Increased dose rate precision in combined $\alpha$ and $\beta$ counting in the $\mu$Dose system - a probabilistic approach to data analysis

Konrad Tudyka$^a$, Andrzej Bluszcz$^a$, Grzegorz Poreba$^a$, Sebastian Miłosz$^a$, Grzegorz Adamiec$^a$, Aleksander Kolarczyk$^b$, Thomas Kolb$^c$, Johanna Lomax$^c$, Markus Fuchs$^c$

$^a$Silesian University of Technology, Institute of Physics - Centre for Science and Education, Division of Radioisotopes, ul. S. Konarskiego 22B, 44-100 Gliwice, Poland
$^b$miDose Solutions, ul. Wolności 234/4, 41-800 Zabrze, Poland
$^c$Justus-Liebig-University Giessen, Department of Geography, 35390 Giessen, Germany

Abstract

The $\mu$Dose system was developed to allow the measurement of environmental levels of natural radioactive isotopes. The system records $\alpha$ and $\beta$ particles along with four decay pairs arising from subsequent decays of $^{214}$Bi/$^{214}$Po, $^{220}$Rn/$^{216}$Po, $^{212}$Bi/$^{212}$Po and $^{219}$Rn/$^{215}$Po. Under the assumption of secular equilibrium this allows to assess the specific radioactivities of $^{238}$U, $^{235}$U, $^{232}$Th decay chains and $^{40}$K. This assessment provides results with uncertainties which are correlated and, thus, require the development of an error estimation methodology which considers this issue. Here we present two different approaches for uncertainty propagation based on Monte Carlo and Bayesian methods. Both approaches produce statistically indistinguishable results and allow significantly better dose rate precision than when the correlations are not accounted for. In the given example, the dose rate precision is improved by a factor of two.

Keywords: Monte Carlo, Bayesian, correlated uncertainties, luminescence dating, dose rate, $\alpha$ counting, $\beta$ counting

1. Introduction

In trapped charge dating, the age is determined from the equivalent of the total absorbed radiation dose and the radiation dose rate. The radiation dose rate is often measured through the detection of ionising radiation or derived from other methods that allow us to assess the specific activities of radioactive elements that contribute to the dose rate in the natural environment. In many cases the major dose rate contributors are $^{238}$U, $^{235}$U, $^{232}$Th decay chains and the $^{40}$K isotope.

The dose rate calculation for trapped charge dating requires a number of corrections, which have been implemented in several tools routinely used in this dating method (Grün 2009, Durcan et al. 2015). In addition, other programs were developed but not formally published, for example ADELE by Kulig (2005) or Dose4Win by Bluszcz (2001). Such programs offer a convenient way of obtaining the dose rate which can be directly used in trapped charge dating.

The $\mu$Dose system (Tudyka et al. 2018) contains an in-built module for obtaining corrected dose rates. In contrast to the above mentioned programs, this module handles correlated uncertainties that arise from the performed measurements of the $\alpha$ and $\beta$ count rates, as well as from four different decay pairs (ibid.). The dose rate calculation procedure is fully controlled by an intuitive graphical user interface making the process convenient and straightforward. In this current work we describe how correlated uncertainties are accounted for in the dose rate calculation algorithms which are implemented in the $\mu$Dose system. Such an approach can be developed for any system characterised by correlated uncertainties. The error propagation can be achieved through either a Monte Carlo method or a probability distribution propagation (based on the Bayes theorem). Here we develop and compare both approaches. Dose rate adjustment for alpha efficiency, grain size distribution, chemical etching, gamma scaling and water content, which are all used in trapped charge dating, are also considered.

Finally, we show how the inclusion of correlated uncertainties allows the precise determination of the dose rate when compared to uncorrelated uncertainties.

2. Dose rate calculation module

In trapped charge dating methods, the annual dose is typically determined under the infinite matrix assumption (e.g. Guérin et al. 2012) or with the use of Monte Carlo simulations of charge transport (e.g. Martin et al. 2015) when the concentrations of radioisotopes and geometry are given.

The dose rate calculation module is organized as shown in the block diagram given as Fig. 1. First, the activities (or concentrations) of $^{238}$U, $^{235}$U, $^{232}$Th decay chains and
their daughters, as well as $^{40}$K isotope are calculated as described in the following section. Next, dose rate distributions are obtained through the application of either a Monte Carlo or a Bayesian approach. Finally, factors like water content, grain size and other effects that are commonly used in trapped charge dating are accounted for. This is briefly described in section 2.4. The module returns the results in the form of reports giving the $\alpha$, $\beta$, $\gamma$ and the total dose rate distributions, with theirs respective means and uncertainties. These values can be directly used for trapped charge dating.

2.1. Decay chains and $^{40}$K activity assessment

$\mu$Dose is equipped with a pulse analyzer (Milosz et al. 2017) that allows detection of $\alpha$ and $\beta$ particles along with four types of decay pairs. A detailed description on how the specific activities of $^{238}$U, $^{235}$U, $^{232}$Th decay chains and $^{40}$K are assessed is given by Tudyka et al. (2018). Before presenting this novel approach, the mathematical procedure can be summarised in the following steps:

$$r_\alpha = k_{\alpha,Th-232}a_{Th-232} + k_{\alpha,U-238}a_{U-238} + k_{\alpha,U-235}a_{U-235}$$

$$= k_{\alpha,Th-232}a_{Th-232} + (k_{\alpha,U-238} + k_{\alpha,U-235}) \frac{\lambda_{U-235}}{137.88 a_{U-238}} a_{U-238},$$

$$r_\beta = k_{\beta,Th-232}a_{Th-232} + k_{\beta,U-238}a_{U-238} + k_{\beta,U-235}a_{U-235} + k_{\beta,K-40}a_{K-40},$$

$$= k_{\beta,Th-232}a_{Th-232} + (k_{\beta,U-238} + k_{\beta,U-235}) \frac{\lambda_{U-235}}{137.88 a_{U-238}} a_{U-238} + k_{\beta,K-40}a_{K-40},$$

$$r_{Bi-212}/Po-212 = k_{Bi-212}/Po-212 a_{Bi-212}/Po-212,$$

$$r_{Bi-214}/Po-214 = k_{Bi-214}/Po-214 a_{Bi-214}/Po-214,$$

$$r_{Rn-220}/Po-216 = k_{Rn-220}/Po-216 a_{Rn-220}/Po-216,$$

$$r_{Rn-219}/Po-215 = k_{Rn-219}/Po-215 a_{Rn-219}/Po-215 \frac{\lambda_{U-235}}{137.88 a_{U-238}} a_{U-238}.$$
Here \( r \) represents the net count rates of the detected events indicated in subscripts, \( k \) is the calibration coefficient for the given system, the decay pairs are indicated in subscripts, and \( a \) refers to specific activities of the isotopes indicated in subscripts. Eqs. (2) can be rewritten in matrix form:

\[
\begin{bmatrix}
\frac{r_\alpha}{r_\beta}
\end{bmatrix} = \begin{bmatrix}
\frac{1}{k_{238,U} - 238} + \frac{1}{k_{235,U} - 238} & \frac{\lambda_{235}}{\lambda_{238}} \\
\frac{1}{k_{238,U} - 238} + \frac{1}{k_{235,U} - 238} & \frac{\lambda_{235}}{\lambda_{238}} \\
0 & k_{232,U} - 232 \\
0 & k_{232,U} - 232 \\
\frac{1}{k_{238,U} - 238} & \frac{1}{k_{238,U} - 238} \\
\frac{1}{k_{238,U} - 238} & \frac{1}{k_{238,U} - 238} \\
\frac{1}{k_{40,K} - 40} & \frac{1}{k_{40,K} - 40} \\
\frac{1}{k_{40,K} - 40} & \frac{1}{k_{40,K} - 40}
\end{bmatrix}
\begin{bmatrix}
\frac{a_{238,U}}{a_{235,U}} \\
\frac{a_{235,U}}{a_{238,U}} \\
\frac{a_{232,U}}{a_{232,U}} \\
\frac{a_{40,K}}{a_{40,K}} \\
\frac{a_{238,U}}{a_{238,U}} \\
\frac{a_{238,U}}{a_{238,U}} \\
\frac{a_{40,K}}{a_{40,K}} \\
\frac{a_{40,K}}{a_{40,K}}
\end{bmatrix}
\]

(3)

or more concisely

\[ r = ka. \]  

(4)

2.2. Monte Carlo module

\( \mu \text{Dose} \) software solves \( a \) in Eq. (4) by means of weighted least squares method yielding an estimate of the activities, \( \mu \), and a corresponding covariance matrix \( \Sigma \). The matrices have the forms where \( \mu = [\mu_{238,U}, \mu_{232,Th}, \mu_{40,K}]^T \) and

\[
\Sigma = \begin{bmatrix}
\sigma_{238,U}^2 & \sigma_{238,U} \sigma_{232,Th} & \sigma_{238,U} \sigma_{40,K} \\
\sigma_{238,U} \sigma_{232,Th} & \sigma_{232,Th}^2 & \sigma_{232,Th} \sigma_{40,K} \\
\sigma_{238,U} \sigma_{40,K} & \sigma_{232,Th} \sigma_{40,K} & \sigma_{40,K}^2
\end{bmatrix}
\]

(5)

All non-diagonal entries in the \( \Sigma \) matrix are typically negative since \( 238,U \), \( 232,Th \) and \( 40,K \) are negatively correlated. This is used in the Monte Carlo module where \( \mu \) and \( \Sigma \) are used do draw \( N \) random values from the multivariate normal distribution \( \text{Duda et al.} [2001] \). This gives \( N \) matrices \( a_i = [a_{i,U} - 238, a_{i,Th} - 232, a_{i,K} - 40]^T \) which contain \( 238,U \), \( 232,Th \) and \( 40,K \) specific activities. Such randomly selected sets of \( 238,U \), \( 232,Th \) and \( 40,K \) values form statistically plausible solutions of Eq. (4) for given count rates, \( r \), and their uncertainties, \( u(r) \). Further processing of random \( a_i \) values is described in detail in the section [2.3]

2.3. Probabilistic module

As an alternative approach to the Monte Carlo module, we can use a probabilistic approach where, instead of generating a population of solutions, we generate a discrete probability density function for solutions of Eq. (4). To do this, we create a set of matrices

\[ a_i = [a_{i,U} - 238, a_{i,Th} - 232, a_{i,K} - 40]^T \]

of statistically plausible solutions, that are evenly spaced. Subsequently, using Eq. (4) we map matrices \( a_i \) to \( r_{i,map} \) by calculating \( r_{i,map} = ka_i \), the mapped \( r_{i,map} \) experimental data \( r \) and their uncertainties \( u(r) \) are then used to assign likelihoods \( P_{i,j} \) for each mapped \( a_i \). This is completed by determining:

\[ P_{i,j} = \prod_{j=1}^{6} f(r_{i,map,j} | r_j, u(r_j)). \]  

(6)

Here, \( f \) is the probability density of the normal distribution with a mean value \( r_j \), standard deviation \( u(r_j) \), \( j \) indicates rows of matrices \( r_{i,map} \), \( r \) and \( u(r) \), i.e. iteration over the \( \alpha, \beta, 212,Bi/212,Po, 214,Bi/214,Po 238,Rn/216,Po \) and \( 219,Rn/215, Po \) count rates respectively.

If \( 238,U \), \( 232,Th \) or \( 40,K \) specific activity values and uncertainties are known from a different measurement (e.g. HPGe) we can assign that prior distribution i.e. \( P_{i,j} \) values assuming a normal distribution. The dissimilarity of two datasets i.e. \( P_i, P_r \) is then measured by the Bhattacharyya distance \( \text{Bhattacharyya} [1943] \). If no \( U, Th \) or \( K \) contents are known then all prior probabilities \( P_{i,j} \) are set to the same value, e.g. 1. Mapped posterior distribution i.e. \( P_{o,i} \) for each \( a_i \) matrix can be calculated as:

\[ P_{o,i} \propto P_{i,j} P_{r,i}. \]  

(7)

Finally, \( P_{o,i} \) values mapped on \( a_i \) matrices are forming discrete probability density function (PDFs) that can be used to assess \( 238,U \), \( 232,Th \) or \( 40,K \) content.

2.4. Dose rate conversion and additional corrections

Dose rate conversion factors had been reported frequently in the literature (e.g. \( \text{Adamiec and Aitken} [1998] \), \( \text{Guérin et al.} [2011] \), \( \text{Liritzis et al.} [2013] \) \( \text{Cresswell et al.} [2018] \). In this module, the user can choose which dose rate conversion factors should be utilised in the calculations. As default, the newest values summarized by \( \text{Cresswell et al.} [2018] \) with their uncertainties are applied.

To obtain dose rates in the Monte Carlo module, all drawn \( a_i \) matrices from the multivariate normal distribution are converted to \( \alpha, \beta \) and \( \gamma \) dose rates. Next, the means with standard deviations of those values are calculated.

In the probabilistic module, data \( a_i \) matrices are converted to \( \alpha, \beta \) and \( \gamma \) dose rates. Next, values \( P_{s,i} \) are assigned to each \( a_i \) used as the weighted means and standard deviations can be determined.

In trapped charge dating, a number of different adjustments are applied to obtain the proper annual dose rate. These adjustments have been summarized and thoroughly discussed previously (e.g. \( \text{Durcan et al.} [2015] \)). Therefore, we will concentrate on a brief overview of the options currently available in the \( \mu \text{Dose} \)-system.
Water content correction is calculated according to the most recent and reliable reports (e.g. Kreutzer et al., 2018. 2014; Mauz et al. 2006; Rees-Jones, 1995; Schmidt et al., 2018). Here we assume a normal distribution for the uncertainty of the a-value.

Grain size correction can be performed for α radiation as given by Bell (1980), Brennan et al. (1991), and Fleming (1979). For the β radiation the user can use data provided by Fleming (1979), Mejdahl (1979), Brennan (2003) or Guérin et al. (2012) data. In addition, Guérin et al. (2012) also provides correction values for feldspar. In this correction, we assume an uniform distribution within the given range of grain sizes.

Chemical HF etching is frequently used to remove the α contribution in grains. This effect can be accounted for optionally by: a) Fleming (1979) for either etching time in minutes or depth in µm for α’s and β’s, b) Bell (1979) for α’s and β’s for etched depth in µm, c) Bell (1979) data for α’s and Brennan (2003) data for β’s for etched depth in µm.

Water content correction is calculated according to Aitken (1985a) or with further modifications (e.g. Aitken and Xie 1990). The user can set an estimated saturation content and fraction of saturation. Moreover, the user may choose the distribution from normal, uniform or various triangular distributions which will affect the propagation of uncertainty.

Gamma dose rate scaling for a given depth is performed as given by Aitken (1985a). This is completed to account for a lower gamma dose at shallow depths up to ca. 30 cm. In addition, the user can scale for a given density and its estimated uncertainty with an assumed normal distribution.

For user convenience, μDose can also calculate cosmic dose rates for given a latitude, longitude, altitude, depth and a given sediment overburden density as well as its uncertainty.

The uncertainties in both Monte Carlo and Probabilistic modules of the final α, β, γ and cosmic dose rates are calculated using a dedicated Monte Carlo simulation.

3. Results and discussion

To compare both modules we used IAEA-RGU-1, IAEA-RGTh-1, and IAEA-RGK-1 standards from the International Atomic Energy Agency (IAEA 1987). 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 are considered to be in secular equilibrium with the parent radioactivity. The IAEA-RGK-1 reference material is produced using high purity (99.8%) potassium sulfate. IAEA-RGU-1, IAEA-RGTh-1, and IAEA-RGK-1 were mixed in equal weight proportions. The activities calculated using reference values IAEA 1987 are 1673 Bq·kg⁻¹ of ²³⁸U, 1083 Bq·kg⁻¹ of ²³²Th and 4699 Bq·kg⁻¹ of ²³⁸K. This sample was next measured on two systems with correlated and uncorrelated uncertainties, namely, μDose and HPGe systems, respectively. The values are reported in Table 1. Note that in the μDose system the covariance matrix Σ is known.

The first variant includes the Monte Carlo calculations as described in the section 2.2 using μDose μ and Σ data (Table 1). This is illustrated on Fig. 2a. The second variant (Fig. 2b) calculates dose rates and uncertainties as described in the section 2.3. We use a set of (30×30×30) nodes where the color of each node indicates the probability. In the third variant (Fig. 2c) we are drawing ²³⁸U, ²³²Th and ⁴⁰K values from three independent normal distributions with standard deviations of σ₂₃⁸U, σ₂₃²Th and σ⁴⁰K. Fig. 2a shows 27,000 of these points. This variant therefore does not take into consideration the correlation of uncertainties. For these cases (Fig. 2a-c) we calculate the dose rates and uncertainties using the newest available dose rate conversion factors (Cresswell et al., 2018). The results are summarized in Table 2.

Note that in Monte Carlo (Fig. 2a) and Probabilistic module (Fig. 2b) the higher a₂₃⁸U specific activities are lower than the corresponding σ₂₃²Th–2₃₂ values and vice versa. Hence, the dose rates arising from the two decay chains will cancel each other out giving lower uncertainties (Table 2 rows 1-2). In the Monte Carlo module, which neglects correlation (Fig. 2b) no such pattern is observed and the uncertainties are significantly higher (Table 2 row 3). This illustrates the advantage of the proposed approaches in the dose rate calculation that yields better, and lower, error estimates. In this particular measurement, the results shown in row 1 and 2 of Table 2 indicate an improvement by a factor of two in the precision, when compared to the uncorrelated values (Table 2 row 3).

The same improvement in dose rate precision arises when we compare measurements performed on μDose and HPGe (Table 1). Here in Table 1 μDose has higher uncertainties when compared to HPGe. This does not indicate a worse dose rate precision. In fact after dose rate conversion with Monte Carlo (Table 2 row 1) and the probabilistic module (Table 2 row 2) we are obtaining higher dose rate precision with μDose when compared to HPGe system where uncertainties are uncorrelated (Table 2 row 4).

Finally, in the probabilistic module we can combine data from the μDose and a different systems like HPGe. Table 2 row 5 and (Fig. 3) shows the merged measurements from Table 2. This increases the precision of the specific radioactivities and the dose rate.

4. Conclusion

The built-in Monte Carlo and probabilistic modules for dose rate calculation allow α, β and γ dose rates to be ob-
Table 1: Specific radioactivity measurements using µDose and HPGe system from Tudyka et al. (2018)

| ID | Device          | $a_{U-238}$ (Bq·kg$^{-1}$) | $a_{Th-232}$ (Bq·kg$^{-1}$) | $a_{K-40}$ (Bq·kg$^{-1}$) |
|----|-----------------|-----------------------------|-----------------------------|-----------------------------|
| 1  | µDose           | 1620 ± 40                   | 1100 ± 60                   | 4480 ± 160                  |
| 2  | HPGe            | 1628 ± 32                   | 1062 ± 37                   | 4610 ± 110                  |
| 3  | known activity  | 1673                        | 1083                        | 4669                        |

Figure 2: Three $^{238}$U, $^{232}$Th and $^{40}$K compositions. Isolines are showing projected 2-D PDF contour-plots. a) A set of 27,000 $^{238}$U, $^{232}$Th and $^{40}$K compositions are drawn by Monte Carlo module from a multivariate normal distribution. Note that the higher $a_{U-238}$ specific activities correspond to lower $a_{Th-232}$ values and vice versa. b) A set of 30×30×30 nodes, where the colour of each node indicates probability of each $^{238}$U, $^{232}$Th and $^{40}$K composition. Note that the higher $a_{U-238}$ specific activities correspond to lower $a_{Th-232}$ values and vice versa. c) A set of 27,000 radioactivities drawn by Monte Carlo module that neglect correlations i.e. setting all off-diagonal elements of $\Sigma$ to zero.

Table 2: Dose rate precision comparison with three calculation procedures for the IAEA reference material mix RGTh-1, RGU-1 RGK-1 in a 1:1:1 ratio

| ID | Calculation module | Device | $\bar{D}_\alpha$ (Gy·ky$^{-1}$) | $u(D)\alpha$ $\bar{D}_\alpha$ (Gy·ky$^{-1}$) | $\bar{D}_\beta$ (Gy·ky$^{-1}$) | $u(D)\beta$ $\bar{D}_\beta$ (Gy·ky$^{-1}$) | $\bar{D}_\gamma$ (Gy·ky$^{-1}$) | $u(D)\gamma$ $\bar{D}_\gamma$ (Gy·ky$^{-1}$) | Distribution |
|----|-------------------|--------|-------------------------------|---------------------------------|-------------------------------|---------------------------------|-------------------------------|---------------------------------|-------------|
| 1  | Monte Carlo        | µDose  | 568.3±7.1                     | 1.2%                            | 38.36±0.55                    | 1.4%                            | 31.59±0.46                    | 1.5%                            | Fig. 2a     |
| 2  | Probabilistic      | µDose  | 568.1±7.2                     | 1.3%                            | 38.36±0.56                    | 1.5%                            | 31.56±0.47                    | 1.5%                            | Fig. 2b     |
| 3  | Monte Carlounc     | µDose  | 568 ± 15                      | 2.6%                            | 38.36±0.88                    | 2.3%                            | 31.59±0.84                    | 2.7%                            | Fig. 2c     |
| 4  | Monte Carlounc     | HPGe   | 561 ± 10                      | 1.8%                            | 38.45±0.66                    | 1.7%                            | 31.17±0.56                    | 1.8%                            | -           |
| 5  | Probabilistic      | HPGe   | 564.1±6.1                     | 1.1%                            | 38.53±0.49                    | 1.3%                            | 31.40±0.35                    | 1.1%                            | Fig. 3      |

unc 27,000 $a_{i,U-238}$, $a_{i,Th-232}$ and $a_{i,K-40}$ drawn from three independent normal distributions
tained with a higher precision when compared to simplified calculations (Table 2). Both implemented methods of dose rate calculation in the µDose software take into account correlations and are consistent. The Monte Carlo module is somewhat less computer intensive and therefore faster than the probabilistic module. The probabilistic approach on the other hand offers a convenient way of combining measurements from different systems.

In the presented example, the dose rate precision was improved by a factor of two when comparing correlated (Table 2 row 1-2) and uncorrelated (Table 2 row 3) uncertainties assessments. When comparing µDose to HPGe, higher uncertainties (Table 1) do not indicate worse dose rate precision. In fact, after dose rate conversion with Monte Carlo (Table 2 row 1) and the probabilistic module (Table 2 row 2) we are obtaining a higher dose rate precision with µDose when compared to the HPGe system, where uncertainties are uncorrelated (Table 2 row 4). This also means that to obtain the same dose rate uncertainty counting time can be significantly reduced.

The Bayesian module can further help to increase the precision if results of independent measurements are available (Table 2 row 5).

The inbuilt dose rate module allows for accounting of the α particle effectiveness with a value, relative grain sizes, chemical etching, water content, relative sample position in the investigated profile and cosmic dose rate. These corrections are needed to obtain reliable annual dose rates in trapped charge dating.

Finally, the µDose dose rate module is fully controlled by an intuitive graphical interface. Moreover, for the user convenience, the input data are pre-filled with the most recently obtained values. This greatly simplifies the dose rate calculation procedure.

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