**Supplementary Information – Single-grain OSL dating methods**

Single-grain De measurements were made using two Risø TL-DA-20 readers (“Riso 2” and “Riso 7”) equipped with blue LED units (Riso 2 = 470 nm, maximum power of 52 mW/cm2; Riso 7 = 470 nm, maximum power 102 mW/cm2), an array of infrared LEDs (Riso 2 = peak emission 870 nm, maximum power of 141 mW/cm2; Riso 7 = peak emission 850 nm, maximum power of 340 mW/cm2), and 10 mW Nd:YVO4 single-grain laser attachments emitting at 532 nm (maximum power of ~50 W/cm2 for both readers). Ultraviolet OSL signals were detected using an EMI 9235QA (Riso 2) or Electron Tubes PDM 9107B (Riso 7) photomultiplier tube, fitted with 7.5 mm-thick Hoya U-340 filters. Samples were irradiated with mounted 90Sr/90Y beta sources that had been calibrated to administer known doses to multi-grain aliquots and single-grain discs (average single-grain dose rate at the time of measurement = 0.10 Gy/s for Riso 2 and 0.08 Gy/s for Riso 7). For single-grain measurements, spatial variations in the beta dose rate across the disc plane were taken into account by undertaking hole-specific calibrations using gamma-irradiated quartz.

The suitability of the SAR De determination procedure used in this study (**Table S1**) was evaluated by undertaking a series of multi-grain aliquot and single-grain dose-recovery tests on sample LM17188-03. Multi-grain aliquot dose-recovery tests were first used to ascertain optimal preheating conditions for bulk grain populations. These tests were performed on ~100-grain aliquots using a series of different regenerative dose preheat (PH1) conditions (ranging between 200 oC for 10 s and 260 oC for 10 s) and different test dose preheat (PH2) combinations (160 oC for 10 s or 220 oC for 10 s). A known laboratory dose of 100 Gy was applied to groups of three aliquots after optically bleaching their natural OSL signals using two 1,000 s blue LED stimulations separated by a 10,000 s pause (to ensure complete decay of phototransferred charge in the 110 oC TL trap). The administered dose was treated as a surrogate natural dose and subsequently measured using a multi-grain version of the SAR sequence shown in **Table S1**, which involved replacing 125°C green laser stimulations with 125°C blue LED stimulations for 60 s, and inserting a 50°C infrared bleach for 40 s prior to each OSL measurement to remove any feldspar signal contamination. **Figure S1** summarises the results of the multi-grain aliquot dose-recovery tests performed on LM17188-03. In general, the mean measured-to-given dose values of this sample exhibit limited dependency on PH1 and PH2 conditions; six of the seven preheat combinations tested yielded mean measured-to-given dose ratios consistent with unity at 1σ or 2σ, though significant inter-aliquot De scatter is observed for some of these preheat combinations (e.g. PH1 of 260 oC for 10 s and a PH2 of 220 oC for 10 s). The most suitable dose-recovery results were obtained using a PH1 of 220 oC for 10 s and a PH2 of 160 oC for 10 s. This preheat combination yielded a weighted mean measured-to-given dose ratio of 1.02 ± 0.06, the lowest inter-aliquot De scatter of all combinations tested, low-dose and high-dose mean recycling ratios in agreement with unity at 1σ (1.00 ± 0.05 and 1.01 ± 0.04, respectively) and a mean recuperation ratio of less than 1%. This optimum multi-grain preheat combination is very similar to that reported for the nearby Hotel California OSL samples by Arnold et al. (2012a, 2013) (i.e. PH1 of 200 oC for 10 s and a PH2 of 160 oC for 10 s). Santamaría et al. (2021) did not provide any details of the dose-recovery test results obtained in their original multi-grain OSL study of the Fuente Mudarra samples.

To confirm the suitability of the SAR procedure at the single-grain scale, we repeated the dose-recovery test on 1100 individual quartz grains from sample LM17188-03 using the optimum multi-grain preheat conditions (PH1 of 220 oC for 10 s and a PH2 of 160 oC for 10 s). A dose of 200 Gy was administered to these quartz grains after bleaching their natural signals using the same procedure described above. This administered dose was chosen as being broadly similar to the expected mean natural dose of LM17188-03, as determined from preliminary De measurements made on 100 grains of this sample. The single-grain OSL dose recovery test yielded a mean measured-to-given dose ratio of 1.00 ± 0.03 (*n* = 90 accepted grains) and an overdispersion value of 15 ± 3%(**Figure S1**). These dose-recovery results confirm the general suitability of the SAR procedure and quality-assurance criteria (see next paragraph) for single-grain De estimation over the natural dose range. They also provide a minimum estimate of the intrinsic single-grain De scatter and overdispersion that is expected to originate from the laboratory procedures themselves, and from grain-to-grain variations in luminescence responses to the fixed SAR conditions.

Between 2200 and 3600 single-grain De measurements were made for each sample. Sensitivity-corrected dose-response curves were constructed using the first 0.17 s of each OSL stimulation after subtracting a mean background count obtained from the last 0.25 s of the signal. Single-grain OSL De estimates were rejected from further consideration if they exhibited one or more of the following properties: (i) weak OSL signals (i.e., the net intensity of the natural test-dose signal (Tn) was less than three times the standard deviation of the late-light background signal); (ii) poor recycling ratios (i.e., the ratios of sensitivity-corrected luminescence responses (Lx/Tx) for two identical regenerative doses were not consistent with unity at 2σ); (iii) high levels of signal recuperation / charge transfer between SAR cycles (i.e., the sensitivity-corrected luminescence response of the 0 Gy regenerative dose point amounted to >5% of the sensitivity-corrected natural signal response (Ln/Tn) at 2σ); (iv) poorly defined or non-monotonic dose-response curves (i.e., those displaying no discernible dose-response curve, or a zero (flat) or negative response with increasing dose) and dose-response curves displaying very scattered Lx/Tx values (i.e., those that could not be successfully fitted with the Monte Carlo procedure and, hence, did not yield finite De values and uncertainty ranges); (v) saturated or non-intersecting natural OSL signals (i.e., Ln/Tn values equal to, or greater than, the *Imax* saturation limit of the dose-response curve at 2σ); (vi) extrapolated natural signals (i.e. Ln/Tn values lying more than 2σ beyond the Lx/Tx value of the largest regenerative-dose administered in the SAR procedure); (vii) contamination by feldspar grains or inclusions (i.e., the ratio of the Lx/Tx values obtained for two identical regenerative doses measured with and without prior infrared stimulation (OSL IR depletion ratio; Duller, 2003) was less than unity at 2σ). The OSL grain classification statistics obtained for each sample after applying these SAR quality assurance criteria are summarised in **Table S2**.

Individual and sample-averaged De estimates are presented throughout this paper with their 1σ uncertainties, which are derived from three sources of uncertainty: (i) a random uncertainty term arising from photon counting statistics for each OSL measurement, calculated using Eq. 3 of Galbraith (2002); (ii) an empirically determined instrument reproducibility uncertainty of 1.9% or 1.5% for each single-grain measurement (calculated specifically for the two Risø readers used in this study according to the approach outlined in Jacobs et al., 2006a); and (iii) a dose-response curve fitting uncertainty determined using 1000 iterations of the Monte Carlo method described by Duller (2007) and implemented in Analyst v4.

Representative OSL dose-response and decay curve for grains that passed the quality assurance criteria are shown in **Figure S2**. The majority of accepted grains display rapidly decaying OSL curves (reaching background levels within 0.5 s), which are characteristic of quartz signals dominated by the most readily bleached (so-called ‘fast’) OSL component **(Figure S2** – compare OSL decay curve shape for a fast-dominated Risø calibration quartz grain; Hansen et al., 2015). The single-grain OSL dose-response curves are generally well-represented by either a single saturating exponential function or a saturating exponential plus linear function, as has been widely reported for quartz grains with fast-dominated OSL signals (e.g., Yoshida et al., 2000; Jacobs et al., 2008; Arnold et al., 2011, 2016). Individual sensitivity-corrected SAR dose-response curves were fitted with the function (either single saturating exponential or saturating exponential plus linear) that yielded the optimum statistical fit for each grain, which was determined from the sum of the squared fitting residuals divided by the number of dose points used to construct the dose-response curve. On average, 43% of accepted grains per sample display moderately bright Tn (20 Gy) OSL signals of 100-1000 cts/0.17 s (e.g., **Figure S2a**), and 5% of accepted grains per sample have relatively bright Tn OSL signals of >1000 cts/0.17 s (e.g., **Figure S2b**). Approximately half of the accepted grains (52%) have relatively dim Tn signal intensities of <100 cts/0.17 s (e.g., **Figure S2d**), while four of the five samples also contain a small number (≤1%) of very bright accepted grains displaying Tn OSL signal intensities >5,000 cts/0.17 s (e.g., **Figure S2c**). The average Tn OSL signal intensities of grains that passed the SAR quality assurance criteria is 315 cts/0.17 s for the five samples.

**Table 3** summarises the environmental dose rate data for the single-grain OSL samples. Where possible, environmental dose rates have been calculated using the same field gamma spectrometry and beta counting datasets as Santamaría et al. (2021). However, as detailed in the main text, it was necessary to make several amendments to the original dose rate calculations of these samples owing to the limited methodological details presented in Santamaría et al. (2021) and to ensure consistency with the single-grain OSL study performed previously at Hotel California. The external gamma dose rates of all five samples have been determined from *in situ* measurements made using a Canberra LaBr3:Ce detector to account for any spatial heterogeneity in the surrounding (~30 cm diameter) gamma radiation field of each sample. The ‘energy windows’ approach described in Arnold et al. (2012b) and Duval and Arnold (2013) was used to derive individual estimates of U, Th and K concentrations from the field gamma-ray spectra collected in the Santamaría et al. (2021) multi-grain OSL study.

The external beta dose rates of samples LM17188-01, LM17188-03 and LM17188-05 have been determined from measurements made using a Risø GM-25-5 beta counter (Bøtter-Jensen and Mejdahl, 1988) on dried and homogenised, bulk sediments collected directly from the OSL sampling positions. This laboratory-based approach was used to ensure that beta dose rates were derived from sample sizes that more closely approximate the very short (~2-3 mm) beta particle radiation fields affecting these samples. Background-subtracted count rates were measured for three aliquots of each sample and compared with net count rates obtained simultaneously for a loess sediment standard with known U, Th and K concentrations (Potts et al., 2003). Beta counting measurements were either not made directly on samples LM17188-02 and LM17188-04 as part of the Santamaría et al. (2021) study, or the measurements originally made on these samples could not be located during our re-evaluation study. Furthermore, the original beta dose rates and uncertainty terms provided for these two samples were identical (indistinguishable at 3 decimal places) and equated to the average beta dose rate of stratigraphically related samples LM17188-03 and LM17188-05 (see Table 2 of Santamaría et al., 2021), suggesting they could represent assumed (averaged) rather than measured values. Given these uncertainties, it has been necessary to recalculate the external beta dose rates of samples LM17188-03 and LM17188-05 using alternative datasets. This has been achieved using U, Th and K concentrations obtained on the dried and homogenised, bulk sediment fractions via a combination of inductively coupled plasma mass spectrometry (ICP-MS) and inductively coupled plasma optical emission spectrometry (ICP-OES), following four acid digestion preparation. The dry beta dose rates obtained for these two samples using ICP-MS/OES are consistent at 2σ with those reported in Table 2 of Santamaría et al. (2021) after adjusting for grain-size attenuation.

Radionuclide concentrations and specific activities have been converted to dose rates using the conversion factors given in Guérin et al. (2011). The final beta dose rate estimates of all five samples have been calculated after making allowance for beta dose attenuation due to grain-size effects and HF etching (Brennan, 2003). Cosmic-ray dose rates have been calculated using the approach described in Prescott and Hutton (1994), after taking into consideration site altitude, geomagnetic latitude, and density / thickness of sediment overburden. A small, assumed internal (alpha plus beta) dose rate of 0.03 ± 0.01 Gy / ka has been included in the final dose rate calculations based on published 238U and 232Th measurements for etched quartz grains from a range of locations (e.g., Mejdahl, 1987; Bowler et al., 2003; Jacobs et al., 2006b; Pawley et al., 2008; Lewis et al., 2020) and an alpha efficiency factor (a-value) of 0.04 ± 0.01 (Rees-Jones, 1995; Rees-Jones and Tite, 1997).

The beta, gamma and cosmic-ray dose rates have been corrected for long-term sediment moisture contents, according to the approach outlined by Aitken (1985). Owing to the relatively low present-day water contents measured for these samples (8–14% dry weight), Santamaría et al. (2021) estimated the long-term moisture contents using proportional saturation assessments. Santamaría et al. (2021) adopted long-term moisture contents of 38–56% (equivalent to 60% of the present-day saturated water contents), which are significantly higher than values reported or assumed for well-preserved silty clay deposits at Atapuerca open-air localities (e.g. Moreno et al., 2012; Arnold et al. 2013). These assumed values are also systematically higher than the present-day water contents reported for freshly preserved silty clay deposits inside the deeper and closed parts of the Atapuerca endokarst system (Arnold et al., 2014, 2015; Demuro et al., 2014, 2019a, 2019b, submitted). The latter is at odds with observations that Atapuerca cave clastic deposits are generally exposed to more humid ambient conditions and are better buffered against seasonal, interannual and longer term dessication cycles (Martín-Chivelet et al., 2011) compared to near-surface, open-air deposits in the surrounding foothills. We consider it unlikely that the Fuente Mudarra deposits consistently experienced water contents in excess of 45% dry weight for their entire burial periods (i.e. the mean assumed value of the five samples from Santamaría et al., 2021); especially given that the burial periods of these samples spanned several glacial stages and pronounced stadial events, which were characterised by significantly reduced effective moisture conditions (e.g. Pellitero et al., 2019; Valero-Garces et al., 2019; Pérez-Díaz and López-Sáez, 2021).

To ensure consistency with luminescence and electron spin resonance dating studies undertaken previously at Atapuerca, including the single-grain OSL study performed at Hotel California, we have adopted more conservative long-term water contents of 19–28% dry weight (equivalent to 30% of the present-day saturated water content estimates), which overlap with the Hotel California long-term water content estimates of 14–26% dry weight at 2σ (Arnold et al., 2013). As the in situ gamma spectrometry measurements for these samples were made under present-day moisture conditions (and hence were already attenuated for sediment water contents of 8–14% dry weight), it was first necessary to correct the as-measured radionuclide concentrations back to dry (i.e. 0% water content) equivalent values before applying the long-term water content corrections of 14–26%. Santamaría et al. (2021) did not indicate whether this necessary step was included in their long-term water-corrected gamma dose rate calculations for the multi-grain OSL study of the Fuente Mudarra samples. A relative uncertainty of 10% has been assigned to our long-term moisture estimates to accommodate any minor variations in hydrologic conditions during burial.

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| --- |
| **Single-grain OSL SAR procedure** |
| **Step** | **Treatment** | **Symbol** |
| 1 | Dose (Natural or laboratory) | N or D |
| 2 a | IRSL stimulation (50ºC for 60 s) |  |
| 3 | Preheat 1 (220ºC for 10 s) | PH1 |
| 4 | Single-grain OSL stimulation (125ºC for 2 s) | Ln or Lx |
| 5 | Test dose (20 Gy) | Td |
| 6 | Preheat 2 (160ºC for 10 s) | PH2 |
| 7 | Single-grain OSL stimulation (125ºC for 2 s) | Tn or Tx |
| 8 | Repeat measurement cycle for different |  |
|  | sized regenerative doses |  |

a Step 2 is only included in the single-grain SAR procedure when measuring the OSL IR depletion ratio (Duller 2003).

**Table S1** Single-aliquot regenerative-dose (SAR) procedures used for dose-recovery measurements and De determination. Each of these SAR measurement cycles was repeated for the natural dose, 5 different sized regenerative doses and a 0 Gy regenerative-dose (to measure OSL signal recuperation). Both the smallest and second largest non-zero regenerative-dose cycles were repeated at the end of the SAR procedure to assess the suitability of the test-dose sensitivity correction. The smallest regenerative-dose cycle was also repeated a second time with the inclusion of step 2 to check for the presence of feldspar contaminants using the OSL IR depletion ratio of Duller (2003). Lx = regenerative dose signal response; Ln = natural dose signal response; Tx = test dose signal response for a laboratory dose cycle Tn = test dose signal response for the natural dose cycle.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Sample name**  | **LM17188-01** | **LM17188-02** | **LM17188-03** | **LM17188-03** | **LM17188-04** | **LM17188-05** |
| **SAR measurement type** | De | De | De | Dose-recovery | De | De |
| **Total measured grains (*n*)** | 2300 | 2700 | 3600 | 1100 | 2200 | 2300 |
| **Grains rejected for failing SAR quality assurance criteria (%)** |  |  |  |  |  |  |
| Weak OSL signals (Tn <3*σ* background) | 42 | 40 | 44 | 50 | 31 | 28 |
| Low-dose recycling ratio ≠ 1 at ±2*σ* | 6 | 6 | 5 | 5 | 7 | 7 |
| High-dose recycling ratio ≠ 1 at ±2*σ* | 4 | 5 | 4 | 5 | 4 | 5 |
| OSL-IR depletion ratio <1 at ±2σ | 6 | 6 | 6 | 4 | 7 | 7 |
| 0 Gy Lx/Tx >5% Ln/Tn  | <1 | <1 | <1 | <1 | <1 | <1 |
| Non-intersecting grains (Ln/Tn > dose response curve saturation) | 2 | 2 | 2 | 0 | 2 | 3 |
| Saturated grains (Ln/Tn ≥ dose response curve *Imax* at ±2σ) | 2 | 3 | 2 | 1 | 3 | 3 |
| Extrapolated grains ( Ln/Tn values >2σ beyond largest Lx/Tx value) | 0 | <1 | <1 | 0 | <1 | <1 |
| Anomalous or no dose response / unable to perform Monte Carlo fit | 29 | 29 | 30 | 27 | 36 | 35 |
| **Sum of rejected grains (%)** | 91 | 91 | 93 | 92 | 91 | 89 |
| **Sum of accepted grains (%)** | 9 | 9 | 7 | 8 | 9 | 11 |

**Table S2** Single-grain OSL classification statistics for the dose recovery and natural De measurements. The proportion of grains that were rejected from final De estimation after applying the various SAR quality assurance criteria are shown in rows 5-13. These criteria were applied to each single-grain measurement in the order listed. Tn = natural test dose signal response; Ln/Tn = sensitivity-corrected natural signal response; Lx/Tx = sensitivity-corrected regenerative-dose signal response; *Imax* = saturation OSL intensity of the fitted dose response curve.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Sample** **name** | **Strat.****level** | **Grain size****(μm)** | **Total****dose rate****(Gy/ka) a** | **Equivalent dose (De) data** | **OSL age** **(ka) a,f** |
| **De type b** | **No. of****grains c** | **Over-****dispersion****(%) d** | **Age** **Model e** | **De****(Gy) a** |
| LM17188-01 | FM4 | 180 – 212 | 2.22 ± 0.12 | SG OSL | 207 / 2300 | 50 ± 3 | CAM | 188 ± 8  | 84.7 ± 5.9 |
|  |  |  |  | Synth. Al. MG OSL | 21 / 23 | 35 ± 7 | CAM | 290 ± 25 | 130.8 ± 13.4 |
| LM17188-02 | FM5 | 180 – 212 | 2.26 ± 0.12 | SG OSL | 240 / 2700 | 45 ± 3 | CAM | 202 ± 7 | 89.2 ± 6.1 |
|  |  |  |  | Synth. Al. MG OSL | 25 / 27 | 31 ± 6 | CAM | 386 ± 28 | 170.3 ± 15.7 |
| LM17188-03 | FM6 | 180 – 212 | 2.25 ± 0.12 | SG OSL | 253 / 3600 | 35 ± 3 | CAM | 251 ± 7 | 111.5 ± 7.2 |
|  |  |  |  | Synth. Al. MG OSL | 34 / 36 | 33 ± 5 | CAM | 331 ± 21 | 147.0 ± 12.7 |
| LM17188-04 | FM7 | 180 – 212 | 2.28 ± 0.12 | SG OSL | 187 / 2200 | 32 ± 3 | CAM | 264 ± 9 | 116.2 ± 7.5 |
|  |  |  |  | Synth. Al. MG OSL | 19 / 22 | 20 ± 5 | CAM | 313 ± 18 | 137.8 ± 11.1 |
| LM17188-05 | FM8 | 180 – 212 | 2.32 ± 0.13 | SG OSL | 249 / 2300 | 32 ± 3 | CAM | 276 ± 8 | 119.2 ± 7.6 |
|  |  |  |  | Synth. Al. MG OSL | 21 / 23 | 12 ± 4 | CAM | 352 ± 14 | 151.8 ± 10.6 |

a Mean ± total uncertainty (68% confidence interval), calculated as the quadratic sum of the random and systematic uncertainties.

b SG OSL = Single-grain OSL; Synth. Al. MG OSL = Synthetic aliquot multi-grain OSL.

c Number of De measurements that passed the SAR rejection criteria and were used for De determination / total number of grains analysed. See Table S2 for single-grain rejection and classification statistics. For each synthetic aliquot dataset, 2-3 De measurements were rejected per sample owing to having saturated, non-intersecting or extrapolated normalised natural OSL signals (i.e., they failed SAR quality assurance criteria (v) or (vi), which precluded finite De interpolation).

d Relative spread in the De dataset beyond that associated with measurement uncertainties of individual De values, calculated using the central age model.

e Age model used to calculate the sample-averaged De value for each sample. The choice of age model for each sample has been made on statistical grounds using the maximum log likelihood score (*Lmax*) criterion outlined by Arnold et al. (2009). CAM = central age model.

r Total uncertainty includes a systematic component of ±2% associated with laboratory beta-source calibration.

**Table S3** Comparison of single-grain OSL results and synthetic aliquot OSL results (equivalent to multi-grain aliquots containing 100-grains each) for the Fuente Mudarra samples. The synthetic aliquot De values were obtained by summing the signals of all accepted and rejected grain types on each single-grain disc.



(a)



(b)

**Figure S1** Multiple-grain and single-grain OSL dose-recovery test results for sample LM17188-03. (a) Measured-to-given dose ratios versus regenerative dose preheat (PH1) and test dose preheat (PH2) temperature (held for 10 s) for ~10-grain aliquots. The natural OSL signals of the multi-grain aliquots were optically bleached with two 1000 s blue LED illuminations at ambient temperature, each separated by a 10 000 s pause. A known dose of 100 Gy was then administered to each aliquot and a multi-grain aliquot version of the SAR procedure shown in **Table S1** was subsequently used to estimate this dose (replacing 125°C green laser stimulations with 125°C blue LED stimulations for 60 s, and inserting a 50°C IR bleach for 40 s prior to each OSL measurement to remove any feldspar signal contamination). (b) Measured-to-given dose ratios obtained for individual quartz grains of sample LM17188-03 in the single-grain SAR dose-recovery test. The grey shaded region is centred on the administered dose for each grain (sample average = 200 Gy). Individual De values that fall within the shaded region are consistent with the administered dose at 2σ.





D0 = 164 ± 7 Gy

De = 221 ± 21 Gy

D0 = 169 ± 17 Gy

De = 281 ± 59 Gy

(a)

(b)





D0 = 125 ± 4 Gy

De = 295 ± 37 Gy

D0 = 298 ± 56 Gy

De = 174 ± 40 Gy

(d)

(c)

**Figure S2** Representative single-grain OSL decay and dose-response curves for quartz grains from sample LM17188-05. The decay curve of a fast-component dominated calibration quartz grain is shown for comparison (Risø calibration quartz standard from Rømø, batch #98; Hansen et al., 2015). In the insets, the open circle denotes the sensitivity-corrected natural OSL signal, and filled circles denote the sensitivity-corrected regenerated OSL signals. The D0 value characterises the rate of signal saturation with respect to administered dose and equates to the dose value for which the saturating exponential dose-response curve slope is 1/*e* (or ~ 0.37) of its initial value. (a) grain with moderate OSL signal brightness (Tn intensity = 100–1,000 counts / 0.17 s). (b) grain with bright OSL signal (Tn intensity = ~1,000–5,000 counts / 0.17 s). (c) grain with very bright OSL signal (Tn intensity = >5,000 counts / 0.17 s). (d) grain with relatively dim OSL signal (Tn intensity = <100 counts / 0.17 s).