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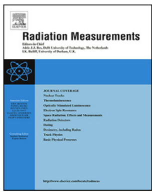

Overcoming crosstalk in luminescence images of mineral grains

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HIGHLIGHTS

- We outline the effects crosstalk in luminescence imaging.
- We design an analytical model for extracting the OSL signal.
- We show the model's benefit by recovering a mixed dose population.

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ABSTRACT

Luminescence imaging systems are becoming available for use in luminescence dating, and could potentially allow the dating of sediment and rock at a microscopic scale. For this to be achieved, analytical methods must be developed for turning the data-rich images into reproducible luminescence signals. At present, luminescence signals are collected from images after identifying Regions of Interest (ROIs) — a group of pixels mapped to a luminescent region or grain; the sum of the net ROI signal provides the measure of luminescence for each grain. However, the design of luminescence imaging systems requires a trade-off between signal focus and signal intensity. To maximise signal intensity, commercial systems use a lens combination which also induces optical aberrations, affecting the focus of the image. The variable focus of the image, combined with sample movement between measurements, means that the ROI signals may suffer from reproducibility problems and that signal crosstalk is a significant problem. Instead, the images should be paramaterised so that the inherent signal from each grain can be decontaminated from nuisance factors. We describe a data reduction method which uses a Bayesian hierarchical model to resolve the signal from each grain, with input from an incrementally expanding ROI. When tested with an artificial mixed population of grains, the method is better at recovering the known doses than the standard ROI approach, and has significant potential if combined with optimised measurement systems and pre-processing software.

1. Introduction

Luminescence dating of sediments has been a key chronometric technique with diverse applications in archaeology and palaeoenvironmental science. Significant interest has also focused upon accurate dating of consolidated samples, such as burned flints (Richter et al., 2014; Schmidt et al., 2015) or stone surfaces (Greilich et al., 2005; Sohbati et al., 2012). Luminescence dating broadly depends on the measurement of two quantities: the equivalent dose (D_e), representing the amount of radiation absorbed by the mineral grains during the burial period; and the dose rate (D_r). The burial age is then defined by Age (ka) = D_e (Gy) / D_r (Gy ka^{-1}). In almost all applications of thermally or Optically Stimulated Luminescence (OSL) dating, the measurement of these quantities follows the disaggregation of the sample. For sediment, mineral grains of interest (either quartz or feldspar) are first isolated through sieving and chemical treatments, yielding a homogeneous grain extract. Rock slices are usually crushed. Optical stimulation is provided either by a cluster of LEDs, in which case the grains are prepared in aliquots of tens to hundreds of grains; or by targeted laser on a grain-by-grain basis, with grains placed in a matrix of holes drilled into the sample carrier (Bøtter-Jensen et al., 2000). Estimates of D_r come from the measured concentration of radionuclides, always performed on a homogenised, bulk sample.

However, the use of bulk estimates for D_e and D_r obscures many complexities in the sediment matrix and radiation field.

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Complexities arise through a combination of dose-rate heterogeneity, poor bleaching and sediment mixing. Both sediment grains and crystals fixed within stone slices are affected by complex dose fields, due to inhomogeneous distribution of particular minerals (e.g. K-feldspars, or heavy minerals such as zircons), or particular micro-horizons (e.g. ash lenses, heavy mineral lags). Measurements that ignore spatial complexity may lead to erroneous estimates of the sample age (Martin et al., 2015).

One route through these difficulties would be to measure both the luminescence and beta dose rates on intact samples, i.e. rock slices or resin-impregnated sediment. With this approach, spatially resolved estimates of both D_e and D_r could be paired in the age equation, leading to age estimates at the sub-millimetre scale. In principle, this approach would make it easier to identify intrusive or poorly bleached grains, and increase the precision and accuracy of the final age estimate. Recent developments in instrumentation and software have made this goal feasible. Spatially resolved estimates of dose rate can be obtained through a combination of 3D scanning and radiation modelling (Martin et al., 2015), or more directly by placing a sediment slice on a pixelated semiconductor detector (Romanyukha et al., 2017). Similarly, technological advances allowing the production of commercially available luminescence imaging systems have only recently occurred. The first purpose-built devices, which used either film cameras (Hashimoto et al., 1986; Malik et al., 1973; Walton and Debenham, 1980) or microchannel plates (Burgraaf and Haskell, 1994) to provide spatial resolution, were superseded by the development of scientific cameras using charge coupled devices (CCDs). CCD-based imaging provided a significant technological breakthrough by allowing the relatively fast and simple collection and digitization of images, therefore much more complex data measurement and handling procedures could be developed (Howell, 2006). CCD-based cameras have been utilized by a number of luminescence laboratories, but signal intensity limitations have again limited the research primarily to luminescence emissions in the visible band (Baril, 2004; Greilich and Wagner, 2006; Spooner, 2000) albeit with a few exceptions (Duller et al., 1997; McCulloch et al., 2011). More recently, further developments in CCD architecture, such as the creation of electron multiplying registers (EM-CCD chips) to improve low intensity imaging, and the use of back-thinned CCDs and better phosphor coatings to improve imaging efficiency in the UV region, have yielded systems that can reliably detect low intensity luminescence emissions (Chauhan et al., 2014; Clark-Balzan and Schwenninger, 2012). These have now been incorporated into automated luminescence readers available from major manufacturers (Kook et al., 2015; Richter et al., 2013).

Quantitative extraction of D_es from images, however, has quickly found some significant issues (Greilich et al., 2015; Gribenski et al., 2015). For luminescence images to be used for routine D_e determination, the information content of the images must be reduced into luminescence signals. For a resin-impregnated sediment, we may wish these to correspond with the luminescence emissions from single grains (analogous to current single grain dating strategies). Rock slices may require a more nuanced approach in which surfaces of homogenous D_e are identified and pooled for calculation (see Greilich et al., 2005). In each case, the key first step involves appropriately assigning the luminescence emissions that arise from specific areas of the sample in order to construct the dose-response curves of the Single Aliquot Regenerative-dose (SAR) protocol (Murray and Wintle, 2000). The data reduction procedure, as currently implemented, has the following steps:

- Correcting image defects caused by cosmic and gamma ray interactions.
- Defining Regions of Interest (ROIs: a group of pixels for each grain) using a reflected light image taken after each luminescence readout.
- Making a correction for displacement of the sample carrier between images.
- Assigning a signal for each grain using the motion-corrected pixels in each ROI.

This ROI approach to data reduction has been implemented for grains scattered on a sample disc (Greilich et al., 2015), and for grains fixed in micro holes (Kook et al., 2015), but has severe limitations when considering intact samples. The signal collected in ROIs is not simply a function of the light emitted from that grain, but is also influenced by the image quality and the crosstalk from other grains. Commercial imaging systems use a set of lenses to focus the light from the sample towards the sensor, and imperfections in the lens system (spherical aberrations and astigmatism) reduce the quality of focus achieved at the detector position. Focussing can be improved by reducing the aperture, but with a cost in signal intensity. The imperfect focus of the luminescence image creates two practical issues with ROI data reduction:

1. *Variability of focus and sample motion*. The focus quality can vary across the image, with typically poorer focussing towards the edge. If the sample carrier is able to move between measurements, then the focus achieved for any one grain will also vary between measurements. An ROI, even if corrected for sample motion, will collect different light signals depending on the position of the grain.
2. *Crosstalk*. With imperfect focussing, the light signal collected from an ROI may contain a contribution from nearby grains. With variable focussing across the image, the degree of crosstalk is also dependent on grain position, which will change between measurements because of sample motion.

This presents a problem for ROI-based data reduction: even with successful image-segmentation and motion-correction algorithms, the inferred signal is sensitive to the grain position and to crosstalk, which must affect signal reproducibility. Crosstalk can be reduced (but not eliminated) by ensuring that grains are spaced apart-either by careful placement on sample discs (Gribenski et al., 2015), or using the single-grain sample discs designed for the Risø XY laser system (Thomsen et al., 2015). However, one of the potential benefits of imaging systems lies in the measurement of still-intact samples, for which grain spacing is not possible, and for which a means of overcoming crosstalk is requied.

This paper seeks a way of defining a grain's luminescence signal that is robust in the presence of variable focusing and independent of crosstalk. A Bayesian hierarchical model has been developed to describe the OSL signal, so that each grain's OSL emission can be disentangled from focus and crosstalk effects. The method is tested by trying to recover a known dose, with grains deliberately separated to simplify analysis. It is hoped the approach will permit future dose evaluation on intact samples.

2. Methods

2.1. Equipment, protocol, and image processing

Measurements were performed with a Freiberg Instruments Lexsyg Research (Richter et al., 2013), a luminescence reader with an automated detector changer and two filter wheels. Optical stimulation was provided by 458 nm (blue) LEDs, with power of 100 mW cm^{-2} at the sample position. Luminescence and reflected light images were detected with a Princeton Instruments

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