintillators, permitting the identification of the source particle of each pulse. This shape is preserved by the PMT and amplifier where the scintillations are transformed into electrical pulses and significantly amplified. The pulse analyzer detects the incoming voltage pulses (Figure 1) and stores them as series of ADC values that represent each pulse. These pulse data are time-stamped and stored for further processing. The acquired data are then transferred to the computer and processed by a dedicated algorithm that determines the pulse height, the pulse shape and the time when each pulse appeared. The pulse height and the pulse shape allow to
discriminate between α and β induced pulses. In addition, the algorithm is also capable of identifying pulses that do not match neither α nor β particles enabling the removal of background pulses arising from electrical noise or other interfering sources. The software also deconvolutes piled up pulses from decays that appeared within a small time interval (ca. 100 ns).

The electronics has a built-in high stability, low ripple, high voltage supply. This is controlled and monitored for system stability by the software. To protect the PMT, the high voltage is automatically switched off when the drawer with the sample container is opened.

α, β and decay pairs detection

Figure 2 shows a typical 2D pulse height vs. pulse shape histogram where the color indicates relative frequency of the recorded pulses obtained from sample 1. Typical counting windows for classifying α and β induced pulses are marked by dashed lines on the same plot.

![Figure 2: Data for a sample no. 1 measured for 44 hours. 2D pulse height vs. pulse histogram where the colour indicates relative frequency of pulses appearing.](image)

In environmental samples, the mean time interval between counts is virtually always much longer than the half-lifes of $^{216}\text{Po}$ ($t_{1/2} = 0.145$ s), $^{215}\text{Po}$ ($t_{1/2} = 1.78$ ms), $^{212}\text{Po}$ ($t_{1/2} = 299$ ns) and $^{214}\text{Po}$ ($t_{1/2} = 164$ µs). This means that after pulses arising from decays of $^{220}\text{Rn}$, $^{219}\text{Rn}$, $^{212}\text{Bi}$ and $^{214}\text{Bi}$, the corresponding daughter radioisotopes will induce pulses appearing in a fast succession giving rise to the so-called decay pairs. In practice, μDose allows identification and counting of those four decay pairs, namely, α-α for $^{220}\text{Rn}/^{216}\text{Po}$ and $^{219}\text{Rn}/^{215}\text{Po}$, and
\(\beta-\alpha\) for \(^{212}\text{Bi}/^{212}\text{Po}\) and \(^{214}\text{Bi}/^{214}\text{Po}\). To detect those decay pairs the system builds a time interval distribution from the recorded time intervals between events classified as \(\beta\) pulses followed in a quick succession of an \(\alpha\) pulse (Figure 3a, b and c). This allows to reveal the exponential decay arising from \(^{212}\text{Bi}/^{212}\text{Po}\) (Figure 3a) and \(^{214}\text{Bi}/^{214}\text{Po}\) (Figure 3b) decay pairs. Similarly, time interval distribution between events classified as \(\alpha\) (Figure 3d, e and f) allow to reveal the exponential decay arising from \(^{219}\text{Rn}/^{215}\text{Po}\) (Figure 3d) and \(^{220}\text{Rn}/^{216}\text{Po}\) (Figure 3e) decay pairs. Figure 3c and f show remaining \(\beta-\alpha\) and \(\alpha-\alpha\) time intervals arising from random decays. On each inset in Figure 3 the fitted signal (sig) and background (bg) are marked. Note that background in Figure 3a, b, c, f is virtually absent.

Figure 3: Data for sample no. 1 measured for 44 hours. Distributions for \(\beta-\alpha\) time intervals in ranges a) 0-3.6 \(\mu\)s, b) 3.6-2100 \(\mu\)s, c) > 2100 \(\mu\)s and \(\alpha-\alpha\) time intervals in ranges d) 0.5-21 ms, e) 21 ms - 1.7 s, c) > 1.7 s. Measured time intervals are shown by the stepped lines, the fitted signal (s) and background (b) are depicted by the filled areas. a), b), d) and e) plots reveal subsequent decays of \(^{220}\text{Rn}/^{216}\text{Po}\), \(^{219}\text{Rn}/^{215}\text{Po}\), \(^{212}\text{Bi}/^{212}\text{Po}\) and \(^{214}\text{Bi}/^{214}\text{Po}\), respectively. d) and g) show remaining, purely random \(\beta-\alpha\) and \(\alpha-\alpha\) time interval distributions. Note the differing horizontal axis scales on plots a-f.
\(^{238}\text{U}, ^{235}\text{U}, ^{232}\text{Th}\) and \(^{40}\text{K}\) activity assessment

The four decay pairs count rates can be used to directly determine the activity per unit of mass

\[
\begin{align*}
r_{\text{Bi}^{\text{212}}/\text{Po}^{\text{212}}} &= k_{\text{Bi}^{\text{212}}/\text{Po}^{\text{212}}} a_{\text{Bi}^{\text{212}}/\text{Po}^{\text{212}}}, \\
r_{\text{Bi}^{\text{214}}/\text{Po}^{\text{214}}} &= k_{\text{Bi}^{\text{214}}/\text{Po}^{\text{214}}} a_{\text{Bi}^{\text{214}}/\text{Po}^{\text{214}}}, \\
r_{\text{Rn}^{\text{220}}/\text{Po}^{\text{216}}} &= k_{\text{Rn}^{\text{220}}/\text{Po}^{\text{216}}} a_{\text{Rn}^{\text{220}}/\text{Po}^{\text{216}}}, \\
r_{\text{Rn}^{\text{219}}/\text{Po}^{\text{215}}} &= k_{\text{Rn}^{\text{219}}/\text{Po}^{\text{215}}} a_{\text{Rn}^{\text{219}}/\text{Po}^{\text{215}}},
\end{align*}
\]

(1)

Here \(r\) are the net count rates of decay pairs indicated in subscripts, \(k\) are calibration parameters for the given system and the given decay pairs indicated in subscripts, and \(a\) are specific activities of decay pairs indicated in subscripts. Equations (1) hold for samples with atomic compositions similar to that of the used calibration standards. Equations (1) remain valid regardless of the state of secular equilibrium in the measured material.

In many cases, samples are close to secular equilibrium and the user may assume the following relationships

\[
\begin{align*}
a_{\text{Th}^{\text{232}}} &= a_{\text{Bi}^{\text{212}}/\text{Po}^{\text{212}}} = a_{\text{Rn}^{\text{220}}/\text{Po}^{\text{216}}}, \\
a_{\text{U}^{\text{238}}} &= a_{\text{Bi}^{\text{214}}/\text{Po}^{\text{214}}}, \\
a_{\text{U}^{\text{235}}} &= a_{\text{Rn}^{\text{219}}/\text{Po}^{\text{215}}}, \\
r_\alpha &= k_{\alpha,\text{Th}^{\text{232}}} a_{\text{Th}^{\text{232}}} + k_{\alpha,\text{U}^{\text{238}}} a_{\text{U}^{\text{238}}} + k_{\alpha,\text{U}^{\text{235}}} a_{\text{U}^{\text{235}}}, \\
r_\beta &= k_{\beta,\text{Th}^{\text{232}}} a_{\text{Th}^{\text{232}}} + k_{\beta,\text{U}^{\text{238}}} a_{\text{U}^{\text{238}}} + k_{\beta,\text{U}^{\text{235}}} a_{\text{U}^{\text{235}}} + k_{\beta,\text{K}^{\text{40}}} a_{\text{K}^{\text{40}}}. 
\end{align*}
\]

(2)

Here \(r_\alpha\) and \(r_\beta\) are the \(\alpha\) and \(\beta\) net count rates. Equations (1) and (2) enable the calculation of the decay chain specific activities in the sample. These equations allow to extract the activity of pure \(\beta\) emitters. In (2), it is assumed that \(^{40}\text{K}\) is the major contributor, which is true for most environmental samples. However, it needs to be borne in mind that in environmental samples also other pure \(\beta\) emitters can be found, e.g. natural \(^{87}\text{Rb}\) or anthropogenic \(^{137}\text{Cs}\).\(^{22-27}\) This needs to be considered individually for each sample as it may
introduce an error in the $^{40}\text{K}$ assessment. E.g., ref 28 assumed a contribution from $^{87}\text{Rb}$ at the level of 50 ppm of natural rubidium per 1% of potassium.

Eqs. (1) and (2) can be further restricted using a known $^{238}\text{U}/^{235}\text{U}$ isotopic ratio$^{29,30}$

$$\frac{^{238}\text{U}}{^{235}\text{U}} = \frac{a_{^{238}\text{U}}/\lambda_{^{238}\text{U}}}{a_{^{235}\text{U}}/\lambda_{^{235}\text{U}}} = 137.88.\quad (3)$$

where $\lambda$ is decay constant of radioisotope indicated in subscript. Eq. (3) removes one degree of freedom and allows obtaining more precise results. In the natural environment, the ratio $^{238}\text{U}/^{235}\text{U}$ ratio can vary by up to ca. 5‰.$^{29-33}$ This however is consistent enough to assume it as constant given the measurement accuracy and precision.

**System calibration**

The $\mu$Dose system needs to be calibrated with reference materials of known radioactivities, as well as a background sample. In the current work we use IAEA-RGU-1, IAEA-RGTh-1, and IAEA-RGK-1 standards from the International Atomic Energy Agency.$^{34}$ The IAEA-RGU-1 and IAEA-RGTh-1 are produced using uranium and thorium ores that are mixed with floated silica powder. Decay chains present in those reference materials can be considered to be in secular equilibrium with parent radioactivity. The IAEA-RGK-1 reference material is produced using high purity (99.8%) potassium sulfate. The $\mu$Dose software contains a dedicated module that allows the user to easily obtain calibration parameters ($k$ in Eqs. 1 and 2). This is done by matching calibration measurements with known radioactivities from the built-in database.
Results and Discussion

Samples and sample preparation

To test the performance of \( \mu \)Dose, activities of five samples were assessed using two additional systems, namely, an high-purity germanium (HPGe) \( \gamma \) spectrometer and a conventional TSAC system.

Sample 1 was an artificial sample composed from IAEA-RGU-1, IAEA-RGTh-1, and IAEA-RGK-1 mixed in equal weight proportions allowing the calculation of its specific activities. Samples 2, 3 and 4 are loess sediments. Sample 5 is a brick from archaeological excavations.

For \( \mu \)Dose and TSAC, the samples were powdered using an agate mill to avoid overcounting of \( \alpha \) particles which may arise from inhomogeneous distribution of radioactive elements in natural samples. After milling grain size distribution was verified using a laser diffractometer Mastersizer 3000 manufactured by Malvern Instruments Ltd. For all samples the median particle size was less than 7 \( \mu \)m. In both \( \mu \)Dose and TSAC systems, we used ground up 1 g samples and 42 mm diameter scintillators. The measurement time was the same for the \( \mu \)Dose and TSAC systems. TSAC measurements were performed using an in-house built system with pulse amplitude-time analyzer.

Measurements

The system set-up and data were evaluated according to ref [2]. \( \mu \)Dose specific uranium, thorium and potassium radioactivities were obtained using equations [1-3].

The \( \gamma \) spectrometry measurements were performed using a low background, high resolution HPGe detector with a resolution at full width at half maximum of 1.8 keV and relative efficiency of 40% at the energy of 1332 keV. The same standards were used as above. The counting times are given in Table [1]. The average \( ^{238} \)U specific radioactivity was calculated from \( ^{234} \)Th, \( ^{234m} \)Pa, \( ^{214} \)Pb, \( ^{214} \)Bi and \( ^{210} \)Pb lines using a weighted mean. The average \( ^{232} \)Th
specific radioactivity was assessed from $^{228}$Ac, $^{212}$Pb, $^{212}$Po and $^{208}$Tl lines using a weighted mean.

The results are summarised in Table 1.

Table 1: Specific radioactivity measurements using $\mu$Dose, a HPGe and a traditional TSAC system. Given uncertainties correspond to $1\sigma$.

|       | Mean $^{238}$U radioactivity (Bq·kg$^{-1}$) | Mean $^{232}$Th radioactivity (Bq·kg$^{-1}$) | $^{40}$K assessment (Bq·kg$^{-1}$) | Measurement time (hr) |
|-------|------------------------------------------|-------------------------------------------|-----------------------------------|---------------------|
| $\mu$Dose | TSAC | HPGe | $\mu$Dose | TSAC | HPGe | $\mu$Dose | TSAC | HPGe | $\mu$Dose & TSAC | HPGe |
| 1*    | 1620 ± 40        | 2400 ± 150   | 1628 ± 32      | 1110 ± 60      | 1300 ± 150      | 1062 ± 37      | 4480 ± 160      | 4610 ± 110      | 44       | 6.5     |
| 2     | 26.3 ± 2.6       | 30.1 ± 4.4   | 26.08 ± 0.59   | 32 ± 4.0       | 31.8 ± 4.3      | 33.90 ± 1.10   | 576 ± 48        | 564 ± 19       | 74       | 25      |
| 3     | 30.9 ± 2.5       | 26.7 ± 4.2   | 26.16 ± 0.55   | 30.1 ± 3.3      | 39.3 ± 4.1      | 32.86 ± 1.05   | 588 ± 20        | 532 ± 18       | 98       | 27      |
| 4     | 38.1 ± 3.9       | 41.6 ± 5.2   | 27.71 ± 0.58   | 33.6 ± 4.5      | 41.1 ± 5.2      | 35.36 ± 1.13   | 618 ± 27        | 570 ± 19       | 66       | 24      |
| 5     | 23.5 ± 2.8       | 26.4 ± 2.4   | 17.80 ± 0.37   | 19.3 ± 2.8      | 24.0 ± 2.4      | 19.48 ± 0.89   | 308 ± 18        | 324 ± 11       | 87       | 29      |

* Sample created form mixing IAEA-RGU-1, IAEA-RGTh-1, and IAEA-RGK-1 in equal weight proportions. The activities calculated using reference values are 1673 Bq·kg$^{-1}$ of $^{238}$U, 1083 Bq·kg$^{-1}$ of $^{232}$Th and 4669 Bq·kg$^{-1}$ of $^{40}$K.

System performance

As seen in Table 1, there is a very good agreement between the values obtained using the $\mu$Dose and gamma spectrometry. The thorium and potassium specific activities agree within 2 standard deviations and uranium for samples 4 and 5 within 3 standard deviations. In the investigated samples there was no indication of possible lack of secular equilibrium in the U and Th decay chains. The results obtained using TSAC are characterised by larger measurement errors for the same counting times, and as mentioned earlier information on the potassium content is unavailable.

When the activities are used to estimate the annual dose in trapped charge dating applications it has to be borne in mind that the values and errors returned by $\mu$Dose are correlated. This fact is taken into account during the calculation of the annual dose and leads to lower errors than in case these values were independent. Such situation is encountered in high resolution gamma spectrometry. The calculations of annual dose will be discussed elsewhere.
Conclusion

The µDose system allows to detect α and β radiation with four different decay pairs arising in $^{238}$U decay chain ($^{214}$Bi/$^{214}$Po), $^{232}$Th decay chain ($^{220}$Rn/$^{216}$Po and $^{212}$Bi/$^{212}$Po) and $^{238}$U decay chain ($^{219}$Rn/$^{215}$Po). If the sample is close enough to secular equilibrium, the obtained α and β counts and four separate decay pairs allow to obtain the $^{238}$U, $^{235}$U and $^{232}$Th decay chains concentration in the given sample. The $^{40}$K activity is assessed from the excess of β counts over what is expected over what is predicted from $^{238}$U, $^{235}$U and $^{232}$Th measurements.

µDose software allows for a convenient system calibration which limits routine work require from the user. The activities are calculated according to various assumptions (details are provided in the section $^{238}$U, $^{238}$U, $^{232}$-Th and $^{40}$K assessment) and measurement reports are automatically created for convenient post processing. The µDose system can be equipped various photomultipliers to assess various sample masses from 0.4–4 g. The software is build with emphasis on EPR/OS/TL dating therefore it includes various modules for dose rate calculation.

Finally the µDose may also provide a new tool for measuring very low levels of $^{226}$Ra, $^{224}$Ra, $^{223}$Ra, $^{222}$Rn, $^{220}$Rn or $^{219}$Rn via decay pair counting. Those isotopes are very frequently investigated because of their importance in medicine, geology or health risk studies.\cite{39,41}

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