

# Optically stimulated luminescence dating using quartz

Andrew Murray, Lee Arnold, Jan-Pieter Buylaert, Guillaume Guérin, Jintang Qin, Ashok Singhvi, Rachel Smedley, Kristina Thomsen

## ▶ To cite this version:

Andrew Murray, Lee Arnold, Jan-Pieter Buylaert, Guillaume Guérin, Jintang Qin, et al.. Optically stimulated luminescence dating using quartz. Nature Reviews Methods Primers, 2021, 1 (1), pp.72. 10.1038/s43586-021-00068-5 . insu-03418831

# HAL Id: insu-03418831 https://insu.hal.science/insu-03418831

Submitted on 26 Oct 2022

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers. L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

# Optically Stimulated Luminescence Dating

2	Andrew Murray <sup>1†</sup> , Lee J. Arnold <sup>2</sup> , Jan-Pieter Buylaert <sup>3</sup> , Guillaume Guérin <sup>4,5</sup> , Jintang Qin <sup>6,7</sup> , Ashok K.
3	Singhvi <sup>8</sup> , Rachel Smedley <sup>9</sup> , Kristina J. Thomsen <sup>10</sup>
4	<sup>1</sup> Department of Geoscience, Aarhus University, Høegh-Guldbergs Gade 2, DK-8000 Aarhus C,
5	Denmark
6	<sup>2</sup> School of Physical Sciences, Environment Institute, Institute for Photonics and Advanced Sensing
7	(IPAS), University of Adelaide, North Terrace Campus, Adelaide, SA 5005, Australia
8 9	<sup>3</sup> Department of Physics, Technical University of Denmark, DTU Risø Campus, DK-4000, Roskilde, Denmark
10	<sup>4</sup> IRAMAT-CRP2A JIMR 5060 CNRS - Université Bordeaux Montaigne Maison de l'archéologie
10	Esplanade des Antilles, 33607 Pessac cedex, France, and Univ Rennes, CNRS, Géosciences Rennes,
12	UMR 6118, 35000 Rennes, France
13	<sup>5</sup> Univ Rennes, CNRS, Géosciences Rennes, UMR 6118, 35000 Rennes, France
14	<sup>6</sup> State Key Laboratory of Earthquake Dynamics, Institute of Geology, China Earthquake
15	Administration, Beijing 100029, China
16	<sup>7</sup> Xinjiang Pamir Intracontinental Subduction National Field Observation and Research Station,
17	Beijing 100029, China
18	<sup>8</sup> AMOPH Division, Physical Research Laboratory, Navarangpura, Ahmedabad 380009, India
19	<sup>9</sup> Department of Geography and Planning, University of Liverpool, L69 7ZT, UK
20	<sup>10</sup> Department of Physics, Technical University of Denmark, DTU Risø Campus, DK-4000, Roskilde,
21	Denmark
22	<sup>†</sup> Corresponding email: anmu@dtu.dk
23	Author contributions

- Introduction (A.M., L.J.A., J-P.B., G.G., J.Q., A.K.S., R.S., K.T.); Experimentation (A.M.,
- 25 L.J.A., J-P.B., G.G., J.Q., A.K.S., R.S., K.T.); Results (A.M., L.J.A., J-P.B., G.G., J.Q.,
- 26 A.K.S., R.S., K.T.); Applications (A.M., L.J.A., J-P.B., G.G., J.Q., A.K.S., R.S., K.T.);
- 27 Reproducibility & data deposition (A.M., L.J.A., J-P.B., G.G., J.Q., A.K.S., R.S., K.T.);

- Limitations and optimisations (A.M., L.J.A., J-P.B., G.G., J.Q., A.K.S., R.S., K.T.); Outlook
   (A.M., L.J.A., J-P.B., G.G., J.Q., A.K.S., R.S., K.T.); A.M. overlooked the Primer.
- 30

## 31 Acknowledgements

The authors thank three unknown referees for their useful suggestions, which certainly improved the 32 manuscript. Stacey-Lynn Pavia steered us through the writing and editing process, and we are very 33 grateful for her support and guidance. AKS acknowledges the Science and Engineering Research Board 34 (DST), India, for a YoS Chair Professorship. Jan-Pieter Buylaert received funding from the European 35 Research Council (ERC) under the European Union's Horizon 2020 research and innovation 36 programme (ERC-2014-StG 639904 – RELOS). Guillaume Guérin received funding from the European 37 Research Council (ERC) under the European Union's Horizon 2020 research and innovation 38 programme (ERC Grant agreement No. 851793 - QuinaWorld). Jintang Qin is funded by National 39 Natural Science Foundation of China (NSFC41671008). 40

41

#### 42 **Competing Interests**

- 43 K.T. and J-P. B. declare a competing interest. They are both employees of the Technical University of
- <sup>44</sup> Denmark, a commercial producer of luminescence dating instrumentation. The remaining authors
- 45 declare no competing interest

46

#### 47 Abstract

Optically Stimulated Luminescence (OSL) signals from quartz can be used to determine the timing of 48 deposition of sedimentary archives. OSL dating uses the accumulation of energy stored in a crystal 49 structure to measure time. This stored energy is absorbed from ionising radiation, and is released (reset) 50 by heat or daylight. The total energy (dose) absorbed since last resetting is measured using OSL, and 51 divided by the rate of storage (dose rate) to give the time elapsed from the last heating or daylight 52 exposure. In this Primer, we introduce the quartz OSL method and outline the signal resetting processes. 53 We describe the origins and quantification of the dose rate and describe the daylight-sensitive OSL 54 signal most appropriate to dose estimation. The most widely used dose measurement method is then 55 introduced, together with quality-control procedures. A broad set of geological and archaeological 56 studies are used to illustrate the wide range of potential applications, and we describe the challenges 57 arising from different deposition environments and summarize evidence for the precision and accuracy 58 of published ages. Uncertainties and minimum reporting are discussed together with methodological 59 limitations, particularly when applied to young and old sediments. Finally, we highlight the anticipated 60 future developments in the field. 61

62

#### 63 [H1] Introduction

The surface of our planet preserves extensive sediment archives recording the history of our species and its environment, but interpreting these archives depends on knowing when the sediments were deposited. Luminescence dating is one of the main methods used to estimate the depositional age of sediments over the last ~0.5 million years. This Primer is targeted at users of luminescence ages who seek greater understanding of how ages are derived and how to interpret their reliability, and at students and academics intending to implement the method in their work.

There are many methods for determining the deposition age of sediments and associated deposits<sup>1</sup>, but 70 radiocarbon, cosmogenic radionuclide and luminescence dating are the most widely applicable to the 71 recent geological past. Other methods, such as U-series and Ar-Ar, are site and sample specific and can 72 only occasionally be employed. Radiocarbon dating was developed first, and it still dominates studies 73 in the Neolithic and more recent archaeology. In other time periods (especially beyond ~45 ka) and in 74 many geological applications, luminescence dating has proved to be the most useful. This method dates 75 the last time quartz (and feldspar) was heated or exposed to daylight. It can be applied to nearly all 76 sediments on the Earth's surface, and used to date material over a wide age range, from almost the last 77 decade to ~500 ka ago. 78

Luminescence is light resulting from the release of energy stored in a crystal structure. This signal can 79 be used for dating because the amount of stored energy increases with continuing exposure to natural 80 ionising radiation (and therefore increases with time), and because luminescence can be reset by heat 81 or daylight. As a result of its known sensitivity to heat, luminescence was first applied to the dating of 82 pottery and other ceramics, but its full potential only became apparent when it was realised that the 83 signal was also reset by daylight exposure<sup>2</sup>. Initial studies employed thermoluminescence (TL), but 84 optically stimulated luminescence (OSL) rapidly took over following the first measurements on quartz 85 in 1985<sup>3</sup>. OSL can be more readily reset by daylight exposure; TL bleaches much more slowly, and is 86 never completely reset. Daylight exposure of sediment grains is ubiquitous during sediment erosion, 87 transport and deposition, and so OSL has the potential to date almost any surficial deposit. This direct 88 dating of the deposition of mineral grains also removes any ambiguity of association, such as occurs 89 with the radiocarbon dating of sediments using organic matter. 90

In nature, luminescence from quartz (and feldspar) arises from the dose absorbed from ionising radiation through time. This radiation is emitted (**Fig. 1a**) primarily during the radioactive decay of <sup>40</sup>K, and the <sup>238</sup>U and <sup>232</sup>Th decay series; it creates free electrons in the quartz crystal lattice, some of which are then trapped for long periods in defects in the lattice structure (**Fig. 1b**). Stimulation with light or heat (**Fig. 1c**) releases these trapped electrons, some of which give up their stored energy as photons; this is luminescence **Fig. 2** illustrates how this luminescence depends on prior bleaching (here, natural 97 exposure to daylight), and on the dose rate . After sample collection and preparation, the dose rate is 98 derived from analyses of radionuclide concentrations, and the dose from OSL. In the laboratory, the 99 natural luminescence signal is first measured by illuminating a sub-sample with light (**Fig. 2a**), and this 91 signal is calibrated in terms of dose. The age in years is then calculated from:

$$Age [a] = \frac{Dose [Gy]}{Dose rate [Gy.a^{-1}]} \qquad \dots \dots \dots (1)$$

This Primer focuses on OSL dating of sand-sized (63-300 µm) quartz grains, using blue (~470 nm) or 102 green (~530 nm) light stimulation and signal detection in the UV band (~340 nm). This choice is based 103 on the relative simplicity of this technique, the signal stability and the ubiquity of the mineral. Other 104 luminescence dating approaches use silt-sized quartz<sup>4, 5</sup>, silt- and sand-sized feldspar<sup>6</sup>, and/or OSL 105 signals stimulated with violet<sup>7</sup>, or infra-red<sup>8</sup> light. We outline the principles and the experimental 106 procedures most commonly used in deriving a quartz luminescence age, summarise methodological 107 challenges, and indicate how these are overcome in practice. We then discuss reproducibility and 108 accuracy, limitations, and finally our expectations for future developments. 109

#### 110 [H1] Experimentation

101

Before a luminescence age is measured, various practical steps must be undertaken. First, the sample 111 must be collected without exposure to daylight – for example, from a wind-blown or fluvial sediment, 112 a deep-sea marine core, an archaeological pottery sherd or an anthropogenic cave deposit. In the 113 laboratory, the quartz grains are separated for measurement in darkroom conditions, because the signal 114 is sensitive to light. Any illumination used must be of a wavelength and intensity that minimises the 115 effect on the potential luminescence signal<sup>9</sup>. The measurement equipment includes a stimulation light 116 source, a source of ionising radiation, a heating system and a photon detector to count the photons 117 118 emitted by the luminescence process. All of these experimental issues are discussed below.

#### 119 [H2] Sampling

The quartz-containing material - usually sediment - must be collected while avoiding daylight exposure 120 because OSL dates the last exposure of sand grains to daylight. In the field, the surface of the sediment, 121 or of the sedimentary sequence of interest, is first cleaned by scraping back<sup>10</sup>, which helps avoid the 122 light-exposed first few mm/cm of material. This also provides a clear view of the stratigraphy, so that 123 locations can be avoided that may have been affected by post-depositional disturbances, such as ice 124 wedges, animal burrows, roots, etc. At least the first few cm must be removed; if possible, it is better to 125 126 cut a section back by >1 m to avoid air-exposed material. This makes it more likely that the oxidation/reduction conditions and moisture content of the sample are representative of the long-term 127 burial. These operations can be undertaken in full daylight, allowing photographs to be taken of the 128

sampling site and of samples before removal, thus providing a record of sampling locations and theirrelationship to the general stratigraphy.

There are then two approaches to collecting a sediment sample from a cleaned terrestrial deposit. The 131 easiest is to hammer tubes (3-5 cm diameter of metal or plastic, depending on the degree of 132 consolidation of the deposit) into the section. This method is only suitable when the unit to be sampled 133 is of sufficient thickness that the tube will not cross sedimentological boundaries. If possible, the tube 134 should be at least 15 cm from any boundary, so that the  $\gamma$  dose rate gradient across the tube is small. A 135 crushed paper/plastic plug can be placed in the tube to support the sediment during insertion, reducing 136 the risk of collapse and the mixing of exposed and unexposed material. The tube must be filled 137 completely, either with sample or by packing with plastic, to ensure that the material cannot move and 138 mix during transport. In the laboratory, the outer  $\sim$ 2-3 cm of each end of the tube that have potentially 139 been light-exposed are set aside for radionuclide concentration measurements (to estimate the dose rate) 140 and for other sedimentological analyses (for example grain size, mineralogy, geochemistry, ancient 141 DNA, tephrochronology, etc.). The inner part of the tube is used for OSL measurements to estimate the 142 dose (see equation 1), and possibly for water content measurements (Box 1). 143

If the unit is too thin to be sampled with a tube, the second approach is to operate in the dark – either 144 under an opaque covering such as a tarpaulin or at night (moonlight has very low intensity and will not 145 significantly affect the OSL during the short sampling exposure<sup>11</sup>). In both cases, a controlled light 146 source, such as a red LED or a headlamp emitting dim orange light, can be used. It is usual to collect 147 two parts for each sample: one for OSL analysis, used to estimate the dose, and the other for 148 radiochemical analysis - to calculate the dose rate. First, the previously cleaned surface is scraped back 149 again (~1 cm) as it is likely to have been exposed to daylight after the initial cleaning of the section; the 150 sediment removed during cleaning can be retained for radioelement concentrations measurements and 151 for any other complementary measurements. The second part of the sample, taken from further into the 152 section, must be protected from light exposure. Although a few hundred grams is usually sufficient for 153 later processing, the actual sample size depends on the concentration and the size distribution of quartz 154 grains. For instance, a carbonate-rich sandstone generally contains a smaller proportion of quartz grains 155 than a desert dune sand, and a marine/lake core usually yields fewer sand-sized grains... If a narrow-156 grain-size fraction is to be targeted for single-grain OSL measurements, the amount of sediment 157 required may be greater than usual to ensure sufficient grains of the desired grain-size range are 158 recovered. This is in contrast to multi-grain aliquots, which can use a different and broader size range, 159 chosen to suit the sample. 160

Marine and lacustrine deposits are often sampled by coring. Although it is desirable to use opaque core barrels, this is not always possible, and successful sampling has been undertaken on poorly-archived light-exposed core material<sup>12</sup>. Whenever possible, cores should be split under controlled lighting. However, undessicated material can often be successfully sampled, even if the core was initially

processed in full daylight, by scrapping off a few mm of the light-exposed surficial material (this should 165 be retained for the dose rate measurement)<sup>13, 14</sup>. It is important to recognise that the depth range that can 166 be treated as a single sample varies with core depth. For a given sedimentation rate, the depth interval 167 representing a period of time that is small compared to the age uncertainty increases with core depth. 168 For instance, consider an age uncertainty of 10%, and a sedimentation rate of 10 mm/ka. For sediment 169 that is 10 ka old (100 mm depth), 10 mm of sample depth is equivalent to the age uncertainty of 10%. 170 But at 100 ka (1000 mm), 10 mm of sample only represents 1% of the age; to achieve the same age 171 sampling resolution as at 10ka, a 100 ka old sample can be 100mm thick. This is important because 172 sample size is often a limiting factor in working with core material. For the same reason, vertical mixing 173 of the outer layer of the core by the core-barrel may be important in shallow material, but can often be 174 ignored in deeper, older material without impacting the age uncertainty. 175

176 If the target is ceramic material such as pottery, bricks, and tiles, or heated or unheated stones and 177 cobbles, all sampling can be undertaken in daylight, although direct sunlight should be avoided. This is 178 because, in the case of ceramics and heated stones, the potentially light-exposed surface of the sample 179 is discarded in the laboratory, while in the case of cobbles, light is attenuated so rapidly with depth that 180 a few minutes of daylight does not affect the average luminescence signal in the outer 1 mm<sup>15</sup>. 181 Nevertheless, some rocks (e.g quartzite) are more translucent than others, and more sensitive to light 182 exposure during sampling; it may be prudent to take extra care with such samples.

The presence of water reduces the dose rate and increases the calculated age. It is therefore important to estimate the average water content of the deposit over the burial period. During sampling one should consider the water content history of the site and collect samples on which to measure the present day water content. A subsample of the OSL sample from inside the tube or from the radioelement sample (see above) can be used for this, and for determining the saturated water contents. In order to measure a reliable present-day water content, sealable bags should be used and the ends of the tubes made airtight by sealing with plastic.

Finally, sampling is the occasion to collect information to measure or estimate the long-term average 190 overburden of a sample. This includes the nature of the overburden, such as a cave roof, sediment, etc., 191 and the density and the thickness of the material covering the sample. The elevation of the site above 192 sea level and geographical coordinates (latitude, longitude) should also be recorded. These field 193 observations are used collectively to calculate the cosmic ray dose rate<sup>16</sup>, which typically contributes 194  $\sim 10\%$  to the total for shallow (<3 m) burial). Furthermore, in cases where the sediment is heterogeneous 195 196 on a scale of 10-50 cm, for example because of the presence of rocks, sediment unconformity, boundary between sediment and air or sediment and bedrock, etc., it may be necessary to measure in situ  $\gamma$  dose 197 rates (see below). It is also often appropriate to collect two or more separate dose rate samples, one 198 from sediment to be dated and one from the adjacent rock/sediment causing heterogeneity, in order to 199

measure the radionuclide concentrations in both components and model the  $\gamma$  dose rate (see Appendix H in<sup>17</sup>).

#### [H2] Sample preparation

In the laboratory, the part of the sample intended for dose rate measurements is homogenised by 203 grinding, so that even the small samples required for radiochemical analyses (typically a few 100 mg) 204 are representative of the whole. Supplementary Table 1 lists typical methods for dose rate 205 determination, and the typical sample size required. Given the usually considerable resource investment 206 207 in sample collection, it is good practice to keep back a portion of the non-light-exposed material as a substitute in case something goes wrong (mis-labelling, light exposure etc.) during sample preparation. 208 The remaining part of the sample intended for OSL measurements undergoes several treatment steps to 209 extract quartz grains (see Fig. 3a). The unexposed inner part of ceramics and heated stones is gently 210 crushed and either it or the sediment is wet sieved to obtain a specific grain-size range. Grain size is 211 important not only because of measurement practicalities, but also because both laboratory and 212 environmental dose rates are, to some degree, grain-size dependent<sup>17</sup>. The selection of a grain-size range 213 is driven by the abundance of that size range in the samples, and the practicality of chemically etching 214 the outer surface of the grains to remove the  $\alpha$ -irradiated component. Typically, grains must not be 215 smaller than fine sand (>  $63 \,\mu$ m) or there is a risk that chemical etching completely dissolves the sample 216 Generally, some choice is made within the range of 63-300 µm (for example 90-125 µm, 150-180 µm 217 or 180-250 µm); the 180-250 µm fraction is the most suitable for measurements using single-grain discs 218 with 300 µm diameter holes. 219

In all the steps outlined below, the sample should remain at or close to room temperature to avoid the risk of thermal loss of the OSL signal, especially during the HF digestions steps. HF is a particularly dangerous substance that can be lethal if used incorrectly. Because of this, the operator should carefully familiarise themselves with all local safety procedures and regulations before using HF.

The selected size fraction is first treated with 10% HCl (aqueous, as are all solutions below) for at least 224 30 minutes, or until gas evolution ceases completely. This removes carbonates, which may impede the 225 reaction with HF. The sample is then treated with 30% H<sub>2</sub>O<sub>2</sub> for up to a few days to remove finely 226 disseminated organics. To more easily disaggregate grains in fine-grained, clayey-silty sediment or 227 carbonate-rich sandstones, the bulk sediment can be soaked in water and treated with HCl and  $H_2O_2$ 228 before wet-sieving. After treating with HCl and H<sub>2</sub>O<sub>2</sub> and sieving, a few gram(s) of sample should be 229 left - mainly a mixture of quartz, feldspar and sometimes heavy-mineral grains, all in the chosen grain-230 size range. If only quartz is desired, the sample may then be placed directly in concentrated HF (40%). 231 Usually ~1 hour is sufficient to remove feldspar and etch the surface of the quartz grains (see below). 232 If the feldspar grains are also required, then prior mineral separation is necessary, usually via density 233

separation. K-feldspars, for example, are commonly used in other approaches to luminescence dating<sup>6</sup>,

<sup>18</sup>. To improve recovery when density separating quartz from feldspar, surface coatings and attached 235 clay grains can be removed using dilute HF (10%). The density separation step normally uses an 236 aqueous heavy liquid such as lithium heteropolytungstate. This is much safer to use than the more 237 traditional organic liquids, and it is also much more convenient, because it can be readily 238 diluted/evaporated to any density between 1 and  $\sim$ 3 g.cm<sup>-3</sup>. A density of 2.70 g.cm<sup>-3</sup> allows any heavy 239 minerals to sink; 2.62 g.cm<sup>-3</sup> allows a quartz-rich fraction to sink; 2.58 g.cm<sup>-3</sup> allows a Na-rich feldspar 240 fraction to sink and leaves a K-rich feldspar fraction floating. Some laboratories use only a single 2.58 241 g.cm<sup>-3</sup> step and rely on the later HF step to remove any denser feldspars that remain in the quartz-rich 242 fraction. Any heavy minerals that persist after the HF treatment rarely, if ever, make a significant 243 contribution to the quartz luminescence signal. Magnetic separation is also used to separate quartz and 244 feldspar grains, instead of density separation<sup>19</sup>. 245

Finally, treating the quartz extract with concentrated HF (40%) for 1 hour removes any remaining 246 feldspar. This treatment also etches  $\sim 10 \,\mu m$  from the outer surface of the quartz grains, thus removing 247 most of the quartz layer exposed to  $\alpha$  particles from the surrounding sedimentary matrix during burial 248 (see Fig. 1). The internal  $\alpha$  activity of quartz is very small and this etching step significantly simplifies 249 the dosimetry calculations. Some laboratories then soak the sample in 10% HCl for several hours to 250 remove any fluorides that might have precipitated on the grain surfaces. Although there is no evidence 251 that such amorphous precipitates are a significant source of contamination in quartz OSL, some fluoride 252 residues can be of sufficient thickness to significantly attenuate the OSL signal. After a final wash, it 253 may then be desirable to re-sieve the samples to define the minimum grain size during  $\beta$ -irradiation and, 254 255 for single grain measurements, to minimise the risk of multiple grains in each hole in the sample holder. For the measurement of multi-grain aliquots, a few mg of quartz may be placed loose in cups, or adhered 256 as a monolayer to stainless steel discs using silicone oil. The quartz is then tested for signal purity using 257 infrared stimulation (see Box 3). If it fails this test, the HF etching stage can be repeated, although the 258 259 extra mass loss has a small additional effect on the average natural  $\beta$  dose to the grains.

#### [H2] Dose rate fundamentals

The dose rate term in equation 1 is derived from the flux of ionising radiation originating with both 261 natural radioactivity and cosmic rays. For sand-sized quartz grains, the dose rate is dominated by <sup>40</sup>K, 262 which emits  $\beta$  particles and  $\gamma$  rays<sup>20</sup>, and members of the <sup>238</sup>U and <sup>232</sup>Th decay series 263  $(\alpha,\beta,\gamma^{17};$  Supplementary Figures 2 and 3). The abundance of these radionuclides in sediments depends 264 on the source rock and on differential transport processes related to weathering, erosion, transport and 265 deposition. Some of these processes are physical, such as sorting by size and/or density, while others 266 separate nuclides as a result of their geochemical properties<sup>21</sup>. The dose rate contribution from cosmic 267 rays is usually small, and depends on latitude, altitude and burial depth<sup>16</sup> (Supplementary Figure 1). 268

Direct measurement of the dose rate is complex, particularly because the various radiations penetrate 269 matter differently; in sediments,  $\alpha$  particles travel distances of up to ~20 µm,  $\beta$  particles up to ~3 mm 270 and  $\gamma$  rays up to ~50 cm (Fig. 1a). Fortunately, in many cases the key dosimetric concept of an infinite 271 matrix can be used to circumvent this difficulty<sup>17, 22</sup>; in an infinite matrix all the energy emitted in the 272 medium must also be absorbed in the medium, provided that the medium is homogeneous over 273 dimensions significantly larger than the distance travelled by the relevant radiation. Thus, one only 274 needs to know the energy emission rate to know the dose rate. The infinite matrix has different 275 dimensions depending on the radiation: for  $\gamma$  rays it is a body of sediment >0.75 m in diameter, and for 276  $\beta$  particles it is only about 5 mm in diameter. 277

The rate of energy emission in an infinite matrix is calculated from the concentrations of all the relevant 278 radionuclides. The activity per unit mass [Bq.kg<sup>-1</sup>] of a certain nuclide can be estimated directly, by for 279 example alpha counting or  $\gamma$  spectrometry (see Supplementary Table 1), but very often only the mass 280 concentrations of K and the series parents U and Th are measured using, for example, inductively 281 coupled plasma mass spectrometry. In order to deduce the activity concentrations of other radionuclides 282 in the U and Th decay series, the condition known as secular equilibrium is often assumed. In some 283 cases this assumption is invalid, because members of the decay series have been separated by their 284 differential geochemistries<sup>21, 23</sup>, and a more detailed knowledge of the state of equilibrium is required. 285 The latter may or may not be time-dependent, depending on whether the system is open or closed to 286 radionuclide migration. 287

The lifetime-averaged water content of the sediment is also important (see Box 1). Water absorbs energy but since it does not usually contain significant radioactivity, it does not emit energy. The net effect is to reduce the dose rate.

#### [H3] In situ γ dose rate measurements

In cases where sediment is heterogeneous on the scale of the penetration distance of  $\gamma$  rays (~30-50 cm, 292 see Figure 1a) the infinite matrix  $\gamma$  dose rate may not be representative of the  $\gamma$  dose rate to which the 293 sample was exposed during burial. These cases include poorly sorted sediment, sediment including 294 pebble and cobbles, or sediment adjacent to a rock wall. As an alternative, the  $\gamma$  dose rate may be 295 measured in situ at the sample location using, for instance, luminescent aluminium oxide (Al<sub>2</sub>O<sub>3</sub>:C) 296 pellets. Aluminium oxide is very similar to quartz in terms of dose absorption characteristics, and 297 doping with carbon gives this material a very sensitive luminescence response to dose<sup>24</sup>. In practice, 298 pellets are left buried in an aluminium tube for 6-12 months, after which the absorbed dose is sufficient 299 to induce a measurable OSL signal<sup>25,26</sup>. The aluminium tube has a  $\sim$ 2-3 mm wall thickness to prevent 300 beta particles from reaching the pellets. This directly-measured dose rate is the sum of  $\gamma$  and cosmic 301 dose rates, as affected by escape of <sup>222</sup>Rn and the water content of the sediment over the period of 302 measurement. Escape of the noble gas <sup>222</sup>Rn can be important because the decay of this radionuclide 303

gives rise to about 60% of the beat dose rate from the <sup>238</sup>U series, and 98% of the gamma dose rate (see Table S2b). In contrast to dose rates derived from radionuclide concentrations, correcting in situ dose rates for past variations in <sup>222</sup>Rn escape or water content requires some independent knowledge of the effect on the calibration of the dosimeter of such variations, and this information is not usually available.

The other common way of determining in situ  $\gamma$  dose rates involves the use of a field  $\gamma$  spectrometer, 308 typically with a NaI(Tl) or LaBr probe. Calibration involves measurements in infinite media of known 309 radionuclide concentrations<sup>27</sup>. Data analysis may consist of estimating effective K, U and Th 310 concentrations using the 'windows technique' (see Appendix L in<sup>17</sup>), and then converting these 311 concentrations into effective infinite matrix dose rates. The concentration of K is based on the emission 312 rate of the  $\gamma$  ray peak at 1461 keV, of U on the 1764 keV peak and of Th on the 2615 keV<sup>28</sup>. 313 Alternatively, the proportionality between total  $\gamma$  dose rate and the count rate above a certain energy is 314 used in the 'threshold' technique<sup>29,30</sup>. The advantage of this approach lies in the inclusion of a greater 315 portion of the recorded spectrum to estimate the  $\gamma$  dose rate, thereby reducing the uncertainties for a 316 given counting time (typically several minutes), but this approach does not give any nuclide-specific 317 information. It is also difficult to correct such threshold data for past variations in <sup>222</sup>Rn escape or water 318 content without independent knowledge of the U, Th and K contributions to the total. 319

320

[H2] Measuring the Equivalent dose (D<sub>e</sub>)

The instrumentation for determining the dose absorbed during burial and its equivalent in terms of producing luminescence includes a luminescence detector, a stimulation light source, a source of ionising radiation and a heating system. **Fig. 3b** summarises the relationship of these four components with respect to the quartz grains to be measured.

The main challenge in designing such a system is instrument sensitivity. The precise measurement of 326 an absorbed dose depends both on the sensitivity of the sample - equivalent to the number of photons 327 emitted per unit mass and per unit absorbed dose [kg<sup>-1</sup>.Gy<sup>-1</sup>], and on the detection efficiency - the 328 fraction of the emitted photons that are actually detected by the instrument. The relationship of the 329 resulting signal to the instrument background noise must also be considered. Detection efficiency 330 depends on the light path, the quantum efficiency of the photon detector, and the wavelength and 331 intensity of the stimulation light source. The latter are important because the more rapidly the charge is 332 released or detrapped (Figure 1c), the stronger the initial luminescence signal compared to background 333 334 noise. For a given detection wavelength, the electron detrapping rate depends on the wavelengthdependent photoionization cross-section<sup>31</sup> and the power density of the stimulation light at the sample. 335

The luminescence detector consists of a photon detector and detection filter(s). To precisely measure weak luminescence signals, a highly sensitive light detector with a low background count rate, such as a photomultiplier, and good light collection are essential. The latter requires that the solid angle subtended by the detector surface at the sample be as large as possible – this is usually achieved by minimising the distance between sample and detector or by focussing through lenses. In OSL, the luminescence signal is emitted as a consequence of optical stimulation, which is  $\sim 10^{15}$  times more intense than the OSL signal itself<sup>32</sup>. Thus appropriate detection filters are necessary, both to separate the luminescence from the stimulation light and to define the spectral detection window.

344

#### 345 [H3] Photon detector

Quartz has a relatively broad OSL emission peaking at ~360 nm, with a full width half maximum ( 346 FWHM) of ~80 nm (see Supplementary Figure 6). Photomultiplier tubes (PMT) operating in photon 347 counting mode and sensitive in the blue/UV range are most commonly used for detecting quartz OSL, 348 because of their high sensitivity, large dynamic range and low dark count rate. A large signal-to-noise 349 ratio is particularly important when measuring insensitive samples and/or samples having absorbed 350 small radiation doses, such as young samples. The quantum efficiency of the PMT is typically up to 351  $\sim$ 30 % depending on the wavelength of the luminescence. However, PMTs do not provide information 352 on the spatial variation of the emitted luminescence. In applications where such spatial information is 353 of importance, such as rock surface luminescence dating, electron-multiplier charge-coupled devices 354 (EMCCD) can be used but they are generally less sensitive than PMTs (see examples in<sup>33-35</sup>). 355

356

#### 357 [H3] Detection filters

The use of detection filters is important to define the wavelength window of interest and to separate the 358 emitted luminescence from scattered stimulation light. In quartz OSL dating, the emission is usually 359 detected through a filter with a peak transmission at 340 nm and a FWHM of 80 nm (Supplementary 360 Figure 7). The main emission from quartz is centred on 365 nm and, in principle, a significant 361 improvement in light detection is available by choosing a longer wavelength detection filter closer to 362 the main emission of quartz. However, moving the detection window closer to the typical stimulation 363 wavelength of 470 nm also gives an increase in background signal resulting from breakthrough of the 364 stimulation light, and hence does not necessarily improve the signal-to-noise ratio<sup>36</sup>. 365

366

#### 367 [H3] Stimulation light source

The electron detrapping rate depends both on the intensity and the wavelength of the optical stimulation light<sup>37, 38</sup> (see Supplementary Figure 8); the more intense and shorter the wavelength, the stronger the OSL signal per unit time. For most applications, optical stimulation is at a constant light intensity (continuous wave mode), but modulating the intensity of the stimulation light, for example in linear modulated OSL<sup>39</sup>, can be used to visually separate the signals from the different traps involved in OSL production. In pulsed OSL [POSL], pulsing the stimulation light and only detecting in between short  $\sim 50 \,\mu s$  bursts of stimulation light can help to separate quartz and feldspar OSL signals instrumentally<sup>40</sup>.

<sup>375</sup> POSL is also used to gain insight into luminescence recombination processes<sup>41</sup>.

Optical stimulation of quartz can be achieved using any visible light source such as filtered halogen 376 lamp systems<sup>36</sup>, lasers<sup>34, 42, 43</sup>, light emitting diodes (LEDs)<sup>32</sup> and laser diodes<sup>34, 44</sup>. In most quartz multi-377 grain OSL dating applications, blue (450-470 nm) or green (~525 nm) LEDs are used, with power 378 densities at the sample position ranging between  $\sim 40$  and  $100 \text{ mW/cm}^2$ . These result in the decay of the 379 fast OSL component within a few seconds (see below). As LEDs have a broad emission at FWHM ~30 380 nm, it is often necessary to stimulate the luminescence through cut-off filters to ensure an effective 381 separation of stimulation light and detection windows<sup>45</sup> (see Supplementary Figure 7). Shorter 382 wavelength light sources such as 405 nm laser diodes<sup>7</sup> can be used to access deeper traps insensitive to 383 blue light. In single-grain applications, stimulation by a green 532 nm solid state laser providing a power 384 density of ~50 W/cm<sup>2</sup> is common<sup>46</sup>. 385

[H3] Source of ionising radiation

The measured natural OSL signal must be calibrated in terms of absorbed dose. As different sub-387 samples/aliquots of quartz can have very different luminescence sensitivities, it is necessary to calibrate 388 aliquots individually. This calibration is performed by comparing the natural OSL signal with that 389 arising from a known laboratory dose given to each aliquot (see below and Figure 4). For convenience, 390 a  $\beta$  source containing the artificial radionuclide <sup>90</sup>Sr with a half-life of 28.8 years and its short-lived 391 progeny <sup>90</sup>Y is usually used to deliver the laboratory dose {Aitken, 1985 #359} {Andersen, 2003 #434}. 392 This combination of emitters has a maximum  $\beta$  energy of 2.27 MeV and a mean energy of ~600 keV, 393 which makes it relatively easy to shield sufficiently for it to be safe to use. 394

The laboratory dose rate to a quartz sample depends on the activity of the source, the source/sample 395 distance, sample grain size, and the substrate material on which the grains sit. Because of this, the source 396 must be calibrated for each irradiation geometry, particularly for different substrates and grain-size 397 ranges. The calibration is usually based on a portion of quartz grains that has been given a known dose 398 using a calibrated  $\gamma$  source. An appropriate measurement protocol is used to determine the duration of 399  $\beta$  irradiation that provides a sensitivity-normalised luminescence intensity equal to that induced by the 400 reference  $\gamma$  dose<sup>47</sup> (see below). Some laboratories provide sensitive quartz that has already absorbed a 401 well-known  $\gamma$  dose<sup>47</sup>, which allows those without access to a calibrated  $\gamma$  source to calibrate their own 402  $\beta$  source as required. 403

404 [H3] Heating system

Finally, to obtain an accurate  $D_e$ , it is important to perform reproducible thermal pre-treatments and to control the temperature of the aliquot during optical stimulation. OSL dating systems usually heat up to 700 °C, controlled by user-defined linear heating rates typically ranging between 0.01 and 10 °C/s. This heating capacity allows for flexibility in application, particularly when measuring TL. In routine quartz

- 409 OSL measurements, temperatures above 280 °C are rarely used and heating rates in the range of 2 to
- 410 5 °C/s are typical.
- 411 [H3] Instrument reproducibility

Very little has been published on formally testing the stability and reproducibility of the various components of an OSL measurement system, but there has been some examination of the overall reproducibility of the measurement of an OSL signal. After allowing for systematic changes in sensitivity in one automated system, repeated cycles of dosing, preheating and OSL measurement using multi-grain aliquots gave a standard deviation of ~1% in the resulting OSL signals <sup>48</sup>.

## 417 [H1] Results

#### 418 [H2] Deriving the dose rate

Radionuclide concentrations vary considerably from one location to another<sup>49</sup> (Supplementary Figure 419 4 a-e) because of changes in grain size and mineralogy, with the result that sample-specific dose rates 420 are required. Because well-sorted sands tend to be lower in radioactivity than clays or silts, the effective 421 age range of the technique can often be extended by targeting sandy sediments - the lower the dose rate, 422 the greater the dating range. Radionuclide concentrations within quartz grains are very low<sup>50</sup>, and the 423 dose rate to such grains is usually derived almost entirely from their surrounding environment. Sand-424 sized quartz grains (usually  $63 - 300 \,\mu\text{m}$ ) are etched in concentrated HF during sample preparation to 425 remove the outer 10-20 µm layer<sup>17, 19</sup>, which reduces the external alpha dose rate contribution to 426 negligible proportions. The effective total dose rate to well-sorted sands is typically ~1-2 Gy.ka<sup>-1</sup> 427 (Supplementary Table 3), although this value can vary considerably<sup>49</sup>. In dry sediment, typically ~60% 428 of the dose rate is from  $\beta$  particles and ~30% is from  $\gamma$  rays; the remaining ~5-10% contribution is from 429 cosmic radiation. The contribution from  ${}^{40}$ K alone may be ~40-50% of the total (Supplementary Figure 430 3), although this is variable (Supplementary Figure 4e,f) because of the considerable range of relative 431 activity concentrations in natural sediments. 432

Depending on how the concentrations of radionuclides have been derived, they may be expressed as activity concentration A  $[Bq\cdot kg^{-1}]$  or mass concentration  $C_0[g\cdot kg^{-1}]$ . The first step in deriving the infinite matrix dose rate is to convert mass concentrations into activity concentrations. Then the atom concentration per unit mass of sediment N  $[kg^{-1}]$  for a particular nuclide is calculated using

- 437  $N = (C_0/m_a) \cdot N_A$  (2)
- where  $m_a$  is the atomic mass (for our purposes with units [g·mol<sup>-1</sup>]) and  $N_A$  is Avogadro's number [mol<sup>-439</sup>]. Then the activity concentration is given by
- 440  $A=N\cdot\lambda$  (3)

- where  $\lambda$  is the relevant decay constant [s<sup>-1</sup>]. Measured concentrations may be reported by external analysts in ppm [ $\mu$ g·g<sup>-1</sup>] for U and Th, and percentage [%, 10 mg·g<sup>-1</sup>] for K; these must first be converted to [g·kg<sup>-1</sup>]. In the case of K, the mass concentration of <sup>40</sup>K is then derived using the isotopic abundance
- (0.0117%) before using equations (2) and (3).
- Finally, for each nuclide and type of emission  $(\alpha, \beta, \gamma)$ , the infinite matrix dose rate is given by the
- 446 product  $A \cdot \overline{E}$ , where  $\overline{E}$  is the average energy emitted per disintegration usually given in MeV (1
- 447 MeV= $1.6 \times 10^{-13}$  J, see <u>The National Nuclear Data Center</u>). Once the infinite dose rate for each nuclide
- and each radiation type has been derived, they are summed over all radionuclides to give the infinite
- 449 matrix dose rates for each type of radiation, as in the example given in Supplementary Table 3.
- 450 Relevant conversion factors  $(Gy\cdot ka^{-1})/(Bq\cdot kg^{-1})$  are summarised in Supplementary Table 2, and are
- <sup>451</sup> regularly compiled and updated<sup>51</sup>.

The example spreadsheet (Supplementary Data) attached to this primer allows the reader to work 452 through the calculation of dose rates using the infinite matrix assumption starting from concentrations 453 of K, U and Th. Such calculations provide estimates of the total amount of energy emitted (and thus 454 absorbed) in the sediment per unit time, regardless of the heterogeneity in the radionuclide 455 concentrations, and thus in the radiation field around each grain. They thus give the arithmetic average 456 dose rate to the grains of interest. Such heterogeneity in dose rate is to be expected in almost all sandy 457 deposits (ref Mayya et al., 2006, RM, Guérin et al., 2015, QG). In these circumstances it can be shown 458 mathematically that an accurate age can only be obtained by using the average absorbed dose in 459 equation 1 (ref Guérin et al., 2017, QG). This problem of the most appropriate dose estimation model 460 to use in the age equation is considered further below. 461

When dose rates vary in time and/or space, the infinite matrix assumption is not  $met^{52}$ . A heterogeneous 462 medium results in a heterogeneous radiation field and thus leads to different doses absorbed by different 463 quartz grains. Small-scale heterogeneity on the µm-mm scale affects radiation travelling only short 464 distances, such as  $\alpha$  and  $\beta$  particles. For example, K-rich feldspar grains in a quartz-dominated sand 465 provide localized high concentrations of  ${}^{40}$ K and different  $\beta$  dose rates to different quartz grains 466 (Supplementary Figure 5b<sup>53, 54</sup>). Larger scale heterogeneity on the cm scale also affects  $\gamma$  dose rates; if 467 limestone blocks, low in  ${}^{40}$ K,  ${}^{238}$ U and  ${}^{232}$ Th, are embedded in a higher activity sediment, the  $\gamma$  dose 468 rates to sand grains within a few tens of cm of the blocks will be lower than those to grains distant from 469 the blocks (see Supplementary Figure 5a). The radiation field may also vary with time, for example as 470 radioactive material is liberated and exported by reducing groundwater or the hydrolysis of feldspar, or 471 as non-radioactive material is imported into the deposit with infiltrating rainwater or groundwater, 472 leading to, for instance, the precipitation of carbonates or silicates<sup>55</sup>. 473

#### 474 [H2] Deriving the equivalent dose D<sub>e</sub>

Estimation of the dose absorbed during burial (termed the natural-, burial- or palaeo-dose) lies at the 475 heart of any luminescence age, and reliable dose estimation depends on the OSL signal used. The 476 luminescence signal from quartz is complex, but with some exceptions<sup>56</sup> there is evidence that the part 477 of the signal known as the fast-component derives from a single type of trap, which appears to empty 478 according to first-order kinetics, if held at an appropriate temperature<sup>57</sup>. As a result, this fast component 479 is largely independent of the other components<sup>58</sup>, and because it follows first-order kinetics, the fast-480 component consistently shows a sample- and dose-independent exponential decay shape with 481 stimulation time (Fig.4c inset)<sup>58, 59</sup>. Importantly, only the luminescence characteristics of the fast-482 component (particularly stability and light sensitivity) are sufficiently well-known to be used in a 483 reliable estimation of natural dose. The fast-component signal is stable (over  $\sim 10^8$  years)<sup>60</sup> and is also 484 most readily reset by daylight, making it most likely to be sufficiently reset in nature. It can also reset 485 by heating<sup>60</sup>. Heating prior to burial increases the sensitivity of the fast-component to dose<sup>59, 61-63</sup>, 486 allowing the measurement of much smaller doses. In particular, ceramics only a few years/decades old 487 can often be dated, and this can also apply to naturally heated sediments such as a lava flow. Although 488 the fast-component OSL signal increases with laboratory dose often up to many hundreds of Gy, natural 489 doses tend to be underestimated above ~150-250 Gy. This effective dose limit is often well below the 490 largest dose that can be measured under laboratory conditions, and the reason for this discrepancy is 491 not well understood<sup>64-66</sup>. When considered together with the value of the dose rate, it is this upper limit 492 to the usable dose that defines the maximum age of the method. This maximum is typically ~150 ka, 493 although ages of >1 Ma have been reported in unusually low dose rate environments<sup>67-69</sup>. In unusually 494 high dose rate environments, the upper age limit will be correspondingly lower. The simplest way to 495 confirm the presence of the fast-component is to compare the initial rate of signal loss during stimulation 496 with that of a standard sample known to be fast-component dominated (Fig. 4c inset using quartz 497 from<sup>47</sup>) or to use the ratio of the initial intensity during stimulation to the intensity remaining later in 498 the stimulation period<sup>70, 71</sup>.

Experimentally, it is important to ensure that the fast-component dominates the numerical estimates of 500 OSL used in dose calculations to avoid contributions from slower components with different bleaching 501 and thermal stability characteristics, and to avoid possible contamination by feldspar OSL. Such signal separation can be achieved by component fitting<sup>37, 72</sup> using a sum of exponential decay terms. In 503 addition, separation can also be improved by stimulating with green light instead of blue because the 504 505 difference between optical cross-sections for release of charge from the fast and slower components is greater for longer wavelengths<sup>73</sup>. Nevertheless, slower components can also be represented by 506 independent decaying exponential curves, and there is an order of magnitude difference in decay rate 507 between the fast and slower components when stimulating with blue light<sup>59</sup>. For most multi-grain 508 applications where the stimulation power is the same from aliquot to aliquot, this difference in decay 509

rates allows the use of simple summation, called early background subtraction<sup>74, 75</sup>, to discriminate against the slower components. In early background subtraction, a signal proportional to the intensity of the fast-component is calculated by subtracting the luminescence observed in the period after the beginning of stimulation from the initial luminescence observed at the beginning of the stimulation period. The signal difference is effectively zero in the absence of any fast-component, because the medium and slower components do not decay significantly in the time between the two summation periods.

Natural quartz has very heterogeneous luminescence and only a few % of grains give a significant fast-517 component signal<sup>76</sup>. Thus there are considerable advantages in measuring aliquots containing hundreds 518 to thousands of grains. This approach is only recommended if it is unlikely that useful information is 519 contained in the distribution of doses recorded by different grains, such as that resulting from 520 521 incomplete resetting or post depositional mixing (this reservation does not apply to dose rate heterogeneity<sup>77</sup>). Luminescence is averaged over many grains when working with such multi-grain 522 aliquots, and the Central Limits theorem suggests that these average OSL signals will be normally 523 distributed. 524

#### [H3] Dose estimation using the Single Aliquot Regenerative (SAR) dose procedure

In regeneration methods, the dose absorbed by an aliquot of quartz grains is estimated by first measuring the natural luminescence resulting from the natural dose. Luminescence signals are then regenerated in the same aliquot by various known laboratory doses given after laboratory resetting. These regenerated signals are then used to construct a curve of luminescence response to dose. Finally, the natural luminescence signal is interpolated onto the laboratory dose response to obtain an estimate of the natural dose. However, sensitivity changes from measurement to measurement have been observed for any regeneration of the signal in the laboratory , rendering interpolation inaccurate. Although such changes can arise from several sources, all are expressed as changes in the sample response to dose.

The Single Aliquot Regenerative (SAR) dose procedure <sup>78</sup> (see **Box 2** and **Fig. 4**) successfully 534 overcomes this problem by using the luminescence response to a fixed test dose to correct for sensitivity 535 changes. SAR is now the most widely used dose estimation method in retrospective dosimetry including 536 dating. It is particularly useful because all the measurements needed for a single dose estimate are made 537 on one aliquot, which can be as small as an individual grain. SAR is thus one of only two procedures 538 that have been used to determine distributions of dose in quartz grains within a sample (the other 539 procedure being the Single Aliquot Added Dose - SAAD  $-^{79}$ ). This sensitivity-change correction, and 540 the assumptions implicit in its use, are discussed using the simple model given in **Box 2**. The most 541 important outcome of this model is that the sensitivity-corrected luminescence signal is independent of 542 the measurement cycle, which is confirmed experimentally (Fig. 5a, data obtained using the 543 measurement sequence of Fig. 5b)<sup>78</sup>. 544

The model in **Box 2** suggests that the luminescence response is unlikely to follow simple exponential saturation. Nevertheless, it is common practice for interpolation to fit the response of  $L_i/T_i$  to dose with an expression of the form:

548 
$$\frac{L_i}{T_i} = \frac{L_s}{T_s} \cdot (1 - e^{-\frac{D}{D_c}}) \dots (6)$$

where the constant  $\frac{L_s}{T_s}$  is the sensitivity-corrected light level at saturation, and the constant D<sub>c</sub> is commonly described as the characteristic saturation dose. D<sub>c</sub>may be referred to as D<sub>0</sub> in the literature, but here we reserve this for the natural dose in a SAR sequence (see **Box 2**). D<sub>c</sub> is only truly constant (and equal to 1/b in b) if  $\chi(D)$  in **Box 2** is constant, which is unusual. Fortunately, interpolation of the natural signal onto a curve built from the regenerated signals ensures that the resulting dose is almost independent of models of luminescence response to dose, in marked contrast to a result obtained by extrapolation.

The signals resulting from the use of the typical SAR measurement sequence (**Fig. 5b**) with a single multi-grain aliquot of quartz extracted from a heated brick are shown in **Fig. 4a-c**, and with quartz from an unheated sediment in Figure 4d-f. The values for  $L_i$  and  $T_i$  were derived by summing the signals observed in the first 0.32 s (~25 mJ) of stimulation and subtracting the next 0.32 s as background. These measurements are discussed below.

In the first SAR cycle (i=0),  $D_0$  was absorbed during burial and the natural signal is measured. This 561 natural OSL signal (L<sub>0</sub>) from the brick quartz is shown as a dashed horizontal line in Fig. 4b, together 562 with the following test dose response ( $T_0$ ) in **Fig. 4a** (solid blue symbol plotted at 0 Gy). At the end of 563 the cycle, the aliquot is bleached to ensure it does not contain any residual signal (step 7 in Fig. 5b), 564 before returning to Step 1 for the second cycle (i=1,  $D_1=0.5$  Gy); and the second cycle signals ( $L_1$  and 565  $T_1$ ) are then measured. Repeating the measurement cycle continues with values of  $D_i$  increasing by 0.5 566 Gy steps until  $D_{11}=10.5$  Gy. In the following cycle,  $D_{12}=0$  Gy to give  $L_{12}$  (downwards open triangle in 567 Fig. 4b) and T<sub>12</sub> in Fig. 4a. The 2.5 Gy point is then repeated (L<sub>13</sub> - open square in Fig. 4b, T<sub>13</sub>), and 568 finally a repeat measurement ( $L_{14}$  – also open square – and  $T_{14}$ ) of the response to a 10.5 Gy regenerative 569 dose. Fig. 4c shows the resulting sensitivity-corrected dose response curve obtained by dividing each 570 value of  $L_i$  by the corresponding  $T_i$ . The intercept of the natural signal,  $L_0/T_0$ , on the smooth curve 571 passing through the regenerated L<sub>i</sub>/T<sub>i</sub> data is 3.8 Gy. Without the sensitivity correction, an incorrect 572 value of 1.8Gy would have been obtained (Fig. 4b). 573

The same process was repeated for the sedimentary quartz aliquot (**Fig. 4d-f**), but with different values

of D<sub>i</sub>, and additional repeated dose points. In this case, the corrected dose estimate is 25 Gy in contrast to a value of 14 Gy without sensitivity correction. It is important to note that the SAR procedure determines the laboratory  $\beta$  dose (as delivered by a relatively simple unscattered  $\beta$  spectrum) that produces the same luminescence response as the natural dose (delivered by complex heavily-scattered  $\alpha$ ,  $\beta$ ,  $\gamma$  and cosmic ray spectra). This matching laboratory dose may not be identical to the natural dose and is therefore termed the equivalent dose D<sub>e</sub> owing to the different luminescence efficiencies of different radiations, especially alpha, cosmic rays and low energy scattered electrons and photons <sup>17, 77, 80</sup>.

#### [H3] Testing the applicability of a SAR protocol

- <sup>584</sup> SAR is an empirical procedure supported by modelling<sup>81</sup>. Reliability can never be assumed a priori and,
- as far as possible, its applicability should be tested on each sample. In routine application, measurements
- of the recycling ratio and recuperation are usually built into any individual SAR dose measurement.
- 587 A recycling ratio is derived from a repeated measurement of at least one dose point (see open symbols
- in Figure 4,  $D \neq 0$  Gy), to test the accuracy of the sensitivity correction. The recycling ratio is the ratio
- of the second regenerated value of L/T to the first  $(L_2/T_2:L_1/T_1)$  and should be close to unity.
- Recuperation is measured by determining L/T for D = 0 Gy (see open symbols in Figure 4 for regeneration dose of 0 Gy). This measurement determines the carry-over of any residual signal from the previous measurement cycle, and it should be small compared to  $(L_n/T_n)$ . In some samples, recuperation may be significant (>5%), and a further optional step added<sup>82</sup> to the original SAR procedure (step 7 in Figure 5b) can reduce it. In very young samples, seemingly significant recuperation (>>5%) is often ignored without apparently compromising the accuracy of the ages<sup>83</sup>.
- Measuring the recycling ratio and recuperation tests the reproducibility of the sensitivity-corrected luminescence response to doses given in the laboratory. In addition, evaluating the importance of thermal pretreatment, thermal transfer and test dose size, and the dose recovery ratio all contribute to the evaluation of the applicability of a SAR protocol.
- Thermal pretreatment or preheating before luminescence measurement is required<sup>84</sup> for accurate 600 estimation of the D<sub>e</sub>. The thermal dependence of D<sub>e</sub> arises largely because of changes in the probability 601 of luminescent recombination<sup>85</sup>, and because of the presence of less-light-sensitive, thermally-shallow 602 unstable traps, which give TL peaks at 110, 180 and 220°C. Charges released from these traps may be 603 recaptured by the deeper fast-component trap and identified with a thermoluminescence peak at 325 604 °C<sup>57, 86</sup>. Alternatively, charge released during stimulation from the fast-component trap may be retained 605 by these shallow traps in a process known as optical transfer. Optical transfer processes are of different 606 significance in measurement following burial compared to measurement after laboratory dosing. For 607 instance, the shallow trap at 110 °C is unstable at environmental temperatures and so remains essentially 608 empty during burial, whereas deeper traps will retain some charge. In contrast, all traps fill during 609

irradiation on a laboratory timescale. Both optical transfer processes affect the intensity of the fast-610 component signal; after irradiation in the laboratory, the shallow trap at 110 °C must be emptied by 611 preheating or else the intensity of the regenerated fast-component signals will be affected and result in 612 a significant error in  $D_e^{87}$ . To this end, preheating is mandatory (step 2 in **Box 2**) because it helps isolate 613 the fast-component by emptying other less-light-sensitive traps that deplete more slowly but contribute 614 to the OSL signal. Preheating also speeds up the transfer of charge after laboratory irradiation, 615 simulating the effect of storage time: it both empties shallow traps and transfers some charge to the fast-616 component trap, as similarly happens during burial. Optical transfer from the 110 °C trap is also why 617 stimulation is usually carried out at a temperature sufficient to prevent the accumulation of charge in 618 the 110 °C trap. In practice, this stimulation temperature is usually 125 °C<sup>57</sup>. 619

However, thermal pretreatment also has undesirable effects: the S and/or  $\chi$  (see Equation 4 in **Box 2**) 620 appropriate to the natural dose may respond differently to preheating compared with the S and/or  $\chi$ 621 appropriate to a laboratory dose. In a process known as thermal transfer, preheating may also induce a 622 fast-component signal in an otherwise well-bleached modern sample through charge transfer from light-623 insensitive shallow traps into a previously empty OSL trap. One way of testing for the significance of 624 all the above-mentioned effects is to look for a plateau in  $D_e$  estimates following preheating to various 625 temperatures. This analysis enables determining the range over which the estimated dose is independent 626 of preheat temperature (Fig. 6a). The sample should also be preheated immediately before measurement 627 of  $T_i$  because the test-dose response can help track the sensitivity of the fast-component. This second 628 preheat step (Step 5, 'cut heat' in Fig. 5b) must be above 130 °C to empty the 110 °C trap, and is usually 629 at least 20 °C below the regeneration preheat temperature in practice. 630

Thermal transfer of charge to the fast-component trap from low-temperature light-insensitive traps can 631 make a well-bleached sample, with negligible charge in the fast-component trap at the time of burial, 632 appear to be incompletely bleached. These low-temperature traps are sufficiently stable in nature to 633 retain a significant trapped charge population, and because they are light-insensitive they are therefore 634 difficult to empty before burial. As a result, they act as charge reservoirs. Thermal transfer can move 635 sufficient charge from these reservoirs into the fast-component trap to result in an overestimation of D<sub>e</sub>. 636 In general, this phenomenon is unlikely to be of significance in older samples, but in young samples, 637 where there is only a small amount of trapped charge in the OSL trap from the most recent burial period, 638 it can be of considerable importance. The potential significance can best be examined by bleaching a 639 set of aliquots in the laboratory either by artificial or daylight stimulation, followed by a preheat plateau 640 test as described above. The apparent dose at low preheat temperatures such as 160°C should be 641 negligible (Fig. 6b). However, the apparent dose increases as the preheat temperature is increased and 642 charge is increasingly transferred to the OSL trap. The choice of maximum acceptable preheat 643 temperature is then made by comparison with the  $D_e^{88, 89}$ . 644

Any dependence of  $D_e$  on test dose size can be similarly examined to find a test dose range over which the  $D_e$  is constant. The OSL from a small test dose can be affected by thermal transfer, especially if the preceding regeneration dose was large, and too large a test dose may lead to dose underestimation<sup>78</sup>. In general, it seems that a test dose of ~30% of the natural dose is a good compromise. However, a value equal to or larger than the  $D_e$  may be necessary for young or insensitive samples to ensure a measurable test dose signal.

Although all the tests described above are important to a greater or lesser degree, the dose recovery 651 ratio<sup>90</sup> provides the single most complete test of SAR performance for any given sample. It can be used 652 to assess whether a specific SAR protocol can accurately determine a known dose administered when 653 the sample is in a condition as close as possible to that when it was initially reset before burial. To 654 achieve this, any natural trapped charge population in an aliquot is first removed by artificial or daylight 655 exposure without any thermal treatment before the aliquot is given a known laboratory dose. While the 656 given laboratory dose is typically similar to the De, young samples may necessitate a larger dose for the 657 precision required. The given dose is then measured using the chosen SAR protocol. The dose recovery 658 ratio is the ratio of the measured dose to the given dose and should have a value close to unity (Fig. 659 **6c,d**). The given dose must be administered before any pretreatment (especially before any preheating) 660 other than light exposure o distinguish the dose recovery ratio from the recycling ratio. 661

Although usually reported as a dose recovery ratio, it is important to realise that the measured quantity 662 is sensitivity-corrected luminescence (L/T), not dose. At low doses, in the more linear part of the dose 663 response curve, dose behaves similarly to sensitivity-corrected OSL (Fig. 7, black OSL distribution at 664 20% of saturation). In contrast, at high doses where the response curve becomes significantly sub-linear, 665 even small uncertainties in L/T are magnified by interpolation and dose uncertainties can become 666 unbounded. For a normal OSL distribution with a relative standard deviation (RSD) of 5% centred on 667 668 86% of saturation (2xD<sub>c</sub>, **Fig. 7**, blue data), almost all signals can be interpolated onto the growth curve. However, the resulting dose distribution is significantly asymmetrical and the RSD is ~20% of the mean 669 dose or 4 times the measurement uncertainty on L/T. In contrast, for an OSL signal with 5% RSD 670 centred on 95% of saturation (3xD<sub>c</sub>), ~15% of aliquots cannot be interpolated onto the dose response 671 curve and the RSD of the apparent dose distribution is ~30% of the mean (Fig. 7, green data). 672

Alternatively in a dose recovery measurement, if the regenerative dose  $D_1$  is chosen equal to the given dose  $D_0$  then the recovery ratio is more usefully expressed as the luminescence ratio  $(L_0/T_0)/(L_1/T_1)$ . This OSL ratio is a more sensitive measure for the reliability of a chosen protocol because the dispersions in the OSL distributions will always be  $\leq$  those in the dose distributions. The OSL ratio also avoids the incorrect rejection of values when measuring close to saturation. In true saturation, the average luminescence ratio should be unity, with 50% of observations (for a normal distribution) lying above unity<sup>91</sup> (**Fig. 7**, red data). However, luminescence ratios above unity cannot be interpolated to

give doses, and so must be ignored if a dose ratio is reported. This incorrectly biases dose estimates to 680 low values (Fig. 7), and produces an inaccurate dose distribution with pronounced asymmetry and a 681 standard deviation of ~30%. As a result, any dose recovery measurement close to saturation is biased 682 and accompanied by a risk of incorrectly accepting/rejecting a sample as suitable for dating. 683

The dose recovery ratio or luminescence recovery ratio is an average sample descriptor, which cannot 684 be applied to the individual aliquots used for dose measurement. The reason for this is that the given 685 dose must be administered before any preheating, but samples must be preheated before measurement 686 of the natural signal. The dose recovery ratio describes the suitability of the chosen SAR protocol to 687 measure a known laboratory dose given to a particular sample prior to any thermal treatment, but it does 688 not establish whether the chosen SAR protocol will measure a dose equivalent to the natural dose 689 accurately; this remains an assumption. To this end, Guérin et al.<sup>92</sup> were unable to establish a correlation 690 between dose recovery ratios and the ratio of OSL ages to independent ages. In the final analysis, the 691 accuracy of OSL ages can only be tested by dating material of known age. 692

#### [H3] Aliquot specific rejection criteria 693

694

Once a suitable SAR protocol has been identified, it is used to measure the De in many aliquots. Not all 695 measurements may be of equal value, and criteria based on recycling ratios, recuperation, absence of 696 effect of prior infra-red stimulation on OSL (known as OSL-IR depletion ratios, see **Box 3**<sup>93</sup>) have all 697 been suggested to identify less reliable values<sup>78</sup>. However, the use of any such criteria should be 698 justified. By normalising doses from each individual aliquot to the sample mean, the correlation 699 between recycling ratio, recuperation and OSL-IR depletion ratio, and De can be examined across a set 700 of samples, each with a different average  $D_e^{91, 94}$ . This correlation is done for 54 loss samples in **Fig.** 701 **8a,b**, and for these samples at least, there is no justification for aliquot-specific rejection based on either 702 poor recycling ratios or significant OSL-IR depletion ratios. Perhaps surprisingly, this is not unusual <sup>95-</sup> 703 <sup>99</sup>. To this end, data should only be considered for rejection if there is evidence to suppoer that doing so 704 improves the average dose estimate. However, identifying appropriate rejection thresholds to identify 705 and reject individual aliquots from a single sample may not always be possible. Experimentally, the 706 aliquot-to-aliquot over-dispersion in these measurables can be considerable, such that in comparison 707 the individual estimates of uncertainty on each aliquot are often small. 708

Quartz dose response curves vary in curvature even within a sample, and some aliquots saturate at 709

lower doses than others<sup>100-104</sup>; this is particularly true for individual grains. For older samples with 710

larger natural doses or when larger given doses are used in dose recovery tests, some aliquots cannot 711

be used to measure the dose of interest because  $D_c$  in Equation 6 is too small. In the worst case, said 712

aliquots that cannot be used will not give estimates of De. In practice, this leads to unbounded 713

estimates of De and/or uncertainty for a number of aliquots, and discarding these values leads to bias. 714

To avoid this biasing of the  $D_e$  distribution towards low dose estimates, aliquots can be sorted by their

ability to record large doses or by increasing  $D_c$  values (**Fig. 8c**), with aliquots only with increasingly

- <sup>717</sup> larger  $D_c$  values selected<sup>91, 94, 105-107</sup>. As  $D_c$  increases, the fraction of aliquots in saturation becomes
- negligible and the estimate of the central tendency of the equivalent dose or given dose distribution
- 719 becomes stable.

#### 720 [H1] Applications

This primer is not intended as a comprehensive review of the OSL application literature, as several such reviews are already available<sup>108-114</sup>. Here, we present illustrative examples of actual and potential applications. Many of these applications were pioneered using thermoluminescence and **IRSL** on both quartz and feldspar, and include the dating of pottery<sup>115</sup>, loess<sup>116</sup>, colluvium<sup>117, 118</sup>, archaeological sediments<sup>119</sup>, periglacial features<sup>120</sup>, fluvial, marine and aeolian systems<sup>121-123</sup>, direct dating of tectonic faults<sup>124</sup>, soils<sup>125</sup> and glacier ice<sup>126</sup>.

#### [H2] Quartz OSL in archaeo- and geochronology

- Quartz grains must give a measurable fast-component OSL signal in response to ionising radiation for 728 reliable dating. Unfortunately, not all sources of quartz have these characteristics. Igneous rocks are the 729 main source of quartz grains in sedimentary deposits, but grains extracted directly from primary rock 730 do not usually show significant luminescence sensitivity<sup>63, 127</sup>. Consequently, sediments in mountain 731 catchments dominated by primary rocks rarely give a measureable fast-component signal<sup>63</sup>. Sensitivity 732 seems to develop during repeated cycles of erosion, transport and deposition<sup>128</sup> and laboratory studies 733 support this inference<sup>63, 129, 130</sup>. Thus repeatedly reworked quartz grains, such as those in large-scale low-734 gradient river systems or in deserts, almost always have a strong luminescence response to dose. Once 735 sensitivity has been confirmed, any dating application must also consider: the reliability of the chosen 736 SAR protocol; the degree of resetting by daylight or heat before burial; and the dose rate during burial. 737 Below we review some illustrative dating applications in this context, and Table 1 summarises different 738 events that can be constrained in time using quartz OSL. 739
- In fired or naturally-heated samples, the luminescence signal follows **Arrhenius** kinetics, where exponential depletion depends on both time and temperature<sup>17</sup>. Typically, heating for one hour at 250 °C or one minute at 300 °C removes 99% of the fast-component OSL<sup>57</sup>. Such heating conditions are commonly exceeded in the firing of pottery, ceramic bricks and tiles<sup>131, 132</sup>, in heated stones<sup>133, 134</sup> and lithics<sup>135</sup>, in sediment heated under archaeological hearths<sup>136</sup>, and in contact with lava flows<sup>137</sup>.
- Local temperatures can also be raised a few hundred degrees by frictional heating associated with formation of fault gouges during tectonic movement, or dike injection due to earthquakes, completely

resetting OSL<sup>138-140</sup>. Lightning strikes can cause localised melting in dry sand, creating fulgurites and
 resetting nearby grains<sup>141</sup>.

Since daylight exposure is ubiquitous (Figure 1), most sedimentary settings can be dated using OSL (Figure 9). The bleaching rate depends on both the daylight flux and spectrum<sup>31, 142</sup>. Under water, the depth and suspended particle load reduce photon flux, especially at shorter wavelengths, and turbulence carries particles into and out of the illuminated zone; all these processes reduce the bleaching rate. Subaerial transport is thus more effective at resetting OSL than sub-aqueous. Nevertheless, dating is often possible in completely dark sub-glacial, cave, deep lake<sup>143</sup> and deep ocean environments<sup>5, 144</sup> because the sediments were sufficiently exposed to daylight before last entrainment.

#### [H2] Landscape evolution and past environments

The rapidity of daylight bleaching implies that usually only the most recent transport and deposition 757 event is dated using OSL. Applications in landscape evolution and palaeo-environmental studies are 758 numerous (Table 1) and have resulted in major advances in understanding a variety of geomorphic 759 processes, including thresholds, leads and lags<sup>145-148</sup>. OSL dating of a wide range of terrestrial sequences 760 has also demonstrated the interrupted nature of terrestrial records<sup>95, 149</sup>. Many deposits record only short 761 and sub-millennial periods of time, often when the climate is changing rapidly from one relatively stable 762 state to another<sup>150-153</sup>. These results have raised questions about the validity of past land-sea and land-763 ice correlations that were usually based on the assumption of continuous deposition in the absence of 764 numerical chronologies<sup>154-156</sup>. OSL dating has changed our view of the Weichselian ice limits in 765 northern Eurasia (QUEEN project<sup>157, 158</sup>) and the retreat of the British-Irish ice sheet (BRITICE-766 CHRONO project<sup>159, 160</sup>). It has also provided one of the foundations of the INOUA Dunes Atlas, a 767 global chronological database on periods of inland or continental sand dune accumulation and 768 stabilization<sup>161, 162</sup>. Dating of frost wedges has demonstrated landscape stability over an entire glacial 769 cycle<sup>163</sup> and constrained palaeotemperatures. In addition, OSL dating of sediment covering faults has 770 enabled the constraining of the timing of past earthquakes<sup>164-167</sup>. 771

772

OSL is also reset by daylight exposure during the vertical reworking of grains by bioturbation (including 773 the formation of biogenic structures such as termite mounds, Fig. 9e), pedoturbation and cryoturbation 774 <sup>168-173</sup>. These additional reset mechanisms have led to new research possibilities of relevance to the 775 Anthropocene (Table 1), particularly in understanding critical zone processes and rates of sediment 776 turnover<sup>174, 175</sup>. In a palaeosol developed in a sediment, turbation continually reset grains during soil 777 development and ceased when the surface was buried. Thus grains in the subsoil below the mixing zone 778 are expected to record the time the sediment was deposited, and the now buried topsoil records the time 779 soil formation stopped<sup>172</sup>. 780

781

All of these processes are climate-driven, or at least climate-dependent. Establishing the absolute timing and rates of change of such landscape development is thus key to identifying periods of climate change, and to establishing the correlations between these local changes and the global climate record<sup>95, 176, 177</sup>. Luminescence is particularly useful among the techniques that can be used to establish such chronologies because it targets the silt and sand grains, which are present in all landscapes.

### 787 [H2] Archaeology and migration studies

Quartz OSL dating has made significant contributions to Holocene archaeology, including dating of 788 Neolithic settlements<sup>178, 179</sup>, Archaic mound complexes in the Americas<sup>180</sup>, Bronze and Iron Age 789 middens <sup>181</sup>, rock art<sup>182, 183</sup>, medieval bricks<sup>184</sup>, mortar <sup>185</sup> and stone surfaces<sup>186-188</sup>. On longer time scales, 790 the diverse sedimentary environments dated using OSL also served as habitats for past human and 791 faunal populations. As a result, quartz OSL is the preferred dating tool in many anthropological and 792 palaeontological applications (Table 1). This preference holds despite the greater precision of other techniques such as argon-argon or U series dating. In contrast to OSL dating, argon-argon or U series 794 dating are very restricted in application (for example, argon-argon dating targets infrequent volcanic 795 ejecta), and even if applicable, usually only give a small number of ages in a section. 796

- OSL dating has made major contributions to debates on the timing of human evolution and migration and on changes in palaeoecology, especially for events beyond the limit of radiocarbon dating (~45 ka). Important results include timelines for Out-of-Africa dispersals of *Homo sapiens*<sup>189-194</sup>, the earliest known human settlement of Australia<sup>195-198</sup> and possibly of the Americas<sup>199, 200</sup>, occupation dynamics of Neanderthals and Denisovans<sup>201, 202</sup>, key evolutionary, behavioural and cultural turning points across different human species<sup>203-206</sup>, and the late Pleistocene megafaunal extinctions in Australia and the
- 803 Americas<sup>207-210</sup>.

#### [H1] Reproducibility and data deposition

Complexities in OSL dating arise from four main factors: limited net daylight flux causing some grains to not completely reset at deposition; different OSL signals from different grains in response to the same radiation, heating and optical stimulation; different radiation fluxes to different grains because of heterogeneity in mineralogy, grain size and burial matrix; and open system deposits leading to timedependent radiation fluxes because of post-depositional mixing, geochemical and diagenetic processes, and variable water contents.

Limited net daylight flux leads to heterogeneity in the apparent D<sub>e</sub>. Different OSL signals from different

grains lead to heterogeneity in the apparent D<sub>e</sub>, and both different radiation fluxes to different grains

- and open system deposits lead to heterogeneity in dose rate and thus in natural dose. All of these factors
- affect the ease with which OSL dating can be applied in different settings (summarised in **Fig. 10**).

#### 815 [H2] Heterogeneity in De

The major factors affecting the complexity of determining  $D_e$  are discussed below. Their relative 816 significance is summarised in Fig. 10a shows that, for example, aeolian sediments are likely to show 817 low complexity (furthest from the centre), because they tend to be well bleached (red band) and have 818 good luminescence characteristics (dark blue band), both with low variability (width of the band). In 819 contrast, glacial sediments are likely to show high complexity (close to origin), because they tend to be 820 poorly bleached and have poor luminescence characteristics with considerable variability (width of red 821 and dark blue bands). Bleaching becomes progressively less important and contributes less to variability 822 as the burial age increases. The complexity arising from bleaching therefore decreases with age, 823 824 whereas complexity from luminescence characteristics is at a maximum for oldest sediments, mainly because of dose saturation effects, and a minimum for medium age material, where signals are strong 825 and the saturation effects are small. 826

The nature of grain transport by wind or water implies that every grain is exposed to a different daylight 827 flux and spectrum. Thus, for short transport duration under attenuated daylight, the latent OSL signals 828 from different grains are reset to different degrees, and only when the exposure is of sufficient duration 829 will all grains be fully reset. Field environment and age are the major factors affecting the completeness 830 of bleaching, and they dictate the position and width of the red band in Fig. 10a. In general, given the 831 wavelength-specific attenuation in a water column, and the wavelength dependence of bleaching, the 832 grain-to-grain variability of bleaching in water-lain sediments is greater than in aeolian sediments. 833 Nevertheless, some flood deposits are surprisingly well-bleached<sup>211, 212</sup> suggesting that significant 834 bleaching occurred before fluvial entrainment or sub-aerially during low flow. 835

If all grains are fully bleached and the dose rate is homogeneous, the D<sub>e</sub> distribution of a sample should 836 be centred on a single value with dispersion accounted for by random measurement uncertainties<sup>48, 213,</sup> 837 <sup>214</sup>. Unfortunately, this is rarely, if ever, the case. Additional dispersion in D<sub>e</sub> originates from differences 838 in the behaviour of OSL signals from different grains, and this compromises both the accuracy and 839 precision of the chosen SAR protocol (see the dark blue band in Figure 10a, which represents intrinsic 840 sources of dispersion). These difficulties arise because the luminescence from different grains responds 841 differently to radiation, heating and optical stimulation <sup>91, 104, 106, 215, 216</sup> or because of different charge 842 transfer properties and different OSL signal compositions<sup>59, 217-219</sup>. In addition, the process of 843 interpolation onto the dose response curve (Fig. 7) will give rise to an asymmetric dose distribution 844 with an increased dose-dependent dispersion compared to that of the OSL distribution. At the limit, a 845 fully saturated sample gives an incorrect, apparently bounded, dose distribution (Fig. 7), with the 846 potential to be misinterpreted as a poorly bleached distribution or as mixture of a finite dose with a 847 848 saturated sample.

In addition to complications arising from these intrinsic factors,  $D_e$  dispersion also arises from extrinsic sources such as variations in bleaching, mixing, and dose rate. When dispersion in  $D_e$  is significant, complex statistical analysis may be necessary<sup>77, 213, 220-225</sup> to extract the dose component of interest. It is not straightforward to differentiate between intrinsic and extrinsic sources of dispersion because we do not have a way of independently measuring the extrinsic component. As a result, any dispersion greater than that determined in laboratory experiments is simply assumed to be extrinsic<sup>53</sup>. However, there are cases where this assumption is incorrect<sup>94</sup>.

Although the size of the apparent residual dose remaining after bleaching is independent of the 856 subsequent burial time, this burial timing affects the residual dose by affecting 'bleaching' (see red band 857 in Burial age in Fig. 10a). There are three approaches to investigating whether a sample is likely to have 858 been sufficiently bleached at deposition: first, modern analogue samples<sup>226</sup> provide site-specific 859 evaluations of bleaching conditions<sup>111, 227, 228</sup>, which assume that the net effects of transport and 860 deposition on bleaching have not changed through time. In general, modern analogues have not 861 completed their transport at the time of collection and because most recently deposited sediment is held 862 in ephemeral storage such as point bars, it is likely to be reworked again before final deposition in long-863 term storage. Thus modern analogues will tend to underestimate the degree of bleaching available in 864 the environment<sup>226</sup>. The second approach involves examining the self-consistency of OSL dating 865 through measurement of different minerals with different luminescence signals. For example, paired 866 dating comparisons of quartz OSL and K-rich feldspar IRSL or post-IR IRSL, where the two feldspar 867 signals are optically reset at different rates, can be used to confirm that all quartz grains in a sample 868 were completely bleached prior to deposition<sup>229-231</sup>. Finally, the third method uses single-grain OSL 869 dating to examine heterogeneity in De in sediments from environments with likely limited daylight 870 exposure or post-depositional alteration<sup>114, 232</sup>. In such cases, heterogeneity in radiation dose rate at the 871 individual grain level must also be considered (see below). 872

Unfortunately, analytical models based on the widely-used Central Age Model (Gablbraith et al., 1999; such as the Minimum Age Model (Ref. Galbaraith et al. 1999) and Finite Mixture Model (Roberts et al., 2000)) do not take this dose rate heterogeneity into account and so risk producing biased estimates of De (Guérin et al., 2017, QG).

This can be illustrated by considering a hypothetical sample in which the distribution of dose-rates 877 dominates the natural dispersion. Suppose this to be a well-bleached, well-behaved sample in which the 878 uncertainty on the measurement of dose is negligibly small (e.g. 1%). All grains must by definition 879 record the same age. Suppose this sample is 10 ka old, and that the environmental dose rate is made up 880 881 of two equal but spatially separated (at the grain scale) contributions, one of 1Gy.ka<sup>-1</sup>, and one of 10 Gy.ka<sup>-1</sup>; we would measure an average dose rate of 5.5 Gy.ka<sup>-1</sup>. Now consider the dose - on average, 882 50% of the grains absorb a burial dose of 10 Gy, and 50% a dose of 100 Gy. If these two doses could 883 be measured individually, each with some similar overall uncertainty, then the unweighted average dose 884 would be 55 Gy, the geometric mean 31.6 Gy and the weighted average 10.9 Gy. Only the average dose 885

gives the correct age of 55/5.5 = 10 ka; the geometric mean age is 31.6/5.5=5.7 ka, and the weighted data give 10.9/5.5=2.0 ka.

Of course such simple bi-modal distributions do not exist in nature, but the point remains – when only an average dose rate is known, and the dose rate is a major factor in dispersion, the dose must also be an average, or the age estimate is likely to be biased. This is particularly true of large aliquots, where the dose is in any case averaged as a result of the large number of grains giving light in each aliquot, and analytical uncertainties on dose estimates from each aliquot are very much smaller than the data dispersion. This point is made in Guerin el al. (2017), where the implications for the analysis of dose distributions are discussed in detail.

895

Site stratigraphy and grain sorting are of considerable importance to reliable OSL dating (Fig. 10a). 896 Sediments affected by significant post-depositional mixing via, for example, bioturbation, freeze/thaw 897 cycles, tectonic activity, or cultivation should be avoided as these processes can lead to significant 898 dispersion in  $D_e$  values, but sampling potentially-mixed sediments may be unavoidable at times. In 899 some cases this mixing is obvious but not always, and the presence of distinguishable components in 900 small-aliquot or single-grain dose distributions has been used to infer mixing<sup>233, 234</sup>. These dose 901 populations can be modelled as a mixture of discrete components using the Finite Mixture Model<sup>235</sup>, 902 and typically the dominant component is chosen to provide the deposition age<sup>234, 236-239</sup>. However, such 903 analyses can imply unrealistic degrees of physical mixing between non-adjacent layers even in deposits 904 where mixing can safely be ruled out<sup>94</sup>, suggesting that the distributions identified by dose mixing 905 models may not necessarily arise from physical mixing<sup>240</sup>. Together with the inability to derive 906 component-specific dose rates for mixed sediments (see below), this calls into question the application 907 of dose mixing models for dating purposes 908

909

#### 910 [H2] Heterogeneity in dose rate

The major factors affecting dose rate complexity are discussed below, and their relative importance is summarised in **Fig. 10b**.

Dose rate evaluations become demanding when the infinite matrix assumption is no longer applicable 913 <sup>52</sup> (red end of the 'Dimension of deposit' bar in **Fig. 10b**). Sand-sized quartz grains have a scattered  $\beta$ 914 spectrum range comparable to that in sediments; the larger the grain, the higher the attenuation of the 915 external  $\beta$  dose rate (Supplementary Table 4). In addition, the distribution of radionuclides at the grain 916 scale in sediments is spatially heterogeneous with variations in both internal and surface-bound 917 radioactivity. Thus 'hotspots' of higher radioactivity, such as K-feldspar and zircons, or 'cold spots' of 918 lower radioactivity, such as carbonate nodules, result in significant variations in the  $\beta$  dose rate to 919 individual grains. This spatial variability can be observed<sup>135, 241, 242</sup>, modelled and, on average, accounted 920 for<sup>53, 54, 243</sup>, but the association of spatially-varying  $\beta$  doses with individual grains from which a D<sub>e</sub> has 921

been estimated is currently not possible. Such variability is usually lower in a well-sorted deposit (green

923 end of 'Sorting' in **Fig. 10b**).

Heterogeneities on a scale of tens of cm affect the  $\gamma$  ray dose rate, which may result from the proximity of a sample to a large rock fragment of a different mineralogy, or from interfaces between different stratigraphic units (see Appendix H in<sup>17</sup>). While in many cases these heterogeneities can be avoided, the choice of sampling location is often restricted in archaeological sites. In that case, the  $\gamma$  dose rate to samples collected close to stratigraphic interfaces may require modelling<sup>244</sup> or in situ  $\gamma$  dose rate measurement using luminescence dosimeters or field  $\gamma$  spectrometers<sup>25-28, 30, 245, 246</sup>.

Water content is particularly important (see **Box 1**) as it reduces the dose rate compared to a dry matrix<sup>17</sup>. 930 <sup>247</sup>, and a 1% change in water mass fraction typically results in a 1% change in apparent OSL age. Thus, 931 a low value for 'Saturated water content' (Fig.10b) reduces the possible range of water contents during 932 burial, and therefore reduces the dependence on assumptions concerning 'Fraction of saturation' of 933 water content (Fig. 10b). Whatever the saturated water content, if it is likely that for the entire burial 934 period the sample has either been dry, for example desert sand, or fully saturated, for example below 935 the water table, then uncertainties arising from assumptions are much reduced. The least desirable 936 condition is a sample in a temperate climate vadose zone, where the average fraction of saturation 937 throughout the burial period is poorly defined (see red centre of band in Fig. 10b). In such cases, there 938 may be no alternative to assuming 50% of saturation with some appropriately large uncertainty on that 939 value. 940

The dose rate is also affected by temporal changes<sup>55, 248</sup>. For instance, post-depositional changes in 941 burial depth can be important for shallow burial (see red end of 'Overburden thickness' in Fig. 10b). 942 Changes due to erosion or deposition increase complexity by affecting both  $\gamma$  and cosmic ray dose 943 rates<sup>16</sup>, but are likely to be less important for deep burial (see green end of band in **Fig. 10b**). Density 944 changes due to porosity alone do not change dose rates because the density of air is too low to absorb 945 significant amounts of energy. However, density changes arising from time-dependent carbonate 946 infilling/leaching of pore space, or oxidation of organic material, change the effective dose rates and 947 the saturated water content (Fig. 10b)<sup>249, 250</sup>, which directly and indirectly increases complexity (see 948 Supplementary Tables 5-6 for correction factors). 949

The dose rate may also change because radionuclides are imported/exported from a volume of sediment. Radionuclide concentrations are affected by water movement, which may import specific radionuclides into secondary minerals or leach them by reduction and/or hydrolysis with time, particularly <sup>40</sup>K, <sup>238</sup>U, and <sup>226</sup>Ra (a shorter-lived member of the <sup>238</sup>U decay series<sup>21, 23</sup>). This import/export leads to disequilibrium in the <sup>238</sup>U decay series and possibly time-dependent dose rates<sup>251</sup>. In addition, the 3.8 day half-life of the noble gas <sup>222</sup>Rn (<sup>238</sup>U series) allows migration through sediment pores before decay.

- <sup>956</sup> Diffusion dominates for partial pore-filling by water, but bulk transport can occur if water moves <sup>957</sup> through a deposit<sup>252</sup>, with losses of 50% at 5 m depth in sand having been reported<sup>253</sup>. Although the <sup>958</sup> post-<sup>222</sup>Rn part of the <sup>238</sup>U decay series contributes the majority (>75 %) of the  $\beta$  and  $\gamma$  dose rate from <sup>959</sup> the <sup>238</sup>U series (Supplementary Figure 3), for a typical sandy sediment containing 20 Bq.kg<sup>-1</sup> <sup>238</sup>U and <sup>960</sup> <sup>232</sup>Th and 300 Bq.kg<sup>-1</sup> <sup>40</sup>K (1.5 and 5 ppm, and 1% respectively), the contribution to the total dose rate <sup>961</sup> is <20%. A combination of thermal ionisation mass spectrometry and high-resolution  $\gamma$  spectrometry
- $_{962}$  can be used to describe the state of equilibrium of, for example, the  $^{238}$ U decay series, but in most cases,
- <sup>963</sup> disequilibria have rather small effects on the OSL age.

Finally, any significant 'post depositional mixing' (see red end of bar in **Fig. 10b**) increases the challenge of determining a time averaged dose rate. In a mixed deposit, grains will have absorbed dose in potentially different dose rate environments before mixing, but only the mixed dose rate is measurable today. Without knowledge of the dose rates in the source deposits contributing to the mixture and of the timing of the mixing, reconstruction of an average dose rate is uncertain.

In summary, dose rates at the grain level are likely to be variable during burial. This variability must be taken into account when using the age equation, and accurate ages should be calculated using a model which delivers average estimates of the absorbed doses during burial (Guérin et al., 2017, QG).

#### 972 [H2] Checking reproducibility

One way to reduce the impact of the various methodological issues discussed above is through internal and external checks on reproducibility. These may, for example, involve collecting a suite of OSL samples targeting different types of sediments preserved in clear stratigraphic relationships, or by collecting replicate samples from the same horizons<sup>95, 99, 198</sup>. Although resource-intensive, multipletechnique and multiple-sample OSL dating strategies are recommended in all dating applications<sup>95-97,</sup> 105, 135, 198, 201, 231.

Statistical uncertainties associated with individual laboratory measurements -conventionally given at 979 980 68% confidence intervals in luminescence dating- are typically 2-3%, both on natural dose measurements involving OSL and on dose rates (as determined from  $\gamma$  spectrometry). These random 981 uncertainties can be reduced by increasing the number of measurements, but this requires a considerable 982 increase in instrument time per sample and is often impractical. Studies involving high-density sampling 983 from large stratigraphic sections frequently report over-dispersed ages, where the ages are more variable 984 than expected from analytical uncertainties alone (see Fig.  $11a^{95}$ ). This suggests that reductions in 985 measurement uncertainty would be of limited benefit. Note that Figure 11a does not include the 986 systematic uncertainties common to all the ages in the section, and that the error bars represent random 987 uncertainties only. 988

The stated uncertainties on calibration standards are also ~2-3%, both for the  $\gamma$ -irradiated quartz <sup>47</sup> used to calibrate laboratory  $\beta$  sources and for the geochemical standards used for the calibration of equipment used in dose rate determination<sup>254</sup>. Together, these contributions result in typical total calculated uncertainties (random and systematic) on an OSL age ranging 5-10% (Supplementary Table 7). Currently, 5% is considered a minimum uncertainty for a sample in an ideal setting and >10% for unfavourable cases involving, for instance, low luminescence sensitivity or saturation of OSL with dose.

- Perhaps because OSL dating is a relatively young method, with the present methodologies dating back 995 to the 2000s and most laboratories small with fully committed instrumentation, the community has yet 996 to develop a routine of inter-laboratory comparisons. The only exception is an exercise in which 26 997 different laboratories dated a beach-ridge sand selected for its good OSL behaviour<sup>255</sup>. The dispersion 998 in the 26 ages (~18%) significantly exceeded the uncertainties on individual ages as estimated by the 999 1000 various participants. Some of the dispersion arose from different assumptions: for example, the use of a 'typical' internal dose rate to quartz by some, while others ignored this possible contribution. In this study, it is clear that the community, as a whole, underestimated the uncertainties they assigned to 1002 individual age estimates. 1003
- In addition, laboratories make use of either their own age calculation software or one of the more widely available packages, such as DRAC<sup>256</sup>, RLuminescence<sup>257</sup> or numOSL<sup>258</sup>. These software all include underlying, often dissimilar, default assumptions, and a rigorous comparison of these packages is highly desirable but not yet established.

Until recently most articles containing luminescence ages presented new data with a variable degree of 1008 detail. However, publications addressing larger scale questions and requiring meta-analyses of 1009 published datasets are becoming more frequent. It is thus increasingly important that all published OSL 1010 ages are accompanied by underlying data of sufficient detail to allow the reader to reproduce the age 1011 calculations and reliably compare ages from different laboratories. An example of the minimum sample 1012 specific information necessary for such calculations is given in **Table 2**. There should also be text 1013 describing the experimental parameters and the results of any additional performance tests, especially 1014 dose recovery experiments. We hope that all journals, even those without specific interests in the 1015 technicalities of dating, will encourage authors to publish such information. 1016

#### 1017 [H1] Limitations and optimizations

The problems encountered in quartz OSL dating often depend on the age of the sample. The ability to date young samples with small doses depends on the sensitivity of the luminescence signal to radiation. For less sensitive quartz, it can be difficult to detect a measurable fast-component signal above the instrument's background and slower decaying OSL signals. Thus dating young desert dunes is much more likely than dating young mountain-valley slope deposits (see Fig. 10a). Also, thermal transfer can
usually be avoided by a low temperature preheat (Fig. 6b).

For older sediments, it may take weeks of instrument time to collect the data required for a single age, 1024 which is a major restriction for laboratories with limited instrumentation. Standardised dose-response 1025 curves are sometimes suggested to reduce this measurement time<sup>100</sup>. For large suites of samples with 1026 similar properties such as, for example, a loess section, the average responses of multi-grain aliquots of 1027 quartz follow a common dose-response curve as demonstrated by examining a sub-set of the suite <sup>259</sup>. 1028 In such cases, individual L<sub>n</sub>/T<sub>n</sub> values can be interpolated on to this common dose-response curve to 1029 determine individual De. This approach is potentially useful for younger, homogeneous samples within 1030 the linear part of the dose-response curve<sup>100, 260, 261</sup>. However, there can be significant reproducibility 1031 issues with less homogeneous material, and particularly as the dose-response curve approaches 1032 1033 saturation.

More generally, the apparent behaviour as the laboratory dose response curve approaches saturation 1034 may be misleading. The controls on luminescence saturation with dose are poorly understood. The 1035 luminescence from individual grains and sometimes from multi-grain aliquots can saturate at very 1036 different doses, continue to grow without apparent saturation, or even decrease at high doses<sup>101, 262</sup>. 1037 However, quartz often underestimates the true age at large doses (150-250 Gy)<sup>64, 66, 263</sup> and it seems 1038 likely that at these large doses the laboratory dose response curve does not reflect the natural dose curve 1039 <sup>65, 264, 265</sup>. One way to address this very important question is to investigate natural dose response curves 1040 using homogeneous material from sections with a well-known age model, such as some well-studied 1041 loess sections<sup>65, 266</sup>. New measurement strategies and statistical tools have also been proposed with the 1042 aim of increasing the range of single-grain OSL<sup>267, 268</sup>. 1043

#### 1044 [H2] Overall accuracy

The overall performance of sand-sized quartz OSL to date samples from a variety of deposition environments, each with independent age control, has been evaluated by others<sup>89, 108, 269</sup>. These data sets are combined in **Fig. 11b**, for 249 ages there is now a considerable body of data suggesting that OSL dating is accurate over a time span of more than four orders of magnitude (**Fig. 11b**), and that the precision and accuracy are largely in the range originally predicted.

#### 1050 **[H1] Outlook**

Over the last twenty years, dating based on the quartz fast-component OSL signal has become an important tool in the geochronology of the past 500 ka. The method is still undergoing rapid development and improvement and exciting new areas of application are waiting to be explored.

- The origins of intrinsic over-dispersion in D<sub>e</sub>, both at the multi- and single-grain level<sup>48, 232</sup>, begs further 1054 investigation. An improved understanding and quantification of these intrinsic sources would, in turn, 1055 enable more reliable quantification and interpretation<sup>242</sup> of the contribution to dispersion from extrinsic 1056 sources, such as incomplete bleaching and mixing. Limits on the upper dose range of the OSL signal are also poorly understood. Laboratory dose response curves often increase to large doses (hundreds of 1058 Gy), but natural doses either do not increase above ~200 Gy, or if they do, they often appear to be an 1059 underestimate<sup>66, 263, 270</sup>. An improved understanding of the fast-component dose response curve at high 1060 doses would increase confidence in older ages and help to extend the datable range. Other quartz signals 1061 such as Violet Stimulated Luminescence<sup>7, 271</sup> and Thermally Transferred OSL<sup>272-274</sup> also have the 1062 potential to date older material, but both require further research. 1063
- Accurate calibration of laboratory radiation sources is an active research area<sup>47, 275</sup> and a regular programme of radiation source comparisons is desirable.
- Addressing these issues should help improve luminescence age accuracy to the probable minimum of ~5%. As the demand for OSL dating continues to grow, the demonstrated benefits of a high stratigraphic age density to site interpretation<sup>148, 198, 276-278</sup> will only increase this demand further.
- Single grain dating has demonstrated its potential for dating incompletely bleached deposits  $^{213, 225, 279, 280}$  but inherent dosimetry uncertainties likely prevent the accurate dating of deposits subject to significant post-depositional mixing<sup>77</sup>. For unmixed deposits, the ability to determine dose rates specific to the individual grains giving a D<sub>e</sub> would likely produce a step change in interpretation and application.
- Field measurements of OSL have been demonstrated<sup>281-283</sup> and the ability to determine approximate ages 'on site' would be valuable. Although there is no technical impediment, calibrating the signal in terms of dose requires a portable ionising radiation source, which has regulatory barriers to overcome.
- Direct dating of buried rock surfaces is rapidly growing<sup>127, 284</sup>, although quartz applications are limited 1076 to sandstones and quartzite clasts because of sensitivity. One advantage of direct dating is that a rock 1077 surface records its history of daylight exposure, which eliminates the uncertainty in whether it was 1078 sufficiently bleached<sup>187, 188, 285, 286</sup>. The potential applications in both geology for cobble fans, moraines, 1079 scree/talus, and rock falls, and in archaeology for megaliths, field boundaries, buildings, and lithics, are 1080 considerable. Interest in the inverse problem of dating the length of daylight exposure of a rock surface 1081 is just beginning<sup>127, 287</sup> and currently requires a known-age calibration surface<sup>15, 287</sup>. However, the 1082 potential of this method is considerable, especially in archaeology. 1083
- Finally there is considerable opportunity to use the signal remaining after daylight exposure to investigate geomorphological processes, such as the relative importance of supra/sub-glacial transport<sup>288</sup> or fluvial residence time<sup>289</sup>.

- 1087 OSL dating of quartz continues to expand its fields of application beyond those mentioned in this
- 1088 Primer. The method continues to develop, and will undoubtedly contribute further to our understanding
- 1089 of Earth surface processes.

1090

1091

### 1092 Glossary

- (Some definitions are specific to the context of this Primer. Where appropriate, units are given in square brackets, e.g.  $[s^{-1}]$ )
- The **Arrhenius** equation states that, for an electron to escape a trap, it must first acquire a minimum amount of energy, called the activation energy  $E_a$ . Then the rate of thermal eviction of trapped
- electrons is given by  $Ae^{-\frac{Ea}{k_BT}}$ . The 'frequency factor' A can be seen as the number of attempts to
- escape made by the trapped electron, and  $e^{-\frac{E_a}{k_B T}}$  as the probability that any given attempt will result in an escape at a particular absolute temperature T.  $k_B$  is Boltzmann's constant.
- $\alpha$  particles: ionising radiation in the form of energetic helium nuclei produced by nuclear decay.
- Anthropocene: a proposed geological epoch covering the period of significant impact on Earth's
- 1102 geology and ecosystems by humans.
- Ar-Ar (or K-Ar) dating: a dating technique based on the decay of  ${}^{40}$ K to  ${}^{40}$ Ar.
- $\beta$  particles: ionising radiation in the form of energetic electrons produced by nuclear decay.
- bioturbation: after initial deposition, the reworking of sediment by biota (plants, animals, etc).
- **Bq:** Becquerel, a nuclear disintegration per second. [s<sup>-1</sup>]
- cal BP: calibrated <sup>14</sup>C years before present (referred to AD1950). Only used for <sup>14</sup>C ages, after
- 1108 conversion to calendar years by calibration. [a]
- 1109 **Central Limits theorem:** states that the sum of independent random variables tends toward a normal
- distribution, even when the original variables themselves are not normally distributed
- cosmogenic radionuclide dating: any one of several methods to date length of exposure of minerals
- at the surface (or sometimes length of burial) using the build-up or decay of radioactivity induced by
- 1113 cosmic rays.
- **coversand:** a continuous layer of sand deposited by wind, often in a coastal environment.
- **crotovina:** An animal burrow that has been filled with material from overlying sediments.
- cryoturbation: the reworking of sediment after initial deposition by the freeze/thaw cycle.
- **dark count rate:** the count rate from a photon detector completely obscured from light. [s<sup>-1</sup>]
- dose: total amount of energy absorbed per unit mass from ionising radiation. [Gy]
- dose rate: the rate of energy absorption from ionising radiation during burial
- 1120  $D_c$  (or  $D_{0 (letter o)}$ ): a constant used to describe the curvature of the luminescence response to dose,
- usually in an expression of the form  $Y=A(1-exp(-D/D_c))$ . [Gy]
- $\mathbf{D}_{e}$ : equivalent dose, the laboratory dose that induces the same luminescence response as the natural
- 1123 dose. [Gy]
- 1124 **ejecta:** material thrown out by volcanic eruption.
- fast-component: the part of the signal that decays most rapidly during stimulation
- feldspar: a group of K-, Na- and Ca-rich aluminosilicate minerals, makes up ~40% of the Earth's
   crust.
- **frost wedges:** cracks formed in frozen ground, often filled with sand; this preserves them in the sedimentary record.
- **FWHM: Full Width at Half Maximum, a numerical description of the width of a peak in data or**
- response.
- $\gamma$  rays: ionising radiation in the form of energetic photons produced by nuclear decay.
- <sup>1133</sup> **Gy:** Gray, energy absorbed per unit mass from ionising radiation. 1 Gy = 1 Joule/kg  $[J.kg^{-1}]$ .
- **igneous rock:** formed by the solidification (crystallisation) of magma or lava.
- 1135 **IRSL**: Infra-red stimulated luminescence
- 1136 **ka:** thousands of years. [a]
- 1137  $\lambda$ : lambda, the nuclide-specific probability of a nuclear decay. [s<sup>-1</sup>]
- leads and lags: in climate science, the difference in timing between changes in two correlated climate
- phenomena (e.g. build-up of global ice and changes in regional dust deposition).
- loess: wind-blown dust (predominantly silt-sized (4-63 μm) grains)
- 1141 **luminescence signal:** the luminescence response,  $L_i$ , to any dose, divided by the response,  $T_i$ , to a
- subsequent fixed test-dose, i.e.  $L_i/T_i$
- 1143 **midden:** in archaeology, a refuse pile of domestic waste.
- 1144 **Neolithic:** the most recent division of the Stone Age (last ~12 ka), and associated with the
- development of farming.
- optically stimulated luminescence OSL: the luminescence signal observed when stored charge is
   stimulated (evicted from traps) by visible or near-visible light.
- 1148 **overburden:** the material covering some geological feature of interest.
- palaeosol: a fossil soil (i.e. one no longer forming); usually found buried within a sequence ofsedimentary units.
- **pedoturbation:** the reworking of sediment after initial deposition by the geochemical and biological processes of soil formation.
- **primary rock:** any rock that formed by solidification (crystallisation) from the melt.
- **Quantum efficiency:** how many photons are required to give a detected event, and the effectiveness of photon to electron conversion
- quartz: the most common natural form of the compound SiO<sub>2</sub>, makes up about 20% of the Earth's
   crust.
- radiocarbon dating (or <sup>14</sup>C dating): a dating technique based on the decay of <sup>14</sup>C. Can only be applied to material of organic origin.
- 1160 Reset: in the case of luminescence, set to zero or 'bleached'
- secular equilibrium: where the production and decay rates of each nuclide in a decay chain are almost identical.

- sedimentological boundaries: strictly, distinct breaks (unconformities) between bodies of sediment.
- Often used more informally to describe gradational boundaries.
- SAAD Single Aliquot Added Dose: a single aliquot procedure for determining D<sub>e</sub>, by adding doses
   to the natural dose and measuring the growth of the resulting luminescence signal.
- 1167 SAR Single Aliquot Regenerative dose: a single aliquot procedure for determining D<sub>e</sub>, based on
- resetting the luminescence signal to zero between each measurement of luminescence, whether resulting from natural or laboratory doses
- 1170  $\tau_{\frac{1}{2}}$  half life: The period of time taken for 50% of a particular radionuclide to undergo radioactive 1171 decay and change into another element. [s]
- thermoluminescence: the thermally stimulated luminescence signal observed when a sample isheated to evict stored charge from traps.
- threshold: In climate science, the point at which a small change in a physical or chemical parameter
- (e.g. global temperature) can lead to a tipping point in outcome and a shift to a new equilibrium state
- of range of average conditions (e.g. massive loss of global ice mass)
- vadose zone: in a porous body (e.g. of sediment), that part lying between the ground surface and the
  water table.

## **Figures captions**

Figure 1: The origins of the luminescence dating signal in quartz. (a) energy is absorbed (dose) as 1181  $\alpha$  and  $\beta$  particles and  $\gamma$  rays emitted during the decay of naturally occurring radionuclides (especially 1182 <sup>238</sup>U and <sup>232</sup>Th series, <sup>40</sup>K) penetrate quartz grains. The different ranges of the various emissions are 1183 indicated, relative to typical grain dimensions. Cosmic rays also contribute to the dose. (Redrawn 1184 from<sup>290</sup>) (b) Energy from ionising radiation is absorbed by an electron (e<sup>-</sup>, filled blue circle), which 1185 migrates through the grain until it is trapped at a defect in the lattice structure (electron trap). The 1186 corresponding 'hole' (h<sup>+</sup>, literally the place where an electron should be, unfilled orange circle) also 1187 migrates until it is trapped in a different kind of defect (hole trap). This process stores energy. (c) 1188 Shining blue light on the crystal gives the trapped electron sufficient energy to escape the trap (optical 1189 stimulation). When it then recombines with the hole, it releases the trapped energy as a more energetic 1190 UV photon (optically stimulated luminescence). 1191

Figure 2: The relationship of the erosion, transport and sedimentation cycle to the acquisition 1192 and loss of a luminescence dating signal (a) any existing energy stored in the quartz crystal structure 1193 is released by daylight exposure (i,iii,v) during different erosion and transport events, by wind, water, 1194 ice, gravity or turbation (see (b)). These periods of resetting are interspersed with periods of burial 1195 (ii,iv,vi), during which dose is absorbed from natural radiation (Figure 1). Sufficient net daylight 1196 exposure completely removes the previously stored energy (e.g. during i and v), otherwise it is only 1197 partially reset (iii). This energy builds up again during the most recent burial (vi), until the quartz sand 1198 grains are sampled (vii) and taken to the laboratory. After cleaning, they are then exposed to blue light, 1199 and the luminescence signal acquired during last burial is measured. (b) illustrates the various transport 1200 agencies that can lead to daylight exposure of mineral grains before final burial (gravity acts as an 1201 independent transport agency only on high gradients).

1203

Figure 3: Sample preparation and instrumentation for OSL measurement. (a) The laboratory steps 1204 used to extract quartz grains from a tube of sediment. Note the alternative processing routes, depending 1205 on whether feldspar is also required. All these steps should be performed under subdued lighting 1206 conditions, using a wavelength and intensity that avoids significant effect on the trapped charge 1207 population<sup>9</sup>. Q: quartz, H: heavy minerals, F: feldspar, NF: sodium-rich feldspar, KF: potassium-rich 1208 feldspar. The combined water drops  $[\checkmark]$  and fan symbol  $[\circledast]$  indicate thorough washing and drying. 1209 1210 (b) The main components in an OSL measurement system. After irradiation in nature or in the laboratory, the quartz is heated to the desired temperature, and the stimulation light turned on. The 1211 photons making up the optically stimulated luminescence pass through the detection filter to the photon 1212 detector, to give the time dependent signal shown at the top. 1213

- Figure 4: Using SAR to determine the equivalent dose, De. (a) Ti and (b) Li plotted against dose for 1214 an aliquot of heated brick quartz; some  $T_i$  are labelled to indicate measurement order. (c) Sensitivity-1215 corrected  $(L_i/T_i)$  growth curve obtained by dividing data from (b) by data from (a); inset, stimulation 1216 curves for natural signal and calibration quartz<sup>47</sup>. (d) - (f) same as (a)-(c) but for quartz extracted from 1217 sediment. (Heated brick - 173701; monastery floor, Sweden. Sediment - 175405; Kalahari Desert, 1218 Namibia.) Open triangles show the response to D=0 Gy. Other open symbols indicate repeated dose 1219 measurements. Note that without sensitivity correction the repeated values of L (in b,e) are inconsistent 1220 with earlier measurements at the same dose; correcting for sensitivity change by diving by T<sub>i</sub> corrects 1221 this (in c,f). The interpolated doses (' $D_e$ ' in b,e and  $D_e$  in c,f) and dose response curve shapes are also significantly different between the uncorrected and sensitivity-corrected data. 1223
- 1224 Figure 5: Correcting for sensitivity changes using the luminescence response to a test dose. (a) The proportional relationship between  $L_i$  and  $T_i$  for a large aliquot (containing hundreds of grains) of 1225 180-250 µm quartz extracted from a fired brick (filled red circles, 19 cycles) and a quartz aliquot from 1226 a desert sediment (filled blue circles, 42 cycles), determined using the SAR sequence in (b). The natural 1227 signals ( $L_0/T_0$ ) are shown as open circles. Same samples as in Figure 4; sediment:  $D_i=44.4$  Gy,  $D_T=5.9$ 1228 Gy; fired brick:  $D_i=3.6$  Gy,  $D_T=1.0$  Gy. Both datasets are consistent with straight lines (dashed) going 1229 through the origin, indicating that the ratio  $L_i/T_i$  is constant for each sample. This is despite the 1230 considerable sensitivity change recorded by T<sub>i</sub> (also shown inset as a function of measurement cycle, 1231 normalized to first test dose measurement). As is common, the heated quartz displays a greater 1232 sensitivity change during SAR measurements than the quartz extracted from sediment. (b) Typical SAR 1233 measurement sequence. In the first cycle, i=0 and D<sub>0</sub> is the natural dose, or the given dose in a dose 1234 recovery experiment. After completing a cycle (step 6 or 7), the sequence returns to step 1 as required. 1235
- Figure 6: Dependence of D<sub>e</sub> and dose recovery ratio on thermal pretreatment (see text for details). 1236 (a) Preheat plateaus for sand-sized quartz extracted from loess (D38141<sup>95</sup>, coversand (OS-B<sup>96</sup>) and 1237 heated sediment from a charcoal kiln (GLL-184301<sup>291</sup>). For all three samples the D<sub>e</sub> is independent of 1238 preheat temperature over a wide temperature interval (dashed line). (b) Thermal transfer plateaus for 1239 sand-sized quartz extracted from a fluvial sand (GLL-160301<sup>292</sup>) and a beach ridge (OSL43<sup>293</sup>). Quartz 1240 aliquots were bleached at room temperature twice using blue light stimulation to remove the natural 1241 OSL signal before measurement of the thermally transferred dose. The effect of thermal transfer 1242 becomes visible for preheats  $\geq 200^{\circ}$ C. (c) Dose recovery plateau for sand-sized quartz extracted from 1243 Chinese loess. The measured to given dose ratio is insensitive to preheat temperature over a wide 1244 temperature range. (d) Histogram of dose recovery results measured with a preheat temperature of 1245 260°C. Adapted from<sup>99</sup>. 1246
- Figure 7: Effect of sublinear dose response on apparent dose distributions (based on<sup>294</sup>). Monte Carlo-modelled normalised OSL distributions shown on vertical axis, centred on 20% (black), 86%

(blue), 95% (green) and 100% (red) of saturation, each assumed normal with a relative standard deviation of 5%. The resulting interpolations onto a dose response curve ( $D_c=45$  Gy, solid pink) are given on the dose axis, with medians connected using intersecting lines. Each dose median is also shown in Gy, followed by the ratio of the median to the expected dose and the % of results giving a non-finite dose in parentheses; the arithmetic means and geometric means are all within 3% of these medians. Note that the dose median shown for 100% is the apparent value, ignoring non-intersecting OSL signals. The true median lies at infinity.

1256

Figure 8: Testing aliquot rejection criteria. The relationship between  $D_e$  and recycling ratio (a) or 1257 OSL IR depletion ratio (b) for large aliquots (containing many hundreds of grains) of 90-180 µm quartz 1258 extracted from Chinese loess. Upper figures show the individual aliquot De values (normalized to the 1259 average De of each sample) as a function of recycling ratio and OSL IR depletion ratio. Lower figures 1260 show the recycling and OSL IR depletion ratios as histograms with mean ratios plotted as blue and red 1261 dashed lines, respectively. From the upper figures it can be seen that there is no relationship between 1262 the measured De and the recycling or OSL IR depletion ratio; rejection of aliquots based on recycling 1263 or OSL IR depletion ratios is not justified. Figure adapted from<sup>95</sup>. (c) Effect of D<sub>c</sub> filtering on grains 1264 with different D<sub>c</sub>. The dose recovery data (red data, given dose 170 Gy) shows the effect on the dose 1265 recovery ratio of only accepting results from aliquots with increasingly larger D<sub>c</sub> values. These data 1266 suggest that only those grains with  $D_c > 125$  Gy give accurate results (red data). Even with this 1267 minimum D<sub>c</sub>, ~3% of the accepted grains still give measured doses in saturation (black data). Sample 1268 from Covalejos cave (Northern Spain)<sup>295</sup> 1269

Figure 9: Opportunities for bleaching of the OSL signal in different environments. (a) Aeolian transport during dust storms (Pavliha-istockphoto.com). (b) fluvial transport (Marek Uliasz race.fit2paddle.com). (c) gravity-driven debris fans (unknown) (d) sediment transported by ice (Michael Hambrey - Glaciers online). (e) bioturbation - termite mounds (ourterritory.com).

Figure 10: Sources of complexity in estimations of dose and dose rate. (a) The likely complexities 1274 in D<sub>e</sub> determination arising from luminescence characteristics (dark blue band) and bleaching (red 1275 band), expressed qualitatively in terms of field observables and expected age (see [H1] "Reproducibility 1276 and data deposition" for details). These observables and estimates are shown on the 10 radial axes, 1277 grouped according to Depositional environment and implied transport mechanism (very light blue), 1278 Sedimentology (light blue) and the expected Burial age of the samples (medium blue). "Young" refers 1279 to quartz grains with expected De values of less than ~10 Gy, "Old" to quartz grains with De values of 1280 larger than ~130 Gy. Each axis is scaled from most complex (centre of the figure) to least complex 1281 (outer edge of figure). The likely variability of each feature is shown by the band width on a particular 1282

axis. There is no reason to expect a correlation between luminescence characteristics and 1283 sorting/stratification, and so there is no dark blue band in that portion of the figure. (b) Bar chart 1284 showing qualitatively the relative influences of different sedimentary characteristics on dose rate 1285 complexity. Each bar depicts an observable sedimentary characteristic (y-axis) that can be evaluated at 1286 the time of OSL sample collection or during sample preparation (white labels – present at the time of 1287 deposition; black labels - post-depositional alteration). Each sedimentary characteristic varies from low 1288 to high (x-axis), and the gradational colouring indicates the degree of beta,  $\gamma$  and cosmic dose rate 1289 complexity (in both space and time) expected for a given category value. 1290

**Figure 11:** Accuracy and precision in quartz OSL dating. (a) Large aliquot 63-90 um quartz OSL ages plotted as a function of depth for a section of Chinese loess encompassing last glacial loess and Holocene soil. Uncertainties are random only. A Bayesian model (with 1 $\sigma$  error band) is also shown illustrating how higher precision can be achieved. Ages excluded from modelling are from samples deliberately taken within **crotovina**. Figure modified from SI in<sup>95</sup>. (b) Comparison of OSL ages determined using quartz from well-bleached samples with independent age control (n = number of samples, followed by the % of samples within  $\pm 2\sigma$  of the line of unit slope)<sup>89, 108, 269</sup>.

#### 1298 Boxes

#### [bH1] Box 1: Water content estimation

Constraining the average water content of a deposit is important to the accuracy of a luminescence age, 1300 because a 1% increase/decrease in lifetime average water content typically leads to a 1% 1301 increase/decrease in derived age<sup>a</sup>. Well-sorted sands have a small variation in porosity, from fully 1302 (~38%) to loosely (46%) compacted<sup>296</sup>, and because the grain surfaces are in physical contact (in 1303 contrast to for example, clay-rich deposits) it is very difficult to over-compress sand by increased 1304 overburden pressure. When fully saturated with water, 42% porosity corresponds to 21% by weight 1305 water content. A saturation water content can be readily estimated using for example, the syringe 1306 method<sup>b</sup> because this packing density is reproducible, in a sandy deposit. If the sediment was below the 1307 ground-water table throughout the burial period, this is the lifetime average water content. Otherwise, 1308 the fraction of saturation during the burial period must be estimated from an understanding of the local 1309 hydrology (drainage, precipitation, etc.) and sediment characteristics<sup>297</sup>. The observed water content, 1310 such as the water content in a field sample measured by drying a known weight of wet material, can 1311 often be taken as a lower limit to lifetime water content because typical sampling sites (cleaned sections, 1312 gully walls, river or road cuts etc.) are usually significantly drier than corresponding material a few 1313 metres deeper into the section. In the worst case, where no further constraints can be placed on water 1314 content history, the mid-point between field and saturated water content can be chosen, and an estimate 1315 of uncertainty adopted, equal to 25% of the range. Then all possible values are covered by  $\pm 2$  times the 1316 uncertainty. For example, a measured water content of, say, 9% (cf. saturation water content of 21%) 1317 would give a mean lifetime water content of 15±3% by weight, implying an overall contribution to the 1318 uncertainty on the age of  $\sim 3\%^{298}$ . This is not large compared to the expected minimum uncertainty of 1319  $\sim$ 5% (uncertainties added in quadrature, see **Table 7**), and so is not of major concern. Similar arguments 1320 can be used to constrain the effects of the infilling of porosity by carbonate. 1321

1322

The water content of a ceramic buried in a sediment is more complicated. The porosity of a ceramic will differ from that of the sediment matrix, but it is readily measured by simply saturating the ceramic in water, weighing, drying and weighing again. Then it is assumed that the fraction of saturation that applies to the sediment matrix also applies to the ceramic<sup>17</sup>. The lifetime average water content estimated for the matrix is applied to the  $\gamma$  dose rate, and that for the ceramic to the beta dose rate.

1328

defined, and increasing the pressure does not extrude more water. The syringe and wet sand are weighed to obtain the

sample during drying to the dry weight is the saturation water content.

<sup>&</sup>lt;sup>1329</sup> <sup>a</sup>The age calculation spreadsheet attached to this paper allows the testing of the sensitivity of the ages to various parameters.

<sup>&</sup>lt;sup>1330</sup> <sup>b</sup>A saturation water content can be measured for sand by adding sediment and excess water to say, a pre-weighed 100 ml

syringe and applying pressure to remove the excess water. The point at which the syringe plunger stops moving is well-

saturated weight, and then placed in an oven and dried to obtain the dry weight: the ratio of the weight loss of the saturated

### [bH1] Box 2: The Single Aliquot Regenerative (SAR) dose procedure

Assuming first order filling of an electron trap during irradiation, the subsequent luminescence signal can then be expressed as (based on<sup>63</sup>):

1339

1347

where L is the number of luminescence photons, S and  $\chi$  describe the dependence of luminescence sensitivity (here photons per recombining electron) on measurement number, *j*, and dose, *D*, respectively,  $N_0$  is an upper limit to the number of trapped electrons (determined by the trap density), and *b* is a constant describing the rate of filling of the electron trap per unit dose. If the OSL trap is fully emptied during measurement, then T(D<sub>T</sub>), the response to a subsequent test dose  $D_T$  can be written as:

1345 
$$T(D_T) = S_{j+1} \cdot \chi(D_T) \cdot N_0 \cdot (1 - e^{-b \cdot D_T})$$

1346 Thus, if  $D_T$  is now fixed

where  $K = 1/(\chi(D_T).(1 - e^{-b.D_T}))$  is a constant. Equation 5 represents the luminescence response to dosing and measurement in any one SAR cycle. If the ratio  $S_j/S_{j+1}$  is constant, the sensitivity-corrected luminescence signal (*L*/*T*) is independent of SAR cycle, and for any given dose, *D*, takes on a unique value. This result has been confirmed by numerical simulations using a more complex model that takes into account competing electron trapping and recombination processes<sup>81</sup>, and by experiment (**Fig. 5a**).

In practice, the sensitivity-corrected luminescence from the 'i'th SAR cycle is usually identified as L<sub>i</sub>/T<sub>i</sub>. 1353 The first SAR cycle is denoted by i=0 because it is different from all later cycles, in that the dose,  $D_0$ , 1354 was absorbed before any subsequent laboratory pretreatment such as preheating. If D<sub>0</sub> was absorbed in 1355 nature, then it is often referred to as  $D_n$  (the measured value of  $D_n$  is  $D_e$ ). In all later cycles (i=1, k), the 1356 aliquot has been preheated and light exposed before absorbing a dose D<sub>i</sub>. Note also that this formulation 1357 assumes that the probability of trapping electrons in this system does not change with repeated dosing 1358 and emptying (b is independent of i and i), and does not change from natural irradiation to laboratory 1359 irradiation, assumptions fundamental to any added or regenerated dose procedure. 1360

Although it is the exponentially saturating population of the electron trap (Equation 4) that records the chronometric information, it is important to realise that the resulting sensitivity-corrected luminescence response curve (Equation 5) does not mirror the growth of the electron trap population. The luminescence response curve may not be a simple saturating exponential and thus may not plateau at high doses; it can continue to increase, or even decrease, with dose, all depending on the behaviour of  $\chi(D)$ .

### [bH3] Box 3: Testing for quartz purity

SAR dose measurements are most successful if the quartz OSL is dominated by the fast-component. 1369 One important step in ensuring this is to remove any other minerals likely to give luminescence. In practice, this means ensuring that all feldspar has been successfully separated in the sample preparation 1371 steps described above. Unfortunately, the chemical pretreatment intended to remove feldspar 1372 contamination is not always completely successful<sup>299, 300</sup>. The reasons for this are unclear, but 1373 presumably involve occluded feldspar grains, or feldspathic-like impurities within individual quartz 1374 grains. Feldspar impurities is a problem because feldspar is sensitive to the wavelengths used to 1375 stimulate quartz (usually 450 to 530 nm), and so can contaminate the quartz OSL signal. Luminescence 1376 from feldspar impurities is also significantly unstable, and much more difficult to bleach. If additional 1377 etching with HF does not remove residual signal contamination, then instrumental separation must be 1378 considered. For stimulation with blue or green wavelengths, the decay rate of the feldspar signal is 1379 considerably slower than that of the quartz fast-component, and so the use of the early background 1380 subtraction approach reduces the importance of any feldspar contamination. But feldspar, in contrast to 1381 quartz<sup>59</sup>, is sensitive to infrared (IR) stimulation. This characteristic can be used to test for, and to 1382 reduce, contamination of the quartz OSL by feldspar OSL; this is done by inserting an IR stimulation 1383 immediately prior to the measurement of  $L_i$  (but not  $T_i$ ) in an additional recycling ratio test<sup>93</sup>. If the 1384 resulting  $(L_i/T_i)/(L_l/T_l)$  recycling ratio (often referred to as the OSL IR depletion ratio in the literature) 1385 is indistinguishable from unity, then feldspar OSL does not contribute significantly to the quartz signal. 1386 In samples where the feldspar contamination remains of significance even after additional etching with 1387 HF, it can be reduced by inserting the IR stimulation step in front of every L and T measurement<sup>301</sup>, 1388 although such a 'double SAR' approach is expensive in measurement time. There are also other 1389 instrumental methods for minimising any feldspar contribution, but these are not in routine use<sup>40, 302</sup>. 1390

# 1 Tables

- **Table 1** Synthesis of different dating applications and events that can be constrained using quartz OSL. Columns 4-5 indicate the general
- suitability of OSL dating in different geological settings and key practical consideration that can influence OSL dating reliability. (Suitability
- <sup>4</sup> classification: O = Optimal; R = routine with appropriate methodological considerations; S = sub-optimal; C = challenging; N = not
- <sup>5</sup> recommended; Nv = novel or not widely used)

~	
6	
~	

Application type	Event being dated	Signal resetting agent	Suitability and age interpretation	Practical considerations
Aeolian (wind- transported)	Formation of desert and coastal dunes <sup>83, 146, 161, 261, 303-311</sup>	Daylight	O; direct dating	Usually well-reset by sub-aerial exposure. Dune fields reflect transitional climates from arid to humid to allow sand movement and subsequent trapping. OSL dates last movement, and can be used to derive dune aggradation and migration rates and desert albedo changes. Bioturbation and pedogenesis processes can affect low-cohesion, near- surface dune deposits. Significant beta-dose heterogeneity possible in low radioactivity dune deposits and/or coarse grained sand dunes. Does not significantly affect average behaviour. Dating of gypsum dunes by targeting embedded quartz grains.
	Loess accumulation <sup>95, 99, 104, 148, 312-321</sup>	Daylight	O; direct dating	Fine-grain deposits with limited sand-sized quartz. Coarse silt can be used, but feldspar contamination may be difficult to remove.

Fluvial, alluvial, beach (high-energy water transport)	Terrace deposition and abandonment <sup>106, 269, 322-324</sup>	Daylight	R-C; direct dating of fluvial deposition; indirect dating of floodplain abandonment	n all cases, both sub-aerial and sub-aqueous daylight exposure likely		
	Channel migration <sup>325, 326</sup>	Daylight	R-C; direct dating of channel deposition; indirect dating of channel erosion	for fluvial sediment transported downstream through many reworking cycles, with intervening periods of direct daylight exposure during low flow. Thus well-sorted distinct sand lenses/layers derived from low energy, shallow water environments are most likely to be well- bleached; they also provide a simplified dosimetry.		
	Channel cutting and filling; floodplain aggradation and alluvial fan formation <sup>327</sup>	Daylight	R-C; direct dating of filling events, indirect dating of cutting or incision events			
	Palaeoflood deposits <sup>211, 212, 328</sup>	Daylight	R-C: direct dating of flood events	Bleaching during flood unlikely, but prior bleaching of sediments before flood entrainment seems common.		
	Beach formation associated with sea level changes <sup>217, 293, 329-331</sup>	Daylight	O-C; direct dating	Usually well reset by sub-aerial and sub-aqueous bleaching. High carbonate contents common in low-latitude beach deposits, may lead to complexity (time-dependent dose rates) if carbonate content changes with time.		
	Tsunami deposits <sup>332-334</sup>	Daylight	S-N; direct dating	Bleaching unlikely during event but sediments entrained in shallow water likely already well-bleached.		
Lacustrine and offshore-marine	Lake level variations <sup>335-340</sup>	Daylight	S-C; direct dating of lake full phases (sedimentation);	Sampling by coring usually precludes <i>in situ</i> $\gamma$ dose rate measurement. Organic-rich, low-energy lakes may have limited coarse-grain quartz yields. Time-dependent dose rates can arise from both U-series		

(low-energy water transport)			indirect dating of lake drying phases (non- deposition)	disequilibria and sediment compaction/dewatering associated with accumulation of overburden (especially true for gyttja).
	Formation of lake-margin deposits (deltas, lake shoreline features) <sup>143, 151, 152, 341-343</sup>	Daylight	R; direct dating	Used to date lake high stands. Marginal deposits may be more accessible and suitable than lake bed deposits (in terms of grain size, bleaching history). Sub-aerial and sub-aqueous bleaching likely for beach deposits.
	Ocean floor sedimentation <sup>5, 13, 14, 144, 344, 345</sup>	Daylight	R-C; direct dating	Bleaching sub-aerial (dust) or sub-aqueous (fluvial input). Time- dependent dose rates due to U-series disequilibria may increase complexity, especially in the presence of high carbonate content. High water content reduces dose rate and extends age range. Usually very low coarse-grained quartz content.
Glacigenic (ice- related)	Moraine / till formation <sup>157, 346-348</sup>	Daylight	R–C; direct dating of ice margin	For all glacial sediments, low OSL signal sensitivity is common in primary ice-eroded material. Limited/no daylight exposure possible. If sediment unexposed to daylight during the last transport, the OSL age will relate to an earlier transport event, but dose rate may have changed.
	Proglacial outwash plain activation <sup>157, 159, 349, 350</sup>	Daylight	R; direct dating of outwash plain; indirect dating of ice margin	Transport distance from ice-margin important for bleaching.

	Glaciofluvial channel activation <sup>150, 351, 352</sup>	Daylight	R; direct dating of channel activation; indirect dating of ice margin	Event-related activation may lead to limited daylight exposure.
	Glaciolacustrine sedimentation <sup>353</sup>	Daylight	R–S; direct dating of proglacial lake sedimentation.	Sediment deposited in ice-marginal turbid lakes may experience little/no daylight exposure; OSL age may therefore reflect previous transport events.
	Glacioaeolian deposition <sup>163, 209,</sup> 354-356	Daylight	O; indirect dating of (peri)-glacial processes	See "sand dune formation" and "loess accumulation" above.
Colluvium, solifluction sheets, debris flows (gravitational mass movements)	Slope failures, formation and progradation <sup>357-360</sup>	Daylight	SR-C; direct dating	Limited transport distances and/or daylight exposure durations can increase the likelihood for heterogeneous bleaching. Poorly sorted deposits may suffer from spatially heterogeneous radiation fields.
Archaeology, palaeontology, palaeoanthropology (including karst deposits)	Site depositional history <sup>97, 105-107, 193, 194, 198, 202, 361-363</sup>	Daylight	O-R; direct dating	Covers a wide-range of depositional contexts (see above for details). Open-air archaeological sites have better bleaching conditions and fewer post-depositional complexities than caves and rock shelters. Complexity from syn-depositional mixing and contamination by older generations of pre-existing deposits may arise in cave settings.
	Internment of autochthonous fossils / cultural materials in sediments <sup>201, 207, 208, 210, 364</sup>	Daylight	O-R; indirect dating but close association with dating event	Multiple cycles of human occupation may cause post-depositional disturbance (digging, burials, fire pits).

	Internment of allochthonous fossils / cultural materials in sediments <sup>230, 365</sup>	Daylight	O-R; indirect dating and poor association with dating event	OSL provides minimum age for fossils and archaeological material. Animal disturbance of host sediments is possible in dens and pitfall traps.
	<i>In situ</i> production of cultural materials, including debitage production <sup>97, 135, 136, 366</sup>	Heat (lithics, fireplaces) Daylight	R, C, Nv; direct dating	Stones (fireplace, pot-boilers) usually straightforward if heated sufficiently to fully reset. Heated lithics with no stratigraphic context (museum specimens) will have poorly constrained $\gamma$ dose rates.
	Heating of pottery, bricks, tiles. Manufacturing of mortars <sup>131, 132,</sup> <sup>367-370</sup>	Heat, Daylight (mortars)	O; direct	Straightforward but extraction of sufficient coarse quartz may not always be possible in pottery. Surrounding sediment/brick should be collected for $\gamma$ dose rate estimation
Soil and regolith processes	Formation of soil A horizons <sup>125,</sup> 169, 172	Daylight	C, Nv; direct dating	Bleaching by turbation ongoing until soil surface buried. Age usually reflects time of burial. Old soils well-bleached, young soils usually incompletely bleached. Scatter in D <sub>e</sub> can be interpreted in terms of turbation rates.
	Sediment mixing between layers <sup>170, 240, 371</sup>	Daylight	C-N; direct dating	Single-grain studies may provide insight into grain movement histories. Reliable dating not usually possible, but identification of mixing may be possible.
	Soil creep rates <sup>168, 372</sup>	Daylight	C, Nv; direct dating	Reliable interpretation of apparent ages challenging.
	Formation of pedogenic and biogenic structures (carbonate nodules, termite mounds, root casts) <sup>171, 173</sup>	Daylight	C-N, Nv; direct dating	Careful sampling important to isolate individual formation events. Problems of interpretation similar to those for soil creep rates. Carbonate-rich structures may involve time-dependent dose rates.

		Daylight	R-C; indirect dating	Sediments associated with palaeo-earthquakes often experienced
Neo-tectonics and	Occurrence of palaeo-	before/after	of seismic event itself	limited transport (and so daylight exposure), particularly colluvium,
volcanic histories	earthquakes <sup>164-167, 373-375</sup>	seismic	but close stratigraphic	debris flows or locally intruded deposits. Targeting stable palaeo-
		events	association	surfaces can improve dating reliability.
	Offset of geomorphic surfaces due to fault movement <sup>376-381</sup>	Daylight before and after faulting event	R-C; indirect dating of faulting event itself but close stratigraphic association	Limited transport distances and daylight exposure if sampling fluvial, colluvium, debris flows or locally intruded deposits. Rock surface dating may be applicable.
	Dating fault gouge and dike intrusions <sup>138</sup>	Frictional heating	R-C; direct dating of faulting or seismic event	Sand-sized quartz grains may not be present in gouge. Gouge may be very thin, making sampling challenging
	Volcanic eruptions – dating proximaldeposits <sup>382-384</sup>	Heating by magma	R-C; indirect dating of volcanic event itself but close stratigraphic association	Quartz from volcanic sources usually insensitive, weak fast- component. But quartz from contact-heated sediments likely to have improved OSL sensitivity.
	Volcanic eruptions – dating distal deposits <sup>104, 351, 354, 364, 385, 386</sup>	Daylight	R-C; indirect dating of volcanic event itself but close stratigraphic association	Sampling above and below discrete tephra horizons constrains ash fall events. Sampling of deposits that host cryptotephra is also possible.
Burial dating of rock surfaces	Construction of megaliths, cairns, stone walls, etc. Dating erratics,	Daylight	R-C, Nv; direct dating	Only likely applicable to sandstones and some quartzite clasts because of poor quartz OSL sensitivity in primary rocks (e.g. granites).

-	scree slopes, rockfalls, cobble			
	beaches and beach ridges <sup>187, 188,</sup>			
	284			
Exposure dating of rock surfaces	Construction of megaliths, cairns, stone walls, etc. Rock art dating. Dating erratics, scree slopes, rockfalls, cobble beaches and beach ridges <sup>15, 187, 287</sup>	Daylight	C, Nv; direct dating	Known-age exposure samples needed for calibration. Only likely applicable to sandstones and some quartzite clasts because of poor quartz OSL sensitivity in primary rocks (e.g. granites).
Thermochronometry	Bedrock exhumation <sup>387, 388</sup>	Cooling below closure temperatures of 30-40°C	C, Nv; direct dating	Signal saturation restricts applications to rapidly exhumed settings or elevated temperature settings. Combination of quartz OSL with other signals (TL, IRSL, post-IR IRSL) may offer more versatility. Only likely applicable to sandstones and some quartzite clasts because of poor quartz OSL sensitivity in primary rocks (e.g. granites).
Palaeo-lightning events	Fulgurite formation <sup>141</sup>	Heating of trapped quartz grains	O-R, Nv; direct dating	Target quartz grains occluded in the fused matrix.

9 **Table 2.** Example tables containing suggested minimum sample-specific information that should be presented in a publication containing quartz OSL

10 luminescence ages. All uncertainties expressed at 1σ confidence level and contain both random and systematic components. More general characteristics

(especially dose recovery) are also required to assess reliability, and can be summarised in associated text.

12

## 13 a)

Lab.code	<sup>238</sup> U, Bq.kg <sup>-1</sup>	<sup>226</sup> Ra, Bq.kg <sup>-1</sup>	<sup>232</sup> Th, Bq.kg <sup>-1</sup>	<sup>40</sup> K, Bq.kg <sup>-1</sup>	*beta dose rate, Gy.ka <sup>-1</sup>	* γ dose rate, Gy.ka <sup>-1</sup>
1	$51 \pm 12$	$46.0~\pm~1.2$	$61.0 \pm 1.2$	$562 \pm 20$	$2.29 ~\pm~ 0.06$	$1.48 \pm 0.04$
2	$72 \pm 12$	$48.2 ~\pm~ 1.1$	$60.6 ~\pm~ 1.2$	$566 \pm 16$	$2.32 ~\pm~ 0.05$	$1.50 \pm 0.04$
3	$58 \pm 9$	$45.2 ~\pm~ 0.9$	$60.2 ~\pm~ 1.0$	$581 \pm 13$	$2.33 ~\pm~ 0.05$	$1.48 \pm 0.04$
4	$46~\pm~8$	$43.8~\pm~0.8$	$32.2 ~\pm~ 0.8$	$158\pm8$	$1.05 ~\pm~ 0.04$	$0.81 \ \pm \ 0.04$

Note: \* dry infinite matrix dose rates

15 b)

14

Lab.code	Site	Depth, cm	w.c. %	OSL D <sub>e</sub> , Gy	(n <sub>r</sub> )	(n <sub>a</sub> )	Total dose rate, Gy/ka	OSL age, ka
1	Test site-A	65	16	$95 \pm 4$	1	23	$3.16 ~\pm~ 0.16$	$30.0~\pm~2.0$
2	Test site-B	65	16	$73 \pm 4$	0	12	$3.19 ~\pm~ 0.16$	$22.9 ~\pm~ 1.7$
3	Test site-C	65	16	$107 \pm 6$	0	24	$3.20 \pm 0.16$	$33.5~\pm~2.5$
4	Test site-C	30	11	$43 \pm 2$	1	35	$1.79 ~\pm~ 0.09$	$23.9~\pm~1.7$

Note: (a) Mean grain size 215µm. (b) Site altitude 0 km. (c) w.c.: water content. (d) n<sub>r</sub>, n<sub>a</sub> number of rejected and accepted aliquots, respectively.

# 1 References

- Noller, J.S.S., J.M.: Lettis, W.R.; Quaternary Geochronology: Methods and Applications
   (American Geophysical Union, 2013).
- Wintle, A. & Huntley, D. Thermoluminescence dating of a deep-sea sediment core. *Nature* 279, 710-712 (1979).
- Huntley, D.J., Godfrey-Smith, D.I. & Thewalt, M.L.W. Optical dating of sediments. *Nature* 313, 105-107 (1985).
- Qiu, F.Y. & Zhou, L.P. A new luminescence chronology for the Mangshan loess-palaeosol sequence on the southern bank of the Yellow River in Henan, central China. *Quaternary Geochronology* 30, 24-33 (2015).
- 5. Sugisaki, S. et al. High resolution optically stimulated luminescence dating of a sediment core from the southwestern Sea of Okhotsk. *Geochemistry, Geophysics, Geosystems* **13** (2012).
- Buylaert, J.-P. et al. A robust feldspar luminescence dating method for Middle and Late
  Pleistocene sediments. *Boreas* 41, 435-451 (2012).
- Jain, M. Extending the dose range: Probing deep traps in quartz with 3.06 eV photons.
   *Radiation Measurements* 44, 445–452 (2009).
- Hütt, G., Jaek, I. & Tchonka, J. Optical dating: K-feldspars optical response stimulation
   spectra. *Quaternary Science Reviews* 7, 381–385 (1988).
- Sohbati, R., Murray, A.S., Lindvold, L., Buylaert, J.-P. & Jain, M. Optimization of laboratory
   illumination in optical dating. *Quaternary Geochronology* 39, 105-111 (2017).
- Nelson, M.S.G., H.J.; Johnson, J.A.; Rittenour, T.M.; Feathers, J.K.; Mahan, S.A. User guide
   for luminescence sampling in archaeological and geological contexts. *Advances in Archaeological Practice* 3, 166-177 (2015).
- Rajapara, H.M., Kumar, V., Chauhan, N., Gajjar, P.N. & Singhvi, A.K. Bleaching of blue
   light stimulated luminescence of quartz by moonlight. *Journal of Earth System Science* 129, 1-7 (2020).
- Chen, Y.W.C., Y.G.; Murray A.S.;Liu, T.K.;Lai, T.C.;. Luminescence dating of neotectonic
   activity on the southwestern coastal plain, Taiwan. *Quaternary Science Reviews* 22, 1223 1229 (2003).
- Sugisaki, S. et al. High resolution OSL dating back to MIS 5e in the central Sea of Okhotsk.
   *Quaternary Geochronology* 5, 293-298 (2010).
- Armitage, S.J. & Pinder, R.C. Testing the applicability of optically stimulated luminescence
   dating to Ocean Drilling Program cores. *Quaternary Geochronology* **39**, 124-130 (2017).
- Gliganic, L.A., Meyer, M.C., Sohbati, R., Jain, M. & Barret, S. OSL surface exposure dating
   of a lithic quarry in Tibet: Laboratory validation and application. *Quaternary Geochronology* 49, 199-204 (2019).
- Prescott, J.R. & Hutton, J.T. Cosmic ray contributions to dose rates for luminescence and
   ESR dating: large depths and long-term time variations. *Radiation Measurements* 23, 497–
   500 (1994).
- 17. Aitken, M.J. Thermoluminescence Dating (Academic Press, London, 1985).
- 18. Li, B.J., Z.; Roberts, R. G.; Li, S. . Review and assessment of the potential of post-IR IRSL
   dating methods to circumvent the problem of anomalous fading in feldspar luminescence.
   *Geochronometria* 41, 178-201 (2014).
- Porat, N., Faerstein, G., Medialdea, A. & Murray, A.S. Re-examination of common extraction and purification methods of quartz and. *Ancient TL* 33, 22-30 (2015).
- 46 20. Chu, S.Y.F., Ekström, L.P. & Firestone, R.B. <u>http://nucleardata.nuclear.lu.se/toi/</u> (1999).
- Ivanovich, M. & Harmon, R.S. Uranium-series disequilibrium: applications to earth, marine,
   and environmental sciences. 2. ed. (Clarendon Press, Oxford, 1992).
- 49 22. Roesch, W.C. & Attix, F.H. in Attix, F.H., Roesch, W.C. (Eds.). Radiation Dosimetry.
  50 Volume I: Fundamentals (Academic Press, New York and London, 1968).
- Bourdon, B., Henderson, G.M., Lundstrom, C.C. & Turner, S.P. (eds) Uranium-series
   Geochemistry, pp. 656 (Mineralogical Society of America, Washington, D.C., 2003).

53	24.	Bøtter-Jensen, L., Agersnap Larsen, N., Markey, B.G. & McKeever, S.W.S. Al2O3:C as a
54		sensitive OSL dosemeter for rapid assessment of environmental photon dose rates. Radiation
55		Measurements 27, 295-298 (1997).
56	25.	Richter, D., Dombrowski, H., Neumaier, S., Guibert, P. & Zink, A.C. Environmental gamma
57		dosimetry with OSL of $\alpha$ -Al2O3: C for in situ sediment measurements. <i>Radiation protection</i>
58		dosimetry, <b>141</b> , 27-35 (2010).
59	26.	Kreutzer, S. et al. Environmental dose rate determination using a passive dosimeter:
60		techniques and workflow for α-Al2O3: C chips. <i>Geochronometria</i> <b>45</b> , 56-67 (2019).
61	27.	Miallier, D., Guérin, G., Mercier, N., Pillevre, T. & Sanzelle, S. The Clermont radiometric
62		reference rocks: a convenient tool for dosimetric purposes. Ancient TL <b>20</b> , 37-43 (2009).
63	28	Arnold L.I. Duval M. Falguères C. Bahain I-I & Demuro M. Portable gamma
64	20.	spectrometry with cerium-doped lanthanum bromide scintillators. Suitability assessments for
65		luminescence and electron spin resonance dating applications Radiation Measurements 6-18
66		47 (2012)
67	20	47 (2012). Layborg L & Kirkegaard P Besponse of 3" x 3" NaI(TI) detectors to terrestrial gamma
07	29.	rediction Nuclear Instruments and Methods <b>121</b> , 230, 251 (1074)
00	20	Guárin G. & Margier N. Determining gamma dosa rates by field gamma spectroscopy in
09	50.	outini, O. & Mercier, N. Determining gamma dose rates by field gamma spectroscopy in
70		sedimentary media. results of Monte Carlo simulations. <i>Kautation Measurements</i> , <b>40</b> , 190-
/1	21	195 (2011). Second N.A. The entired detine signal form month $\mathbf{P}_{i}$ is $M_{i} = (22, 502, 600)$
72	31.	Spooner, N.A. The optical dating signal from quartz. <i>Radiation Measurements</i> 23, 593-600
73	22	
74	32.	Bøtter-Jensen, L., Thomsen, K.J. & Jain, M. Review of optically stimulated luminescence
75		(OSL) instrumental developments for retrospective dosimetry. Rad. Meas 45, 253–257
76		
77	33.	Clark-Balzan, L. & Schwenninger, J. First steps toward spatially resolved OSL dating with
78		electron multiplying charge-coupled devices (EMCCDs): system design and image analysis.
79		Radiation Measurements 47, 797–802 (2012).
80	34.	Richter, D., Richter, A. & Dornich, K. Lexsyg – a new instrument for luminescence research.
81		Geochronometria <b>40</b> , 220–228 (2013).
82	35.	Thomsen, K.J., Kook, M., Murray, A.S., Jain, M. & Lapp, T. Single-grain results from an
83		EMCCD-based imaging system. Radiation Measurements 81, 185–191 (2015).
84	36.	Bøtter-Jensen, L. & Duller, G.A.T. A new system for measuring OSL from quartz samples.
85		Nucl. Tracks. Radiat. Meas 20, 549–553 (1992).
86	37.	Bulur, E., Bøtter-Jensen, L. & Murray, A.S. Optically stimulated luminescence from quartz
87		measured using linear modulation technique. Radiation Measurements 32, 407-411 (2000).
88	38.	Huntley, D.J., Short, M.A. & Dunphy, K. Deep traps in quartz and their use for optical dating.
89		<i>Can. J. Phys</i> <b>74</b> , 81–91 (1996).
90	39.	Bulur, E. An alternative technique for optically stimulated luminescence (OSL) experiment.
91		<i>Radiat. Meas</i> <b>26</b> , 701–709 (1996).
92	40.	Thomsen, K.J. et al. Minimizing feldspar OSL contamination in quartz UV-OSL using pulsed
93		blue stimulation. Radiation Measurements 43, 752–757 (2008).
94	41.	Ankjærgaard, C. & Jain, M. Optically stimulated phosphorescence in quartz over the
95		millisecond to second time scale: insights into the role of shallow traps in delaying
96		luminescent recombination. Journal of Physics D: Applied Physics 43, 255502 (2010).
97	42.	Huntley, D.J., Godfrey-Smith, D.I. & Thewalt, M.L.W. Optically dating of sediments, <i>Nature</i>
98		<b>313</b> . 105–107 (1985).
99	43.	Duller, G.A.T., Bøtter-Jensen, L., Murray, A.S. & Truscott, A.J. Single grain laser
100		luminescence (SGLL) measurement using a novel automated reader Nuclear Instruments and
101		Methods B 155, 506–514 (1999).
102	44	Bøtter-Jensen L. Bulur E. Duller GAT & Murray AS Advances in luminescence
103		instrument systems Radiation Measurements <b>37</b> , 573–578 (2000)
10/	45	Bøtter-Jensen I Duller G A T Murray A S & Baneriee D Blue light emitting diodes for
105	-IJ.	ontical stimulation of quartz in retrospective dosimetry and dating <i>Radiation Protection</i>
106		Dosimetry 84 335_340 (1999)
100		$\mathcal{L}_{OSUMCHY} \mathbf{J}_{\mathbf{J}}, \mathcal{J}_{\mathbf{J}}, \mathcal{J}, $

46. Duller, G.A.T., Bøtter-Jensen, L. & Murray, A.S. Combining infrared- and green-laser 107 stimulation sources in single-grain luminescence measurements of feldspar and quartz. 108 Radiation Measurements 37, 543–550 (2003). 109 47. Hansen, V., Murray, A., Buylaert, J.P., Yeo, E.Y. & Thomsen, K. A new irradiated quartz for 110 beta source calibration. Radiation Measurements 81, 123-127 (2015). 111 Thomsen, K.J., Murray, A.S. & Bøtter-Jensen, L. Sources of variability in OSL dose 48. 112 measurements using single grains of quartz. Radiation Measurements **39**, 47–61 (2005). 113 49. Ankjærgaard, C. & Murray, A.S. Total beta and gamma dose rates in trapped charge dating 114 115 based on beta counting. Radiation Measurements 42, 352-359 (2007). 50. Vandenberghe, D., De Corte, F., Buylaert, J.-P. & Kučera, J. On the internal radioactivity in 116 quartz. Radiation Measurements 43, 771-775 (2008). 117 51. Guérin, G., Mercier, N. & Adamiec, G. Dose-rate conversion factors: update. Ancient TL 29, 118 5-8 (2011). 119 Guérin, G., Mercier, N., Nathan, R., Adamiec, G. & Lefrais, Y. On the use of the infinite 52. 120 matrix assumption and associated concepts: a critical review. Radiation Measurements 47, 121 778-785 (2012). 122 Guérin, G., Jain, M., Thomsen, K.J., Murray, A.S. & Mercier, N. Modelling dose rate to 53. 123 single grains of quartz in well-sorted sand samples: The dispersion arising from the presence 124 of potassium feldspars and implications for single grain OSL dating. Quaternary 125 Geochronology 27, 52-65 (2015). 126 Mayya, R.M., Morthekai, P., Murari, M.K. & Singhvi, A.K. Towards quantifying beta 54. 127 microdosimetric effects in single grain quartz dose distribution. Radiation Measurements 41, 128 1032-1039 (2006). 129 Guibert, P., Lahaye, C. & Bechtel, F. The importance of U-series disequilibrium of sediments 55. 130 in luminescence dating: a case study at the Roc de Marsal cave (Dordogne, France). 131 Radiation Measurements 44, 223-231 (2009). 132 Jain, M., Choi, J.H. & Thomas, P.J. The ultrafast OSL component in quartz: Origins and 133 56. implications. Radiation Measurements 43, 709-714 (2008). 134 57. Wintle, A.G. & Murray, A.S. The relationship between quartz TL, PTTL and OSL. Radiation 135 Measurements 27, 611–624 (1997). 136 Jain, M., Murray, A.S., Bøtter-Jensen, L. & Wintle, A.G. A single-aliquot regenerative-dose 58. 137 method based on IR (1.49 eV) bleaching of the fast OSL component in quartz. Radiation 138 Measurements 39, 309-318 (2005). 59. Jain, M., Murray, A.S. & Bøtter-Jensen, L. Characterisation of blue-light stimulated 140 luminescence components in different quartz samples: implications for dose measurement. 141 Radiation Measurements 37, 441–449 (2003). 142 60. Murray, A.S. & Wintle, A.G. Isothermal decay of optically stimulated luminescence in 143 quartz. Radiation Measurements 30, 119–125 (1999). 144 Bøtter-Jensen, L. et al. Luminescence sensitivity changes in quartz as a result of annealing. 61. 145 Radiation Measurements 24, 535-541 (1995). 146 Wintle, A.G. & Murray, A.S. Quartz OSL: effects of thermal treatment and their relevance to 62. 147 laboratory dating procedures. Radiation Measurements 32, 387-400 (2000). 148 Moska, P. & Murray, A.S. Stability of the quartz fast-component in insensitive samples. 63. 149 Radiation Measurements 41, 878-885 (2006). 150 Buylaert, J.P. et al. Luminescence dating of old (>70 ka) Chinese loess: A comparison of 64. 151 single-aliquot OSL and IRSL techniques. *Quaternary Geochronology* **2**, 9-14 (2007). 152 Chapot, M., Roberts, H.M., Duller, G.A. & Lai, Z.P. A comparison of natural- and laboratory-65. 153 generated dose response curves for quartz optically stimulated luminescence signals from 154 Chinese Loess. Radiation Measurements 47, 1045-1052 (2012). 155 Lowick, S.E., Preusser, F. & Wintle, A.G. Investigating quartz optically stimulated 66. 156 luminescence dose-response curves at high doses. Radiation Measurements 45, 975-984 157 (2010). 158 Ellerton, D. et al. An 800 kyr record of dune emplacement in relationship to high sea level. 67. 159 Geomorphology 354, 106999 (2020). 160

161	68.	Pawley, S.M., Bailey, R.M., Rose, J., Moorlock, B.S.P. & Hamblin, R.J.O. Age limits on
162		Middle Pleistocene glacial sediments from OSL dating, north Norfolk, UK. Quaternary
163		Science reviews 27, 1363-1377 (2008).
164	69.	Beerten, K.V., K. et al. Electron spin resonance (ESR), optically stimulated luminescence
165		(OSL) and terrestrial cosmogenic radionuclide (TCN) dating of quartz from a Plio-Pleistocene
166		sandy formation in the Campine area, NE Belgium. <i>Quaternary International</i> 556, 144-158
167		(2020).
168	70.	Madsen, A.T., Duller, G.A.T., Donnelly, J.P., Roberts, H.M. & Wintle, A.G. A chronology of
169		hurricane landfalls at Little Sippewisset Marsh, Massachusetts, USA, using optical dating.
170		<i>Geomorphology</i> <b>109</b> , 36-45 (2009).
171	71.	Durcan, J.A. & Duller, G.A.T. The fast ratio: A rapid measure for testing the dominance of
172		the fast component in the initial OSL signal from quartz. Radiation Measurements 46, 1965-
173		1072 (2011).
174	72.	Ankjærgaard, C., Jain, M., Hansen, P.C. & Nielsen, H.B. Towards multi-exponential analysis
175		in optically stimulated luminescence. Journal of Physics D: Applied Physics 43, 195501
176		(2010).
177	73.	Bailey, R.M., Yukihara, E.G. & McKeever, S.W.S. Separation of quartz optically stimulated
178		luminescence components using green (525 nm) stimulation. Radiation Measurements 46.
179		643-648 (2011).
180	74.	Ballarini, M., Wallinga, J., Wintle, A. G. & Bos, A. J. J. A modified SAR protocol for optical
181	,	dating of individual grains from young quartz samples. <i>Radiation Measurements</i> , 360-369
182		(2007).
183	75.	Cunningham, A. & Wallinga, J. Selection of integration time intervals for quartz OSL decay
184	101	curves. <i>Quaternary Geochronology</i> <b>5</b> , 657-666 (2010).
185	76.	Duller, G.A.T., Bøtter-Jensen, L. & Murray, A.S. Optical dating of single sand-sized grains of
186	/ 01	quartz: sources of variability <i>Radiation Measurements</i> <b>32</b> 453-457 (2000)
187	77	Guérin G et al. Absorbed dose, equivalent dose, measured dose rates, and implications for
188	,,,	OSL age estimates: Introducing the Average Dose Model. <i>Quaternary Geochronology</i> <b>41</b> .
189		163-173 (2017)
190	78	Murray A S & Wintle A G Luminescence dating of quartz using an improved single-
101	70.	aliquot regenerative-dose protocol <i>Radiation Measurements</i> <b>32</b> , 57–73 (2000)
192	79	Murray A S Roberts R G & Wintle A G Equivalent dose measurement using a single
193	12.	aliquot of quartz. Radiation Measurements 27, 171-184 (1997)
194	80	Huntley D I Note: some notes on language Ancient TL <b>19</b> 27-28 (2001)
195	81	Bailey R M Towards a general kinetic model for optically and thermally stimulated
196	01.	luminescence of quartz Radiation Measurements <b>33</b> 17-45 (2001)
197	82	Murray A S & Wintle A G The single aliquot regenerative dose protocol: potential for
198	02.	improvements in reliability <i>Radiation Measurements</i> <b>37</b> 377–381 (2003)
199	83	Madsen A T Murray A S & Andersen T I Ontical dating of dune ridges on Rømø a
200	05.	hadsen, Miri, Maray, More Pindersen, M.S. Optical during of durie Hages on Romp, a harrier island in the Wadden Sea Denmark <i>Journal of Coastal Research</i> 23, 1259–1269
200		(2007)
201	84	Godfrey-Smith DI Thermal effects in the optically stimulated luminescence of quartz and
202	04.	mixed feldspars from sediments <i>Journal of Physics D: applied Physics</i> <b>27</b> , 1737-1746
203		(1994)
204	85	Wintle $\Delta G M \Delta S$ Luminescence sensitivity changes in quartz Radiation Measurements
200	05.	<b>30</b> 107 118 (1999)
200	86	Smith BW & Rhodes E I Charge movements in quartz and their relevance to optical
207	80.	dating Radiation Measurements <b>73</b> 329-333 (1994)
200	87	Ankigragard C Murray A S & Denby PM Thermal pre-treatment (preheat) in the OSI
209	07.	dating of quartz: is it processary? <i>Radiation Protaction Dosimatry</i> <b>110</b> A70 A73 (2006)
210	88	Report E L Observations of thermal transfer OSL signals in classication and the matter and the second station
211	00.	Magsuraments <b>32</b> , 505, 602 (2000)
212	80	Madson & T. Murray & S. Anderson T.I. Dairun M. & Browning Madson U. Onticelly
213	07.	stimulated luminescence dating of young actuaring addimental a comparison with 210Db and
214		127Cs deting Marine Geology <b>214</b> , 251, 269 (2005)
215		13705 uaung. Murine Geology <b>214</b> , 231–208 (2003).

90. Murray, A.S. Developments in optically transferred luminescence and photo-transferred 216 thermoluminescence dating: application to a 2000-year sequence of flood deposits. 217 Geochimica et Cosmochimica Acta 60, 565–576 (1996). 218 91. Singh, A. et al. Counter-intuitive influence of Himalayan river morphodynamics on Indus 219 Civilisation urban settlements. Nature Communications 8, 1-14 (2017). 220 92. Guérin, G.C., B.; Lahaye, C.; Thomsen, K. J.; Tribolo, C.; Urbanova, P.; Guibert, P.; Mercier, 221 N.; Valladas, H.;. Testing the accuracy of a single grain OSL Bayesian central dose model 222 with known-age samples. Radiation Measurements 81, 62-70 (2015). 223 224 93. Duller, G.A.T. Distinguishing quartz and feldspar in single grain luminescence measurements. Radiation Measurements 37, 161-165 (2003). 225 Thomsen, K.J. et al. Testing single-grain quartz OSL methods using sediment samples with 94. 226 independent age control from the Bordes-Fitte rockshelter (Roches d'Abilly site, Central 227 France. Quaternary Geochronology 31, 77–96 (2016). 228 Stevens, T. et al. Ice-volume-forced erosion of the Chinese Loess Plateau global Quaternary 95. 229 stratotype site. Nature Communications 9, 983–983 (2018). 230 Vandenberghe, D. et al. Exploring the method of optical dating and comparison of optical and 96. 231 14C ages of Late Weichselian coversands in the southern Netherlands. Journal of Quaternary 232 Science 19, 73-86 (2004). 233 97. Guérin, G. et al. The complementarity of luminescence dating methods illustrated on the 234 Mousterian sequence of the Roc de Marsal: A series of reindeer-dominated, Quina 235 Mousterian Layers dated to MIS 3. Quaternary International 433, 102-115 (2017). 236 98. Nielsen, A., Murray, A.S., Pejrup, M. & Elberling, B. Optically stimulated luminescence 237 dating of a Holocene beach ridge plain in Northern Jutland, Denmark. Quaternary 238 Geochronology 1, 305-312 (2006). 239 Buylaert, J.P. et al. Optical dating of Chinese loess using sand-sized quartz: Establishing a 99. 240 time frame for Late Pleistocene climate changes in the western part of the Chinese Loess 241 Plateau. Quaternary Geochronology 3, 99-113 (2008). 242 100. Roberts, H.M. & Duller, G.A.T. Standardised growth curves for optical dating of sediment 243 using multiple-grain aliquots. Radiation Measurements 38, 241-252 (2004). 244 101. Duller, G.A.T. Improving the accuracy and precision of equivalent doses determined using 245 the optically stimulated luminescence signal from single grains of quartz. Radiation 246 Measurements 47, 770-777 (2012). 247 Jacobs, Z., Roberts, R.G., Nespoulet, R., El Hajraoui, M.A. & Debénath, A. Single-grain OSL 102. 248 chronologies for Middle Palaeolithic deposits at El Mnasra and El Harhoura 2, Morocco: 249 Implications forLate Pleistocene huma-environment interactions along the Atlantic coast of 250 northwest Africa. Journal of Human Evolution 62, 377-394 (2012). 251 103. Gliganic, L.A., Jacobs, Z. & Roberts, R.G. Luminescence characteristics and dose 252 distributions. Archaeological and Anthropological Science 4, 115-135 (2012). 253 Demuro, M., Arnold, L.E., Froese, D.G. & Roberts, R.G. OSL dating of loess deposits 104. 254 bracketing Sheep Creek tephra beds, northwest Canada: Dim and problematic single-grain 255 OSL characteristics and their effect on multi-grain age estimates. Quaternary Geochronology 256 15, 67-87 (2013). 257 105. Guérin, G. et al. A multi-method luminescence dating of the Palaeolithic sequence of La 258 Ferrassie based on new excavations adjacent to the La Ferrassie 1 and 2 skeletons. Journal of 259 Archaeological Science 58, 147-166 (2015). 260 Arnold, L.J. et al. OSL dating of individual quartz 'supergrains' from the Ancient Middle 106. 261 Palaeolithic site of Cuesta de la Bajada, Spain. Quaternary Geochronology 36, 78-101 262 (2016). 263 107. Demuro, M., Arnold, L.J., Aranburu, A., Gómez-Olivencia, A. & Arsuaga, J.L. Single-grain 264 OSL dating of the Middle Palaeolithic site of Galería de las Estatuas, Atapuerca (Burgos, 265 Spain). Quaternary Geochronology 49, 138-145 (2019). 266 108. Murray, A.S. & Olley, J.M. Precision and accuracy in the optically stimulated luminescence 267 dating of sedimentary quartz. Geochronometria 21, 1–16 (2002). 268 109. Preusser, F. et al. Quartz as a natural luminescence dosimeter. Earth-Science Reviews 97, 269 184-214 (2009). 270

110. Rhodes, E.J. Optically stimulated luminescence dating of sediments over the past 200,000 271 years. Annual Review of Earth and Planetary Sciences 39, 461-488 (2011). 272 Madsen, A.T. & Murray, A.S. Optically stimulated luminescence dating of young sediments: 111. 273 a review. Geomorphology 109, 3–16 (2009). 274 112. Wallinga, J. Optically stimulated luminescence dating of fluvial deposits: A review. Boreas 275 31, 303-322 (2003). 276 113. Thrasher, I.M., Mauz, B., Chiverrell, R.C. & Lang, A. Luminescence dating of glaciofluvial 277 deposits: A review. Earth-Science Reviews 97, 133-146 (2009). 278 279 114. Jacobs, Z. & Roberts, R.G. Advances in optically stimulated luminescence dating of individual grains of quartz from archeological deposits. Evolutionary Athropology 16, 210-280 223 (2007). 281 115. Fleming, S.J. The pre-dose technique: a new thermo-luminescent dating method. 282 Archaeometry 15, 13-30 (1973). 283 Wintle, A.G. Thermoluminescence dating of late Devensian loesses in Southern England. 116. 284 Nature 289, 479-480 (1981). 285 Wintle, A.G., Li, S.H. & Botha, G.A. Luminescence dating of colluvial deposits from Natal, 117. 286 South Africa. South African Journal of Science 89, 77-82 (1993). 287 Lang, A. Infra-red stimulated luminescence dating of holocene reworked silty sediments. 118. 288 Quaternary Science Reviews 13, 525-528 (1994). 289 119. Chawla, S. & Singhvi, A.K. Thermoluminescence dating of Archaeological Sediments. 290 Naturwissenschaften 76, 416-419 (1989). 291 120. Owen, L.A. et al. Relic permafrost structures in the Gobi of Mongolia: Age and significance. 292 Journal of Ouaternary Science 13, 539-547 (1998). 293 121. Balescu, S. & Lamothe, M. Comparison of TL and IRSL age estimates of feldspar coarse 294 grains from waterlain sediments. Quaternary Science Reviews 13, 437-444 (1994). 295 Edwards, S.R. Luminescence dating of sand from the Kelso Dunes, California. The dynamics 122. 296 and environmental context of aeolian sedimentary systems, 59-68 (1993). 297 123. Lamothe, M. & Huntley, D.J. Thermoluminescence dating of Late Pleistocene sediments, St. 298 Lawrence Lowland, Quebec. *Geographie Physique et Quaternaire* **42**, 33-44 (1988). 299 124. Banerjee, D., Singhvi, A.K., Pande, K., Chandra, B.P. & Gogte, V. Towards a direct dating of 300 fault gouges using luminescence dating techniques - methodological aspects. Current Science 301 77, 256-268 (1999). 302 125. Singhvi, A.K., Bronger, A., Pant, R.K. & Sauer, W. Thermoluminescence dating and its 303 implications for the chronostratigraphy of loess-paleosol sequences in the Kashmir Valley 304 (India). Chemical Geology: Isotope Geoscience Section 65, 45-56 (1987). 305 126. Bhandari, N., Sen Gupta, D., Singhvi, A.K., Nijampurkar, V.N. & Vohra, C.P. 306 Thermoluminescene dating of glaciers. PACT 9, 513-521 (1983). 307 Sohbati, R., Murray, A.S., Jain, M., Buylaert, J.P. & Thomsen, K.J. Investigating the resetting 127. 308 of OSL signals in rock surfaces. Geochronometria 30, 249-258 (2011). 309 Pietsch, T.J., Olley, J.M. & Nanson, G.C. Fluvial transport as a natural luminescence 128. 310 sensitiser of quartz. Quaternary Geochronology 3, 365-76 (2008). 311 129. Li, S.-H. Luminescence sensitivity changes of quartz by bleaching, annealing and UV 312 exposure. Radiation Effects and Defects in Solids 157, 357-364 (2002). 313 130. Koul, D.K. & Chougaonkar, M.P. The pre-dose phenomenon in the OSL signal of quartz. 314 Radiation Measurements 42, 1265-1272 (2007). 315 Benea, V. et al. Luminescence dating of neolithic ceramics from lumea nouă, Romania. 131. 316 *Geochronometria* **28**, 9-16 (2007). 317 Ideker, C.J., Byrd Finley, J., Rittenour, T.M. & Nelson, M.S. Single-grain optically stimulated 132. 318 luminescence dating of quartz temper from prehistoric Intermountain Ware ceramics, 319 northwestern Wyoming, USA. *Ouaternary Geochronology* **42**, 42-55 (2017). 320 133. Armitage, S.J. & King, G.E. Optically stimulated luminescence dating of hearths from the 321 Fazzan Basin, Libya: A tool for determining the timing and pattern of Holocene occupation of 322 the Sahara. Quaternary Geochronology 15, 88-97 (2013). 323

324	134.	al Khasawneh, S., Murray, A., Bonatz, D. & Freiesleben, T. Testing the application of post IR
325		IRSL dating to Iron- and Viking-age ceramics and heated stones from Denmark. <i>Quaternary</i>
326		Geochronology <b>30</b> , 386-391 (2015).
327	135.	Guérin, G. et al. Multi-method (TL and OSL), multi-material (quartz and flint) dating of the
328		Mousterian site of Roc de Marsal (Dordogne, France): correlating Neanderthal occupations
329		with the climatic variability of MIS 5–3. Journal of Archaeological Science 39, 3071-3084
330		(2012).
331	136.	Moayed, N.K. et al. Bypassing the Suess-effect: Age determination of charcoal kiln remains
332		using OSL dating. Journal of Archaeological Science 120, 105176 (2020).
333	137.	Ahn, HS. et al. Magnetic assessment of OSL and radiocarbon ages of sediments beneath a
334		lava in Jeju Island, Korea: Implication of possible resetting of OSL signals and age constraint
335		of the late Quaternary lava. Quaternary Geochronology 48, 45-63 (2018).
336	138.	Porat, N., Levi, T. & Weinberger, R. Possible resetting of quartz OSL signals during
337		earthquakes—Evidence from late Pleistocene injection dikes, Dead Sea basin, Israel.
338		Quaternary Geochronology 2, 272-277 (2007).
339	139.	$\widetilde{K}$ im, J.H. et al. Experimental investigations on dating the last earthquake event using OSL
340		signals of quartz from fault gouges. <i>Tectonophysics</i> <b>769</b> , 228191 (2019).
341	140.	Tsakalos, E. et al. Absolute dating of past seismic events using the Optically Stimulated
342		Luminescence (OSL) technique on fault gouge material-A case study of the Nojima Fault
343		Zone, SW Japan. Journal of Geophysical Research: Solid Earth 125, 019257 (2019).
344	141.	Navarro-González, R. et al. Paleoecology reconstruction from trapped gases in a fulgurite
345		from the late Pleistocence of the Libiyan Desert. <i>Geology</i> <b>35</b> , 117–120 (2007).
346	142.	Godfrey-Smith, D.I., Huntley, D.J. & Chen, W.H. Optical dating studies of quartz and
347		feldspar sediment extracts. Quaternary Science Reviews 7, 373-380 (1988).
348	143.	Li, G. et al. Quantitative precipitation reconstructions from Chagan Nur revealed lag response
349		of East Asian summer monsoon precipitation to summer insolation during the Holocene in
350		arid northern China. Quaternary Science Reviews 239, 106365 (2020).
351	144.	Stokes, S. et al. Alternative chronologies for Late Quaternary (Last Interglacial-Holocene)
352		deep sea sediments via optical dating of silt-sized quartz. Quaternary Science Reviews 22,
353		925–941 (2003).
354	145.	Singhvi, A.K. & Porat, N. Impact of luminescence dating on geomorphological and
355		palaeoclimate research in drylands. Boreas 37, 536-558 (2008).
356	146.	Telfer, M.W. & Thomas, D.S.G. Late Quaternary linear dune accumulation and
357		chronostratigraphy of the southwestern Kalahari: implications for aeolian palaeoclimatic
358		reconstructions and predictions of future dynamics. Quaternary Science Reviews 26, 2617-
359		2630 (2007).
360	147.	Stevens, T., Thomas, D.S.G., Armitage, S.J., Lunn, H.R. & Lu, H. Reinterpreting climate
361		proxy records from late Quaternary Chinese loess: A detailed OSL investigation. Earth-
362		Science Reviews <b>80</b> , 111-136 (2007).
363	148.	Kang, S. et al. Early Holocene weakening and mid- to late Holocene strengthening of the East
364		Asian winter monsoon. <i>Geology</i> <b>48</b> , 1043–1047 (2020).
365	149.	Zeng, L. et al. Response of dune mobility and pedogenesis to fluctuations in monsoon
366		precipitation and human activity in the Hulunbuir dune field, northeastern China, since the
367		last deglaciation. <i>Global and Planetary Change</i> <b>168</b> , 1-14 (2018).
368	150.	Murton, J.B., Bateman, M.D., Dallimore, S.R., Teller, J.T. & Yang, Z. Identification of
369		Younger Dryas outburst flood path from lake Agassiz to the arctic Ocean. <i>Nature</i> <b>464</b> , 740–
370		43 (2010).
371	151.	Armitage, S.J., Bristow, C.S. & Drake, N.A. West African monsoon dynamics inferred from
372		abrupt fluctuations of Lake Mega-Chad. Proceedings of the National Academy of Sciences of
373	1.50	the United States of America 112, 8543–8548 (2015).
374	152.	Cohen, T.J. et al. Hydrological transformation coincided with megataunal extinction in
375	1.52	central Australia. <i>Geology</i> <b>43</b> , 195–199 (2015).
376	153.	De Deckker, P. et al. Marine Isotope Stage 4 in Australasia: a full glacial culminating 65,000
377		years ago – global connections and implications for human dispersal. <i>Quaternary Science</i>
378		<i>Keviews</i> <b>204</b> , 187–207 (2019).

379	154.	Cohen, K.M. & Gibbard, P.L. Global chronostratigraphical correlation table for the last 2.7
380		million years, version 2019 QI-500. Quaternary International 500, 20-31 (2019).
381	155.	Vandenberghe, J. Multi-proxy analysis: a reflection on essence and potential pitfalls.
382		Netherlands Journal of Geosciences — Geologie en Mijnbouw <b>91</b> , 263-269 (2012).
383	156.	Marković, S.B. et al. Loess correlations – Between myth and reality. Palaeogeography,
384		Palaeoclimatology, Palaeoecology <b>509</b> , 4-23 (2018).
385	157.	Svendsen, J.I. et al. Late Quaternary ice sheet history of northern Eurasia. <i>Quaternary Science</i>
386		<i>Reviews</i> 23, 1229-1271 (2004).
387	158.	Thiede, J. https://en.wikipedia.org/wiki/Quaternary Environment of the Eurasian North
388		(2003).
389	159.	Smedley, R.K. et al. New age constraints for the limit of the British-Irish ice sheet on the Isles
390		of scilly, Journal of Quaternary Science <b>32</b> , 48–62 (2017).
391	160.	Clark, C.D. http://www.britice-chrono.group.shef.ac.uk/ (2018).
392	161.	Lancaster, N. et al. The INOUA Dunes Atlas chronologic database. <i>Quaternary International</i>
393	101.	<b>410</b> 3-10 (2016)
30/	162	Lancaster N https://www.dri.edu/inquadunesatlas/
305	162.	Buylaert IP et al Ontical dating of relict sand wedges and composite-wedge pseudomorphs
306	105.	in Flanders Belgium <i>Boreas</i> <b>38</b> 160–175 (2009)
207	164	Chaong C S at al. Determination of slip rate by optical dating of fluxial deposits from the
397	104.	Wongson foult SE Koroo. Quaternamy Science Provided dating of Indvia deposits from the
390	165	Choi S. L et al. Estimation of possible maximum earthqueke magnitudes of Queternery faults.
399	105.	choi, SJ. et al. Estimation of possible maximum earliquake magnitudes of Quaternary faults
400	166	In the southern Korean Pennisula. Qualernary International <b>344</b> , 55-65 (2014).
401	100.	foult in NE Delaium, OSL and constraints. Quatum and Holocene earlinguake events along the Geleen
402	1.67	Tault in NE Belgium: OSL age constraints. <i>Quaternary International</i> <b>199</b> , 56-74 (2009).
403	107.	Fattani, M. et al. Holocene sup-rate on the Sabzevar thrust raut, NE fran, determined using
404		(2006)
405	160	(2000). Heimseth A.M. Chennell I. Snooner N.A. & Questioux D.C. Creening soil Coolean <b>30</b>
406	108.	111 114 (2002)
407	160	111-114 (2002).
408	109.	distribution to identify phases of podegonic mixing on dryland alluvial for Oustaman
409		Intermetional <b>262</b> , 22, (2015)
410	170	International <b>302</b> , 23–35 (2013).
411	170.	Dateman, M.D., Fleuenck, C.D., Jaiswal, M.K. & Singhvi, A.K. Investigations into the
412		potential effects of pedoturbation on fummescence dating. <i>Quaternary Science Reviews 22</i> , 1160–76 (2002)
413	171	1109–70 (2003). Dials W. I. at al. Subtarrangen transport and denosition of quartz by ents in conduction
414	1/1.	Klink, w.J. et al. Subternation in onticel luminessence deting. Low all of Archaeological
415		Seimes <b>40</b> , 2217, 2226 (2012)
416	170	Science 40, 2217-2220 (2015).
417	1/2.	Giay, H.J., Keen-Zebert, A., Fuldish, D.J., Tucker, G.E. & Mahan, S.A. Depth dependent son
418		mixing persists across chinatic zone. <i>Proceedings of the National Academy of Sciences</i> <b>11</b> 7, 9750, 9756 (2020)
419	172	0750-0750 (2020).
420	175.	Ansteinsen, J.A. et al. Qualification of termine bioturbation in a savannah ecosystem.
421	174	application of OSL dating. Quaternary Geochronology <b>50D</b> , 534-541 (2015).
422	1/4.	Geometric de la complete de la compl
423	175	Geomorphology 307, 107296 (2020).
424	175.	Madsen, A. I., Murray, A.S., Jain, M., Andersen, I.J. & Pejrup, M. A new method for
425		Estumine Constal and Shalf Science <b>02</b> A(A, 472 (2011))
426	176	Estuarine, Coastal and Shelf Science 92, 404–472 (2011).
427	1/0.	Au, Z. et al. Chinate-driven changes to dure activity during the Last Glacial Maximum and declarization in the Mu Lie dure field months equated $China = E_{\rm cut} + E_{\rm cut}$
428		degraciation in the Niu US dune field, north-central Unina,. Earth and Planetary Science
429	177	Letters, 441, 149-139 (2013). Stations T at al. Abrupt last algorid duct fall over contheast English descripted with
430	1//.	Stevens, 1. et al. Adrupt last glacial dust fall over southeast England associated with dynamics of the British Irish ice shoet. Our term are Science British 250, 106641 (2020)
431	170	uynamics of the Difusit-firsh ice sheet, Quaternary Science Kevlews, 250, 100041 (2020).
432	1/8.	rotat, IN., Rosell, S.A., Doareuo, E. & Avill, I. Dauling the Kalinal Sanaromini Late Neolithic desort cult site. <i>Journal of Archaeological Science</i> <b>22</b> , 1241, 1255 (2006)
433		uesen cun sne. <i>Journal of Archaeological Science</i> <b>33</b> , 1541-1555 (2000).

179. Huang, C.C. et al. Extraordinary Floods of 4100-4000 a BP recorded at the Late Neolithic 434 Ruins in the Jinghe River Gorges, Middle Reach of the Yellow River, China. 435 Palaeogeography, Palaeoclimatology, Palaeoecology 289, 1-9 (2010). 436 180. Saunders, J.W. et al. A mound complex in Louisiana at 5400-5000 years before present. 437 Science 277, 1796-1799 (1997). 438 181. Burbidge, C.I., Batt, C.M., Barnett, S.M. & Dockrill, S.J. The potential for dating the old 439 Scatness site, Shetland, by optically stimulated luminescence. Archaeometry 43, 589-596 440 (2001). 441 182. Roberts, R.G. et al. Luminescence dating of rock art and past environments using mud-wasp 442 nests in northern Australia. Nature 387, 696–699 (1997). 443 183. Pederson, J.L. et al. Age of Barrier Canyon-style rock art constrained by cross-cutting 444 relations and luminescence dating techniques. Proceedings of the National Academy of 445 Sciences 111, 12986–12991 (2014). 446 Blain, S., Bailiff, I.K., Guibert, P., Bouvier, A. & Baylé, M. An intercomparison study of 184. 447 luminescence dating protocols and techniques applied to medieval brick samples from 448 Normandy (France). Quaternary Geochronology 5, 311-316 (2007). 449 Guibert, P., Christophe, C., Urbanova, P., Guérin, G. & Blain, S. Modeling incomplete and 185. 450 heterogeneous bleaching of mobile grains partially exposed to the light: towards a new tool 451 for single grain OSL dating of poorly bleached mortars. Radiation Measurements 107, 48-57 452 (2017). 453 Greilich, S., Glasmacher, U.A. & Wagner, G.A. Optical dating of granitic stone surfaces. 186. 454 Archaeometry 47, 645-665 (2005). 455 187. Chapot, M.S., Sohbati, R., Murray, A.S., Pederson, J.L. & Rittenour, T.M. Constraining the 456 age of rock art by dating a rockfall event using sediment and rock-surface luminescence 457 dating techniques. Quaternary Geochronology 13, 18-25 (2012). 458 Sohbati, R., Murray, A.S., Buylaert, J.-P., Almeida, N.A.C. & Cunha, P.P. Optically 188. 459 stimulated luminescence (OSL) dating of quartzite cobbles from the Tapada do Montinho 460 archaeological site (east-central Portugal). Boreas 41, 452-562 (2012). 461 189. Petraglia, M. et al. Middle Paleolithic Assemblages from the Indian Subcontinent Before and 462 After the Toba Super-Eruption. Science 317, 114–116 (2007). 463 190. Armitage, S.J. et al. The southern route 'out of Africa': evidence for an early expansion of 464 modern humans into Arabia. Science 331, 453-456 (2011). 465 191. Mishra, S., Chauhan, N. & Singhvi, A.K. Continuity of Microblade Technology in the Indian 466 Subcontinent since at least 45 ka: Implications for the Dispersal of Modern humans. PLoS 1 8 467 (2013). 468 192. Meyer, M.C. et al. Permanent human occupation of the central Tibetan Plateau in the early 469 Holocene. Science 355, 64-67 (2017). 470 193. Hu, Y. et al. Late Middle Pleistocene Levallois stone-tool technology in southwest China. 471 Nature 565, 82–85 (2019). 472 Zhang, X.L. et al. The earliest human occupation of the high-altitude Tibetan Plateau 40 194. 473 thousand to 30 thousand years ago. Science 362, 1049-1051 (2018). 474 195. Roberts, R. et al. Optical and radiocarbon dating at Jinmium rock shelter in northern 475 Australia. Nature 393, 358–362 (1998). 476 196. Bowler, J.M. et al. New ages for human occupation and climate change at Lake Mungo, 477 Australia. Nature 421, 837-840 (2003). 478 197. Hamm, G. et al. Cultural innovation and megafauna interaction in the early settlement of arid 479 Australia. Nature 539, 280–283 (2016). 480 Clarkson, C. et al. New evidence for human occupation of northern Australia by 65,000 years 198. 481 ago. Nature 547, 306-310 (2017). 482 199. Lahave, C. et al. Human occupation in South America by 20,000 BC: the Toca da Tira Peia 483 site, Piauí, Brazil. Journal of Archaeological Science 40, 2840-2847 (2013). 484 Lahaye, C. et al. Another site, same old song: the Pleistocene-Holocene archaeological 200. 485 sequence of Toca da Janela da Barra do Antonião-North, Piauí, Brazil. Quaternary 486 Geochronology 49, 223-229 (2019). 487

488	201.	Jacobs, Z. et al. Timing of archaic hominin occupation of Denisova Cave in southern Siberia.
489		<i>Nature</i> <b>565</b> , 594–599 (2019).
490	202.	Zilhão, J. et al. Last Interglacial Iberian Neandertals as Fisher-Hunter-Gatherers. Science 367,
491		1443 (2020).
492	203.	Bouzouggar, A. et al. 82,000-year-old shell beads from North Africa and implications for the
493		origins of modern human behavior. <i>Proceedings of the National Academy of Sciences</i> <b>104</b> .
494		9964–9969 (2007)
495	204	Carter, T et al. Earliest occupation of the Central Aegean (Naxos). Greece: Implications for
196	2011	hominin and Homo saniens' behavior and dispersals <i>Science advances</i> <b>5</b> eaax0997 (2019)
407	205	Petraglia M et al Population increase and environmental deterioration correspond with
497	205.	microlithic inpositions in South Asia ca. 25,000 years ago. <i>Proceedings of the National</i>
490		Academy of Sciences <b>106</b> , 12261, 12266 (2000)
499	206	Henchilwood C S. at al. A 100,000 Year Old Oahra Processing Workshop at Plambos Cava
500	200.	South A frice, Science <b>324</b> , 210, 222 (2011)
501	207	South Annea. Science <b>334</b> , 219–222 (2011).
502	207.	Roberts, R.G. et al. New ages for the fast Australian megalauna: continent-wide extinction
503	200	about 46,000 years ago. <i>Science</i> <b>292</b> , 1888–1892 (2001).
504	208.	Prideaux, G.J. et al. An arid-adapted middle Pleistocene vertebrate fauna from south-central
505		Australia. <i>Nature</i> <b>445</b> , 422–425 (2007).
506	209.	Haile, J. et al. Ancient DNA reveals late survival of mammoth and horse in interior Alaska.
507		Proceedings of the National Academy of Sciences <b>106</b> , 22363–22368 (2009).
508	210.	Hocknull, S.A. et al. Extinction of eastern Sahul megafauna coincides with sustained
509		environmental deterioration. Nature Communications 11, 2250 (2020).
510	211.	Medialdea, A., Thomsen, K.J., Murray, A.S. & Benito, G. Reliability of equivalent-dose
511		determination and age-models in the OSL dating of historical and modern palaeoflood
512		sediments. Quaternary Geochronology 22, 11-24 (2014).
513	212.	Kale, V.S., Singhvi, A.K., Mishra, P.K. & Banerjee, D. Sedimentary records and
514		luminescence chronology of Late Holocene palaeofloods in the Luni River, Thar Desert,
515		northwest India. Catena 40, 337-358 (2000).
516	213.	Arnold, L.J., Roberts, R.G., Galbraith, R.F. & DeLong, S.B. A revised burial dose estimation
517		procedure for optical dating of young and modern-age sediments. <i>Quaternary Geochronology</i>
518		4, 306–325 (2009).
519	214.	Galbraith, R.F. & Roberts, R.G. Statistical aspects of equivalent dose and error calculation
520		and display in OSL dating: an overview and some recommendations. <i>Quaternary</i>
521		Geochronology 11. $1-27$ (2012).
522	215.	Arnold, L.J., Demuro, M. & Ruiz, N. Empirical Insights into multi-grain averaging effects
523		from 'pseudo' single-grain OSL measurements. <i>Radiation Measurements</i> <b>47</b> , 652–658
524		(2012a)
525	216	Timar-Gabor A et al. On the importance of grain size in luminescence dating using quartz
526	210.	Radiation Measurements <b>106</b> 464-471 (2017)
527	217	Choi I H Murray A S Cheong C S Hong D G & Chang H W The resolution of
528	217.	stratigraphic inconsistency in the luminescence ages of marine terrace sediments from Korea
520		Quaternary Science Reviews 22, 1201–1206 (2003)
525	218	Singeraver IS & Bailey B M Component resolved bleaching spectra of quartz ontically
530	210.	stimulated luminosconos: preliminary results and implications for dating. <i>Padiation</i>
531		Maggurements <b>29</b> , 111, 119 (2004)
532	210	Meusurements <b>30</b> , 111-110 (2004).
533	219.	Zhou, L.P., Fu, D.P. & Zhang, J.F. An analysis of the components of the huminescence signals
534		Taillitetan and their suitability for artical dating $Q_{\rm eff}$ ( $Q_{\rm eff}$ ) ( $Q_{\rm $
535		rajikistan, and their suitability for optical dating. Quaternary Geochronology 5, 149–153
536	000	
537	220.	Galbraith, K.F., Roberts, R.G., Laslett, G.M., Yoshida, H. & Olley, J.M. Optical dating of
538		single and multiple grains of quartz from Jinmium rock shelter, northern Australia: Part I,
539	001	experimental design and statistical models. Archaeometry 41, 339-364 (1999).
540	221.	Bailey, R.M. & Arnold, L.J. Statistical modelling of single grain quartz De distributions and
541		an assessment of procedures for estimating burial dose. <i>Quaternary Science Reviews</i> 25,
542		2475–2502 (2006).

- 222. Combès, B. & Philippe, A. Bayesian analysis of individual and systematic multiplicative
   errors for estimating ages with stratigraphic constraints in optically stimulated luminescence
   dating. *Quaternary Geochronology* 39, 24–34 (2017).
- Philippe, A., Guérin, G. & Kreutzer, S. BayLum-An R package for Bayesian analysis of OSL ages: An introduction. *Quaternary Geochronology* 49, 16–24 (2019).
- 224. Combès, B. et al. A Bayesian central equivalent dose model for optically stimulated
   luminescence dating. *Quaternary Geochronology* 28, 62-70 (2015).
- Thomsen, K.J., Murray, A.S., Bøtter-Jensen, L. & Kinahan, J. Determination of burial dose in incompletely bleached fluvial samples using single grains of quartz. *Radiation Measurements* 42, 370-379. (2007).
- Jain, M., Murray, A.S. & Bøtter-Jensen, L. Optically stimulated luminescence dating: How significant is incomplete light exposure in fluvial environments? *Quaternaire* 15, 143-157 (2004).
- Gliganic, L.A., Cohen, T.J., Meyer, M. & Molenaar, A. Variations in luminescence properties
   of quartz and feldspar from modern fluvial sediments in three rivers. *Quaternary Geochronology* 41, 70–82 (2017).
- Arnold, L.J. et al. Single-grain TT-OSL bleaching characteristics: Insights from modern analogues and OSL dating comparisons. *Quaternary Geochronology* 49, 45–51 (2019).
- Murray, A.S., Thomsen, K.J., Masuda, N., Buylaert, J.P. & Jain, M. Identifying well-bleached
   quartz using the different bleaching rates of quartz and feldspar luminescence signals.
   *Radiation Measurements* 47, 688–695 (2012).
- Demuro, M., Arnold, L.J., Parés, J.M. & Sala, R. Extended-range luminescence chronologies
   suggest potentially complex bone accumulation histories at the Early-to-Middle Pleistocene
   palaeontological site of Huéscar-1 (Guadix-Baza basin, Spain). *Quaternary International* 389,
   191–212 (2015).
- Möller, P. & Murray, A.S. Drumlinised glaciofluvial and glaciolacustrine sediments on the
   Småland peneplain, South Sweden new information on the growth and decay history of the
   Fennoscandian Ice Sheets during MIS 3. *Quaternary Science Reviews* 122, 1–29, (2015).
- 571 232. Duller, G.A.T. Single-grain optical dating of Quaternary sediments: why aliquot size matters 572 in luminescence dating. *Boreas* **37**, 589–612 (2008).
- Jacobs, Z., Wintle, A.G., Duller, G.A., Roberts, R.G. & Wadley, L. New ages for the postHowiesons Poort, late and final Middle stone age at Sibudu, South Africa. *Journal of Archaeological Science* 35, 1790-1807 (2008).
- Fitzsimmons, K.E., Stern, N. & Murray-Wallace, C.V. Depositional history and archaeology
   of the central Lake Mungo lunette, Willandra Lakes, southeast Australia. *Journal of Archaeological Science* 41, 349-364 (2014).
- Galbraith, R.G. & Galbraith, R.F. Estimating the component ages in a finite mixture. *Nucl. Tracks Radiat. Meas.* 17, 197-206 (1990).
- Jacobs, Z., Duller, G.A.T., Wintle, A.G. & Henshilwood, C.S. Extending the chronology of
   deposits at Blombos Cave, South Africa, back to 140 ka using optical dating of single and
   multiple grains of quartz. *Journal of Human Evolution* 51, 255-273 (2006).
- David, B. et al. Sediment mixing at Nonda Rock: investigations of stratigraphic integrity at an
   early archaeological site in northern Australia and implications for the human colonisation of
   the continent. *Journal of Quaternary Science* 22, 449–479 (2007).
- Bateman, M.D. et al. Detecting post-depositional sediment disturbance in sandy deposits
  using optical luminescence. *Quaternary Geochronology* 2, 57–64 (2007).
- Wolfe, S., Murton, J., Bateman, M. & Barlow, J. Oriented-lake development in the context of
   late Quaternary landscape evolution, McKinley Bay Coastal Plain, western Arctic Canada.
   *Quaternary Science Reviews* 242, 106414 (2020).
- Arnold, L.J. & Roberts, R.G. Stochastic modelling of multi-grain equivalent dose (De)
   distributions: implications for OSL dating of sediment mixtures. *Quaternary Geochronology* 4, 204-230 (2009).
- Rufer, D. & Preusser, F. Potential of autoradiography to detect spatially resolved radiation
   patterns in the context of trapped charge dating. *Geochronometria* 34, 1-13 (2009).

597	242.	Smedley, R.K., Duller, G.A.T., Rufer, D. & Utley, J. Empirical assessment of beta dose
598		heterogeneity in sediments: implications for luminescence dating. Quaternary Geochronology
599		<b>56</b> , 101052 (2020).
600	243.	Nathan, R., Puthusserry, T., Jain, M. & Murray, A.S. Environmental dose rate heterogeneity
601		due to beta microdosimetry and its implications for luminescence dating: Monte Carlo
602		modelling and experimental validation. <i>Radiation Measurements</i> <b>37</b> , 305–313 (2003).
603	244.	Guibert, P., Bechtel, F., Schvoerer, M., Müller, P. & Balescu, S. A new method for gamma
604		dose-rate estimation of heterogeneous media in TL dating. <i>Radiation Measurements</i> . 29, 561-
605		572 (1998).
606	245.	Burbidge, C.I. & Duller, G.A.T. Combined gamma and beta dosimetry, using Al2O3:C. for in
607		situ measurements on a sequence of archaeological deposits. <i>Radiation Measurements</i> 37.
608		285-291 (2003).
609	246	Guérin G & Mercier N Field gamma spectrometry Monte Carlo simulations and potential
610	210.	of non-invasive measurements. <i>Geochronometria</i> <b>39</b> 40-47 (2012)
611	247	Guérin G & Mercier N Preliminary insight into dose deposition processes in sedimentary
610	247.	media on a scale of single grains: Monte Carlo modelling of the effect of water on the gamma
612		dose rate <i>Radiation Magsuraments</i> <b>A7</b> 541 547 (2012)
013	210	use rate. Relation measurements, $47$ , $541-547$ (2012).
014	240.	begennig, D. & Degennig, A. Change is the only constant - time-dependent dose rates in
615	240	Sinchui A.K. Denenice D. Demech D. Deiserum S.N. & Coste V. A.Luminessence
616	249.	Singnvi, A.K., Banerjee, D., Ramesn, R., Rajaguru, S.N. & Gogie, V. A Luminescence
617		the for dating dirty pedogenic carbonates for paleoenvironmental reconstruction. Earth
618	250	and Planetary Science Letters 139, 321, (1996).
619	250.	Nathan, R.P. & Mauz, B. On the dose-rate estimate of carbonate-rich sediments for trapped
620	0.5.1	charge dating. Radiation Measurements 43, 14–25 (2008).
621	251.	Olley, J.M., Murray, A.S. & Roberts, R.G. The effects of disequilibria in the uranium and
622		thorium decay chains on burial dose rates in fluvial sediments. <i>Quaternary Geochronology</i>
623		<b>15</b> , 751–760 (1996).
624	252.	Tanner, A.B. in The Natural radiation Environment III, Conf. 780422, Dept. of Commerce
625		(Viginia, 1978).
626	253.	Olley, J.M., Roberts, R.G. & Murray, A.S. Disequilibria in the uranium decay series in
627		sedimentary deposits at Allens cave, Nullarbor Plain, Australia: Implications for dose rate
628		determinations. <i>Radiation Measurements</i> 27, 433–443 (1997).
629	254.	Murray, A.S., Helsted, L.M., Autzen, M., Jain, M. & Buylaert, J.P. Measurement of natural
630		radioactivity: Calibration and performance of a high-resolution gamma spectrometry facility.
631		Radiation Measurements 15, 215-220 (2018).
632	255.	Murray, A.S., Buylaert, J.P. & Thiel, C. A luminescence dating intercomparison based on a
633		Danish beach-ridge sand. Radiation Measurements 81, 32-38 (2015).
634	256.	Durcan, J.A., King, G.E. & Duller, G.A. DRAC: Dose Rate and Age Calculator for trapped
635		charge dating. Quaternary Geochronology 28, 54-61 (2015).
636	257.	Kreutzer, S. et al. Introducing an R package for luminescence dating analysis. Ancient TL 30,
637		1-8 (2012).
638	258.	Peng, J., Dong, Z., Han, F., Long, H. & Liu, X. R package numOSL: numeric routines for
639		optically stimulated luminescence dating. Ancient TL 31, 41-48 (2013).
640	259.	Lai, Z.P., Brückner, H., Zöller, L. & Füllinga, A. Existence of a common growth curve for
641		silt-sized quartz OSL of loess from different continents. Radiation Measurements 42, 1432-
642		1440 (2007).
643	260.	Burbidge, C.I., Duller, G.A.T. & Roberts, H.M. De determination for young samples using
644		the standardised OSL response of coarse-grain quartz. Radiation Measurements 41, 278-288
645		(2006).
646	261.	Telfer, M.W. & Hesse, P.P. Palaeoenvironmental reconstructions from linear dunefields:
647		Recent progress, current challenges and future directions. <i>Ouaternary Science Reviews</i> 78. 1-
648		21 (2013).
649	262.	Thomsen, K.J., Murray, A. & Jain, M. The dose dependency of the over-dispersion of quartz
650		OSL single grain dose distributions. Radiation Measurements 47, 732–739 (2012).

651	263.	Timar-Gabor, A. et al. Optical dating of Romanian loess: A comparison between silt-sized
652		and sand-sized quartz. Quaternary International 240, 62-70 (2011).
653	264.	Qin, J.T. & Zhou, L.P. Stepped-irradiation SAR: A viable approach to circumvent OSL
654		equivalent dose underestimation in last glacial loess of northwestern China. Radiation
655		<i>Measurements</i> <b>44</b> , 417–422 (2009).
656	265.	Timar-Gabor, A. & Wintle, A.G. On natural and laboratory generated dose response curves
657		for quartz of different grain sizes from Romanian loess. Quaternary Geochronology 18, 34-40
658		(2013).
659	266.	Timar-Gabor, A. et al. Fundamental investigations of natural and laboratory generated SAR
660		dose response curves for quartz OSL in the high dose range. <i>Radiation Measurements</i> 81,
661	0.47	150-156 (2015).
662	267.	Chauhan, N., Anand, S., Selvam, P., Mayya, Y.S. & Singhvi, A.K. Extending the upper age
663		limits in the luminescence dating of sediments: A feasibility study of using large grains. $D_{i}$ is $M_{i}$ is $M_{i}$ in the luminescence dating of sediments: A feasibility study of using large grains.
664	269	Radiation Measurements 44, 629-633 (2009).
665	208.	heydari, M. & Guerin, G. OSL signal saturation and dose rate variability: investigating the
666	260	Denaviour of different statistical models. <i>Radiation Measurements</i> <b>120</b> , 96-105 (2018).
667	209.	Rittenour, 1. Luminescence dating of fluvial deposits: applications to geomorphic,
000	270	L ai Z. Chronology and the upper dating limit for loss samples from Lucchuan section in the
670	270.	Chinese Loess Plateau using quartz OSL SAR protocol <i>Journal of Asian Earth Sciences</i> 37
671		176-185 (2010)
672	271	Ankiærgaard $C$ et al. Violet stimulated luminescence: geo- or thermochronometer?
673	271.	Radiation Measurements 81 78-84 (2015)
674	272.	Wang, X.L., Lu, Y.C. & Wintle, A.G. Recuperated OSL dating of fine-grained quartz in
675		Chinese loess. <i>Quaternary Geochronology</i> <b>1</b> , 89-100 (2006).
676	273.	Arnold, L.J. & Demuro, M. Insights into TT-OSL signal stability from single-grain analyses
677		of known-age deposits at Atapuerca, Spain. Quaternary Geochronology 30, 472-478 (2015).
678	274.	Duller, G.A.T. & Wintle, A.G. A review of the thermally transferred optically stimulated
679		luminescence signal from quartz for dating sediments. Quaternary Geochronology 7, 6-20
680		(2012).
681	275.	Hansen, V. et al. Towards the origins of over-dispersion in beta source calibration. Radiation
682		<i>Measurements</i> <b>120</b> , 157-162 (2018).
683	276.	Stevens, T., Armitage, S.J., Lu, H. & Thomas, D.S.G. Sedimentation and diagenesis of
684		Chinese loess: Implications for the preservation of continuous, high-resolution climate
685	077	records. <i>Geology</i> <b>34</b> , 849-852 (2006).
686	277.	Y1, S. et al. High resolution USL and post-IR IRSL dating of the last interglacial-glacial cycle
687		at the Sandanuo loess site (northeastern China). Quaternary Geochronology <b>30</b> , 200-206
688	270	(2013). Sucieski S. et al. OSI define of fine grained quartz from Helegone Vangtze delte sediments
600	270.	Sugisari, S. et al. OSE dating of fine-granied quartz from from from engize dena sediments.
690	279	Olley IM Caitcheon GG & Murray AS The distribution of apparent doses as
692	21).	determined by optically stimulated luminescence in small aliquots of fluxial quartz.
693		implications of dating young sediments. <i>Quaternary Science Reviews</i> <b>17</b> , 1033-1040 (1998)
694	280.	Olley, I.M., Caitcheon, G.G. & Roberts, R.G. The origin of dose distributions in fluvial
695		sediments, and the prospect of dating single grains from fluvial deposits using optically
696		stimulated luminescence. Radiation Measurements <b>30</b> , 207-217 (1999).
697	281.	Sanderson, D.C.W. & Murphy, S. Using simple portable OSL measurements and laboratory
698		characterisation to help understand complex and heterogeneous sediment sequences for
699		luminescence dating. Quaternary Geochronology 5, 299-305 (2010).
700	282.	Muños-Salinas, E., Bishop, P., Sanderson, D.C.W. & Zamorano, J. Interpreting luminescence
701		data from a portable OSL reader: three case studies in fluvial settings. Earth Surface
702		Processes and Landforms <b>36</b> , 651-660 (2010).
703	283.	Stone, A.E.C., Bateman, M.D. & Thomas, D.S.G. Rapid age assessment in the Namib Sand
704		Sea using a portable luminescence reader. <i>Quaternary Geochronology</i> <b>30</b> , 134-140 (2015).

Simms, A.R., DeWitt, R., Kouremenos, P. & Drewry, A.M. A new approach to reconstructing 284. 705 sea levels in Antarctica using optically stimulated luminescence of cobble surfaces. 706 Quaternary Geochronology 6, 50-60 (2011). 707 Souza, P.E. et al. Luminescence dating of buried cobble surfaces from sandy beach ridges: a 285. 708 case study from Denmark. . Boreas 48, 841-855 (2019). 709 Jenkins, G.T.H., Duller, G.A.T., Roberts, H.M., Chiverrell, R.C. & Glasser, N.F. A new 286. 710 approach for luminescence dating glaciofluvial deposits - High precision optical dating of 711 cobbles. Quaternary Science Reviews 192, 263-273 (2018). 712 Sohbati, R., Murray, A.S., Chapot, M.S., Jain, M. & Pederson, J. Optically stimulated 713 287. luminescence (OSL) as a chronometer for surface exposure dating. Journal of Geophysical 714 Research 117, B09202 (2012). 715 288. Rades, E.F., Sohbati, R., Lüthgens, C., Jain, M. & Murray, A. First luminescence-depth 716 profiles from boulders from moraine deposits: Insights into glaciation chronology and 717 transport dynamics in Malta valley, Austria. Radiation Measurements 120, 281-289 (2018). 718 289. Gray, H.J., Jain, M., Sawakuchi, A.O., Mahan, S. & Tucker, G. Luminescence as a sediment 719 tracer and provenance tool. Reviews of Geophysics 57, 987-1017 (2019). 720 Aitken, M.J. An Introduction to Optical Dating. The Dating of Quaternary Sediments by the 721 290. Use of Photon-Stimulated Luminescence (Oxford University Press, Oxford, 1998). 722 291. Karimi Moayed, N. et al. Bypassing the Suess-effect : age determination of charcoal kiln 723 remains using OSL dating. Journal of Archaeological Science 120, 105176 (2020). 724 292. Vandenberghe, D., Derese, C. & Houbrechts, G. Residual doses in recent alluvial sediments 725 from the Ardenne (S Belgium). Geochronometria 28, 1-8 (2007). 726 293. Rémillard, A.M. et al. Ouartz OSL dating of late Holocene beach ridges from the Magdalen 727 Islands (Quebec, Canada). Quaternary Geochronology 30, 264-269 (2015). 728 Murray, A.S. & Funder, S. OSL dating of a Danish Eemian coastal marine deposit: a test of 294. 729 accuracy. Quaternary Science Reviews 22, 1177-1183 (2003). 730 295. Sanguino, J. & Montes, R. in Neandertales cantábricos, estado de la cuestión 489-504 (Museo 731 de Altamira, Santander, 2005). 732 296. Curry, C.W., Bennett, R.H., Hulbert, M.H., Curry, K.J. & Faas, R.W. Comparative Study of 733 Sand Porosity and a Technique for Determining Porosity of Undisturbed Marine Sediment. 734 Marine Georesources & Geotechnology 22, 231–252 (2004). 735 297. Nelson, M.S. & Rittenour, T.M. Using grain-size characteristics to model soil water content: 736 Application to dose-rate calculation for luminescence dating. Radiation Measurements 81, 737 142-149 (2015). 738 Zimmerman, D.W. Thermoluminescent dating using fine grains from pottery. Archaeometry 298. 739 13, 29-52 (1971). 740 299. Lawson, M.J., Daniels, J.T.M. & Rhodes, E.J. Assessing Optically Stimulated Luminescence 741 (OSL) signal contamination within small aliquots and single grain measurements utilizing the 742 composition test Quaternary International 362, 34-41 (2015). 743 Antohi-Trandafir, O. et al. Luminescence properties of natural muscovite relevant to optical 300. 744 dating of contaminated quartz samples. Radiation Measurements 109, 1-7 (2018). 745 301. Banerjee, D., Murray, A.S., Bøtter-Jensen, L. & Lang, A. Equivalent dose estimation from a 746 single aliquot of polymineral fine-grains. Radiation Measurements 33, 73-93 (2001). 747 302. Denby, P.M., Bøtter-Jensen, L., Murray, A.S. & Moska, P. Application of pulsed OSL to the 748 separation of the luminescence components from a mixed quartz/feldspar sample. Radiation 749 Measurements 41, 774–779 (2006). 750 Fan, Y. et al. History and mechanisms for the expansion of the Badain Jaran Desert, northern 303. 751 China, since 20 ka: Geological and luminescence chronological evidence. The Holocene 26, 752 532-548 (2015). 753 304. Yu, L., Lai, Z., An, P., Pan, T. & Chang, Q. Aeolian sediments evolution controlled by fluvial 754 processes, climate change and human activities since LGM in the Qaidam Basin, Qinghai-755 Tibetan Plateau. Quaternary International 372, 23-32 (2015). 756 Hesse, P.P. How do longitudinal dunes respond to climate forcing? Insights from 25 years of 305. 757 luminescence dating of the Australian desert dunefields. Quaternary International 410, 11-29 758 (2016). 759

306. Wang, X.F., Zhao, H., Yang, H.Y. & Wang, K.Q. Optical dating reveals that the height of 760 Earth's tallest megadunes in the Badain Jaran Desert of NW China is increasing. Journal of 761 Asian Earth Sciences 185, 104025 (2019). 762 Xu, Z. et al. Critical transitions in Chinese dunes during the past 12,000 years. Science 307. 763 Advances 6, 8020 (2020). 764 308. Kocurek, G. et al. White Sands Dune Field, New Mexico: Age, dune dynamics and recent 765 accumulations. Sedimentary Geology 197, 313-331 (2007). 766 Li, G.Q. et al. Environmental changes in the Ulan Buh Desert, southern Inner Mongolia, 309. 767 China since the middle Pleistocene based on sedimentology, chronology and proxy indexes. 768 Quaternary Science Reviews 128, 69-80 (2015). 769 310. Tamura, T. et al. Seasonal control on coastal dune morphostratigraphy under a monsoon 770 climate, Mui Ne dunefield, SE Vietnam. Geomorphology 370, 107371 (2020). 771 311. Yang, L., Wang, T., Long, H. & He, Z. Late Holocene dune mobilization in the Horgin 772 dunefield of northern China. Journal of Asian Earth Sciences 138, 136-147 (2017). 773 312. Roberts, H.M., Wintle, A.G., Maher, B.A. & Hu, M. Holocene sediment-accumulation rates 774 in the western Loess Plateau, China, and a 2500-year record of agricultural activity, revealed 775 by OSL dating. The Holocene 11, 477-483 (2001). 776 Lai, Z.P. & Wintle, A.G. Locating the boundary between the Pleistocene and the Holocene in 313. 777 Chinese loess using luminescence. The Holocene 16, 893-899 (2006). 778 314. Kang, S. et al. Increasing effective moisture during the Holocene in the semiarid regions of 779 the Yili Basin, Central Asia: Evidence from loess sections. Quaternary Science Reviews 246, 780 781 106553 (2020). 315. Tecsa, V. et al. Revisiting the chronostratigraphy of Late Pleistocene loess-paleosol 782 sequences in southwestern Ukraine: OSL dating of Kurortne section. Quaternary 783 International 542, 65-79 (2020). 784 316. Stevens, T. et al. Abrupt last glacial dust fall over southeast England associated with 785 dynamics of the British-Irish ice sheet. Quaternary Science Reviews 250, 106641 (2020). 786 317. Xu, Z., Stevens, T., Yi, S., Mason, J.A. & Lu, H. Seesaw pattern in dust accumulation on the 787 Chinese Loess Plateau forced by late glacial shifts in the East Asian monsoon. Geology 46, 788 871-874 (2018). 789 318. Constantin, D. et al. High-resolution OSL dating of the Costinesti section (Dobrogea, SE 790 Romania) using fine and coarse quartz. Quaternary International 334-335, 20-29 (2014). 791 319. Fuchs, M. et al. The loess sequence of Dolní Věstonice, Czech Republic: A new OSL-based 792 chronology of the Last Climatic Cycle. Boreas 42, 664-677 (2013). 793 320. Roberts, H.M., Muhs, D.R., Wintle, A.G., Duller, G.A.T. & Bettis III., E.A. Unprecedented 794 last-glacial mass accumulation rates determined by luminescence dating of loess from western 795 Nebraska. Quaternary Research 59, 411-419 (2003). 796 Bosq, M. et al. Chronostratigraphy of two Late Pleistocene loess-palaeosol sequences in the 321. 797 Rhône Valley (southeast France). Quaternary Science Reviews 245, 106473 (2020). Srivastava, P., Juyal, N., Singhvi, A.K. & Wasson, R.J. Luminescence chronology of river 322. 799 adjustment and incision of Quaternary sediments in the alluvial plain of the Sabarmati River, 800 north Gujarat, India. Geomorphology 36, 217-229 (2001). 801 323. Zhang, J.F. et al. Optical dating of a hyperconcentrated flow deposit on a Yellow River 802 terrace in Hukou, Shaanxi, China. *Ouaternary Geochronology* 5, 194-199 (2010). 803 324. Sim, A.K. et al. Dating recent floodplain sediments in the Hawkesbury-Nepean River system, 804 eastern Australia using single-grain quartz OSL. Boreas 43, 1-21 (2014). 805 Arnold, L.J., Bailey, R.M. & Tucker, G.E. Statistical treatment of fluvial dose distributions 325. 806 from southern Colorado arroyo deposits. Quaternary Geochronology 2, 162-167 (2007). 807 Srivastava, P., Singh, I.B., Sharma, M. & Singhvi, A.K. Luminescence chronometry and Late 326. 808 Quaternary geomorphic history stratigraphic record of the Ganga plains, India: A review. 809 Palaeogeography, Palaeoclimatology, Palaeoecology 197, 13-41 (2003). 810 Townsend, K.F.N., M.S.; Rittenour, T.M.; Pederson, J.L. Anatomy and evolution of a 327. 811 dynamic arroyo system, Kanab Creek, southern Utah, USA GSA Bulletin 131, 2094–2109 812 (2019). 813

814	328.	Shen, H., Yu, L., Zhang, H., Zhao, M. & Lai, Z. OSL and radiocarbon dating of flood
815		deposits and its paleoclimatic and archaeological implications in the Yihe River Basin, East
816		China. Ouaternary Geochronology <b>30</b> , 398-404 (2015).
817	329.	Jacobs, Z. Luminescence chronologies for coastal and marine sediments. <i>Boreas</i> 37, 508-535
818		(2008).
819	330.	Bateman, M.D. et al. The evolution of coastal barrier systems: a case study of the Middle-
820	220.	Late Pleistocene Wilderness barriers. South Africa <i>Quaternary Science Reviews</i> <b>30</b> , 63-81
020		(2010)
021	331	(2010). Tamura T et al Luminescence dating of heach ridges for characterizing multi-decadal to
022	551.	contemporal deltais shoreling shanges during Late Helesene. Makeng Diver delta, Maring
023 824		Geology <b>326-328</b> 140-153 (2012)
825	332	Murari MK Achyuthan H & Singhyi A K Luminescence studies on the sediments laid
826	552.	down by the December 2004 tsunami event: Prospects for the dating of palaeo tsunamis and
020 927		for the estimation of sediment fluxes. Current Science <b>92</b> , 367-371 (2007)
027	222	Curba D D at al Optical dating of electic deposite generated by an extreme marine coestal
020	555.	flood: The 1755 taynami denosits in the Algerva (Dertugal). Ougtarman Caechronology 5
829		100d: The 1755 isunann deposits in the Algarve (Politigal). Qualernary Geochronology 5,
830	224	529-555 (2010).
831	334.	Tamura, T., Sawai, Y. & Ito, K. OSL dating of the AD 869 Jogan tsunami deposit,
832	~~~	northeastern Japan. Quaternary Geochronology <b>30</b> , 294-298 (2015).
833	335.	Lukas, S., Preusser, F., Anselmetti, F.S. & Tinner, W. Testing the potential of luminescence
834		dating of high-alpine lake sediments. <i>Quaternary Geochronology</i> 8, 23-32 (2012).
835	336.	Fu, X., Cohen, T.J. & Arnold, L.J. Extending the record of lacustrine phases beyond the last
836		interglacial for Lake Eyre in central Australia using luminescence dating. Quaternary Science
837		<i>Reviews</i> <b>162</b> , 88-110 (2017).
838	337.	Kemp, C.W. et al. Climates of the last three interglacials in subtropical eastern Australia
839		inferred from wetland sediment geochemistry. Palaeogeography, Palaeoclimatology,
840		Palaeoecology <b>538</b> , 109463 (2020).
841	338.	Lewis, R. et al. Insights into subtropical Australian aridity from Welsby Lagoon, North
842		Stradbroke Island, over the past 80,000 years. Quaternary Science Reviews 234, 106262
843		(2020).
844	339.	Wallinga, J. & Bos, I.J. Optical dating of fluvio-deltaic clastic lake-fill sediments - A
845		feasibility study in the Holocene Rhine delta (western Netherlands). <i>Quaternary</i>
846		Geochronology <b>5</b> , 602-610 (2010).
847	340.	Kortekaas, M., Murray, A.S., Sandgren, P. & Biörck, S. OSL chronology for a sediment core
848	6.01	from the southern Baltic Sea: A continuous sedimentation record since deglaciation
849		Ouaternary Geochronology 2, 95-101 (2007).
850	341	Gutiérrez E et al Late Holocene evolution of playa lakes in the central sector of the Ebro
851	0111	Depression based on geophysical surveys and morpho-stratigraphic analysis of lacustrine
852		terraces Geomorphology <b>196</b> 177-197 (2013)
052	312	Long H. Shen I. Wang V. Gao I. & Freehen M. High resolution OSL dating of a late
000	J <del>4</del> 2.	Quaternary sequence from Vingkai Lake (NE Asia): Chronological challenge of the "MIS 3a
004		Maga palaolaka" hypothesis in China, Earth and Planatam Saianaa Lattars <b>428</b> , 281, 202
000		(2015)
856	242	(2015). Swith J. N. et al. Timing of laber level along from a deep last along it laber Missensley and ital.
857	343.	Smith, L.N. et al. Timing of lake-level changes for a deep last-glacial Lake Missoula: optical
858		dating of the Garden Guich area, Montana, USA. Quaternary Science Reviews 183, 23-35
859	244	(2018).
860	344.	Olley, J.M. et al. Optical dating of deep-sea sediments using single grains of quartz: a
861	215	comparison with radiocarbon. Sedimentary Geology 169, 175-189 (2004).
862	345.	Armitage, S.J. Optically stimulated luminescence dating of Ocean Drilling Program core
863		658B: Complications arising from authigenic uranium uptake and lateral sediment movement.
864		<i>Quaternary Geochronology</i> <b>30</b> , 270-274 (2015).
865	346.	Houmark-Nielsen, M. Testing OSL failures against a regional Weichselian glaciation
866		chronology from southern Scandinavia. Boreas 37, 660-677 (2008).

867	347.	Hu, G., Yi, CL., Zhang, JF., Liu, JH. & Jiang, T. Luminescence dating of glacial deposits
868		near the eastern Himalayan syntaxis using different grain-size fractions. Quaternary Science
869		<i>Reviews</i> <b>124</b> , 124-144 (2015).
870	348.	Ou, X. et al. Single grain optically stimulated luminescence dating of glacial sediments from
871		the Baiyu Valley, southeastern Tibet. Quaternary Geochronology 30, 314-319 (2015).
872	349.	Fuchs, M. & Owen, L.A. Luminescence dating of glacial and associated sediments: review,
873		recommendations and future directions. Boreas 37, 636-659 (2008).
874	350.	Alexanderson, H. & Murray, A.S. Luminescence signals from modern sediments in a
875		glaciated bay, NW Svalbard. Quaternary Geochronology 10, 250-256 (2012).
876	351.	Demuro, M., Froese, D.G., Arnold, L.J. & Roberts, R.G. Single-grain OSL dating of
877		glaciofluvial quartz constrains Reid glaciation in NW Canada to MIS 6. Quaternary Research
878		77, 305-316 (2012).
879	352.	Rowan, A.V. et al. Optically stimulated luminescence dating of glaciofluvial sediments on the
880		Canterbury Plains, South Island, New Zealand. <i>Quaternary Geochronology</i> <b>8</b> , 10-22 (2012).
881	353.	Thomas, P.J., Murray, A.S., Kjær, K.H., Funder, S. & Larsen, E. Optically Stimulated
882		Luminescence (OSL) dating of glacial sediments from Arctic Russia - depositional bleaching
883		and methodological aspects. Boreas 35, 587-599 (2006).
884	354.	Demuro, M. et al. Optically stimulated luminescence dating of single and multiple grains of
885		quartz from perennially-frozen loess in western Yukon Territory. Canada: Comparison with
886		radiocarbon chronologies for the late Pleistocene Dawson tephra. <i>Ouaternary Geochronology</i>
887		<b>3</b> , 346-364 (2008).
888	355.	Bateman, M.D., Murton, J.B. & Boulter, C. The source of De variability in periglacial sand
889		wedges: Depositional processes versus measurement issues. <i>Quaternary Geochronology</i> 5,
890		250-256 (2010).
891	356.	Liu, X.J. & Lai, Z.P. Optical dating of sand wedges and ice-wedge casts from Qinghai Lake
892		area on the northeastern Qinghai-Tibetan Plateau and its palaeoenvironmental implications.
893		Boreas 42, 333-341 (2013).
894	357.	Arnold, L.J. et al. Optical dating of perennially frozen deposits associated with preserved
895		ancient plant and animal DNA in north-central Siberia. <i>Ouaternary Geochronology</i> <b>3</b> , 114-
896		2 = 136 (2008).
897	358.	Fuchs, M. & Lang, A. Luminescence dating of hillslope deposits-A review. <i>Geomorphology</i>
898		<b>109</b> , 17-26 (2009).
899	359.	Guerrero, J. et al. Landslide-dam paleolakes in the central Pyrenees, Upper Gállego River
900		Valley, NE Spain: Timing and relationship with deglaciation. <i>Landslides</i> 15, 1975-1989
901		(2018).
902	360.	Völkel, J., Murray, A., Leopold, M. & Hürkamp, K. Colluvial filling of a glacial bypass
903		channel near the Chiemsee (Stöttham) and its function as geoarchive. Zeitschrift für
904		Geomorphologie <b>56</b> , 371-386 (2012).
905	361.	Arnold, L.J., Demuro, M., Navazo Ruiz, M., Benito-Calvo, A. & Pérez-González, A. OSL
906		dating of the Middle Palaeolithic Hotel California site, Sierra de Atapuerca, north-central
907		Spain. Boreas 42, 285-305 (2013).
908	362.	Li, ZY. et al. Late Pleistocene archaic human crania from Xuchang, China. Science 355,
909		969–972 (2017).
910	363.	Feathers, J.K., Rhodes, E.J., Huot, S. & Mcavoy, J.M. Luminescence dating of sand deposits
911		related to late Pleistocene human occupation at the Cactus Hill Site, Virginia, USA.
912		Quaternary Geochronology 1, 167-187 (2006).
913	364.	Froese, D.G. et al. Fossil and genomic evidence constrains the timing of bison arrival in North
914		America. Proceedings of the National Academy of Sciences 114, 3457-3462 (2017).
915	365.	Méndez-Quintas, E. et al. First evidence of an extensive Acheulean large cutting tool
916		accumulation in Europe from Porto Maior (Galicia, Spain). Scientific Reports 8, 3082 (2018).
917	366.	Susino, G.J. Optical dating and microwaste – archaeological applications. <i>Quaternary</i>
918		Geochronology 5, 306-310 (2010).
919	367.	Bate, S. et al. Pottery versus sediment: Optically stimulated luminescence dating of the
920		Neolithic Vinča culture, Serbia. Quaternary International 429, 45-53 (2017).
		- · · · · · · · · · · · · · · · · · · ·

368. Bailiff, I.K. Methodological developments in the luminescence dating of brick from English 921 late-medieval and post-medieval buildings. Archaeometry 49, 827-851 (2007). 922 Urbanová, P. et al. A novel interdisciplinary approach for building archaeology: The 369. 923 integration of mortar "single grain" luminescence dating into archaeological research, the 924 example of Saint Seurin Basilica, Bordeaux. Journal of Archaeological Science: Reports 20, 925 307-323 (2018). 926 370. Feathers, J.K., Johnson, J. & Kembel, S.R. Luminescence dating of Monumental stone 927 architecture at Chavín de Huántar, Perú. Journal of Archaeological Method and Theory 15, 928 929 266-296 (2008). 371. Johnson, M.O. et al. Quantifying the rate and depth dependence of bioturbation based on 930 optically-stimulated luminescence (OSL) dates and meteoric 10Be. Earth Surface Processes 931 and Landforms 39, 1188-1196 (2014). 932 372. Furbish, D.J. et al. Soil Particle Transport and Mixing Near a Hillslope Crest: 2. Cosmogenic 933 Nuclide and Optically Stimulated Luminescence Tracers. Journal of Geophysical Research: 934 Earth Surface 123, 1078-1093 (2018). 935 Liu, J.F. et al. OSL and AMS Dating of the Penultimate Earthquake at the Leigu Trench along 373. 936 937 the Beichuan Fault, Longmen Shan, in the Northeast Margin of the Tibetan Plateau. Bulletin of the Seismological Society of America 100, 2681-2688 (2010). 938 Yang, H.L., Chen, J., Thompson, J.A. & Liu, J.F. Optical dating of the 12 May 2008, Ms 8.0 374. 939 Wenchuan earthquake-related sediments: Tests of zeroing assumptions. Quaternary 940 Geochronology 10, 273-279 (2012). 941 375. Torabi, M., Fattahi, M., Amini, H., Ghassemi, M.R. & Karimi, N. OSL dating of landslide-942 dammed-lake deposits in the North of Tehran, Iran: 958 Ray-Taleghan/Ruyan earthquake. 943 *Ouaternary International* In press. 944 Rizza, M. et al. Using luminescence dating of coarse matrix material to estimate the slip rate 945 376. of the Astaneh fault, Iran. Quaternary Geochronology 6, 390-406 (2011). 946 377. Chen, Y., Li, S.-H., Sun, J. & Fu, B. OSL dating of offset streams across the Altyn Tagh 947 Fault: Channel deflection, loess deposition and implication for the slip rate. Tectonophysics 948 594, 182-194 (2013). 949 378. Gong, Z.J., Li, S.-H. & Li, B. Late Quaternary faulting on the Manas and Hutubi reverse 950 faults in the northern foreland basin of Tian Shan, China. Earth and Planetary Science Letters 951 424, 212-225 (2015). 952 379. Stockmeyer, J.M. et al. Active thrust sheet deformation over multiple rupture cycles: a 953 quantitative basis for relating terrace folds to fault slip rates. Geological Society of America 954 Bulletin 129, 1337-1356 (2017). 955 380. Peltzer, G. et al. Stable Rate of Slip Along the Karakax Section of the Altyn Tagh Fault from 956 Observation of Interglacial and Postglacial Offset Morphology and Surface Dating. Journal of 957 Geophysical Research: Solid Earth 125, e2019JB018893 (2020). 958 381. Sohbati, R., Borella, J., Murray, A., Quigley, M. & Buylaert, J.-P. Optical dating of loessic 959 hillslope sediments constrains timing of prehistoric rockfalls, Christchurch, New Zealand. 960 Journal of Quaternary Science 31, 678-690 (2016). 961 Preusser, F., Rufer, D. & Schreurs, G. Direct dating of Ouaternary phreatic maar eruptions. 382. 962 Geology 39, 1135-1138 (2011). 963 Rittenour, T.M., Riggs, N.R. & Kennedy, L.E. Application of single-grain OSL to quartz 383. 964 xenocrysts within a basalt flow, San Francisco volcanic field, northern Arizona, USA. 965 Quaternary Geochronology 10, 300-307 (2012). 966 Liu, Z., Zhao, H., Wang, C.M. & Li, S.H. Estimation of paleo-firing temperatures using 384. 967 luminescence signals for the volcanic lava baked layer in Datong, China. Quaternary 968 Geochronology 30, 363-368 (2015). 969 385. Westgate, J.A. et al. Changing ideas on the identity and stratigraphic significance of the 970 Sheep Creek tephra beds in Alaska and the Yukon Territory, northwestern North America. 971 Quaternary International 178, 183-209 (2008). 972 Anechitei-Deacu, V., Timar-Gabor, A., Fitzsimmons, K.E., Veres, D. & Hambach, U. Multi-386. 973 method luminescence investigations on quartz grains of different sizes extracted from a loess 974
section in southeast Romania interbedding the campanian ignimbrite ash layer. 975 *Geochronometria* **41**, 1-14 (2014). 976 387. Herman, F., Rhodes, E.J., Braun, J. & Heiniger, L. Uniform erosion rates and relief amplitude 977 during glacial cycles in the Southern Alps of New Zealand, as revealed from OSL-978 thermochronology. Earth and Planetary Science Letters 297, 183-189 (2010). 979 388. Guralnik, B. et al. OSL-thermochronometry using bedrock quartz: A note of caution. 980 Quaternary Geochronology 25, 37-48 (2015). 981 982

# **Supplementary Information**

## (Section numbering corresponds to the relevant section number in the main text)

# 2. Experimentation

# (a) Sampling

Luminescence dating obviously starts in the field, where the sample containing quartz grains - usually sediment - must be collected. Since OSL dates the last exposure of sediment to daylight, one must avoid light exposure during sampling. The surface of the sediment, or of the sedimentary sequence of interest, is first cleaned by scraping back. This is not only to avoid the light exposed first few mm of the exposure, but also to provide a clear view of the stratigraphy and so avoid locations that may have been affected by post-depositional disturbances (ice wedges, animal burrows, roots, etc.). At least the first few cm must be removed; it is better to cut a section back by > 1 m to avoid air-exposed material and so to take samples where oxidation/reduction conditions and moisture content are more likely to be representative of the long term burial (see Section 2.1 on dose rates). All these operations can be done in full daylight and taking photographs of sampling site (overview) and sampling locations (detail) is recommended.

There are then two approaches to collecting a sediment sample: the simplest is to hammer tubes (metal or plastic, depending on the deposit) into the cleaned section; these are often intended for one-time use, but may be reusable [1]. This method is only suitable when the unit to be sampled is of sufficient thickness that the tube will not cross sedimentological boundaries. Ideally the gamma dose rate gradient across the tube should also be small; if possible, the tube should be at least 15 cm from any boundary. A plug of wadded paper or crumpled plastic bag should be placed in the tube, at the end closest to the section. This will support the sediment as it enters the tube and reduce the risk of it collapsing and mixing exposed and unexposed material. The tube must be filled completely, either with sample or by packing with plastic, to ensure that the material cannot move and mix during transport. In the laboratory the potentially light-exposed outer ~2-3 cm of each end of the tube is retained for radionuclide concentration measurements, and any other sedimentological analyses (e.g. grain size, mineralogy, geochemistry, ancient DNA, tephrochronology, etc.). The inner part of the tube is used for OSL measurements, and possibly for water content measurements.

If the unit is too thin to be sampled with a tube, then the second approach is to operate in the dark – either at night (moonlight is very low intensity, and will not significantly affect the OSL during the

short sampling exposure, see [2]), or under an opaque covering such as a tarpaulin. In both cases, it is safe to use a controlled light source, such as headlamp emitting dim orange light, or a red LED (e.g. bicycle rear light). It is usual to collect two parts for each sample: one mainly for OSL analysis and the other mainly for radiochemical analysis. The previously cleaned surface is first scraped to remove a further cm or so. This part is likely to have been exposed to daylight after the cleaning of the section, and is not suitable for OSL measurement; it is removed and retained for measurement of the radioelement concentrations and for any complementary measurements. Since exposure to light is unimportant for such measurements, this part of the sample may be collected in transparent bags. The second part of the sample, taken from further into the section, must be protected from light exposure, either by packing into metal containers, or fully opaque plastic bags. Usually a few hundred grams is sufficient – but that of course depends on the concentration and grain-size distribution of quartz grains in the sample. A carbonate-rich sandstone will generally yield less quartz than a desert sand, and a marine/lake core fewer sand-sized grains than a beach sand. If a narrow-grain-size fraction is to be targeted (e.g. for single-grain OSL), the amount of sediment required may be greater than usual.

If the target is ceramic material (pottery, bricks, tiles) or cobbles, heated stones etc., all sampling can be undertaken in daylight, although direct sunlight should be avoided. This is because, in the case of ceramics and heated stones, the potentially light-exposed surface of the sample is discarded in the laboratory, and in the case of cobbles, light is attenuated so rapidly that a few minutes of daylight does not affect the average luminescence signal in the outer 1 mm.

During sampling one must also consider the water content history of the site and the collection of (separate) samples for the present day water content. A subsample of the OSL samples (from inside tube) or the transparent bags (see above) can be used to determine this and the saturated water content (see in section 2.1.2 below). In order to retain as much moisture as possible sealable bags should be used and the ends of the tubes covered with plastic and sealed with tape.

Finally, sampling is the occasion to collect information relevant for calculating cosmic dose rates (Figure S1) by measuring or estimating the overburden of a sample. This includes the nature of the overburden (e.g. cave roof, sediment, etc.), the density and above all the thickness of the material covering every sample. The elevation of the site above sea level and geographical coordinates (latitude, longitude) should also be recorded. These field observations are used to calculate the cosmic dose rate [3]. Furthermore, in some cases (see section S2.1 below) where the sediment is heterogeneous on a scale of 10-50 cm (presence of rocks, sediment unconformity, boundary between sediment and air, or sediment and bedrock, etc.) it may be necessary to measure in situ gamma dose rates. It is often also possible to collect two separate dose rate samples (one from sediment to be dated and one from the adjacent rock/sediment causing heterogeneity), measure the radionuclide concentrations for both samples and then model the gamma dose rate (see Appendix H in [4]).

#### (b) Sample preparation

In the laboratory, that part of the sample intended for dose rate measurements is homogenised (usually by grinding) so that even a small sample of less than few grams is representative of the whole. This part may be from the ends of a sampling tube, or the outer few mm of a ceramic or heated stone. Section S2.1 lists typical methods for dose rate determination. It is good practice to keep a portion of the non-light-exposed material as back-up in case something goes wrong in sample preparation. The remaining part of the sample intended for OSL measurements undergoes several treatment steps to extract quartz grains (see Figure S2). The unexposed inner part of ceramics and heated stones is gently crushed. Then the crushed material, or the sediment, is wet sieved - a constrained grain size fraction is of importance not only because of measurement practicalities, but also because both laboratory and environmental dose rates are, to some degree, grain-size dependent - see section S2.1 below. The factors driving the selection of grain size are: (i) the abundance of that fraction in the samples; (ii) the ability to chemically etch the outer surface of the grains to remove the alpha-irradiated component (typically not smaller than fine sand, i.e. > 63  $\mu$ m – grains must be large enough, or else nothing is left after the HF treatment described below); (iii) ease of manipulation of the grains. Generally, some choice is made within the range  $63-300 \text{ }\mu\text{m}$  (e.g.  $90-125 \text{ }\mu\text{m}$  or  $180-250 \text{ }\mu\text{m}$ ); the fraction  $150-180 \text{ }\mu\text{m}$ is most suitable for measurements using single-grain discs with e.g.  $\emptyset$  200 µm holes.

The sample should remain at or close to room temperature in all the steps outlined below, to avoid the risk of thermal fading of the OSL signal; this applies particularly to the HF digestions steps. The selected size fraction is treated with 10% HCl for at least 30 minutes (until no reaction is visible) to remove carbonates - these may impede the reaction with HF - and with 10% H<sub>2</sub>O<sub>2</sub> (for up to a few days) to remove finely disseminated organics. After HCl and H<sub>2</sub>O<sub>2</sub> treatments, the grain fractions are washed with water (usually 2-3 times). Note that it is usually easier to soak more compact fine-grained (clayey-silty) sediment in water and treat with HCl, H<sub>2</sub>O<sub>2</sub> first, to disaggregate the grains, before wet sieving. This is also true for carbonate-rich sandstones (e.g. aeolianites).

After sieving and treating with HCl and H<sub>2</sub>O<sub>2</sub>, there should be at least a few gram(s) of sample, at this point mainly a mixture of quartz, feldspar and possibly heavy-mineral grains in the chosen grain size fraction. If only quartz is desired, the sample may be placed directly in concentrated HF (40%); usually  $\sim$ 1 hour is sufficient to remove feldspar and etch the surface of the quartz grains (see below). If the feldspar grains are also required, then prior mineral separation is necessary (K-feldspars in particular are commonly used in other approaches to luminescence dating). Prior to mineral separation the mixed quartz/feldspar fraction can be etched for up to 40 min in 10% HF to remove surface coatings (e.g. iron oxides) and attached clay grains, washed in 10% HCl for  $\sim$ 20 mins, and then with water and dried. This step improves recovery in the following separation step. Density separation using aqueous heavy liquids (e.g. lithium heteropolytungstate, or LST) is a safe and convenient

approach; a density of 2.70 g.cm<sup>-3</sup> allows the heavy minerals to sink; 2.62 g.cm<sup>-3</sup> allows a quartz-rich fraction to sink; 2.58 g.cm<sup>-3</sup> allows a Na-rich feldspar fraction to sink, and floats mainly K-rich feldspar grains. Some laboratories use only a single 2.58 g.cm<sup>-3</sup> step, and rely on HF to remove any denser feldspars that remain in the quartz-rich fraction. Any heavy minerals surviving the HF treatment rarely, if ever, make a significant contribution to the quartz luminescence signal. Magnetic separation is also used to separate quartz and feldspar grains [5].

Finally, treatment with concentrated HF (40%) for 1 hour removes any remaining feldspar, and ~10  $\mu$ m from the outer surface of the quartz grains, thus removing most of the quartz layer exposed to alpha particles from the surrounding sedimentary matrix during burial. The internal alpha activity of quartz is very small, and so this step significantly simplifies the dosimetry calculations. Some laboratories then soak the sample in 10% HCl for several hours to remove any fluorides that might have precipitated on the grain surfaces, because some fluoride salts are luminescent. Although there is no evidence that these amorphous precipitates are in fact a significantly attenuate the OSL signal. Finally, the samples are washed (typically 3 times) with water and dried; it may be desirable then to sieve the samples again, to define the minimum grain size during beta irradiation, and, for single grain measurements, to avoid the risk of multiple grains in each hole in the sample holder. The quartz is then tested for signal purity (Section S2.2.1(a)); if it fails this test, the HF etch can be repeated (the additional mass loss will have a small effect on the average natural beta dose to the grains).

### 2.1. Dose Rate

#### 2.1.1. Basic concepts

## (a) Dose rate calculation using the infinite matrix assumption.

Various methods for determining the dose rate are summarised in Table S1. Concentrations of radionuclides may be expressed in activity concentrations [Bq.kg<sup>-1</sup>] or mass concentrations. To calculate the activity concentration from a mass concentration ( $C_0$ , [g.kg<sup>-1</sup>]) measurement, the atom concentration per unit mass of sediment (N, [kg<sup>-1</sup>]) is first calculated using N=C<sub>0</sub> [g.kg<sup>-1</sup>]/ Z (atomic weight [g.mol<sup>-1</sup>]) x N<sub>A</sub> (Avogadro's number [mol<sup>-1</sup>]). N is then multiplied by the relevant decay constant ( $\lambda$ , [s<sup>-1</sup>]) to give the radionuclide activity concentration. Measured concentrations may be reported in ppm [µg.g<sup>-1</sup>] for U and Th, and percentage [%] for K; these must first be converted to [g.kg<sup>-1</sup>]. In the case of K, the mass concentration of <sup>40</sup>K must then be derived using the isotopic abundance (0.0117%).

Finally, for each nuclide, multiplying the activity concentration by the average energy emitted per disintegration (see e.g. <u>https://www.nndc.bnl.gov/nudat2/</u> given in MeV, 1 MeV=1.6×10<sup>-13</sup> J;) gives

what is known as the infinite matrix dose rate. Relevant conversion factors ( $C_0 \times N_A \times \lambda/Z$ ) (summarised in Table S2) are regularly compiled and updated (e.g. [6]). Table S3 uses these data to derive infinite matrix dose rates from a sand containing representative concentrations of <sup>40</sup>K, <sup>238</sup>U and <sup>232</sup>Th. Figure S3 shows the <sup>238</sup>U and <sup>232</sup>Th radioactive decay chains, and Figure S4 groups radionuclides according to their potential mobility in sediment, and presents the relative contribution of each group to the total dose rate, assuming the relative activity concentrations of Table 3. Figure S5 summarises the potential variability in <sup>40</sup>K, <sup>238</sup>U and <sup>232</sup>Th concentrations, and their relative contributions to dose rates. Figure S6b) illustrates how variable beta dose rates can be at the 100 µm scale.

#### (b) In situ gamma dose rate measurements

In some cases, sediment is heterogeneous at the scale of the penetration distance of gamma rays ( $\sim$ 30-50 cm, see Figure S6a), e.g. if it is poorly sorted, and includes pebble and cobbles, or is adjacent to a rock wall. Then the infinite matrix gamma dose rate may not be representative of the gamma dose rate to which the sample was exposed during burial. As an alternative, the gamma dose rate may be measured in situ, at the sample location, e.g. with luminescent Al<sub>2</sub>O<sub>3</sub>:C pellets. Aluminium oxide is very similar to quartz in terms of dose absorption characteristics, and doping with carbon gives this material a very sensitive luminescence response to dose [7]. In practice, pellets are left buried in an aluminium tube of  $\sim$ 2-3 mm thickness (to prevent beta particles from reaching the pellets) for 6-12 months, after which the absorbed dose is sufficient to induce a measurable OSL signal [8], [9]. This directly measured dose rate is the sum of gamma and cosmic dose rates, as affected by escape of <sup>222</sup>Rn and the water content of the sediment over the period of measurement. The other common way of determining in situ gamma dose rates involves the use of a field gamma spectrometer, typically with a NaI(TI) or LaBr probe. Calibration involves measurements in infinite media with known radionuclide concentrations [10]. Data analysis may consist of estimating effective K, U and Th concentrations using the 'windows technique' (see Appendix L in [4]), and then converting these concentrations into effective infinite matrix dose rates. The concentration of K is based on the emission rate of the 1461 keV peak, of U of the 1764 keV peak and of Th of the 2615 keV (e.g. [11]). Otherwise, one may use the proportionality between gamma dose rate and the count rate above a certain energy channel, in what is called the threshold technique [12], [13]. The advantage of the threshold technique lies in the inclusion of a greater portion of the recorded spectra to estimate the gamma dose rates, thereby reducing the uncertainties for a given counting time (typically several minutes), but this approach does not give any nuclide-specific information.

# 2.1.2. Practicalities

*Water content measurements* – Constraining the average water content of a deposit is very important to the accuracy of a luminescence age, because a 1% increase/decrease in life-time average

water content typically leads to a 1% increase/decrease in derived age<sup>1</sup>. Well-sorted sands have only a small variation in porosity, from fully (~38%) to loosely (46%) compacted [14], and because the grain surfaces are in physical contact (in contrast to e.g. clay-rich deposits) it is very difficult to overcompress sand by increased overburden pressure. When fully saturated with water, 42% porosity corresponds to 21% by weight water content. Because the packing density is reproducible, a saturation water content can be measured for a sandy deposit by adding sediment and excess water to say, a preweighed 100 ml syringe and applying pressure to remove the excess water. The point at which the syringe plunger stops moving is well-defined, and increasing the pressure does not extrude more water. The syringe and wet sand are weighed to obtain the saturated weight, and then placed in an oven and dried to obtain the dry weight – the ratio of the weight loss of the saturated sample during drying to the dry weight is the saturation water content. If the sediment was likely to have been below the ground-water table throughout the burial period, this is the lifetime average water content. Otherwise, the likely fraction of saturation during the burial period must be estimated from a knowledge of the local hydrology (drainage, precipitation, etc.) and sediment characteristics [15]. The observed water content (*i.e.* the water content in a field sample, measured simply by drying a known weight of wet material) can often be taken as a lower limit to lifetime water content, since typical sampling sites (cleaned sections, gully walls, river or road cuts etc.) are usually significantly drier than corresponding material a few metres deeper into the section. In the worst case, where no further constraints can be placed on water content history, the mid-point between field and saturated water content can be chosen, and an estimate of uncertainty adopted, equal to 25% of the range. Then all possible values are covered by  $\pm 2$  times the uncertainty. For example a measured water content of, say, 9% (cf. saturation water content of 21%) would give a mean lifetime water content of  $15\pm3\%$  by weight, implying an overall contribution to the uncertainty on the age of  $\sim 3\%$  [16]. This is not large compared to the expected minimum uncertainty of ~5% (uncertainties added in quadrature, see Table S8), and so is not of major concern. Similar arguments can be used to constrain the effects of the infilling of porosity by carbonate.

The water content of a ceramic buried in a sediment is more complicated. The porosity of a ceramic will differ from that of the sediment matrix, but it is readily measured by simply saturating the ceramic in water, weighing, drying and weighing again. Then it is assumed that the fraction of saturation that applies to the matrix also applies to the ceramic [4]. The lifetime average water content estimated for the matrix is applied to the gamma dose rate, and that for the ceramic to the beta dose rate.

<sup>&</sup>lt;sup>1</sup> The age calculation spreadsheet attached to this paper allows the reader to test the sensitivity of the ages to various parameters.

# 2.2. Testing the applicability of a SAR protocol

#### 2.2.1. Dose estimation using SAR

#### (a) Testing for quartz purity

The chemical pretreatment intended to remove any feldspar contamination is not always completely successful [17, 18]. The reasons for this are unclear, but presumably involve occluded feldspar grains, or even feldspathic-like impurities within individual quartz grains. This is a problem because the luminescence from feldspar impurities is significantly unstable, and much more difficult to bleach. Unfortunately, feldspar is sensitive to the wavelengths used to stimulate quartz (usually 450 to 530 nm), and so can contaminate the quartz signal. If additional etching with HF does not remove residual signal contamination, then instrumental separation must be considered. For stimulation with blue or green wavelengths, the decay rate of the feldspar signal is considerably slower than that of the quartz fast component, and so the use of the early background subtraction approach helps to reduce the importance of any feldspar contamination. But, feldspar, in contrast to quartz [19], is sensitive to infrared (IR) stimulation. This characteristic can be used to test for, and to reduce, contamination of the quartz OSL by inserting an IR stimulation immediately prior to the measurement of  $L_i$  (but not  $T_i$ ) in an additional recycling ratio test [20]. If this  $(L_i/T_i)/(L_i/T_i)$  ratio is indistinguishable from that measured without IR (see section 2.2.2 in main text), then feldspar does not contribute significantly to the quartz signal. In samples where the feldspar contamination is of significance, it can be reduced by inserting the IR stimulation step in front of every L and T measurement [21], although this is expensive in measurement time. There are also other instrumental methods for minimising any feldspar contribution, but these are not in routine use [22, 23, 24].

## (b) Instrumentation for determining the $D_e$

The instrumentation for determining the absorbed dose is relatively simple and comprises four main components: 1) luminescence detector, 2) stimulation light source, 3) source of ionising radiation and 4) heating system. **Error! Reference source not found.** summarises the relationship of these four components to the quartz grains to be measured.

The main challenge in designing such a system is the instrumental sensitivity. The ability to measure an absorbed dose depends both on the sensitivity of the sample (i.e. the number of photons emitted per unit mass [kg<sup>-1</sup>] and per unit absorbed dose [kg<sup>-1</sup>.Gy<sup>-1</sup>]), on the instrument sensitivity, i.e. the fraction of the emitted photons that are actually detected by the instrument (detection efficiency), and the relationship of this signal to the instrument background noise. Detection efficiency depends on the light path (controlling the fraction of the emitted signal reaching the detector, and dependent on geometry, mirrors, lenses and filters), the choice of photon detector (how many photons arriving at the detector are required to give a detected event – the quantum efficiency) and the wavelength and intensity of the stimulation light source (the more rapidly the charge is released (detrapped), the stronger the initial luminescence signal compared to background. For a given detection wavelength, the electron detrapping rate depends on the wavelength-dependent photoionization cross-section and the power density of the stimulation light).

The luminescence detector consists of a photon detector and detection filter(s). Natural phosphors such as quartz are relatively inefficient in converting absorbed dose into luminescence and thus the emitted luminescence signal is usually too weak to be detected by the naked eye. To measure such signals precisely, a highly sensitive light detector with a low noise (background) level and good light collection are essential. The latter requires as large as possible solid angle subtended by the detector surface at the sample – this is usually achieved by as short as possible distance between sample and detector, or by focussing through lenses. In OSL the luminescence signal is emitted as a consequence of optical stimulation, which is ~10<sup>15</sup> times more intense than the OSL signal itself. OSL is an anti-Stokes emission, i.e. the energy of the optical stimulation is less than that of the emitted luminescence. Appropriate detection filters must be used both to separate the luminescence from the stimulation light and to define the spectral detection window.

**Photon detector** - Figure S8 shows the emission spectrum from quartz obtained by stimulation with 647 nm laser light. Quartz has a relatively broad OSL emission peaking at ~360 nm (full width half maximum, FWHM ~80 nm). Photomultiplier tubes (PMT) operating in photon-counting mode, sensitive in the blue/UV range are most commonly used for detecting quartz OSL, mainly because of their high sensitivity, large dynamic range and low dark count rate. A high signal-to-noise ratio is particularly important when measuring insensitive samples and/or samples having absorbed small radiation doses (i.e. young samples). The quantum efficiency (the effectiveness of the PMT at converting photons into electrons) is typically up to ~30 %, depending on the wavelength of the luminescence. The only major drawback of using PMTs is that they do not provide information on the spatial variation of the emitted luminescence. In applications where such spatial information is of importance (e.g. rock surface luminescence dating) electron-multiplier charge-coupled devices (EMCCD) can be used, although they are in general less sensitive than PMTs (e.g. [25], [26], [27]).

**Detection filters** - The use of detection filters is important both to define the wavelength window of interest and to separate the emitted luminescence from scattered stimulation light. In quartz OSL dating, the UV/blue emission is usually detected through a broadband Hoya U-340 glass filter, which has its peak transmission at 340 nm with a FWHM of 80 nm (see Figure S9). The main emission from quartz is centred at 365 nm and thus, in principle, a significant improvement in light detection is available by choosing a detection filter closer to the main emission of quartz. However, moving the

detection window closer to the stimulation wavelength (usually 470 nm, see below) is likely to lead to a significant increase in background signal (from breakthrough of the stimulation light) and hence the signal-to-noise ratio is not necessarily improved [28].

Stimulation light source – In quartz OSL, the electron detrapping rate depends both on the intensity and the wavelength of the optical stimulation light (see Figure S10; [29, 30]); the more intense and the shorter the wavelength the stronger the OSL signal per unit time. For most OSL applications, optical stimulation is at a constant light intensity (continuous wave mode), but modulating the intensity of the stimulation light (e.g. linear modulated OSL, LM-OSL; [31]) can be used to visually separate the traps involved in the OSL production. In some applications, pulsing the stimulation light and only detecting in between short (~50  $\mu$ s) bursts of stimulation light (Pulsed OSL, POSL) can help to separate quartz and feldspar OSL signals instrumentally [23]. POSL stimulation can also be used to gain insight into the physics of recombination processes (e.g. [32]).

Optical stimulation of quartz can be achieved using any visible light source such as filtered lamp systems [28], lasers [33, 34, 26], light emitting diodes (LEDs) [35] and laser diodes [36, 26]. In most quartz multi-grain OSL dating applications, blue (450-470 nm) or green (~525 nm) LEDs with power densities at the sample position ranging between ~40 and 100 mW/cm<sup>2</sup> are used, resulting in the decay of the fast OSL component in a few seconds. As LEDs have a broad emission (FWHM of ~30 nm) it is often necessary to stimulate through cut-off filters to ensure an effective separation of stimulation light and detection windows ( [37], see Figure S9). Higher energy light sources (e.g. 405 nm laser diode [38]) can be used to access deeper traps insensitive to blue light. In single-grain applications, stimulation by a green (532 nm) solid state laser providing a power density of ~50 W/cm<sup>2</sup> is commonly used [39].

*Source of ionising radiation* - The measured natural OSL signal must be calibrated in terms of absorbed dose. As different aliquots can have very different luminescence sensitivities, it is necessary to calibrate aliquots individually. This calibration is performed by comparing the natural OSL signal to that arising from a known laboratory dose given to each aliquots (using a SAR protocol, Section 2.2 in main text). This known dose can be given by any source of ionising radiation, but for convenience this is usually a beta source containing the artificial radionuclide <sup>90</sup>Sr ( $\tau_{\frac{1}{2}}$  28.8 years) and its short lived progeny <sup>90</sup>Y. This combination of emitters has a maximum beta energy of 2.27 MeV and a mean energy of ~600 keV, which makes it relatively easy to shield sufficiently for it to be safe to use.

The laboratory dose rate to a quartz sample depends on the activity of the source, the source/sample distance, sample grain size, and on the substrate material on which the grains sit. Because of this the source must be calibrated for each irradiation geometry (particularly for different substrates and grain-size range). The calibration is usually done by using a portion of quartz grains that has been given a

known dose using a standardised gamma source. An appropriate SAR protocol then aims at determining the duration of beta irradiation that provides a sensitivity-normalised luminescence intensity equal to that induced by the reference gamma dose [40]. Some laboratories make available sensitive quartz which has absorbed a well-known dose, so that others without access to a calibrated gamma source can calibrate their own beta source as required.

*Heating system* – to obtain accurate measurements of  $D_e$ , it is important to perform reproducible thermal pre-treatments and to be able to keep the aliquot at a given temperature during optical stimulation. OSL dating systems will usually heat up to 700 °C using heating rates typically ranging between 0.01 and 10 °C /s, although temperatures above 350 °C are rarely used.

# 4. Reproducibility and data deposition

(a) *Data reporting:* It is common in the literature to read articles describing equivalent dose determination in detail, while dose rate data are often summarised in a way that renders recalculation or reproducibility studies difficult. Until recently most articles containing luminescence ages presented new data (with variable degree of detail). However, publications addressing larger scale questions by presenting meta-analyses of published datasets are becoming more frequent. To encourage this, detailed information behind the OSL ages is required, both to allow the assessment of the quality of the data, and to ensure that the same underlying assumptions are adopted, and so enable the derivation of a coherent data set. Bayesian modelling of OSL ages under stratigraphic constraints may require a combination of raw measurement files and details of calibration uncertainties, both for equivalent dose and dose rate measurements [41], [42]. Here we discuss data reporting in terms of good practice, in the sense that it provides sufficient information for later comparisons to be made between studies that focus on the same research question – and thus, on the same samples, stratigraphic records or dated events.

When publishing OSL results, it should be made clear to the reader whether the signal is fast component-dominated (perhaps with a figure showing a comparison with known fast-component dominated material such as calibration quartz [40]), and that the interpolation is not compromised by saturation effects (e.g. using an illustrative dose response curve, see Figure 4(c,f)). Although recuperation and recycling are important (and can be shown on the dose response curve), it is most important to document the dose or light recovery ratio for the material under consideration (Section 2.2.2(vi)). In general, it is not necessary to give a value for each sample from the same sedimentary suite, a single average with standard error and number of measurements (as in Figure 6(b)) is usually sufficient.

In presenting dose rates, the methods adopted should be presented first, starting with the type of instruments and experimental conditions (Table S1). Second, we recommend that the measured values are reported: for example, if high resolution gamma spectrometry has been used, then concentrations in K, U and Th should be given; if a beta counting system is used, then count-rates should be given. Reference to the calibration standards and procedures is of course desirable – laboratories commonly publish their protocols, e.g. [43], [44], [45].

The source of the conversion factors is important: in the two examples above, conversion is from (activity) concentrations to dose rates and from count rates to dose rates, for gamma spectrometry and beta counting, respectively (Table S2). The reader should also be able to find (i) which factors were used to account for grain size attenuation of beta dose rates (e.g. Table S4 below taken from [46]); (ii) if alpha dose rates were taken into account (and if so, which alpha dose rate efficiency factors were used); (iii) what water content value was used, how it was determined and which factors were used to account for the effect of water on dose rates (see Tables S5 and S6, and e.g. [47], [48], [16]); (iv) if an internal dose rate was considered - and if so, if it was measured or simply assumed [49]; (v) geographical coordinates, altitude and burial depth, together with the derived cosmic dose rates<sup>2</sup> (unless gamma and cosmic dose rates were measured directly using e.g. Al<sub>2</sub>O<sub>3</sub>:C pellets). In some cases, sediment may be rich in organic matter or carbonates; these may have changed through time, perhaps by oxidation (organic material) or by the formation of secondary carbonates in pore spaces. This leads to complexities resulting from sediment chemical composition; since quartz is the dosimeter, differential absorption of dose in the surrounding non-quartz material renders the infinite matrix assumption invalid, or at least approximate. If the amount of organic or carbonate material is quantified, differential dose rate absorption coefficients may then be used [47], and reported. Table S5 below gives attenuation factors to account for the effect of water and carbonates in infinite matrix conditions, while Table S6 details variations of the water correction factors in well-sorted sediment as a function of grain size.

To illustrate our suggestions, we provide Table S7(a,b); this represents our view of the minimum numerical reporting that allows the transparency required. The various underlying assumptions and the factors used should be given in the main text or as footnotes, together with any other relevant data (particularly the dose recovery summary).

For the reader to follow the details of an age calculation, and understand how the various effects and parameters are taken into account in practice, an age calculation spreadsheet is provide as Supplementary Material. This is not proposed as a standard data processing option (there are many more sophisticated software packages available) but it does provide a transparent and practical

 $<sup>^{2}</sup>$  The age calculation spreadsheet provided here allows the calculation of cosmic dose rates as a function of latitude, altitude and burial depth following [3].

implementation of age calculation, parameterized functions to estimate cosmic dose rates and correction factors for the effect of water, grain size attenuation factors, etc.

(a) Uncertainty analysis of luminescence data follows guidelines such as those of the Bureau International des Poids et Mesures (the intergovernmental organisation through which Member States act together on matters related to measurement science and measurement standards). The Bureau's guide on the expression of uncertainty in measurement distinguishes between random and systematic uncertainties [50]. The measurement error is defined as the difference between the measured, or observed value, and the true value (measurand); it is unknown (since the true value is unknown) but it can be characterised by a probability density. Repetition of a measurement leads to random fluctuations of the observed values (and thus characterises random uncertainties), while the use of a given instrument, calibrated with a given standard, my lead to systematic bias in measurements. In other words, the scale of analysis dictates whether an uncertainty is random or systematic - for example, uncertainties that are systematic within each of several laboratories (e.g. calibration of a gamma spectrometer), will give rise to random fluctuations at the scale of an intercomparison study involving different laboratories (Table S8).

In both cases (systematic and random), provided everything has been done to reduce systematic uncertainties, both systematic and random errors may be assumed to follow a normal distribution. Table S8 lists the main parameters needed to calculate an OSL age (other than measurement uncertainties, which can, in principle, be reduced arbitrarily by simply making more measurements) and the typical standard deviation of the error, as well as comments on its nature (systematic or random).

Formally, one may write for all quantities at stake:

$$\tilde{x} = x + e$$

where  $\tilde{x}$  is the measurement or observed value, x the true value and e the measurement error. We then introduce  $\sigma$ , the standard deviation of e (and  $\tilde{x}$ ). For example, if we measure the equivalent dose on n aliquots, we generally approximate  $\sigma$  by the standard deviation of the D<sub>e</sub> values, divided by the square root of n. Many quantities must be determined and added, multiplied or divided to finally obtain an age; for uncertainty propagation, if z = x + y (or z = x - y), it is standard practice to approximate

$$\sigma_z$$
 by  $\sqrt{\sigma_x^2 + \sigma_y^2}$ . If  $z = y/x$  (or  $z = yx$ ), then  $\frac{\sigma_z}{z}$  is approximated by  $\sqrt{\left(\frac{\sigma_x}{x}\right)^2 + \left(\frac{\sigma_y}{y}\right)^2}$ .

Although there is no formal convention, OSL ages are almost exclusively reported with an uncertainty of one standard deviation of the error, i.e. the interval  $[A - \sigma_A; A + \sigma_A]$  is the 68% confidence interval.





Variation of cosmic ray dose rate with altitude



Figure S2. Schematic overview of the laboratory treatment to extract quartz grains from the sample. All these steps should be performed under subdued lighting conditions (using light of wavelength and intensity without a significant effect on the trapped charge population [51]). Q: quartz, H: heavy minerals, F: feldspar, NF: sodium-rich feldspar, KF: potassium-rich feldspar. The combined water drops [ $\checkmark$ ] and fan symbol [ $\circledast$ ] indicate thorough washing and drying.



Figure S3. Radioactive decay chains of <sup>238</sup>U (left) and <sup>232</sup>Th (right). Long-lived radioisotopes of the same element are marked with rings of one colour. In secular equilibrium, the activity of each radionuclide is almost identical and follows the parent decay rate. While the <sup>232</sup>Th chain is unlikely to suffer from disequilibria (because of the short half lives of all of its progeny), the <sup>238</sup>U chain may be affected by <sup>222</sup>Rn escape (<sup>222</sup>Rn is a gas and the half-life of several days makes it possible for it to move before decaying), or by loss or gain of <sup>226</sup>Ra and <sup>238</sup>U – both elements can be mobilised or deposited by changes in porewater geochemistry (e.g. salinity, dissolved carbonate, reduction/oxidation conditions).



Figure S4. The radionuclides in the decay series are combined in groups reflecting the presumed potential for disequilibria based on half-lives and physico-chemical parameters (nuclides within each group can be deduced by comparison with Figure S3). The relative contribution of these groups to the total dose rate for a sand sample with the activities given in Table S2 is shown as %; these values can give some indication of the potential importance of radioactive disequilibria (bearing in mind that this is most likely to occur in the U series). Unless sediment is unusually rich in U (relative to K and Th), the contribution of the U series to the total dose rate is sufficiently small for most samples to make unlikely errors of more than 5% in the final age arising from disequilibrium.



Figure S5 (reproduced from [52]). Variability in radionuclide concentrations in 3578 samples used for luminescence dating. Left – histograms of activity concentrations of  $^{238}$ U (a),  $^{226}$ Ra (b),  $^{232}$ Th (c) and  $^{40}$ K (d). Right – relative contributions to beta (a) and gamma (b) dose rates from  $^{238}$ U,  $^{232}$ Th and  $^{40}$ K. The table summarises the concentration values.



Mode, percentiles and median for the distributions of concentrations of  $^{238}\rm{U},$   $^{226}\rm{Ra},$   $^{232}\rm{Th}$  and  $^{40}\rm{K}$  in 3758 samples

	<sup>238</sup> U	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K
Mode, $Bq kg^{-1}$	8	5	4	355
0.01-percentile, $Bq kg^{-1}$	0.20	2.72	2.84	40
Median, $Bq kg^{-1}$	15.30	14.35	15.34	406
0.99-percentile, $Bq kg^{-1}$	90.37	86.69	150.4	1182

Figure S6. Examples of heterogeneous dose rate distributions at the Palaeolithic site of Roc de Marsal [53, 54].

a) Gamma dose rates. The stratigraphic section (left panel) comprises sediment between two calcareous rocky horizons: bedrock at the bottom and collapsed rocks from the cave roof on top; the right panel is a schematic view of the section. The graph (inset) illustrates that gamma dose rates measured in situ with Al<sub>2</sub>O<sub>3</sub>:C dosimeters show large variations, because the limestone contains very low radionuclide concentrations compared to the sediment. In such cases, dating the sediment at or close to the interface requires careful assessment of the gamma dose rate.



b) Beta dose rates. Beta autoradiography [55] (right panel) allows qualitative estimation of dose rate heterogeneities. In the example shown here, the limestone gravel and effects of dissolution ('Loose brown patch') shown in left panel induce large variations in beta dose rates (right panel). 'Hotspots' appear in the form of small dark circular patches on the right image, in this case presumably K-feldspar grains. Generally low dose rates, poor sorting and coarser grain size distributions tend to indicate greater variability in dose rates [56, 57, 58, 59].



5 cm



Figure S7. The main components in an OSL measurement system.

Figure S8. Emission spectra of several sedimentary quartz samples from South Australia obtained using stimulation at 647 nm (reproduced from [60]).



Figure S9. Detection: transmission of 7.5 mm of Hoya U-340 detection filter; Emission: Quartz OSL; Stimulation: emission spectra of typical blue and green LEDs. Cut-off filters: Typical cut-off filters used in front of blue and green stimulation LEDs to more effectively separate the stimulation light from the detection window.



Figure S10. Optical stimulation (excitation) spectrum for a sedimentary quartz annealed at 850°C and detected through the Hoya U-340 glass filter. Reproduced from [61].



Table S1. Methods for the determination of dose rates. Where relevant (marked with a \* below), it is usually desirable to ensure that  $^{222}$ Rn is in equilibrium with its parent  $^{226}$ Ra before measurement.

Method	Output	Comment	References
High-resolution gamma	[K], [U], [Th]	Allows some investigation of disequilibrium.	[44], [62],
spectrometry*		Depending on detector, requires >10 g of sample.	[63]
Low-resolution gamma spectrometry*	[K], [U], [Th]	Slightly less sensitive than high resolution, but does not require cooling. Typically ~200 g sample	[64]
Beta counting*	Beta activity concentration	Conversion from count rate to dose rate is radionuclide dependent. Requires 10s of g sample.	[52], [65]
Alpha counting*	Alpha activity concentration	<sup>222</sup> Rn difficult to control; lithium borate glassification can be used to ensure homogeneity	[4]; [66]
ICP-MS (Inductively Coupled Plasma-Mass Spectrometry), or OES (Optical Emission Spectrometry) for K	[K], [U], [Th]	Only a few hundred mg of material analysed, so homogenisation is of great importance.	[67] [68]
NAA (Neutron Activation Analysis)	[K], [U], [Th]	Only a few hundred mg of material analysed, so homogenisation is of great importance.	[69] [70]
In situ dosimeters	Gamma and cosmic dose rate at sample location	Requires long storage time on site (6-12 months). Useful in heterogeneous settings at the dm- or m-scale.	[8] [9] [71]
Field gamma spectrometer	effective [K], [U], [Th], or direct gamma dose rate at sample location	Useful when working in heterogeneous settings at the dm- or m-scale.	[12], [13]; [10]; [11]

Table S2. Energy release and dose rates in a) the  $^{232}$ Th decay series, and b) the Uranium ( $^{238}$ U and  $^{235}$ U) decay series, and c) Potassium ( $^{40}$ K) and Rubidium ( $^{87}$ Rb; only beta dose rates). Adapted from [6]

a) <sup>232</sup>Th decay series.

Isotope	Half-life	Alpha		В	eta	Gamma	
	(s)	Е	DR	Е	DR	Е	DR
<sup>232</sup> Th	4.43 1017	4.003	0.0821	0.0113	0.0002	0.0011	0.0000
<sup>228</sup> Ra	$1.81 \ 10^8$	-	-	0.0092	0.0002	0.0004	0.0000
<sup>228</sup> Ac	2.21 104	-	-	0.4171	0.0086	0.8602	0.0176
228Th	6.03 107	5.406	0.1109	0.0195	0.0004	0.0031	0.0001
<sup>224</sup> Ra	3.16 10 <sup>5</sup>	5.673	0.1164	0.0023	0.0000	0.0104	0.0002
<sup>220</sup> Rn	5.56 10 <sup>1</sup>	6.288	0.1290	-	-	0.0006	0.0000
<sup>216</sup> Po	1.45 10-1	6.778	0.1390	-	-	0.0000	0.0000
<sup>212</sup> Pb	3.83 10 <sup>4</sup>	-	-	0.1721	0.0035	0.1437	0.0029
<sup>212</sup> Bi	3.63 10 <sup>3</sup>	2.175	0.0446	0.5034	0.0103	0.1039	0.0021
<sup>212</sup> Po (0.641)	2.99 10-4	5.631	0.1155	-	-	-	-
<sup>208</sup> Tl (0.359)	1.83 10 <sup>2</sup>	-	-	0.2140	0.0044	1.2136	0.0249
total			0.7375		0.0277		0.0479
Pre-Rn total			0.3093		0.0094		0.0180

#### Notes.

1. Energies ("E) are given in MeV and represent the energy emitted per disintegration.

2. Branching ratios are shown in parenthesis against the radioelements in the branches; associated values given for energy release are after adjustment for branching. Note that the branching also affects the energy release of the radioelement at which the branching occurs; thus the value given for the alpha release by <sup>212</sup>Bi is 35.9% of the full energy - because <sup>208</sup>Tl is formed by alpha emission from <sup>212</sup>Bi.

3. Beta components include Auger electrons and internal conversion; gamma components include X-rays and annihilation radiation; alpha recoil and neutrinos are not included due to their insignificant contribution to dose-rates (cf. [72]).

5. A dash indicates that no radiation of that type is mentioned by the National Nuclear Data Centre.

6. Dose rate values ("DR") are given in Gy.ka<sup>-1</sup> per ppm of parent (i.e. mg of parent per kg of sample), assuming equilibrium in the decay chains. The activity of the parent is 4.057 Bq per kg of sample.

7. The rows labelled 'pre-Rn' give the values for 100% escape of radon.

8. Relative differences are calculated between this paper and values from [72].

9. <sup>216</sup>At has been omitted since its contribution to the total energy is insignificant.

Table S2 continued.

Instanc	Half-life	Alpha			Beta	Ļ		Gamma		
Isotope	(s)	Е	DR	DR, nat. U	Е	DR	DR, nat. U	Е	DR	DR, nat. U
<sup>238</sup> U	$1.41 \ 10^{17}$	4.193	0.264	0.262	0.007	0.0005	0.0004	0.001	0.0001	0.0001
<sup>234</sup> Th	2.08 106	-	-	-	0.059	0.0037	0.0037	0.008	0.0005	0.0005
$^{234}$ Pa <sub>m</sub>	6.95 10 <sup>1</sup>	-	-	-	0.810	0.0509	0.0506	0.016	0.0010	0.0010
<sup>234</sup> Pa (0.0016)	2.41 104	-	-	-	0.001	0.0001	0.0001	0.001	0.0001	0.0001
<sup>234</sup> U	$7.75 \ 10^{12}$	4.759	0.299	0.297	0.012	0.0007	0.0007	0.001	0.0001	0.0001
<sup>230</sup> Th	2.38 10 <sup>12</sup>	4.664	0.293	0.291	0.013	0.0008	0.0008	0.001	0.0001	0.0001
<sup>226</sup> Ra	$5.05 \ 10^{10}$	4.775	0.300	0.298	0.004	0.0002	0.0002	0.007	0.0005	0.0005
<sup>222</sup> Rn	$3.30\ 10^5$	5.489	0.345	0.343	-	-	-	0.000	0.0000	0.0000
<sup>218</sup> Po	$1.86\ 10^2$	6.001	0.377	0.375	-	-	-	-	-	-
<sup>214</sup> Pb	$1.61\ 10^3$	-	-	-	0.291	0.0183	0.0182	0.239	0.0150	0.0149
<sup>214</sup> Bi	$1.19\ 10^3$	0.001	0.000	0.000	0.654	0.0411	0.0408	1.475	0.0928	0.0921
<sup>214</sup> Po	1.64 10 <sup>-4</sup>	7.687	0.483	0.480	-	-	-	0.000	0.0000	0.0000
<sup>210</sup> Pb	$7.01\ 10^8$	-	-	-	0.033	0.0021	0.0021	0.005	0.0003	0.0003
<sup>210</sup> Bi	$4.33\ 10^5$	-	-	-	0.389	0.0245	0.0243	-	-	-
<sup>210</sup> Po	$1.20\ 10^7$	5.304	0.333	0.331	0.000	0.0000	0.0000	0.000	0.0000	0.0000
<sup>238</sup> U total			2.695	2.676		0.1429	0.1419		0.1104	0.1096
<sup>238</sup> U Pre-Rn			1 1 5 6	1 148		0.0570	0.0566		0.0022	0.0022
total				111.10		010070	0.00000		0.0022	0.0022
235* *	2 22 1016	4 1 1 4	1 ((2	0.012	0.020	0.0117	0.0001	0.164	0.0005	0.0005
2350	2.22 10.0	4.114	1.003	0.012	0.029	0.011/	0.0001	0.104	0.0005	0.0005
<sup>231</sup> Ih	2.20 10°	-	-	-	0.146	0.0591	0.0004	0.023	0.0094	0.0001
<sup>231</sup> Pa	1.03 10.2	4.924	1.990	0.014	0.032	0.0130	0.0001	0.040	0.0160	0.0001
<sup>227</sup> Ac	$0.8/10^{\circ}$	0.070	0.028	0.000	0.012	0.0049	0.0000	0.001	0.0002	0.0000
$^{223}$ En (0.014)	$1.01\ 10^{\circ}$ $1\ 22\ 10^{3}$	5.808	2.547	0.017	0.050	0.0202	0.0001	0.134	0.0021	0.0004
223 <b>D</b> -	$1.52\ 10^{\circ}$	5.664	2 280	0.000	0.000	0.0002	0.0000	0.001	0.0003	0.0000
219 <b>P</b> n	<b>3</b> 06 10 <sup>0</sup>	6 753	2.289	0.010	0.008	0.0273	0.0002	0.133	0.0340	0.0004
215 <b>D</b> O	1 78 10 <sup>-3</sup>	7 302	2.729	0.019	0.007	0.0027	0.0000	0.058	0.0255	0.0002
211 <b>Dh</b>	$2.17.10^3$	1.392	2.987	0.021	0.450	- 0 1817	0.0013	0.064	0.0258	0.0002
211 <b>B</b> i	$1.28 \ 10^2$	6 5 4 9	2 647	0.019	0.13	0.0053	0.0013	0.004	0.0258	0.0002
211 <b>P</b> O	5 16 10 <sup>-1</sup>	0.071	2.047	0.019	0.013	-	0.0000	-	-	-
<sup>207</sup> T1	$2.86 \ 10^2$	-	-	-	0.495	0.2002	0.0014	0.002	0.0009	0.0000
<sup>235</sup> U total	2.00 10		16 690	0.1185	55	0.5265	0.0037		0.2807	0.0020
total			10.070	2 795		0.0200	0.1457		0.2007	0.1116
Pre-Rn total				1 267			0.0603			0.0042
				1.407			0.0000			0.0012

b) Uranium ( $^{238}$ U and  $^{235}$ U) decay series.

Notes.

1. See notes 1-8 of Table S2(a).

2. The mass abundances used in the natural Uranium calculations for  $^{238}$ U and  $^{235}$ U (respectively 99.29% and 0.71%) correspond to the natural atomic abundances of 99.28% and 0.72% respectively.

3. The activity of the parent (per ppm of parent) is 12.44 Bq per kg of sample for <sup>238</sup>U, 79.94 for <sup>235</sup>U and 12.92 for natural Uranium.

4. The rows labelled 'pre-Rn' give the values for 100% escape of radon in the case of  $^{238}$ U series, but because of the short half-life of  $^{219}$ Rn the values given for natural Uranium include contributions of that gas and its daughters.

5. <sup>218</sup>At, <sup>218</sup>Rn, <sup>210</sup>Tl, <sup>206</sup>Tl and <sup>215</sup>At have been omitted since their contribution to the total is insignificant.

Table S2 continued.

c) Dose-rate data for Potassium and Rubidium.

		<sup>40</sup> K	<sup>87</sup> Rb
Natural abundance (mg.g <sup>-1</sup> )		0.119	283
Half-life (Ga)		1.248	48.1
Average energy per disintegration	Beta	0.499	0.0817
(MeV)	Gamma	0.1557	
Specific activity (Bq.kg <sup>-1</sup> ) for	Total	316.4	44.8
concentration $af 19$ and $K and 50$ new afrest Ph	Beta	282.5	44.8
of 1% hat. K and 50 ppm of hat. Ko	Gamma	33.73	
Dose-rate (Gy.ka <sup>-1</sup> ) for	Beta	0.7982	0.0185
concentrations as above	Gamma	0.2491	

Notes.

1. The energy given for Potassium is that released per disintegration, i.e. after allowance for branching between beta and gamma (89.28% and 10.72% respectively).

2. The contents given in row 1 correspond to natural atomic abundances of 116.7 ppm and 27.8%.

Table S3. Typical concentrations of radionuclides in a well-sorted sand, and the derived infinite matrix dose rates (without grain size attenuation taken into account). In this case, <sup>40</sup>K alone contributes about 55% to the total, and the gamma dose rate about 35%.

Radio-	Activity	Concentration	Beta dose rate,	Gamma dose rate,
nuclide	Bq.kg <sup>-1</sup>		Gy.ka <sup>-1</sup>	Gy.ka <sup>-1</sup>
<sup>40</sup> K	300	~1%	0.76	0.24
<sup>238</sup> U	20	~1.5 ppm	0.23	0.17
<sup>232</sup> Th	20	~5 ppm	0.14	0.24
		Total:	1.12	0.65

Table S4. Beta self-dose values ( $\varphi$ ) used to derive attenuation factors (1- $\varphi$ ) following the superposition
theorem [73]. Values were calculated with Geant4 for the <sup>232</sup> Th decay series, the Uranium ( <sup>238</sup> U and <sup>235</sup> U)
decay series (all in secular equilibrium), and $^{40}$ K, for two different minerals: quartz and potassium feldspar.

GEANT4								
	Pota	ssium	Ura	nium		Thorium		
Grain size (ø, μm)	Quartz (d=2.65 g.cm <sup>-3</sup> )	KAlSi <sub>3</sub> O <sub>8</sub> (d=2.60 g.cm <sup>-3</sup> )	Quartz (d=2.65 g.cm <sup>-3</sup> )	KAlSi <sub>3</sub> O <sub>8</sub> (d=2.60 g.cm <sup>-3</sup> )		Quartz (d=2.65 g.cm <sup>-3</sup> )	KAlSi <sub>3</sub> O <sub>8</sub> (d=2.60 g.cm <sup>-3</sup> )	
20	0.007	0.007	0.038	0.037		0.048	0.047	
40	0.014	0.014	0.057	0.056		0.077	0.075	
60	0.022	0.021	0.072	0.07		0.098	0.097	
80	0.029	0.028	0.084	0.082		0.117	0.115	
100	0.037	0.035	0.095	0.093		0.134	0.131	
120	0.044	0.042	0.105	0.103		0.149	0.146	
140	0.052	0.05	0.115	0.113		0.163	0.16	
160	0.059	0.057	0.125	0.122		0.176	0.173	
180	0.067	0.064	0.134	0.131		0.188	0.185	
200	0.075	0.072	0.143	0.14		0.2	0.196	
250	0.095	0.091	0.165	0.161		0.225	0.221	
300	0.114	0.11	0.185	0.181		0.247	0.243	
400	0.154	0.148	0.223	0.217		0.286	0.281	
600	0.231	0.223	0.289	0.282		0.35	0.343	
800	0.304	0.294	0.346	0.338		0.403	0.395	
1000	0.372	0.36	0.395	0.387		0.449	0.441	

Table S5. Water and carbonate correction factors for a) beta radiation and b) gamma radiation. Reproduced from [47].

Water content $(m_w/m_s)$	Carbonate	e content $(m_c/m_s)$	)						
	0		0.04	0.04		0.20		1.00	
0	-		K Th U	0.99 0.99 0.99	K Th U	0.99 0.99 0.99	K Th U	0.99 0.99 0.99	
0.04	K	1.19	K	1.09	K	1.03	K	1.00	
	Th	1.20	Th	1.09	Th	1.03	Th	1.00	
	U	1.19	U	1.09	U	1.02	U	1.00	
0.20	K	1.20	K	1.16	K	1.09	K	1.02	
	Th	1.20	Th	1.17	Th	1.09	Th	1.02	
	U	1.20	U	1.16	U	1.09	U	1.02	
1.00	K	1.19	K	1.18	K	1.15	K	1.09	
	Th	1.19	Th	1.18	Th	1.16	Th	1.09	
	U	1.19	U	1.18	U	1.15	U	1.08	

## a) The water and carbonate correction factors for beta radiation.

The water and carbonate content of the sediment is given as mass ( $m_w$  and  $m_c$ , respectively) per mass sediment ( $m_s$ ). The factures are tabulated for independent determination of water and carbonate. The use of these tables requires the assessment of the mass of the dry sediment (the fraction having no water or carbonate). It is suggested that intermediate values are calculated using log interpolation.

## b) The water and carbonate correction factors for gamma radiation.

Water content $(m_w/m_s)$	Carbonate content $(m_c/m_s)$								
	0		0.04	0.04		0.20		1.00	
0	_		К	1.02	K	1.02	K	1.01	
			Th	1.04	Th	1.04	Th	1.03	
			U	1.04	U	1.04	U	1.04	
0.04	K	1.06	К	1.04	K	1.03	K	1.02	
	Th	1.02	Th	1.03	Th	1.04	Th	1.03	
	U	1.02	U	1.03	U	1.04	U	1.04	
0.20	K	1.06	K	1.05	K	1.04	K	1.02	
	Th	1.02	Th	1.02	Th	1.03	Th	1.03	
	U	1.01	U	1.02	U	1.03	U	1.04	
1.00	K	1.04	K	1.04	K	1.04	K	1.04	
	Th	0.99	Th	0.99	Th	1.00	Th	1.03	
	U	0.98	U	0.98	U	1.01	U	1.03	

The water and carbonate content of the sediment is given as mass ( $m_w$  and  $m_c$ , respectively) per mass sediment ( $m_s$ ).

Grain radius	Cubic simple			Cubic simple Body-centred cubic				Fac	Face-centred cubic			
(µm)	K	U	Th	K	U	Th	Κ	U	Th			
20	1.181	1.157	1.164	1.194	1.176	1.185	1.205	1.193	1.188			
	<i>0.002</i>	<i>0.017</i>	<i>0.019</i>	0.008	<i>0.030</i>	0.034	<i>0.002</i>	0.034	<i>0.032</i>			
40	1.165	1.132	1.137	1.184	1.160	1.163	1.198	1.176	1.173			
	<i>0.002</i>	<i>0.016</i>	0.019	<i>0.010</i>	<i>0.027</i>	0.027	<i>0.002</i>	0.027	0.033			
60	1.154	1.112	1.118	1.177	1.152	1.167	1.189	1.165	1.169			
	<i>0.003</i>	0.015	<i>0.021</i>	0.011	0.024	<i>0.022</i>	<i>0.003</i>	0.032	0.038			
80	1.146	1.102	1.105	1.170	1.134	1.137	1.177	1.138	1.153			
	<i>0.003</i>	<i>0.014</i>	0.017	<i>0.013</i>	<i>0.024</i>	0.017	0.003	<i>0.030</i>	<i>0.031</i>			
100	1.140	1.086	1.104	1.164	1.128	1.139	1.173	1.139	1.154			
	<i>0.003</i>	<i>0.014</i>	<i>0.019</i>	<i>0.014</i>	0.025	<i>0.032</i>	0.003	<i>0.029</i>	0.045			
150	1.131	1.071	1.085	1.154	1.113	1.127	1.162	1.116	1.137			
	<i>0.003</i>	<i>0.017</i>	<i>0.016</i>	0.016	<i>0.027</i>	0.036	0.004	<i>0.032</i>	0.030			
200	1.124	1.061	1.074	1.144	1.094	1.106	1.156	1.095	1.111			
	<i>0.004</i>	<i>0.015</i>	<i>0.016</i>	0.018	<i>0.022</i>	<i>0.028</i>	0.004	<i>0.031</i>	0.038			
300	1.115	1.058	1.073	1.137	1.086	1.095	1.147	1.101	1.107			
	0.005	<i>0.015</i>	0.017	<i>0.02</i>	<i>0.025</i>	0.027	0.005	0.032	0.033			
400	1.109	1.044	1.054	1.129	1.082	1.087	1.148	1.106	1.112			
	0.005	<i>0.014</i>	0.017	0.023	<i>0.023</i>	0.035	0.005	<i>0.029</i>	0.038			
500	1.106	1.042	1.053	1.127	1.062	1.082	1.143	1.088	1.084			
	<i>0.006</i>	<i>0.017</i>	<i>0.016</i>	0.025	<i>0.022</i>	<i>0.031</i>	<i>0.006</i>	<i>0.029</i>	<i>0.031</i>			

Table S6 (reproduced from [48]). Water correction factors for gamma radiation obtained using Monte Carlo simulations for the three radioelements found in sediments, as a function of the packing system and grain size.

Table S7. Example tables containing suggested minimum information that should be presented in a publication containing quartz OSL luminescence ages.

	1
я	۱
u	,

Lab.code	<sup>238</sup> U, Bq.kg <sup>-1</sup>	<sup>226</sup> Ra, Bq.kg <sup>-1</sup>	<sup>232</sup> Th, Bq.kg <sup>-1</sup>	<sup>40</sup> K, Bq.kg <sup>-1</sup>	*beta dose rate, Gy.ka <sup>-1</sup>	*gamma dose rate, Gy.ka <sup>-1</sup>
1	51 ± 12	46.0 ± 1.2	61.0 ± 1.2	562 ± 20	2.29 ± 0.06	1.48 ± 0.04
2	72 ± 12	48.2 ± 1.1	60.6 ± 1.2	566 ± 16	2.32 ± 0.05	$1.50 \pm 0.04$
3	58 ± 9	45.2 ± 0.9	60.2 ± 1.0	581 ± 13	2.33 ± 0.05	$1.48 \pm 0.04$
4	46 ± 8	43.8 ± 0.8	32.2 ± 0.8	158 ± 8	1.05 ± 0.04	0.81 ± 0.04

Note: \* dry infinite matrix dose rates

b)

Lab.code	Site	Depth, cm	w.c. %	OSL D <sub>e</sub> , Gy	(n <sub>r</sub> )	(n <sub>a</sub> )	Total dose rate, Gy/ka	OSL age, ka
1	Test site-A	65	16	95 ± 4	1	23	3.16 ± 0.16	30.0 ± 2.0
2	Test site-B	65	16	73 ± 4	0	12	3.19 ± 0.16	22.9 ± 1.7
3	Test site-C	65	16	107 ± 6	0	24	3.20 ± 0.16	33.5 ± 2.5
4	Test site-C	30	11	43 ± 2	1	35	1.79 ± 0.09	23.9 ± 1.7



(a) Mean grain size 215μm. (b) Site altitude 0 km. (c) w.c.: water content. (d) n<sub>r</sub>, n<sub>a</sub>: number of rejected and accepted aliquots, respectively.

Table S8. Typical dose rate uncertainty budget for a 50 ka sample with dose rate of 2 Gy.ka<sup>-1</sup> (neglecting sample measurement uncertainties). Dose rate derived from gamma spectrometry measurements of K,U,Th.

	¤Est. Relative	elative Absolute Nature of uncertainty when ages compared with:				
	Uncertainty	uncertainty,	#OSL ages	#OSL ages from	Non-OSL	
	%	ka	from same section	other sites	ages	
Saturation water content	2	1	Usually <b>Random</b> , (but likely <b>Systematic</b> if same grain-size distribution in all samples)	Random	Random	
Fraction of saturation	4	2	<b>Random</b> (assumed values for each sample chosen independently of each other) and/or <b>Systematic</b> (e.g. if site was wetter than assumed).	Random	Random	
Gamma spectrometer calibration	2	1	#Systematic for a single spectrometer (but each spectrometer has its own uncertainty on the counting of the calibration standards)	<i>Systematic</i> for a single gamma spectrometer (but see cell to left)	<i>Systematic</i> for a single gamma spectrometer (but see cell to left)	
Conversion - activity to dose rate	2	1	Systematic	Systematic	Systematic	
Beta source calibration	2	1	<i>*Systematic</i> for a single source (but each source has its own uncertainty on measurement of the known-dose standard)	<i>Systematic</i> for a single source (but see cell to left)	<i>Systematic</i> for a single source (but see cell to left)	
Cosmic ray uncertainty	0.02 Gy.ka <sup>-1</sup> *	0.5	<i>Systematic</i> for given elevation, relative depth and latitude.	<b>Random</b> (unknown depth variation between sites)	Random	
Beta attenuation factor	2	1	Systematic	Systematic	Systematic	
Internal activity of quartz (assumed 0.02 Gy ka <sup>-1</sup> )	0.01 Gy.ka <sup>-1</sup> *	0.03	Systematic	Systematic	Systematic	
	Total:	3 (i.e. 6%)				

¤ Estimates only

\* these two contributions are fixed, rather than relative to the dose rate.

# for all OSL ages from the same laboratory (i.e. assuming a single set of calibration materials).

## References

- H. N. Chandel, A. D. Patel, H. R. Vaghela and G. P. Ubale, »An effective and reusable sampling pipe for luminescence dating, *Ancient TL*, vol. 24, nr. 1, pp. 21-22, 2006.
- [2] H. M. Rajapara, V. Kumar, N. Chauhan, P. N. Gajjar and A. K. Singhvi, »Bleaching of blue light stimulated luminescence of quartz by moonlight," *Journal of Earth System Science*, vol. 129, pp. 1-7, 2020.
- [3] J. R. Prescott and J. T. Hutton, "Cosmic ray contributions to dose rates for luminescence and ESR dating: large depths and long-term time variations," *Radiation Measurements*, vol. 23, p. 497–500, 1994.
- [4] M. Aitken, Thermoluminescence Dating, London: Academic Press, 1985, p. 153.
- [5] N. Porat, G. Faerstein, A. Medialdea and A. Murray, »Re-examination of common extraction and purification methods of quartz and, *Ancient TL*, vol. 33, pp. 22-30, 2015.
- [6] G. Guérin, N. Mercier and G. Adamiec, »Dose-rate conversion factors: update., « Ancient TL, vol. 29, nr. 1, pp. 5-8, 2011.
- [7] L. Bøtter-Jensen, N. Agersnap Larsen, B. G. Markey and S. W. McKeever, »Al2O3:C as a sensitive OSL dosemeter for rapid assessment of environmental photon dose rates, *Radiation Measurements*, vol. 27, nr. 2, pp. 295-298, 1997.
- [8] D. Richter, H. Dombrowski, S. Neumaier, P. Guibert and A. C. Zink, "Environmental gamma dosimetry with OSL of α-Al2O3:C for in situ sediment measurements," *Radiation protection dosimetry*, vol. 141, p. 27–35, 2010.
- [9] S. Kreutzer, L. Martin, G. Guérin, C. Tribolo, P. Selva and N. Mercier, "Environmental dose rate determination using a passive dosimeter: techniques and workflow for α-Al2O3:C chips," *Geochronometria*, vol. 45, p. 56–67, 2018.
- [10] D. Miallier, G. Guérin, N. Mercier, T. Pilleyre and S. Sanzelle, "The Clermont radiometric reference rocks: a convenient tool for dosimetric purposes," *Ancient TL*, vol. 27, p. 37–42, 2009.
- [11] L. J. Arnold, M. Duval, C. Falguères, J. J. Bahain and M. Demuro, "Portable gamma spectrometry with cerium-doped lanthanum bromide scintillators: Suitability assessments for luminescence and electron spin resonance dating applications," *Radiation Measurements*, vol. 47, p. 6–18, 2012.
- [12] L. Løvborg and P. Kirkegaard, »Response of 3" x 3" NaI(Tl) detectors to terrestrial gamma radiation., *Nuclear Instrumensts and Methods*, vol. 121, pp. 239-251, 1974.
- [13] G. Guérin and N. Mercier, "Determining gamma dose rates by field gamma spectroscopy in sedimentary media: results of Monte Carlo simulations," *Radiation Measurements*, vol. 46, p. 190–195, 2011.
- [14] C. W. Curry, R. H. Bennett, M. H. Hulbert, K. J. Curry and R. W. Faas, »Comparative Study of Sand Porosity and a Technique for Determining Porosity of Undisturbed Marine Sediment,« *Marine Georesources & Geotechnology*, vol. 22, p. 231–252, 10 2004.
- [15] M. Nelson and T. Rittenour, »Using grain-size characteristics to model soil water content:

Application to dose-rate calculation for luminescence dating., *Radiation Measurements*, vol. 81, pp. 142-149, 2015.

- [16] D. Zimmerman, »Thermoluminescent dating using fine grains from pottery.,« Archaeometry, vol. 13, pp. 29-52, 1971.
- [17] M. Lawson, J. Daniels and E. Rhodes, »Assessing Optically Stimulated Luminescence (OSL) signal contamination within small aliquots and single grain measurements utilizing the composition test,« *Quaternary International*, vol. 362, pp. 34-41, 2015.
- [18] O. Antohi-Trandafir, A. Timar-Gabor, A. Vulpoi, R. Bălc, J. Longman, D. Veres and S. Simon, »Luminescence properties of natural muscovite relevant to optical dating of contaminated quartz samples," *Radiation Measurements*, vol. 109, pp. 1-7, 2018.
- [19] M. Jain, A. S. Murray and L. Bøtter-Jensen, "Characterisation of blue-light stimulated luminescence components in different quartz samples," *implications for dose measurement Radiation Measurements*, vol. 37, p. 441–449, 2003.
- [20] G. A. Duller, »Distinguishing quartz and feldspar in single grain luminescence measurements, *Radiation Measurements*, vol. 37, pp. 161-165, 2003.
- [21] D. Banerjee, A. S. Murray, L. Bøtter-Jensen and A. Lang, "Equivalent dose estimation from a single aliquot of polymineral fine-grains," *Radiation Measurements*, vol. 33, p. 73–93, 2001.
- [22] P. M. Denby, L. Bøtter-Jensen, A. S. Murray and P. Moska, "Application of pulsed OSL to the separation of the luminescence components from a mixed quartz/feldspar sample," *Radiation Measurements*, vol. 41, p. 774–779, 2006.
- [23] K. J. Thomsen, M. Jain, A. S. Murray, P. M. Denby, N. Roy and L. Bøtter-Jensen, "Minimizing feldspar OSL contamination in quartz UV-OSL using pulsed blue stimulation," *Radiation Measurements*, vol. 43, p. 752–757, 2008.
- [24] M. Jain and A. Singhvi, »Limits to depletion of blue–green light stimulated luminescence in feldspars: implications for quartz dating.,« *Radiation Measurements*, vol. 33, p. 883–892, 2001.
- [25] L. Clark-Balzan and J. Schwenninger, "First steps toward spatially resolved OSL dating with electron multiplying charge-coupled devices (EMCCDs): system design and image analysis," *Radiation Measurements*, vol. 47, p. 797–802, 2012.
- [26] D. Richter, A. Richter and K. Dornich, "Lexsyg a new instrument for luminescence research," *Geochronometria*, vol. 40, p. 220–228, 2013.
- [27] K. J. Thomsen, M. Kook, A. S. Murray, M. Jain and T. Lapp, "Single-grain results from an EMCCD-based imaging system," *Radiation Measurements*, vol. 81, p. 185–191, 2015.
- [28] L. Bøtter-Jensen and D. G.A.T, "A new system for measuring OSL from quartz samples," Nucl. Tracks. Radiat. Meas, vol. 20, p. 549–553, 1992.
- [29] E. Bulur, L. Bøtter-Jensen and A. S. Murray, "Optically stimulated luminescence from quartz measured using the linear modulation technique," *Radiation Measurements*, vol. 32, p. 407–411, 2000.
- [30] D. J. Huntley, M. A. Short and K. Dunphy, "Deep traps in quartz and their use for optical dating," *Canadian Journal of Physics*, vol. 74, p. 81–91, 1996.

- [31] E. Bulur, "An alternative technique for optically stimulated luminescence (OSL) experiment," *Radiation Measurements*, vol. 26, p. 701–709, 1996.
- [32] C. Ankjærgaard and M. Jain, "Optically stimulated phosphorescence in quartz over the millisecond to second time scale: insights into the role of shallow traps in delaying luminescent recombination," *Journal of Physics D: Applied Physics*, vol. 43, p. 255502, 2010.
- [33] D. J. Huntley, D. I. Godfrey-Smith and M. L. W. Thewalt, "Optically dating of sediments," *Nature*, vol. 313, p. 105–107, 1985.
- [34] G. A. T. Duller, L. Bøtter-Jensen, A. S. Murray and A. J. Truscott, "Single grain laser luminescence (SGLL) measurement using a novel automated reader," *Nuclear Instruments and Methods B*, vol. 155, p. 506–514, 1999.
- [35] L. Bøtter-Jensen, K. J. Thomsen and M. Jain, "Review of optically stimulated luminescence (OSL) instrumental developments for retrospective dosimetry," *Radiation Measurements*, vol. 45, p. 253–257, 2010.
- [36] L. Bøtter-Jensen, E. Bulur, G. A. T. Duler and A. S. Murray, "Advances in luminescence instrument systems," *Radiation Measurements*, vol. 32, p. 523–528, 2000.
- [37] L. Bøtter-Jensen, G. A. T. Duller, A. S. Murray and D. Banerjee, "Blue light emitting diodes for optical stimulation of quartz in retrospective dosimetry and dating," *Radiation Protection Dosimetry*, vol. 84, p. 335–340, 1999.
- [38] M. Jain, "Extending the dose range: Probing deep traps in quartz with 3.06 eV photons," *Radiation Measurements*, vol. 44, p. 445–452, 2009.
- [39] G. A. T. Duller, L. Bøtter-Jensen and A. S. Murray, "Combining infrared- and green-laser stimulation sources in single-grain luminescence measurements of feldspar and quartz," *Radiation Measurements*, vol. 37, p. 543–550, 2003.
- [40] V. Hansen, A. Murray, J. P. Buylaert, E. Y. Yeo and K. Thomsen, "A new irradiated quartz for beta source calibration," *Radiation Measurements*, vol. 81, pp. 123-127, 2015.
- [41] B. Combès and A. Philippe, "Bayesian analysis of individual and systematic multiplicative errors for estimating ages with stratigraphic constraints in optically stimulated luminescence dating," *Quaternary Geochronology*, vol. 39, p. 24–34, 2017.
- [42] A. Philippe, G. Guérin and S. Kreutzer, "BayLum-An R package for Bayesian analysis of OSL ages: An introduction," *Quaternary Geochronology*, vol. 49, p. 16–24, 2019.
- [43] A. S. Murray, R. Marten, A. Johnston and P. Martin, »Analysis for naturally occurring radionuclides at environmental concentrations by gamma spectrometry., *Journal of Radioanalytical and Radiochemistry.*, vol. 115, pp. 263-288, 1987.
- [44] P. Guibert and M. Schvoerer, »TL dating: Low background gamma spectrometry as a tool for the determination of the annual dose, *«International Journal of Radiation Applications and Instrumentation. Part D. Nuclear Tracks and Radiation Measurements*, pp. 231-238, 1991.
- [45] Z. Jacobs and R. Roberts, »Single-grain chronologies for for the Still Bay and Howieson's Poort industries and the transition between them: Further analyses and statistical modelling, *Journal* of Human Evolution, vol. 107, pp. 1-13, 2017.
- [46] G. Guérin, N. Mercier, R. Nathan, G. Adamiec and Y. Lefrais, »On the use of the infinite matrix assumption and associated concepts: a critical review., *Radiation Measurements*, vol. 47, nr. 9, pp. 778-785, 2012.
- [47] R. P. Nathan and B. Mauz, "On the dose-rate estimate of carbonate-rich sediments for trapped charge dating," *Radiation Measurements*, vol. 43, p. 14–25, 2008.
- [48] G. Guérin and N. Mercier, »Preliminary insight into dose deposition processes in sedimentary media on a scale of single grains: Monte Carlo modelling of the effect of water on the gamma dose rate.,« *Radiation Measurements*,, vol. 47, nr. 7, pp. 541-547, 2012.
- [49] A. S. Murray, J. P. Buylaert and C. Thiel, "A Luminescence Dating Intercomparison Based on a Danish Beach-ridge Sand," *Radiation Measurements*, vol. 81, pp. 32-38, 2015.
- [50] I. I. I. BIPM, »Evaluation of measurement data-guide to the expression of uncertainty in measurement, *«JCGM 100: 2008 GUM 1995 with minor corrections. Joint Committe for Guides in Metrology*, pp. 1-132, 2008.
- [51] R. Sohbati, A. S. Murray, L. Lindvold, J.-P. Buylaert and M. Jain, »Optimization of laboratory illumination in optical dating, *Quaternary Geochronology*, vol. 39, pp. 105-111, 2017.
- [52] C. Ankjærgaard and A. S. Murray, "Total beta and gamma dose rates based on beta counting in trapped charge dating," *Radiation Measurements*, vol. 42, p. 352–359, 2007.
- [53] G. Guérin, E. Discamps, C. Lahaye, N. Mercier, P. Guibert, A. Turq, H. L. Dibble, S. P. McPherron, D. Sandgathe, P. Goldberg, M. Jain, K. J. Thomsen, M. Patou-Mathis, J.-C. Castel and M.-C. Soulier, »Multi-method (TL and OSL), multi-material (quartz and flint) dating of the Mousterian site of Roc de Marsal (Dordogne, France): correlating Neanderthal occupations with the climatic variability of MIS 5–3,« *Journal of Archaeological Science*, vol. 39, pp. 3071-3084, 2012.
- [54] G. Guérin, M. Frouin, J. Tuquoi, K. J. Thomsen, P. Goldberg, V. Aldeias, C. Lahaye, N. Mercier, P. Guibert, M. Jain, D. Sandgathe, S. J. McPherron, A. Turq and H. L. Dibble, »The complementarity of luminescence dating methods illustrated on the Mousterian sequence of the Roc de Marsal: A series of reindeer-dominated, Quina Mousterian Layers dated to MIS 3,« *Quaternary International*, vol. 433, pp. 102-115, 2017.
- [55] D. Rufer and F. Preusser, »Potential of autoradiography to detect spatially resolved radiation patterns in the context of trapped charge dating., *Geochronometria*, vol. 34, pp. 1-13, 2009.
- [56] R. M. Mayya, P. Morthekai, M. K. Murari and A. K. Singhvi, "Towards quantifying beta microdosimetric effects in single grain quartz dose distribution," *Radiation Measurements*, vol. 41, p. 1032–1039, 2006.
- [57] G. Guérin, M. Jain, K. J. Thomsen, A. S. Murray and N. Mercier, "Modelling dose rate to single grains of quartz in well-sorted sand samples: The dispersion arising from the presence of potassium feldspars and implications for single grain OSL dating," *Quaternary Geochronology*, vol. 27, pp. 52-65, 2015.
- [58] R. K. Smedley, G. A. T. Duller, D. Rufer and J. Utley, "Empirical assessment of beta dose heterogeneity in sediments: implications for luminescence dating," *Quaternary Geochronology*, vol. 56, 2020.

- [59] L. Martin, N. Mercier, S. Incerti, Y. Lefrais, C. Pecheyran, G. Guérin, M. Jarry, L. Bruxelles, F. Bon og C. Pallier, »Dosimetric study of sediments at the beta dose rate scale: Characterization and modelization with the DosiVox software.,« *Radiation Measurements*, vol. 81, pp. 134-141, 2015.
- [60] D. J. Huntley, D. I. Godfrey-Smith and E. H. Haskell, "Light-induced emission spectra from some quartz and feldspars," *Nuclear Tracks and Radiation Measurements*, vol. 18, p. 127–131, 1991.
- [61] L. Bøtter-Jensen, D. G.A.T. and P. N.R.J, "Excitation and emission spectrometry of stimulated luminescence from quartz and feldspars," *Radiation Measurements*, vol. 23, p. 613–616, 1994.
- [62] A. S. Murray, L. Helsted, M. Autzen, M. Jain og J. Buylaert, »Measurement of natural radioactivity: Calibration and performance of a high-resolution gamma spectrometry facility,« *Radiation Measurements*, vol. 15, pp. 215-220, 2018.
- [63] A. Murray og M. Aitken, »Analysis of low-level natural radioactivity in small mineral samples for use in thermoluminescence dating, using high-resolution gamma spectrometry, «*International Journal of Radiation Applications and Instrumentation. Part A. Applied Radiation and Isotopes*, vol. 39, pp. 145-158, 1988.
- [64] M. Bu, A. S. Murray, M. Kook, L. M. Helsted, J.-P. Buylaert og K. J. Thomsen, »Characterisation of scintillator-based gamma spectrometers for," *Radiation Measurements*, vol. 120, pp. 253-259, 2018.
- [65] A. Cunningham, A. S. Murray, S. J. Armitage og M. Autzen, »High-precision natural dose rate estimates through beta counting, *Radiation Measurements*, vol. 120, pp. 209-214, 2018.
- [66] H. E. Jensen og J. R. Prescott, »The thick-source alpha particle countig technique: comparison with other techniques and solutions to the problem of overcounting, *PACT (Rixensart, Belg.)*, vol. 9, pp. 25-35, 1983.
- [67] M. Sargent, H. Goenaga-Infante, K. Inagaki, L. Ma, J. Meija, A. Pramann, O. Rienitz, R. Sturgeon, J. Vogl, J. Wang og L. Yang, "The role of ICP-MS in inorganic chemical metrology," *Meterologia*, vol. 56, p. 034005, 2019.
- [68] R. Bailey, S. Stokes og H. Bray, »Inductively-Coupled Plasma Mass Spectrometry (ICP-MS) for dose rate determination: some guidelines for sample preparation and analysis.,« Ancient TL, vol. 21, pp. 11-15, 2003.
- [69] S. Hossain, F. De Corte, D. Vandenberghe og P. Van den haute, »A comparison of methods for the annual radiation dose determination in the luminescence dating of loess sediment, « Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, vol. 490, nr. 3, pp. 598-613, 2002.
- [70] S. M. Hossain, F. De Corte, D. Vandenberghe og P. Van den Haute, »The role of k0-NAA in the assessment of the annual radiation dose for use in TL/OSL dating of sediments, *« Journal of Radioanalytical and Nuclear Chemistry*, vol. 257, pp. 639-642, 2003.
- [71] C. Burbidge og G. Duller, »Combined gamma and beta dosimetry, using Al2O3:C, for in situ measurements on a sequence of archaeological deposits.,« *Radiation Measurements*, vol. 37, pp. 285-291, 2003.

- [72] G. Adamiec og M. J. Aitken, »Dose-Rate Conversion Factors: Update., « Ancient TL, vol. 16, pp. 37-50, 1998.
- [73] V. Mejdahl, »Thermoluminescence dating; beta-dose attenuation in quartz grains, « *Archaeometry*, vol. 21, pp. 61-72, 1979.



Figure 1: The erosion, transport and sedimentation cycle, and its relationship to the acquisition of a luminescence dating signal. (a) illustrates the various transport agencies that can lead to daylight exposure of mineral grains before final burial\*. (b) any existing energy (i) stored in the quartz crystal structure is released by repeated daylight exposure (ii) during erosion and transport by wind, water, ice, gravity or turbation, interspersed with periods of burial. Sufficient net daylight exposure completely removes the previously stored energy. This energy builds up again (iii) during the most recent burial, until the quartz sand grains are sampled and taken to the laboratory. (c) Stimulating the sand grains with blue light releases the stored energy, some of which gives the luminescence signal. This signal decays rapidly with measurement time, as all the energy is released.

\*gravity acts as an independent transport agency only on high gradients.



Figure 2. Dose rate components in luminescence dating. Decay of naturally occurring radionuclides (<sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K) produces ionising radiation (a-particles, b-particles, g-photons), each with a specific range in sediment. A small portion of the dose rate comes from cosmic rays hitting the Earth's surface. Figure redrawn from [206].



Figure 3. The proportional relationship between  $L_i$  and  $T_i$  for a large aliquot (containing hundreds of grains) of 180-250 µm quartz extracted from a fired brick (filled red circles, 19 cycles) and a quartz aliquot from a desert sediment (filled blue circles, 42 cycles). The sediment sample comes from the Kalahari Desert in Namibia (lab code: 175405) and the brick from a monastery floor in Sweden (lab code: 173701). The natural signals ( $L_0$ ) are shown as open circles. Both datasets are consistent with straight lines (dashed) going through the origin, indicating that the ratio  $L_i/T_i$  is constant for each sample. The inset shows the sensitivity change as measured by  $T_i$  (normalized to first test dose measurement) as a function of measurement cycle. Note that heated quartz typically displays a greater sensitivity change during SAR measurements compared to quartz extracted from sediment. Sediment (175405):  $D_i=44.4$  Gy,  $D_T=5.9$  Gy. Fired brick (173701):  $D_i=3.6$  Gy,  $D_T=1.0$  Gy.



Figure 4. (a)  $T_i$  and (b)  $L_i$  plotted against dose for an aliquot of heated brick quartz; some  $T_i$  are labelled to indicate measurement order. (c) Sensitivity corrected ( $L_i/T_i$ ) growth curve obtained by dividing data from (b) by data from (a); inset, stimulation curves for natural signal and calibration quartz [36]. (d) - (f) same as (a)-(c) but for quartz extracted from sediment. Open triangles show the response to D=0 Gy. Other open symbols indicate repeated dose measurements. Note that without sensitivity correction (b, e) the repeated values of L are inconsistent with earlier measurements at the same dose; correcting for sensitivity change by diving by  $T_i$  corrects this (c, f). The interpolated doses ('D<sub>e</sub>' in b,e and D<sub>e</sub> in c,f) and growth curve shapes are also significantly different for the uncorrected and sensitivity corrected data. Samples are the same as used in Figure 3.



Figure 5. (a) Preheat plateaus for sand-sized quartz extracted from loess (D38141 [60], coversand (OS-B; [61]) and heated sediment from a charcoal kiln (GLL-184301; [259]). For all three samples the  $D_e$  is independent of preheat temperature over a wide temperature interval (dashed line). (b) Thermal transfer plateaus for sand-sized quartz extracted from a fluvial sand (GLL-160301; [260]) and a beach ridge (OSL43; [261]). Quartz aliquots were bleached at room temperature twice using blue light stimulation to remove the natural OSL signal before measurement of the thermally transferred dose. The effect of thermal transfer becomes visible for preheats  $\geq 200^{\circ}C$ .



Figure 6. (a) Dose recovery plateau for sand-sized quartz extracted from Chinese loess. The measured to given dose ratio is insensitive to preheat temperature over a wide temperature range. (b) Histogram of dose recovery results measured with a preheat temperature of 260°C. Adapted from [64].



Figure 7. Relationship between large aliquot (containing many hundreds of grains)  $D_e$  and recycling ratio (a) or OSL IR depletion ratio (b) for 90-180 µm quartz extracted from Chinese loess. Upper figures show the individual aliquot  $D_e$  values (normalized to the average  $D_e$  of each sample) as a function of recycling ratio and OSL IR depletion ratio. Lower figures show the recycling and OSL IR depletion ratios as histograms with mean ratios plotted as blue and red dashed lines, respectively. From the upper figures it can be seen that there is no relationship between the measured  $D_e$  and the recycling or OSL IR depletion ratio; rejection of aliquots based on recycling or OSL IR depletion ratios does not improve these sample  $D_e$ . Figure adapted from [60].



Figure 8. Effect of  $D_C$  filtering on grains with different  $D_C$  [237]. The dose recovery data (red data, given dose 170 Gy) shows the effect on the dose recovery ratio of only accepting results from aliquots with increasingly larger  $D_c$  values. These data suggest that only those grains with  $D_C > 125$  Gy give accurate results (red data). Even with this minimum  $D_C$ , ~3% of the accepted grains still give doses in measured doses in saturation (black data).



Figure 9. Opportunities for bleaching of the OSL signal in different environments. (a) aeolian transport during dust storms (Pavliha-istockphoto.com). (b) fluvial transport (Marek Uliasz - race.fit2paddle.com). (c) gravity-driven debris fans (unknown) (d) sediment transported by ice (Michael Hambrey - Glaciers online)(e) bioturbation - termite mounds (ourterritory.com).



Figure 10a. The likely complexities in  $D_e$  determination arising from luminescence characteristics (dark blue band) and bleaching (red band), expressed qualitatively in terms of field observables and expected age (see Section 3 for details). These observables and estimates are shown on the 10 radial axes, grouped according to Depositional environment and implied transport mechanism (very light blue), Sedimentology (light blue) and the expected Burial age of the samples (median blue). "Young" refers to quartz grains with expected  $D_e$  values of less than ~10 Gy, "Old" to quartz grains with  $D_e$  values of larger than ~130 Gy. Each axis is scaled from most complex (centre of the figure) to least complex (outer edge of figure). The likely variability of each feature is shown by the band width on a particular axis. There is no reason to expect a correlation between luminescence characteristics and sorting/stratification, and so there is no dark blue band in that portion of the figure.

For example, the figure shows that aeolian sediments are likely to show low complexity, because they tend to be well bleached and have good luminescence characteristics, both with low variability. In contrast, glacial sediments are likely to show high complexity, because they tend to be poorly bleached and have poor luminescence characteristics although the variability in this conclusion is considerable. Bleaching becomes progressively less important (and less variable) as the Burial age increases, and so the complexity arising from this parameter decreases with age, whereas complexity from luminescence characteristics is at a maximum for oldest sediments (mainly because of saturation effects), and a minimum for medium age material, where signals are strong, and the saturation effects are small.



Figure 10b. Bar chart showing the relative influences of different sedimentary characteristics on dose rate complexity. Each bar depicts an observable sedimentary characteristic (y-axis) that can be evaluated at the time of OSL sample collection or during sample preparation (white labels – present at the time of deposition; black labels – post-depositional alteration). Each bar is gradationally coloured according to the degree of beta, gamma and cosmic dose rate complexity (in both space and time) expected for a given category value (x-axis).



Figure 11. [77], [262], [80]: Comparison of quartz OSL ages with independent age control (n = number of samples, followed by the % of samples within  $\pm 2 \sigma$  of the line of unit slope).



Figure 12. Large aliquot 63-90 um quartz OSL ages plotted as a function of depth for a section of Chinese loess encompassing last glacial loess and Holocene soil. Uncertainties are random only. A Bayesian model (with  $1\sigma$  error band) is also shown illustrating how higher precision can be achieved. Ages excluded from modelling are from samples deliberately sampled in crotovina. Figure modified from SI in [60].