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Abstract

Luminescence dating methods currently allow for the evaluation of the distribution of equivalent dose (D_e) values for individual sand-sized grains of quartz and feldspar from a given sample, but the environmental dose rate is still derived from the bulk sample. Additionally, single-grain optically stimulated luminescence (OSL) dating is performed on disaggregated samples, resulting in the loss of micro-stratigraphic context. To enhance the interpretation of D_e distributions, we aim to estimate the beta dose rate to sub-millimetre regions of intact samples using the Timepix pixelated semiconductor detector. The Timepix contains an array of 256 × 256 pixels, each 55 × 55 µm in size and with its own

preamplifier, discriminator and digital counter. The detector has a total sensitive area of 1.98 cm², and 65,536 independent channels. The output of each measurement is a matrix containing the position and pixel-by-pixel count rate (or deposited energy) of each particle that interacted in the sensitive volume of the detector. The main challenge in using the Timepix detector is low natural sample activity, and the goal of this work is to acquire data with minimal background contribution. With an experimental setup guided by Geant4 simulations, progress has been made to greatly reduce background noise using ad hoc shielding and post-acquisition particle analysis. We have established a Timepix measurement procedure applicable to resin-impregnated sediment samples, including sample preparation, measurement, and data processing and analysis. These steps have been tested on an artificial micro-stratified sample (composed of quartz and biotite grains held together by resin) to derive the corresponding spatially resolved beta dose rates.

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Deriving spatially resolved beta dose rates in sediment using the Timepix pixelated detector

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Keywords

Spatially resolved radioactivity, beta dose rate, OSL, Medipix, Timepix, Geant4 simulation

Highlights

- Timepix detector used to map beta dose rates in sediment
- Geant4 simulations used to design measurement procedure
- Method tested on an artificial micro-stratified sample
- We show that natural dose rates can be mapped at sub-millimetre scales

Abstract

Luminescence dating methods currently allow for the evaluation of the distribution of equivalent dose (D_e) values for individual sand-sized grains of quartz and feldspar from a given sample, but the environmental dose rate is still derived from the bulk sample. Additionally, single-grain optically stimulated luminescence (OSL) dating is performed on disaggregated samples, resulting in the loss of micro-stratigraphic context. To enhance the interpretation of D_e distributions, we aim to estimate the beta dose rate to sub-millimetre regions of intact samples using the Timepix pixelated semiconductor detector. The Timepix contains an array of 256 x 256 pixels, each 55 x 55 µm in size and with its own preamplifier, discriminator and digital counter. The detector has a total sensitive area of 1.98 cm², and 65,536 independent channels. The output of each measurement is a matrix containing the position and pixel-by-pixel count rate (or deposited energy) of each particle that interacted in the sensitive volume of the detector. The main challenge in using the Timepix detector is low natural sample activity, and the goal of this work is to acquire data with minimal background contribution. With an experimental setup guided by Geant4 simulations, progress has been made to greatly reduce background noise using ad hoc shielding and post-acquisition particle analysis. We have established a Timepix measurement procedure applicable to resin-impregnated sediment samples, including sample preparation, measurement, and data processing and analysis. These steps have been tested on an artificial micro-stratified sample (composed of quartz and biotite grains held together by resin) to derive the corresponding spatially resolved beta dose rates.

1. Introduction

Optically stimulated luminescence (OSL) dosimetry has been widely used over the past 30 years as a dating method for naturally deposited sediments and implemented successfully on individual sand-sized grains of quartz and feldspar (Roberts et al., 2015). In the single-aliquot regenerative-dose protocol (Murray and Roberts, 1998; Galbraith et al., 1999; Murray and Wintle, 2000), the equivalent dose (D_e) for a sample can be estimated from the distribution of single-grain D_e values. Single-grain dating enables the OSL behaviour of individual grains to be investigated and unsuitable grains rejected. It also permits an assessment of the extent to which grains were adequately bleached by sunlight before deposition and remained undisturbed thereafter.

The environmental dose rate, in contrast, is represented by an average value obtained from the bulk sample, usually after it has been homogenised in the laboratory. Beta particles can penetrate up to ~3 mm through most sediments and the coefficient of variation of beta dose rates in sediments can be large, owing to the spatially non-uniform distribution of radionuclides in the ²³⁸U, ²³⁵U, ²³²Th decay chains and of ⁴⁰K. At low dose rates, the impact of beta dose heterogeneity will likely be high, such as in the Murray Basin of South Australia where the beta dose rates contributed 55% of the total activity to the dune sands (Lomax et al., 2007). Substantial spatial variations in beta dose rate may also arise in deposits with heterogeneous compositions, such as at Diepkloof Rockshelter in South Africa where the beta component constituted 50–70% of the total dose rate (Jacobs and Roberts, 2015). The use of an average beta dose rate may, therefore, not be appropriate for all single-grain D_e distributions (Jacobs et al., 2008; Galbraith, 2015; Martin et al., 2015; Roberts et al., 2015). Additionally, the extraction of grains for D_e estimation results in the loss of micro-stratigraphic context, so there would be benefits in being able to make measurements of the spatial distribution of beta dose rates on intact samples.

Measuring beta dose rates on intact sediment samples is difficult, because the low levels of radioactivity in nature make signal-to-noise ratios problematic as sample size decreases. For this reason, estimates of dose rate distributions have necessarily come through modelling, most prominently using Monte Carlo transport codes (Nathan et al., 2003; Martin et al., 2014, 2015; Guérin et al., 2015). These models use a simplified geometry of packed grains (usually spheres or ellipsoids), each with a given composition, and the deposited energy is recorded in a selection of 'dosimeter' grains.

Experimental validation of such models has required the artificial raising of the dose rate, so that the dose received by mineral grains can be detected (Nathan et al., 2003; Cunningham et al., 2012). Nathan et al. (2003) prepared sand boxes with uranium ore as the radioactive mineral, and allowed many months for the dose to build in the dosimeter

grains; Cunningham et al. (2012) sped up the experimental process by using neutronactivated NaOH grains as the radioactive source. Unfortunately, these experimental enhancements are not possible for natural samples. A Monte Carlo model of the dose rate distribution is unlikely to capture the complexity of natural sediments, where mineral inhomogeneities and micro-stratification are common occurrences (e.g., Roberts et al., 2015: Fig. 5). Furthermore, since we are dealing with dose rates only slightly higher than background levels, it is essential to use a detector that has high sensitivity and is stable over long measurement periods (7–40 days).

Here we investigate the use of a measurement device, the Timepix, which has the potential to resolve this problem. The Timepix hybrid-pixel detector, developed by the Medipix2 collaboration, consists of a pixelated semiconductor detector sensor coupled to the Timepix readout chip (Llopart et al., 2007; Jakůbek, 2009). The sensor is made of 300 µm-thick high resistivity silicon, which consists of 256 x 256 pixels, each with dimensions 55 x 55 μ m. Sensor pixels are individually Flip Chip ® (Lau, 1996) bump-bonded to corresponding pixels in the Timepix readout chip that lies underneath. Each readout pixel has its own preamplifier, threshold discriminator and digital counter. The complete assembly has a total sensitive area of 1.98 cm² and 65,536 independent readout channels (Fig. 1). With sub-millimetre spatial resolution, the detector has the potential to register emissions from individual mineral grains, as well as the sediment matrix in which they are embedded. Additionally, the detector is able to discriminate between particle types (alpha, beta, gamma and cosmic radiation), based on the morphology and energy deposition of the resulting clusters (Bouchami et al., 2010). This means that sources of background radiation (mostly from the gamma- and cosmic-ray components) can be suppressed, and the distribution of beta-like hits can be separated for analysis.

In this paper, we develop a procedure for using the Timepix to estimate the beta dose rate distribution in natural sediments. The proposed method for achieving spatially resolved dose rates is tested using an artificial micro-stratified sample prepared from grains of quartz and biotite (a dark mica mineral). To quantify the beta dose rate from the emitter grains, a calibration is performed using a sample prepared using an international standard of known ⁴⁰K radioactivity concentration distributed uniformly within the sample.

- 2. Materials and Methods
- 2.1 Materials: the Timepix detector and simulation platform
- 2.1.1 Timepix measurement

The Timepix contains a matrix of semiconductor detectors – that is, pixels – and each pixel is able to measure the energy deposition in the "time-over-threshold" mode, where the charge collected in each pixel is recorded while the preamplifier output is over the threshold (Llopart et al., 2007); or each pixel can function simply as an event counter. Measurements are stored as a matrix containing information on the x-y location and energy deposition in each pixel (when an energy calibration is provided). The Timepix chip used in

this experiment is part of the Jablotron MX-10 unit, which is factory-calibrated and comes with calibration parameters that can be imported into the acquisition software (Pixelman: Turecek et al., 2011), so that particle energy can be registered. Prior to data acquisition, a threshold equalisation procedure (Llopart et al., 2007) is carried out to compensate for pixel-to-pixel variations, and dead pixels are masked to avoid over-response.

Particle types are recognised based on cluster morphology (Holy, 2008). Beta particles usually have a cluster of pixel hits that appears as a long, curly track, or as single, double, triple and quadruple hits (Fig. 2). The shape of the cluster depends on the charge, velocity and mass of the incoming particle, as well as its angle of incidence. Clusters produced by gamma rays are similar to those of beta particles, as they mainly deposit energy by producing electrons (Teyssier et al., 2011).

Due to the typically low natural activity of environmental samples, background radiation contributes significantly to the total measured dose. We used several techniques to minimise incident radioactivity outside of the sample of interest, including shielding and cluster analysis. In this study, the Timepix detector was embedded in a 3 cm-thick lead castle for measurement acquisition, which stops most of the background radiation from reaching the detector (Fig. 1). A post-measurement cluster analysis was performed, based on the Medipix Analysis Framework (MAFalda: Bouchami et al., 2011; Idaragga, 2012), to ensure that only beta particle tracks were counted in the dose rate evaluation. Hits registered above a specific threshold (which was determined in the equalisation process) were grouped into clusters. The clusters were then characterised based on cluster length in the x-y directions, energy deposition, and energy-weighted centres of mass. Beta particles were classified as 'light tracks' consisting of non-straight line morphology, as opposed to the straight tracks of minimum ionising particles (MIPs). Due to the low activity nature of the measured samples, we used a long frame length (30 s) and overlapping tracks were not observed.

Resinated samples were placed directly on the detector chip to maximise sample emissions reaching the silicon sensor, with a thin layer of mylar film placed between the detector and the sample to protect the sensor. Total measurement time was adjusted to minimise counting error, varying between 7 and 14 days in duration for the data obtained in this study. For all measurements, the length and width of the sample were each approximately 10 mm and the thickness was 6 mm.

2.1.2 Simulation study

Geant4 simulations (Geant4 version 10.1) were developed and used as an aid to both establish the measurement procedure and analyse the acquired data. Geant4 is a Monte Carlo radiation transport code (Agostinelli et al., 2003; Allison et al., 2016), which facilitated the design of the experimental setup geometry specific to our study. The Timepix detector was modelled as a silicon chip with a voxelised readout geometry containing the 256 x 256 Timepix pixels. *EmStandardPhysics_option3 Physics List* was adopted to model the electromagnetic interactions of particles in the experimental setup. The Geant4 Radioactive Decay Model was also included in the physics list. The energy deposition in

each pixel and its corresponding x-y coordinates on the voxel grid were scored for each incident particle.

One of the first steps in establishing the Timepix measurement procedure was to verify the optimal sample thickness to maximise particle emissions incident on the detector. Geant4 was used to achieve this goal. A 10 x 10 mm SiO₂ block containing a uniformly distributed 40 K source was set on top of the Timepix detector. Samples of 0.1, 1–10, and 15, 20 and 30 mm thicknesses were simulated, modifying only the z-dimension. Simulation results demonstrated a plateau in the recorded hits per pixel for sample thickness above 3 mm, similar to the maximum range of its primary beta particle, due to beta particle absorption inside the sample (Fig. 3).

2.2 Using the Timepix to derive spatially resolved dose rates

2.2.1 Edge correction

The particle hit rate was found to vary depending on pixel proximity to the edge of the sample. This is explained by the lack of 2π geometry of hits for pixels close to the sample edge, where particles incident on these pixels are not originating in equal amounts from all directions. We derived a correction method to adjust the pixel count rate for the diminishing detection rate with increased proximity to the sample edge. Geant4 simulations were used to extract a correction function, where the simulation output matrix of detected particle positions on the 256 x 256 pixel map was converted into a binary matrix, treating pixels in the area directly beneath the sample as 1 and pixels outside of this area as 0. Pixels were then grouped into clusters at a sub-millimetre spatial resolution, and the proximity of each cluster to the sample edge was defined by the sum of the binary matrix, weighted by a Gaussian kernel. Higher values indicated a closer proximity to the centre of the sample, and lower values indicated proximity to the sample edge. Fig. 4 shows the results of the simulation, with each point corresponding to one pixel and its hit rate as a function of proximity to the sample edge, represented by the distance parameter d. A polynomial fit to this data was used to derive a correction factor to adjust the count rate acquired by the Timepix as a function of pixel location beneath the sample area.

2.2.2 Dose rate calibration of the Timepix

For many natural sediments, ⁴⁰K accounts for the largest component of the total dose rate. We used the IAEA standard material (IAEA-RGK-1) composed of potassium sulfate with a known ⁴⁰K concentration to make three calibration samples of known activity. Magnesium oxide (MgO) material was used as the nonradioactive sample component and distributed uniformly in quantified proportions with the IAEA-RGK-1, where IAEA-RGK-1 material constituted 100%, 24%, and 8% of the sample, respectively. The corresponding dose rates were quantified using established conversion factors (Guérin et al., 2011), adjusting for the higher ⁴⁰K content of the IAEA standard compared to the concentrations in natural sediments, resulting in calibration sample dose rates of 39.6, 9.6, and 3.0 Gy/ka, respectively.

Resin impregnation of the calibration samples was necessary to simulate an intact sediment sample, and to permit sample placement directly on the sensitive Timepix detector window. Calibration samples were produced in plaster moulds with small openings to allow resin to penetrate the sample material via capillary action. A calibration function (Fig. 5) was derived to convert the Timepix-measured count rate into the corresponding beta dose rate:

 $D_r = 3E + 06 * C_{Timepix}$

where D_r is the beta dose rate (in Gy/ka) and $C_{Timepix}$ is the count rate measured in the Timepix detector (counts/s/pixel).

2.2.3 Application to a simulated micro-stratified sample

Once we had established a procedure for Timepix measurements, a custom-made microstratified sample was chosen as a simplified representation of a real sediment sample (Fig. 6b). Two geological minerals, biotite and quartz, were selected as the radioactive and nonradioactive components, respectively. Biotite was chosen as an easily obtainable mineral with a sufficiently high concentration of ⁴⁰K for detection; the decay of ⁴⁰K results in the emission of a beta particle with a maximum energy of 1.3 MeV and a 1.5 MeV gamma ray. Prior to data acquisition, an ²⁴¹Am alpha source was used to delineate the boundaries of the sample in order to establish its exact location on the Timepix pixel grid and to enable the edge adjustment to be performed subsequently. This was done following sample placement on the Timepix sensor, by holding the ²⁴¹Am source over the setup until sufficient counts were observed to precisely outline sample shape on an integral measurement frame displayed in Pixelman software. The sample was then measured on the Timepix continuously for 7 days.

Measurement output was subjected to cluster analysis, where clusters were differentiated based on their tracks of energy deposition (i.e., cluster shape). Only curly tracks corresponding to beta and gamma particles were considered for subsequent data analysis. The data generated by the cluster analysis were then taken through a series of steps before determining a spatially resolved dose rate distribution for the sample:

- Pixel binning: an average count was obtained for square regions of interest to ensure that counts were statistically significant, while retaining sub-millimetre spatial resolution.

- Pixels outside the area directly beneath the sample were excluded, using the sample delineation data generated beforehand with the ²⁴¹Am alpha source.

- Edge correction: hit rates were adjusted for pixels near the edge of the sample area (section 2.2.1).

- The calibration function was applied to derive the corresponding spatially resolved pixel dose rates (section 2.2.2).

3. Results

The Timepix measurement count rates are plotted as a 2D hit map in Fig. 6a, where the scale on the right shows the number of counts per pixel derived in the cluster analysis. The biotite and quartz components can be clearly distinguished based on their spatially resolved count rates at a 0.1 mm resolution, plotted as cluster x-y centroids. The count rates range from 0 to 60 counts/pixel (after 7 days of measurement).

Fig. 6c depicts the same data after making the edge adjustment of the count rate in the region of interest, and converting the counts/pixel to the corresponding dose rates at a 0.9 mm resolution. The 2D map displays only pixel values directly beneath the sample area. The calculated dose rates range between -0.16 and 3.56 Gy/ka, with the highest-dose areas corresponding to biotite and the lower-dose areas to quartz.

The range of beta dose rate values can be observed in Fig. 7, where the x-profile corresponds to the non-radioactive quartz component and the y-profile captures both quartz and biotite. The quartz values are distributed around a dose rate of 0.05 Gy/ka, varying between -0.16 and 0.32 Gy/ka, whereas the biotite regions have dose rates ranging from 0.82 to 3.57 Gy/ka.

4. Discussion

4.1 Assumptions and uncertainties

We have assumed that the IAEA-RGK-1 calibration sample used to derive the Timepix calibration function contains uniformly distributed 40 K, with resin occupying only the pore spaces. Furthermore, the dose rate conversion factors given by Guérin et al. (2011) are based on an infinite dose matrix, whereas the Timepix is only able to measure the surface distribution of sample radioactivity. Here we relate the surface count rate determined by the Timepix to the 4π geometry applicable to buried sediments.

Due to the Poisson statistics pertinent to the Timepix detector, it is not possible to completely avoid negative dose rate results when a background adjustment is applied to the measurement. In these cases it is possible to either increase measurement time or decrease the spatial resolution of the derived data. Dose rate values below 0 are extracted for statistically insignificant count regions, and therefore do not yield statistically meaningful dose rates. The error bars on Fig. 7 further demonstrate this by showing that the "real" 0 Gy/ka values for the inactive quartz component are within the statistical uncertainty of the negative values.

It is also assumed that the final dose rate distribution of the micro-stratified sample is due solely to particles arising from within the sample. This assumption should be generally valid, as we used physical shielding by a 3 cm-thick lead container, followed by cluster analysis of the acquired data to select only those tracks corresponding to the energy deposition of beta particles.

4.2 Detector uncertainty calculations

Timepix count rate uncertainty was estimated by assuming a Poisson distribution, and calculated as the square root of the total number of detected counts per unit area. The average relative error associated with converting the Timepix count rate to beta dose rate was calculated as 24.3% for the Timepix detector (assuming a 14-day measurement) for single pixel resolution (55 μ m). Addition of a relative uncertainty of 2.86% for the IAEA-RGK-1 standard gives a total relative uncertainty of 24.5% per pixel for the calibration function. The uncertainty decreases at lower spatial resolutions, falling to 8.1% and 5.7% at 0.5 mm and 1 mm resolution, respectively. Since Timepix uncertainty varies as a function of detected counts, it is also strongly dependent on the sample dose rate. For the stratified artificial sample, a spatial resolution of 1 mm² resulted in relative uncertainties of 19.8% and 30.3% for beta dose rates of 3.5 and 1 Gy/ka, respectively, assuming 14 days of measurement time.

4.3 Future application to sediment samples

The next phase of investigation is to apply this Timepix measurement and analysis procedure to real sedimentary samples. The measurement time will need to take account of the typically low radioactivity content of natural samples, as described in section 4.2 above. Additionally, to derive the dose distribution to quartz grains, the mineral composition of the sediment slice will need to be mapped to establish the position of the dosimeter grains in relation to the emitter grains.

Previous efforts to derive spatially resolved dose rates include the use of α -Al₂O₃:C grains (Kalchgruber et al., 2003), fission tracks (Wagner et al., 2005) and autoradiography (Rufer and Preusser, 2009). Evaluating our method against these previous investigations reveals a few strengths and some shortcomings. The α -Al₂O₃:C study proved successful at obtaining a beta dose rate distribution at the scale of individual sand-sized grains, but it required the sample to be disaggregated, thereby losing the original spatial context. Our proposed Timepix method allows for the measurement of intact samples, but has the uncertainty of converting the detected count rate into an estimate of the beta dose rate. The fission track method can evaluate the alpha and beta components within a sample, but necessitates the use of a nuclear reactor. Autoradiography procedures are analogous to Timepix methods in being able to visualise the radioactivity within a sample and convert the acquired data into an estimate of dose rate using a derived calibration. However, autoradiography cannot distinguish between particle types, so it requires a longer length of measurement time to overcome the higher background. Autoradiography calibration also demands knowledge of the radionuclide composition of the sample to estimate the dose rate. In contrast, Timepix directly outputs the particle rate incident on the detector, where individual particle tracks can be differentiated and the count rate can be converted to dose rate.

5. Conclusions

Spatially resolved beta dose rate distributions can be determined with the Timepix pixelated detector to assist with the interpretation of D_e values obtained using single-grain OSL dating methods.

We have described a procedure for deriving a dose rate distribution for an intact, custommade sediment sample using a calibration function to convert a Timepix-determined count rate into a 2D map of beta dose rate. By discriminating by particle type, the Timepix is able to identify the beta contribution to the dose rate with sub-millimetre spatial resolution from measurements collected over a period of 2–3 weeks. When applied to natural samples, this method has the potential to greatly improve the accuracy of single-grain ages for complex, micro-stratified sedimentary deposits.

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Fig. 1. (a) Experimental setup containing the Timepix detector embedded in a 3 cm-thick lead castle and connected to Pixelman readout software. (b) Sample positioned directly over the detector sensor, which is protected by a mylar film.



Fig. 2. Two-dimensional map of particle tracks detected by the Timepix sensor, representing (A) muon, (B) alpha and (C) electron tracks (George, 2015).



Fig. 3. Results of Geant4 simulation showing a plateau in the particles detected by the Timepix sensor for samples thicker than \sim 3 mm; this corresponds to the maximum range of beta particles emitted by 40 K decay. The inset figure shows the simulation geometry. The orange block represents the Timepix detector chip modelled as a slab of 0.3 mm-thick silicon that scores incoming emissions in each of its 256 x 256 pixels. The orange block represents the sample (modelled as a 10x10 mm SiO₂ block of various thicknesses) placed directly on top of the sensor with uniform 40 K gamma and beta particle emissions.



Fig. 4. Results of the Geant4 simulation of particle hit rate as a function of distance to sample edge on the Timepix sensor, expressed in arbitrary units (a.u.). A polynomial function fitted to these data was used to correct pixel hits for the decrease in detected counts as a function of pixel proximity to the sample edge: $f(d) = -2.3875d^3 + 4.1156d^2 - 1.0169d + 0.2888$, where *d* represents pixel distance from the sample edge. Pixel clusters were identified with sub-millimetre spatial resolution and their proximity to the sample edge was derived using a Gaussian fit.



Fig. 5 Calibration curve to convert Timepix counts (counts/pixel/second) into dose rate (Gy/ka) derived using the IAEA standard material composed of potassium sulfate with a known ⁴⁰K concentration (IAEA-RGK-1). Three samples were made consisting of various IAEA-RGK-1 concentrations: a pure sample (39.6 Gy/ka), 24% concentration of IAEA-RGK-1 (9.6 Gy/ka), and 8% concentration of IAEA-RGK-1 (3.0 Gy/ka). MgO material was used as the nonradioactive sample component to obtain lower sample activities. The corresponding dose rates were quantified using established conversion factors (Guérin et al., 2011), adjusting for the higher ⁴⁰K content of the IAEA standard compared to the concentrations in natural sediments.



Fig 6. a) Post-cluster analysis 2D hit map of the micro-stratified sample, showing spatially resolved Timepix count rates (counts/pixel) at a 0.1 mm resolution. Biotite layers range between 25 and 60 counts/pixel, while the non-radioactive quartz layers range from 0 to 25 counts/pixel. b) Custom-made micro-stratified sample composed of radioactive biotite (dark layers) and non-radioactive quartz. The x-y-z dimensions of the sample are 10 x 10 x 6 mm. c) Edge-corrected distribution of beta dose rates (Gy/ka) for the micro-stratified sample at a 0.9 mm resolution. This plot shows only the pixel clusters located directly beneath the sample.



Fig. 7. Two profiles of the beta dose rate across the micro-stratified sample. The blue line corresponds to the x-profile and the red line to the y-profile, as shown on the sample image in the centre of the plot. The x-profile values correspond to the non-radioactive quartz component and are distributed around a beta dose rate of 0.05 Gy/ka (range: -0.16 to 0.32 Gy/ka). The y-profile values peak at the radioactive biotite positions and decrease to the x-profile values for quartz (range: 0.09 to 3.57 Gy/ka). Dose rate uncertainty increases with higher spatial resolution. For the 0.9 mm regions, the uncertainty ranged between 16.7% (for a dose rate of 3.56 Gy/ka) to 32.8% (for a dose rate of 1.44 Gy/ka).