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Sources of uncertainties in OSL dating of archaeological mortars: The case study of the Roman amphitheatre "*Palais-Gallien*" in Bordeaux

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HIGHLIGHTS

- Fewer than 8% of all measured grains exhibited an OSL signal.
- The heterogeneity of β-irradiation arises principally from variations of K-content.
- Beta-imaging confirmed inhomogeneous distribution of beta sources in studied samples.
- The samples are highly affected by heterogeneous bleaching.
- Standard age models are not adapted for the studied series of samples.

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ABSTRACT

Archaeological mortars are more convenient and much more representative for the chronology of buildings than brick or wood constructions that can be re-used from older buildings. Before dating unknown samples of mortars, further investigation of OSL from mortars is required and the most efficient methodology needs to be established. In this study we compared the ages obtained by OSL dating of quartz extracted from mortars of the Roman amphitheatre *Palais-Gallien* in Bordeaux with independent age information.

Resetting of the OSL signal occurred during the preparation of mortar when grains of sand (quartz) were extracted and mixed with lime and water. The mortar was subsequently hidden from light by embedding within the structure which is the event to be dated.

Various factors contribute to uncertainties in the age determination. The frequency of measured equivalent doses reveals a large scattering. Optical bleaching of certain grains can be partial due to the short duration of the exposure to light. We worked with the single grain technique in order to find and select the grains that were sufficiently exposed to daylight. To determine the average equivalent dose, we tried three different approaches: a calculation of an arithmetic mean and one following either the central age model or the 3-parameter minimum age model, the latter turned out to be the only relevant way to evaluate the experimental data. The proportion of grains included in the calculation of the average equivalent dose represents 2.7–4.7 % of the overall analysed grains. The results obtained for the three out of four samples are approaching the expected age, however, the minimum doses and the corresponding ages are significantly over-estimated in case of two samples.

The studied material is very coarse, which causes heterogeneity of irradiation at the single grain scale, and contributes to a dispersion of equivalent doses. Different analytical methods (scanning electron microscopy with energy dispersive X-ray spectroscopy cartography, Beta-radiography imaging, *inductively coupled plasma mass spectrometry*) were employed to demonstrate the presence of this phenomenon.

Despite the extremely large proportion of high equivalent doses in equivalent dose distributions, there is an apparent presence of well-bleached grains at the beginning of equivalent dose distributions. The study shows the potential of dating mortars by single grain OSL.

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1. Introduction

1.1. Luminescence dating in building archaeology

Luminescence dating is a powerful tool for age determination of the last heating or the last exposure to light of ancient inorganic materials. Apart from its widespread use in paleo-environmental studies, prehistory and other related fields, it plays a substantial role in recent archaeology. From the 1980's there has been an increasing interest in thermoluminescence dating applied to fired pottery and then to fired ceramic building materials, such as architectural ceramics (for instance, Goedicke et al., 1981; Guibert et al., 1998; Bailiff and Holland, 2000). In many cases TL or OSL was used for brick dating as an ideal alternative to the radiocarbon method in order to date the construction phases where organic elements are absent from a dated structure (assuming the production of bricks is contemporary to the building). The current trends in building and urban archaeology imply that the combination of diverse dating techniques with an archaeological approach allows a better understanding of architectural structures. Many recent studies have highlighted the complementarity of various dating methods such as dendrochronology, radiocarbon, luminescence and archaeomagnetism (Gallo et al., 1999; Blain et al., 2007; Guibert et al., 2009a; Blain et al., 2011). Besides the enhancement of former interpretations and the improvement in accuracy if a convergence in dating results is observed, the combination of dating techniques can, for example, be a unique tool to detect a re-use of hard building elements like bricks (Galli et al., 2004; Bailiff et al., 2010; Bouvier et al., 2013). Thus, to a certain extent dating methods can bring information about the recycling of building materials and from a more general point of view they contribute potentially to a better knowledge of the history of the construction (Sapin et al., 2008; Guibert et al., 2012).

The possibility of reusing ceramic building materials means there is a risk that they are not contemporary to the construction of the structure under study. In addition, many historical buildings are constructed from stone, mud bricks or *pisé*, thus they do not contain any fired building element which could be dated.

To obtain the date of the construction it is possible to use luminescence techniques which have already been performed on the surface of siliceous stones and bricks that were exposed to light when building elements were embedded in masonry. In general, this technique is rather difficult to carry out and the success depends greatly on the transparency of the material being sampled, on the possibility of sampling interior parts of walls of heritage classified monuments, and finally on OSL properties of the material itself (Habermann et al., 2000; Greilich et al., 2005; Liritzis and Vafiadou, 2005; Greilich and Wagner, 2006; Vieillevigne et al., 2006; Liritzis et al., 2007, 2010; Liritzis, 2011; Cassen et al., 2013). Despite its potential, surface dating cannot be easily employed and that is why our attention is now being turned towards mortar.

The mortar is common to a majority of historical buildings and it represents the only building material that cannot be replaced without being destroyed, because of its mechanical properties. Thus, it is more suitable and much more representative for the chronology of buildings when compared to brick or wood constructions, since its making is undoubtedly contemporary to the building.

1.2. OSL dating versus radiocarbon dating of mortars

The unique advantage of mortar dating for building archaeology was recognized many years ago. For a long time attempts were focused on the radiocarbon method that has its limitations however. While the dating of organic remains from mortar may prove not to be contemporary to the mortar itself, the analyses of calcite formed through the carbonation process in lime mortar (Pesce and Ball, 2012) is often complicated by a problematic separation of the studied calcite from the geological calcite – either due to inefficient lime burning (calcination) or due to the adjunction of pieces of limestone to the aggregates – and by the risk that the hardening might not have occurred fast enough. Due to all the difficulties mentioned the radiocarbon method cannot be universally used. Thus, optically stimulated luminescence (OSL) becomes a promising alternative.

1.3. OSL of mortars: particularities

Lime mortars composed of a mixture of sand, lime and water can be dated by optically stimulated luminescence: the basic premise in such an analysis is that the quartz in the sand used for making mortar is optically zeroed during the process of quarrying and mixing. When the production process is over, the mortar is hidden from light because it is embedded within the structure – the moment to be dated.

The problem of mortar dating by OSL actually reveals certain common characteristics with the dating of fluvial sediments, such as partial bleaching and variable beta dose rate. Further complication is due to the young age of the mortar (2500 years maximum in Europe) implying dim luminescence signals. It is therefore necessary to take into account these potential obstacles so that a suitable methodology can be developed.

1.4. Current state of the problem & objectives of the study

A couple of isolated solitary experiments of mortar dating by OSL were performed between 2000 and 2013 (Bøtter-Jensen et al., 2000a; Goedicke, 2002; Zacharias et al., 2002; Jain et al., 2004; Goedicke, 2011; Gueli et al., 2010; Panzeri, 2013). Mortar was first recognized as a suitable dosimeter (for the reconstruction of accident doses after incidents at nuclear power plants) by Bøtter-Jensen et al. (2000a). A first attempt to perform optical dating on mortars was carried out by Zacharias et al. (2002) on 90–250 μm quartz inclusions extracted from samples taken from two Byzantine churches. In the 2002 paper Ch. Goedicke (2002) discussed the possible degree of bleaching of mortar and presents single aliquot measurements of 19 mortar samples from the last millennium. For 5 out of 19 samples the archaeological dose can be determined, leading to the suggestion of carrying out a single grain analysis for heterogeneously bleached samples. For the other 14 samples he compared evaluation methods (weighted histograms, radial plots) of frequency distributions, obtaining more asymmetric distributions for larger grain sizes, thus suggesting coarser grains are better bleached. However, none of the evaluation methods proved to be universal for all the samples.

In 2010 Goedicke (2011) performed a complete dating procedure on the set of samples from the Roman city of *Mogontiacum*, now modern Mainz, Germany. He concludes that for half of the 14 cases a classical SAR dating protocol was not suitable. Therefore, he recommends the determination of the ratio of the fast to slow component of the OSL emission and of the degree of bleaching in order to predict a dating potential for the mortar. He underlines the necessity of the single grain analysis for poorly bleached samples.

Another approach is discussed in the article of A. Gueli et al. (2010). They perform the OSL dating working on a fine-grained fraction of mortar samples from the 12th and 15th centuries AD and observe a good agreement between the ages obtained for mortar samples and bricks.

Jain et al. (2004) focused on the analysis of modern mortar samples comparing the results of single aliquots and single grains.