

## THE STATE-OF-THE-ART OF DATING TECHNIQUES APPLIED TO ANCIENT MORTARS AND BINDERS: A REVIEW

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**ABSTRACT.** The most recent workshop on mortar dating (25–27 Oct. 2018, Bordeaux, Montaigne University, France), which closely followed the publication of an extensive round robin-exercise involving several laboratories, was an opportunity to review the history and challenges of mortar dating methods and procedures currently in use. This review stems from the keynote lectures presented at the meeting, and wishes to summarize recent results, present trends, and future challenges. Three major areas are brought into focus (1) radiocarbon ( $^{14}\text{C}$ ) dating of complex mortars: can we assess the chances of successful dating?, (2)  $^{14}\text{C}$  dating of archaeological carbonate materials: difficulties, new directions and applications, and (3) single grain optically stimulated luminescence (OSL) dating of mortars in architectural archaeology: the current state of the art. This paper reflects the material presented by the authors and discussed at the workshop.

**KEYWORDS:** mortar characterization, mortar dating, radiocarbon dating, review, single grain OSL dating.

### INTRODUCTION

Ancient binders used for architectural purposes (i.e. binding and waterproofing agents in constructions, plasters, etc.) are mostly rather complex materials (Table 1) and through the history of architecture they developed from simple clays to modern concrete systems (Artioli et al. 2019). The necessity to find reliable ways of dating architectural binding systems derives from the fact that they are direct testaments to structural architecture developments and changes, they are the plastered support of our visual past (as frescoes and decorations) and they are the fundamental technical components of hydric reservoirs and waterways. Determining the chronology of the architectural components, including binders, is thus of great importance in decoding the construction technologies and building history of the past.

Two physical dating methods currently enable us to date binders: radiocarbon ( $^{14}\text{C}$ ) dating and optically stimulated luminescence (OSL). The aim of the  $^{14}\text{C}$  method is to date the moment of lime carbonation and so it applies to the binder component in lime-based mortars. On the other hand, OSL dating addresses the analysis of the aggregate in mortars, with the objective of determining the moment of its last exposure to light, prior to the embedding of the mortar within a built structure. This paper attempts to review the current state-of-the-art of both applications.

### $^{14}\text{C}$ DATING OF COMPLEX MORTARS: CAN WE ASSESS THE CHANCES OF SUCCESSFUL DATING?

The possibility of  $^{14}\text{C}$  dating mortars based on simple lime systems was first proposed more than fifty years ago (Labeyrie and Delibrias 1964). It is regarded as a potentially very

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Table 1 Main classes of binding compounds produced by pyrotechnology (modified from Artioli 2010 and Artioli et al. 2019).

Starting reactive material	Production process	Material-water mixture	Final product	Mineral phases in the hardened aged material
Lime-plaster (quicklime)	Calcinations of limestone	Slaked lime (lime putty)	Lime plaster	Calcite, aragonite
		Slaked lime + fine aggregate	Lime mortar	Calcite, aragonite + aggregate
		Slaked lime + fine aggregate + pozzolanic material (natural volcanics and/or pottery)	Hydraulic mortar (Roman opus caementitium)	Calcite, aragonite, zeolites, C-S-H + aggregate, M-S-H + aggregate
Gypsum-plaster (plaster of Paris)	Calcination of dolomite	Slaked magnesia-lime	Dolomitic or magnesian plaster	Calcite, brucite, periclase
	Calcination of gypsum	Bassanite ( $\pm$ anhydrite)	Gypsum plaster	Gypsum
Natural hydraulic lime (NHL)	Calcination of impure limestones	Bassanite + fine aggregate	Gypsum mortar	Gypsum + aggregate
		Slaked lime + activated clays	Hydraulic mortar	Calcite + C-S-H + Afm/AFt
Portland-clinker	Calcinations of limestones+clay	Portland cement paste	Portland cement	Portlandite, C-S-H, calcite
		Portland cement paste + fine aggregate	Portland cement mortar	Portlandite, C-S-H, calcite + aggregate
		Portland cement paste + fine and coarse aggregate	Concrete	Portlandite, C-S-H, calcite + aggregate
		Portland cement paste + fine aggregate + pozzolan	Pozzolanic Portland cement mortar	Portlandite, C-S-H, calcite, Ca-aluminosilicates
		Portland cement paste + fine aggregate + SCM (supplementary cementitious materials)	Blended cements	Portlandite, C-S-H, calcite, complex phases in the C-A-S-H system

C-S-H: calcium silica hydrate; C-A-S-H: calcium-aluminum-silica-water system; M-S-H: magnesium silica hydrate; AFm: aluminate ferrite monosulfate phases of hydrocalumite type; Aft: calcium tri-sulfo aluminate hydrate phases.

useful technique for archeological and conservation activities. The method is based on the reliable assumption that throughout history the lime binders used in construction were invariably obtained by the calcination of calcium carbonate (both of geological origin, that is limestones and marbles, or of biological origin, such as shells and corals). Heating any polymorph of  $\text{CaCO}_3$  (calcite, aragonite) above approximately  $850^\circ\text{C}$  releases  $\text{CO}_2$  and produces lime ( $\text{CaO}$ ), which is normally converted to portlandite ( $\text{Ca(OH)}_2$ ) by mixing it with a sufficient amount of water, a process called slaking. The slaked lime (or lime putty) is then used as the binder in masonry or as plaster (Barba and Villaseñor Alonso 2013; Artioli et al. 2019). When in place, the portlandite is converted back to calcium carbonate by  $\text{CO}_2$  absorption from the atmosphere: it is the carbonation process that makes a durable and resistant binder through  $\text{CaCO}_3$  crystallization. This dating method assumes that after the emplacement of the lime binder the carbonation process occurs rapidly (i.e. weeks or months) with respect to the architectural history of the building. Therefore, the measurement of the  $^{14}\text{C}$  content of the binder should yield the age of the corresponding construction phase. Indeed, simple lime systems that do not contain carbonate-containing phases other than those derived from the slaked lime carbonation (i.e. the pristine binder carbonate) can actually be straightforwardly and reliably dated (e.g. sample 1, Finnish Mortar in the MODIS exercise: Hajdas et al. 2017; Hayen et al. 2017). Problems arise when carbonate impurities of a different origin are present, as discussed below.

Van Strydonck (2016) recently reviewed the conceptual and technical developments in the history of the method. Most dating approaches largely rely on the step-dissolution strategy, in an attempt to separately date the pristine fraction of the binder carbonate (see for example Ringbom et al. 2014). However, in the last decade establishing an agreed measurement protocol potentially applicable to all ancient lime mortars has proven to be a very challenging task. The key issue is how to extract, identify, characterize, and measure the original fraction of carbonated portlandite that carries the  $^{14}\text{C}$  atmospheric signal relative to the date of the building. The original calcite (sometimes defined as the anthropogenic carbonate) may be mixed or contaminated with geological limestone (geogenic carbonate), secondary calcite due to dissolution and re-precipitation, and even secondary phases containing fossil  $\text{CO}_2$ , such as the layered double hydroxides (LDH) of the hydrocalcite-hydrocalumite type (Artioli et al. 2017; Ponce-Antón et al. 2018). In all of these cases (Figure 1) the obtained  $^{14}\text{C}$  dates are incompatible with the expected chronology of the material, as clearly shown by the recent round-robin exercise (Hajdas et al. 2017; Hayen et al. 2017).

Furthermore, it has become apparent that the presently adopted sample preparation and  $^{14}\text{C}$  extraction techniques are unsuitable for binding systems that are more complex than pure lime binders (Table 1). This includes all the materials undergoing some kind of hydraulic reaction. The compelling question in ancient mortar dating therefore is “can we really predict whether and to what extent  $^{14}\text{C}$  dating is bound to be successful and reliable for a specific sample?”

The cited recent round-robin exercise, called MODIS (mortar dating inter-comparison study: Hajdas et al. 2017; Hayen et al. 2017), clearly established that the  $^{14}\text{C}$  dating of lime mortars is straightforward and reliable only for suitably pure lime systems. Such is the reference case study of the Finnish sample from the bedding mortar of a wall from the church of Nagu, in the Åland archipelago, Finland. This sample (Sample 1) is the only one that yielded reasonably correct dates (i.e. dates compatible with the  $^{14}\text{C}$  results obtained on a coeval fragment of wood), independent of the binder separation method and the measurement

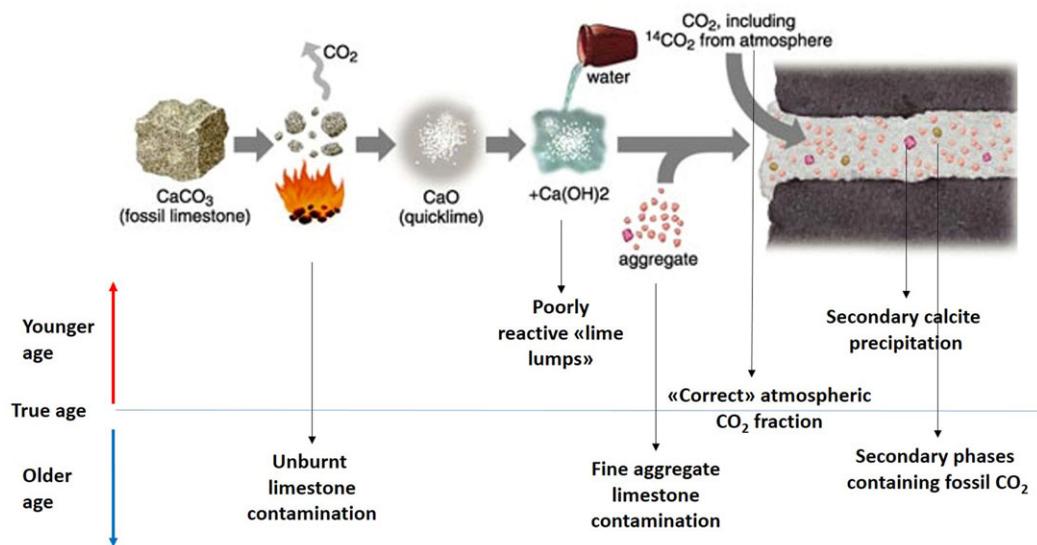


Figure 1 The lime cycle (modified from Hale et al. 2003) showing some of the possible contamination that can bias the dating results.

protocol. All other samples, namely a lime conglomerate from a burial of Cova S'Estora, Son Pellisser on the island of Mallorca, Spain (sample 2, Van Strydonck et al. 2017); the remains of a medieval mortar mixer from Basel Cathedral Hill, Switzerland (Sample 3); and a rendering from a Roman wall excavated in the city of Tongeren, Belgium (Sample 4), yielded inaccurate results, depending on the composition of the separated binding fraction subject to  $^{14}\text{C}$  measurement. The inter-comparison exercise has now provided a reasonable idea for why the results were biased. In the case of both sample 2 (the Mallorca burial) and sample 3 (the medieval mortar mixer from Basel) a fine fraction of geological carbonate pervaded the binder, for different reasons. Since it was impossible to separate the geologic carbonate from the anthropogenic one with simple mechanical or chemical processes, only the very first dissolution fractions lead to reliable dates, and most of the resulting dates were substantially older than expected. Sample 3 shows the presence of older carbon-containing materials (bones, wood) mixed with the mortar. Sample 2 is further complicated by the presence of late reactions involving dolomite and hydromagnesite likely entrapping older  $\text{CO}_2$ . The late de-dolomitization reaction in an alkaline environment (the so-called alkali-carbonate reaction: Katayama 2004, 2010) is well studied in modern concrete, but it is actually a rather frequent process in ancient mortars too. The consequence of the dolomite decomposition is the release of Mg in different forms (especially brucite in the early stages) which rapidly converts into  $\text{CO}_3$  containing phases, such as LDHs and hydromagnesite. The "old" carbonate released may also be incorporated in the precipitating secondary calcite, with deleterious consequences for dating.

In the case of sample 4 (the Roman cocciopesto mortar), all of the contributing laboratories obtained younger ages with respect to the dates in agreement with the coeval charcoal. We here propose that the bias may be systematically caused by the presence of younger  $\text{CO}_2$  entrapped in the layered double hydroxides (LDH, Pöllmann 2006; Mills et al. 2012), which are commonly formed during late hydraulic reactions in cocciopesto-containing mortars (Artioli et al. 2017).

The fact that the same sample processed by different laboratories may yield different results was initially recognized quite a while ago and caused a good deal of concern (Hodgins et al. 2011; Lindroos et al. 2011). It is evident that the binder fraction cannot be simply extracted and dated, and some kind of prescreening procedure is needed to assess the adequacy of the sample for the dating method, and its chance of success. It is envisaged that the correct dating strategy conceptually ought to include the following steps:

1. An in-depth characterization of the sample;
2. The preliminary interpretation of the phases present, the chemical-physical history of the sample, and the assessment of whether a “dateable fraction” can be separated by chemical or physical means;
3. The practical extraction of the binder fraction, and finally;
4. The  $^{14}\text{C}$  measurement.

The joint Padova-Caserta research group also advocates a further checking of the purity of the separated binder fraction between steps 3 and 4 in order to control the material that is actually bound to be dated (see Figure 2 of Addis et al. 2019). The *dateable fraction* of the carbonate binder may be tentatively defined as “the separated fine fraction of anthropogenic carbonate that has been experimentally screened for the absence of secondary carbonate, geogenic carbonate, and spurious  $\text{CO}_2$ -containing phases”. The use of separation techniques and the necessity to have sufficient material available for the screening of the dateable fraction implies that a substantial amount of the original sample is required in order to have enough material for dating. The total sample amount required of course depends on the purity of the lime binder and on the original binder to aggregate ratio in the mortar. For average samples based on a traditional 1:2 binder to aggregate ratio it is estimated that a few grams of mortar could be required.

Most developments in the past two decades focused on the extraction methods (see for example: Van Strydonck et al. 1992; Sonninen and Jungner 2001; Marzaioli et al. 2011; Ortega et al. 2012; Ringbom et al. 2014), so that in most cases at present only steps 3 and 4 are commonly carried out. Judgement of the reliability of the resulting dates is thus often based on expectations or external considerations, such as supporting archaeological information, stratigraphy, or dates derived from stratigraphically consistent materials.

Previous studies have shown that a thorough mineralogical, petrological and geochemical characterization of the mortar sample is advised in order to plan a better sample preparation and dating strategy, or at least to have a basis for the correct interpretation of the results (Addis et al. 2016, 2019; Van Strydonck 2016; Marzaioli et al. 2019). Hayen et al (2017) even stated that “Prescreening and characterization of the mortars should become a standard approach prior to the mortar dating in order to understand the failure risks of the dating process and to evaluate the data obtained”.

What, then, are the techniques and protocols that should be used to characterize ancient mortars?

The mineralogical and petrological investigation of historical mortars is well established (Vendrell-Saz et al. 1996; Franzini et al. 1999; Crisci et al. 2004; Elsen 2006; Ortega et al. 2008), and commonly encompasses a number of traditional mineralogical techniques. Building on the protocol introduced by Crisci et al. (2004) and leaving aside the

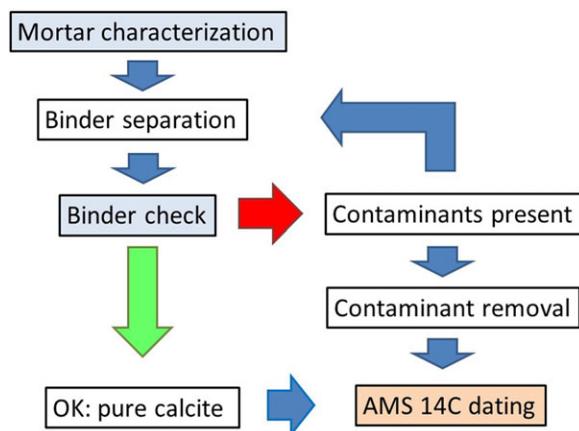


Figure 2 Flow chart of the characterization and processing activities prior to  $^{14}\text{C}$  dating.

measurement of the bulk physical properties of the material, the following techniques are considered to be suitable for a preliminary mineralogical assessment of the material: (a) thin section analysis of the phases, micro-texture, and reaction layers by optical and electron microscopies (reflected-light optical microscopy, RLOM, scanning electron microscopy with energy dispersive spectrometry, SEM/EDS); (b) a quantitative assessment of the crystalline phases by X-ray powder diffraction (XRPD) and an evaluation of the amorphous content by advanced full-profile analysis using internal standard (Rietveld-based quantitative phase analysis, QPA) and Fourier Transform Infrared spectroscopy, FTIR, and; (c) a quantitative assessment of the  $\text{H}_2\text{O}$  and  $\text{CO}_2$  containing phases by thermal analyses (thermo-gravimetric analysis, TGA, differential thermal analysis, DTA, differential scanning calorimetry, DSC).

Use of these techniques (Jones 1987; Mukherjee 2012) is expected to provide a good overview of (1) the nature of the crystalline and amorphous phases present in the material, (2) their history in terms of crystallization sequence, equilibrium state, or reaction history. Based on such information, especially the mineralogical and micro-textural features, it is normally possible to define if there is sufficient dateable calcite, if this calcite is the pristine anthropogenic carbonate, and if possible contaminants are present, such as secondary carbonates, geological carbonates, or  $\text{CO}_2$ -containing phases, as discussed above. This first step should indicate the most suitable method for the separation of the binder calcite (Figure 2).

If the binder separation is carried out chemically or thermally directly in the  $\text{CO}_2$  separating pipeline, then there is no possibility of further examination. If a substantial amount of carbonate is present in the mortar, then a physical separation method is usually adopted, generally a combination of cryo-breaking, sonication, centrifugation, sieving, and/or sedimentation techniques. The ultimate aim is to separate the binder particles from any aggregate component. In rare optimal cases, the separated fraction is pure and ready for  $^{14}\text{C}$  measurements (step 4). However, this fraction is often contaminated by fine particles of the aggregate (frequently clays and carbonates) or secondary phases such as compounds of the LDH family discussed above, which are naturally nanostructured. To date, detection of these contaminant phases during the “binder check” phase (Figure 2) is relatively easy. The real

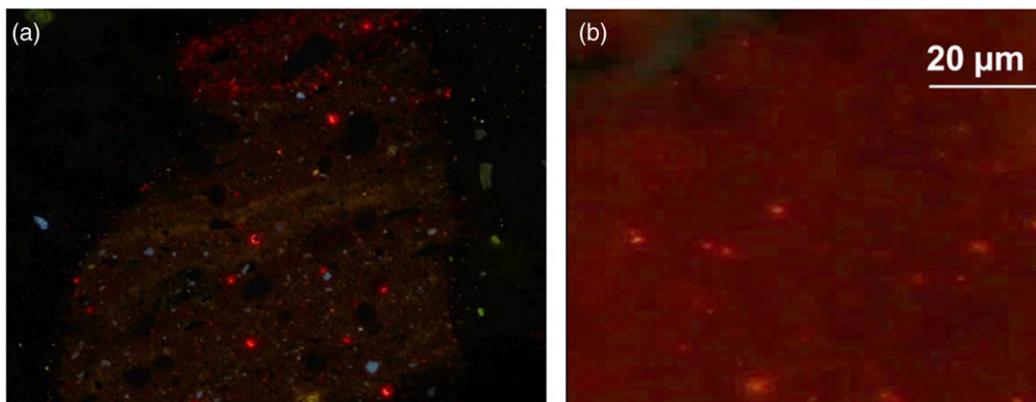


Figure 3 Optical images of electron-induced luminescence in geological carbonate particles. The technique can be efficiently used to detect residual fine carbonate grains in the binder (left). The right image is actually of a so-called “lime lump”, showing that they are not immune from geological carbonate contamination.

difficulty is to further purify the fine fraction, in order to remove the contaminants from the binder carbonate fraction. This is the most critical step, and an investigation is in progress by several research groups (Addis et al. 2016, 2019; Ponce-Antón et al. 2018). It is expected that there will not be a single solution to this problem, but an array of different methods adapted to specific mineral mixtures might work. As an example, thermal treatments seem to be very efficient for the removal of LDH phases from the carbonate mixture (Ricci et al. 2020 in this issue).

We may briefly mention cathodoluminescence spectroscopy (Machel 2000) as a powerful method to detect minute quantities of geologic carbonate in the sample or in the separated fraction (Figure 3). The technique is very well known in geology and sedimentology, but to date it has found limited application in the mortar dating field (Lindroos et al. 2007; Ortega et al. 2012; Murakami et al. 2013). In principle, the detailed understanding of the relationship between the crystal-chemical substitutions in the calcium carbonate lattice and the measured luminescence signal may be a useful and highly sensitive tool for the identification and detection of geologically derived carbonate (Toffolo et al. 2019).

Solid state nuclear magnetic resonance (SS-MAS-NMR) is also a powerful tool to detect minor traces of hydraulic reactions in the sample, for example in the presence of reactive silica-rich components (volcanics, pozzolans, cocciopesto). The NMR signal is element specific and highly significant with regards to the local environment of the probed cations, which in binder systems are normally  $^{27}\text{Al}$  and  $^{29}\text{Si}$  (Richardson et al. 2010). The technique therefore may greatly help in clarifying the role and presence of specific reaction products in the binder. As a matter of fact, NMR spectroscopy is especially powerful in detecting LDH phases in the separated binder fractions, when the amount of the contaminant is below the detection limit of XRPD. Thus, it may prove a key technique in determining the purity of the binder fraction that is used for  $^{14}\text{C}$  dating.

The fractionation of the stable isotopes of oxygen and carbon have been tentatively used to characterize anomalous carbonation processes of the lime binder (Pachiaudi et al. 1986; Van Strydonck et al. 1986, 1989, 1992; Ambers 1987; Dotsika et al. 2009). The isotopic fractionation sequence has been experimentally reproduced and interpreted (Kosednar-

Legenstein et al. 2008) so that there is a conceptual framework for interpreting the fractionation patterns. However, in many practical cases the measured fractionation values seem to result from the combination of several reaction processes, testifying to the complexity of the mortar systems.

#### **<sup>14</sup>C DATING OF ARCHAEOLOGICAL CARBONATE MATERIALS: DIFFICULTIES, NEW DIRECTIONS AND APPLICATIONS**

The methodological difficulties presented before related to the separation of the calcite fraction for dating even after extensive characterization do not hold us back from studying this material found so often in the archaeological and historical records. The necessity for a fine and detailed characterization indicates that the dating of plaster/mortar/cement cannot be regarded as a routine procedure. An alternative approach is to try to identify the pristine or altered by diagenesis status of the material. The origins of different calcitic fractions in archaeological mortar can be distinguished based on a Fourier transform infrared spectroscopy (FTIR) technique that characterizes the extent of molecular disorder in the calcite crystal (Gueta et al. 2007). This technique consists of repeated grinding of the material followed by FTIR analyses to separate the contributions of particle size and molecular disorder to the peak widths characteristic of calcium carbonate. Freshly made plaster/mortar has proved to be extremely disordered (Chu et al. 2008; Regev et al. 2010; Xu et al. 2015, 2016), and hence it can be expected that the most disordered fraction with a degree of disorder comparable to modern plaster, is composed mainly of the original plaster fraction. This FTIR analysis can therefore provide one way to identify the original anthropogenic component if it is preserved at all (Regev et al. 2010) and therefore separate this fraction for <sup>14</sup>C date. This approach was first applied to dating samples at the site of Yiftahel (Poduska et al. 2012), in Israel. Based on the FTIR parameter obtained on different parts of the sample, the disordered fraction was separated from the bulk material and pretreated for <sup>14</sup>C dating (Poduska et al. 2012). The results were promising, but there was still a discrepancy of a few hundred years between the binder dating of the best-preserved samples and the <sup>14</sup>C determination-based short-lived material from the same layer. Unfortunately, such a difference is not compatible with high precision chronology based on <sup>14</sup>C dating.

It seems that a better understanding of the mineralogical properties of freshly prepared lime/mortar is still needed, and in particular the diagenetic reactions that occur once buried. The working hypothesis when approaching such archaeological material is that the calcitic anthropogenic fraction continuously undergoes reactions and hence changes over time, like “a living organism”. Dissolution and reprecipitation as calcite might change the crystallinity order with the calcite becoming more stable. In this process exchange with a new carbon source might change the original <sup>14</sup>C concentration making its interpretation for dating impossible.

We would like to also propose that <sup>14</sup>C concentrations can be effectively used in order to monitor the post-burial diagenetic reactions that take place in an archaeological site. For example, if fractions are separated by density, and then characterized by the molecular disorder of the calcite using FTIR, will the <sup>14</sup>C concentrations of the fraction correlate linearly with decreasing molecular disorder? It would also be interesting to isolate the so called geogenic fraction of the binder, based on its morphological appearance in thin sections and/or on it being relatively well crystallized, and measure its <sup>14</sup>C concentration. If there is appreciable <sup>14</sup>C then clearly even the geogenic fraction is, in such environment,

reactive. To resolve these questions,  $^{14}\text{C}$  should be used as a tracer of processes, and its concentration expressed in pMC, not as age.

The fact that a better understanding of plaster mineralogy at its pristine formation is still needed can be seen in the recent observation that aragonite is formed in small amounts in freshly prepared plaster and can also be found in archaeological plasters and ash layers (Toffolo and Boaretto 2014; Toffolo et al. 2017). This aragonite is formed in the same pyrogenic process where calcite is found and therefore will have the atmospheric  $^{14}\text{C}$  concentration at the time of formation. Pyrogenic-formed aragonite could be a perfect material for  $^{14}\text{C}$  dating as pyrogenic calcite. The formation mechanisms need to be understood, especially in view of the fact that, unlike the calcite crystals of plaster, the aragonite crystals are large and adopt a well-defined acicular morphology (Toffolo et al. 2017). Note that inorganically formed aragonite crystals are usually acicular, but they are extremely small. The fact that the aragonite crystals are large (Toffolo et al. 2017) implies that they are relatively stable. This may be the reason why they are preserved in archaeological samples, even though aragonite is thermodynamically less stable than calcite (Lippmann 1973). The presence of aragonite opens up new opportunities for  $^{14}\text{C}$  dating mortar, as the aragonite crystals are formed during the plaster formation process, and if they are preserved, they are likely to be pristine. Toffolo et al. (2017) developed a method to separate aragonite crystals based on density and thermal decomposition and have shown that their  $^{14}\text{C}$  dates are consistent with the short-lived charcoal dates obtained from the same contexts. While this dating fraction depends on the presence of aragonite, it also raises a question about which conditions favor the formation and preservation of aragonite.

These are questions that are of interest for the conservation of carbonate material in the environment, for archaeological site preservation but also for modern structures.  $^{14}\text{C}$  has the sensitivity to address these questions and might open new possibilities for dating and for cultural heritage conservation.

#### **SINGLE GRAIN OSL DATING OF MORTARS IN ARCHAEOLOGY OF ARCHITECTURE: CURRENT STATE OF THE ART**

Optically stimulated luminescence (OSL) is a paleodosimetric dating method that has been applied to geology and prehistoric archaeology since the 1980s. It dates the last heating or the last exposure of minerals such as quartz or feldspar to light, which makes it an ideal candidate for dating the burial of sediment layers or dating the firing of inorganic artefacts. Furthermore, with the development of robust methodological concepts and the technological improvements of measurement systems at the beginning of the new millennia, OSL can also be used for the dating of mortars.

In the case of mortars, the OSL method is applied to the aggregate which makes it theoretically applicable not only to the dating of lime-based materials, but also to any other types of binding materials containing quartz or feldspars. The basic premise for dating is that the quartz in the sand used for making mortar was optically zeroed by light—“bleached”—during the process leading to mortar fabrication. Thus, the moment dated is the last exposure of the mortar to light, called bleaching. The OSL age of any dated material is then calculated as the ratio of the archaeological dose (or paleodose), accumulated by quartz minerals since the last light exposure and determined by the measurement of the corresponding luminescence, to the annual dose rate, usually assessed by determining the radiochemical composition of the

sample and of its nearest environment. Although different parameters may affect the accuracy and precision of OSL ages, the fundamental concern of the researchers dealing with the OSL dating of mortars is the degree of bleaching.

### **Pioneering Publications of OSL Mortar Dating**

At the time when the possibility to use quartz grains in mortar as a natural dosimeter in retrospective dosimetry had first been identified (Bøtter-Jensen et al. 2000), the usual practice of conventional OSL dating of sediments consisted of measuring the average luminescence signal, emitted by several hundreds or thousands of grains. While some early experiments on mortars with independent age control provided reasonable dating results even with this classical multigrain procedure (Zacharias et al. 2002; Goedicke 2003), others failed to reach the expected dates (Goedicke 2003, 2011). The reasons for this were quickly understood and highlighted. A conventional multigrain approach can only be successful if all the quartz grains in mortar are properly reset by light during the manipulation of the sand aggregate when making mortar. In reality, many historical mortars contain grains with varying degrees of optical bleaching, and so emit luminescent signals of different intensities (Jain et al. 2004). For such cases, the authors of the first publications on OSL mortar dating suggested that “large sets of single—or ideally single-grain—aliquots have to be measured to exclude insufficiently bleached signal components from the De estimation” (Zacharias et al. 2002).

However, if we consider a signal from only one “single grain” of quartz extracted from mortar, the luminescence emitted is generally several orders of magnitude weaker than conventional luminescence readers were able to detect. In that time, the efficient measurement systems to detect luminescence signals from individual grains were under development (Duller et al. 1999; Bøtter-Jensen et al. 2000).

### **Dating of Fine-Grained and Coarse-Grained Quartz Fractions with the Multigrain Procedure**

In 2010, Gueli et al. (2010) proposed an alternative to overcome the problem of low luminescence signal intensity arising from single grains. Instead of individual sand sized grains, the authors attempted to date a fine-grained quartz fraction (4–11  $\mu\text{m}$ ) using the classical multigrain procedure. Since then, it has been tested on a total of 15 mortar samples originating from five different monuments in southern Europe, namely Italy (Stella et al. 2013), France (Stella et al. 2013) and Portugal (Stella et al. 2018). The majority of the dating results is in agreement with TL dating of the adjacent bricks, which was systematically applied for a cross-check of the chronological data. Such concordant results are indicative of a good bleaching degree of the mortars studied. This argument is also supported by the Gaussian forms of equivalent dose distributions obtained for these samples.

However, when dating a fine grain fraction, thousands of grains are deposited on one disc and measured at the same time. Consequently, only average luminescence signals are detected and the true dispersion existing between individual grains is much less observable. Therefore, one cannot be completely sure about the bleaching degree when dating mortars in such a manner. The authors conclude that “the methodology is then only indicative but not exhaustive to evaluate the bleaching degree of fine grains” (Stella et al. 2018). Hence, due to this small risk, Stella et al. (2018) emphasize that in case of using the fine grain fraction to date mortars, “the association between dates of bricks and dates of mortars coupled with

historical and architectural survey represents a useful approach to obtain indications about the accuracy of the dating results.”

The surprisingly good bleaching degree of fine grain fraction extracted from the Mediterranean mortars by Stella et al. (2013, 2018) does not match with the conclusions of Goedicke (2003) or Jain et al. (2004). According to Goedicke, “larger, heavier grains are more likely to be bleached, since they tend to move to the surface when mixed together with the smaller ones and hereby more exposed to light” (Goedicke 2003). Apart from these considerations based on a physical model of mortar mixtures, Goedicke performed several comparative tests by measuring equivalent doses from different granulometric fractions of the same mortar samples. He obtained much more asymmetric distributions for coarser quartz and concluded, as did Jain et al. (2004), that coarser quartz grains analyzed in their study appear to be better-bleached.

It is, however, necessary to underline that Goedicke (2003) and Jain et al. (2004) did not study the quartz from the same geographical area as Stella et al. (2013, 2018). As suggested by Urbanová (2019) and discussed further in this paper, the mortars from the coastal areas of southern Europe currently seem to have more favorable characteristics for luminescence dating, which may be linked to the origin, sedimentary history and processing of the sand aggregates used to prepare mortar. It is thus possible that the mortars studied by Gueli et al. (2010) and Stella et al. (2013, 2018) were globally well-bleached and also that a coarser fraction of these samples would provide satisfactory results with the multigrain procedure. On the other hand, the methodology might prove to be problematic for heterogeneously bleached mortars. This is one of the aspects to be verified during the inter-laboratory mortar dating inter-comparison, carried out within the framework of the MODIS 2 study in which all participating research laboratories work on the same mortar samples.

Goedicke considered one major advantage of using coarse quartz for dating. In fact, the probability of identifying the well bleached grains in multigrain discs can be easily increased by reducing their number on a disc (e.g. Jain et al. 2004). Despite this improvement, consisting in the reduction of the number of grains per disc, only 7 out of 14 mortars in a subsequent study (Goedicke 2011) could have been reliably dated by the conventional multigrain procedure. The authors conclude that if very low levels of bleaching are encountered, the only way to find the archaeological dose is by single grain analysis (Jain et al. 2004; Feathers et al. 2008; Goedicke 2011).

### Switchover to “Single Grain” Analyses

In the first single grain studies (e.g. by Lamothe et al. 1994; Murray et al. 1997; Roberts et al. 1998; Olley et al. 1999), the authors were using conventional diodes for the optical stimulation. Progressively, the instrumentation specific for the “single grain” analyses has been introduced (Duller et al. 1999, 2000; Bøtter-Jensen et al. 2000) which allowed much faster stimulation and detection of separate luminescence signals from individual grains. A laser beam (Nd:YVO<sub>4</sub>), emitting at 532 nm and focused on single grains, has been implemented in luminescence readers as a source of optical excitation. The laser beam “causes the OSL signal to decay 100 times faster (within 0.4 s) than in conventional OSL” (Goedicke 2011) which allowed the efficient measurement of the luminescence signals emitted by individual grains and so opened new perspectives to the OSL dating of mortars.

Shortly after its development, the instrumentation for “single grain” OSL dating (SG-OSL) started to enter research practice, in particular for dating sand-sized, heterogeneously bleached sediments (Olley et al. 2004; Jacobs et al. 2013; Sim et al. 2013; Medialdea et al. 2014). In this context, important methodological research has been undertaken to understand the scattering in equivalent dose distributions arising from the “single grains” (Thomsen et al. 2003; Jacobs et al. 2006) and new ways of estimating archaeological dose in heterogeneously bleached quartz have been suggested since then (Thomsen et al. 2005, 2007; Guibert et al. 2017).

The first “single grain” tests performed on construction mortars were linked to the methodological research on accidental dosimetry (Jain et al. 2002, 2004) and applied in particular to modern materials. Some studies (Jain et al. 2004; Arnold et al. 2012) compared the distributions obtained from small “multigrain” aliquots and “single grains” of quartz extracted from modern binders which were stated to be dim and heterogeneously bleached. Small aliquot results were over-estimated when compared to the single-grains. Goedicke (2011) concluded that “if very low levels of bleaching are encountered, the only way to find the archaeological dose is by single grain analysis”.

#### *“Single Grain” OSL Dating and Data Treatment*

In SG-OSL dating, the procedure leading to the determination of the archaeological dose involves several stages:

- Extraction of the mineral (quartz or feldspar) grains from the sample (which must not be exposed to light);
- Verification of the purity of the mineral extracted (e.g. Bøtter-Jensen et al. 2003);
- Measurement of the natural luminescence signal emitted by individual quartz grains as a result of the optical stimulation (Wintle and Murray 2006);
- Construction of the dose-response curve and conversion of the natural luminescence signal from each quartz grain to the respective radiation dose received by this same grain (Wintle and Murray 2006);
- Statistical treatment of the distribution of equivalent doses arising from the luminescence of individual grains which aims to identify the well-bleached grains, subsequently used for the calculation of the final archaeological dose (paleodose) and so the age.

The first four steps follow standard methodologies that are unified within the research luminescence community. In particular, the acquisition of the natural luminescence signals from individual grains are currently conditioned by using the same “single grain” measurement system of the DTU Risø laboratory and the same single-aliquot regeneration (SAR) measurement protocol is generally employed (Murray and Roberts 1998). On the other hand, differences may appear in the data treatment methodologies, leading to the final determination of the archeological dose and so the age.

Several so-called “age” models to determine the final archeological dose (paleodose) have been developed in the past for both well-bleached (CAM: central Age Model by Galbraith et al. 1999; BaSar model by Guérin et al. 2015) and heterogeneously bleached samples (MAM: Minimum Age Model by Galbraith et al. 1999; IEU: Internal-External consistency criterion by Thomsen et al. 2005, EED: Exponential Exposure Model by Guibert et al. 2017; Christophe et al. 2018; Guibert et al. 2020 in this issue). The selection of the most

convenient model to calculate the archaeological dose (paleodose) is a crucial step in the dating procedure since it directly affects the accuracy of the final age. In this respect, some pieces of information are critical to assess the reliability of the SG-OSL or the OSL age of mortar and should always be provided in the articles dealing with the luminescence dating of mortars: the discussion on the sources of scattering in dose distributions (intrinsic variability between grains, extrinsic sources of the dispersion: bleaching degree, microdosimetric heterogeneity), and the justification of the statistical model used for the calculation of the final archaeological dose. The reader can find a more detailed discussion on this subject with respect to mortars in Urbanová and Guibert (2017) or in more fundamental studies, e.g. by Thomsen et al. (2005) or Guérin et al. (2017).

#### *“Single Grain” OSL Dating of Historical Monuments*

After the first series of exploratory papers applying mainly a classical “multigrain” procedure and discussed at the beginning of this paper, larger “single grain” OSL dating studies were performed on mud mortars (Feathers et al. 2008; 16 prehistoric samples), on lime mortars (Urbanová and Guibert 2017; 33 samples with the known historical age) and on earthen mortars (Panzeri et al. 2017; 10 samples with the known historical age).

Feathers et al. (2008) presented the first larger “single grain” OSL dating study on mortars which was integrated in an archaeological research project on the prehistoric site Chavín De Huántar in Peru. It is the only paper on this topic that studies materials outside Europe. Feathers et al. (2008) deal with a particularly difficult coarse grained, un-processed material, which is highly affected by both extrinsic sources of dispersion in the data: beta heterogeneity and heterogeneous bleaching. The study indicates the potential of the “single grain” procedure for mortar dating and pinpoints all the problematic points that will be explored afterwards.

Nine years later, Urbanová and Guibert (2017) came up with the methodological study on 33 lime mortars from 3 Roman, 2 early medieval and 2 medieval monuments from different parts of France and Switzerland, aiming in this way to diversify the types of quartz tested and compare its behavior. Similar to the method in Feathers et al. (2008), all samples within the study were dated exclusively with the “single grain” procedure combined with the qualitative assessment of beta dose rate variability through beta imaging and K-content cartography. In conclusion, the authors provide the discussion on the specificities of sampling, preparation and parameters in OSL dating of mortars, which are in some aspects different from OSL dating of sediments. Based on the findings, the authors divide the studied mortars from different monuments into several categories according to their bleaching degree and beta dose rate variability. Conclusive results are achieved for all mortars showing rather good degrees of bleaching, and also for poorly bleached mortars which are not significantly affected by the beta dose rate heterogeneity. Insufficiently bleached mortars which are largely affected by microdosimetric variations (in particular *cocciopesto* mortars and coarse-grained materials with inhomogeneous distribution of K-feldspars in the matrix) are stated to be difficult to date.

Finally, Panzeri et al. (2017) reports the SG-OSL dating of 10 earthen mortars from two 15th and 18th century monuments in the Cremona, north-central Italy. The IEU and the unlogged-MAM model were stated as the most appropriate for the calculation of the archaeological dose for the given set of samples. The over-dispersion value obtained from the dose recovery measurements (DRT, e.g. Thomsen et al. 2012) was used as an input for the estimation of

the archaeological dose. The discussion on the microdosimetric heterogeneity of the studied samples which might potentially have some effect on the choice of the input is not provided. The authors point out the unfavorable characteristics of the quartz studied (low sensitivity and very low bleaching degrees), implying an age precision which is considered as “relatively low” by the authors.

The common range of absolute precision obtained by luminescence dating techniques is usually between 5–10% for one individual dating result. In the case of mortars with less favorable characteristics for dating (low sensitivity, poor bleaching, beta dose rate heterogeneity), the error may be higher. The multiplication of the samples and dating of several stratigraphic levels are the main strategies to reduce the statistical uncertainties.

The level of contribution that the resulting date brings to the historical knowledge of the monument depends on the historical and archaeological context. For “younger” monuments, which may be usually dated by more precise approaches (from the 13th century onwards, e.g. Panzeri et al. 2017), the precision of the methodology may be judged as “low” or insufficient in some cases. On the contrary, in older periods such as the prehistoric era or the Early Middle Ages (4th–11th century AD) for which we often lack reliable written and archaeological sources, it was clearly demonstrated that the SG-OSL dating of mortars can enrich the historical knowledge of the monument and provide historically relevant dates (e.g. Feathers et al. 2008; Čaušević-Bully et al. 2018; Urbanová et al. 2018; Javel et al. 2019; Panzeri et al. 2019), thereby opening new perspectives for dating of buildings in archaeology. In highly favorable environmental contexts (regions with high environmental dose rates and providing quartz with high sensitivity to the optical stimulation), OSL dating with the classical multigrain procedure also seems to provide reasonable historic dates (Sanjez-Pardo et al. 2017).

#### *“Single Grain” OSL Dating Potential of Mortar and Provenance of Quartz?*

Based on the SG-OSL studies of European historical mortars published up to now, we have an insight on four late medieval sites from central-north Italy (Panzeri 2013; Panzeri et al. 2017, 2019), three medieval sites from Switzerland (Urbanová and Guibert 2017) and many examples of early medieval architecture from southern France (Urbanová and Guibert 2017; Urbanová et al. 2018; Javel et al. 2019), northern Croatia (Čausević-Bully et al. 2018), northern Italy (Panzeri et al. 2017; Urbanová 2019) and southern Spain (Urbanová 2019). Apart from a study of one pre-Columbian site from Peru (Feathers et al. 2008), other research on the SG-OSL dating of mortars outside Europe is lacking.

Figure 5a illustrates the geographical distribution of the European sites, whose mortars were studied up to now with the “single grain” procedure, with respect to the degree of bleaching. To visualize the general tendency of the bleaching degree for individual European sites, the samples were classified into categories:

- a) Yellow circles: mortars that may be considered well-bleached, i.e. all accepted luminescent grains were taken into account for the calculation of the archeological dose;
- b) Gray circles: mortars that may be considered as partially bleached, i.e. more than 30% of accepted luminescent grains were considered well-bleached and taken into account for the calculation of the archeological dose (Figure 4f–h);

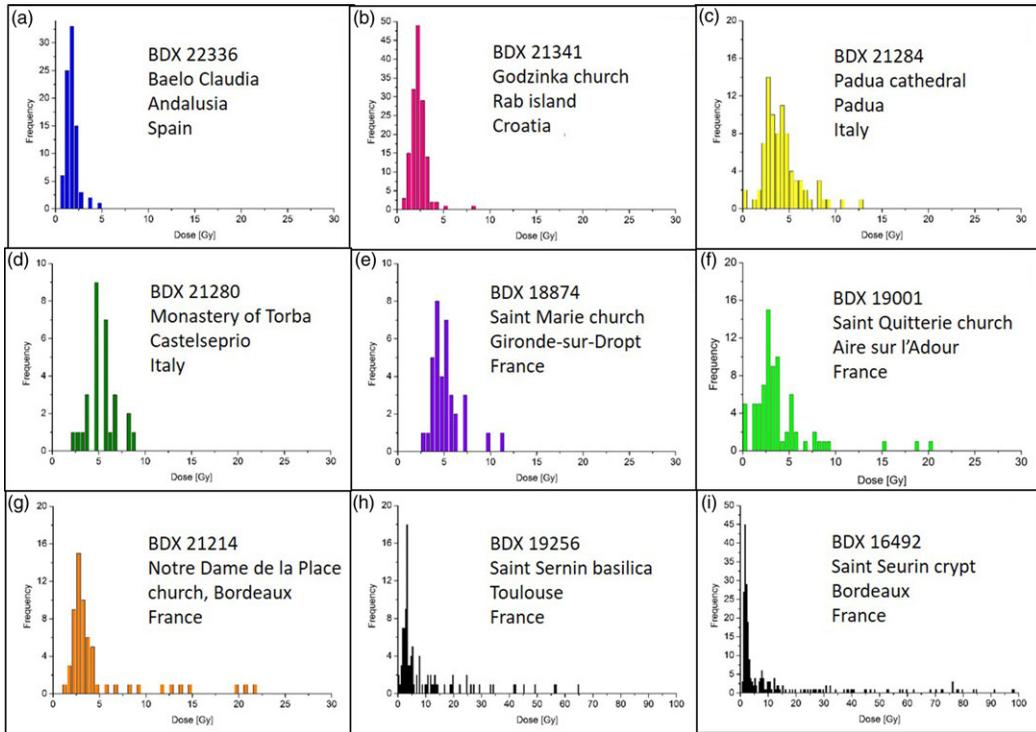


Figure 4 Some examples of the equivalent dose distributions obtained with the “single grain” OSL procedure for mortars originating from different parts of Europe.

- c) Black circles: mortars that may be considered poorly bleached, i.e. less than 30% of accepted luminescent grains were considered well-bleached and taken into account for the calculation of the archeological dose (Figure 4i);
- d) White circles: mortars that did not emit any luminescence signal after optical stimulation.

In addition, Figure 5b shows the distribution of these sites, but with regard to the sensitivity of the quartz to the SG-OSL stimulation (i.e. the percentage of the grains emitting an exploitable luminescence signal). Also, in these cases the categories were defined as follows:

- a) Yellow circles: mortars showing high sensitivity, i.e. mortars for which more than 5% of all analyzed quartz grains emit the luminescence signal suitable for SG-OSL dating (i.e. the signal meeting the acceptance criteria as defined in Urbanová and Guibert 2017)
- b) Gray circles: mortars showing standard sensitivity, i.e. mortars for which between 2 and 5% of all analyzed quartz grains emit the luminescence signal suitable for SG-OSL dating
- c) Black circles: mortars showing low sensitivity, i.e. mortars for which less than 2% of all analyzed quartz grains emit the luminescence signal suitable for SG-OSL dating
- d) White circles: mortars that did not emit any luminescence signal after optical stimulation.

We must add, however, that for certain monuments, quartz sands present in mortars sampled in different construction phases of the same site may show some differences in terms of SG-OSL dating potential (e.g. n. 5, 6, 8, 15).

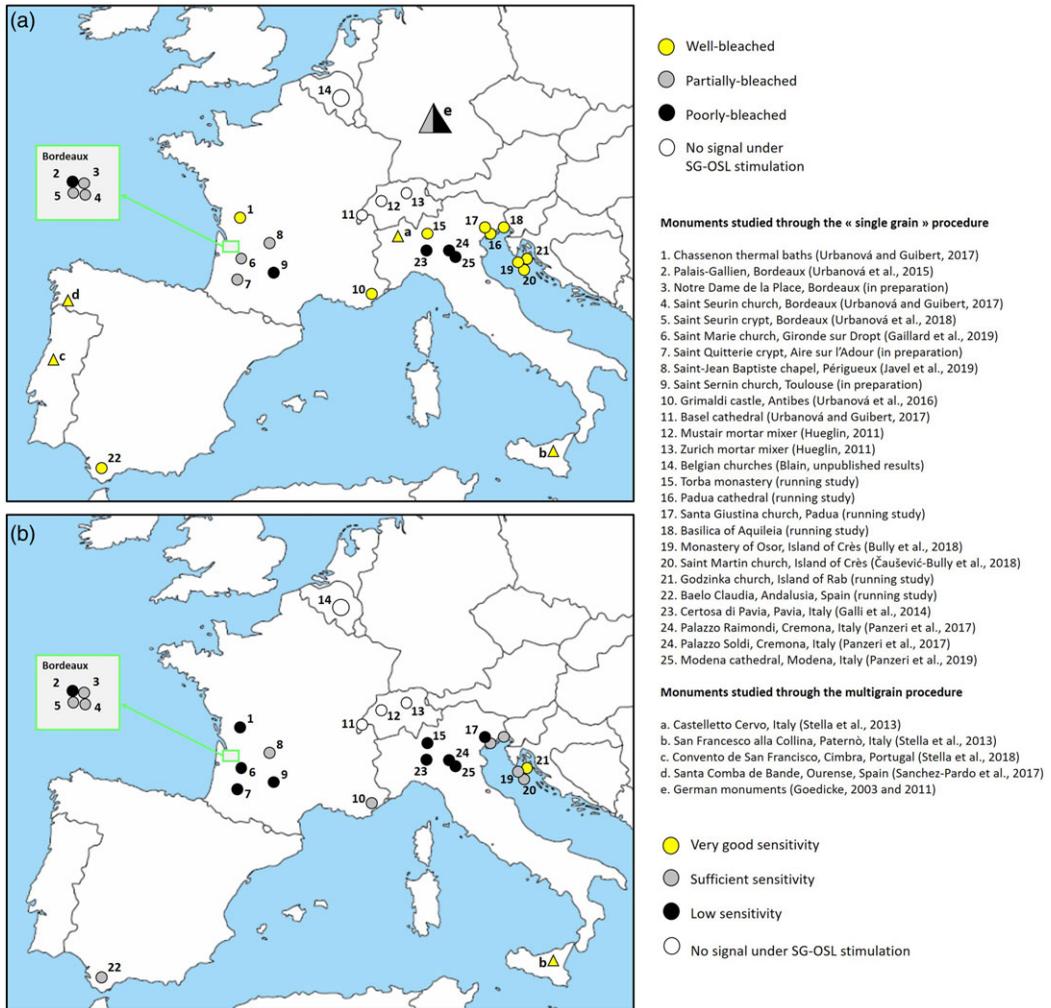


Figure 5 Geographical distribution of the European sites whose mortars were studied up to now through the “single grain” OSL procedure with respect to the degree of bleaching (a) and to sensitivity of quartz to the SG-OSL stimulation (b). For certain monuments, quartz sands present in mortars sampled in different construction phases of the same site may show differences in terms of SG-OSL dating potential (e.g. n. 5, 6, 8, 15), which are closely linked to the variations in mortar composition and preparation technology.

In general, it is logically supposed, and it was also many times demonstrated, that the charge used to prepare mortars comes from local materials with primary sources located no further than several kilometers from the site. Based on the information that can be read in the Figure 5, we can hypothesize that the quartz from the monuments in southern Europe located close to or on the Mediterranean coast tend to show sufficient or very good sensitivity overall to SG-OSL stimulation and is often well-bleached. We can presume the use of coastal sands for the majority of these mortars. The quartz originating from the historical buildings located closer to large mountain formations (Alpes, Pyrenées, French *Massif Central*) show very low or no sensitivity to SG-OSL and variable bleaching degrees. The aggregates used for

mortar making in these cases may originate from the sediments with shorter sedimentary histories (closer to river sources etc.), as the geographical position of these sites suggest.

It is known and has been shown by many laboratory studies that the sensitivity of quartz minerals rises after its exposure to repeated irradiation, illumination and heating (e.g. Murray and Roberts 1998; Wintle and Murray 2006). The research carried out by Pietch et al. (2008) and Sawakuchi et al. (2011) on natural samples with the “single grain” procedure have equally demonstrated that, in natural conditions, the sensitivity of quartz grains as well as the proportion of well-bleached grains increase significantly with the repeated irradiation, illumination and heating to which the sediments are exposed in nature. In the research presented by Sawakuchi et al. (2011), fluvial sands with short sedimentary histories (located a short distance from their primary rock sources) had overall quite low SG-OSL sensitivity. Coastal sediments with longer sedimentary histories generally demonstrated higher sensitivity and also higher variability between individual grains. Even if more detailed research needs to be undertaken into these phenomena, current general observations on mortar’s behavior seem to match the findings by Sawakuchi et al. (2011).

The findings can be complemented by the results from the “multigrain” studies of the well-dated monuments marked by triangles in Figure 5a. If we assume that obtaining a Gaussian distribution with the “multigrain procedure” indicates a good degree of bleaching, the monuments from Southern Europe (yellow triangles, Stella et al. 2013, 2018; Sanchez-Pardo et al. 2017) mainly enter this category. On the other hand, strongly asymmetric distributions obtained with the “multigrain procedure” imply partial or poor bleaching. This is the case for the majority of the sites studied in Central Europe by Goedicke (2003, 2011). Furthermore, it is necessary to mention that the mortars from three different medieval sites located in Switzerland (Urbanová and Guibert 2017) provide examples of quartz with no detectable SG-OSL signal after optical stimulation. Consequently, this quartz cannot be currently dated with SG-OSL.

In case of poorly sensitive quartz, one might consider the use of feldspar for mortar dating. The feldspars could provide better luminescence signals and are also less sensitive to the microdosimetric variations due to their internal content of radioelements. On the other hand, if we consider a very low age of mortar as a construction material and very short exposure of mineral grains to light during the dynamic process of mortar making, the slower bleaching rate and anomalous fading of the signal characteristic for feldspars might potentially represent obstacles for this application which, however, has never been really tested on mortars yet.

Although many more examples need to be studied to provide an overall view on the SG-OSL dating potential of European mortars, the research on this topic has underwent significant progress in recent two decades. The current findings indicate a very good dating potential for historical mortars in certain regions and in particular in the Mediterranean area while the few examples that we have from Switzerland, Germany and Belgium show less favorable characteristics for dating. Systematic research of a closer link between quartz sensitivity, its geological history, sand processing and the bleaching degree of mortar seems to be a promising clue for the future to better understand the differences in behavior between mortars of different origins and of varying mineralogical compositions.

### *“Single Grain” versus “Multigrain” in OSL Dating of Mortars*

Research on mortar dating through optically stimulated luminescence has considerably advanced in recent years. One of the latest improvements consists in the possibility to detect the luminescence signals from each quartz grain extracted from the mortar individually, thanks to the automatization and democratization of the “single grain” measurement systems. Although the use of this technique is widespread in sediment dating, its use on mortars is still relatively scarce.

Various undesirable phenomena may affect the SG-OSL and OSL mortar dating procedures as summed up recently by Sanjurgo-Sanchez et al. ([in press](#)). Nevertheless, there are two important factors that imply if the dating process will be successful or not: degree of bleaching and the sensitivity of quartz to SG-OSL stimulation (i.e. the percentage of the grains emitting an exploitable luminescence signal).

Parallel to the studies that use exclusively the “single grain” measurement systems, conventional multigrain analyses still continues to be used for mortar dating (e.g. Sanchez-Pardo et al. [2017](#); Moroupolou et al. [2018](#); Stella et al. [2018](#); Panzeri et al. [2019](#)). Some of the recent studies deal in different ways with this situation. Sanchez-Pardo et al. ([2017](#)), Stella et al. ([2018](#)) or Panzeri et al. ([2019](#)) show equivalent dose distributions which indicate a Gaussian form, and the resulting OSL dates are then based on the weighted average of several tens of individual multigrain aliquots (from 30 up to 90). On the other hand, Moropoulou et al. ([2018](#)) provides the final archeological doses based on the weighted average of only 6–8 aliquots, without providing any information about the scattering of the equivalent dose distributions and so about the bleaching degree of the studied mortars. From the given data, it is hereby not possible for the reader to evaluate the bleaching degree of the samples presented.

In the cases when a bleaching degree of studied mortars cannot be assessed with the “single grain” procedure, analyses of several tens of individual aliquots (i.e. aliquots with only few grains) is highly recommended in order to obtain a statistical representativeness from which it is possible to deduce the dispersion in the data. Nevertheless, the advice is relevant if the OSL dating of mortar with the “multigrain” procedure can be combined with other dating methods to cross-check the chronological data. If the goal is to establish an independent dating approach for the OSL dating of mortar that could be used for dating of monuments with unknown chronology, the “single grain” procedure is the most secure way of evaluating with absolute certainty the bleaching degree.

### **CONCLUDING NOTES**

The research on  $^{14}\text{C}$  and OSL dating of mortars has made considerable progress in recent years. As the two methods are applied to different components of mortar, the lime binder for  $^{14}\text{C}$  and the aggregate for OSL, they might well be complementary and their application mostly depends on the nature of the mortar material.

Concerning practical aspects, the quantity of the sample needed for OSL dating depends on the quantity of the specific granulometric fractions of quartz. In general, about 100 g of the bulk mortar not exposed to light generally allows for the extraction of a sufficient amount of quartz for dating purposes.

OSL dating is applied on quartz aggregate extracted from mortar which is theoretically an inert component of the mortar system. The sample preparation for luminescence dating thus consists in the extraction of the quartz grains from the material which is simple in principle. The purity of the extract can be easily checked.

In the case of OSL dating, two premises are required for successful dating: the quartz has to emit an exploitable luminescence signal and at least a part of the luminescent grains has to show a sufficient bleaching degree. Both characteristics are specific to the material and cannot be predicted without proper OSL analyses. If the grains show variable degrees of bleaching, which is generally the case, the grains with different luminescent signals can be distinguished thanks to the use of the “single grain” procedure (analyses of grains one by one). If the OSL is supposed to be used as an independent dating technique, the “single grain” procedure is the most secure way of evaluating with absolute certainty the bleaching degree and thus establishing a reliable dating result.

In the case of mortar  $^{14}\text{C}$  dating, the characterization of the material, composition, pristine quality and preservation of the carbonate fraction in the mortar is fundamental for qualifying the material for  $^{14}\text{C}$  dating. Therefore, while for dating only less than a mg of carbon is necessary, the characterization of the material and the study of the different fractions from the material might require a substantial sample to start. The amount needed can be roughly estimated in a few grams of material at least, to allow for characterization, separation, and dating. As the nature of the anthropogenic carbonate is very variable due to the original preparation and diagenesis through time, a “standard” chemical preparation for dating is, at present, not yet possible. Tailoring the pretreatment for the separation of the pristine fraction is therefore one of the most important steps for successful  $^{14}\text{C}$  dating.

Dating materials in archaeology are of course fundamental for building a chronology. Anthropogenic carbonate has been of great interest for this reason and it has been studied extensively as far as  $^{14}\text{C}$  dating is concerned. Yet, dating cannot yet be easily obtained for these complex materials. In any case the presence of  $^{14}\text{C}$  in mortar is always related to a chemical or physical process which, beyond the date of the material, is there to be deciphered and it is of great help in understanding the history and the interaction of the mortar with the environment through time.

Both  $^{14}\text{C}$  and OSL dating of mortars are not yet routine applications, mostly because of the complexity of the procedures. However, many recent studies demonstrate the relevance of mortar dating for the chronological study of historical buildings and show that they can provide historically relevant dates as a useful aid to archaeology and architectural history.

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