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ORIGINAL PAPER

Mortars, plasters and pigments—research questions and sampling criteria

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Abstract
Within the Topical Collection, this paper represents an introductory contribution aimed at describing and discussing the research questions and the sampling criteria in the field of mortars, plasters and pigments studies. The paper is divided into three parts. In the first part, some terminological issues are clarified and the building archaeology is introduced as an indispensable method for sampling and interpreting archaeometric results. In the second part, the most common research questions are presented and discussed. Some case studies are also reported to clarify what the expected results may be. The sampling problem is faced in the third part, where the criteria for a representative, functional and suitable selection are provided.

Keywords Mortars and plasters · Pigment analysis · Building archaeology · Archaeometry and archaeology · Research questions · Sampling criteria

Premise

This Topical Collection (TC) covers several topics in the field of study, in which ancient architecture, art history, archaeology and material analyses intersect. The chosen perspective is that of a multidisciplinary scenario, capable of combining, integrating and solving the research issues raised by the study of mortars, plasters and pigments.

The first group of contributions explains how mortars have been made and used through the ages (Arizzi and Cultrone 2021, Ergenç et al. 2021; Lancaster 2021; Vitti 2021). An insight into their production, transport and on-site organisation is further provided by DeLaine (2021). Furthermore, several issues concerning the degradation and conservation of mortars and plasters are addressed from practical and technical standpoints (La Russa and Ruffolo 2021; Caroselli et al. 2021).

The second group of contributions is focused on pigments, starting from a philological essay on terminology (Becker 2021). Three archaeological reviews on prehistoric (Domingo Sanz and Chieli 2021), Roman (Salvadori and Sbrolli 2021) and Medieval (Murat 2021) wall paintings clarify the archaeological and historical/cultural framework. A series of archaeometric reviews illustrate the state of the art of the studies carried out on Fe-based red, yellow and brown ochres (Mastrotheodoros et al. forthcoming); Cu-based greens and blues (Švarcová et al. 2021); As-based yellows and reds (Gliozzo and Burgio 2021); Pb-based whites, reds, yellows and oranges (Gliozzo and Ionescu 2021); Hg-based red and white (Gliozzo 2021); and organic pigments (Aceto 2021). An overview of the use of inks, pigments and dyes in manuscripts, their scientific examination and analysis protocol (Burgio 2021) as well as an overview of glass-based pigments (Cavallo and Riccardi forthcoming) is also presented. Furthermore, two papers on cosmetic (Pérez-Arantegui 2021) and bioactive (antibacterial) pigments (Knapp et al. 2021) provide insights into the variety and different uses of these materials.

This article is part of the Topical Collection on *Mortars, plasters and pigments: Research questions and answers*

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Table 1 Definitions and uses of pigment, ink, dye and de/colouring agents to be used in archaeometry

	Pigment (Ink)	Dye*	De/colouring agent
Definition	Substance which, finely dispersed in water or other solvent, colours the substrate by overlapping	Fine substance which penetrates the substrate to which it is being applied	Substance which chemically bonds the material to which is added
Characteristics	- Chiefly inorganic - Fine/coarse grained - Insoluble - Dispersed suspension that covers the substrate	- Chiefly organic - Fine grained - Soluble - Solution that is absorbed by the substrate	- Only inorganic - Different types of compounds and grain sizes -Insoluble -Incorporated by the object
Few examples	Ochres (Mastrotheodoros et al. forthc.) Cu-based (Švarcová et al. 2021) Hg-based (Gliozzo 2021) As-based (Gliozzo and Burgio 2021) Pb-based (Gliozzo and Ionescu 2021) Smalt (Cavallo & Riccardi forthc.)	Carmine; Gamboge Indigo; Lac**; Madder; *Purpurissimum* Sepia	Metallic Cu Oxydes and hydroxides containing Cu^{2+} Fe^{2+}, Fe^{3+} Mn^{4+}, Co^{2+} Bronze Bindheimite
Use	Painting, writing	Textiles and leathers (painting, writing***)	Glass

* For a review on organic colouring materials used in wall painting, see Aceto (2021) in this TC

**The term lac may refer to two very different types of inorganic compounds: a) the gommalacca or shellac, i.e. "a pink-red–purple organic colourant derived from an insect and used as a lake pigment or a dye" (Berbers et al. 2019); b) a mixture of nitrocellulose and resin/wax

*** Limited to specific chronologies and artefacts

Introduction

This paper serves as an anchor for the numerous contributions of this TC. It intends to provide the criteria for both the formulation of sound archaeometric questions and the execution of a suitable sampling in the field of mortars, plasters and pigments studies.

To frame these issues in a methodologically clear and exhaustive panorama, two brief explanatory sections are provided at the beginning: (1) what is meant by mortar, plaster and pigment and (2) what is meant by building archaeology.

Both these introductory texts function to explain which archaeometric questions are valuable for historical reconstruction and to guide researchers in the direction of representative sampling. These last two issues represent the bulk of the present contribution and are both presented in light of the most recent advances made in this field. The interpretative problems that frequently arise from the archaeometric study of mortars are discussed in the last section of this paper, exploring the value and potentiality of archaeometric research for historically meaningful reconstructions.

Mortars and pigments: basic definitions

The term mortar basically defines a mixture of different components used to bond bricks or stones (Table 1). Terminological issues may arise when trying to distinguish a mortar from concrete and cement; therefore, the definitions are provided here, based on the International Standards Organization (ISO 6707–1:2020[1]):

(a) **Mortar** is described as a mixture of binder, aggregate and water;
(b) **Concrete** is a mixture of aggregate, cement and water;
(c) **Cement** is an inorganic binder mixed with water to form a paste "that sets by means of hydration reactions and processes, and that, after hardening, retains its strength and stability".

Furthermore, in the context of Roman architecture, concrete is defined as the mixture of lime, aggregates and water to form the so-called *opus-caementicium* (see Vitti 2021 in this TC).

In this context, mortar is a recipe where the components are the variables and its end-use is the archaeometric study's object. This basic distinction is fundamental in organising the research questions and provides a key tool for their interpretation. Consequently, correct identification and characterisation of the variables represent the starting point for any technological and provenance issue. Moreover, while the recipe's reconstruction mostly regards the technological issue, the end-use of a mortar provides the necessary link to establish its suitability and functionality. Basically, there

[1] https://www.iso.org/obp/ui/#iso:std:iso:6707:-1:ed-6:v1:en:term:3.4.4.27

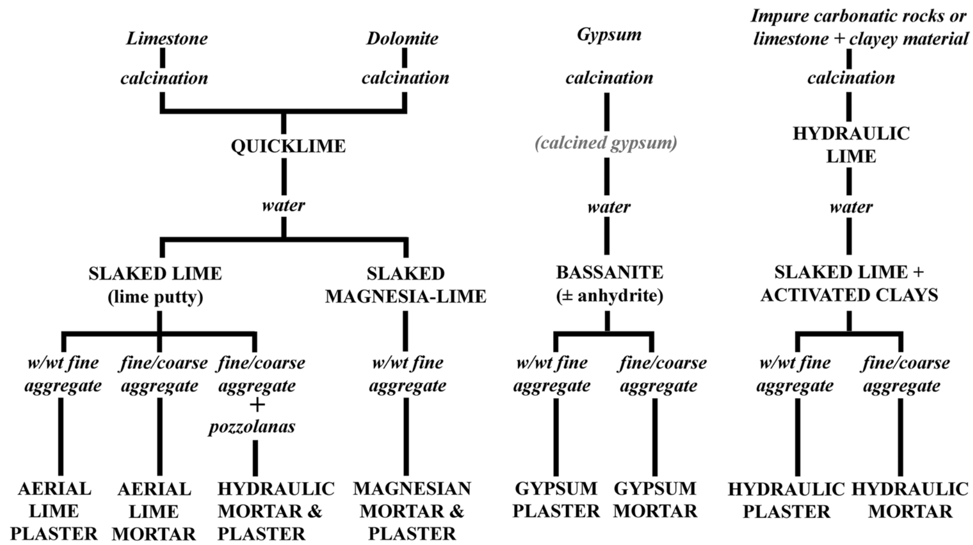


Fig. 1 Main types of plasters and mortars, depending on the starting raw material

are two types of mortars: the aerial (Ergenç et al. 2021 in this TC) and the hydraulic ones (Arizzi and Cultrone 2021 in this TC).

Aerial mortars are characterised by higher flexibility, plasticity and permeability than the hydraulic ones; however, the former shows a greater shrinkage during hardening, lower mechanical strength and resistance to moisture, salt attack and frost compared with the latter. Based on these main properties, aerial mortars are mainly used in sheltered areas, while hydraulic ones are favoured in exposed buildings, such as port infrastructures (Vicat 1837; Ashurst and Ashurst 1988; Gibbons et al. 1995; Holmes and Wingate 1997; Cowper 1998).

The term plaster includes several types of mixtures, depending on the type of binder and aggregate used. Technical terminology (ISO 6707–1:2020[2]) distinguishes between mixtures of one or more binders, defined as plasters, and mixtures of one or more binders with aggregate (and other possible admixtures), defined as renders. Moreover, the terms plaster and render should apply to mixtures used for the internal and external finish, respectively.

In archaeometry, plaster is used as a more general term, including the meaning and properties of both modern plasters and renders. This should perhaps remain so to maintain terminological coherence with the history of studies and highlight the distinction between ancient materials and technical materials used, for example, for restoration and conservation. On the other hand, the modern classification of mortar and cement (*e.g.* UNI EN 197/1) foresee subdivisions that do not apply to the ancient world's materials and could not be extensively used tout court.

Typical examples in ancient buildings include:

- Clay plasters, mixing clay, sand and plant fibres;
- Aerial lime mortars and plasters, mixingslaked lime and aggregate (*e.g.* sand or a mixture of sand and other inert);
- (Feebly hydraulic[3]) magnesian mortars and plasters, mixing slaked magnesia-lime and aggregate;
- Gypsum mortars and plasters, consisting of calcium sulfate hemihydrates and sand;
- Hydraulic pozzolanic mortars and plasters, mixing hydrated lime with natural or artificial pozzolanas (*e.g.* fly ashes and/or powdered ceramics);
- Hydraulic mortars and plasters, mixing a natural hydraulic lime and aggregate.

A schematic distinction between the various types of plasters and mortars is provided in Fig. 1, while the main reactions addressed henceforth are listed in Fig. 2.

The use of the term pigment may be tricky, as several other terms, such as colouring agent, ink and dye may appear to be synonyms; however, several differences exist among them regarding grain size, composition, application and use. The definitions provided by different authorities such as the ETAD,[4] the CPMA[5] and the DIN Standards Committee Pigments and Extenders[6] are based on current commercial use and classification of colour pigments. They sometimes

[2] https://www.iso.org/obp/ui/#iso:std:iso:6707:-1:ed-6:v1:en:term:3.4.4.27

[3] The hydraulicity of magnesia-limes is debated. Its characterisation as feebly hydraulic relies on Chever et al. (2010).

[4] Ecological and Toxicological Association of Dyes and Organic Pigments Manufacturers (https://etad.com/).

[5] Color Pigments Manufacturers Association, Inc. (https://www.pigments.org/).

[6] Responsible for the European standardization (https://www.din.de/en/getting-involved/standards-committees/npf).

Fig. 2 Chemical reactions occurring during mortar production and weathering (from Davidson et al. 1965; Böke et al. 2006; Uğurlu and Böke 2009; Jakić et al. 2016; Ponce-Antón et al. 2018; Li et al. 2020). The magnesian lime cycle is oversimplified because the formation of the different phases (*e.g.* artinite, brucite, calcite, dolomite, dypingite, huntite, hydromagnesite, lansfordite, magnesite, nesquehonite, periclase and portlandite) depends on the temperature, the CO_2 concentration and the pH and RH (*i.e.* relative humidity). Therefore, it is not straightforward to indicate which phases are formed for each slaking, setting and hardening phase (see also Lanas and Alvarez 2004 on this topic)

Lime production

calcination: $CaCO_3 \xrightarrow{heat} \overset{quicklime}{CaO} + CO_2$

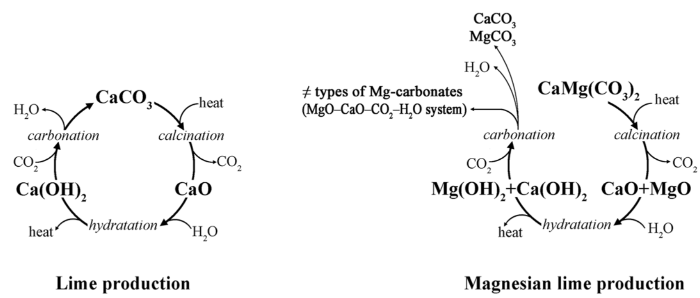
hydratation: $CaO + H_2O \rightarrow \overset{slaked\ lime}{Ca(OH)_2}$

Magnesian lime production

calcination (above 315°C): $CaMg(CO_3)_2 \xrightarrow{heat} MgO + CaCO_3 + CO_2$

calcination (above 528°C): $CaMg(CO_3)_2 \xrightarrow{heat} MgO + CaO + 2CO_2$

hydratation: $MgO + CaO + H_2O \rightarrow Mg(OH)_2 + Ca(OH)_2$



Gypsum production

calcination: $CaSO_4 \cdot 2H_2O \xrightarrow{heat} CaSO_4 \cdot ½H_2O + \frac{3}{2}H_2O_{(g)}$

rehydration: $CaSO_4 \cdot ½H_2O + H_2O \rightarrow CaSO_4 \cdot 2H_2O + H_2O$

Additives production

kaolinite→metakaolin. $Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O \longrightarrow Al_2O_3 \cdot 2SiO_2 + 2H_2O$

Reactions

aerial mortar $\overset{slaked\ lime}{Ca(OH)_2} + CO_2 \rightarrow CaCO_3 + H_2O$

using metakaolinite: $\overset{metakaolin}{Al_2O_3 \cdot 2SiO_2} + \overset{slaked\ lime}{7Ca(OH)_2} + 19H_2O \rightarrow \overset{CAH}{4CaO \cdot Al_2O_3 \cdot 19H_2O} + \overset{CSH}{3CaO \cdot 2SiO_2 \cdot 7H_2O}$

Effect of salt

$$2NaCl + Ca(OH)_2 \rightarrow CaCl_2 + 2NaOH$$

$$CaCl_2 + 3Ca(OH)_2 + 12H_2O \rightarrow 3Ca(OH)_2 \cdot CaCl_2 \cdot 12H_2O$$

include both organic (*i.e.* containing carbon) and inorganic substances under the general definitions of "pigment" and "dye". In the current regulation (ISO 18,451–1:2019[7]), a pigment is defined as a "*colourant consisting of particles, insoluble in the application medium (e.g. coating material or plastic)*" where colour is a "*generic term for all colouring substances*" and subcategories are allowed based on their chemical composition and properties (*e.g.* inorganic, organic, coloured, white, effect, corrosion-inhibiting, magnetic). Main distinctions are traced as follows:

– Dyes ("colourant*, soluble in the application medium*");
– Pigments used for ceramics and glass (called stains);
– Extenders ("*material in granular or powder form, practically insoluble to somewhat soluble in the application medium and used to modify or influence certain physical properties*");
– Fillers ("*coating material with a high proportion of extender, intended primarily to even out irregularities in substrates to be painted and to improve surface appearance*").

Undoubtedly, the increase and the variation in terminology that we have witnessed in recent years follow the development of technological products that did not exist in ancient times and the creation of products used for conservation.

[7] https://www.iso.org/obp/ui/#iso:std:iso:18451:-1:ed-2:v1:en:term:3.19

It is not uncommon, in modern literature, to find the term inorganic dyes used in place of pigment or the term colouring agent used in the sense of chromophore and applied to both pigments and dyes. Therefore, while it is necessary to consider and adopt it when necessary, it is also worth providing adequate terminological explanations and maintaining a certain consistency with our study field, hence avoiding unnecessary anachronisms.

To make an example, each definition produced by the sector authorities agrees on the basic distinction: pigment-insoluble, dye-soluble; however, no further specification is given on the state (solid or liquid) or nature (organic/inorganic and natural/synthetic), meaning that all combinations appear possible. Conversely, in archaeometry, these distinctions have important implications regarding the analytical techniques to be used for their characterisation and the problems related to conservation.

In the current archaeometric literature and practice (Table 1), pigments are considered inorganic materials (chiefly minerals and earths). They may be either natural or synthetic (*e.g.* Egyptian blue) and colour the surfaces to which they are applied by simply covering it. Their use for different types of paintings (*e.g.* paintings, frescoes and icons) is the most common one; however, when used for dark writing in manuscripts, the name switches from pigment to ink. Conversely, dyes mainly include organic materials, frequently soluble (also insoluble, *e.g.* indigo), that can chemically disperse into and bind to the materials. They are frequently obtained from plants (*e.g.* leaves, berries and roots) or animals (*e.g.* insects and molluscs), and their multiple uses range from textiles and food colouring to painting. Finally, the terms de/colouring agents are used only in the field of glass studies and are sometimes synonymous with chromophore, to identify elements and phases—exclusively inorganic so far—that chemically bond the material to which they are added.

To conclude, we must add that (a) mixed inorganic–organic compounds are also known, as evidenced by the mixture of palygorskite clay and indigo used for Maya blue; (b) the term colourant should be avoided in scientific literature, as it is a general term that includes both pigments and dyes.

Building archaeology

The building archaeology (BA) is a research methodology and a discipline that provides a reading of the macro transformations of the investigated building, such as changes in the original project, added structures, interventions for the redistribution of spaces, use and functions of buildings and analysis of the environmental or built context related to the building.

Born at the end of the 1980s by Italian medieval archaeologists (Siena, Genova and Padova universities), the discipline picked up the need to renew the archaeological methods applied to historical architecture.

BA applies the stratigraphic method to any building type, whether or not preserved in its entirety. The theoretical and methodological system used in BA has been, by now, thoroughly discussed and formalised. The main objective is to reconstruct the historical diachrony (phases of construction, use, transformation, abandonment and destruction) of individual structures or entire buildings.

Moreover, in addition to identifying the historical sequences of architectural artefacts, the BA offers a fundamental tool for conservation, eventual restoration and, overall, the protection and safeguarding of historic buildings.

To achieve a diachronic reading of the elevation, the basic method of the BA includes:

- The stratigraphy, for the definition of relative chronologies;
- The study of building techniques, which, in turn, includes:
  - The characterisation of the building materials (stone, bricks, mortar, plaster, etc.);
  - The technical and technological study of both materials and structures and the way materials are assembled.

In practice, the BA consists of (a) distinguishing the constructively coherent parts on a wall/structure, (b) identifying their contours and (c) organising the individual stratigraphic units in a diagram according to a relative chronology. This procedure represents the basis of archaeological stratigraphy, as it allows us to understand the before and after of each action.

To better understand the role of mortars and pigments, it is necessary to clarify the role of building techniques under a theoretical (Mannoni 1997) and methodological (Parenti 1988a) point of view, as to be aware of the limitations in using building techniques to date buildings (Parenti 1988b).

Moreover, this is the study's field, where a fruitful collaboration between different expertise is essential, as it includes the investigation of mortars, plasters and pigments.

The analysis of building techniques has to be multi-level, from the scale of the entire building to the materials constituting the wall, the mortars, the plasters, the binders and the pigments.

In this sense, it is an indispensable premise to any archaeometric research. To interpret the results at the large scale of the entire building, it is necessary to start from sampling at the small scale of the single wall.

At the small scale, BA provides information on the construction morphology, details of the workforce and interpretations of the structures.

All these features, combined together, provide the **sampling criteria** for archaeometric research.

After the completion of archaeometric analyses, the results complete and improve the description of the technique itself. In substance, BA and archaeometry initially work separated and then converge towards an agreed, accurate and comprehensive result.

At the large scale, the construction techniques and the stratigraphy provide the **instruments for the interpretation of results**. The contextualisation of the results is the passage that allows us to reconstruct the history of the building. By integrating all the results obtained from the joint research, it is possible to reconstruct all stages of processing: from the quarry chosen to supply the most suitable material, to the production of lime and mortar, to assembling of materials in the masonry, to the decoration, to the uses and, eventually, to the collapse of the building.

At both the small and large scale, the building technique represents the main instrument to establish the absolute chronology of a built context. Archaeological dating uses direct and indirect sources. The former includes historical, cartographic or iconographic sources, while the latter are deduced from BA or from structural elements (*e.g.* stamped bricks of known chronology or coins; see also Mannoni 1984). Fundamental information is also obtained by studying local chronological clusters, *i.e.* construction techniques closely linked to a territory and the consistency of the material used. Archaeological dating is, thus, the main tool to decide whether the archaeometric analysis of dating is a path to be attempted or not and, if it is, to select the most significant samples.

It goes without saying that, in practice, the chronological sequence of our studies generally corresponds to the small/large scale distinction proposed above.

The archaeometric questions

Taking up the concept already expressed in Gliozzo (2020a), the archaeometric questions guide both sampling and experimental choices, besides providing the key criterion of evaluation for the entire research project. Research questions should be wide and necessarily contextualised in a historical perspective (the "big picture" in Gliozzo 2020a). The traditional—now anachronistic—distinction between archaeological and scientific questions finds a practical compendium in the archaeometric question. Consequently, archaeometric research implies and requires a multidisciplinary team to provide the necessary skill-sets on a permanent basis.

In the field of mortar studies, this unity of purposes and practices is particularly evident and essential. The close link between research phases, such as sampling, analytical study and results interpretation, with the history and intended use of the investigated "structure" implies a close collaboration in planning the best research strategy.

The sections below clarify the main research question raised during the study of the masonry works, including both those that are possible to solve through archaeometric methods and those in which archeometry plays a marginal role. The aim is to provide as complete a picture as possible, albeit within the single article's limits.

The provenance of raw materials used for building materials

While building material characterisation provides precious information on the type of raw material used, the identification and localisation of the supply area inform us on the aspects that determined its selection. This is the main reason why we should consider this question as intended to investigate a territory rather than limited archaeological samples and geological outcrops. Moreover, this is a multi-fold question since it regards all different components: mortars, plasters, pigments, bonded stones or clayey raw materials used for brick making. While it is possible to limit the study to mortars, plasters or pigments, an in-depth understanding of the whole context certainly grants higher quality research. As a matter of fact, this approach makes the difference between mere material analysis and meaningful archaeometric research.

In practice, the localisation of the supply areas or quarries allows a deepening of our knowledge on various issues:

(1) To define what was the builders' awareness of the resources available in their territory;
(2) To determine when the functionality criterion prevailed over that of availability or vice versa;
(3) To find rational explanations to technologically controversial choices;
(4) To answer all those "whys" that follow the provenance issue's resolution and require a thorough knowledge of both the territory and the ancient production techniques.

In addition, the reconstruction of georesources supply strategy and trade patterns clarifies which was the commercial network of reference for the site in a given chronological period and, therefore, allows the researcher to evaluate the choices made by the builders in a broader framework that includes the social and market economy, the geology of the territory and the complex production activities carried out on the site.

In the practice of archaeometric research, this question involves a preliminary in-depth knowledge of the geological setting of a large territory, including the archaeological site, the archaeometric analysis of archaeological materials and that of numerous types of natural georesources that are believed to have been used. All this information provides the indispensable reference database.

To make a shortlist, materials analyses and investigation may regard:

A. Mortars, plasters and pigments from which the study starts (henceforth, "the study's object"), further divided into:

  A(1). Geological sands, which can be compared to those used in plasters and mortars;
  A(2). Geological raw materials suitable for the production of the binder;
  A(3). Any material that may have been added to both mortars and plasters (*e.g.* plants, ground ceramic fragments, or rock fragments) to confer particular properties;
  A(4). The water;

B. In stonework, building stones and relative geological samples. In brickwork, bricks and geological clayey materials, which can be compared to those used in bricks;
C. Minerals and pigments naturally present in the area under investigation.

To better explain how the research deals with the "provenance of raw materials", we propose some examples from published case studies, but, for details, we refer to Ergenç et al. (2021) and Arizzi and Cultrone (2021 in this TC).

A. The study's objects

Undoubtedly, the study's objects, whether they are mortars, plasters or pigments, are typically the first to be investigated; otherwise, we would have no information available to guide the search for the raw materials that have allowed their realization. Indeed, the research begins with the basic question: "**What is it made of?**".

The specific methodologies are addressed in the various contributions of this TC; therefore, we do not dwell on the subject here. However, we would like to highlight two procedural aspects that we believe are important but not always clear to newbies: (1) it is important to perform an accurate visual inspection of mortars and plaster before any sampling is planned, and (2) analyses are not a quick procedure. In most cases, it is not enough to insert the sample into an instrument to get the answers we want. Still from a procedural point of view, the researcher must bear in mind that sampling and subsequent analyses should only be carried out after the stratigraphic and the typological study have been completed; otherwise, the risk is to select a collection of non-representative samples. Moreover, (a) once the first round of materials analyses is completed, the need for targeted field campaigns arises, both to study the territory and to sample the geological materials to be analysed for comparison, and (b) after the characterisation of archaeological and geological materials, a third experimental phase may follow, aimed at verifying some working hypotheses. This is the procedure generally adopted because the archaeometric approach is sometimes intended as a "tool" to find some answers and not as a consolidated procedure, intimately linked to the archaeological research methodology.

On the contrary, if the archaeometrist is involved from the beginning of the excavations, he/she can carry out the geological "reading" of the territory and a part of the samplings in advance of the usual timetable, the objectives can be concerted and programmed more systematically and effectively and well-integrated results may become expendable in less time.

A(1) The aggregate: sands Based on the International Standards Organization (ISO 6707–1:2020[8]), the aggregate is an "inert granular material" that can be divided into fine and heavy, the latter having oven dry-particle ≥ 3000 kg/m^3.

In historical mortars, most of the aggregate is usually constituted by sands; therefore, investigating their nature—"**which kind of sand is it?**"—typically represents the first step.

These are usually classified as fluvial, lacustrine, marine, fossil and quarry sands, depending on whether they are taken from rivers, lakes, sea beaches or other terrestrial deposits. The canonical particle size classification defined by Wentworth (1922) distinguishes sands as very fine (> 63 < 125 μm), fine (> 125 < 250 μm), medium (> 250 < 500 μm), coarse (> 500 μm < 1 mm) and very coarse (> 1 < 2 mm) and the presence of the coarser gravel fraction distinguishes concretes from mortars.[9] It is possible to use other classifications, but the reader must bear in mind that, while the Wentworth classification was specifically formulated in the geological field, others such as the ISO 14,688–1:2002 have been elaborated for engineering purposes. The shape and rounding of the grains, the sorting and the composition represent other important characteristics that, combined with previous ones, provide the necessary information to determine the nature of sands and guide the search for natural outcrops to be sampled for comparison.

8 https://www.iso.org/obp/ui/#iso:std:iso:6707:-1:ed-6:v1:en:term:3.4.4.27

9 In this regard, it is also worth underlining that "*whereas concrete is itself a building material, mortar is a bonding agent that typically holds bricks, tiles and other masonry units together*" (Allen and Iano 2013).

In this way, it is possible to answer the second question aimed at locating the quarry, site, or area of supply of the sand: "**where does it outcrop and from where was it taken?**".

The following questions depend on the results obtained by characterising the mortar/plaster. For example, if the sand that was used has a significant clay component, we may wonder ***why*** the type of sand that is least suited to making these products was used. If, on the other hand, the sand is rich in an earthy component, we may ask ourselves ***why*** the ancient artisans did not feel the need to wash them before use. If, finally, the particle size is poorly/well sorted, we may ask ourselves ***how*** to explain this evidence: comparable with the natural supply deposit or the result of a technological procedure (sieving)?

These are just a few examples of questions that may arise from the initial two. Therefore, it is good to remain flexible when planning the overall research and, especially, the initial sampling because it is not possible to predict them in advance. In several cases, in-depth knowledge of the territory likely provides the key to answer the various research questions. Still, the researcher should be aware that this knowledge often makes it necessary for a supplement of the investigation—aimed at the characterisation of natural materials—because literature data may be insufficient for archaeometric research. Apart from this, an equal in-depth knowledge on the different yield of fine sands (*e.g.* requiring high amounts of binder and water and thus resulting in less workable, too porous and less resistant mortars) and coarse sands (*e.g.* requiring less binder and water, thus resulting in better workable, adequately porous and highly resistant mortars) and, in general, the importance of sorting, or preferences tuned to certain types of sands (*e.g.* river and lake) over others (*e.g.* quarry sands due to impurities; sea sands for aerial mortars due to salts) is preliminarily required.

A(2) The binder The characterisation of the binder always represents a key question: "**which kind of binder was used?**". The analyses combined with the knowledge of the geological settings provide the necessary clues to the identification and localisation of the raw materials used for binder making, *i.e.* to solve the related question: "**where do the raw materials to make the binder come from?**".

Since binders may be a variety of materials, several types of sources should be targeted, such as earths (with earth mortars), clays (with clay mortars), limestone and/or sea-shells (with lime-based mortars/plasters), dolomite and dolostones (with magnesian mortars/plasters), gypsum deposits (with gypsum mortars/plasters) and marls (with lime-based mortars with feeble hydraulic properties).

However, even if, in most cases, the material is of a geogenic nature, its study implies an additional difficulty since we no longer see the "original" material but only its product, further reacted after use. For example, when seashells are used (*e.g.* Gleize et al. 2009), lumps of un-burnt lime are absent since they easily burn completely. Conversely, when carbonatic rocks are used, the presence of these relics is more frequent and may favour raw materials' search; however, the researcher is frequently obliged to deal with very small quantities of little dimensions even in this case. Furthermore, different types of binders may have been used in combination (*e.g.* lime and gypsum). In these cases, determining the nature and origin of the binder may be a complicated task, which requires the use of numerous analytical techniques, capable of providing complementary information.

The finding of the lime kiln or a dump of waste materials would represent an ideal situation. A micro-stratigraphic sampling of contextual materials may guarantee the identification of the raw material, provide information about its processing and, when it is possible to establish a direct connection between the place of production and the structure of use, indicate the degree of mortars elaboration. In such an advantageous situation, the field campaign may be precisely directed towards specific outcrops, and the ensuing laboratory analyses may provide results ranging from a high likelihood to certainty.

In the absence of this archaeological evidence, the level of accuracy of the answers is strongly conditioned by the characteristics of the binder itself (*e.g.* nature, composition, abundance and size) and is, therefore, not predictable.

A(3) The additives (~ modern additives, admixtures and aggregates) In modern terminology, the additive is a material "*added in small quantities to a liquid or granular material to produce some desired modification to its characteristics*" (ISO 6707–1:2020), while the admixture is a material "*added in small quantities before or during a mixing process in order to modify the characteristics of a mixture*". The two terms describe apparently similar materials that, added in small quantities, can modify the properties of the starting mixture. However, the difference between the two terms lies mainly in that the additive is added during the manufacturing of the cement, while the admixture is added to the concrete during mixing. Moreover, the UNI EN 934–2 standard states that the admixture for concrete should be in an amount not greater than 5% by mass, compared with the cement. Therefore, under the current rules, the addition of materials, such as ground ceramic fragments, in considerable proportions would fall into the category of aggregates and not that of additives.

However, one of the main aims of the archaeometric study on ancient mortars and plasters is to reconstruct the production's phases and the supply modalities of each component. In this case, the term additive, used in its etymological sense (from *addere*, to add) has the advantage of clearly recalling the difference between a primary component (frequently sand) and an added component (*i.e.* the additive) in the aggregate. This is the meaning we have given to the term additive in this article. Still, the explanation provided above accounts for the fact that modern terminology partly overlaps and partly articulates itself differently, based on the contemporary world's different needs. It follows that, as long as terminological uncertainties persist, it is always good to specify the choice made in the description of the results.

Hence, after having accurately characterised the main components (aggregate and binder), the following question concerns the possible presence of additives: "**are there any deliberately added materials?**". If yes: "**which types of materials were added?**" and "**where did they come from or how were they made?**" (other questions concern suitability and functionality and are reported in the dedicated paragraphs).

The addition of inclusions to improve the final product's performance was a common practice that frequently left recognisable traces in literary sources and archaeological evidence. Consequently, citable examples are numerous and provide us with an extremely varied picture in which inorganic and organic materials are enlisted. Therefore, it is possible to formulate the characterisation question in terms of: "**are the additives of an inorganic or organic nature?**" (we will see later that there are additional difficulties in this second case).

To properly solve this question, it is necessary, perhaps even more than in previous cases, to know which types of additives were used to facilitate—or even allow—their identification during the analysis of the "study's object". For this reason, we present a shortlist of inorganic and organic additives (and admixtures); however, we refer to the other contributions for the necessary insights on their characteristics and properties (Arizzi and Cultrone 2021; Ergenç et al. 2021).

Beginning with inorganic additives, the most common were natural and artificial pozzolans that are defined as "*siliceous or siliceous and aluminous materials which in themselves possess little or no cementitious value but will, in finely divided form and in the presence of moisture, chemically react with calcium hydroxide at ordinary temperatures to form compounds possessing cementitious properties*" (Dodson 1990).

Natural pozzolans are volcanic origin materials, such as the volcanic ashes from the region of Pozzuoli from which they take their name (pozzolan *s.s.*). These are often found in the mortars/plasters of sites located within or close to volcanic regions (Barba et al. 2009; Özkaya and Böke 2009; Villaseñor and Graham 2010; Kurugöl and Güleç 2012), but when they are found in sites far from supply areas, archaeometric research is called upon to localise their origin and reconstruct the trade routes that allowed their import and usage. The provenance question may thus remain confined to the scale of the site and its territory or expand to long-scale range imports and cross other issues such as those concerning the methods and extent of the marketing of raw materials in a given period (*e.g.* "**what was the diffusion of these materials?**"; "**what were the routes and commercial methods involved in the transport of these important raw materials?**").

The discovery of recycled amphorae containing north-Latial pozzolans in the ship B of Pisa (Augustan Age) is a clear example of how the archaeometric analyses can effectively trace the movements of these raw materials, apparently also used as ballast despite its low weight. As further evidence that the investigation of the provenance is only the first step to seek historically much more significant reconstructions, the studies conducted on volcanic scoria in some buildings of ancient Rome (Lancaster et al. 2011) have not only updated our knowledge on the chronologically diversified supply of these materials from Rome, the Vesuvius and the Campi Flegrei, but also imposed a revision of the land and sea transport system and hypothesised an imperial involvement in the trade of these raw materials. Similarly, the studies conducted on Tunisian (from Sardinia or Pantelleria) and Turkish (from short-range resources) analogous materials have shed new light on the methods of importation (primary or secondary loads associated with other goods, such as grain mills), as well as demonstrated their long-range transportation (Lancaster et al. 2010).

In a nutshell, from the provenance investigation of a particular additive in the mortar, it is possible to find ourselves investigating the favourability of winds along one sea route compared to another, the distribution of primary and secondary ports, the type and capacity of ancient boats, the circulation of associated goods (*e.g.* foodstuffs, ceramic, millstones, building materials) and the ancient navigation methods (e.g. coastal navigation): it is precisely here that the study becomes interesting and fruitful.

Proceeding further, it is worth adding that the term pozzolan may also apply to non-volcanic, natural and artificial materials with similar characteristics (chiefly silica and alumina, along with ferrites). Therefore, the question may turn into: "**what type of pozzolan was added?**".

Main examples are represented by:

– Ceramic powder (especially tiles). Based on literary sources, this was one of the most typical inclusion ("cocciopesto" and *opus signinum* in the Roman world; *Khorasan* in Turkey; *Surkhi* in India; *semen merah* in Indonesia; *Homra* in Arabic countries; Spence 1979). During the Roman age, both *Vitruvius* and *Plinius* reported on the effectiveness of such inclusions in constructions built along a river or the sea exposed to humidity and marine sprays.[10] During the Middle Ages, Villard de Honnecourt (French master mason of the thirteenth century

[10] *Vitruvius*, *De Architectura* II, 5, 1 "*etiam in fluviatica aut marina si qui testam tunsam et succretam ex tertia parte adiecerit, efficiet materiae temperaturam ad usum meliorem*". (If to river or sea sand, potsherds ground and passed through a sieve, in the proportion of one third part, be added, the mortar will be better for use). Pliny, *Naturalis Historia*, XXXVI, 54, 175 "*si et testae tusae tertia pars addatur, melior materia erit*" (If, too, one third of the mortar is composed of bruised earthenware, it will be all the better).

AD) reported a hydraulic paste recipe based on lime, pounded fragments of bricks and linseed oil.[11] In the Renaissance period, the famous architect Leon Battista Alberti (1404–1472) reported the common opinion that, if you add a third of crushed brick, the resulting mixture becomes much more tenacious.[12] A century later, Pietro di Giacomo Cataneo (Sienese architect, 1510–1574) handed down the recipe for mortar, recommending two lime parts and two other parts of tile powder, with half a part of iron flakes.[13] The list could go on for long but we believe it is already clear how the masters configured the use of this material as "typical" over the centuries. The archaeometric analyses support this reconstruction. Ceramic powders, sometimes mixed with other pozzolans and volcanic materials, have been frequently found in mortars and plaster dated to the:

- Late Bronze Age (Theodoridou et al. 2013);
- Hellenistic (see, *e.g.* in Italy, Montana et al. 2016);
- Roman and early Byzantine periods (see, *e.g.* in Italy, Bugini et al. 1993; Damiani et al. 2003; Bertolini et al. 2013; Izzo et al. 2016; Columbu and Garau 2017; Graziano et al. 2018; Miriello et al. 2018, Montana et al. 2018; Sitzia et al. 2020; in Spain Alonso-Olazabal et al. 2020; in Tunisia, Farci et al. 2005; and in Turkey, Bakolas et al. 1998; Miriello et al. 2011);
- Medieval periods (see, *e.g.* in Greece Moropoulou et al. 2000; in Italy, Lezzerini et al. 2014; in Portugal, Adriano et al. 2009; in Turkey, Kurugöl and Güleç 2012) and beyond (*e.g.* in the Czech Republic, Přikryl et al. 2011; in Italy, Cantù et al. 2016; in Turkey, Böke et al. 2006; Uğurlu and Böke 2009; Binici et al. 2010);

- Processed slags from metalworking (see, *e.g.* Diekamp et al. 2006; Cacciotti et al. 2015; and Kropáč and Dolníček 2013 for correlated evidence);
- Processed clays such as metakaolins (see, *e.g.* Baronio and Binda 1997; particularly used for restoration, *e.g.* Aggelakopoulou et al. 2011; Vejmelková et al. 2012a; Loureiro et al. 2020);
- Diatomaceous earths (see, *e.g.* Franzini et al. 1999, 2000) and opal-A (Sarp Tunçoku and Caner-Saltık 2006);
- Specific soils in earthen mortars (see, *e.g.* Cantù et al. 2016).

Finally, marble (or travertine) dust was used in mixtures with sand and lime for plasters to be decorated or for the bed mortar layer of mosaics or for relief decorations (stuccos) that had the splendour and luster of marble (*opus albarium* or *caementum marmoreum*; see *Vitruvius*, *De Architectura*, VII 2, 1–2 and Cataneo 1567, II 11; for a review on some historical treatises see Salavessa et al. 2013; for some archaeometric evidence see Toniolo et al. 1998; Riccardi et al. 2007; Kriznar et al. 2008; Weber et al. 2009; Duran et al. 2010; Miriello et al. 2011; De Luca et al. 2012; Robador and Arroyo 2013; Lezzerini et al. 2014, 2019).[14]

As for the identification of organic additives, additional difficulties need to be known in advance. The substantial difference between inorganic and organic materials is that the former are generally visible to the naked eye or at medium magnification. In contrast, many of the organic ones (*e.g.* milk and egg whites) are not visible. Therefore, the research question "**are there any organic additives?**" stems more from the preliminary knowledge of ancient methods and techniques than from macroscopic and microscopic observation but is archaeometrically resolvable through an ad hoc analytical strategy.

The next step concerns the characterisation: "**what kind of organic substance was used?**".

The ancient authors documented the extensive use of both plant and animal origin substances to improve the performance of mortars and plasters. For example, *Vitruvius* suggested the use of lime tempered with oil (and dregs of oil) for waterproofing and preventing frosting (*De Architectura*, VII 1, 6–7; VIII 6, 8), thus demonstrating that the water-repellent properties of natural oils and fats were well known in Antiquity. Pliny reported that the temple of Minerva of Elis had been plastered by *Panænus* (brother of *Phidias*) with the addition of milk and saffron.[15]

[11] Album de Villard de Honnecourt, Plate XLII "*On prend chaux et tuile de paiens pilee, et vous ferez autant de l'une que de l'autre, mettant un peu plus de tuile de paiens, jusqu'a, ce que sa couleur domine l'autre. Detrempez ce ciment d'huile de lin, et vous en pourrez faire un vaisseau a contenir l'eau*." (Take lime and pounded pagan (Roman) tiles in equal quantities, adding a little more of the latter until its colour predominates. Moisten this cement with linseed oil, and with it you can make a vessel that will hold water)".

[12] *De re aedificatoria*, III, 4 "*tertiam si tunsae testae partem adieceris, affirmant omnes futuram multo tenaciorem*".

[13] *I Quattro Primi Libri di Architettura*, II, 12 "*piglisi per ogni due staia di calcina due altre staia di polvere di tegole, con mezzo staio di scaglia di ferro*".

[14] Waste generated during mining and processing of marble is nowadays re-evaluated for the same use (see *e.g.* Kore et al. 2020).

[15] *Naturalis Historia* XXXVI, 55, 177 "*Elide aedis est Minervae, in qua frater Phidiae Panaenus tectorium induxit lacte et croco subactum, ut ferunt; ideo, si teratur hodie in eo saliva pollice, odorem croci saporemque reddit*".

The archaeometric research allowed for the individuation of several others organic additives in archaeological finds, for example:

- Wood, straw and charcoal in Roman mortars and plasters from Petra in Jordan (Al-Bashaireh and Hodgins 2011);
- A saccharide material-based additive of plant origin and a natural gum in the mortars of the medieval shipyard of Amalfi Arsenale (Rampazzi et al. 2016);
- Egg white and urea in the mortars from the Yoros Castle (13^{th}–14^{th} AD) at Anadolukavağı (Kurugöl and Güleç 2012);
- Fibers in 13^{th}–18^{th} AD mortars from Erzurum in Turkey (Binici et al. 2010);
- Proteins and/or animal blood in post-medieval earthen mortars from Cremona in Italy (Cantù et al. 2016);
- Egg yolk, oil and some resin in mortars repairs from the St. Engracia Basilica at Zaragoza in Spain (Luxán et al. 1995);
- Sticky rice, egg white and tung oil, brown sugar, pig blood and tung oil in several Chinese mortars, variously dated from 563 to 1381 AD (Yang et al. 2009; Yang et al. 2010; Zhang et al. 2014; Zhao et al. 2014a, b, 2015).

These few examples should have clarified that the use of organic additives is ancient and seamlessly has come down to us, in practically every part of the world. The second aspect to consider is that not all archaeometric methods allow for accurate identification of the organic substances; therefore, the analytical procedure must be strategically targeted. This appears even more evident if we try to draw up a list of possible organic substances used for mortars and plasters. Using only the reference quoted here, along with those discussed by Sickels (1981, 1982), it is possible to list (in alphabetical order) animal glue, barley, beer, beeswax, blood (also of hippopotamus), butter, charcoal, cheese, cotton, curd, dung, egg white and yolk, elm bark, fibres, fruit juices (*e.g.* fig), gluten, gum Arabic or tragacanth, hair, hogs' lard, keratin, malt, milk (casein), molasses, oil (*e.g.* linseed oil and tung oil), resin, rice, rye flour, saffron, shellac, starch, straw, suet, sugar, tallow, tannin, urea, wine and wort.

It goes without saying that the variety of materials is wide and that, most likely, we do not know it fully yet. What is certain, however, is that the provenance question may not directly regard a geographic area but a certain animal or vegetal species, *e.g.* "**from which animal does the blood come?**"; "**which plant does the wood come from?**"; "**where was the species from which the oil was extracted grown or fished/bred?**".

As a final example, we propose a comprehensive study on Chinese organic–inorganic composite mortars performed by Li and Zhang (2019). Their research focused on 358 mortar samples taken from 159 buildings (city walls and forts, monumental buildings and offices, temples, pagodas, houses and tombs) dated from the Taosi phase to the Qing Dynasty (2300 BC–1911 AD). Different analytical approaches allowed these authors to determine the presence of starch in 112 samples, oils in 87, proteins in 59, sugar in 14 and blood in 5; furthermore, 48 and 5 samples showed two and three organic additives, respectively. Apart from the comprehensive and diachronic reconstruction proposed by these authors, it is also interesting to learn how the choices made by the ancient artisans were directly correlated with (a) the variable climatic environments, (b) the different seismicity of the various areas, (c) the development of agriculture and (d) socio-cultural factors linked to the relationship between man and nature and Confucianism.

Also in this case, therefore, the characterization of additives may lead to studies regarding ancient agriculture, beliefs, philosophies and religions, the seismicity of specific territories, the societies, understood as cultural ensembles, and the dynamics of trade in perishable products.

Finally, it can be concluded that the provenance of an additive is more likely a starting point rather than an arrival point: "**beyond the technical aspect, what further information can I deduce from the presence of these additives in mortars/plasters, starting from their provenance?**".

A(4) The water Water supply is often taken for granted, especially if the archaeological site is located along a watercourse or near the sea. However, water procurement deserves a closer study—"**which kind of water did they use and where did they get it from?**"—as it was necessary to supply considerable quantities, and different types of water (*i.e.* from wells, springs, rivers and sea) have different yield over time. For example, salt content can damage an aerial mortar while enhancing the binding properties of a hydraulic mortar (Davidson et al. 1965; Karim et al. 2017; Li et al. 2020; see also Fig. 2).

The presence of chlorine and sodium in the analysed mortars may give a clue on this aspect and clarify specific technological choices. For example, these two components in the binding matrices and pozzolanic lumps of some Nora plasters (used for waterproofing this Punic-Roman site's cisterns in Sardinia, Italy) made the authors suggest the use of seawater for their preparation (Secco et al. 2020).

Looking at the same topic from the point of view of conservation, the decay induced by salt crystallisation cycles (salt weathering) is a factor that puts monuments at risk; moreover, it is foreseeable that it will be more and more significant due to the climate breakdown we are witnessing (see below. On salt weathering also see Ergenç et al. 2020; Randazzo et al. 2020). Based on these examples, the question "**where was the water for the mortar/plaster mix from?**" certainly acquires a much more meaningful motivation and it would also be useful to investigate "**what quantities were needed?**" for the masonry under examination.

B. Stones and bricks

In this case, the materials under investigation may either be worked stones and/or ceramic bricks and tiles. The questions arising from their study are only partially different: (a) dealing with stoneworks, the main questions regard the characterisation ("**what kind of stone was used?**") and the localisation of the geological outcrops ("**where do the stones come from?**"); (b) when ceramic bricks were used, the questions regard the characterisation of the raw materials ("**what kind of clayey raw materials were used?**"), the localisation of the clay outcrop that supplied them ("**from which outcrop were they taken?**") and, possibly, the localisation of the production plant ("**where were the raw materials prepared and fired?**"). In this second case, the study of mortars is closely linked with both that of stone materials and that of ceramic productions and, therefore, it becomes obvious how a global approach to an archaeological site may prove economically much more sensible, as well as historically more interesting, than a single material approach. It should also be added that the additives found in mortars may result from waste from stone processing or from the shredding of local ceramics; therefore, the investigation of stone materials may provide information that can be used in more than one field of investigation (see Fort et al. 2021).

In this case, the example we propose is regarding the archaeological site of *Thamusida*-Sidi Ali ben Ahmed in Morocco (Fig. 3), where both stones and bricks were used for the masonries and a global archaeometric approach was applied to its Cultural Heritage remains. In this case, the presentation goes through the series of questions that typically arise during the research and the provided answers (Q&A), as summarised in Table 2.

The processing of raw materials used for mortars and plasters

In the study of masonry works, the preparation concerns every single component found in the mortar but also, as seen in the previous example, the processing of stones and bricks (*i.e.* "**is it possible to recognize traces left by stoneworking?**"; "**how were raw clays prepared for brick making?**").

Focusing on mortars and plasters, the questions, therefore, concern:

- The sands (*e.g.* "**were the sands prepared or used as they were?**"; "**were they washed to eliminate sodium chlorides?**"; "**were they ground, sieved or mixed to obtain the desired granulometry and sorting?**")
- The binder and then the lime processing (*e.g.* "**are there unburnt portions?**"; "**is it possible to trace the kiln and evaluate its function and effectiveness?**")
- The additives (*e.g.* "**have they been chopped or pulverized?**")

It is useless to deny that the answers that we can realistically obtain may be very uncertain or partial, often dictated by common sense than by real archaeometric evidence. In the most fortunate cases, the evidence of production structures such as lime kilns are found (*e.g.* Vaschalde et al. 2016; Toffolo et al. 2017a; Casas et al. 2020; Goguitchaichvili et al. 2020), and the researcher can thus reconstruct the lime production process. There are also particular cases in which the slags, sintered during the burning process inside the inner lining of the lime kiln, have been reused inside the mortars. Their discovery may indirectly provide information about the production and preparation of both the binder and the additives (Kropáč and Dolníček 2013). A similar fortunate case—although very rare—is represented by layers of abandoned raw materials from which we can obtain important information about sand processing.

Despite the difficulty in tracing this information, its use for both conservation and formulation of compatible mortars is undeniable. It is well known, for example, that the use of unwashed marine sands introduces harmful Cl-salts into aerial mortars; moreover, the evaporation of saline solutions can lead to discontinuities. Indeed, saline solutions represent a critical factor that may enter the mortar also a posteriori, for example, during flooding events or the circulation of thermal waters.

Finally, this question and the following one imply a whole series of questions related to the organisation of the production, through which we can move from the analysis of the material culture to the study of workers, observing what the production relationships were, how it was their work, what surfaces they were able to achieve at a certain time and so on (DeLaine 2021).

The reconstruction of mortars and plasters recipes

This technological issue includes a simple question to which a necessarily simplistic answer is usually provided. The question "**in what proportions were the various components inserted and mixed?**" may be misleading if we consider some non-computable variables, such as:

a. The compositional variability that is certainly present in the different days of work;

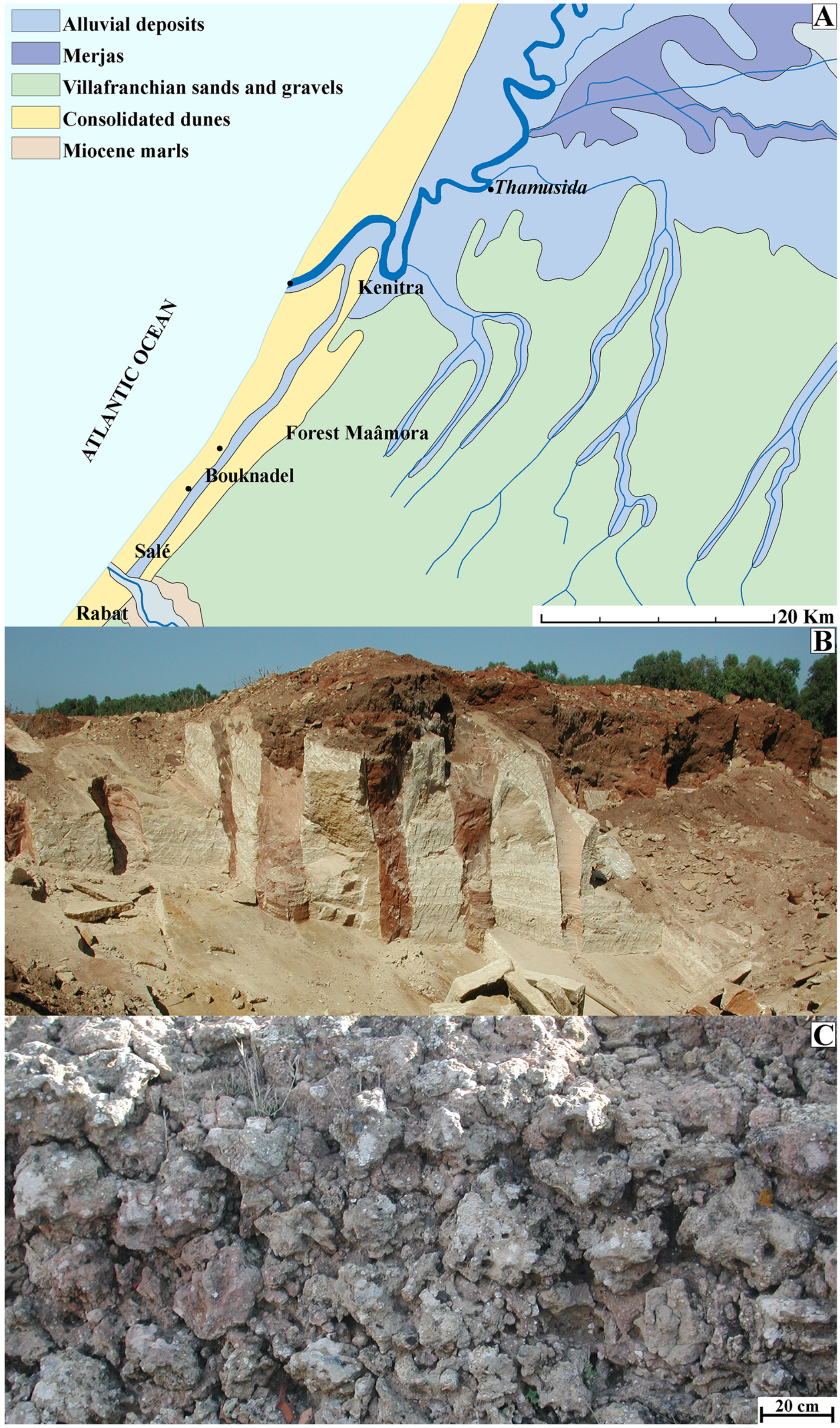


Fig. 3 (a) The Moroccan area including the archaeological site of *Thamusida* in Morocco. *Thamusida* corresponds to the present area of Sidi Ali ben Ahmed and is located along the left bank of the Sebou River (15 km north of the city of Kénitra and about 20 km north of the estuary. The widest supply area for calcarenite is that of Sidi Bouknadel (about 30 km SW as the crow flies). The two black dots above and below Bouknadel correspond to two quarries whose calcarenite was characterised for comparison. (**b**) An open quarry of calcarenite at Bouk Nadel. (**c**) A wall of the archaeological site of Thamusida where calcarenite was used

Table 2 Q&A on building materials provenance raised and provided during the development of the *Thamusida* research project (years 2003–2008)

Questions	Actions required	Answers
What kind of stone was used?	Representative sampling of stones in *Thamusida*'s masonries	Rocks macroscopically the same in the site. Samples distinguished into quarry cuts (squared), roughly hewn stones (with a flat face) and stones used as they are (shapeless)
	Characterization through OM, SEM–EDS, XRD, XRD	The samples consist of polygenic clasts, rounded in shape and constant granulometry (150 and 200 µm). The clasts are mainly of quartz, calcite, fragments of calcareous shells and lithic fragments (esp. sandstone with a clayey matrix). The rock used for stone masonry is a calcarenite (for details Giorgetti and Gliozzo 2009)
Does calcarenite outcrop at the site?	Short-range distance field campaign	No
So, where did they get it from?	Geological study of the area and longer-range field campaign	Calcarenite outcrops are common along a large part of the coastal strip (approximately corresponding to the yellow strip in Fig. 3). Numerous quarries are still active along National Route 1, especially in the area comprised between Salè and Sidi Bouknadel. Compared to the site, the most consistent and close outcrops are those of Sidi Bouknadel
Is there any way to know exactly where the artisans took it from?	Sampling campaign of un-worked rocks	Samples were taken along the coast, in several sites north and south of the site
	Characterization through OM, SEM–EDS, XRD, XRD	Set up of an ad hoc database of unworked calcarenites to be used for compositional comparison. The analyses showed both a great overall similarity between worked and un-worked samples and an intra-site compositional heterogeneity that prevented a more precise localisation (for details Giorgetti and Gliozzo 2009)
Can we try to figure out how they transported it to the site, although we don't know the exact spot where it came from?	Knowledge on the modes of transport adopted in antiquity and GIS-based network analysis	The most likely path includes a first section of marine navigation along the coast, moving from north or south to the Sebou River's mouth. The second section of river navigation runs against the current, moving from the mouth to the site. In this second section, they can have used the (alaggio) hauling method, i.e. boats like the *naves caudicariae* pulled with ropes by animals or by men who proceeded on roads parallel to the course of the river, specially made for this purpose. GIS-based network analysis also informs that all possible land paths require much more time than the navigated ones
So, why did they go to get that kind of stone off the site?	Detailed knowledge of the geological setting of the territory	Although this lithotype is not readily available in the surroundings of *Thamusida*, it still represents the lithotype available at the shortest distance from the site. Therefore, calcarenite choice was still the cheapest one, in terms of time and effort to transport it to the site (compared for instance with limestones and basalts). Moreover, this is the most common type of stone in the area and, in fact, the same stone is also used in today's city of Rabat (e.g. for the Tour Hassan or the Kasbah des Oudayas), although its carbonatic composition and porous structure cause significant conservation issues (for details Zaouia et al. 2005)

Table 2 (continued)

Questions	Actions required	Answers
On the other hand, the bricks with what were they produced and where they come from?	Representative sampling of bricks and tiles in *Thamusida*'s masonries and brick wastes from the ceramic kiln	Collection of 67 bricks and tiles: 27 from the area of the Islamic ceramic workshop (active from the 8th cen. AD onwards); 40 from bricks and tile in the Roman settlement (1st-3rd AD), mostly in situ
	Characterization of the ceramics through OM, SEM–EDS, XRD, XRD	Both Roman and Islamic bricks used local grey tirs (illitic clays) as received, *i.e.* without manipulation or temper addition and fired them at 600–950 °C (for details Gliozzo et al. 2011)
	Short-range distance field campaign for the individuation and sampling of clay outcrops	Two were the raw materials closely available for ceramic production: the grey and black tirs that are clay soils more (black) or less (grey) rich in humus (for details Arnoldus Huyzenveld 2008)
	Characterization of the clayey samples through OM, SEM–EDS, XRD, XRD	Set up of an ad hoc database of clayey raw materials to be used for compositional comparison. Local ceramists selected the grey *tirs* and were particularly active in the earlier phase of Roman occupation (first–third centuries AD). Conversely, the later phase witnesses a contraction of production favouring imports which end up supplanting local production (for details Gliozzo et al. 2011)
	Epigraphic study of stamps on bricks	Imports from Banasa (bricks stamped QAP and APP) and, likely, from the bay of Tangiers (bricks stamped CN and HADRIAVG) (for details Gliozzo et al. 2011)
In conclusion, what was the operational criterion underlying the construction of the masonry works of Thamusida?	Integration of all results achieved	The best criterion, combining economy and functionality at the same time

Abbreviations: *OM* optical microscopy, *SEM–EDS scanning electron microscopy coupled* with *energy dispersive* X-ray spectroscopy, *XRD* X-ray diffraction, *XRF* X-ray fluorescence

b. A dismissal that led to the replacement of the person in charge with another worker who introduced a partially different recipe;
c. A momentary shortage of one of the raw materials that led to the mortar's completion by not respecting the original recipe, and other cases of this type, that are possible but not traceable.

Furthermore, mortars and plasters are materials that tolerate a wide compositional variability. By changing the ratio of components, various properties may change, such as setting time, durability or mechanical properties. Still, it is not certain that the ancient workers understood a decrease in these properties as we are used to interpreting them today. The error of anachronism—that is to evaluate the quality of a product based on modern parameters—must always be kept in mind. In several cases, this distinction does not make much sense or is not drawn based on realistic or sensible parameters.

Another, often discouraging, phenomenon concerns the approach with ancient texts. For a long time, researchers have debated whether and how the indications of *Vitruvius*—or other ancient architects—swere respected or neglected, giving rise to a series of interpretations of little use. The text of *Vitruvius* has often been read with all the rigidity of a black and white text, in order to deny the validity of his teaching or to identify any detachments from the norm. How ancient texts should be read is explained by Lancaster (2021) in this TC.

Operationally, archeometry can answer—with different levels of approximation—the questions: "**how much aggregate, how much binder and how many additives (if any) were mixed to make this mortar/plaster?**", or "**what is the ratio between the various components?**" and the results relate to the individual sample analysed and not to the entire masonry work. To obtain a realistic estimate of the proportions adopted for the entire work, it is necessary to select and collect a stratigraphically appropriate and numerically representative collection of samples and to evaluate homogeneity/heterogeneity induced by random and non-calculable factors, such as those described above.

The characterisation of the components and the evaluation of the relationship between them is a tool to evaluate changes made during the work or diversified choices, based on the type or location of the structure under eximination. For example, if the ratio varies significantly within the plasters of the same room, it may be possible to imagine a change of the workers or a series of renovations. If the ratio changes significantly between buildings with different use (*e.g.* dry/humid, internal/external environment), a deliberate diversification (*i.e.* to confer different properties to the materials) becomes possible.

The suitability and functionality of mortars and plasters

These issues include several questions that may widely vary depending on the types of examined structure.

For example, assessing the hydraulic properties of mortars/plasters is a typical research question, especially when exposed to high moisture levels, such as thermal baths and cisterns, or wet environments, such as harbours. The questions that can be made in these cases are various, for example: "**is the plaster suitable for the environment in which the workers applied it?**"; "**is the composition of the mortar suitable for setting underwater?**"; "**is the plastering of the cistern functional to water containment?**"; and "**is the mortar/plaster suitable for the intended use?**". Questions about the functionality of a mortar/plaster are all the more complicated, the greater the number of variables to consider. Mortars and plasters must be regarded as systems in which each component plays a specific (but possibly different) role, depending on the components with which it is mixed. Aerial lime-based mortars/plasters appear as the most straightforward system, as they are reduced to two components, *i.e.* the aggregate and the binder; however, even assuming an invariable binder's composition, the result is not obvious. The aggregate may vary in terms of composition, grain size, roundness and sorting and each of these properties corresponds to a different yield of the final product (see, *e.g.* Grassl et al. 2010; Idiart et al. 2012). Nevertheless, the binder's composition may vary when the lime is obtained from impure limestones. In this case, the type and amount of impurities present in the raw material will determine the characteristics of the binder.

If additives are added to a lime-based mortar/plaster, the number of variables and, consequently, the complexity of the system increase. Each type of additive will trigger specific reactions depending on the system and different characteristics will be achieved by the final mortar/plaster.

This reasoning serves to convey that the characterisation of a mortar/plaster, as aerial or hydraulic, is only a first step to defining its suitability and functionality. The latter properties require the evaluation of numerous features such as, for example, shrinkage, workability, plasticity, stiffness, brittleness, hardening with high relative humidity, pozzolanic activity, thermal conductivity, porosity, water permeability, adhesion, mechanical strength, tensile strength, compressive strength, moisture resistance and frost resistance.

Each of these parameters varies according to the number and type of variables that constitute the system. To give just a few examples related to the strength of a mortar, it is possible to list:

- Using natural hydraulic lime (NHL), high strength is obtained when a suitable grain size distribution of the aggregate is achieved; however, the strength decreases

if the aggregates are siliceous, while it increases if the aggregates consist of limestone and/or the percentage of binder also increases (Lanas et al. 2004);
- The compressive strength varies, for example, with the variation of the binder's metakaolin content, while it remains almost unchanged if crushed brick particles are added (Nežerka et al. 2014). The addition of metakaolin can increase the compressive strength of lime-based plasters up to five times (Vejmelková et al. 2012b);
- Using Mg-limes, the strength is comparable to that of natural feebly hydraulic lime, according to the parameters established by EN459-1. With respect to pure calcium-lime, the flexural strength and the compressive strength can be 1.75 and 2.4 times higher (Pavia et al. 2005; Chever et al. 2010);
- The strength is low in pure lime matrix pastes, due to the formation of cracks around the aggregate; therefore, the characteristics of the aggregate becomes crucial (Nežerka et al. 2014);
- The mechanical strength can also be increased through the introduction of casein or animal glue (Ventolà et al. 2011) or even sticky rice soup (Yang et al. 2009, 2016).

This is an example related to a single property, the strength, but highlights how suitability and functionality depend not only on multiple variables but also on the characteristics of each component.

To make just one example of the suitability/functionality of historic mortars, Singh and Kumar (2020) investigated a series of plasters from the late fifteenth century Mughal's summer palace of Farah Bagh (India). Although the most striking feature of this monument is perhaps that of being equipped with a complex system of air cooling, the equally complex preparation and application of different types and layers of plasters are indicative of an advanced technology, aimed at their suitability and durability during processing and use in an environment that is repeatedly subjected to moisture. The workers obtained flexibility, strength and permeability by adding several types of inclusions, ranging from jute fibres and dry paddy stems to grinded ceramics and basaltic rocks.

Of a very different nature are cases in which the hydraulicity of a mortar/plaster is involuntary or not functional for any purpose (*e.g.* in some residential rooms in predominantly dry climates). The question thus becomes "**why was a hydraulic mortar used when it was not necessary?**" and/or "**did they really want to hydraulicise the mortar, or recycle materials already available, resulting, for example, from demolition of old structures?**".

This is a more common situation than imagined and clarifies how a mortar/plaster's hydraulicity is a characteristic to be interpreted with caution and above all else, not individually. For example, we know that some precautions such as the introduction of fibres made these materials more suitable for use in seismic areas and therefore, in these cases, the question may become: "**are mortars and plasters suitable for that given structure, considering the seismicity of the area?**".

There are also cultural factors to consider when the structure in question is large enough to include both environments open to external visitors and work environments open to owners and professionals only. In this case, a question may concern the value given to suitability/functionality with respect to the owners' self-representation (*e.g.* "**is there a variation in the suitability/functionality of mortars/plasters based on the destination of the rooms?**").

Undoubtedly, this case is closely linked to the use of decorations and other questions may arise on the suitability/functionality of the plasterwork for paintings (*e.g.* "**is the plaster suitable / functional to the realisation of wall painting?**").

Furthermore, from a diachronic perspective, the reconstruction of the recipe may also be tackled in chronological terms and, thus, the questions become "**how have the recipes evolved?**"; "**from the beginning of pyrotechnology in the distant Epi-Paleolithic Geometric Kebaran** (Gourdin and Kingery 1975; Kingery et al. 1988) **is it possible to observe radical changes, progressive adaptations or conservative choices?**".

Hence, the various questions that can be raised concern a whole series of how, when and why, the solution of which helps us to understand better a wide range of aspects related to the cost-effectiveness of the work, the technical skills of the workers, the priorities and the variable tastes of clients bound to the fashions of the time and the relationship between the intended use of a structure and its functional performance.

The archaeometric answers that can be obtained for these questions are often decisive, thanks to the numerous experimental works that have addressed the technical characteristics of ancient and modern mortars/plasters.

Essential for this topic are the works carried out by Baronio and Binda (1997) and Böke et al. (2006) on the hydraulicity of different types of bricks and clays for cocciopesto. These studies led to clarification of the conditions necessary for the reaction to occur and the ceramic material to be configured as pozzolanic:

a. Ceramic firing must not exceed 900 °C; otherwise minerals destabilization can lead to the formation of new thermodynamically stable phases (Gliozzo 2020b);
b. The temperatures typically range between 450 and 800 °C according to the type of clay minerals (He et al. 1995);
c. Ceramic fragments must be finely ground;
d. Not all clays fired at low temperatures develop pozzolanic properties (e.g. when they have a low percentage of the clayey fraction).

Numerous outstanding insights on colour variations, waterproofing and mechanical properties that can be conferred to a mortar/plaster through the addition of specific additives have also been gained through experimental reproductions. For example:

- Centauro et al. (2017) carried out a series of ageing tests adding linseed oil, brown sugar and cow's milk;
- Nunes et al. (2018) investigated the microstructure and composition of lime and metakaolin pastes with linseed oil added and aged for 68 months;
- Zendri et al. (2004) reproduced "cocciopesto" mortars, in order to investigate the reactions responsible for conferring hydraulic properties to these products;
- Nežerka et al. (2014) compared the pozzolanic activity of metakaolin and crushed bricks;
- Işikdağ and Topçu (2013) compared the mechanical strength of mortars, including tile powder, crushed tile, lime and granulate blast furnace slag;
- According to ancient recipes, Salavessa et al. (2013) compared the compressive and flexural strength of plasters prepared with either waste marble or limestone dust;
- Vejmelková et al. (2012b) investigated the mechanical and fracture-mechanical properties, hydric parameters, durability characteristics and thermal properties of several lime–pozzolana composites, using a particular clay shale that is available in the Czech Republic that may have been the natural substitute of volcanic pozzolanas (absent in Central Europe);
- Yang et al. (2009) and Yang et al. (2016) reproduced the mechanism of solidification of sticky rice mortar;
- Ventolà et al. (2011) tested different types of non-hydraulic lime mortars, adding animal glue, casein, nopal and olive oil to develop new compatible products for repairs;
- Lima et al. (2020) investigated the effects of clay mineralogy on drying behaviour, pore size distribution, mechanical strength, vapour adsorption, desorption capacity, water capillary absorption, linear dry shrinkage, cracking and thermal conductivity. They also verified the greater suitability of illitic clayish earths compared with montmorillonitic or kaolinitic ones.

From these studies, it becomes clear that the sampling of archaeological finds and comparative geological materials may not be sufficient and research must be open to experimentation to validate/discard the working hypotheses. In practice, this procedure leads to an increase in samples, analyses and time, but it certainly offers the necessary tools for correct and meaningful interpretations.

Indeed, not all studies require an additional experimental procedure. Knowledge of the properties conferred by the different constituents is already largely known (see also Moropoulou et al. 2005) and must be deeply known in advance. Furthermore, it is also possible to gather useful information from industrial research (therefore, not related to archeometry or restoration). For example, some experimental programs aimed at the development of commercial materials may provide useful information for a better understanding of the properties of ancient mortars and plasters (*e.g.* Grist et al. 2013; Ergenç et al. 2018).

Particular cases

The study of mortars and plasters may also present particular suitability/functionality cases depending on the archaeological context or find in which they are found.

For example, the archaeometric investigations made on the wall plasters of a round installation (Feature 6), found at the Late Natufian village of Nahal Ein Gev II (NEG II, ca. 12,000 cal BP) helped to shed light on the likely use of that structure as one of the very first storage installations (Grosman et al. 2020). Firstly, the authors reconstructed the morphology of the installation as a plastered domed pit. Then, they characterised the walls as an alternation of mud and lime plastering (the latter was absent in other areas of the site). The heating experiments verified that NEG II inhabitants did not use the installation for pyrotechnological processes and the filling was characterised as waste dumped inside after its original use. By integrating all the various information, the authors were finally able to identify the use of "Feature 6" installation for cereals and legumes storage. The discovery is of outstanding importance, as it testified the practice of (short-term) storage just before the Neolithic and subsequent expansion of agriculture. In this case, however, the research question appears somehow reverted: "**can the plaster provide information about the function of the structure it was associated with?**".

Another rather frequent case concerns the possibly contextual use of lime for other purposes. The lime produced for masonry works could have been used in tobacco processing (see, *e.g.* Villaseñor and Graham 2010), in maize processing (see, *e.g.* Katz et al. 1974), in waterproofing amphorae (see, *e.g.* Dorrego et al. 2004) and boats, in glass production and tanning (Foy et al. 2000; Heth 2015), not to mention that calcium oxide was also a thrown weapon (a smokescreen irritating to the eyes similar to modern smoke bombs or pepper stinging sprays) in battles against opponents. Taking a look at other materials found at the same time, for example, in an excavation, may therefore raise new questions (*e.g.* "**what was the organization of lime production in light of the many possible uses of this material?**") and provide interesting information on the versatility of a single material.

Mortars and plasters dating

Mortars dating, including radiocarbon and optically stimulated luminescence (OSL) dating, has been comprehensively addressed in the paper by Ergenç et al. (2021) in this TC. We will, therefore, limit ourselves to discussing some problematic aspects related to the radiocarbon technique on which numerous publications are recently flourishing.

In principle, the method is straightforward. It is based on the extraction and measurement of pyrogenic $CaCO_3$, formed after the incorporation (fixation) of atmospheric CO_2 (which contains ^{14}C) by slaked lime ($Ca(OH)_2$) (Labeyrie and Delibrias 1964).

The binder's age corresponds to that of the mortar's hardening, and, consequently, its ^{14}C date corresponds to that of the mortar from which the sample was extracted. However, this "simple" concept brings with it several practical complications, essentially due to the very individuation and extraction of appropriate samples from the carbonaceous binder.

Calcite may have different origins, not exclusively related to the hardening process; therefore, the possible "contaminants" are many:

a. Geogenic calcite, *i.e.* calcite naturally present in the aggregate, introduced during mixing the aggregate with the binder;
b. Calcite contained in the ashes;
c. Unreacted calcite, *i.e.* limestone not completely transformed during the lime production cycle;
d. Lime lumps, *i.e.* lime that did not react to form calcite after hardening of the mortar;
e. Secondary calcite, *i.e.* calcite formed by recrystallisation beginning from the mortar's use up to the present day.

The whole range of contaminants can move the date far back, as it happens with geogenic calcite (*i.e.* dating refers to the age of formation of the carbonates present in the sand), or far forward, as it happens with secondary calcite.

This problem became immediately evident. Stuiver and Smith (1965) highlighted chronological discrepancies related to ^{14}C dilution due to residues of limestones or calcareous sands used to prepare CaO (Baxter and Walton 1970). Moreover, the method was initially presented as unsuitable for hydraulic mortars whose solidification, in the presence of water, led to the formation of calcium silicates and aluminates (Delibrias and Labeyrie 1965). Subsequently, other researchers have confirmed that this difficulty is mainly due to the poor permeability to atmospheric CO_2, the enduring reactivity of this type of mortar and the presence of lime lumps or phases formed from hydraulic reactions, such as layered double hydroxides (Artioli et al. 2017; Toffolo et al. 2020).

Hence, it soon became clear that these carbonatic residues had to be eliminated before proceeding with the analysis and ensuing attempts "*to refine the technique to make it workable*" involved the mechanical separation of the binder from the aggregate (Folk and Valastro 1976).

The difficulty in obtaining reliable dating of well-dated samples have held back the application of this technique (Malone et al. 1980; Zouridakis et al. 1987; Heinemeier et al. 2010; see also Van Strydonck et al. 2011, 2015 and De Mulder et al. 2014 on lime burials) but did not stop its experimentation and consequent development (Van Strydonck et al. 1982, 1983, 1986, 1989, 1992; Pachiaudi et al. 1986; Hajdas et al. 2017; Hayen et al. 2017).

Sampling and analytical problems are well known to date (Boaretto and Poduska 2013; Ringbom et al. 2014; Toffolo et al. 2020; Urbanová et al. 2020) and partially obviated through the preliminary analysis of the samples. On the other hand, the development of accelerator mass spectrometry (AMS) has made it possible to step forward further, thanks to reducing the sample required for the analysis (less than 1 mg).

Several treatments and analyses are required both for the selection and measurement of the samples and the validation of the proposed date. In the first category, it is possible to list:

- Dry/wet sieving and purification treatments, to separate specific fractions and eliminate part of the contaminants (Addis et al. 2019; Michalska 2019; Ricci et al. 2019);
- Density separation, to separate components, based on their specific gravity (see, *e.g.* Moropoulou et al. 1995; Toffolo et al. 2017b);
- Optical and scanning electron microscopy, to identify the different components of the mortar (authors are too numerous in this case because this represents a quasi-mandatory research step);
- Cathodoluminescence, to identify the different generations of carbonates (see, *e.g.* Heinemeier et al. 1997; Lindroos et al. 2007; Murakami et al. 2013; Toffolo et al. 2019b):
- Fourier transform infrared spectroscopy (FTIR), to individuate geogenic, biogenic and pyrogenic calcites and thus, avoid samples showing extensive recrystallisation (see, *e.g.* Anastasiou et al. 2006; Regev et al. 2010; Poduska et al. 2011, 2012; Toffolo et al. 2019a);
- X-ray diffraction (XRD), to obtain a qualitative and quantitative measurement of both crystalline phases and amorphous fraction. This technique is particularly advantageous to investigate the possible presence of secondary phases, especially those occurring in hydraulic mortars such as the double-layer hydroxide (LDH) minerals (*i.e.* typical products of the pozzolanic reaction; see, *e.g.* Artioli et al. 2017; Ponce-Antón et al. 2018); however, this task can also be aided by the following techniques;
- Solid state nuclear magnetic resonance (SS-MAS-NMR), to identify traces of LDH phases (Richardson et al.

2010), whose contents are below the detection limits of the XRD;
- Differential thermal analysis and thermogravimetric analysis (DTA-TGA), to study the thermal decomposition of the various types of carbonates, based on the experimental evidence that lower temperatures are required to decompose pyrogenic $CaCO_3$ than geogenic $CaCO_3$ (see, *e.g.* Moropoulou et al. 1995; Anastasiou et al. 2006; Toffolo et al. 2017b);
- Sequential dissolution, to provide representative CO_2 samples of the binder, excluding contaminants (Lindroos et al. 2007; Ringbom et al. 2014). This is a fundamental preparation stage of the AMS dating technique (Tubbs and Kinder 1990; Hale et al. 2003; Regev et al. 2017), chiefly because the sequentiality of carbonates dissolution (secondary carbonates → pyrogenic $CaCO_3$ → geogenic aggregates) allows for CO_2 isolation during hydrolysis. The times with which the sequential dissolution takes place are not known with certainty. Therefore, further investigations are needed to verify the reliability of the selected sample and, consequently, of the proposed dating. In an attempt to solve this problem, Toffolo et al. (2017a) and Toffolo and Boaretto (2014) started from the observations made on aragonite and developed a new procedure involving the thermal decomposition (at 500 °C) of pyrogenic calcite.

Among all these methods, there is no one that, taken individually, guarantees a correct selection of the sample. The combined use of two or more techniques is always necessary, even if not necessarily decisive. As clearly demonstrated by experimental and cross-laboratory testing, different or analogous treatments can yield very different dates (Hayen et al. 2017; Michalska 2019). In the former study (Hayen et al. 2017), a detailed compositional analysis of the samples was preliminarily performed and the same four mortars were investigated using different techniques and laboratories. The dates obtained on different parts of the sample and/or with different analytical techniques varied by many centuries between them and there is no possibility of establishing which is the "right" one, except with a combined investigation of a numerically high and compositionally heterogeneous collection of samples (*e.g.* lime lumps, charcoal fragments, specific grain fractions, etc.). In the latter study, Michalska (2019) compared the ^{14}C results obtained for 37 mortars with different compositions and enucleated three groups: (1) air-hardening and slightly hydraulic mortars that can be successfully dated, (2) highly hydraulic mortars affected by rejuvenation effects and (3) mortars with very high content of ageing components, for which "*obtaining the true age of mortar production is not possible with the current state of knowledge*".

In conclusion, despite all the advances made, the method is very laborious and frequently inconclusive. It is not yet able to provide a date that unquestionably relates to the mortar under investigation; consequently, a series of age control investigations are made for comparison with other techniques, which may, however present other types of problems. In the second category, it is possible to list:

- TL and OSL, to compare mortar dating with that obtained on silica-based fragments such as those included in cocciopesto or, specifically, to date their last exposure to light before being put "in the dark" inside a mortar (OSL applications in Moropoulou et al. 2018 and Ergenç et al. 2021);
- Dendrochronology and palaeoenvironmental studies, to compare mortar dating with that obtained on other contextual materials;
- Archaeological stratigraphy for a relative frame.

Despite all these limitations, researchers are certainly not discouraged. Future research will surely solve the raised issues; however, it was necessary to describe the criticalities of the method to not misguide researchers on chronological issues.

Mortars and plasters alteration and degradation

The alteration and degradation of stone materials, mortars and plasters represent a critical issue in the field of restoration and conservation of built heritage (Price and Doehne 2011). Alteration and degradation are often used as synonyms, but they are not. The term alteration refers to a physico-chemical change that generally occurs on the surfaces, for example, a colour change. It can be seen, but hardly can it be linked to something that threatens the integrity and the function of the stone. On the contrary, the degradation of a mortar/plaster represents something that must be taken into account to preserve the material itself. In other words, degradation implies a physico-chemical modification of the materials, which leads to a worsening of its properties and, if it has a great entity, a restoration/conservation intervention is needed. It is important to establish a common language and let the researchers and stakeholders understand each other. For this reason, national and international organizations have set up common glossaries to give the same terminology to alteration and degradation forms. One of the most used is the glossary issued by ICOMOS (Vergès-Belmin 2008). Chemical, physical and biological agents can cause the alteration of mortars and plasters; however, it is common that an alteration/degradation pattern is due to a combination of the abovementioned agents.

The characterisation of the alteration and degradation forms, both on a macroscopic and microscopic level, requires a multidisciplinary approach since chemical, mineralogical, and biological skills are needed (La Russa and Ruffolo 2021). The interventions are planned based on the materials' conservation state (Caroselli et al. 2021). That is why alteration/degradation analyses should be carried out before restoration.

When we deal with archaeometric analysis of mortar and plaster samples, a particular attention has to be paid to their degradation since it can induce wrong interpretations. For example, when the dating of mortars is performed on degraded samples, it is easy to observe a rejuvenation of the ^{14}C age because of the generation of secondary carbonates (Michalska 2019). Generally speaking, the degradation of mortars induces a change in terms of elemental and mineralogical composition, as well as microstructural (*i.e.* porosity, binder-aggregate ratio), which must be taken into account when approaching an archaeometric study based on such parameters.

C. The characterisation of pigments

The first questions concern pigments identification ("**what pigment is it?**") and nature ("**is it a natural or synthetic pigment?**").

Most of the time, answering the first question also provides the answer to the second question. For example, if the answer is "Egyptian blue" or "lime white", we already know from previous studies that these compounds were artificially produced (see Gliozzo and Burgio 2021; Gliozzo and Ionescu 2021; Cavallo and Riccardi forthcoming, and Švarcová et al. 2021 in this TC for futher examples).

Conversely, if the answer is "yellow ochre" or "green earth", the natural origin will be straightforwardly determined (see Mastrotheodoros et al. forthcoming in this TC).

On the other hand, however, even pigments of natural origin, such as ochers, could be processed to vary their colour or consistency. For example, it is well known that goethite-based yellow ochres could turn to hematite-red upon heating.

Still another case is presented by arsenic-based pigments for which, after a certain period, both natural and artificial compounds of realgar and orpiment were in use (Gliozzo and Burgio 2021 in this TC).

Therefore, if the answer to the second question is "natural", we should proceed further with the other research questions. On the contrary, if the answer is "processed" or "artificial", the next question will be aimed at investigating "**how was the pigment processed and produced**".

The provenance of minerals and pigments

The provenance study is often very complicated and not always conclusive. On the one hand, the great heterogeneity of widespread raw materials, such as ochers, makes it difficult to distinguish supply areas that are very distant from each other. On the other hand, the great compositional homogeneity of certain other pigments, such as cinnabar, makes it difficult to discriminate between geographically close resources. In practice, it all depends on the composition and/or the information we have available for a compositional comparison (see the specific contributions, pigment by pigment, of this TC).

The pictorial technique

Understanding "**how was the pigment applied?**" and/or "**how was the surface prepared?**" means investigating the link between the pigment and its support. Each technique corresponds to different archaeometric evidence that must be investigated and unravelled (see Salvadori and Sbrolli 2021 and Murat 2021 in this TC).

For example, some pigments, such as ochers, lend themselves to being used with the *a fresco* technique, while for others, such as those based on lead, a *secco* technique is preferred. Therefore, investigating the painting technique means investigating the palette and technique of the painter and verifying the appropriateness and effectiveness of some techniques compared to others.

Moreover, we know that some pigments, such as those based on arsenic, are particularly sensitive to light and sometimes obliged to adopt particular techniques for their application. Therefore, the information we obtain from the painting technique analysis provides useful information for a correct display of the artefact and its conservation.

Finally, the information that we can obtain from the study of a single artefact or artwork represents a piece of the wider mosaic that describes the evolution of painting techniques and is, therefore, of value in the broad field of art history.

Linking pigments with the social status of the clients

Up to this point, the questions posed to pigments are all technical. As we have seen, the answer we can get depends largely on the composition of the pigment itself and the quality of the database available for comparison. Nevertheless, there are also other questions related to the use of specific pigments that go beyond technological choices. For example, "**is it possible to establish whether the type of pigment used corresponds to a desire for representation?**" or, in other words, "**can the low/high cost of a pigment faithfully reflect the social status of the clients?**".

The use of cinnabar instead of ocher, for example, has often been referred to as a desire for social self-representation on the part of the clients rather than to a different—more vivid and brilliant—rendering of cinnabar compared to ocher. Similar assumptions have been made regarding the use of Egyptian blue and lapis lazuli, although there were no other choices in the ancient world with which to obtain blue.

To avoid possible anachronism, the study of the relationship between pigment and client may help clarify if pigments were used as a demagogic and self-representative tool and cast an eye on the tastes of the time.

Sampling criteria and methods

In the cases that are addressed below, the sampling aims to answer the questions that we have analysed in the previous chapters; however, the most important aspect to keep in mind concerns contextualisation.

Mortars, plasters and pigments constitute a complex system of structural and aesthetic importance that must be contextualised within the various phases of a historic building. Attributing a specific context to the various components is essential for correct sampling for archaeometric purposes. To this end, it is necessary to apply the method of "reading" historical buildings established precisely by the BA to correct extrapolation of the components according to their space–time dimension.

As anticipated above, we must also consider that the research in progress may raise new questions not stated from the beginning. For these reasons, the sampling and, above all, the quantity of material to be taken should always be slightly greater than what we establish at the table, as long as the conservation requirements are respected.

How to obtain a representative sample set of mortars and plasters

The first assumption is that there is no arbitrary number of samples that can be considered representative tout court. Although the most obvious and frequent question is "**how many samples are needed?**", not only is there no straightforward answer but it is worth bypassing this question with another type of question: "**what samples are needed and from where should they be taken?**".

The main characteristics of sampling must be.

a. Representativeness, which is given by stratigraphic, typological and chronological criteria;
b. Functionality, given by sample collection's intrinsic ability to answer established research questions (*e.g.* production technology, provenance, alteration and degradation);
c. Suitability, which concerns the suitability of the material sample to be investigated by the analytical techniques (Gliozzo 2020a, Fig. 4).

A sampling that complies with the representativeness criterion is first and foremost guided by the definition of the construction techniques and the stratigraphic sequence. The parameters to be evaluated are many and the resulting sampling must thus be representative of (1) the types and characteristics of all construction materials (*e.g.* lithotypes/bricks and mortars), (2) the way they were assembled, *i.e.* the construction techniques and (3) the construction phases in which they were employed as delineated by the stratigraphy.

In the first case, a preliminary evaluation is essential to identify macroscopic similarities/differences between building materials. A range of simple tests that can be performed in the field (*e.g.* the effervescence in dilute hydrochloric acid) and the use of a magnifying lens may help this first phase. A preliminary catalogue of the mortars is also essential. It should take into account the broadest range of visible characteristics, such as colour, texture, clasts types, dimensions and distribution, state of aggregation, and the possible presence of cracks and traces of weathering. These characteristics, combined with the parameters used for the definition of construction techniques (*e.g.* structure, equipment, size of elements, percentage of pieces placed at the tip or end, size distribution of segments, the possible presence of plasters), provide the framework on which to set the selection of representative samples based on the first two parameters.

In the third case, the stratigraphic sequence may suggest duplicating the selection of some types of materials or construction techniques, in order to evaluate any changes or persistence on a diachronic basis. For example, the acquisition of two samples of the same lithotype from structures dated to chronologically different periods could demonstrate slight variations in raw materials supply.

Finally, the criterion of representativeness must guide a selection that accounts for all types of materials used, with all construction techniques, for each period considered.

A sampling that conforms to the functionality criterion is guided by the research questions. The representativeness criterion has previously provided useful samples to answer some questions such as "what materials were used for constructing the walls?" but it cannot be considered sufficient. If, for example, we want to know where the workers took the raw materials from, it is frequently necessary to expand the sampling outside the building under investigation. The investigation moves to the surrounding territory to an extent that the researcher can establish, based on geological maps and historical information. Since a detailed characterisation of the lithotypes is required to focus the search for supply basins, it is also foreseeable that sampling will be a multistep activity conducted on several occasions.

For other types of questions, such as those aimed at reconstructing the production technology of mortars, the "representative sampling" already considers numerous features derived from the production processes and their eventual diachronic evolution. Still, only laboratory analyses can decide if the sampled collection is sufficient, or an expansion is necessary. Furthermore, evaluating the production processes and the mortars' physical–mechanical characteristics may require a series of experimental reproductions for which further sampling of raw materials may be necessary.

Similarly, it may be possible to answer questions relating to alteration and degradation through the set of samples obtained with the first criterion. In these cases, however, it is more frequent to carry out targeted sampling. In fact, while unaltered samples are preferred for the characterisation of building materials, altered specimens are also required to evaluate their conservation state.

As far as the dating question is concerned, it was previously explained why the sampling issue is crucial for radiocarbon dating and why it may be necessary to carry out multiple samplings before obtaining a suitable sample for analysis.

In summary, while the "representative sampling" may already provide the necessary material to answer some research questions, the functionality criterion aims precisely to test the completeness of the overall sampling from the point of view of research questions.

The research methodology guides sampling compliant with the third criterion, *i.e.* the suitability. In addition to the example recalled above on selecting suitable samples for radiocarbon dating, another clarifying example concerns visibly altered mortars. By adding all the three criteria together, the sample must be (a) representative of both the original composition and the altered composition; (b) functional to solve research questions, such as those relating to the recipe and those relating to the evaluation of degradation; and, finally, (c) suitable for the analytical techniques that the researcher will use to investigate it.

In this case, it is well known that (1) the thickness of the surface alteration layer is variable, (2) the preparation of a thin section requires a small volume but a suitable surface and (3) an accurate chemical analysis may require a variable volume depending on the chosen technique. Therefore, the sampling methods will be crucial to obtain a suitable sample while observing the fundamental principle of conservation, or in other terms, of minimum sampling.

Archaeometric analyses can move along the path of non-destructiveness or destructiveness. Between these two end-members, different levels of destructivity and accuracy can be achieved (*e.g.* highly destructive, minimally-destructive). Provided the same quality of the results obtained, the most obvious approach would be that of non-destructiveness; however, it should be emphasised that, often, non-destructive techniques cannot provide sufficiently accurate or comparable results to those obtained with destructive methods. Therefore, on the one hand, the researcher risks collecting a lot of quick, cheap and non-destructive data that cannot provide unquestionable answers and, on the other hand, data inaccuracy can lead to a waste of time and money.

Therefore, the choice of analytical techniques should first find a balance between the accuracy requirements of quality research and the conservation needs and then guide sampling to the selection of suitable samples. In this regard and based on what has been said previously, it is good to underscore that sampling does not necessarily involve the physical taking of material. In the case of non-destructive techniques, sampling may simply address the selection of the analysis spots.

To conclude, while <u>before</u> proceeding with sampling, it is necessary to collect all available data from any field of study; <u>during sampling</u>, it is mandatory to properly record and document all phases and, <u>after sampling</u>, it is required to remain open and flexible to answer new questions that may arise. Before deciding and after taking the samples, the overall research should record the various actions and use a critical survey. This survey should be equipped with specific signs that provide a cartographic mapping functional to sampling. Regardless of the use of new technologies for the graphic representation of historical buildings, we should create standardised protocols to record sampling through, for example, a precision topographic positioning.

How to obtain a representative sample set of pigments

A sampling of pigments is carried out primarily based on colours. In this way, by counting the number of colours visible in the painting, the number of samples to be investigated is also obtained. In large-scale paintings such as wall paintings, the sampling of a single sample per colour may prove insufficient. Therefore, it will be necessary to carry out multiple samplings for each colour to evaluate any changes. This reasoning is valid both in the case of natural and artificial pigments, as it provides the possibility of identifying different technologies and/or provenances.

In complex structures with several decoration phases, the stratigraphic sequence of superimposed layers provides the tool for guiding sampling.

When the analysis involves non-destructive instrumentation, it is undoubtedly desirable to consider a large increase in the spots to be analysed. When, on the other hand, the research question requires the use of destructive investigations, the number of samples will adapt to the minimum necessary and will possibly be taken in the least exposed portion.

For the study of the painting technique, there is an additional difficulty, as the analysis needs to go beyond the pictorial film, in order to reach the preparatory layer. In this case, the researcher may choose either to analyze the gaps or take a sample that shows the cross-section. The first method does not necessarily damage the artefact but is not always resolutive, while the second has the advantage of ensuring an exhaustive response but is destructive. The researcher must, therefore, choose the first or second method based on conservation needs. A third option is to be considered when the analysis concerns mobile artwork and consists of instruments with a higher penetration capacity than conventional techniques, such as Raman or portable X-ray fluorescence.

In any case, while it is not acceptable that the availability/absence of specific instrumentation or funds decides the analytical protocol, the diversity offered by the artworks and their needs (wall painting/mobile artefact, alteration issues and conservation needs, the impossibility of transferring the find to the laboratory, etc.) always guide the selection of the most suitable sampling and analytical techniques.

Concluding summary of key concepts

The archaeometric study of mortars, plasters and pigments provides information about:

- The origin of the raw materials used as aggregate, additives or pigments or as raw material for the production of lime;
- The production technology of lime, mortars, plasters and pigments as well as the painting technique;
- The state of alteration and/or degradation of both buildings and paintings, as well as the yield of products for restoration;
- The chronology of some components of the mortar with different levels of likelihood depending on whether they are siliceous materials, datable through TL-OSL (relatively accurate results), or carbonaceous, datable through radiocarbon (results not trustable as received but to be verified).

From the first series of questions addressing the "study's object" directly, the integration of laboratory results with the geological knowledge of the territory provides information about:

- The exploitation of the territory itself over the centuries or in a specific period;
- The relationship between knowledge of the raw materials available and their exploitation for production purposes;
- The tools to understand better certain technical choices relating to the use of certain materials compared to others.

The integration with the stratigraphy allows the observation of changes during the same phase and/or specify the chronological seriation and/or provide indirect information on the workers and the construction site's organisation ("cantiere").

The integration with other historical information on economic trades sheds light on the commercial dynamics and on the routes of trade, as well as on the status of the clients and the functional destination of some buildings.

Finally, the integration with technological knowledge can add useful pieces of information to reconstruct the history of technologies, in a diachronically and geographically wide perspective.

The main assumption that derives from these examples must be that of a plurality of questions that follow one another, rather than a rigid and pre-set list of questions. This last method is functional to the start of the research but must then open up to the unexpected how, when and why that arise during the advances/progress of the research.

Consequently, sampling must remain flexible and, subject to conservation requirements, must answer both pre-set and unexpected questions. Mortar sampling should be guided by three main parameters: representativeness, functionality and suitability. Pigment's sampling should be based on colours and, when applicable, guided by stratigraphy.

Non-destructive methodologies are certainly an advantage in the study of pigments. Still, they cannot always provide sufficiently accurate measurements and, being superficial analyses, they can mislead due to surface alterations. On the other hand, destructive methodologies are almost always preferable (if not indispensable) in the study of mortars and plasters. A reasoned sampling can minimise the damage.

Finally, three important aspects should be born in mind. First, the results obtained on a single sample or a poorly selected sample collection cannot be extended to entire rooms or buildings.

Secondly, mortars/plasters that are compositionally very different from each other may show comparable performance, at least adopting ancient standards. We should avoid anachronisms putting ourselves in the shoes of people who do not necessarily think with our parameters. Leaving aside qualitative evaluations (that we cannot estimate except with current criteria) is likely the best approach in most cases.

Thirdly, there is an unquantifiable level of randomness that we cannot in any way reconstruct; therefore, we must be careful in not falling into attractive overinterpretations. Some choices or variations of the "products" we analyse should not necessarily be interpreted as a decline in quality or an evolution of the technique. They may (simply) represent the best choice at that given moment.

Authors' contributions Building Archaeology by AP; Mortars and plasters alteration and degradation by MFLR; Concluding summary of key concepts AP-MFLR-EG; other contents by EG. English professional revision provided by MFLR.

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Declarations

Conflict of interest The authors have no conflicts of interest to declare.

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