Methodological developments in the luminescence dating of brick from English late-medieval and post-medieval buildings

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Abstract

Fired clay brick samples, obtained from a group of seven high status late-medieval and post-medieval buildings in England ranging in age from ca AD 1390 to 1740, were dated by the luminescence method using an optically stimulated luminescence (OSL) technique. The results obtained indicate that, when applied to quartz extracted from brick, the technique is capable of producing dates that are in consistently good agreement with independent dating evidence for the buildings. For six samples taken from a group of four dating 'control' buildings the mean difference between the central values of luminescence and assigned ages was 5±10 years (s.d., n= 6). The methodology used is appropriate for application to other standing buildings in other temporal and geographic regions, and may be used with confidence where conventional dating methods are less certain. The study also examines the luminescence characteristics of quartz and the characteristics of the lithogenic radionuclides in brick samples and identifies various aspects related to the assessment of experimental uncertainty in testing the reliability of the method.

INTRODUCTION

During the last two millennia fired clay brick has become a ubiquitous building material in many parts of Europe and its chemical and physical robustness enable it to remain within the archaeological record for many centuries, whether as a structural element in a standing building or as an artefact in a buried context. Many surviving late medieval buildings erected wholly or partially in brick have been substantially altered since their original construction and the cumulative effect of many alterations requires detailed examination to unravel the complex structural history of the building. Such analysis, particularly when combined with documentary evidence and tree-ring dating of timbers, comprises the conventional approach to historic buildings analysis, and one which has the capability to date the original construction and subsequent alterations to within several years or better. However, problems in dating may arise, particularly in the study of vernacular buildings, where documentary evidence has not survived, or may have never existed, where tree-ring dates are not available (such as the replacement of original structural timbers, insufficient number of rings, etc.), and where there is an absence of diagnostic architectural features. In these circumstances the uncertainty in dating may rise to at least several decades, depending on the nature of the available building evidence. This paper discusses the results obtained from a program of luminescence dating of brick from a group of independently dated late-medieval and post-medieval English buildings with the purpose of evaluating the potential of the method in the interpretation and phasing of buildings of this period.

The use of brick in medieval England

Unlike the building traditions of continental W. Europe, there was a long interregnum following the departure of the Romans from Britain, during which brick was not manufactured until its relatively late reintroduction towards the end of the C12th (Moore, 1991; Salzman, 1967). Evidence of the earliest known local production of medieval brick is found in buildings in Essex, although there are examples elsewhere of early medieval buildings where it is uncertain whether ceramic tiles and bricks incorporated into the structure were re-used or of local contemporary manufacture (Clifton-Taylor, 1962; Wight, 1972; Ryan, 1996). It is known that some brick was imported from the Low Countries during the C13th and C14th for the construction of high status buildings, but there is documentary evidence for the emergence of a commerce in brick manufacture in detailed records related to brick production in Hull (Yorkshire) and Wisbech (Cambridgeshire) during the early C14th (Brooks, 1939; Sherlock, 1998; Smith, 1985). In the county of Lincolnshire, lying between these two known examples of production centres, locally produced brick is found in standing buildings that survive from the late C14th, although many of these buildings tend to be isolated rather than in urban settings (Smith, 1985). During the C15th the use of brick became fashionable in E. England and the number of brickyards and brickmaking sites expanded to meet local needs, rising to a peak of nearly 200 production centres in the late C19th (Robinson, 1999), facilitated by the widespread availability of suitable clays (Miller and Robinson, 1989). However it is only recently that archaeological work to classify basic brick fabric types has commenced (A. Vince, pers. comm.).

Apart from the work performed in our laboratory on buildings in Newcastle upon Tyne (Bailiff and Holland, 2000) and Suffolk (Antrobus, 2004), only very limited work on the luminescence dating of brick from buildings in England has been performed (Cramp *et al.*, 1977). This in contrast to work performed elsewhere in Europe where the potential of the application of the method to the study the architectural history of buildings was recognised at an earlier stage in the development of luminescence dating (Goedicke *et al.*, 1981). More recent work on standing buildings in Europe has included applications in Finland (Jungner, 1987; Hütt *et al.*, 2001), Germany (Göksu and Schwenk, 2001), Italy (Martini and Sibilia, 2001), Denmark (Abrahamsen *et al.*, 1998) and the Czech Republic (Čechak *et al.*, 2000).

Whereas the earlier work exploited higher levels of precision that can be obtained with the thermoluminescence (TL) fine grain technique (polymineral grains of 4-11 μm dia.), some of the more recent applications (e.g., Hütt *et al.*, 2001) have employed the quartz inclusion technique (quartz grains of typically 90-150 μm dia.) that is based on the measurement of the 210˚C TL peak. The latter was found to be a sensitive technique for use in retrospective dosimetry (Bailiff and Petrov, 1999) with modern bricks, prompting its application to dating. Studies of the kinetics of the trap(s) associated with the peak (Petrov and Bailiff, 1997; Göksu *et al.*, 2001) indicated its suitability in terms of trap lifetime for application to dating during the last millennium.

The use of quartz avoids anomalous fading that is associated with feldspar minerals and which, if present, causes an underestimation of the age due to loss of the latent luminescence signal (Aitken, 1985). The degree of signal loss is unpredictable and needs to be assessed for each sample, although recent studies suggest that it is positively correlated with K content in feldspars (Meisl and Huntley, 2005). When performing measurements with fine grain samples extracted from brick, the proportions of the detected luminescence due to feldspar and quartz will depend on the type of clay and the firing conditions, and the extent of the influence of the former may vary considerably. Although the results obtained with north Italian brick appear to have been both precise and accurate using the polymineral fine-grain technique (Goedicke *et al.*,1981), we have found its reliability when applied to English ceramic materials to be inconsistent, and attempts to remove feldspars using various acid etching procedures have generally not yielded satisfactory samples. The extraction of feldspar inclusions is also a further possibility to consider, but still requires testing for anomalous fading. Athough we have not undertaken a comprehensive survey of the performance of different techniques, dating work with English brick in this laboratory has focused on the extraction of quartz inclusions, and in earlier work the measurement of optically stimulated luminescence (OSL) with quartz was found to offer advantages in terms of signal sensitivity (Bailiff and Holland, 2000) compared with thermoluminescence (TL).

THE BUILDINGS

Brick samples were obtained from a group of seven late- and post-medieval buildings in England, six of which are located in Lincolnshire and one in Wiltshire. They were selected from a wider group of buildings currently under study and include late- and post- medieval buildings for which independent dating evidence is available. The assigned dates derived from the latter were based on a combination of structural analysis, documentary records and, for some of the buildings, included tree-ring dates for structural or roof timbers. The principal details of the buildings sampled, comprising Tattershall Castle, Doddington Hall, Ayscoughfee Hall, Alford Manor, Boston Guildhall and Fydell House located in Lincolnshire, and Clarendon House located in S E Wiltshire, are summarised in Table 1. Of these, five (Alford Manor, Ayscoughfee, Doddington, Boston Guildhall and Tattershall) are sufficiently closely dated to be considered as dating 'controls', and the later buildings, Fydell House and Clarendon House, although chronologically less tightly constrained, provide examples of testing phases of construction.

The construction of the Great Tower of Tattershall Castle (Lab. ref. 318) started in ca A.D. 1434 (Salzman, 1967). The massive brick solar tower, four stories in height (35.5 m), represented a stunning addition to a group of earlier manorial buildings, and aspects of its construction and function within the built landscape are discussed by Everson and Stocker (2007). Surviving accounts for 1445-46 record that the tower was constructed under the supervision of a Flemish 'brekemaker', Baldwin, and that 322,000 bricks were supplied for the construction of its base. (Simpson, 1955). Two brick samples were obtained from the interior walls of, one each from the NE and NW towers at the ground floor and the basement levels respectively.

Doddington Hall (Lab. ref. 317), Doddington, is a late Elizabethan house that was built in brick between 1593 and 1600 by Thomas Tailor; the design, based on an elongated H plan, is attributed to Robert Smythson who was the distinguished designer of a group of advanced late Elizabethan Midland houses (Pevsner *et al.*, 1989). An indenture dated 1593 records the sale of the Doddington Estate to Thomas Tailor (see note to Table 1), and a sheet of lead marked 'IW 1600' recovered from the cladding of the centre dome marks the completion of the Hall in 1600 (Jarvis, pers. comm.). Two samples (317-1a, -1b) were obtained from an internal foundation wall in the south wing directly below the ground floor. The bricks were produced locally with clay obtained within local beds.

St Mary's Guildhall (Lab. ref. 310), Boston, although having undergone much rebuilding and alteration, has extant original timber framework and external brickwork are considered to be largely original (Pevsner *et al.*, 1989). A recent structural investigation (Clark *et al.* 2003) placed the date of its initial construction to 1390 on the basis of tree-ring analysis of the framing and roof timbers (Howard, 2003). As a result of this work it has a special status as one of the earliest surviving brick buildings in Lincolnshire. Two samples of brick were taken from the exterior of the N elevation of the building, one associated with original phase of construction the building (310-1) and the other an adjoining mid C19th extension (310-2), taken for comparison.

Ayscoughfee Hall (Lab. ref. 319), Spalding, is a much altered building that was originally built on an H-plan. Previously considered to have been constructed in 1429 (Pevsner *et al.*, 1989), a recent structural assessment of the building, Clark and Mellor, 2005), supported by tree-ring dating of 34 roof timbers, proposes that the house was erected during the early 1450's. Two cores were taken from an internal gable wall associated with the original structure and located within the roof space (319 -1, -2).

Alford Manor, Alford (Lab. ref. 301), was until recently considered to have originated as a 'mud and stud' C16th house built on an H-plan with a thatched roof, and later (1661) encased in brick (Pevsner et al., 1989). Samples of brick were obtained from two locations, one at the rear corner of what was considered to be the mid C17th brick encasement (301-1) and the other, for comparison, from a later C18th extension (301-2). A recent analysis by Clark and Nash (2003) that includes tree-ring dates for structural timbers from both the roof and frame elements (Arnold *et al.*, 2003) provides a detailed account of the construction history. It concludes that the building was erected as a composite timber frame and brick structure and that construction followed shortly after A.D. 1611, which corresponds to the date obtained by tree-ring analysis for the felling of the timbers.

Fydell House (Lab. ref. 311), Boston, is described as a grand early C18th house (Pevsner *et al.*, 1989) constructed of brick on two levels and located adjacent to Boston Guildhall. Ironwork, embossed with the year 1726, and a date impressed brick mark the transfer of ownership of the house to Mr Fydell in that year, and alterations in the brickwork suggest some structural alterations were made. Apart from restoration of the upper floor following fire damage during the Second World War, the building has remained largely original since 1726 and is considered to be a fine example of the style of its period. Of six samples taken from this building at ground floor level and the garden walls, three (311 -2, -4, -6) from the main house are discussed in this paper, one of which (311-6) is putatively associated with reconstruction of the front façade.

Clarendon House, Wiltshire (Lab. Ref. 315) is reported to have been built in 1737 (Pevsner, 1975), but is considered to be stylistically earlier. It has been the subject of recent architectural and documentary research as part of a wider study of the medieval royal deer park and later estate (Beaumont James and Gerrard, 2006). Structural analysis indicates that the mansion has two main phases of construction. A brick core (315-4) was taken from an internal wall associated with the first phase and further core (315-5) was obtained from the internal face of the front elevation associated with the second phase of construction. Traditions of brickmaking on the estate suggest that the materials were locally sourced. The earliest phase is dated stylistically to the mid–late C17th, supported by tree-ring analysis that indicates the youngest structural timbers were felled after 1667 (Tyers, 1999). The date of

1737 given by Pevsner derives from a datestone, but in a recent expert assessment the third numeral of the datestone was judged to be sufficiently unclear to give rise to uncertainty as to whether the date was 1717 or 1737 (Gerrard, pers. comm.). The earlier date coincides with a change in ownership of the estate to the Bathurst family. Taking into account the range of available evidence, the current date range for the second phase cannot be refined to closer than 1717-1737.

EXPERIMENTAL

As discussed in more detail below, the quartz inclusion technique (Aitken, 1985; 1998) was applied to determine the luminescence age of the brick samples discussed in this paper. The age, A, is calculated by determining experimentally the values of the palaeodose, P, and the total dose-rate, $\dot{D}_{tot}$, to enable the age equation to be evaluated:

$$A = \frac{P}{\dot{D}_{tot}} \pm \sigma_A \, ; \pm \sigma_B \qquad (1)$$

where $\dot{D}_{tot} = a\dot{D}_{\alpha} + b\dot{D}_{\beta} + g\dot{D}_{\gamma} + \dot{D}_{cos}$.

$\dot{D}_{\alpha}$ is the alpha dose-rate due to alpha emitters within the interior of the quartz grains, a is the *a* value (Aitken, 1985) that accounts for the lower yield of luminescence per unit of absorbed dose, $\dot{D}_{\beta}$ and $\dot{D}_{\gamma}$ are the point-absorber infinite medium beta and gamma dose-rates respectively, b is a lumped correction factor related to the attenuation of beta radiation by quartz grains, taking into account the reduction in grain size due to HF etching, and differences in the absorption coefficient between ceramic and water, g is a lumped correction factor related the geometry of the sources of gamma radiation and to differences in the absorption coefficient between ceramic and water, and where $\dot{D}_{cos}$ is the cosmic ray dose-rate (Prescott and Hutton, 1988). The process of HF etching quartz grains is assumed to reduce the dose-rate contribution from external sources of alpha particles to a negligible level.

The age is given with two error terms based on the specification by Aitken (1985). The first error term, σ_A, is a type A standard uncertainty (ISO, 1993) obtained by an analysis of repeated observations and the second error term, σ_B, is a type B standard uncertainty based on an assessment of uncertainty associated with all the quantities employed in the calculation of the age, including those of type A, and is equivalent to the overall error described by Aitken (1985) . Expressions for both terms are derived from an analysis of the propagation of errors (see Aitken, 1985) when calculating the age using measured and calculated values, and hitherto this approach has been generally considered to be sufficiently robust. Unless stated otherwise, all the uncertainties discussed in this paper are given as $\pm 1\sigma$.

Brick sampling

Samples of brick were obtained using a diamond faced core drill of 50 mm diameter designed for dry cutting except in two cases where a whole brick was extracted (318-1b) and where a smaller core drill (38 mm dia.) was used (311-6). The drill speed and pressure applied during the cutting were adjusted to prevent excessive heating. With one exception (318), the contexts were generally dry for the locations discussed in this paper. The cores, once cut, were marked to indicate their location and orientation and packed in heavy gauge plastic tubing. Where the sampled location required cosmetic restoration, the brick cavities were back-filled with lime-based mortar and the surface impregnated with brick dust, and in some cases a cap cut from the core was inserted to reduce the amount of exposed mortar within the brick. A dosemeter capsule (glass tube, 3 mm-thick walls) containing dosimetry grade aluminium oxide (Al_2O_3:C; Landauer Inc., USA) was inserted into a hole drilled near to the core location to a measured depth (usually 10 cm), that was sufficient to place it in the rear half of the brick. The aluminium oxide records the combined dose–rate due to gamma and cosmic radiation while the beta rays from the surrounding material are absorbed within the tube walls. At some of the locations the gamma ray spectrum was measured using a NaI detector, Ametek) placed into the core cavity – the data obtained were used as a back-up to the dosemeter measurements and were not applied to dose-rate calculations discussed below.

Preparation of Samples

Following the removal of any residual mortar deposits, the cores (or bricks) were cut using a water lubricated diamond saw to obtain slices of controlled depth and thickness. A general assessment of the composition of the brick fabric was made using a low power stereo microscope by examining a cut surface (usually an end cap cut from the inner core surface) to examine for heterogeneity of the fabric due to rock fragments or agglomerations (Kaipa *et al.*, 1988), both of which may cause significant local variations in the beta dose-rate to individual grains. A ~10 mm thick slice was cut between 8 and 10 cm from the front surface of the brick, the exact depth depending on the composition and condition of the core. This thickness of slice usually provided sufficient material for both beta dose-rate and luminescence measurements, sampling for both from the same material volume being relevant in the case of non-uniform fabric composition. The slices were subsequently examined under red light to compare the fabric composition against the initial assessment.

Quartz inclusions

Quartz inclusions were extracted from the selected sieved fraction (as discussed below) by following a procedure based on that developed for the quartz inclusion technique (Aitken, 1985). After removal of the outer 1- 2 mm rim of the slice and a portion for dose-rate assessment, the slice was mechanically crushed and the material dry sieved to isolate the grain size fractions in the ranges <90 µm, 90-150 µm and 150-355 µm. Although the 90-150 µm fraction is preferred in this laboratory because it corresponds to the grain size range used in the primary beta source calibration, its availability in sufficient quantity depends on the coarseness of the temper, and in the case of bricks from location 319 the use of a finer temper necessitated the extraction of a smaller grain size range (53-90 μm).

Following a test for the presence of carbonates, the sieved fraction was etched in HF (40%, for 45 mins), immersed in HCl for 60 mins to remove precipitates, and finally re-sieved, with appropriate washing procedures applied at each stage. The quality of the etched material was assessed by visual examination of one or more aliquots under a low power microscope. The presence of feldspars in the etched material was tested by measuring the response of aliquots to infra-red stimulation following the application of a laboratory beta dose and pre-heat treatment – no significant IRSL was detected for the samples discussed in this paper and heavy liquid separation was considered to be unnecessary.

Polymineral fine grains

Polymineral fine grain discs were also prepared with several samples (samples 310-1,-2; 301-1,-2 and 311-2) to obtain dates for comparison with the quartz inclusion results. They were prepared following the conventional fine-grain procedure (Aitken, 1985).

Determination of palaeodose

Instrumentation

Luminescence measurements were performed using a TL-DA-12 semi-automated reader (Risø National Laboratory, Denmark). Optically stimulated luminescence (OSL) was detected after passing through a Hoya U340 filter (7.5 mm), and OSL decay curves were recorded using either blue diode (470 nm; ~50 mW cm^{-2}) or filtered tungsten-halogen (450-550 nm; ~30 mW cm^{-2}) stimulation sources. As part of the initial testing procedure, the TL signal from quartz inclusions was also measured using a heating rate of 5˚C s^{-1} (no optical filter was inserted in the detection system). Laboratory beta doses were administered to luminescence samples using a $^{90}Sr/^{90}Y$ beta source mounted on the reader that had been calibrated against a secondary standard ^{60}Co source (Göksu *et al.*, 1995). The beta irradiator was also used to administer doses to aliquots of Al_2O_3:C grains (*in situ* gamma-ray dosimetry), where an aluminium absorber was inserted below the irradiator aperture to provide a sufficiently low dose-rate (calibrated dose-rate, 224 μGy min^{-1}), in this case primarily due to bremmstrahlung rather than beta particles. Alpha doses were administered to polymineral fine grain samples using an irradiator containing ^{241}Am foil sources that had been sub-calibrated against the Oxford Research Laboratory for Archaeology alpha source (Aitken, 1985).

Procedures: quartz inclusions

Sample aliquots of typically 1-2 mg of quartz were deposited as a near monolayer onto stainless steel discs that had previously been coated with a thin layer of silicone oil and spread within a diameter of 6-8 mm. Initial tests were performed to establish the basic OSL and TL characteristics of each sample. On the basis of relatively poorer TL signal strength and levels of sensitisation in some samples, the palaeodose was determined using OSL for all the samples discussed here.

The absorbed dose, P, to quartz grains was determined using a single aliquot OSL regenerative procedure (Proc. A, Table 2) based on a procedure described previously (Bailiff and Holland, 2000). It is similar to the SAR procedure described by Murray and Wintle, 2000; 2003), but handles corrections for sensitization effects and thermal transfer differently, to suit the characteristics of comparatively young ceramic samples. The main steps are given in Table 2. A second OSL decay curve was recorded at each stage of the regenerative procedure (Proc. A, steps 3, 5, 7, 11) to monitor the OSL signal due to thermal transfer, referred to as a pre-heat monitor, and this was used to define the background signal. The degree of sensitization was measured at two levels of dose within each sequence (Proc A, steps 9 and 11) The preheat treatment, comprising a 10s hold at the selected temperature, T_P, was applied for temperatures between 200 and 260 ˚C to establish the form of the palaeodose-preheat characteristic (P vs T_P). The pattern of sensitivity change with cumulative dose was measured by repeating a series of regeneration cycles using the same dose and pre-heat temperature. The same procedure was applied to separate aliquots using a different preheat temperature. The beta doses administered in regeneration measurements ranged from 0.8 to 1.2 of the estimated cumulative dose (P), and this range was extended if further investigation of the growth characteristic was required. The measurements detailed in Procedure B (Table 1) were performed with further aliquots to provide data equivalent to the SAR procedure described by Murray and Wintle (2003).

Additive dose measurements were also applied to test for changes in luminescence properties during the first OSL measurement using Procedure B with one pre-heat temperature (220˚C). The 'additive' beta dose was administered before Step 1 (see Table 2) where three levels of additive dose were applied (β = 0.5P, P and 1.5P) to separate sets of aliquots.

Dose-rate assessment

The components of the total dose-rate, $\dot{D}_{tot}$, were determined by direct measurement of the contemporary dose-rate in pulverised brick samples (beta radiation) and, as described above, in the sampled walls (gamma and cosmic radiation) by employing luminescent dosemeters. Indirect methods based on the measurement of activity using thick source alpha counting (TSAC) and high resolution gamma spectrometry were also used to determine the concentrations of lithogenic radionuclides in the same pulverised samples of brick. Samples of mortar recovered from some of the locations during drilling were also analysed.

Instrumentation

The beta dose-rate was determined using the technique of β-TLD (Bailiff, 1982; Aitken, 1985). The system employs a 10 mm dia. calcium fluoride dosemeter to measure externally the beta dose-rate close to the surface of the sample and interposed between which is a mylar screen to absorb alpha particles. The gamma-ray activity of brick samples was measured using a gamma-ray spectrometer (configured with shielding for low background, Canberra high purity germanium coaxial detector type GR2018 of 20% efficiency and with a Be window). The spectrometer was calibrated using silica rich sands containing lithogenic radionuclides of certified concentrations (New Brunswick Laboratories, USA). The alpha activity of pulverised brick samples was determined using thick source alpha counting (TSAC), as described in Aitken (1985), with prefabricated ZnS scintillator screens of 42 mm diameter (Daybreak Nuclear Ltd, USA). The apparatus was calibrated using SiO_2 sands of certified U and Th concentrations (as above).

Procedures

β-TLD measurements were performed with several aliquots (~ 2 cm^3) of a pulverised portion of the same brick slice. The combined gamma and cosmic dose-rate was measured with the Al_2O_3:C dosemeter capsules for periods ranging from 0.5 to 5 years. Following retrieval of the dosemeters, the accrued absorbed dose was determined by measuring the TL signal and applying a regeneration procedure.

Thick source alpha counting (TSAC) was performed with pulverised brick samples (~ 5 g), in an unsealed state (α_0) and during the first 24 h following sealing (α_1). The alpha activity of HF etched quartz was also measured using the same measurement technique but where ~10 mg of pulverised quartz was deposited onto a 26.5 mm dia. mylar disc by sedimentation and placed into contact with a ZnS scintillator screen of the same diameter. Larger quantities (25 g) of the pulverised brick were measured to determine the activities of the ^{232}Th and ^{238}U decay series and ^{40}K using the high resolution gamma-ray spectrometer. The specific activities of the ^{232}Th and ^{238}U decay series were determined by measurement of the gamma-ray emissions by ^{228}Ac, ^{212}Bi, ^{212}Pb and ^{206}Tl (^{232}Th series), and ^{226}Ra, ^{214}Pb, ^{214}Bi and ^{210}Pb (^{238}U series).

A measure of the saturation water uptake by the brick fabric was obtained by immersing cut sections of brick in water at room temperature for a period of 5-10 days. The weight of water absorbed is expressed as a percentage of the weight of air-dried (50˚C) material.

RESULTS

Brick fabric

Examination of cut surfaces of brick indicated the absence of agglomerations of sand-size grains in all the samples. The fabrics of samples taken from 301, 315, 317, 319, 311, were predominantly uniform, and generally of fine composition with little distinguishing features. Sample 315-5 had a noticeably higher concentration of sand-size temper. The fabrics of samples from 318 were coarser with some stone fragments and various sizes of crystalline inclusions in the temper. All the bricks were of soft composition, but well fired, and of generally uniform red colour with no evidence of reduction or vitrification.

Palaeodose

Quartz inclusions

With the exception of three samples, the OSL intensities recorded were weak, and to obtain signals of adequate strength the stimulation source was adjusted to 90% of maximum power to allow the OSL signal to be resolved. A high proportion of the OSL decayed within several seconds and the decay curves were dominated by components (Bailey, 2001) judged to be 'fast'/'medium'; the 'slow' component was either absent or not resolved above the background signal. The relative strength of the natural OSL signal for each sample is indicated by the signal-to-background ratio, R_{SB}, the values of which are given in Table 3. Very bright emissions were obtained from three samples (311-6, 317-1a and 319-1), whereas sample 301-1 was the weakest, approaching a level where fluctuations due to poor signal statistics intrude (R_{SB} = ~0.5 - 1). The latter becomes noticeable at higher preheat temperatures and where, for example, a 50% reduction in intensity was observed when the preheat temperature was increased from 220 to 260°C in the case of sample 301.

The OSL signal was obtained by integrating the photon counts recorded during the first few seconds of stimulation (typically 5s), which was sufficient to account for at least 80% of the emission associated with the 'fast'/'medium' components. Once selected, the same period of integration was used for all measurements performed with one sample. The OSL data calculated in this manner were used to generate a growth characteristic, providing there was no significant change in the form of the decay curve within the integration range throughout the series of regeneration measurements. Where aliquots failed to meet this requirement, the results were excluded. On average, about 14% of all the aliquots measured were excluded from the calculation of P due to either change in the form of the decay curve, poor reproducibility in the growth characteristic data or large statistical fluctuations in the signal after subtraction of the background signal.

The estimate of P for each aliquot was obtained using the interpolation procedure applied in the standard regenerative technique and included corrections for changes in sensitivity during the repeated measurement cycles. The average degree of sensitivity change for these samples is low (Table 3), but in view of the variability between aliquots of the same sample (e.g. in half the cases the s.d. is ~ ±10% or greater) corrective procedures were applied. In the first, the sensitivity per unit dose was plotted against cumulative dose and, where there was a consistent pattern of change, a coefficient of sensitization was calculated and applied as a cumulative correction factor to the measured OSL signal at each measured point in the procedure. The second correction procedure followed that described by Murray and Wintle (2003), except that the 'test' dose was comparable to the size of P (as indicated in Table 2). The growth characteristics plotted with the sensitivity corrected values were found to be linear and with indication of negligible supralinear growth at low doses.

Two examples of the palaeodose-preheat temperature characteristic are shown in Fig. 1a,b for samples 311-6 and 315-5. Concordance of the values of P for a range of pre-heat temperatures is generally used as a qualitative test to confirm that i) the measured OSL is associated with the release of charge from traps that are thermally stable over the dating timescale of interest and ii) the regenerative procedure applied is self-consistent in terms of correcting for changes in sensitivity during the experiment. For preheat temperatures above 200˚C, the OSL signal is predicted by calculation to be stable for periods beyond a million years at ambient temperatures (Smith *et al.*, 1990), and thermal stability is consequently not an issue for dating during the last 1000 years. The behaviour observed ranged from samples exhibiting a resolved plateau with small dispersion in P (Fig. 1a), to cases where the dispersion was relatively high (Fig. 1b), and where the definition of a plateau over an equivalent range of preheat temperatures is not clear-cut.

The values of P used in the evaluation of the age equation correspond to the mean of the values obtained for a selected range of preheat temperatures (Table 3). The standard error of the mean value was used as the measure of uncertainty (col. 6). In some cases, where the values of P obtained for the highest and lowest preheat temperatures agreed within the limits of experimental error, the average value of P was calculated using data for only one pre-heat temperature selected within the range of the other preheat temperatures applied (col. 4) to optimize the precision in P. Using this procedure the standard error for the majority of samples ranges is less than ±1.5% (rising to ±3.2%, 311-4). It is interesting to note that bricks taken from the same wall had differing characteristics (317-1a, ±1% vs 317-1b, ±2.4%). The values of the range, ∆P, of the P values, expressed as a standardized value, obtained for each sample (Table 3, col. 5), were less than 4 and this is consistent with expectation for normally distributed values of P.

The results of the additive dose measurements were used to construct a characteristic similar to that developed for use in the pre-dose technique (Haskell and Bailiff, 1985) and also later adapted in the OSL SARA procedure (Mejdahl and Bøtter-Jensen, 1994). The estimate of the palaeodose, P_{Add}, was obtained by extrapolating a linear curve fitted to a plot of the total dose determined using a regenerative procedure vs. additional laboratory dose. A comparison of the values of palaeodose obtained using the additive (P_{Add}) and the standard regenerative procedure (denoted P_{Regen}) plotted in Fig. 2 indicate agreement within experimental uncertainty (slope = 1.06±0.06; s.d., n=10). The two procedures are expected to give the same value of palaeodose unless a change in the luminescence sensitivity occurs during the first OSL measurement, which would not be detected using the standard regenerative procedure. The need to extrapolate the characteristic obtained using additive doses gives rise to inferior precision in the palaeodose and hence it has not been used in the age calculation.

Amongst several potential causes of elevated levels of dispersion in measured values of P is spatial heterogeneity in dose-rate, in particular the beta dose-rate. Elevated and significantly varying concentrations of uranium and/or thorium within quartz grains may also lead to differences in the internal grain alpha dose-rate, but the alpha counting screening tests applied to the samples discussed here suggest low internal activity from these sources. In addition to poor mixing of coarse aggregates of significantly differing radionuclide content (e.g. flint and sandstone) of the type considered by Nathan *et al.* (2003) in sedimentary media for example, or the presence of grains with concentrations of beta emitting radionuclides that are significantly higher than in the ceramic matrix (e.g. potassic feldspars), agglomerations of quartz grains (Kaipa *et al.*, 1988) may occur in bricks. Agglomerations that break up during sample preparation can lead to a significant variation in the beta dose-rate (e.g. by 50%) to individual grains. Sensitivity to this type of heterogeneity is likely to be increased if the luminescence measured is dominated by a few ultra bright quartz grains within each aliquot. Tests using an OSL scanner (Bailiff and Mikhailik, 2003; Fig. 3) with aliquots of some of the samples in this study indicated that the luminescence detected was dominated by a small proportion of bright grains – similar to that widely observed in unheated single grain measurements with quartz extracted from sedimentary samples. Although visual inspection of cut surfaces of brick indicated the absence of agglomerations, which is supported by finding generally low dispersion in the values of P, some samples (e.g., 317-1b and 315-5) exhibited higher levels of dispersion in P (Fig. 1a,b). A proportionately wide range of values of R_{SB} due to poor signal statistics obtained with some aliquots probably accounts for the higher dispersion in P for sample 317-1b, and it is interesting to note that a sample taken from the same wall (317-1a) and separated by a couple of meters has markedly different characteristics. In the case of 315-5, the reproducibility of the signals (R_{SB} = 2.3±0.1) suggests that the dispersion in P is associated with heterogeneity in the beta dose-rate rather than with poor signal statistics. Pinpointing the source of variation requires more detailed work to establish the nature of the spatial relationship between quartz grains (as radiation dosemeters), other constituents in the fabric (absorbers, such as agglomerations) and the radionuclide sources. Recent work on the spatially resolved determination of absorbed dose in quartz grains in slices of brick has indicated that visually complex fabrics are not necessarily associated with significant dispersion in P (Bailiff, 2006).

Polymineral fine grain

The palaeodose was determined using an OSL additive dose, single aliquot regenerative, procedure. Tests of the stability of the signal after a 4-month storage period at ambient temperatures indicated loss of signal due to anomalous fading in all cases (10-40%). In view of this outcome further technical details of these measurements are not presented.

Dose-rate

The total dose-rate, $\dot{D}_{tot}$, was determined primarily from the results of β- and γ- TLD respectively. Data obtained using thick source alpha counting and high resolution gamma-ray spectrometry provided further detail of the contributions made to the dose-rate by different radionuclides and the extent of disequilibrium. The thick source alpha activity results were also applied in the calculation of the effective alpha dose-rate for the subset of samples that were tested using the fine-grain technique.

The values of $\dot{D}_{tot}$, used in the calculation of the luminescence age, are given for each sample in Table 4, together with a breakdown of the percentage contribution due to beta and combined gamma and cosmic radiation dose-rates. The majority of $\dot{D}_{tot}$ is due to beta radiation (average, 64%) and hence is associated with sources located within the brick. Measurements of the alpha activity of pulverised HF etched quartz performed where there was sufficient material (one sample from each building) indicated a count-rate approaching the background rate and corresponding to an effective internal alpha dose-rate of about 0.03 mGy a^{-1} assuming an efficiency of 5%. An allowance of 0.03±0.02 mGy a^{-1} to account for the internal grain dose-rate is included in the total dose-rate for all the samples. The screening of quartz for uranium and/or thorium indicated negligible internal radioactivity for the samples discussed here (samples of etched quartz are submitted for analysis using mass spectrometry if elevated activity is detected).

The specific activities of ^{238}U, ^{232}Th and ^{40}K and the value of the ^{210}Pb:^{226}Ra ratio, obtained by gamma spectrometry, are given in Table 4 (cols 4-7). These activities were converted to point absorber beta dose-rates (col. 9) using conversion factors taken from Adamiec and Aitken (1998) for the Th and U series and Nambi and Aitken (1986) for ^{40}K, and taking into account suggested revisions to the contribution from ^{87}Rb (Readhead, 2002a,b).

The measured ^{210}Pb: ^{226}Ra activity ratios (col. 7) indicate a partial loss of gaseous ^{222}Rn from all the bricks (*in situ*), leading to a lower dose-rate from the uranium series than would be obtained if the radon had been fully retained. (Aitken, 1985; Olley *et al.*, 1996). On the basis of the energy released by the ^{238}U decay series (Adameic and Aitken, 1998), full loss of ^{222}Rn within the sample volume of interest is expected to lead to reductions of 60% and 96% in the beta and gamma dose rates respectively due to natural uranium. In the case of sample 318-2, for example, this would lead to a reduction in beta and gamma dose-rates due to all the lithogenic sources of 9% and 29% respectively and, for the observed 30% deficit in ^{222}Rn, ~3% and ~9% respectively (^{210}Pb: ^{226}Ra activity ratio = 0.7; Table 4). Hence dose-rates calculated on the basis of the concentrations of the lithogenic radionuclides assuming secular equilibrium in the U and Th series are expected to be systematically higher than the experimentally determined dose rates (β TLD and γ TLD). The average value of the ratio of the infinite medium beta dose-rate calculated using concentration values and measured by β TLD (Table 4, col. 9) for the samples tested is 1.00±0.07 (s.d., n=12), with a range of 0.91-1.03. Although this level of precision is not sufficient to test for the expected differences discussed above, the data indicate overall consistency between the two methods.

A similar comparison of gamma and cosmic dose-rate at the dosemeter location is more complex because adjustments for the irradiation geometry are required. The combined gamma and cosmic dose-rate at each dosemeter location was calculated on the basis of the measured radionuclide concentrations (Table 4 and notes) and using data published by Prescott and Hutton (1988). The average value for the equivalent ratios of the combined gamma and cosmic dose-rate at the dosemeter locations (Table 4, col. 8) is 1.06±0.13 (s.d., n = 11) and, within the limitations of the comparison, this is consistent with the expected difference discussed above. A more precise comparison requires, in addition to the considerations of radon release from the bricks *in situ*, knowledge of the radionuclide concentrations within the other bricks surrounding the sample location that contribute to the gamma dose-rate, the composition and uniformity of which are presumed to be similar in the above calculations.

Moisture content

The moisture contents of the bricks when sampled were less than 2%; except at location 318 where they were higher (3-4%). The values assigned for the average moisture content since construction were 3±1% (all locations except 318) and 5±1% (318), and corrections to the dose-rate for differences between the absorption coefficients of water and ceramic were made using the factors (Table 4, notes) derived by Zimmerman (1971) and given in Aitken (1985). The measured values of saturation water uptake are also given in Table 4 (col. 10). These moisture content values were used to: i) adjust the beta dose-rate (Aitken, 1985), ii) calculate the uncertainty in both beta and gamma dose-rate due to fluctuations in moisture content and iii) adjust the gamma dose-rate where it was calculated using radionuclide concentration data and combined with the published conversion factors discussed above to compare with the dose-rate measured by the dosemeters.

Age calculation

The dose-rate, $\dot{D}_{tot}$, in the age equation represents a time-averaged value that is derived from contemporary measurements. Various assumptions and estimations concerning the uncertainty in the dose-rate associated with this extrapolation to the date of construction of the sampled part of the structure are made when calculating the age. It is assumed that the bricks were used shortly after manufacture and that the structure from which they were taken remained intact – i.e., that the bricks were not re-used from an earlier building. A relatively restricted range of the average moisture content is specified for the sampled locations (3±1% except 318, 5±1%), and these margins make a small contribution (<3% of the overall error) to the overall uncertainty in the age. This seems to be reasonable in the light of recent studies (Raimondo *et al.*, 2006) of the effect of microstructure and pore morphology on the equilibrium moisture content (MEq; absorbed water per dry volume) of modern fired clay bricks, where a value of MEq converted to absorbed water per unit dry weight of ~3% was obtained for bricks with a total porosity of ~35% (70% RH).

Nonetheless, since larger variations in average moisture content have a potentially strong effect on the value of the dose-rate, it is important to be aware of the sensitivity in the age calculation. The calculated effect of changes in the average moisture content on the age for a given value of palaeodose is illustrated in Fig. 4, using data for sample 318-2. As the average moisture content rises, the age increases due to the reduction in dose-rate, and the width of the age distribution is related to the uncertainty specified in the moisture content. In moving from a dry condition to 10% moisture content (saturation level for 318-2), the relationship is linear and the age increases from 530±32 to 574±37 years, corresponding to approximately 5 years increase in age per % increment in average moisture content. The moisture content values are assumed to be normally distributed about the mean value in these calculations, but for larger variations other distributions, such as log-normal, may be more appropriate. Significant departures from steady-state conditions in the past would consequently have potentially important implications on the interpretation of dates and hence this needs to be considered when selecting samples. A more quantitative approach to arriving at an estimate of the average moisture content is likely to require an analysis of the pore size distribution (Raimondo *et al.*, 2006) and this type of investigation has yet to be explored.

The luminescence properties of the quartz extracted from the additional samples obtained from Ayscoughfee Hall (319) and Boston Guildhall (310), were not favourable for dating measurements (due to weak luminescence signals) and this reflects the variability of properties that can occur between bricks of similar age and manufacture taken from the same building.

DISCUSSION

Consistently good agreement between the luminescence and assigned date ranges was obtained within the time span investigated of about 350 years (Fig. 5; Table 5). For the dating ‘control’ locations of Boston Guildhall (310), Doddington Hall (317), Tattershall (318) and

Ayscoughfee (319) the mean difference, Δd, between the central values of luminescence and assigned ages is 5±10 years, (s.d., n= 6), taking each result to provide an independent test of comparison. Calculation of the χ^2 statistic for the differences indicates agreement between the assigned and luminescence dates at a significance level of 0.05 (χ^2 = 6.2; d.f.=4). The larger difference observed in the case of Alford Manor is discussed further below. The variation in Δd for this group was also calculated by means of a simple Monte Carlo simulation that assumed normal and uniform distributions associated with the luminescence and assigned dates respectively; it produced a distribution that is approximately normal, with a central value of +5 years and with a standard deviation of ±10 years. Using either approach, the average difference compares favourably with the value of the error term σ_B associated with the luminescence dates (±17 to ±37 years) calculated using the standard uncertainty assessment procedure.

The luminescence dates for the pairs of samples taken from the same phase of the same building (Doddington, 317; Tattershall, 318) overlap within the σ_A uncertainty limits, indicating self-consistency of the dating results for each of these buildings. However, in the case of Alford (301) and Ayscoughfee (319), one of the two core samples taken could not be dated because the luminescence signals from quartz inclusions were too weak. Unfortunately it has not been possible so far to identify ‘good’ luminescence samples from an inspection of the external brick surface.

The central value of the Alford Manor luminescence date, although some 55 years earlier than the assigned date (Clark and Nash, 2003), lies just within $2\sigma_B$ limits ($\Delta d = 1.9\sigma_B$) of the assigned date range of 1611-1615. However, it is now understood (J. Clark, pers. comm.) that the sampled brick was located in an area of brickwork at a corner of the building that had been inserted during the early C18th following removal of the original timber corner post. The altered nature of the brickwork had not been realised at the time of sampling, which preceded the structural analysis by at least a year. This raises both technical luminescence and building historical issues. If it is assumed that the brick was re-used, the dose-rate history of the brick before the C18th alteration is uncertain. The main component of the dose-rate that is uncertain is the gamma and cosmic ray dose-rate (30-40% of the total dose-rate) during the period beyond ~300 years in age. Providing the brick was amongst other bricks (in a wall or within a discarded pile), the average combined gamma and cosmic dose-rate for the sampled brick during the period in question is likely to be within 20% of the currently assessed average dose-rate. If the average gamma and cosmic dose-rate is changed from 110 to 90% of the value given in Table 4 to account for this, the central value of the date changes from 1530 to 1570; in other words, making only a slight change to that given in Table 5. It is possible that the brick sampled was a re-used Alford Manor brick (ca 1611), but a 50% increase in the average combined gamma and cosmic dose-rate would be required to yield a date that is consistent with the assigned date range, and this is considered improbable. The dating evidence points more towards the use of brick from another structure of age similar to or greater than that of Alford Manor. It may be relevant that the highest ratio of calculated to measured gamma dosemeter dose-rate (Table 4) is for Alford, indicating differences in the composition of the bricks within the repaired section. Although the more pressing issue for the application of luminescence to this particular building is the testing of brickwork firmly associated with the 1611 construction, the detection of the practice of re-use of brick from other structures of differing age presents a potentially interesting area of investigation.

The luminescence date of 1730 ±11; ±18 (315-5) for the later phase sample from Clarendon House falls within the assigned range of 1717-1737, although the overall uncertainty is not sufficient to resolve the issue of interpretation of the datestone. However, the luminescence date of 1688 ±8; ±18 (315-4) for the sample taken from the interior wall confirms that it is associated with an earlier phase of the building since the difference of 42 years between the central values of the two ages is shown to be significant using Ward and Wilson’s (1978) test statistic T (T=9.5; $\chi^2_{1,0.05}$ = 3.84), where σ_A was used as the weighting factor in calculating the pooled date.

The current resolving power of the method applied in this study is more severely tested in the case of the three Fydell House samples 311-2, -4 and -6 (1727±8;±17, 1709±12±20, 1721±10±17 respectively). The pooled age for these samples is 1719 ±6; ±12 and the value of the T statistic, applied as discussed above, is 1.6 which indicates that the dates are not significantly different ($\chi^2_{2,0.05}$ = 5.99) as a group. Hence the range of the pooled date (1707-1731) accommodates both of the assigned date ranges for the construction and suggested alterations.

An examination of the calculations for all the dates discussed above indicates that the major contributor to the error term σ_B is the systematic error associated with the calibration of the laboratory radiation sources and the various coefficients related to dose-rate assessment, equivalent to the uncertainties σ_4 and σ_5 in Aitken (1985). Following Aitken, a margin of ±5% associated with calibration factors and the various coefficients has been retained in these calculations, although the indication from the above control group comparison is that a lower value may be appropriate. However, given the potential for variation in the various parameters to exceed this margin, particularly in respect of the dose-rate, it would be prudent to retain the ±5% margin until a major methodological reassessment of calibrations and uncertainty assessment is completed.

The average of the error terms σ_A and σ_B for the samples tested correspond to about ±3% and ±6% of their luminescence ages respectively. At this level of calculated uncertainty (σ_B; corresponding to 1σ ranges of ~35 to ~80 years for the period 1700-1300 respectively), application of the method to the dating of late medieval brick buildings is potentially appropriate where conventional analysis or tree-ring dating has been unable to provide absolute chronological placement to better than about 50 years. Also, the provision of absolute dates for studying the re-use of brick, may offer a new direction of investigation. Although this was unwittingly detected at Alford, it has been more systematically investigated in a study of a group of buildings in Newcastle upon Tyne (Bailiff *et al.*, in preparation). The level of precision (σ_A) that is routinely achievable with English brick requires further testing by applying the method more widely and, in particular, to the analysis of multiple phase buildings. When producing and comparing a series of dates from the same building, for which the error term σ_A can be used, it is likely that the application of Bayesian analysis (e.g., Millard, 2006) to a sequence of dates would enhance the resolving power of the method. Also, since different bricks of the same manufacture may have widely differing luminescence responses and may not yield measurable signals in some cases, the sampling of several bricks is advisable even if the surviving structure is believed to be original and of one phase. The opportunities for multiple sampling of this type, while usually limited in fully restored buildings, are likely to be significantly improved in terms of access and the structural detail revealed where buildings are undergoing restoration, or subject to formal recording required by legislated planning guidance.

The detection of anomalous fading in the fine grain samples tested in this study indicates that the long term stability tests should form part of initial tests if the fine grain method is to be used. However, the occurrence of this characteristic is likely to vary according to clay source and, for example, in one case of a brick sampled from a building in North East England where anomalous fading was absent in the fine grain sample, good agreement between fine grain and quartz inclusion dates was obtained. The fine grain method has the advantage of requiring less sample, offering greater flexibility where cosmetic damage to walls is an issue, and a lower dependence of the total dose-rate on sources external to the brick, which is desirable where there are doubts concerning the integrity of the brickwork or structural changes in the local environment. On the other hand, the introduction of a third component of dosimetry (alpha) increases the overall error due to the addition of a further set of experimental uncertainties.

As with the determination of the palaeodose, the reliability of the assessment of dose-rate can be expected to be improved by using two independent methods of measurement, and derivation of the total dose rate by both experimental dosimetry and by calculation based on measured radionuclide concentrations are desirable. The long term radon emanation from undisturbed brick fabric indicated by the gamma spectrometry measurements affects both systems of measurement but requires adjusted conversion factors to be applied to the concentration values to obtain the current dose-rate. The assumption that the latter is representative of the lifetime average for the sampled structure is susceptible to unknown variations in radon emanation and moisture content in the past (and noting the former is related to the latter; Aitken, 1985), and this affects both methods of measurement. The concordance of the luminescence and assigned dates indicate that the assumptions made concerning this extrapolation are reasonable for this study. Similar agreement elsewhere, is likely to be influenced by the possibility of the selecting sample locations that are currently dry and where there have not been prolonged episodes of elevated moisture levels in the past. Amongst the various coefficients and factors that are applied in the calculation of the dose-rate using both methods, three in particular need further refinement to reduce the experimental uncertainties in general application: i) the wall geometry factor which is currently limited to a plane wall geometry, ii) the reduction in the average dose-rate due radon release and iii) the dosemeter wall attenuation factor (Aznar et al, 2003), given that the design of encapsulation varies between laboratories.

One further point is that by testing material towards the rear of the sampled brick (~10 cm from the surface for these samples), the effect of contributions to the palaeodose from radionuclide sources located beyond the immediate vicinity of the sampled core such as adjacent walls and layers in the form of plaster that may have been added at a later stage during the history of the building are reduced due to shielding by the material closer to the external surface.

CONCLUSION

The results of this study indicate that the OSL technique applied to quartz extracted from brick is capable of producing dates that are in good agreement with independent dating evidence. For six samples taken from a group of four 'control' buildings the mean difference between the central values of luminescence and assigned ages was 5±10 years (s.d., n= 6). This was tested with a group of high status buildings in Lincolnshire as a precursor to a wider study of vernacular English buildings, but the methodology used is appropriate for application to other standing buildings in other temporal and geographic regions, and might be used with confidence where conventional dating methods are less certain. The study also examined the luminescence characteristics of quartz and the characteristics of the lithogenic radionuclides in brick samples and identified various aspects related to the assessment of experimental uncertainty that will be examined in wider testing of the method.

Although the potential of applying TL in architectural historical studies was demonstrated over two decades ago by the Berlin group in their study on northern Italian villas, this work provides the basis for applying the method to English buildings, particularly of the late medieval period, where hitherto the method has not been exploited. The dating of brick from earlier medieval English buildings is also a long standing research question awaiting further study. Although luminescence cannot provide the degree of chronological resolution comparable to the best provided by conventional buildings analysis combined with tree-ring analysis, there are many instances where the dating of brickwork in vernacular buildings is uncertain due to a lack of diagnostic features or suitable timber structural elements for tree-ring analysis. Moreover, by providing a direct means of dating of bricks as artefacts, luminescence has a role that is complementary to conventional buildings analysis. It also opens up the possibility of contributing to the study of the re-use of brick in structures, and more generally how this may relate to lost buildings in the archaeological record.

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Table 1. Summary of buildings and dating assessment

Lab. ref.	Building	Assigned date range A.D.	Assessment
301	Alford Manor House Alford, Lincs.	1611-1615	Buildings analysis incl. dendrochronology. (Clark and Nash, 2003; Arnold *et al.*, 2003; Pevsner *et al.*, 1989)
310	St Mary's Guildhall Boston, Lincs.	1390-1395	Buildings analysis incl. dendrochronology. (Clark *et al.*, 2003; Pevsner *et al.*, 1989)
311	Fydell House Boston, Lincs.	Ph 1. 1705-?1710 Ph 2. 1725-1726	Documentary sources and date marked ironwork (Ph. 2) (Pevsner *et al.*, 1989)
315	Clarendon House, Wilts.	Ph 1. 1650-?1675 Ph 2. 1727-1737	Documentary sources, stylistic dating and datestone. (Beaumont James and Gerrard, 2006; Pevsner, 1975)
317	Doddington Hall Doddington, Lincs.	1593-1600	Documentary sources and architectural style. (Jarvis, pers. comm.[1]; Pevsner *et al.*, 1989)
318	Tattershall Castle Tattershall, Lincs	1445-1450	Documentary sources and architectural style. (Simpson, 1955; Salzman, 1967; Pevsner *et al.*, 1989)
319	Ayscoughfee Hall Spalding, Lincs.	1450-1455	Buildings analysis incl. dendrochronology. (Clark and Mellor, 2005; Pevsner *et al.*, 1989)

1. An indenture, dated 1 June Eliz 1593 between John Savyle and Thomas Tailor, who built the Hall, records the purchase of the Estate (Public Record Office, Close Rolls 35 Eliz Part 4; Ref C54/1440).

Table 2. OSL measurement procedures

	Proc. A	Measurement		Proc. B	Measurement
1	PH; OSL	Pre heat using a selected temperature within the range 200-260 °C; measure OSL.	1	PH; OSL	Pre heat using a selected temperature within the range 200-260 °C; measure OSL.
2	PH; OSL	Pre-heat monitor (PHM)	2	PH; OSL	Pre-heat monitor (PHM)
3	+ 0.8 ß; PH; OSL	1st dose point / Sensitivity Monitor.	3	+ 0.8 ß; PH; OSL	1st dose point / Sensitivity Monitor
4	PH; OSL	PHM	4	PH; OSL	PHM
5	+ ß; PH; OSL	2nd dose point	5	+ ß; PH; OSL	2nd dose point
6	PH; OSL	PHM	6	PH; OSL	PHM
7	+1.2 ß; PH; OSL	3rd dose point	7	+ 0.8 ß; PH; OSL	Sensitivity Monitor
8	PH; OSL	PHM	8	PH; OSL	PHM
9	+ 0.8 ß; PH; OSL	Sensitivity Monitor 1	9	+ 1.2 ß; PH; OSL	3rd dose point
10	PH; OSL	PHM	10	PH; OSL	PHM
11	+ 1.2 ß; PH; OSL	Sensitivity Monitor 2	11-19		Repeat Steps 3-10
12	PH; OSL	PHM	20	+ 0.8 ß; PH; OSL	Sensitivity Monitor

Notes

1. The OSL decay curve was measured for 50-100 s depending on the characteristics of the particular sample; the sample temperature was held at 125ºC during stimulation.
2. The preheat (PH) was performed by heating the aliquot (5˚C s^{-1}) to a maximum temperature selected in the range 200-260 ˚C and holding the sample at that temperature for 10s.
3. The symbol ß indicates the administration of a laboratory beta dose corresponding to the value of the estimated palaeodose, P.
4. The additive dose procedure (applying either Proc. A or Proc. B) followed the same sequence except that a laboratory beta dose ß' was administered before step 1. The value of ß in the subsequent steps (3, 5, 7, etc.) was adjusted to reflect the combined dose (P+ ß') received before the first OSL measurement.

Table 3. Palaeodose determination

Lab. ref.	R_{SB} ± s.d.	Sensitivity change ± s.d.	P eval. PhT range (˚C)	P range, $\Delta P/\sigma$	P ± s.e. (mGy)	n
301-1	1.2±0.4	0.98±0.13	220-250	3.4	1270±20	16
310-1	3.6±1.0	1.06±0.09	220-240	3.6	2698±31	10
311-2	1.8±0.8	1.00±0.11	220-250	2.8	1054±18	11
311-4	1.8±0.6	1.10±0.16	210-240	3.3	1152±37	14
311-6	38±1	1.04±0.02	220-240	3.5	971±10	14
315-4	2.9±0.4	0.98±0.08	210-230	2.9	1120±16	6
315-5	2.3±0.1	1.01±0.14	220 (200-240)	3.5	948±33	8
317-1a	51±11	1.03±0.03	220 (200-240)	2.5	1357±12	8
317-1b	3.8±3.4	1.05±0.09	210-230	2.6	1558±38	9
318-1	6.7±1.5	1.01±0.03	220 (200-240)	2.9	1783±22	11
318-2	4.5±0.4	1.03±0.05	220 (200-240)	3.2	1840±21	8
319-1	28±14	1.10±0.03	220 (200-240)	3.1	1930±19	7

Notes.

1. The sensitivity change corresponds to the ratio of the OSL signals measured at point 9 to that measured at point 1 of Proc. A.

2. The range in P, ΔP, is expressed as a standardized value, $\Delta P/\sigma$, for the results obtained using the preheat temperature indicated in the adjacent column.

Table 4. Dose-rate determination

Lab. ref.	TLD: $b\dot{D}_{\beta}$ mGy a^{-1}	TLD: $g\dot{D}_{\gamma}+\dot{D}_{cos}$ mGy a^{-1}	Gamma Spectrometer: Th Bq kg^{-1}	U Bq kg^{-1}	K Bq kg^{-1}	Ratio $^{210}Pb/^{226}Ra$	Ratio D_{γ} Conc./ γTLD	Ratio D_{β} Conc./ βTLD	H_20 sat. % dry weight
301-1	1.89 (67%)	0.93 (33%)	45.6 ±3.2	40.2 ±1.6	527 ±8	0.9±0.1	1.27	1.03	17
310-1	2.99 (68%)	1.38 (32%)	62.0 ±3.5	45.7 ±1.7	868 ±10	0.8±0.1	1.16	0.98	17
311-2	2.48 (65%)	1.31 (35%)	47.8 ±3.2	36.9 ±1.6	700 ±9	0.9±0.1	1.04	0.94	22
311-4	2.58 (66%)	1.31 (34%)	51.5 ±3.2	38.0 ±1.6	735 ±9	1.0±0.1	1.06	0.95	15
311-6	1.96 (57%)	1.26 (37%)	35.0 ±4.0	36.8 ±2.0	600 ±9	0.9±0.1	-	1.03	-
315-4	1.98 (58%)	1.41 (42%)	40.7 ±3.1	34.6 ±1.6	760 ±10	0.9±0.1	0.90	1.14	18
315-5	2.11 (61%)	1.35 (39%)	40.5 ±3.1	35.1 ±1.6	700 ±9	0.8±0.1	0.87	0.94	19
317-1a	2.05 (63%)	1.19 (37%)	56.6 ±3.3	43.8 ±1.7	610 ±9	0.8±0.1	1.14	1.10	16
317-1b	2.42 (67%)	1.21 (33%)	61.6 ±6.3	40.4 ±3.0	673 ±17	0.8±0.1	1.17	0.98	11
318-1	2.09 (64%)	1.16 (36%)	41.1 ±3.1	32.0 ±1.5	654 ±9	0.9±0.1	1.01	1.0	14
318-2	2.26 (68%)	1.07 (32%)	41.1 ±3.1	32.8 ±1.5	665 ±9	0.7±0.1	1.10	0.88	13
319-1	2.14 (62%)	1.32 (38%)	37.8 ±3.1	29.0 ±1.5	672 ±9	0.8±0.1	0.90	1.0	26

Notes.

1. The dose-rates given in the first two cols. are the experimentally determined values to which corrections for moisture content (β dose rate) and attenuation (β and γ dose rates) have been applied, as discussed in the main text. Details of the attenuation factors incorporated in the lumped correction factors b and g in Eqn. 1 are given below; the alpha dose contribution is assumed to be negligible for these samples.

2. The values of the point absorber beta dose-rate within the tested material, as measured using β-TLD, were reduced (by 7% for 90-150 μm grains) to account for the effects of attenuation due to the finite size of the quartz grains using data published by Brennan (2003).

3. The gamma dose-rates, as measured by γ-TLD, were increased by 8% (after making an allowance for the estimated cosmic dose contribution) to account for the attenuation of gamma radiation by

the dosemeter capsule wall. This value was derived from Monte Carlo simulations for a dosemeter located within a brick wall. Where the dosemeter was located within 1 cm of the depth of the core section tested, no further adjustment to the measured dose-rate was performed. Otherwise, a correction (< 5%) was made to the dose-rate to adjust for the rise in the gamma dose-rate with depth using dose-rate profiles (Bailiff, 2001) that employ coefficients calculated by Løvborg and presented in Aitken (1985, Appdx. H).

3. The combined gamma and cosmic dose-rate at the dosemeter location was calculated using the concentrations of the lithogenic radionuclides measured in brick by gamma spectrometry, using a general value for mortar (U/Th/K: 30/30/300 Bq kg^{-1}). The calculations took into account the relative masses of brick and mortar within the wall structure, estimated on the basis of the average dimensions of brick and mortar thickness. The infinite medium gamma dose-rate was calculated from concentration values using published conversion factors, as discussed in the main text, and the dose-rate at the dosemeter position was obtained by application of a wall geometry factor, the value of which is related to dosemeter depth and wall thickness (e.g. 0.85 for at 10 cm depth in a 50 cm thick wall). The estimated values of the cosmic dose-rate were in the range 0.15 – 0.2 mGy a^{-1}, where the upper limit was used for external samples and samples in roof spaces. For buildings where the roof material had been changed from thatch to mineral tile, it was assumed that the average cosmic dose-rates at a depth of 10 cm in brick were likely to be similar (typical reed thatch thickness is 35 kg m^{-2}).

4. Corrections to dose-rates for moisture content were made using factors (ratio of absorption coefficients for water and ceramic) of 1.25 and 1.14 for the beta and gamma dose-rates respectively, as calculated by Zimmerman (1971).

Table 5. Luminescence and assigned dates

	Luminescence			Independent Assessment
Lab. Ref.	$\dot{D}_{tot}$ ± s.e.	P ±s.e.	Date $\pm\sigma_A$; $\pm\sigma_B$	Assigned Date Range
Dur05 OSLqi-	mGy a^{-1}	mGy	A.D.	A.D.
301-1	2.82±0.06	1270±20	1555 ±12; ±27	1611-1615
310-1	4.37±0.10	2698±31	1388 ±16; ±37	1390-1395
311-2	3.79±0.08	1054±18	1727 ±8; ±17	1700-1726
311-4	3.89±0.09	1152±37	1709 ±12; ±20	1700-1726
311-6	3.42±0.12	971±10	1721 ±10; ±17	1724-1726
315-4	3.39±0.07	1120±16	1688 ±8; ±18	1667-1690
315-5	3.46±0.08	948±33	1730 ±11; ±18	1717-1737
317-1a	3.24±0.07	1357±12	1586 ±10; ±24	1593-1600
317-1b	3.64±0.08	1558±38	1576 ±14; ±27	1593-1600
318-1	3.25±0.07	1790±24	1455 ±14; ±33	1445-1450
318-2	3.33±0.08	1840±28	1453 ±15; ±34	1445-1450
319-1	3.46±0.08	1930±19	1447 ±13; ±32	1450-1455

Notes.

The luminescence ages were calculated from the year of measurement, 2005, as indicated in the sample reference code (i.e., Dur05OSLqi-301-1).

Figure captions

Figure 1a.
Palaeodose vs pre-heat temperature characteristic, sample 311-6. Each data point represents the value of P evaluated using one aliquot and error bars fall within the data symbols (shaded circles).

Figure 1b.
Palaeodose vs pre-heat temperature characteristic for sample 315-5. Each data point represents the value of P evaluated using one aliquot. The average value obtained at each of the three preheat temperatures is indicated by an open square and the error bar represents the standard deviation.

Figure 2.
Comparison of values of palaeodose obtained using an additive dose regenerative procedure, P_{Add}, plotted against those obtained using a regenerative procedure, P_{Regen}, as discussed in the main text. The dotted line represents the line of concordance. The error bars correspond to the standard error of the mean value, some of which (P_{Regen}) fall within the shaded symbols.

Figure 3.
Map of OSL emission from an aliquot of etched quartz extracted from sample 319-1 that had been used in routine dating measurements. The aliquot was irradiated (~2 Gy) and preheated (220˚C, 10s) before the aliquot was scanned. The aliquot comprised ~ 0.5 mg of quartz grains distributed within a diameter of ~4 mm and the scanned area is 10 x 10 mm.

Figure 4.
Effect of change in average moisture content on the calculated luminescence age, shown for sample 318-2. The error bars for the moisture content values are ±20% of the central value used in the age calculation; the distribution is assumed to be normal. The dotted line is shown to indicate that the relationship between age and variation in moisture content is linear in the range shown here.

Figure 5.

Plot showing luminescence date vs assigned date for: (1) Boston Guildhall, (2) Tattershall Castle, (3) Ayscoughfee Hall, (4) Doddington Hall, (5) Fydell House (pooled date) and (6) Clarendon House. Concordance of the two dating systems is indicated by the dotted line. The error bars correspond to the overall uncertainty associated with the luminescence date, σ_B.

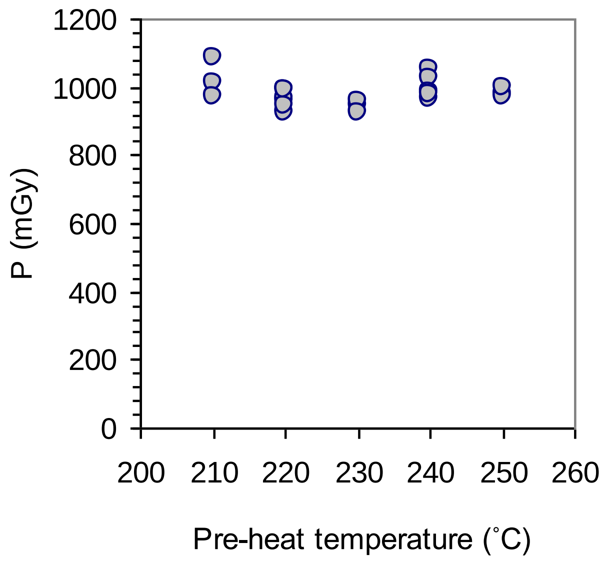


Figure 1a.

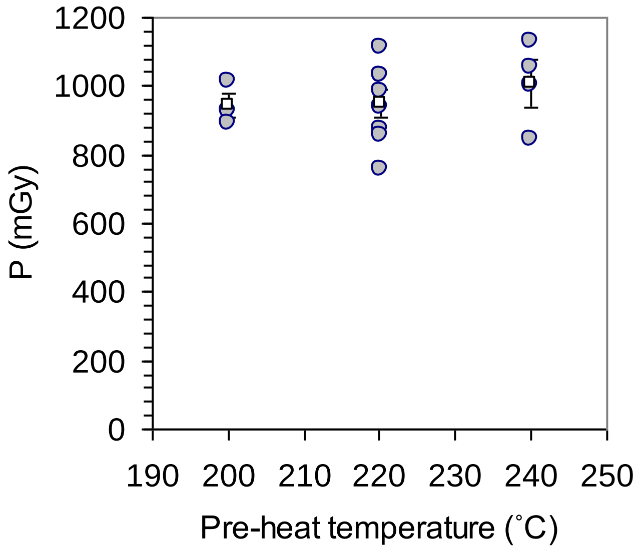


Figure 1b.

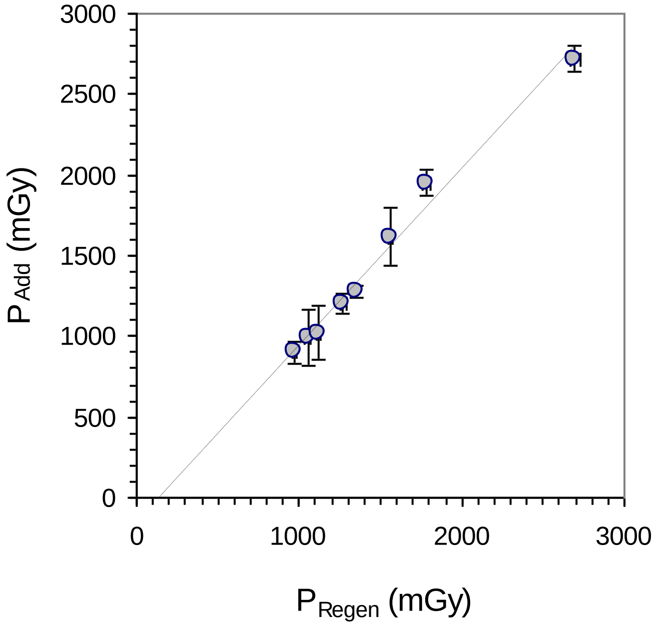

3000
2500
2000
1500
1000
500
0
0
1000
2000
3000
P Add (mGy)
P Regen (mGy)

Figure 2.

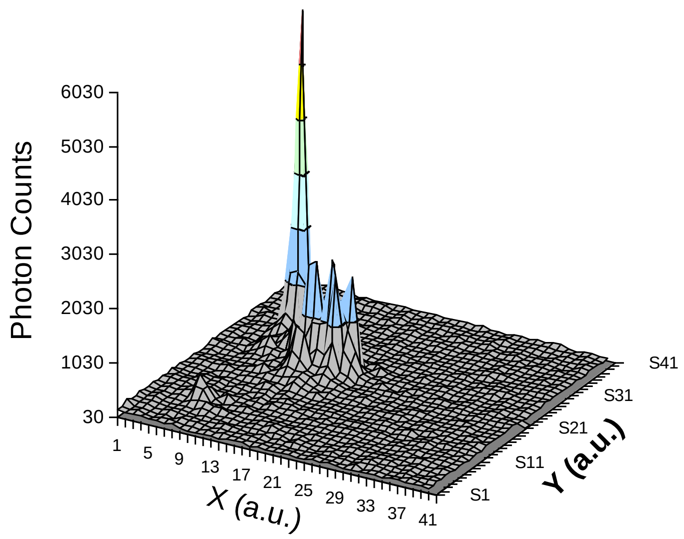
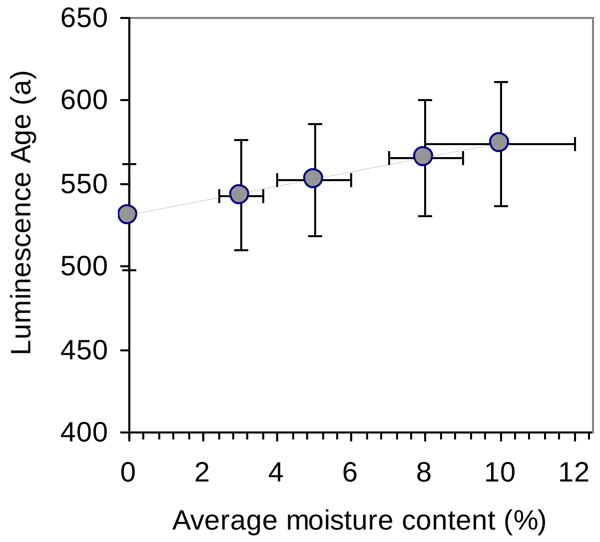

Photon Counts
6030
5030
4030
3030
2030
1030
30
1
5
9
13
17
21
25
29
33
37
41
X (a.u.)
S1
S11
S21
S31
S41
Y (a.u.)

Figure 3.



650
600
550
500
450
400
Luminescence Age (a)
0
2
4
6
8
10
12
Average moisture content (%)

Figure 4.

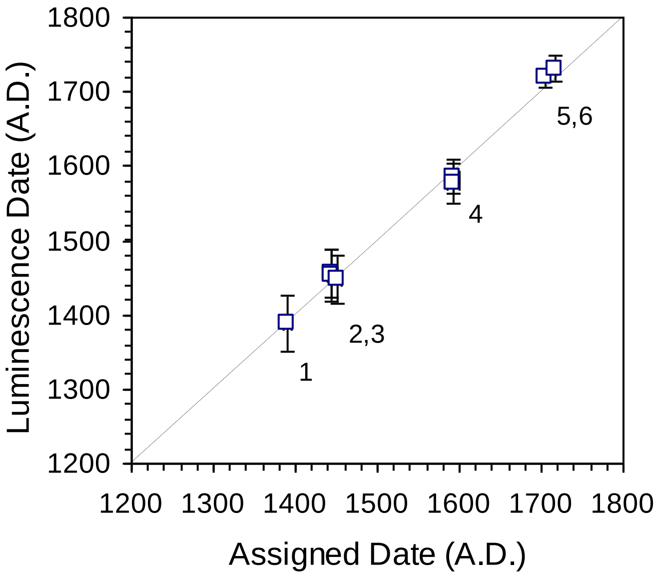

Luminescence Date (A.D.)
1800
1700
1600
1500
1400
1300
1200
5,6
4
2,3
1
1200 1300 1400 1500 1600 1700 1800
Assigned Date (A.D.)

Figure 5.