Testing the applicability of optically stimulated luminescence dating to Ocean Drilling Program cores.

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Abstract

Chronologies for marine sediments are usually constructed by tuning marine proxies for global ice volume ($\delta^{18}O$) to the well understood variations in the Earth's orbit, by the identification of event horizons and/or by radiocarbon dating. However, these techniques are not universally applicable. Optically stimulated luminescence dating (OSL) is potentially widely applicable to marine cores and may offer significant advantages over more conventional chronometric techniques but methodological considerations regarding the application of the techniques have yet to be systematically explored. Using material from core Ocean Drilling Program (ODP) core 658B, we assess the applicability of OSL dating to deep ocean sediments. For this core, equivalent dose does not change with depth below the split core face beyond the upper 1 mm, indicating that retrieval and prolonged storage of ODP material does not compromise the OSL signal. However equivalent dose decreases with increasing particle size, reaching a plateau at ~30-40 µm. These data suggest that ocean floor sediment reworking causes the deposition of old material at the sediment-water interface, potentially resulting in OSL age overestimates. This observation strongly suggests that seafloor reworking processes should be considered both when selecting target cores and when interpreting results. Nonetheless, we observe a good general agreement between OSL ages and

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independent age estimates for a suite of sediments from the Marine Isotope Stage 6-5e transition, suggesting that the application of luminescence dating techniques to deep-sea sediments merits further investigation.

Keywords: Optically stimulated luminescence; Geochronology; Marine sediments; North Africa.

1. Introduction

Marine sediments are widely exploited archives of palaeoenvironmental information. Chronologies for marine sediments are usually constructed by tuning marine proxies for global ice volume ($\delta^{18}O$) to the well understood variations in the Earth's orbit (Lisiecki and Raymo, 2005), by the identification of event horizons such as tephras (e.g. Matthews et al., 2015) and geomagnetic excursions (e.g. Collins et al., 2012) and/or by radiocarbon dating. While tremendously powerful, these techniques are not universally applicable. Dating marine sediments using radiocarbon methods or the $\delta^{18}O$ signal is difficult for sites below the carbonate compensation depth (e.g. much of the Southern Ocean), and radiocarbon dating is not applicable beyond ~50-60 ka even where carbonate is preserved. Correlation of the $\delta^{18}O$ record from a particular core to a "stacked" record (e.g. Lisiecki and Raymo, 2005) is often complicated by site specific factors such as: 1) hiatuses in the depositional record due to bottom current reworking; 2) smoothing of the $\delta^{18}O$ signal by bioturbation, such that only the most pronounced events are discernible and 3) misidentification of events in complex isotopic stages such marine isotope stage (MIS) 5. Event horizons such as tephras are invaluable in integrating marine and terrestrial records, but are only useful where their occurrence is frequent in the period of interest. In addition, accurate identification of both tephra horizons and geomagnetic excursions in marine cores frequently requires an initial lower-precision age model e.g. where a single source emits multiple geochemically similar tephras, such as the Campi Flegrei between c. 17-14 ka BP (Matthews et al., 2015). These limitations have rendered some timeframes and regions difficult to date. For example, in the carbonate free regions of the Arctic Ocean, competing chronostratigraphic interpretations yield

sedimentation rates which differ by an order of magnitude (Berger, 2006). Consequently, several authors (e.g. Armitage, 2015; Berger, 2006; Jakobsson et al., 2003; Stokes et al., 2003) have advocated the use of optically stimulated luminescence (OSL) as an additional method to provide age information for deep-sea sediments.

OSL is a radiometric dating technique which determines the time elapsed since a mineral grain (usually quartz or feldspar) was last exposed to sunlight (i.e. burial period) and has gained wide acceptance as a tool for developing terrestrial chronologies (Olley et al., 2004). An OSL age is calculated using Equation 1:

$$\text{Age (ka)} = \text{Equivalent dose (Gy)} / \text{Dose rate (Gy/ka)} \quad \text{(Eq. 1)}$$

where the equivalent dose (D_e) is the laboratory estimate of the cumulative exposure of a sample to ionising radiation since deposition, and the dose rate is the rate of exposure to ionising radiation. In principle, luminescence methods can date any marine sediment containing quartz (to ~200 ka) or feldspar (to ~500 ka), provided that these minerals were subaerially exposed immediately prior to deposition. However, a number of potential impediments to the routine application of OSL dating to deep-sea sediments, relating to determination of both the equivalent dose and the dose rate, have been identified. Determination of a chronologically useful equivalent dose may be hampered by: 1) the incorporation of material which was not exposed to sufficient sunlight prior to deposition to completely reset the OSL signal (Berger, 2006); 2) depletion of the OSL signal due to light exposure during core retrieval and storage, and 3) reworking of older material into sediments during deposition, leading to an overestimation of the depositional age (Armitage, 2015; Berger, 2006). Accurate determination of the dose rate for deep-sea sediments is complicated by disequilibrium in the uranium decay series due to incorporation of excess long-lived insoluble isotopes (^{230}Th in the ^{238}U series and ^{231}Pa in the ^{235}U series) into seafloor sediments (Stokes et al., 2003; Wintle and Huntley, 1979) and

authigenic uranium uptake at the sediment-water interface under reducing conditions (Armitage, 2015).

In this study we use sediment from Ocean Drilling Program (ODP) core 658B (Ruddiman et al., 1988) to test the applicability of optically stimulated luminescence dating to marine sediments. Specifically we aim to: 1) assess directly the depletion of the OSL signal due to light exposure during core retrieval and storage; 2) test the hypothesis (Armitage, 2015) that coarser quartz grains are less prone to seafloor reworking and hence are the preferred dosimeter for OSL dating of deep-sea sediments, and 3) test the applicability of OSL to sediments beyond the range of ^{14}C by dating known-age material from marine isotope stage MIS 6-5e.

2 Materials and methods

Core 658B was recovered in 1986 from a water depth of 2,263 m off Cap Blanc, Mauritania (20°45'N, 18°35'W) during ODP Leg 108 (Ruddiman et al., 1988). Trade winds cause strong upwelling over the site, leading to high surface productivity and high biogenic particle fluxes to the seafloor. Biogenic carbonate comprises 40-60% by mass of the sediment, and until recently the remainder was thought to consist of terrigenous dust (deMenocal et al., 2000) due to the site's location beneath the axis of the summer African dust plume (Figure 1). However, recent analysis of orbital radar satellite imagery has been used to propose that during the late-glacial to Holocene African Humid Period (~11.5-5 ka, Armitage et al., 2015; deMenocal et al., 2000; McGee et al., 2013) a major river within the Tamanrasset palaeowatershed delivered large amounts of fluvial sediment to this site (Skonieczny et al., 2015). Irrespective of the transport pathway followed by terrigenous sediments in ODP Core 658B, Armitage (2015) was able to measure OSL ages which were consistent with independent chronological information over the timeframe ~10-50 ka, suggesting that the OSL signal in this material was completely reset by subaerial exposure immediately prior to deposition.

Core 658B is an ideal target for testing OSL dating in the marine realm since: 1) It has a high accumulation rate (c.18 cm/ka,deMenocal et al., 2000); 2) The high terrigenous sediment flux provides a substantial well-bleached quartz component; 3) The terrigenous sediments have a sufficiently wide grain-size range to test the hypothesis that coarse grains are less prone to seafloor reworking; 4) The excess ^{230}Th ($^{230}Th_{xs}$) record for core Core 658C (Adkins et al., 2006) may be used to calculate excess activity in Core 658B and 5) Core 658B was 16 years old when sampled so the deleterious effects (if any) of light exposure during storage should be pronounced.

2.1 Equivalent dose determination

Samples were collected in July 2002 from a 66 mm diameter split core at the ODP East Coast Repository, Lamont-Doherty Earth Observatory, USA. Paired samples were taken every ~25 cm through the uppermost 11 m of the core by inserting short sections of 20 mm diameter opaque tubing. This approach yielded two ~7 cm^3 sub-samples per level. Samples were processed under subdued orange light at the Royal Holloway luminescence laboratory. In all cases the outer (closest to the core barrel) ~8 mm was discarded to avoid core barrel smearing (Figure 2). For the samples presented in Sections 4 and 5 the light exposed upper (split core face) ~8 mm was removed and used for carbonate content and dose rate analysis, while the remaining middle ~8 mm of the sample was dispersed in deionised water and sieved at 150 µm. The >150 µm fraction consisted of foraminiferal tests, and <150 µm fraction was used for equivalent dose measurements. For the samples presented in Section 3, the upper 5 mm of sediment was removed in 1 mm thick slices, and an equivalent dose was measured for material from each slice. Equivalent doses were measured on quartz isolated from samples by treating them with HCl and H_2O_2 to remove carbonate and organic matter respectively. The resulting material was then separated into the required size fraction using Stokes settling for fractions ≤20 µm and wet sieving for larger fractions, and feldspar was subsequently removed using H_2SiF_6 followed by an HCl rinse.

All OSL measurements presented in this study were carried out using a Risø TL/OSL-DA-20 automated dating system. Optical stimulation of single aliquots was carried out using a blue (470 ± 30 nm) light emitting diode (LED) array with a power density of ~90 mW/cm^2. Infra-red (IR) stimulation was carried out using an IR (870 nm) laser diode array with a power density of ~200 mW/cm^2. OSL was measured using an Electron Tubes Ltd 9235QB photomultiplier tube with 7.5 mm of Hoya U-340 filter interposed between the sample and photomultiplier. Irradiation was carried out using a 40 mCi ^{90}Sr/^{90}Y beta source, calibrated relative to the National Physical Laboratory, Teddington Hotspot 800 ^{60}Co γ-source (Armitage and Bailey, 2005). Equivalent doses (D_e) were determined using the single-aliquot regenerative-dose (SAR) method (Galbraith et al., 1999; Murray and Wintle, 2000). OSL signals were measured at 125 °C and growth curves were fitted using a saturating-exponential-plus-linear function. The standard error associated with each individual D_e determination was estimated by Monte Carlo simulation using Luminescence Analyst Software (Duller, 2007). Previous dose recovery experiments (Roberts et al., 1999; Wallinga et al., 2000) on quartz from ODP Core 658B found no dependence of D_e upon preheating regime for three samples with equivalent doses ranging from 23-77 Gy (Armitage, 2015). An additional dose recovery experiment was performed on sample 73B (D_e = 198 Gy), which confirmed that this finding is also true for the older samples reported in Section 5. Preheating regimes of 260 °C, 10 s for PH1 (the pre-heat prior to measurement of L_n or L_x) and 220 °C, 10 s for PH2 (the pre-heat prior to measurement of T_n or T_x) or 200 °C, 10 s for PH1 and 160 °C, 0 s for PH2 were adopted for all subsequent measurements. When calculating D_e, aliquots were rejected where the recycling ratio (Murray and Wintle, 2000) or IR depletion ratio (Duller, 2003) differed from unity by more than two standard deviations, or where the sensitivity corrected luminescence intensity in response to a 0 Gy regeneration dose exceeded 5% of the sensitivity corrected natural luminescence intensity.

2.2 Dose rate calculation

The environmental dose rate for MIS 6-5e samples from core 658B was calculated using a version of the $Marine_{xs+auth}$ model described in Armitage (2015). Briefly, ^{238}U and ^{232}Th and bulk K concentrations were measured using ICP-MS. Authigenic uranium (U_{auth}) activity was calculated using the $^{238}U/^{232}Th$ activity ratio of crustal rocks and pelagic marine sediments of 0.8±0.2 (Anderson et al., 1989), and corrected for the effects of ingrowth of ^{230}Th from $^{234}U_{auth}$ (Section S1). Burial $^{230}Th_{xs}$ for all samples was assumed to be the mean value (36.6±9.3 Bq/kg) measured by Adkins et al. (2006) for Core 658C over the time period 2-18.5 ka. Initial $^{231}Pa_{xs}$ (3.38±0.87 Bq/kg) was calculated using the 0.093 production activity ratio of $^{231}Pa/^{230}Th$ (Henderson and Anderson, 2003). Time independent dose rates were calculated from detrital ^{238}U (measured ^{238}U minus U_{auth}), U_{auth}, ^{232}Th and K concentrations, assuming equilibrium for ^{238}U and ^{232}Th, and equilibrium from ^{238}U-^{234}U for U_{auth}, using the standard conversion factors (Guérin et al., 2011). The additional time dependant dose contribution due to $^{230}Th_{xs}$ and $^{231}Pa_{xs}$, and the final age calculation, were performed using the method outlined by Stokes et al. (2003). Dose rates were corrected for alpha efficiency (0.02±0.02), alpha and beta attenuation and water content (Aitken, 1985). Water contents were taken from the relevant ODP initial report (Ruddiman et al., 1988). Dose rate data and ages are presented in Tables S1 and S2. The dose rate values for individual isotopes within the uranium and thorium decay series provided in Tables 5-12 of Stokes et al. (2003) are based upon outdated dose rate conversion factors (Adamiec and Aitken, 1998). Updated values based upon the conversion factors of Guérin et al. (2011) are provided in supplementary data Tables S3-S6.

3 Assessment of OSL signal depletion during core retrieval and storage

For OSL dating of marine sediments to become a useful tool for the palaeoclimate community, the technique must be applicable to curated material. However, few marine cores have been retrieved or stored with the intention of preserving their luminescence signal, and consequently most have been exposed to unquantified amounts of light, possibly compromising their use for OSL dating. ODP cores are stored under cool (4 °C) moist conditions, preventing desiccation and shrinkage, meaning

that only the split core face is exposed to laboratory light. Consequently signal loss due to light exposure during storage should be greatest at the core face, and reduce to a negligible level with depth. To determine the depth below which negligible signal loss occurred during storage, D_e-depth profiles were constructed for two samples (Figure 3). The fine silt (4-11 μm) fraction was used for these measurements since it is sufficiently abundant to allow assessment of very small sample volumes. The D_e-depth profile for sample 6B is flat, while the profiles for sample 7B has a plateau in D_e below 1 mm depth.

We interpret these data to indicate that for ODP core 658B, ≤1 mm of sediment is required to completely shield underlying material from bleaching by laboratory light. This result is consistent with similar studies which have demonstrated that the effects of brief sunlight bleaching on TL only extended ~2 mm into a quartzite pebble (Richards, 1994), while <10 mm of sandy limestone prevented bleaching of the OSL signal during ~5,000 years of exposure to direct Saharan sunlight (Armitage and King, 2013). Since core 658B was sampled 16 years after it was split, it is probable that similarly curated cores also preserve a useable amount of material which is suitable for OSL dating, and the technique may be applied retrospectively to a wide range of archived marine cores.

4. Equivalent dose versus grain-size

The fine-silt component of marine sediment is prone to seafloor reworking by bottom-currents. Since this reworking occurs at depth and in darkness, the reworked sediment is deposited with a luminescence signal due to the cumulative radiation exposure experienced since sub-aerial exposure to sunlight. Consequently, the equivalent dose for this material does not relate solely to the time elapsed since final deposition, leading to age overestimates when luminescence dating is applied to minerals extracted from this size-fraction (Armitage, 2015; Berger, 2006, 2009, 2011). However, bottom-current reworking processes should be most important for finer grain-size fractions, owing to the strong dependence of settling velocity upon particle size. For example, 90% of grains within the

nepheloid (cloudy) layer in non-polar deep oceans, which results from bottom-current reworking processes, consists of grains with a diameter for 0.5-8.5 µm (Berger, 2006). These considerations suggest that where bottom-current reworking may have occurred, deep-sea sediments should be dated using mineral grains extracted from the coarsest fraction possible. Unfortunately, the dominant minerogenic fraction of many marine sediments is fine silt sized, and few cores are physically large enough to allow the study of fractions which are not abundant. Consequently, it is important to determine the minimum grain size which is not subject to bottom-current reworking.

ODP site 658 is ideal for studying seafloor reworking since despite being located on a ridge between two major submarine canyon systems (Ruddiman et al., 1988), $^{230}Th_{xs}$ measurements suggest that core 658C has received three time more sediment laterally than vertically over the last 20 ka (Adkins et al., 2006). This observation led Armitage (2015) to argue that the discrepancy between paired fine silt (4-11 µm) and coarse silt (40-63 µm) luminescence ages for core 658B were due to the incorporation of "old" reworked material into the former. In addition, ODP site 658's location beneath the summer African dust plume (deMenocal et al., 2000) and offshore of a major river (Skonieczny et al., 2015) mean that it contains dateable quantities of fine and coarse silt-sized quartz.

Equivalent doses were measured from the 0-5, 5-10, 10-15, 15-20, 20-30, 30-40 and 40-60 µm fractions of samples 12B and 15B (Figure 4). These samples were measured due to the discrepancy between the fine and coarse silt luminescence ages reported by Armitage (2015), and due to the agreement between the latter and the independent chronology for core 658B. For each sample, equivalent doses were normalised to the D_e measured from the 40-63 µm fraction after correction for grain size dependant variation in both instrument (Armitage and Bailey, 2005) and environmental (Durcan et al., 2015) dose rates. For both samples, the normalised D_e decreases with increasing grain size, and D_e for the 30-40 µm grains is indistinguishable from that for 40-60 µm. However, the D_e for the finest fraction (0-5 µm) of 12B was nearly twice that for the coarsest fraction (40-60 µm) whereas the discrepancy was 20% for sample 15B.

From these data we infer that at ODP site 658, reworked older material is present in size fractions as coarse as 20-30 μm, and is not restricted to the fine silt fraction as might have been expected from the composition of the nepheloid layer. The progressive decrease in D_e with increasing grain size suggests that the proportion of reworked material present is similarly decreasing. Consequently it is possible that at sites which are subject to less lateral sediment movement than ODP 658, grain sizes finer than 20-30 μm might produce reliable ages. The main tool for determining the presence or absence of lateral sediment movement is $^{230}Th_{xs}$ measurements (Adkins et al., 2006). However, since this technique relies on discrepancies between the abundance of $^{230}Th_{xs}$ in samples, and the rate of $^{230}Th_{xs}$ production in the overlying water column, it may only be used for well-dated depth intervals (Adkins et al., 2006). Consequently, an alternative method for testing for reworked sediment is required. Where a sufficiently wide range of grain sizes are present in a single sample, the absence of change in D_e with grain size provides strong circumstantial evidence for the absence of older reworked material. Nonetheless the discrepancy between the scale of grain size dependence for samples 12B and 15B suggests that the incorporation of reworked material is highly temporally variable. Consequently, D_e versus grain size measurements would need to be made on a number of samples from any given site to make a strong case that reworked sediment was absent. These conclusions do not preclude the application of luminescence dating to marine sediments, but they do strongly suggest that seafloor reworking processes should be considered both when selecting target cores and when interpreting results. However, since fine grained particles may be redeposited into deep-sea sediments by turbidity (downslope) and contour (along-slope) currents, consideration of contour current velocity alone is unlikely to be sufficient (Faugères and Mulder, 2011).

5. Dating samples from MIS 6-5

A number of studies have demonstrated good agreement between independent chronologies and luminescence ages from deep-sea sediments. In most cases these studies used material retrieved from

259 relatively shallow depths where $^{230}Th_{xs}$ and $^{231}Pa_{xs}$ is insignificant, and/or have used cores where
260 detrital terrigenous inputs are sufficiently large to render these excess activities insignificant in the
261 dose rate calculation. Of the studies that have accounted for $^{230}Th_{xs}$ and $^{231}Pa_{xs}$, few provide
262 convincing ages beyond the range of ^{14}C (e.g. Stokes et al., 2003). However, it is essential to
263 demonstrate the applicability of luminescence dating to these older samples if the technique is to
264 become established as a useful chronometer for marine sediments. To provide a convincing test of
265 luminescence dating as applied to deep-sea sediments beyond the range of ^{14}C, it is necessary to
266 identify samples with precisely and accurately known ages. Samples which meet these criteria have
267 proved difficult to identify in terrestrial settings (e.g. Murray et al., 2008; Murray and Funder, 2003;
268 Murray et al., 2007) but ought to be relatively easy to identify in marine settings with continuous
269 sedimentation and a well-resolved $\delta^{18}O$ record or (well-dated) unambiguously identifiable event
270 horizons. For example, Stokes et al. (2003) used the Toba Ash and last appearance of *Globigerinoides*
271 *ruber* (pink) to provide know age horizons at ~74 and ~120 ka. In the present case, an independent
272 age was estimated for each sample by identifying MIS 5e (sample 69B) and MIS 6 (sample 77A) in
273 our $\delta^{18}O$ record, and assigning these samples ages of 123 and 140 ka respectively (Lisiecki and
274 Raymo, 2005). Ages for samples between these two points were estimated assuming a constant
275 accumulation rate. Ages were measured for eight samples, using coarse silt-sized (40-63 μm) quartz.
276 Equivalent doses, dose rates and ages for these samples are presented in Tables S1 and S2. Coarse
277 silt OSL ages are shown alongside the independent chronological data in Figure 5.
278
279 All the luminescence ages are in agreement with the independent ages estimated from the $\delta^{18}O$ record.
280 However, although individual pairs of luminescence and independent ages are indistinguishable,
281 principally due to the ~6-9% uncertainty on the former, all eight luminescence ages are lower than
282 their respective independent ages. Dose recovery experiments were performed on 10 aliquots of
283 samples 74B and 75B, using given doses of 220 and 240 Gy respectively, and yielded mean ratios
284 (measured/given) of 1.04±0.02 and 1.02±0.02. These data suggest that the measurement procedure

adopted is appropriate for these samples. However, the mean ratio of OSL to independent ages is 0.93±0.04. In an attempt to determine the cause of this age underestimate, the OSL data were repeatedly reanalysed. Firstly, aliquots were rejected where the recycling ratio or IR depletion ratio differed from unity by more than one standard deviation. This approach reduced the yield of acceptable aliquots from 86 to 59%, but the mean ratio of original: recalculated equivalent doses was 1.00±0.01, suggesting that insufficiently stringent rejection criteria do not account for the observed age underestimate. Using an early-background subtraction (Cunningham and Wallinga, 2010) yielded a ratio of 1.01±0.01 (signal and background intervals of 0-0.2 and 0.2-0.7 s as opposed to 0-0.2 and 36-40 s in the original analysis) suggesting that the intial signal is fast-component dominated. Similarly, integrating the signal from 0.1-0.3 s yielded a ratio of 0.98±0.01, indicating that an unstable ultrafast component is not present (Jain et al., 2008). Lastly, excluding aliquots where D_e is greater than twice the curve fitting parameter D_0 when the growth curve is fitted using a single saturating exponential fit (Durcan, 2012; Wintle and Murray, 2006) yielded a ratio of original to recalculated equivalent doses of 0.96±0.02. This, combined with the observation that for all aliquots the sensitivity-corrected luminescence intensity continues to grow with dose beyond D_e, leads us to infer that the observed age underestimate is not caused by dose-response saturation.

The mean ratio of OSL to independent ages (0.93±0.04) is similar in magnitude to the discrepancy between OSL and "known" ages reported for several terrestrial Eemian sites (e.g. Murray and Funder, 2003; Murray et al., 2007) which are approximately contemporaneous with our material. However, quartz OSL age underestimates are not universally found for Eemain and pre-Eemian sites (e.g. Murray et al., 2008), and despite considerable research it has proved difficult to identify a mechanism explaining the age underestimates which are found (e.g. Lowick and Preusser, 2011). It is possible that our ages for MIS 6-5e samples from core 658B are another example of this phenomenon. It is equally plausible that age underestimates result from the use of $^{230}Th_{xs}$ data from the 2-18.5 ka portion of the core (Adkins et al., 2006), which may not be representative of conditions during the MIS 6-5e

transition. Nonetheless, the general agreement between our ages and independent age estimates indicates that luminescence dating of deep-sea sediments beyond the range of ^{14}C is possible.

6. Conclusions

Our data from ODP core 658B suggest that a wide range of marine sediments may be suitable for OSL dating. In particular, prolonged storage in core repositories does not reduce the OSL signal, except for material less than 1 mm below the exposed surface, meaning that most archived cores should be suitable for OSL dating. However, seafloor reworking is not restricted to fine silt sized particles as had been suggested by previous authors. At Site 658, this process appears to be responsible for the redeposition of particles as large as 20-30 μm in diameter. This finding suggests that seafloor reworking may be a primary determinant of the applicability of luminescence dating to deep ocean sediments. Nonetheless, it may frequently be possible to work on sites or particle sizes which are not subject to seafloor reworking. Lastly, our data indicate that accurate luminescence dating of deep-sea sediments beyond the range of ^{14}C is possible, and suggest that this application of the technique merits further investigation.

7 Acknowledgements

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Figure captions

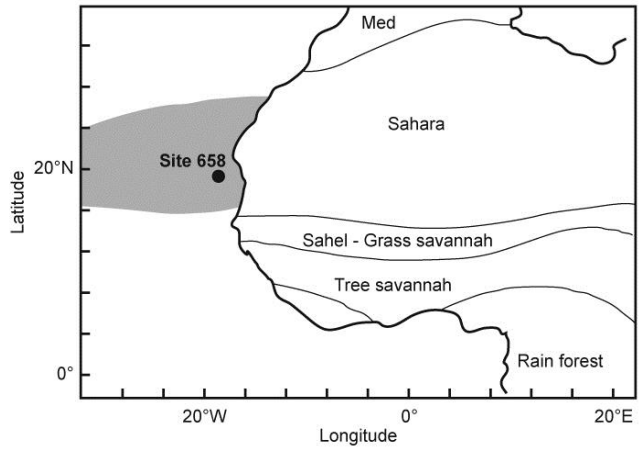

Figure 1: Location of ODP Site 658 under the summer African dust plume (grey). Redrawn from deMenocal et al. (2000).

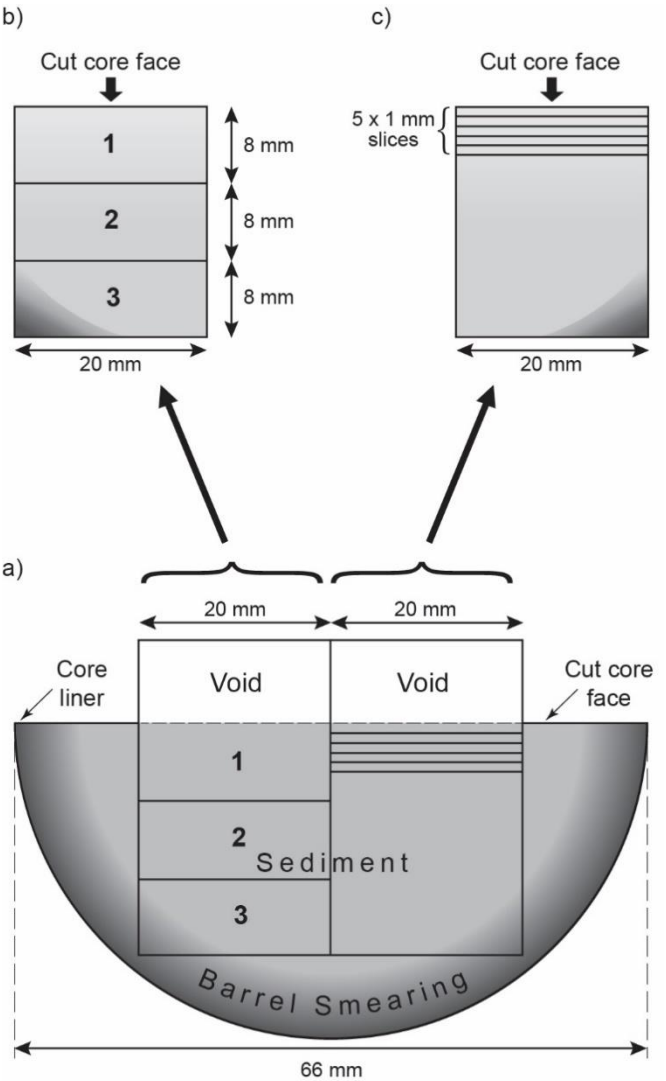

Figure 2: Sampling of ODP core 658B. a) The split core showing core dimensions, the cut core face, which is assumed to be bleached by core repository lighting, and barrel smearing i.e. sediment intruded into underlying material during coring. OSL samples were taken by pushing two 20 mm diameter opaque tubes into the cut core face. b) Typical sub-samples taken from a single OSL

sample tube. This sub-sampling scheme was followed for material presented in Sections 4 and 5. Section 1 contains the light exposed core face and was used for carbonate content and dose rate evaluation. Section 2 was used for equivalent dose determination while section 3 was discarded as containing some contamination due to barrel smearing. c) Sub-sampling scheme used to produce the D_e-depth profiles presented in Section 3.

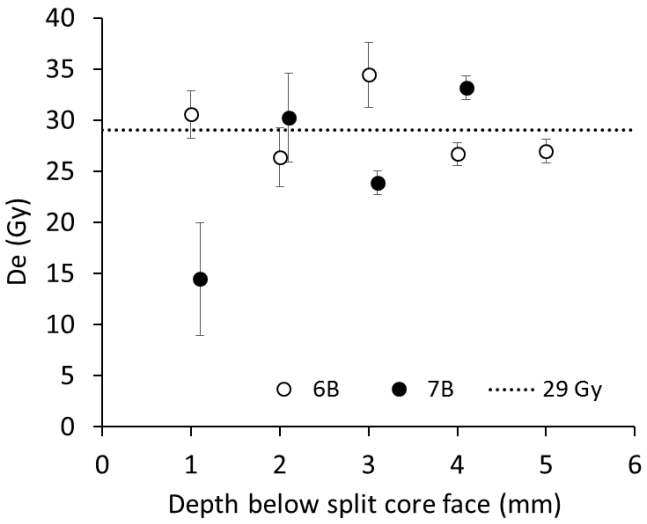

Figure 3: Equivalent dose versus depth below split core face for samples 6B and 7B. Markers are centred upon the mean D_e on the y-axis, and the deepest depth represented in the sample on the x-axis. Each point is the mean of 3-6 aliquots, and 6 aliquots were measured where sufficient material was available. Data for sample 7B are offset by 0.2 mm on the x-axis for clarity. The dashed line is at 29 Gy, the approximate D_e for the 8-16 mm depth subsample of both samples (6B = 28.99±1.07 Gy and 7B = 29.08±1.07 Gy, Armitage, 2015)

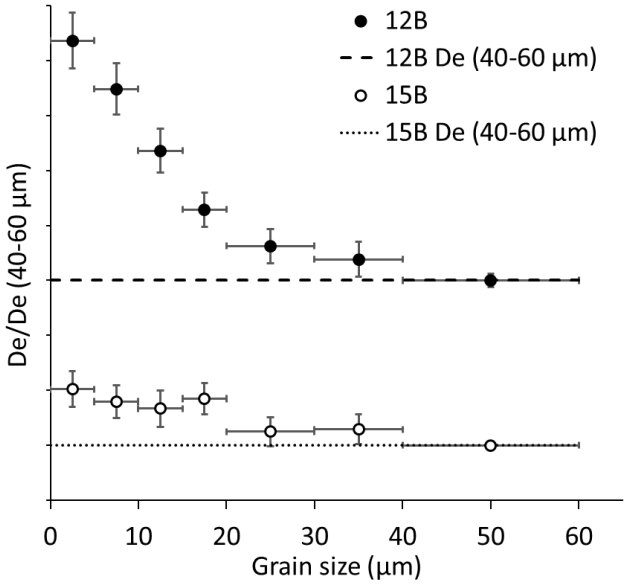

Figure 4: Equivalent dose (normalised to the equivalent dose for the 40-60 µm fraction) versus grain size for samples 12B and 15B. Markers are centred upon the mean normalised D_e on the y-axis, and the mean grain size on the x-axis. Each point is the mean of 6-9 aliquots, and 9 aliquots were measured where sufficient material was available. Equivalent doses were corrected for the grain size dependency of instrument and environmental dose rates (Armitage and Bailey, 2005; Durcan et al., 2015) and normalised to the data for 40-60 µm. A 3% uncertainty was assumed on each correction factor. X-axis uncertainties indicate the range of grain sizes in a fraction, while the

y-axis uncertainties were calculated via the propagation in quadrature of errors on the measured equivalent dose, instrument beta source calibration (3%), grain-size specific instrument dose rate correction factor (3%) and grain size specific environmental dose rate correction factor. Y-axis tick marks are spaced at 0.2 (20%) intervals.

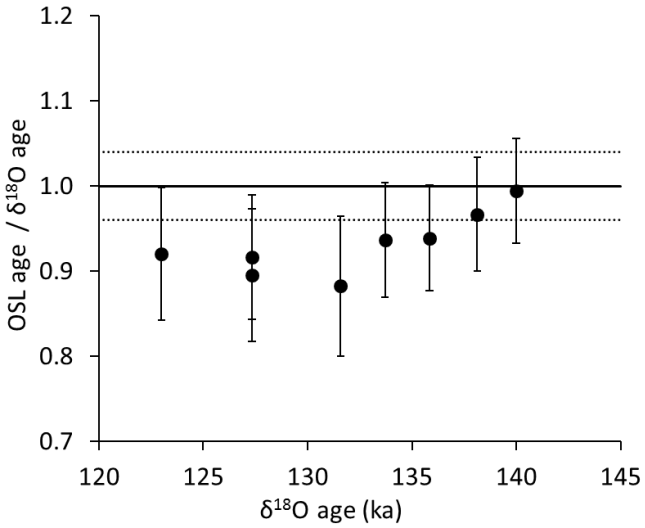

Figure 5: OSL age (normalised to the independent age determined via comparison of our $\delta^{18}O$ record to the LR04 stack – "$\delta^{18}O$ age") versus independent age for coarse silt-sized quartz samples from the MIS6-5e section of core 658B. Nine aliquots were measured for each sample, and two separately prepared sub-samples of sample 71 (127 ka) were measured. The solid line represents a normalised OSL age of unity (OSL age = independent age) while the dashed lines at ± 4% of this value represent the claimed uncertainty on ages attributed to isotopic events in the 0-1 Ma portion of the LR04 stack (Lisiecki and Raymo, 2005). Non-normalised OSL and $\delta^{18}O$ ages are presented in Table S2.

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