Refining the chronology of Acheulean deposits at Porto Maior in the River Miño basin (Galicia, Spain) using a comparative luminescence and ESR dating approach

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Abstract

Trapped charge dating of optically bleached quartz, which involves techniques such as electron spin resonance (ESR) and thermally transferred optically stimulated luminescence (TT-OSL), is increasingly being used to establish Middle and Early Pleistocene chronologies for archaeological, palaeontological and palaeoenvironmental sedimentary sequences. Comparative dating studies that make use of several independent or semi-independent radiometric techniques in tandem are an invaluable means of assessing the reliability of optically bleached quartz dating methods and ascertaining empirical relationships between different quartz dating signals. In this study, we undertake a comparative dating evaluation of single-grain TT-OSL and various ESR signals that exhibit different optical resetting properties (Al, Ti-Li and, where measurable, Ti-H centres) at the Middle Pleistocene archaeological site of Porto Maior, northwest Spain, which contains a distinctive, regionally significant Acheulean stone tool assemblage. We expand the existing chronological framework for Porto Maior using single-grain TT-OSL dating, derive combined numerical ages for individual depositional units using Bayesian modelling, and further examine the relationship between ESR and TT-OSL signals by reviewing a range of published comparative dating studies. The new single-grain TT-OSL ages obtained for unit PM3 at Porto Maior (225 ± 31 ka and 242 ± 32 ka) are in close agreement with previously published ESR Ti-Li ages for this unit (266 ± 23 ka and 279 ± 26 ka) and significantly younger than those reported for the Al centre (441 ± 75 ka and 452 ± 62 ka). Our broader comparison of published TT-OSL – ESR dating results also shows statistically significant agreement between single-grain TT-OSL and Ti-Li centre ages in other sedimentary settings, suggesting a potentially closer relationship between the TT-OSL-producing traps and the Ti-Li paramagnetic centres in quartz, rather than between the TT-OSL and Al centre signals. The Bayesian modelling results for Porto Maior enable us to refine the existing bracketing depositional age for the archaeological accumulation at Porto Maior and provide a stronger statistical foundation for chronostratigraphic correlations with other Acheulean archaeological deposits found in the Miño River basin. The Bayesian model indicates that the entire Porto Maior sedimentary sequence was deposited between 312.6 ± 32.6 ka and $18.8 \pm$

7.7 ka, and that the *in situ* large cutting tool assemblage from level PM4 most likely accumulated during Marine Isotope Stage 7 (mean depositional age = 210.7 ± 24.7 ka). The results of this study reinforce the usefulness of applying the multiple centre (MC) ESR dating approach in many dating contexts, and demonstrate the value of undertaking comparisons between semi-independent quartz dating signals in order to maximise the reliability of ESR and single-grain TT-OSL quartz chronologies.

1. Introduction

The last two decades have seen major advances in the development of trapped charge dating methods that can be applied to optically bleached sedimentary quartz from Early and Middle Pleistocene archaeological, palaeontological and palaeoenvironmental sites (Toyoda et al., 2000; Voinchet et al. 2004; Wang et al., 2006; Duller and Wintle, 2012; Arnold et al., 2015; Duval et al., 2015; Wintle and Adamiec, 2017). In particular, quartz dating techniques such as electron spin resonance (ESR) and thermally transferred optically stimulated luminescence (TT-OSL) represent invaluable geochronology tools for a wide range of Quaternary research applications, including archaeological dating, as they (i) make use of ubiquitously preserved sedimentary materials (silicate minerals), (ii) cover an important timeframe in human evolution (>1 Ma – 100 ka) that is typically outside the range of alternative dating techniques (or for which the application of other dating methods is often not practical due to the absence of suitably preserved materials), and (iii) provide numerical age estimates instead of relative chronologies (unlike palaeomagnetism or biochronology).

ESR dating was first applied to optically bleached quartz during the mid-1980s (Yokoyama et al., 1985) but its use over the following two decades remained limited as ESR quartz dating applications continued to focus on volcanic and intra-fault contexts (see review by Grün, 1989). Methodological research undertaken during these early years did, however, pave the way for an improved general understanding of quartz ESR signals used in optically bleached dating contexts, particularly with regards to thermal stability considerations and optical bleaching kinetics (e.g. Toyoda and Ikeya, 1991; Falguères et al., 1991; Walther and Zilles, 1994). ESR dating of optically bleached quartz from Quaternary sedimentary deposits has traditionally focused on the use of the aluminium (Al) centre (e.g., Falguères, 1986; Laurent, 1993; Tanaka et al., 1995; Yoshida, 1996; Voinchet, 2002 and references therein), which is related to the most abundant trace element in quartz (Preusser et al., 2009) and gives rise to a strong ESR signal when measured at liquid nitrogen temperature. However, the relatively slow bleaching kinetics of the Al centre and the presence of a residual (optically unbleachable) Al signal component (Walther and Zilles, 1994) has led to increasing use of alternative quartz ESR dating signals, particularly titanium (Ti) centre signals in conjunction with Al centre signals (Toyoda et al., 2000), with encouraging results (e.g. Rink et al., 2007; Tissoux et al., 2008). Comparative chronological studies involving independent or semi-independent numerical dating methods (e.g., luminescence, Ar-Ar, combined U-series/ESR dating of fossil teeth) have also contributed to continued improvements in the reliability of ESR quartz dating methods over the last decade (e.g., Rink et al., 2007; Moreno et al., 2015; Arnold et al., 2014, 2015; Bartz et al., 2019; Duval et al., this volume). As a result, ESR dating of optically bleached quartz has now been reliably applied to a large number of archaeological records and associated terrestrial archives (typically fluvial deposits) from Europe (Voinchet et al. 2015; Moreno et al., 2015; Duval et al., 2017; Pereira et al., 2018;

Bahain et al., this volume; Duval et al., this volume), Asia-Oceania (Beerten et al., 2006; Liu et al., 2013; 2014; Ingicco et al., 2018) and northern Africa (Sahnouni et al., 2018), among other regions.

In parallel, there has been an increase in TT-OSL dating applications in archaeological and geomorphological contexts over the last decade (see review by Arnold et al. 2015). TT-OSL dating originally emerged as an alternative luminescence dating technique with the potential to extend the traditional upper age limit of quartz OSL (Wang et al., 2006). TT-OSL signals are measured after optically removing the conventional OSL signal and heating the sample (typically to ≥ 260 °C) to induce a transfer of charge from the TT-OSL electron trap into the main OSL dating trap (Wintle and Duller, 2012). The dose saturation limits of the TT-OSL signal is typically an order of magnitude higher than for conventional OSL and at least double that reported for pIR-IRSL (Arnold et al., 2015), permitting finite age determination over Middle Pleistocene timescales (e.g., Jacobs et al., 2011; Arnold et al., 2014). Several different TT-OSL dating protocols have been developed to enable burial dose estimation using individual aliquots composed of hundreds or thousands of quartz grains (e.g., Wang et al., 2007; Tsukamoto et al., 2008; Porat et al., 2009; Stevens et al., 2009; Adamiec et al., 2010; Jacobs et al., 2011). Recent developments in TT-OSL research have seen the emergence of single-grain dating approaches (e.g., Arnold et al., 2013, 2014; Demuro et al., 2015), which offer a number of potential advantages over traditional multi-grain TT-OSL approaches for quartz samples with inherently bright luminescence signals. Such benefits include the ability to isolate grains with favourable dating properties (e.g., high dose saturation limits, charge transfer into ‘fast’ component OSL traps), the detection of inter-grain differences in problematic TT-OSL behaviours (including low thermal stabilities), and a means of circumventing averaging effects arising from simultaneously measuring grains with different bleaching histories, signal compositions or TT-OSL source trap properties (Arnold et al., 2014; Arnold and Demuro, 2015; Arnold et al. 2019).

The successful application of both ESR and TT-OSL dating methods depends on adequate signal bleaching by sunlight prior to the burial event being dated. This is an important consideration in any dating study because the resetting of ESR and TT-OSL signal down to low or constant residual levels has been shown to require up to several weeks or months of natural daylight exposure (e.g., Voinchet et al., 2003; Jacobs et al., 2011; Demuro et al., 2015; Duval et al., 2017). Since incomplete signal resetting or residual subtraction can lead to non-trivial age overestimations, it is necessary to undertake site-specific assessments of bleaching sufficiency to ensure meaningful interpretations of resultant ESR or luminescence chronologies. For TT-OSL dating studies, single-grain applications offer an ideal opportunity to better characterise TT-OSL signal resetting at the most fundamental scale of analysis (e.g., Arnold et al., 2019). Assessments of TT-OSL bleaching adequacy can also be undertaken via dating comparisons with more rapidly bleaching luminescence signals (e.g., post infrared infrared stimulated luminescence (pIR-IRSL and OSL signals); with parity in ages being used to support adequate resetting of the slower bleaching TT-OSL signal. In ESR dating studies, the multiple centre (MC) approach (Toyoda et al., 2000) has progressively become a standard requirement for assessing bleaching suitability prior to sediment deposition (see a recent overview in Duval et al., 2017). This comparative dating approach takes advantage of the differential bleaching rate of ESR signals associated with different light-sensitive paramagnetic centres in quartz (see bleaching kinetics comparison in Fig. 1 from Duval et al., 2017):

namely, the slowly and incompletely bleached Al centre and the more rapidly (and fully) bleached Ti centres (Ti-Li and, if measurable, Ti-H centres). As part of the MC approach, various ESR signals are measured in a given quartz sample and, under ideal bleaching and transport conditions, they are expected to provide similar estimates of the dose absorbed by the sample during its burial history (e.g., see Sahnouni et al., 2018, and Voinchet et al., this volume, for examples of consistent Al and Ti ESR MC ages). Statistically significant differences in the resultant ESR chronologies of different paramagnetic centres are usually interpreted as indicating that the slowly bleaching Al centre provides a maximum age for the deposits, with the rapidly bleaching Ti centres most likely providing the closest estimate for the true age of sediment deposition (Toyoda et al., 2000; Tissoux et al., 2007; Voinchet et al., 2003; Duval et al., 2017).

The MC (Al and Ti) approach has recently been applied in a combined ESR-luminescence quartz dating study of the Porto Maior archaeological site, which is set in a fluvial terrace sequence of the Miño River (Galicia, northwest Spain; Méndez-Quintas et al., 2018). In that study, the ESR Al centre ages for the lower half of the fluvial sequence were found to be more than 50% older than the corresponding Ti-Li centre ages. The Ti centre ages at Porto Maior appear to be well-supported by paired post-infrared infrared stimulated luminescence (pIR-IRSL) dating of potassium feldspar grains from closely associated samples (Méndez-Quintas et al., 2018), with both techniques converging on an age of ~240 ka for the main archaeological layer. This site provides an ideal opportunity for evaluating the usefulness of the ESR MC approach and for further investigating the age disparities obtained for the ESR Al and Ti centres by applying additional extended-range dating techniques such as single-grain TT-OSL. It also represents a suitable context for exploring the potential relationships between different quartz trapped charge dating signals (paramagnetic and luminescence), and for examining the potential and pitfalls of using these signals to resolve Middle Pleistocene human occupation histories at other archaeological sites in the region. These various issues are explored further in the present study.

Porto Maior represents the first reported Acheulean site in Europe to be characterised by an extensive accumulation of large cutting tools (LCTs). The density of LCTs at this site is higher than at any other western European site, and parallels that recorded at several sites from Africa and the Near East. Similarly, the large average size of the LCTs (predominantly handaxes) from Porto Maior, as well as their technological attributes, are comparable to those commonly found in Africa and the Near East (Méndez-Quintas et al., 2018). These shared lithic assemblage characteristics have been interpreted as representing evidence for a direct link between the Acheulean tradition from Africa and that of southwest Europe (Méndez-Quintas et al., 2018). Refining the accuracy and precision of existing age estimates at Porto Maior is therefore important for better understanding the chronological context of this important Lower Palaeolithic site within a regional archaeological framework.

Building on the recent archaeological evaluation of Porto Maior by Méndez-Quintas et al. (2018), the present study aims to undertake additional chronological assessments of the site using luminescence and ESR quartz dating comparisons. The specific aims of the paper are to: (i) expand the existing dating framework by obtaining new single-grain TT-OSL ages from selected Porto Maior samples and compare these with corresponding ESR Al and Ti quartz ages; (ii) derive a Bayesian modelled chronostratigraphic sequence for

Porto Maior in order to derive combined ages for individual depositional units at the site, refine the timing of the *in situ* archaeological accumulation, and provide a stronger statistical foundation for future chronostratigraphic correlations across the basin; and (iii) examine the comparative optically bleached quartz dating results obtained from Porto Maior and a range of other published sites to better evaluate the empirical relationship between the bleaching properties and corresponding ages of ESR and TT-OSL signals in different sedimentary contexts.

2. Study site, stratigraphy, lithic assemblage and existing chronology

The archaeological site of Porto Maior is located along the northern margins of the Miño River (Galicia, Spain) on a fluvial terrace found +34 m above the current river level (terrace T4 of the local fluvial terrace system). The >6 m stratigraphic sequence is made up of five stratigraphic levels, characterised from bottom to top as follows: levels PM1 and PM2 – fluvial facies composed of clast-supported gravels; levels PM3 and PM4 – fluvial facies composed of massive fine-grain (silts and sands) overbank deposits affected by pedogenesis, and; level PM5 – comprises a colluvium gravel deposit at its base (with re-deposited lithic industry) and fine loam sediments of aeolian origin in the upper section (**Figure 1**). The *in situ* anthropological accumulation of lithic tools at Porto Maior has been recovered from an 11.8 m² excavated area at the base of PM4, which sits uncomfortably on PM3. In total, 290 lithic artefacts have been recovered from this fine-grain layer, of which 159 show no evidence of fluvial abrasion and are considered to represent localised human activity (denoted the main assemblage or MA in Méndez-Quintas et al., 2018). The LCT lithic assemblage has been described as large flake blank (LFB *sensu* Sharon) Acheulean and is dominated by handaxes, although it also contains cleavers and picks. The LCTs are among the largest reported for the Iberian Peninsula and Europe (mean length of handaxes = 186.4 cm). and constitute the densest Acheulean assemblage published thus far for an European site (9.5 pieces per m²). As noted in the Introduction section, both the technological attributes (including size) of the LCTs and accumulation patterns of the Porto Maior assemblage closely match those usually described for African and Near Eastern sites, potentially indicating an African affinity in the Acheulean tradition of southwest Europe. There are no preserved organic or faunal remains at Porto Maior (a common occurrence for open-air lower Palaeolithic sites in the region; e.g. Méndez-Quintas et al., in press), which precludes biochronological assessments of the archaeological sequence.

The existing chronology for the Porto Maior sedimentary sequence has been determined using a combination of three semi-independent quartz dating methods (**Table 1**): ESR dating of six quartz samples from units PM1, PM3, PM4 and PM5; potassium feldspar pIR-IRSL dating of four samples from units PM3, PM4 and PM5 using a stimulation temperature of 225°C and a preheat of 250°C for 60 s (herein denoted pIR-IRSL₂₂₅); and replicate single-grain OSL quartz dating of the luminescence sample collected from unit PM5 (Méndez-Quintas et al., 2018). The MC approach has previously been applied to each of the ESR quartz samples, resulting in two replicate ages per sample using the Al and Ti-Li (option D; Duval and Guilarte, 2015) centres. The suitability of the Ti-H signal has been additionally evaluated for the Porto Maior ESR samples by Méndez-Quintas et al. (2018). The five samples from the fluvial units (PM1, PM3 and PM4) did not yield sufficiently reliable Ti-H data to enable calculation of meaningful burial dose estimates. However, the uppermost sample collected from

the colluvial and aeolian deposits of unit PM5 did produce a measurable Ti-H signal, and the resultant age was considered in the final dating evaluations of Méndez-Quintas et al. (2018), albeit with caveats (see details in section 3.2).

For the lower half of the sedimentary sequence (i.e., levels PM1 and PM3), the ESR ages from the Al centre were significantly (i.e., 58–70%) older than those obtained for the Ti-Li centre. However, for the upper part of the sequence (i.e., levels PM4 and PM5), the Al and Ti-Li age estimates were in agreement at 2σ (**Table 1**). In general, Méndez-Quintas et al. (2018) interpreted the ESR ages obtained using the Al centre as maximum (*terminus post quem*) ages due to the slow and incomplete bleaching characteristics of the Al signal (Duval et al., 2017), and considered the chronology derived using the more rapidly and completely bleachable Ti-Li centre as the most accurate estimate for the timing of sediment deposition. This assessment was confirmed by the pIR-IRSL₂₂₅ ages obtained for levels PM3 and PM4 (231 ± 15 ka to 268 ± 24 ka), which were in closer agreement with corresponding ESR ages obtained for the Ti-Li centre (206 ± 20 ka to 279 ± 26 ka) than those from the Al centre (181 ± 25 ka to 451 ± 66 ka). The only ESR age available for level PM1 (264 ± 27 ka, based on the Li-Ti centre) indicates that the basal unit of the sediment sequence was likely deposited during marine isotope stage (MIS) 8. Using the various ESR Ti-Li and pIR-IRSL ages, Méndez-Quintas et al. (2018) calculated weighted mean ages of 268 ± 12 ka and 226 ± 10 ka for units PM3 and PM4, respectively. The uppermost unit (PM5) was dated to MIS 2 using both single-grain OSL and pIR-IRSL. In contrast, the ESR Ti-H, Ti-Li and Al centres yielded systematically older ages of between 48 ± 10 ka and 138 ± 25 ka for related deposits in the same unit (**Table 1**), which were interpreted as maximum age estimates for PM5. The difference between the ESR and luminescence chronologies for this unit were considered to primarily reflect the limitations of applying ESR quartz dating over low natural dose ranges (<100 Gy) when using the multiple aliquot additive dose method; although minor differences in the ESR and luminescence sampling positions within unit PM5 may have also contributed to these age inconsistencies. The difference in the combined age estimates of units PM4 and PM5 indicates the presence of a prolonged sedimentary hiatus at Porto Maior during the Late Pleistocene, which is consistent with a major change in sedimentary processes observed between levels PM4 and PM5 (Méndez-Quintas et al., 2018). The existing age range for the human occupation layer at the base of level PM4 is 293–205 ka, based on the 2σ confidence interval of the weighted mean Ti centre and pIR-IRSL ages for units PM3-PM4.

3. Methods

3.1. TT-OSL dating of sedimentary quartz grains from Porto Maior

As part of an expanded quartz dating study of Porto Maior, we have applied single-grain TT-OSL to all four of the luminescence dating samples (PM16-3, PM16-4, PM16-5 and PM16-6) that were previously dated using pIR-IRSL₂₂₅ (Méndez-Quintas et al., 2018). Samples PM16-3 and PM16-5 come from the lower and upper part of unit PM3, respectively, and are closely associated with ESR samples VI1205 and VI1204. Samples PM16-6 and PM16-4 come from units PM4 and PM5, respectively, and are associated with ESR samples MIN1402 and MIN1401 (**Table 1**). Measurements of quartz TT-OSL signals were made using a Risø TL/OSL-DA-20 reader equipped with a spatially calibrated $^{90}\text{Sr}/^{90}\text{Y}$ β radiation source delivering a mean dose rate of ~ 0.106 Gy/s. The

TT-OSL signal of individual grains were stimulated using a 10 mW Nd:YVO4 laser attachment (532 nm, maximum power of $\sim 50 \text{ W cm}^2$) at 90% power. Measurements were performed on 90-125 μm quartz grains carefully brushed onto aluminium discs drilled with an array of $300 \times 300 \mu\text{m}$ holes. This type of ‘pseudo’ single-grain approach was chosen because the proportion of grains yielding TT-OSL signals is relatively low for these samples (74–81% of grain-hole positions did not produce any statistically distinguishable TT-OSL T_n signal; **Table S2**). It is estimated that ~ 18 grains were placed in each hole position when using this configuration (Arnold et al., 2012a), but the low frequency of TT-OSL-producing grains for these samples (see section 4.1) effectively ensures single-grain resolution has been maintained in this study (Demuro et al., 2013). Ultraviolet emissions were detected using an EMI 9235QB photomultiplier fitted with a 7.5 mm-thick U-340 filter.

TT-OSL D_e values were obtained using the single-aliquot regenerative-dose (SAR) protocol of Stevens et al. (2009), which has been modified to enable the measurement of individual grains (i.e., to enable single-grain measurements in steps 3, 5, 9 and 11 of **Table 2**; Demuro et al., 2014). Net L_x and T_x values were calculated by integrating the first 0.15 s of the TT-OSL decay curves and subtracting a background count from the last 0.25 s of the signal. The SAR dose-response curves were fitted using a single-saturating exponential function. Individual D_e values were accepted if they passed the following quality assurance criteria: (i) the luminescence signals were statistically distinguishable from background (the net intensity of the natural test dose signal, T_n , was $>3\sigma$ above the late-light background signal); (ii) the recycling ratio (i.e., sensitivity-corrected luminescence responses (L_x/T_x) for two identical high regenerative doses) was consistent with unity at 2σ ; (iii) the recuperation ratio, calculated as the ratio of the sensitivity-corrected 0 Gy dose point (L_0/T_x) to the sensitivity-corrected natural (L_n/T_n), was $<5\%$; (iv) no contamination by feldspar grains or inclusions was detected using the OSL IR depletion ratio (i.e., the ratio of the L_x/T_x values obtained for two identical regenerative doses measured with and without prior IR stimulation overlapped with unity at 2σ ; Duller, 2003); (v) the net T_n signal had a relative error of $<30\%$; (vi) the sensitivity-corrected natural signal intercepted the sensitivity-corrected dose-response curve and it did not intercept the saturated part of the dose-response curve (i.e., the L_n/T_n value was lower than the I_{max} saturation limit of the dose-response curve at 2σ); (vii) the dose-response curve did not display anomalous properties (e.g., zero or negative responses with increasing dose) and resulted in good Monte Carlo fits.

Dose recovery tests were performed on sample PM16-3 after administering a dose of $578 \pm 4 \text{ Gy}$ on top of the naturally accumulated dose for a sub-set of grains (see Demuro et al., 2019 for full details of procedure). A net dose recovery (measured-to-given dose) ratio of 0.98 ± 0.26 was obtained after subtracting the weighted mean natural D_e of sample PM16-3 (**Table 3**) from the weighted mean D_e of the unbleached and dosed grains, and dividing this value by the given dose (**Table S1**). Although this dose recovery result is based on a limited number of grains ($n = 18$), the ratio obtained is within 2σ of unity and supports the suitability of the chosen measurement conditions for dating these samples.

The environmental dose rates used to calculate the final single-grain TT-OSL ages of samples PM16-4, PM16-6, PM16-5 and PM16-3 have been obtained using a combination of *in situ* and laboratory measurements (**Table 3**). Gamma dose rates have been derived from *in situ* gamma-ray spectrometry measured immediately after collecting the luminescence dating samples (using the windows approach; Arnold et al., 2012b). Additionally, sediment sub-samples were collected from each sample position to assess sample moisture content and beta dose rate values. The water content values (expressed as percentage of dry weight) were set to between 20% and 29% for samples PM16-5, PM16-3 and PM16-6 (from levels PM3 and PM4) and 15% for sample PM16-4 (from the uppermost level PM5). These water content values were equivalent to those used for calculating pIR-IR and ESR ages reported in Méndez-Quintas et al. (2018). Beta dose rates were determined from low-level beta counting measurements undertaken on dry and homogenised sediment samples (Bøtter-Jensen and Mejdahl, 1988). The contribution from cosmic rays was calculated using the approach outlined in Prescott and Hutton (1994). Full details of the dose rate measurement conditions and calculations can be found in the footnotes of **Table 3**.

3.2 Bayesian age modelling methodology

Bayesian age modelling has been used to combine the reliable luminescence and ESR dating results (likelihoods) from the present study and Méndez-Quintas et al. (2018) with all known stratigraphic information (priors) for Porto Maior, in order to derive a unified chronostratigraphic framework. The specific aims of the Porto Maior Bayesian modelling assessments are threefold: (i) to calculate amalgamated (posterior) probabilistic chronologies for individual stratigraphic units; (ii) to improve the existing age estimate for the *in situ* archaeology-bearing deposits, which is currently based on the weighted mean age estimate of bracketing Ti centre and pIR-IRSL ages from units PM3–PM4, and; (iii) to undertake sensitivity analyses of different types of dating uncertainty representation within OSL and ESR chronological modelling frameworks. The Porto Maior Bayesian age model has been constructed using OxCal v4.3.2 (Bronk-Ramsey, 2009a), and is based on the approach outlined in Hamm et al. (2016) and Demuro et al. (2019). A *sequence* deposition model with nested sub-*sequences* of uniform prior duration and associated *boundaries* has been used to capture the chronostratigraphic features of the site. The main depositional *sequence* incorporates four stratigraphic units in an ordered succession according to depth (units PM1-2, PM3, PM4 and PM5, from the base of the sequence upwards). Units PM1 and PM2 are represented as a combined unit in the model as they are sedimentologically similar, comprise archaeologically sterile fluvial facies, and they are collectively constrained by only a single likelihood age estimate (ESR sample MIN1403 from PM1). Individual stratigraphic units have been represented as nested *sequences* within the broader depositional framework since their overall stratigraphic ordering is sufficiently well-preserved. The relative ordering of dating samples from individual units is also sufficiently clear and therefore all likelihood groupings have been modelled as single *sequences* rather than *sub-phases*. *Boundaries* have been used to delineate the beginning and end of each stratigraphic unit. The entire site *sequence* has been constrained with a minimum age of 0 years for the uppermost *boundary* (above the PM5 *sequence*) and a maximum age of 424 ka for the lowermost boundary (below the PM1-2 *sequence*). The latter is based on the ESR Al-centre age for sample MIN1403, which provides a maximum age estimate for the deposition of unit PM1 according to the MC ESR dating approach (Méndez-Quintas et al., 2018).

Fifteen numerical dating results obtained using five different luminescence and ESR signals (ESR Ti-Li centre, ESR Ti-H centre, single-grain OSL, single-grain TT-OSL and pIR-IRSL) and two different preserved minerals (quartz and potassium feldspar extracts) have been included as likelihoods in the Porto Maior Bayesian model (**Table 4**). The six ESR likelihoods included in the model have been derived using the most rapidly bleached Ti centre signal measured for each dating sample (Ti-Li in the case of samples MIN1402, MIN1403, V1204, V1205, V1206 and Ti-H for MIN1401; see Méndez-Quintas et al., 2018 for full details). The more slowly (and incompletely) bleached Al centre signals are considered to yield *terminus post quem* ages according to ESR dating MC approach employed by Méndez-Quintas et al. (2018), and have therefore not been included as likelihoods in the Bayesian model to ensure quality control. For sample MIN1401, Méndez-Quintas et al. (2018) consider the Ti-H age to be more appropriate than the replicate Ti-Li or Al centre ages but caution that the reliability of the Ti-H likelihood may be compromised by the proximity of the natural D_e value to the practical lower dating limit of the ESR technique and the poor goodness-of-fit of the extrapolated multiple aliquot additive dose response curve. These complications have been accounted for in our model by assigning sample MIN1401 as a maximum age likelihood for the lowermost (bottom) *boundary* of the PM-5 *sequence* using the OxCal *after* command.

The replicate pIR-IRSL and TT-OSL dating results for samples PM16-3, PM16-5 and PM16-6, and the replicate pIR-IRSL, OSL and TT-OSL dating results for sample PM16-4 are considered equally reliable on methodological grounds (see section 4.1 and discussions in Méndez-Quintas et al., 2018), and have therefore been included in the Bayesian model as combined luminescence age estimates for each sample. Since the replicate age determinations for these samples do not have fully independent dose rate uncertainties, they were combined after excluding all shared systematic uncertainty terms (including beta source calibration uncertainty, long-term water content uncertainty, cosmic-ray and internal dose rate errors, and uncertainties on dose rate conversion factors). This was achieved in OxCal by inputting the TT-OSL D_e estimate, pIR-IRSL D_e estimate, OSL D_e estimate and shared dose rate term as individual parameters with associated random uncertainty terms. The pIR-IRSL+TT-OSL and pIR-IRSL+OSL+TT-OSL ages of individual samples were then calculated as dependent parameters, and a weighted average was derived using the OxCal *combine* function. The additional component of shared systematic uncertainty was subsequently added in quadrature to the combined age for samples PM16-3, PM16-4, PM16-5 and PM16-6 for the purpose of Bayesian modelling.

The luminescence and ESR likelihoods were input into the model as calendar ages before AD2014 (the average year of sample collection), together with their associated 1σ uncertainty ranges, using the *date* command. Individual likelihoods have been represented in the main Bayesian model (denoted hereafter as “Model 1”) using their combined systematic and random uncertainty terms, following the approach adopted in similar studies of multi-method chronological datasets (e.g., Falguères et al., 2013; Higham et al., 2014; Kennett et al., 2015; Hamm et al., 2016; Demuro et al., 2019). While this approach may be considered sub-optimal for certain Bayesian age modelling applications (e.g., Rhodes et al., 2003), alternative modelling of shared systematic uncertainties in Bayesian frameworks such as OxCal can be limited by practical considerations (e.g.,

subsequent recombination of systematic uncertainty components with modelled age distributions may be complicated by non-Gaussian posterior distributions). For the type of prior and likelihood datasets considered in the present study, it has also been reported that Bayesian age modelling of *ex ante* combined systematic and random uncertainty terms may not significantly bias posterior age estimates (e.g., Higham et al., 2014; Groucutt et al., 2018; Demuro et al., 2019). However, in order to better assess the sensitivity of the Porto Maior Bayesian modelling results to prior incorporation of the shared and systematic dating uncertainties, we have implemented a second version of the model ('Model 2') in which only the random luminescence and ESR dating uncertainty ranges (i.e., those shown in parentheses in **Table 1**) have been included as likelihood input parameters. This modelling approach follows the procedure outlined in Clarkson et al. (2017), and involves subsequently recombining the average relative systematic uncertainty of the fifteen likelihood estimates with the modelled posterior *boundary* age distributions of each stratigraphic unit *ex post facto*. For the fifteen numerical dating estimates included in our model, the mean random and systematic uncertainty components account for $8.1 \pm 1.1\%$ and $6.3 \pm 0.3\%$ (mean $\pm$ standard error), respectively, of the total (random plus systematic) relative 1σ uncertainties before combining in quadrature. The total (random plus systematic) uncertainties associated with the modelled upper and lower age ranges of each unit (top and bottom *boundary* posterior distributions) have therefore been calculated by combining, in quadrature, the modelled (random) uncertainties and the 6.3% mean relative systematic uncertainty derived from the original likelihood dataset.

The two versions of the Porto Maior Bayesian age model were run using the general *outlier* function (Bronk Ramsey, 2009b), which is based on a Student t-test distribution with 5 degrees of freedom. Prior outlier probabilities of 5% were equally assigned to all dating samples to identify potentially significant statistical outliers. Likelihood estimates that yielded posterior outlier probabilities $>5\%$ were not excluded from the final model but were proportionally down-weighted in the iterative Monte Carlo runs, thereby producing an averaged chronological model (Bronk Ramsey, 2009b).

The OxCal *date* and *difference* functions (Bronk Ramsey, 2009a) were used to calculate the duration of each stratigraphic unit and potential hiatuses between sedimentary deposits from the posterior probabilities of their associated upper and lower *boundaries*. For Model 2, the *difference* and *date* queries have been run on the posterior *boundary* distributions after recombining, in quadrature, the mean relative systematic uncertainty of the thirteen likelihood estimates with the modelled (random) uncertainty ranges.

4. Results

4.1. Single-grain TT-OSL characteristics and ages

Relatively few of the quartz grains measured for samples PM16-4, PM16-6, PM16-5 and PM16-3 yield useable TT-OSL signals; ~87-92% of grain-hole positions ('pseudo' single-grain measurements) exhibit absent or very weak TT-OSL signals (failing quality assurance criteria (i) and (v)), and only ~1-2% of measured D_e values are considered suitable for burial dose estimation (**Table S2**). Low-to-moderate sensitivity change in the TT-OSL signal was observed through the SAR measurement procedure, with more than half of the grains accepted for final age estimation displaying sensitivity changes (i.e., an increase in T_x signal relative to T_n) of less than 50%.

The accepted grains from these samples generally exhibit fast-decaying TT-OSL signals, consistent with charge transfer into the most readily bleached (so-called ‘fast’) OSL dating trap, and the dose-response curves display high saturation dose limits (D_0 values >500 Gy; i.e., sufficient to accommodate the expected burial doses range of 700-900 Gy for these samples according to the associated pIR-IRSL ages) (**Figure 2a and b**).

The single-grain TT-OSL D_e distributions display a moderate degree of scatter (**Figure 2c–f**), although the relative uncertainties of the individual D_e values are generally high (20-60%) due to the dim TT-OSL signal intensities of accepted grains. None of the D_e datasets are considered to be significantly positively skewed according to the criterion outlined by Arnold and Roberts (2011). The overdispersion values obtained for samples PM16-4, PM16-6, PM16-5 and PM16-3 range between 32% and 47% (**Table 3**). These values are higher than those reported elsewhere for well-bleached and unmixed single-grain D_e datasets (e.g., the meta-analysis overdispersion averages of $20 \pm 1\%$ and $21 \pm 2\%$ reported for single-grain OSL and single-grain TT-OSL datasets by Arnold and Roberts, 2009 and Arnold et al., 2019, respectively). However, they are in agreement with these published average overdispersion ranges when compared at 2σ . The above-average overdispersion values for the Porto Maior samples may be related to intrinsic ‘between-grain’ scatter associated with our experimental procedures (e.g., grain-to-grain variations in luminescence responses due to the fixed SAR conditions; e.g., Demuro et al., 2013) or microdosimetry effects (e.g., Nathan et al., 2003; Mayya et al., 2006; Arnold et al., 2014) arising from variable proximities of quartz grains to either quartzite pebbles with relatively low radioactivities or potassium feldspars grains with relatively high radioactivities. Taking into consideration the stratigraphic context and D_e characteristics of the Porto Maior samples, we have derived representative single-grain TT-OSL burial dose estimates using the weighted mean D_e values for each sample, which has been calculated using the central age model (CAM) of Galbraith et al. (1999). The final quartz TT-OSL ages for the lower (sample PM16-3) and upper (sample PM16-5) sections of unit PM3 are 242 ± 32 ka and 225 ± 31 ka, respectively (**Table 3**), which are consistent with the existing pIR-IRSL ages for this unit at 1σ (**Table 1**). For level PM4 (sample PM16-6) the final single-grain TT-OSL ages are 172 ± 21 ka and this age is within 2σ of the equivalent pIR-IRSL age for this unit. For level PM 5 (sample PM16-4) the final TT-OSL age is 26 ± 3 ka, which is within 2σ the single-grain OSL age obtained for this sample, but slightly older than the corresponding pIR-IRSL age at 2σ (**Table 1**).

4.2. Bayesian modelling results

The results of the Porto Maior Bayesian model (Model 1) are summarised in **Table 4** and **Figure 3**. All modelled age ranges have been rounded to the nearest 50 years and are reported as the 68.2% and 95.4% highest probability density function (PDF) ranges, and the mean and 1σ uncertainty ranges of the modelled posterior distributions. The Bayesian modelled posterior age ranges of the upper and lower (top and bottom) *boundaries* were used to constrain the end and beginning periods of each depositional event at Porto Maior, as well as the likely age and duration of each stratigraphic unit and potential hiatuses between sedimentary deposits. These chronological relationships are summarised in **Figure 4** and **Table 5**, and have been calculated using the *difference* and *date* query functions in OxCal (see **Table 5** footnote for further details).

The statistical validity of Model 1 is supported by a model agreement index (A_{model}) of 128.8%, which is considerably higher than the preferable internal consistency threshold value of 60% (Bronk-Ramsey, 2009a), and indicates significant overlap between the measurement data and the modelled posterior distributions as a whole. The overall agreement index ($A_{overall}$) of Model 1 is 126%, which reflects relatively high individual agreement indices (A_i = correspondence between individual likelihood and posterior distributions) for each modelled dating sample, as well as a strong convergence integral (an assessment of whether the modelling algorithm has generated a truly representative posterior probability distribution) (Bronk-Ramsey, 2009a). The individual A_i values were also significantly higher than 60% for all modelled likelihoods, ranging from 78.4% for ESR MIN1403 to 128.6% for pIR+TT-OSL PM16-5 with a likelihood average A_i of 107.5%. The average convergence integral for all individual posterior distributions is 96.4% (C_i range = 85.6 – 100%), supporting the overall effectiveness of the Monte Carlo solutions. Sample ESR VI1204 was the only likelihood to exhibit a posterior outlier probability higher than the specified prior threshold of 5% (**Table 4**); though this occurrence is not considered extraordinary as one in twenty individual age estimates are expected to be significantly offset from the event of interest when adopting a prior outlier threshold of 5% (Bronk Ramsey, 2009b). This sample also exhibits an acceptable A_i value of 96.8% suggesting that it does not constitute a major statistical outlier. The overall suitability of the modelling indices and absence of major statistical outliers supports the appropriateness of the Bayesian modelling results for deriving meaningful combined age estimates for individual depositional units at Porto Maior.

The modelled posterior distributions of all the numerical dating samples (shown in dark blue on **Figure 3**) encompass narrower age ranges in comparison to their original likelihood age ranges (shown in light blue on **Figure 3**). Overall, the relative uncertainty ranges of the posterior likelihood distributions are reduced by an average of 32% when compared with the original dating sample uncertainties. The improvement in precision is most apparent for the four posterior likelihood estimates from unit PM3 (pIR+TT-OSL PM16-5, ESR VI1204, ESR VI1205, pIR+TT-OSL PM16-3), which exhibit 47–55% reductions when compared with the original dating sample uncertainties. For the fifteen dating samples included in our model, the random and systematic uncertainty components account for $8.1 \pm 1.2\%$ and $2.6 \pm 0.3\%$ (mean $\pm$ standard error), respectively, of the final (random plus systematic) relative 1σ uncertainties (mean = $10.6 \pm 0.9\%$) after combining in quadrature. The modelled posterior age distributions of these samples exhibit optimised total relative uncertainties of 4.2–20.0% (mean = $6.6 \pm 0.5\%$) (**Table 4**), which remain significantly higher than the relative systematic uncertainty contributions to the original likelihood ranges (1.1–4.7%; mean = $2.6 \pm 0.3\%$). As the overall improvements in chronological precision for Model 1 do not exceed the underlying systematic uncertainty contributions to the likelihood estimates, it seems unlikely that our modelling approach will have significantly biased the posterior age estimates. This interpretation is further supported by the results of Model 2, as detailed below.

The results of Porto Maior Bayesian Model 2, which was run after excluding all shared systematic uncertainty components from each luminescence and ESR likelihood input parameter, are summarised in **Table S3-S4** and **Figures S1-S2**. The Model 2 posterior *boundary* distributions in **Table S3** are expressed in two formats: the

first (shown outside parentheses) represents the raw OxCal modelling output and is based on propagation of random dating uncertainties only, while the second (shown in parentheses) includes propagation of the modelled (random) dating uncertainties and the mean systematic uncertainty (6.3%) of the fifteen numerical dating estimates in quadrature. The latter modelled age estimate is therefore the most appropriate to use for direct comparisons with independent chronologies and with the results of Model 1. Model 2 exhibits slightly lower A_{model} and $A_{overall}$ indices (101.1 and 102.7, respectively) compared to Model 1, though both exceed the preferable acceptance threshold value of 60% (Bronk-Ramsey, 2009) and are indicative of adequate modelling agreement. As with Model 1, none of the posterior likelihood estimates are identified as major statistical outliers and sample ESR VI1204 is again the only likelihood exhibiting a posterior outlier probability higher than the specified prior threshold of 5% (**Table S3**).

After recombining the shared systematic uncertainties with the posterior age distributions of Model 2, the resultant *boundary* age ranges of each stratigraphic unit reveal relatively minor differences between the two modelling scenarios (**Table 6**). The Model 2 mean posterior age ranges for the upper and lower boundaries (top and bottom) of each stratigraphic unit differ from the corresponding estimates of Model 1 by -800 years on average (a mean difference of -0.3% between the modelled *boundary* ages of Model 2 and 1). The total 1σ relative uncertainties on the Model 2 mean *boundary* ages were on average 1% higher than their Model 1 counterparts. Application of a paired-age homogeneity test (adapted from Galbraith, 2003; see Appendix A8 of Galbraith and Roberts, 2012) reveals that all of the corresponding mean boundary ages calculated using Model 1 and Model 2 are in agreement with a common value at their 2σ uncertainty ranges (**Table 6**; see footnote for further methodological details). To further examine the consistency of the modelling results as a combined dataset, we have applied a one-sample two-tailed Student's t -test following the approach detailed in Arnold et al. (2015). Standardised normal values ($\tilde{x}$) were first calculated as the difference between the Model 1 and Model 2 mean age of each boundary divided by the square root of the summed squares of the corresponding 1σ age uncertainties. The observed value of the test statistic (t_{obs}) was then calculated from the standardised normal values of each dataset according to the equation:

$$t_{obs} = \frac{(\tilde{x} - \mu_0)}{\left(\frac{s}{\sqrt{n}}\right)}$$

where $\tilde{x}$ is the sample mean standardised normal value, μ_0 is the reference value (in this case 0), and s is the standard deviation of the standardised normal values. The observed t value (t_{obs}) was compared with critical test statistic value (t_{crit}) at the 0.05 significance level with $n-1$ degree of freedom. The results of this statistical test reveal that the paired Model 1 – Model 2 *boundary* age estimates do not differ from each other at the 95.4% confidence level ($t_{obs} = 0.26 < t_{crit(\alpha:0.05)} = \pm 2.36$) (i.e., the null hypothesis is accepted).

In assessing the relative merits of Model 2 and Model 1 for deriving the final chronological sequence of Porto Maior it is worth noting that several of the modelled *boundaries* exhibit non-Gaussian posterior age distributions. This is particularly true for the upper and lower boundary of PM5, the lower boundary of PM4

and the lower boundary of PM1-2 (**Figures 3, S1**). For such non-Gaussian posterior age distributions, the recombination of a simplified (effectively Gaussian) mean relative systematic uncertainty derived from the original likelihood dataset may not adequately represent or capture the true underlying complexity of the total modelled uncertainty ranges. Model 2 is also limited by the fact that it requires recombination of a single, generalised estimate of the likelihood relative systematic uncertainty (6.3%) with the modelled (random) *boundary* age uncertainties *ex post facto*. This approach does not directly capture the inherent inter-sample variability in relative systematic uncertainties for the Porto Maior sequence, which vary between 4.9% and 7.7% for individual likelihood estimates (before combining in quadrature). Both of these practical complications are avoided in Model 1, as the final non-Gaussian posterior *boundary* distributions have been calculated by considering sample-specific systematic uncertainties and prior incorporation of all relevant empirical uncertainty components upfront. As such, we favour Model 1 for reconstructing the final chronostratigraphic sequence of Porto Maior. While acknowledging the assumptions and simplified likelihood representations implicit in the Bayesian approach of Model 1, it is worth emphasising that the resultant posterior *boundary* distributions are statistically indistinguishable from those obtained using the Model 2 configuration. The modelling results are therefore relatively insensitive to the treatment of shared likelihood uncertainties in this instance, and can be considered as a useful probabilistic means of evaluating age-depth relationships at the site.

5. Discussion

5.1. Comparison between TT-OSL and ESR chronologies for Porto Maior

This study (together with Méndez-Quintas et al., 2018) represents one of the few available comparisons of ages obtained using two ESR centres in combination with TT-OSL. The new dating results presented here therefore provide a useful means of assessing the empirical relationship between the ESR and TT-OSL signals for optically bleached quartz. ESR sample MIN1402, which was collected from the upper part of the Porto Maior sequence (level PM4), produced similar ages (within 1σ errors) for the Al (181 ± 25 ka) and Ti-Li (option D; 206 ± 20 ka) centres. The single-grain TT-OSL age obtained for the correlative luminescence sample in level PM4 was 172 ± 21 ka, which is within 1σ of both the Al and Ti-Li ages. In the lower section (levels PM3 and PM2), the ESR ages of samples VI1204, VI1205 and MIN1403 obtained using the Al centre were older by a factor of 1.6–1.8 (~ 424 – 452 ka) when compared to the ages obtained with the Ti-Li centre (~ 264 – 279 ka). The two TT-OSL ages of 225 ± 31 ka and 242 ± 32 ka presented here for the luminescence samples collected from level PM3 (PM16-3 and PM16-5, respectively), as well as the pIR-IRSL ages presented in Méndez-Quintas et al. (2018) for this level (~ 259 – 268 ka), are in closer agreement with the ESR ages obtained with the Ti-Li centre than those obtained with the Al centre (**Table 1**). These results indicate a potentially weaker link between the ESR Al centre and the trap giving rise to the TT-OSL signal in quartz, when compared to the Ti-Li centre.

One aspect worth considering further when evaluating the comparative quartz dating results for Porto Maior is that the TT-OSL measurements have been made at the single-grain scale of analysis while the ESR measurements have been made on bulk aliquots containing between 50 and 250 mg of quartz grains, and

therefore reflect composite signals derived from >10,000 grains (see Duval et al., 2017). The ESR experimental set-up therefore precludes direct assessment of grains with potentially problematic ESR signals that could have affected the resulting age estimates. By definition, bulk ESR D_e measurements are only likely to be reliable if the combined multi-grain signal is dominated by individual quartz grains with similar natural bleaching histories and radiation sensitivity behaviours. However, the proportion of grains that actually contribute to the main radiation-induced ESR signals measured from multigrain aliquots is presently unknown, and the information available regarding grain-to-grain ESR signal variability is scarce. Single-grain quartz ESR measurements made using Q-band spectroscopy (Beerten and Stesman, 2006) have showed that individual grains in a given sample can display significant variations in behaviours, suggesting that the ESR signal intensity measured on a given multigrain aliquot may potentially be composed of grains that contribute differently to the bulk signal. However, as with single-grain quartz OSL characteristics, it is feasible that different samples may exhibit significant variability in their grain-to-grain ESR signal properties and relative signal contributions. Unfortunately, limitations in the standard instrumentation currently available for ESR dating mean it is virtually impossible to evaluate possible grain averaging effects on our ESR D_e evaluations for the Porto Maior samples. It therefore remains difficult to ascertain a direct correspondence between TT-OSL and ESR signal properties at the individual grain scale in this study.

Although this study does not directly investigate the causes behind the closer age agreement between the TT-OSL and Ti-Li signals when compared to the Al signal, one possible explanation for this observation may relate to partial bleaching of the Al centre for the lower deposits. This possibility would be consistent with the recent study of Duval et al. (2017), which directly measured the bleaching rates of the ESR Al, Ti-Li and Ti-H centres, as well as TT-OSL signals, for a range of Spanish and French sedimentary quartz samples. The bleaching rate of the various ESR signals and the TT-OSL signal was measured after exposing aliquots of naturally-irradiated quartz to UV light in a UVACUBE 400 (Dr Honle) sunlight simulator for different time-periods. The results showed that the TT-OSL signal was optically reset at a rate similar to the Ti-H and Ti-Li centres combined, while the bleaching rate of the Al centre was substantially slower when compared to all other ESR signals and the TT-OSL signals. It is worth noting, however, that the results presented in Duval et al. (2017) relate to ESR and TT-OSL measurements made on different samples, and therefore they may mask inter-sample variations in bleaching properties. Several other ESR quartz studies have shown that the Al centre bleaches more slowly than any of the Ti centres when exposed to UV light and that an unbleachable component is prevalent in the Al centre signal (Walther and Zilles, 1994; Toyoda et al., 2000; Voinchet et al., 2003; Tissoux et al., 2007, 2008). Gao et al. (2009) showed that the ESR Ti-Li centre (option A *sensu* Duval and Guilarte, 2015) signal is readily bleached by UV exposure and that signal depletion occurs more rapidly as the UV intensity is increased. Although equivalent data is not available for the TT-OSL signal, sunlight bleaching experiments performed in different natural environments are consistent with similar dependencies on UV light intensity. In particular, comparison of the results presented by Demuro et al. (2015) and Jacobs et al. (2011) may indicate that the TT-OSL signal bleaches twice as fast at higher altitudes (950 m.a.s.l), where the UV radiance is considered to be more intense, than at sea level (i.e., 90% of the original TT-OSL signal was depleted by 6 weeks in the former study versus 12 weeks in the latter study). Again, it would be worth replicating these UV dependency tests

using the same samples and the same experimental set-up to eliminate any sample-specific or empirical biases. Nevertheless, the comparative ESR and luminescence datasets obtained for the two Porto Maior samples are outwardly consistent with existing research on the relative bleaching characteristics of the Al centre, Ti-Li centre, Ti-H centre and the TT-OSL signals of quartz, which also suggest a closer relationship between the trap giving rise to the TT-OSL signal and the ESR Ti centres, rather than the Al centre.

5.2. Comparison of published TT-OSL and ESR chronologies for different sites

To better evaluate the relationship between ESR and single-grain TT-OSL dating signals across a broader range of sites and sedimentary contexts, we have synthesised the comparative ages published so far as part of combined ESR–TT-OSL dating studies (**Table 7, Figure 5**). In total, ESR and single-grain TT-OSL dating have been applied in conjunction at eight archaeological and palaeoenvironmental sites, with most of these comparative studies focusing on sedimentary sequences from the Iberian Peninsula. The MC ESR dating approach has been employed at five of these study sites, with the remaining sites being ESR dated using Al centre signals alone (**Table 7**). Arnold et al. (2013) reported single-grain and multigrain TT-OSL ages of ~352 and ~332 ka, respectively, for the formation of the +19–30 m terrace of the Pico River (Duero basin, Spain). These ages were in agreement with ESR Al centre ages of ~370 and ~400 ka derived from a correlative terrace in the neighbouring Arlanzon River valley (terrace Tazn8; Moreno et al., 2012). There has been less success, however, in obtaining reproducible ESR Al centre and single-grain TT-OSL ages at the nearby karstic infill deposits of Atapuerca, which are located adjacent to the Arlanzon River (~10 km away). In the combined single-grain TT-OSL and ESR dating study of the Middle Pleistocene hominin site of Sima de los Huesos, two of the three dating samples collected from stratigraphic layer LU-7 produced ESR Al centre ages that were systematically older than corresponding single-grain TT-OSL ages by a factor of two or three (i.e., ~1.5 Ma and ~945 ka compared to a weighted mean TT-OSL age of 416 ± 19 ka). These two Al centre ages were also significantly older than U-series ages obtained on calcite rafts (436 ± 30 ka; $n=4$) from the underlying stratigraphic layer (LU-6) (Arsuaga et al., 2014). This systematic age overestimation relative to several independent dating methods (U-series, single-grain TT-OSL, pIR-IR₂₂₅) may be partially explained by the deep karst setting of the site and the potentially reduced optical bleaching possibilities of the Al centre signal in this depositional setting when compared to open-air sites (i.e., fluvial deposits). Interestingly, no evidence of partial bleaching has been observed in the single-grain TT-OSL D_e datasets from Sima de los Huesos (Arnold et al., 2014; Demuro et al., 2019), further supporting our observations at Porto Maior that there seems to be a weak link between the ESR Al centre and the trap giving rise to the single-grain TT-OSL signal. Elsewhere in the Atapuerca complex, Moreno et al. (2015) obtained three ESR Al centre ages of 591 ± 71 ka to 947 ± 90 ka for Gran Dolina level ATD6-3, which has been independently dated to >780 ka using palaeomagnetism (Parés et al., 2013), 780–949 ka using combined US-ESR dating on a *Homo antecessor* tooth (Duval et al., 2018), and ~831–856 ka using single-grain TT-OSL dating (Arnold et al., 2015; Arnold and Demuro, 2015). The weighted mean ESR and TT-OSL ages for Gran Dolina level TD6-3 are in broad agreement at 2σ , though the three Al centre ages exhibit a significant amount of scatter that is likely related to the heterogeneous dosimetric environment and the lack of *in situ* dosimetry measurements for two of the samples.

The ESR MC approach has been applied in conjunction with single-grain TT-OSL dating at a series of Early Pleistocene (~1–1.4 Ma) fluvial deposits from Morocco, as well as Middle Pleistocene fluvial deposits from France and Spain (**Table 7**). For the Early Pleistocene fluvial sequences of the Moulouya River basin, Morocco, both the Al and Ti-Li centres produced ESR ages that are in agreement with the paired TT-OSL ages at 2σ ; although the Ti-Li centre (option D) produced slightly younger ages that are in closer agreement with those obtained with single-grain TT-OSL (Bartz et al., 2018; 2019). Similar trends are apparent when comparing the combined Al centre, Ti centre and single-grain TT-OSL ages obtained for the Lower Palaeolithic fluvial sequences of Lunery-la Terre-des-Sablons and Brinay-la Noira in the Middle Loire basin, France (Duval et al., this volume). Finally, fluvial deposits corresponding to the formation of the +20 m terrace in the Deba River system of Cantabria (northern Spain) have been dated to 216 ± 15 ka using single-grain TT-OSL (Arriolabengoa et al., 2018). Comparison of this TT-OSL age with the MC ESR results presented for the same terrace (del Val et al., 2019), reveals close agreement with the both Ti-Li centre option D and Ti-H centre ages of 259 ± 20 ka and 192 ± 21 ka. In contrast, the corresponding Al centre age of this terrace is 421 ± 27 ka (del Val et al., 2019), which is older than the TT-OSL age by a factor of two.

Collectively, the published datasets synthesised in **Figure 5** and **Table 7** indicate that the comparative Ti-Li centre (option D) and single-grain TT-OSL ages obtained so far on optically bleached quartz are in agreement at 2σ . Though based on a limited sample dataset ($n = 9$), the combined Ti-Li centre – single-grain TT-OSL dataset is well-represented by a weighted linear regression function ($R^2 = 0.942$) that is consistent with the 1:1 line at 2σ (slope = 1.15 ± 0.08 ; intercept = 101 ± 20 ka) (**Figure 5a**). Application of a paired-age homogeneity test (adapted from Galbraith, 2003; see Appendix A8 of Galbraith and Roberts, 2012) reveals that 89% of the samples have Ti centre and single-grain TT-OSL ages in agreement with a common value at their 2σ uncertainty ranges; while 33% of these samples yielded paired ages that are consistent with a common value at their 1σ uncertainty ranges. To assess the general consistency of the Ti-Li centre and single-grain TT-OSL age estimates as a single dataset, we applied the one-sample two-tailed Student's t -test outlined in Section 4.2, following the approach detailed in Arnold et al. (2015). The results of this statistical test reveal that the paired Ti-Li centre – single-grain TT-OSL ages estimates do not differ from each other at the 95.4% confidence level ($t_{obs} = 1.82 < t_{crit}(\alpha:0.05) = \pm 2.36$) (i.e., the null hypothesis is accepted). A similar comparison between the combined Al centre and single-grain TT-OSL dataset reveals dating agreement for some of the study sites, but also a tendency for the Al centre ESR ages to be significantly older than the paired TT-OSL ages, as well as associated Ti-Li centre ages or other independent age control, in some depositional contexts (**Figure 5b**; **Table 7**). These mixed comparative dating results are borne out by the regression analyses and paired statistical tests. The Al centre and single-grain TT-OSL ages display more noticeable scatter and are weakly correlated ($R^2 = 0.556$) when fitted with a weighted linear function (**Figure 5b**). According to the paired homogeneity test, only 41% of the Al centre ages are in agreement with their single-grain TT-OSL counterparts at $\pm 2\sigma$. When analysed as a combined dataset, the Al centre and single-grain TT-OSL ages are considered to be statistically different from each other at the 95.4% confidence level ($t_{obs} = 2.88 > t_{crit}(\alpha:0.05) = \pm 2.16$).

Additional studies are clearly needed to increase the number and temporal coverage of these comparative ESR – TT-OSL datasets. However, our initial synthesis appears to highlight a potentially closer relationship between the ages of the TT-OSL and the ESR Ti-Li centre signals, rather than between the TT-OSL and Al centre signals. These datasets also reinforce the usefulness of employing the MC dating approach and emphasise the value of undertaking comparisons with semi-independent quartz dating signals, particularly single-grain TT-OSL dating, in order to maximise the reliability of ESR quartz chronologies.

5.3. Refining the chronology framework of Porto Maior using Bayesian modelling

The Bayesian modelling results obtained in this study have enabled us to refine the scope of the existing luminescence-ESR dating framework for Porto Maior by deriving more statistically meaningful combined ages for individual depositional units at the site, and, most importantly, by permitting a revision of the existing age estimate for the *in situ* archaeological layer (PM4) presented by Méndez-Quintas et al. (2018). Additionally, our Bayesian modelling results at Porto Maior have provided insights into the relative merits of employing different schemes for dealing with shared dating uncertainties in OSL and ESR chronological modelling applications, which may be of practical use for similar quartz dating studies performed at other archaeological sites.

While our Bayesian modelling results show that the prior and posterior modelled age ranges of individual dating likelihood estimates are generally not significantly different from each other at the 95.4% C.I. (**Figure 5; Table 4**), the modelled *boundary* age ranges for the various depositional units nevertheless provide a stronger statistical foundation for improved chronostratigraphic correlations of geographically disparate archaeological deposits across different parts of the Miño River basin. With considerable efforts currently ongoing to systematically date similar Acheulean deposits elsewhere in the Miño basin using ESR and luminescence techniques, (e.g. Méndez-Quintas et al., submitted), such improved chronostratigraphic correlations will be critical for future reconstructions of human occupation dynamics across the region during the Middle Pleistocene.

The results of the *difference* and *date* query analysis on the Model 1 posterior boundary probabilities reveals that the lowermost archaeological level (Unit PM3) was deposited between 252.0–276.9 ka and 239.3–263.5 ka (68.2% boundary probability ranges) with a mean depositional age of 258.3 ± 12.1 ka (**Figure 4; Table 5**). The uppermost archaeological level, which contains the *in situ* LCT accumulations, was deposited between 218.9–245.9 ka and 179.3–223 ka, with a mean depositional age of 210.7 ± 24.7 ka. The basal sterile fluvial deposits (Unit 1-2) have lower and upper boundary age ranges of 283.8–424.0 ka and 262.9–296.5 ka, respectively, and a mean depositional age of 312.6 ± 32.6 ka. While the uppermost unit (PM5), which is devoid of *in situ* artefacts, was deposited between 15.9–30.0 ka and 7.8–17.4 ka, and has a mean depositional age of 18.8 ± 7.7 ka. Application of the *difference* query to the posterior probabilities of the PM1-2 and PM3 boundaries, and the PM3 and PM4 boundaries, confirms the absence of any statistically significant temporal gap between these two groups of successive units at the 95.4% probability ranges (**Table 5**). This observation suggests that the preserved contacts between these units spanned relatively short timeframes that cannot be fully resolved beyond the modelled likelihood uncertainty ranges. The only statistically significant temporal hiatus identified in the

depositional sequence took place between the accumulation of *in situ* archaeological unit PM4 and the overlying sterile layer (PM5). The *difference* query indicates a mean temporal gap of 161.7 ± 36.4 ka between these two units, which confirms a prolonged period of non-deposition and / or a statistically significant amount of eroded material in this part of the profile between MIS 6 to MIS 3.

The ages for the lower sedimentary sequence at Porto Maior, with associated LFB Acheulean industry in levels PM3 and PM4, confirm the existence of this technocomplex during the Late Middle Pleistocene in the Iberian Peninsula, which is similar to that of other Iberian Acheulean sites dated to MIS 9–6 (Santonja et al., 2016), including Pinedo (López-Recio et al., 2015), Torralba (Santonja et al., 2016), Valdocarros (Rubio-Jara et al., 2016; Moreno et al., in press). The Bayesian modelled age for the *in situ* Porto Maior LCT accumulation (211 ± 25 ka at 1σ) overlaps with, but is slightly younger than, the age range of 293–205 ka originally inferred by Méndez Quintas et al. (2018), which was based on the 2σ confidence interval of the weighted mean Ti centre and pIR-IRSL ages for units PM3–PM4. The Bayesian modelling chronostratigraphic framework therefore suggests the *in situ* lithic-bearing deposits at the base of Unit PM4 most likely accumulated during MIS 7 rather than during MIS 7-8 as originally proposed by Méndez-Quintas et al. (2018). These results also confirm the presence of LFB Acheulean at a time when non-LFB Acheulean and early Middle Palaeolithic assemblages are found elsewhere on the Iberian Peninsula (Santonja et al., 2014).

6. Conclusion

This study has added several new facets to our chronological understanding of the Porto Maior archaeological site: (i) It has enhanced the scope of the original dating framework presented by Méndez-Quintas et al. (2018) via the establishment of new single-grain TT-OSL ages for level PM3 and (ii) it has derived more statistically meaningful combined age ranges for the various depositional units and archaeological finds at the site using an integrated Bayesian modelling approach. In doing so it has confirmed the suitability of the original ESR MC dating interpretations of Méndez-Quintas et al. (2018), and enabled refinement of the depositional age for the *in situ* LCT accumulation at Porto Maior (210.7 ± 24.7 ka at 1σ).

The results of this study suggest a potentially close relationship between TT-OSL and Ti-Li paramagnetic centres. This interpretation appears to be supported by our synthesis of comparative ESR–single-grain TT-OSL dating studies performed at other sites. From an ESR dating perspective, our new results and analyses of published datasets imply that the MC dating approach is likely to be advantageous in many dating contexts and that there is good potential for obtaining reliable Middle to Early Pleistocene chronologies using appropriate ESR quartz dating methodologies; especially where they are applied in combination with other extended-range quartz dating techniques such as single-grain TT-OSL.

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Figure Captions

Figure 1. (A) Location of Porto Maior (black star), Galicia, Spain. (B) Stratigraphic section showing the previously published pIR-IRSL, single-grain OSL and ESR ages for Porto Maior, together with the new TT-OSL ages obtained in the present study (modified from Méndez-Quintas et al., 2018).

Figure 2. (A, B) Example of a sensitivity-corrected TT-OSL dose-response and decay curve for an individual grain from sample PM16-3. White square denotes the sensitivity-corrected natural OSL signal; filled circles denote the sensitivity-corrected regenerated OSL signals; white circle denotes the repeated regenerative dose point used to calculate the recycling ratio. The D_0 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. (C–F) The single-grain TT-OSL D_e distributions obtained for samples PM16-4, PM16-6, PM16-5 and PM16-3. The grey bars on the radial plots are centred on the central age model (CAM) D_e value used to derive the final burial dose estimate for each sample. Also shown are the overdispersion (OD) values associated with each D_e dataset.

Figure 3. Bayesian modelling results for the Porto Maior stratigraphic sequence (Model 1; see main text). The prior age distributions for the dating samples (likelihoods) are shown as light blue probability density functions (PDFs). The modelled posterior distributions for the dating samples and stratigraphic unit boundaries are shown as dark blue and grey PDFs, respectively. Unmodelled and modelled ages are shown on a calendar year timescale and both are expressed in years before AD2014 (average sample collection date). The white circles and associated error bars represent the mean ages and 1σ uncertainty ranges of the PDFs. The 68.2% and 95.4% ranges of the highest posterior probabilities are indicated by the horizontal bars underneath the PDFs.

Figure 4. Bayesian-modelled durations of the stratigraphic units at Porto Maior (Model 1; see main text). The PDFs have been calculated from the modelled posterior probabilities of the upper and lower boundaries of each stratigraphic unit (shown as grey PDFs in Figure 5) using the date query function in OxCal v4.3.2.

Figure 5. ESR and TT-OSL age comparisons based on published dating studies conducted on a range of sites from Spain, France and Morocco (based on data shown in **Table 7**). The uncertainties for each age estimate represent the 1σ errors. The type of ESR centre used to derive comparative ages for each TT-OSL sample correspond to the Ti-Li centre (option D) in plot (A) and the Al centre in plot (B). Each dataset has been fitted with a linear least-squares function (dashed line) weighted according to the inverse squared relative errors of both coordinates, following the approach of Reed (1989). The confidence intervals about the linear regression functions (long-dashed lines) have been calculated using the Student's t-statistic for a two-tailed test with $(n-2)$ degrees of freedom and are presented at the 95.4% confidence level ($\alpha = 0.05$). The solid line represents the 1:1 line for each dataset.

Level	Sample	ESR age (ka)			Luminescence age (ka)			
		Al centre	Ti-Li centre (op. D)	Ti-H centre	Sample	Single-grain OSL	pIR-IRSL ₂₂₅	Single-grain TT-OSL
PM5	MIN1401	138.5 ± 25.2(23.3)	124.2 ± 12.2(8.5)	47.5 ± 10.0(9.5)	PM16-4	17.1 ± 1.3(1.0)	16.6 ± 0.9(0.5)	25.8 ± 3.4(3.2) (This study)
PM4	MIN1402	181.5 ± 24.0(19.6)	205.5 ± 20.0(12.3)		PM16-6		230.6 ± 14.8(7.7)	172.1 ± 20.5(17.6) (This study)
PM4	VI1206	349.7 ± 45.7(37.2)	237.8 ± 21.1(10.5)					
PM3	VI1204	441.3 ± 74.8(66.8)	278.8 ± 26.1(15.0)		PM16-5		268.3 ± 24.0(19.8)	225.3 ± 30.6(27.9) (This study)
PM3	VI1205	452.4 ± 66.4(56.8)	265.5 ± 22.9(10.4)		PM16-3		259.1 ± 28.8(24.4)	241.5 ± 32.2(28.0) (This study)
PM1	MIN1403	424.4 ± 85.1(78.7)	264.3 ± 26.8(18.0)					

Table 1. ESR (quartz) and luminescence chronologies (quartz and potassium feldspars) obtained previously for Porto Maior, Spain (Méndez-Quintas et al., 2018). The new single-grain TT-OSL quartz ages obtained in this study are also provided in the last column. The dating uncertainties shown outside parentheses represent the total (random plus systematic) 1σ range for each sample, whereas those shown in parentheses represent the random 1σ dating uncertainties only.

Step	Single-grain TT-OSL SAR protocol	Signal
1 ^a	Give dose	
2	Preheat to 260°C for 10 s	
3	Stimulate with green laser at 125°C for 3 s (90% power)	
4	Preheat to 260°C for 10 s	
5	Stimulate with green laser at 125°C for 3 s (90% power)	TT-OSL L _n or L _x
6	Stimulate with blue LEDs at 280°C for 400 s	
7	Give test dose (250 Gy)	
8	Preheat to 260°C for 10 s	
9	Stimulate with green laser at 125°C for 3 s (90% power)	
10	Preheat to 260°C for 10 s	
11	Stimulate with green laser at 125°C for 3 s (90% power)	TT-OSL T _n or T _x
12	Stimulate with blue LEDs at 290°C for 400 s	
13	Return to 1	

^a Step omitted when measuring the natural signal (L_n).

Table 2. SAR protocol used in this study to obtain single-grain quartz TT-OSL ages for the Porto Maior samples. L_x = regenerative dose signal response; L_n = natural dose signal response; T_x = test dose signal response for a laboratory dose cycle T_n = test dose signal response for the natural dose cycle.

Sample	Accepted/ measured ^a	Overdis- persion (%) ^b	CAM D _e (Gy)	Water content (%) ^c	Beta dose rate (Gy / ka) ^d	Gamma dose rate (Gy / ka) ^e	Cosmic dose rate (Gy / ka) ^f	Total dose rate (Gy / ka) ^g	Age (ka) ^h
PM16-4	27 / 2600	47 ± 12	95 ± 12	15.0	1.99 ± 0.05	1.49 ± 0.05	0.17 ± 0.02	3.68 ± 0.15	26 ± 3
PM16-6	22 / 1300	32 ± 10	556 ± 56	24.9	1.75 ± 0.09	1.30 ± 0.05	0.15 ± 0.02	3.23 ± 0.20	172 ± 21
PM16-5	14 / 1000	32 ± 12	732 ± 89	19.8	1.70 ± 0.08	1.38 ± 0.05	0.14 ± 0.02	3.25 ± 0.18	225 ± 31
PM16-3	23 / 1300	38 ± 10	804 ± 92	28.5	1.80 ± 0.09	1.39 ± 0.05	0.11 ± 0.01	3.33 ± 0.22	242 ± 32

^a Number of D_e measurements that passed the standard SAR rejection criteria / total number of D_e measured made per sample.

^b The relative spread in the D_e dataset beyond that associated with the measurement uncertainties for individual D_e values, calculated using the central age model (CAM) of Galbraith et al. (1999).

^c Water content used for calculating environmental dose rates, expressed as % of dry mass of sample and assigned a relative uncertainty of ± 20%. For samples PM16-3, PM16-5 and PM16-6 the long-term water contents are taken as being equivalent to 50% of saturated values, while for sample PM16-4 the water content was set to 15% (see justification in Méndez-Quintas et al., 2018). An associated uncertainty of 20% was added to water content values when calculating dose rates.

^d Beta dose rates were calculated using a Risø GM-25-5 low-level beta counter (Bøtter-Jensen and Mejdahl, 1988), after making allowance for beta dose attenuation due to grain-size effects and HF etching (Brennan, 2003).

^e Gamma dose rates were calculated from in situ measurements made at each sample position with a NaI:Tl detector using the ‘energy windows’ method detailed in Arnold et al. (2012b). Radionuclide concentrations were converted to dose rates using published conversion factors (Guérin et al., 2011)

^f Cosmic-ray dose rates were calculated according to Prescott and Hutton (1994) and assigned a relative uncertainty of ± 10%.

^g Includes an internal dose rate of 0.03 ± 0.01 Gy / ka after Bowler et al. (2003).

^h Mean ± total uncertainty (68% confidence interval), calculated as the quadratic sum of the random and systematic uncertainties. Total uncertainty includes a systematic component of ± 2% associated with laboratory beta-source calibration.

Table 3. TT-OSL dose rate data, single-grain equivalent doses and final ages for luminescence samples PM16-4, PM16-6, PM16-5 and PM16-3.

Boundary	Dating sample	Unmodelled age (years) ^a			Modelled age (years)			Agreement index (A _i) (%)	Posterior outlier probability (%)
		68.2% range	95.4% range	Mean ± 1σ	68.2% range	95.4% range	Mean ± 1σ		
Unit PM5 top	pIR+OSL+TT-OSL PM16-4 ^b	17750 – 15900	18600 – 15050	16850 ± 900	17400 – 7750	17600 – 1050	10700 ± 4950	100.2	5
					17750 – 15900	18650 – 15000	16850 ± 950		
Unit PM5 bottom	ESR MIN1401	57800 – 37250	67600 – 27400	47500 ± 10050	29950 – 15900	47900 – 15250	26850 ± 9900	101.1	0
					58650 – 39100	68600 – 29550	49000 ± 9700		
Unit PM4 top	pIR+TT-OSL PM16-6 ^b	225900 – 185150	245450 – 165600	205500 ± 19950	223400 – 179300	236550 – 111750	188550 ± 35150	118.0	3
					223500 – 195150	234500 – 176550	206900 ± 14600		
					228750 – 207050	238900 – 195450	217350 ± 10750		
					237200 – 213950	249000 – 202350	225600 ± 11500		
Unit PM4 bottom	ESR VII206	259300 – 216250	280000 – 195550	237800 ± 21100	237200 – 213950	249000 – 202350	225600 ± 11500	110.5	5
Unit PM3 top	pIR+TT-OSL PM16-5 ^b	275400 – 232400	296050 – 211700	253900 ± 21100	245900 – 218900	259750 – 206900	232750 ± 13500	128.6	2
					263450 – 239250	274600 – 226600	250950 ± 12150		
					265650 – 242650	276400 – 231900	254100 ± 11100		
					267600 – 246450	278550 – 235750	257150 ± 10750		
					270900 – 248500	281900 – 238000	259900 ± 10950		
Unit PM3 bottom	pIR+TT-OSL PM16-3 ^b	276100 – 226900	299700 – 203250	251500 ± 24100	276900 – 251950	291600 – 241050	265650 ± 12900	121.1	2
					274000 – 250150	286300 – 239300	262550 ± 11550		
					296500 – 262850	317800 – 249650	282350 ± 17400		
Unit PM1-2 top	ESR MIN1403	291650 – 236950	317950 – 210650	264300 ± 26800	311650 – 274350	332800 – 260150	295250 ± 18400	78.4	1
					424000 – 283800	424000 – 280700	342650 ± 43500		
Unit PM1-2 bottom									

^a The original dating results are all expressed in years before AD2014 (average sample collection year) for modelling purposes to ensure the likelihoods are directly comparable.

^b The combined (unmodelled) ages for luminescence samples PM16-3, PM16-4, PM16-5 and PM16-6 have each been calculated from their replicate pIR-IRSL and TT-OSL ages, or replicate pIR-IRSL, OSL and TT-OSL ages, using the OxCal *Combine* function. See Supplementary Information text for further details.

Table 4. Summary of Bayesian modelling results for Porto Maior (Model 1; see main text). The likelihood (unmodelled) and posterior (modelled) age ranges are presented for each of the numerical dating samples. Posterior (modelled) age ranges are also shown for the boundaries of each stratigraphic unit. Posterior ages are presented as the 68.2% and 95.4% highest probability density ranges. The mean and 1σ uncertainty ranges of the modelled posterior distributions are shown for comparison (assuming a normally distributed probability density function). The unmodelled and modelled age estimates have been rounded to the nearest 50 years.

Unit / boundary	Time variable	Modelled age (years) ^a			Modelled duration (years) ^{a, b}		
		68.2% range	95.4% range	Mean $\pm$ 1 σ	68.2% range	95.4% range	Mean $\pm$ 1 σ
PM5	age	23050 – 11350	36100 – 3900	18800 $\pm$ 7650			
PM5	duration				-50 – 20450	-50 – 39700	16150 $\pm$ 12050
PM5 – PM4 boundary	duration				149500 – 200250	82200 – 215400	161650 $\pm$ 36350
PM4	age	234300 – 200300	251050 – 159650	210650 $\pm$ 24650			
PM4	duration				-50 – 51250	-50 – 127450	44200 $\pm$ 39900
PM4 – PM3 boundary	duration				-50 – 23100	-50 – 43900	18250 $\pm$ 13300
PM3	age	269900 – 246150	282300 – 234200	258300 $\pm$ 12100			
PM3	duration				-50 – 17600	-50 – 41750	14700 $\pm$ 13050
PM3 – PM2 boundary	duration				-50 – 20550	-50 – 4600	16700 $\pm$ 14350
PM1-2	age	330050 – 272250	382750 – 259650	312550 $\pm$ 32600			
PM1-2	duration				-50 – 81300	-50 – 141500	60300 $\pm$ 45200

^a Modelled age ranges were calculated from the posterior probabilities of the upper and lower boundaries (top and bottom) of each stratigraphic unit (see Table 4) using the *date* query function in OxCal v4.3. Modelled durations were calculated using the *difference* query function, and provide the temporal range between the posterior probability density distributions of successive stratigraphic boundaries.

^b The OxCal *difference* function can be used to test whether or not the posterior probability distributions of successive stratigraphic boundaries are significantly different from each other at a given confidence interval. When the *difference* function is applied to adjacent boundaries of two different stratigraphic units (e.g. LU-6 – LU-5 boundary), it provides a statistical indication of the presence or absence of potential depositional hiatuses (given the available dating evidence). Calculated duration ranges that overlap with 0 at the 95.4% confidence interval suggest that the boundaries of successive units are not separated by a statistically significant temporal hiatus. Calculated duration ranges that do not overlap with 0 at the 95.4% confidence interval indicate potentially missing material and / or a temporal gap between the boundaries of successive units. When the *difference* function is applied to the upper and lower boundaries of the same stratigraphic unit (as opposed to the adjacent boundaries of successive units), a calculated duration of >0 years indicate a statistically significant temporal difference between the onset and termination of the depositional event. In contrast, calculated duration ranges that overlap with 0 indicate statistically indistinguishable onset and termination ages for the depositional event at a given confidence interval.

Table 5. Bayesian modelled posterior age ranges and depositional durations for the Porto Maior stratigraphic units (Model 1). Posterior ages/durations are presented as the 68.2% and 95.4% highest probability density ranges. The mean and 1 σ uncertainty ranges of the modelled posterior distributions are shown for comparison (assuming a normally distributed probability density function). The unmodelled and modelled age estimates have been rounded to the nearest 50 years.

Boundary	Model	Modelled age ^a				Difference in modelling results (Model 2 – Model 1)				Paired-value homogeneity test ^a			
		68.2% range (years)	95.4% range (years)	Mean $\pm 1\sigma$ (years)	Relative uncertainty ($\pm 1\sigma$) (%)	Δ mean age (years)	% change in mean age (%)	Δ mean age uncertainty ($\pm 1\sigma$) (years)	Δ relative uncertainty ($\pm 1\sigma$) (%)	G-value	G-value < $X^2_{crit}(\alpha:0.05)$	G-value < $X^2_{crit}(\alpha:0.32)$	P-value
Unit PM5 top	1	17400 – 7750	17600 – 1050	10700 $\pm$ 4950	46								
	2	17450 – 10400	17600 – 1850	12050 $\pm$ 4750	39	1350	13	-200	-7	0.04	Yes	Yes	0.84
Unit PM5 bottom	1	29950 – 15900	47900 – 15250	26850 $\pm$ 9900	37								
	2	25200 – 16000	44950 – 15400	24250 $\pm$ 9150	38	-2600	-10	-750	-1	0.04	Yes	Yes	0.85
Unit PM4 top	1	223400 – 179300	236550 – 111750	188550 $\pm$ 35150	19								
	2	226300 – 180200	234400 – 111300	189300 $\pm$ 36350	19	750	<1	1200	0	<0.01	Yes	Yes	0.99
Unit PM4 bottom	1	245900 – 218900	259750 – 206900	232750 $\pm$ 13500	6								
	2	255700 – 218800	307100 – 206750	238150 $\pm$ 18350	8	5400	2	4850	2	0.06	Yes	Yes	0.81
Unit PM3 top	1	263450 – 239250	274600 – 226600	250950 $\pm$ 12150	5								
	2	274500 – 238100	290800 – 218150	255200 $\pm$ 18250	7	4250	2	6100	2	0.04	Yes	Yes	0.85
Unit PM3 bottom	1	276900 – 251950	291600 – 241050	265650 $\pm$ 12900	5								
	2	284400 – 247400	304300 – 229600	266500 $\pm$ 18700	7	850	<1	5800	2	<0.01	Yes	Yes	0.97
Unit PM1-2 top	1	296500 – 262850	317800 – 249650	282350 $\pm$ 17400	6								
	2	296300 – 255400	320450 – 237300	277700 $\pm$ 20850	8	-4650	-2	3450	2	0.03	Yes	Yes	0.86
Unit PM1-2 bottom	1	424000 – 283800	424000 – 280700	342650 $\pm$ 43500	13								
	2	427050 – 269700	435450 – 259350	330700 $\pm$ 49400	15	-11950	-4	5900	2	0.03	Yes	Yes	0.86
Average						-800	0.3	3300	1				

^a The homogeneity test of Galbraith (2003) was used to formally test whether the posterior boundary ages of Model 1 and Model 2 were consistent with a shared value at their mean $\pm 1\sigma$ and $\pm 2\sigma$ ranges. For this purpose, G-values were calculated for each posterior boundary mean $\pm 1\sigma$ age range (column 5) as the square of the standardised difference between the two comparative age estimates obtained using Model 1 and Model 2 (Appendix A8 of Galbraith and Roberts, 2012). The observed G-values were compared with the critical value of a X^2 probability distribution at the 0.32 and 0.05 significance levels (α) with 1 degree of freedom. Note – significance level (%) = 100(1 – α). Individual G-values lower than the $X^2_{crit}(\alpha:0.32)$ and $X^2_{crit}(\alpha:0.05)$ scores indicate that the Model 1 and Model 2 posterior age estimates of a given boundary are in agreement with a common value at the 1σ (68% C.I.) and 2σ (95% C.I.) uncertainty ranges, respectively. The P-value is the probability that a random value drawn from a X^2 distribution with $n - 1$ degrees of freedom is larger than the observed G-value for each posterior paired boundary age. A small P-value (conventionally less than 0.05) would provide strong statistical evidence that the Model 1 and Model 2 posterior age estimates of a given boundary are not consistent with a common value (Galbraith and Roberts, 2012).

Table 6. Comparison of the Bayesian modelled posterior age ranges for the upper and lower boundaries (top and bottom) of each stratigraphic unit obtained using Model 1 and Model 2. Model 1 was run using the combined (random + systematic) empirical dating uncertainties of each luminescence and ESR sample as likelihood input parameters. Model 2 was run after excluding all shared systematic error terms from each luminescence and ESR likelihood input parameter. For the model 2 results shown here, the average shared systematic uncertainty of the 15 likelihood (unmodelled) ages has subsequently been added in quadrature

to the posterior (modelled) probabilities of the upper and lower boundaries of each unit to enable direct comparisons with the posterior (modelled) probability ranges of Model 1. The modelled age estimates have been rounded to the nearest 50 years.

Site	Level / fluvial terrace	Sample	ESR-Al centre age (ka)	ESR-Ti-Li centre age (ka)	Sample	Single-grain TT-OSL age (ka)	Reference
<i>Molouya River (Morocco)</i>	BOU	C-E3825	1160 ± 250	1100 ± 110	C-L3825	1150 ± 100	Bartz et al. (2018); Bartz et al. (2019)
	BOU	C-E3824	1530 ± 190	1260 ± 100	C-L3824	1090 ± 100	Bartz et al. (2018); Bartz et al. (2019)
<i>Lunery (France)</i>	Unit 3	LUN1101	866 ± 100	654 ± 64	CRF11-2	727 ± 51	Duval et al. (this volume)
<i>Noira (France)</i>	Unit 6	NOI1104	940 ± 119	739 ± 72	CRF11-4	644 ± 47	Duval et al. (this volume)
<i>Porto Maior (Spain)</i>	PM5	MIN1401	138 ± 25	124 ± 12	PM16-4	26 ± 3	Méndez-Quintas et al. (2018)
	PM4	MIN1402	181 ± 25	206 ± 20	PM16-6	172 ± 21	Méndez-Quintas et al. (2018)
	PM3	VI1204	441 ± 75	279 ± 26	PM16-5	225 ± 31	Méndez-Quintas et al. (2018); This study
	PM3	VI1205	451 ± 66	266 ± 23	PM16-3	242 ± 32	Méndez-Quintas et al. (2018); This study
<i>Deba River Terrace (Spain)</i>	+20 m / Layer K in pit Lezetxiki 2	CANT15130	421 ± 27	259 ± 20	LZ12-6	216 ± 15	del Val et al. (2019); Arriolabengoa et al. (2018)
<i>Atapuerca – Sima de los Huesos (Spain)</i>	LU7	SH12-1B	945 ± 100		SH12-1A	429 ± 32	Arsuaga et al. (2014); Arnold et al. (2014)
	LU7	SH12-2B	1502 ± 170		SH12-2A	419 ± 34	Arsuaga et al. (2014); Arnold et al. (2014)
	LU7	SH12-3B	443 ± 90		SH12-3A	396 ± 35	Arsuaga et al. (2014); Arnold et al. (2014)
<i>Atapuerca – Gran Dolina (Spain)</i>	TD6	TD08-15	591 ± 71		F13	856 ± 75	Moreno et al. (2015); Arnold et al. (2015)
	TD6	TD08-14	601 ± 88		F14	831 ± 90	Moreno et al. (2015); Arnold et al. (2015)
	TD6	TD08-13	947 ± 90				Moreno et al. (2015)
<i>Pico River / Arlanzón River terraces (Spain)</i>	Tazn 8 / Tp1	AZN08-03	400 ± 90		PT10-1	352 ± 23 332 ± 28 ^a	Moreno et al. (2012); Arnold et al. (2013)
	Tazn 8	AZN08-04	370 ± 70				Moreno et al. (2012)

^a This age was derived from measurements made on multi-grain aliquots.

Table 7. Comparative chronologies obtained using single-grain TT-OSL and ESR dating (based on the Al and Ti-Li centres) of optically-bleached quartz from a range of archaeological sites and fluvial deposits from Spain, France and Morocco.

Figure 1. (A) Location of Porto Maior (black star), Galicia, Spain. (B) Stratigraphic section showing the previously published pIR-IRSL, single-grain OSL and ESR ages for Porto Maior, together with the new TT-OSL ages obtained in the present study (modified from Méndez-Quintas et al., 2018).

Figure 2. (A, B) Example of a sensitivity-corrected TT-OSL dose-response and decay curve for an individual grain from sample PM16-3. White square denotes the sensitivity-corrected natural OSL signal; filled circles denote the sensitivity-corrected regenerated OSL signals; white circle denotes the repeated regenerative dose point used to calculate the recycling ratio. The D_0 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. (C–F) The single-grain TT-OSL D_e distributions obtained for samples PM16-4, PM16-6, PM16-5 and PM16-3. The grey bars on the radial plots are centred on the central age model (CAM) D_e value used to derive the final burial dose estimate for each sample. Also shown are the overdispersion (OD) values associated with each D_e dataset.

Figure 3. Bayesian modelling results for the Porto Maior stratigraphic sequence (Model 1; see main text). The prior age distributions for the dating samples (likelihoods) are shown as light blue probability density functions (PDFs). The modelled posterior distributions for the dating samples and stratigraphic unit boundaries are shown as dark blue and grey PDFs, respectively. Unmodelled and modelled ages are shown on a calendar year timescale and both are expressed in years before AD2014 (average sample

collection date). The white circles and associated error bars represent the mean ages and 1σ uncertainty ranges of the PDFs. The 68.2% and 95.4% ranges of the highest posterior probabilities are indicated by the horizontal bars underneath the PDFs.

Figure 4. Bayesian-modelled durations of the stratigraphic units at Porto Maior (Model 1; see main text). The PDFs have been calculated from the modelled posterior probabilities of the upper and lower boundaries of each stratigraphic unit (shown as grey PDFs in **Figure 5**) using the date query function in OxCal v4.3.2.

Figure 5. ESR and TT-OSL age comparisons based on published dating studies conducted on a range of sites from Spain, France and Morocco (based on data shown in **Table 7**). The uncertainties for each age estimate represent the 1σ errors. The type of ESR centre used to derive comparative ages for each TT-OSL sample correspond to the Ti-Li centre (option D) in plot (A) and the Al centre in plot (B). Each dataset has been fitted with a linear least-squares function (dashed line) weighted according to the inverse squared relative errors of both coordinates, following the approach of Reed (1989). The confidence intervals about the linear regression functions (long-dashed lines) have been calculated using the Student's t-statistic for a two-tailed test with $(n-2)$ degrees of freedom and are presented at the 95.4% confidence level ($\alpha = 0.05$). The solid line represents the 1:1 line for each dataset.

Supplementary Information - Demuro et al. Refining the chronology of Acheulean deposits at Porto Maior in the River Miño basin (Galicia, Spain) using a comparative luminescence and ESR dating approach.

Figure S1 – Bayesian modelling results for the Porto Maior stratigraphic sequence (Model 2; see main text). The prior age distributions for the dating samples (likelihoods) are shown as light blue probability density functions (PDFs). The modelled posterior distributions for the dating samples and stratigraphic unit boundaries are shown as dark blue and grey PDFs, respectively. Unmodelled and modelled ages are shown on a calendar year timescale and both are expressed in years before AD2014 (average sample collection date). The white circles and associated error bars represent the mean ages and 1 σ uncertainty ranges of the PDFs. The 68.2% and 95.4% ranges of the highest posterior probabilities are indicated by the horizontal bars underneath the PDFs.

Figure S2 – Bayesian-modelled durations of the stratigraphic units at Porto Maior (Model 2; see main text). The PDFs have been calculated from the modelled posterior probabilities of the upper and lower boundaries of each stratigraphic unit (shown as grey PDFs in **Figure S1**) using the date query function in OxCal v4.3.2.

Natural dose assessment (un-dosed grains)			Dose recovery test				
Sample	Accepted / measured	CAM D _e (Gy)	Given dose (Gy)	Accepted / measured	CAM D _e (Gy)	OD (%)	Measured / given dose ratio (after residual subtraction)
PM16-3	23 / 1300	804 ± 92	578 ± 4	18 / 500	1368 ± 117	19 ± 11	0.98 ± 0.26

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

Table S1. Single-grain TT-OSL dose recovery test results obtained for sample PM16-3 using the SAR protocol shown in **Table 2**.

Sample name	PM16-4		PM16-6		PM16-5		PM16-3	
Total number of grains measured	2600		1300		1000		1300	
	n	%	n	%	n	%	n	%
<u>Reason for rejecting grains from D_e analysis:</u>								
(i) T _n < 3σ background (non-luminescent grains)	1924	74.0	994	76.5	805	80.5	1009	77.6
(ii) Poor dose recycling ratio	228	8.8	147	11.3	62	6.2	76	5.8
(iii) Recuperation (0 Gy L _x /T _x > 5% of L _n /T _n)	25	1.0	0	0	0	0	5	0.4
(iv) Poor OSL IR depletion ratio	7	0.3	0	0	2	0.2	1	0.1
(v) Relative error on the net T _n signal > 30%	387	14.9	133	10.2	114	11.4	176	13.5
(vi) Saturated or non-intersecting grains (L _n /T _n ≈ or > dose-response curve saturation)	0	0	2	0.2	1	0.1	3	0.2
(vii) Anomalous dose-response / unable to perform Monte Carlo fit ^a	2	0.1	2	0.2	2	0.2	7	0.5
Accepted grains (used for D_e determination)	27	1.0	22	1.7	14	1.4	23	1.8

Includes grains with zero or negative changes in L_x/T_x with increasing dose, and grains displaying very scattered L_x/T_x values that could not be successfully fitted with the Monte Carlo procedure.

Table S2. Single-grain TT-OSL quality assurance statistics obtained for natural D_e measurements of samples PM16-4, PM16-6, PM16-5 and PM16-3.

Boundary	Dating sample	Unmodelled age (years) ^a			Modelled age (years) ^b			Agreement index (A _i) (%)	Posterior outlier probability (%)
		68.2% range	95.4% range	Mean ± 1σ	68.2% range	95.4% range	Mean ± 1σ		
Unit PM5 top	pIR+OSL+TT-OSL PM16-4 ^c	17250 – 16400	17650 – 16000	16850 ± 400	17350 – 10500	17500 – 1950	12050 ± 4700	104.8	0
					(17450 – 10400)	(17600 – 1850)	(12050 ± 4750)		
Unit PM5 bottom	ESR MIN1401	57200 – 37850	66500 – 28550	47500 ± 9500	25000 – 16200	44450 – 15900	24250 ± 9000	100.7	0
					(25200 – 16000)	(44950 – 15400)	(24250 ± 9150)		
Unit PM4 top	ESR MIN1402	218050 – 193000	230050 – 181000	205500 ± 12250	222500 – 184050	230500 – 115200	189300 ± 34350	106.8	2
					(226300 – 180200)	(234400 – 111300)	(189300 ± 36350)		
	pIR+TT-OSL PM16-6 ^c	228400 – 214000	235350 – 207050	221200 ± 7050	221850 – 201950	228950 – 187350	209600 ± 10700	112.6	1
					227300 – 215000	232450 – 208450	220950 ± 6200		
	ESR VII206	248500 – 227100	258800 – 216800	237800 ± 10500	238100 – 221850	247150 – 215150	230600 ± 8050	99.2	3
Unit PM4 bottom					248200 – 226300	259450 – 218400	238150 ± 10750		
					(255700 – 218800)	(307100 – 206750)	(238150 ± 18350)		
Unit PM3 top	pIR+TTOSL PM16-5 ^c	270350 – 237400	286200 – 221600	253900 ± 16150	264900 – 247700	271950 – 237000	255200 ± 8850	129.5	1
					(274500 – 238100)	(290800 – 218150)	(255200 ± 18250)		
	ESR VII204	294100 – 263600	308750 – 248900	278850 ± 14950	265900 – 250550	273150 – 242000	257750 ± 7650	68.4	9
					267600 – 253400	274450 – 246050	260250 ± 7050		
	ESR VII205	276100 – 254950	286300 – 244750	265500 ± 10400	269350 – 255450	276400 – 248300	262350 ± 6950	118.5	2
Unit PM3 bottom	pIR+TTOSL PM16-3 ^c	270300 – 232700	288350 – 214650	251500 ± 18450	271650 – 256500	279750 – 249150	264250 ± 7450	111.1	1
					274000 – 257800	283750 – 250150	266500 ± 8450		
					(284400 – 247400)	(304300 – 229600)	(266500 ± 18700)		
Unit PM1-2 top	ESR MIN1403	282600 – 246000	300200 – 228400	264300 ± 17950	286850 – 264800	301550 – 256200	277700 ± 11550	74.9	1
					(296300 – 255400)	(320450 – 237300)	(277700 ± 20850)		
					424000 – 272750	424000 – 270800	330700 ± 44850		
Unit PM1-2 bottom					(427050 – 269700)	(435450 – 259350)	(330700 ± 49400)		

^a The original dating results are all expressed in years before AD2014 (average sample collection year) for modelling purposes to ensure the likelihoods are directly comparable.

^b The modelled *boundary* age ranges and uncertainties shown outside parentheses represents the raw OxCal modelling output and are based on propagation of random dating uncertainties only. The modelled *boundary* age ranges and uncertainties shown in parentheses includes propagation of the modelled (random) dating uncertainties and the mean systematic uncertainty (6.4%) of the thirteen numerical dating estimates in quadrature.

c The combined (unmodelled) ages for luminescence samples PM16-3, PM16-4, PM16-5 and PM16-6 have each been calculated from their replicate pIR-IRSL and TT-OSL ages, or replicate pIR-IRSL, OSL and TT-OSL ages, using the OxCal *Combine* function. See Supplementary Information text for further details.

Table S3. Summary of Bayesian modelling results for Porto Maior (Model 2; see main text). The likelihood (unmodelled) and posterior (modelled) age ranges are presented for each of the numerical dating samples. Posterior (modelled) age ranges are also shown for the boundaries of each stratigraphic unit. Posterior ages are presented as the 68.2% and 95.4% highest probability density ranges. The mean and 1σ uncertainty ranges of the modelled posterior distributions are shown for comparison (assuming a normally distributed probability density function). The unmodelled and modelled age estimates have been rounded to the nearest 50 years.

Unit / boundary	Time variable	Modelled age (years) ^a			Modelled duration (years) ^{a, b}		
		68.2% range	95.4% range	Mean $\pm$ 1 σ	68.2% range	95.4% range	Mean $\pm$ 1 σ
PM5	age	24050 – 11050	32950 – 6150	18800 $\pm$ 6900			
PM5	duration				3250 – 21050	-150 – 30700	14550 $\pm$ 8650
PM5 – PM4 boundary	duration				126650 – 193500	89250 – 218950	156300 $\pm$ 32650
PM4	age	241000 – 192100	259800 – 157450	211150 $\pm$ 27250			
PM4	duration				15900 – 84200	-450 – 120550	57600 $\pm$ 34050
PM4 – PM3 boundary	duration				-14950 – 33150	-39550 – 56100	8500 $\pm$ 23850
PM3	age	276350 – 245750	292000 – 230450	260950 $\pm$ 15850			
PM3	duration				-250 – 31850	-250 – 62300	25050 $\pm$ 18600
PM3 – PM2 boundary	duration				-23750 – 28550	-50500 – 53600	1850 $\pm$ 25950
PM1-2	age	332000 – 271300	376800 – 251150	309600 $\pm$ 33700			
PM1-2	duration				9200 – 94650	-3150 – 149450	68550 $\pm$ 42750

^a Modelled age ranges were calculated from the posterior probabilities of the upper and lower boundaries (top and bottom) of each stratigraphic unit (see Table 5) using the *date* query function in OxCal v4.3. Modelled durations were calculated using the *difference* query function, and provide the temporal range between the posterior probability density distributions of successive stratigraphic boundaries.

^b The OxCal *difference* function can be used to test whether or not the posterior probability distributions of successive stratigraphic boundaries are significantly different from each other at a given confidence interval. When the *difference* function is applied to adjacent boundaries of two different stratigraphic units (e.g. LU-6 – LU-5 boundary), it provides a statistical indication of the presence or absence of potential depositional hiatuses (given the available dating evidence). Calculated duration ranges that overlap with 0 at the 95.4% confidence interval suggest that the boundaries of successive units are not separated by a statistically significant temporal hiatus. Calculated duration ranges that do not overlap with 0 at the 95.4% confidence interval indicate potentially missing material and / or a temporal gap between the boundaries of successive units. When the *difference* function is applied to the upper and lower boundaries of the same stratigraphic unit (as opposed to the adjacent boundaries of successive units), a calculated duration of >0 years indicate a statistically significant temporal difference between the onset and termination of the depositional event. In contrast, calculated duration ranges that overlap with 0 indicate statistically indistinguishable onset and termination ages for the depositional event at a given confidence interval.

Table S4. Bayesian modelled posterior age ranges and depositional durations for the Porto Maior stratigraphic units (Model 2). Posterior ages/durations are presented as the 68.2% and 95.4% highest probability density ranges. The mean and 1 σ uncertainty ranges of the modelled posterior distributions are shown for comparison (assuming a normally distributed probability density function). For the model 2 results shown here, the average shared systematic uncertainty of the 15 likelihood (unmodelled) ages has subsequently been added in quadrature to the posterior (modelled) probabilities of the upper and lower boundaries of each unit to enable direct comparisons with the posterior (modelled) probability ranges of Model 1. The unmodelled and modelled age estimates have been rounded to the nearest 50 years.