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## A POTTERY BIOGRAPHY: CONSIDERING THE ENTIRE LIFECYCLE OF A POT IN ORGANIC RESIDUE ANALYSIS

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# A pottery biography: considering the entire lifecycle of a pot in organic residue analysis

*Biographie d'une céramique : prendre en compte l'ensemble du cycle de vie d'un récipient lors de l'analyse des résidus organiques amorphes*

Léa DRIEU<sup>a</sup>

**Abstract:** Over the past thirty years, the study of the exploitation of natural substances by ancient societies has been a major focus of organic residue analysis in ceramics. However, the mechanisms controlling the absorption and preservation of organic matter within the walls of pottery at all stages of a vessel's lifecycle are still very poorly understood. Organic and mineral composition of the paste, surface treatments, porosity, environmental conditions of disposal of the vessel are likely to influence the organic signal preserved over time. Storage conditions of the archaeological material can also affect the preservation of organic matter after excavation. Recent studies are beginning to address these issues, and the first results are already refining the interpretations of organic residue analysis and opening up new fields of investigation, particularly with regard to the study of the ceramic manufacturing *chaîne opératoire*, burial environments and potential contamination.

**Résumé :** Depuis une trentaine d'années, l'exploitation des substances naturelles par les sociétés anciennes a été le principal objet des analyses de résidus organiques amorphes dans les céramiques. Les mécanismes régissant l'absorption et la préservation de la matière organique au sein des récipients à toutes les étapes du cycle de vie d'un récipient restent cependant encore très mal compris. Composition organique et minérale de la pâte, traitements de surface, porosité, conditions environnementales d'abandon du récipient, conditions de stockage du matériel archéologique sont ainsi susceptibles d'influencer le signal organique détecté lors de l'analyse. De récentes études commencent à explorer ces questions, et les premiers résultats permettent d'ores et déjà d'affiner les interprétations de l'analyse des résidus organiques amorphes et d'ouvrir de nouveaux champs d'investigation, notamment en termes d'étude de la chaîne opératoire céramique, des environnements de dégradation et des potentielles contaminations.

**Keywords:** residue, ceramic, ceramic paste, organic analysis, clay.

**Mots clés :** résidu, céramique, pâte céramique, analyse organique, argile.

## 1. INTRODUCTION

Organic residue analysis, a now well-established field, has revealed the content of ceramics from a wide variety of contexts, from prehistoric to modern times all over the world. These studies were made possible by the properties of pottery, a receptacle at different scales. First of all, it is a containing utensil, central in many societies, and used in a very wide range of human activities (exploitation of natural substances, food, technical activities, trade, etc.). In addition,

as a porous material, pottery records the signal of this activity, in the form of organic molecules and protects it, at least partially, from degradation. Finally, ceramic is a material strong enough to be well preserved in most archaeological contexts. As a result, organic residues in pottery were mainly oriented towards pottery rather than other types of artefacts, and focused mainly on their use.

Pottery, this remarkable tool for studying ancient human societies is however much more complex than its simple

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role as a container. The lifecycle of the ceramic object begins well before its first use, through the choices made during the manufacturing *chaîne opératoire* and continues after its disposal, through burial, excavation and analysis (Figure 1).

The chemical signal extracted from the ceramic potsherd in organic residue analysis is the result of all the processes that successively occurred since the selection of the raw material by the potter. While the first publications that established the basis of organic residue analysis generally mention these parameters (influence of the paste mineralogy, potential surface treatment, risk of contamination by the sediment, etc.), the quest to explore the content of pottery has somewhat neglected the complexity of the ceramic object. Now that organic residue analysis is well established as an archaeological science, we observe a new interest in these issues, and recent studies are beginning to explore again the parameters that impact the organic signal, beyond the use of the pots (e.g. Admiraal *et al.*, 2020; Drieu *et al.*, 2019; Goldenberg *et al.*, 2014; Reber *et al.*, 2018). For once, the present paper will focus on all the stages of the ceramic lifecycle, trying to highlight as many parameters that influence the absorption and preservation of organic matter at all stages of the life cycle of the vessels. Since these aspects remain difficult to untangle at the moment, the aim is to synthesize these data from the most pioneering to the most recent literature, and complete them with original experimental and archaeological data, which, although preliminary, provide new insights to be considered.

## 2. THE HIDDEN SIDE OF THE CERAMIC CHAÎNE OPÉRATOIRE: ORGANIC MATTER RELATED TO MANUFACTURING

### Composition of the raw clay paste

Organic matter is a natural component of clay soils, present in variable quantities depending on the type of deposit (Gabasio *et al.*, 1986; Johnson *et al.*, 1988; Rice, 1987, p. 37). This amount of carbon related to decomposed or decomposing organic matter is sometimes large enough to bias radiocarbon dating (e.g. Gabasio *et al.*, 1986; Tankersley *et al.*, 1987). Organic materials are also sometimes added voluntarily by the potter, often to modify the properties of the raw paste or of the fired pot (e.g. modify the plasticity of the paste, control shrinkage during drying, create a porous vessel, etc.; Gosselain, 2002, p. 36; London, 1981; Maritan *et al.*, 2006; Oberweiler, 2006; Schiffer and Skibo, 1987; Sestier *et al.*, 2011). In addition to the common mineral additions, temper can be of plant or animal origin. Such

add-ons are easily identified in a vessel when of macroscopic dimensions (e.g. stem fragments, mosses, seeds, fleece fibres, etc.). They are likely to contribute to the organic matter content of the paste, for example as waxes, fats or terpenes. Ethnographic research attests to practices that also involve amorphous organic substances: manure, which includes a liquid fraction (e.g. Glick, 1936; London, 1981), blood or marine oils (Harry *et al.*, 2009; Rice, 1987, p. 119), beeswax (Vellard, 1939, p. 81, 98) and plant extracts (Glick, 1936; Shepard, 1956, p. 52) are mentioned.

Studies of the content of ceramics using organic residue analysis are based on the fundamental assumption that all organic matter related to manufacturing is reset during the firing of the vessel. However, several studies mention that, depending on the parameters involved in the firing process, part of the organic matter could be preserved (Gabasio *et al.*, 1986; Johnson *et al.*, 1988; Rice, 1987). Recently, several experiments have been set up to test this hypothesis (Admiraal *et al.*, 2020; Reber *et al.*, 2018). Clay sherds naturally rich in organic matter or enriched have been fired at different temperatures, in muffle furnaces or in fireplaces. During these experiments, all the organic matter in the raw paste was proved to be degraded by the firing process, which seems to justify the initial assumptions of “resetting” the ceramic. However, it is important to note that, due to the novelty of this research topic, only few variations in parameters have yet been tested. New firing experiments involving clays that are more or less rich in organic matter suggest that lipids can be preserved under certain conditions of temperature, firing time and type of clay paste (Drieu, 2017). Future experiments should be developed according to a specific archaeological context, for which information is available on the type of clay paste and firing atmospheres, and even, if possible, on firing structures and temperatures.

### Surface treatments

Ethnography and archaeology demonstrate that the application of organic matter on surfaces and its thermal transformation is a common practice for the treatment of ceramic surfaces. Application can be made on raw surfaces, directly (Bollong *et al.*, 1993; Harry *et al.*, 2009), or indirectly (e.g. furbishing with animal hair which deposits grease on surfaces; Lepère, 2014). Immediately after firing, the still hot surface of the vessels can also be exposed to organic matter via various processes (soaking, rubbing, spraying, smudging; e.g. Gosselain, 2002, p. 183-198; Longacre *et al.*, 2000; Rice, 1987, p. 158, 163-164; Skibo, 2013, p. 80). Ethnography has highlighted the great diversity of organic materials (of plant or animal origin, in liquid or solid form)

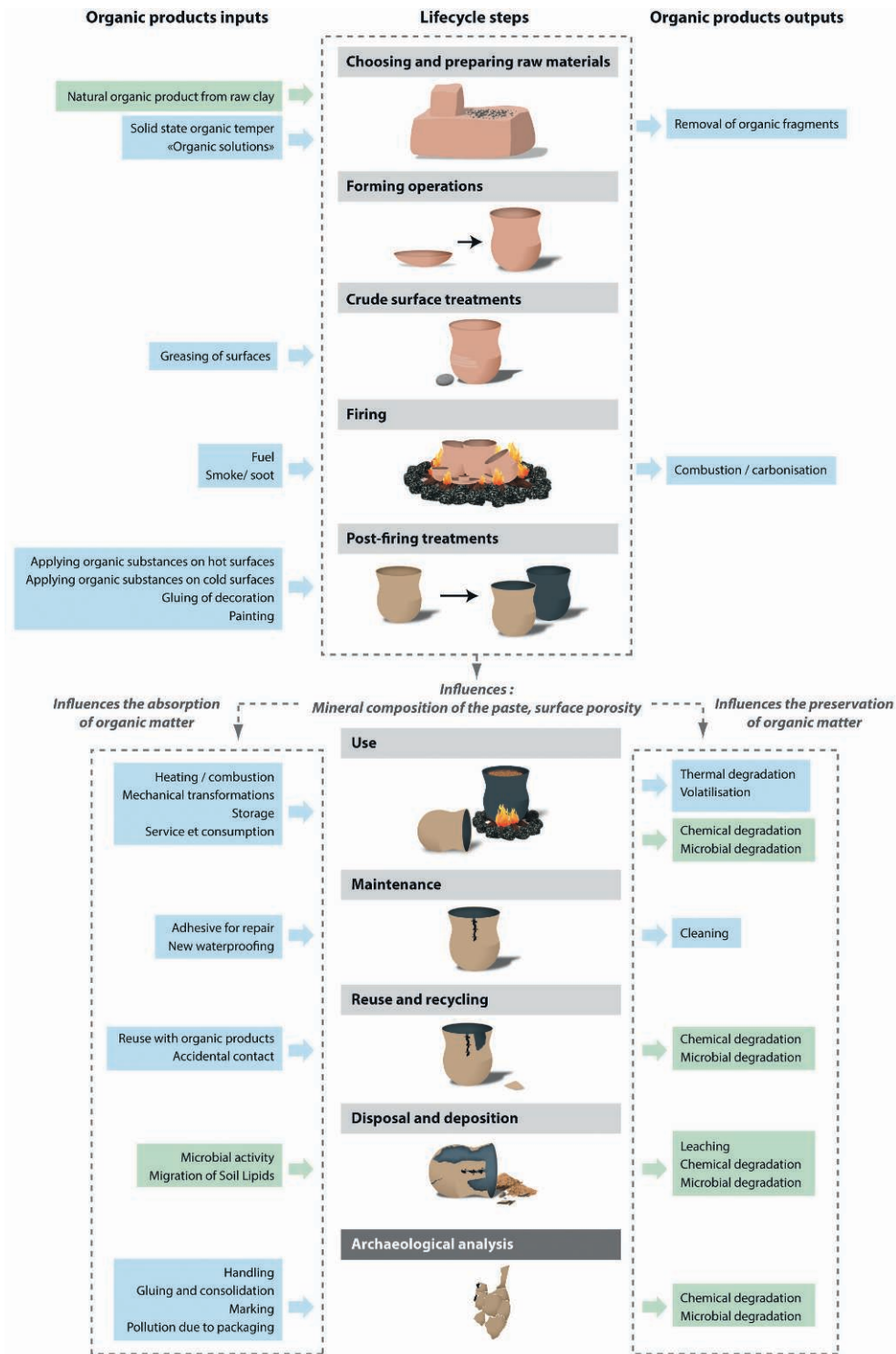


Figure 1: Inputs and outputs of organic matter during the ceramic lifecycle. In blue, inputs and outputs from human activities, in green, inputs and outputs from natural processes (adapted from Gabasio *et al.*, 1986; Oudemans & Boon, 1991; Roffet-Salque *et al.*, 2017; Stacey, 2009).

Figure 1 : Apports et pertes de matière organique au cours du cycle de vie d'un récipient. En bleu, les apports et les pertes liés à des activités anthropiques, en vert, les apports et les pertes liés à des processus naturels (adapté d'après Gabasio *et al.*, 1986 ; Oudemans & Boon, 1991 ; Roffet-Salque *et al.*, 2017 ; Stacey, 2009).

that can be used in this type of process, which usually generates a more or less homogeneous blackening of the surfaces and the appearance of dark margins in the edge of the walls (Drieu *et al.*, 2019; Martineau & Pétrequin, 2000). Two studies of high-temperature surface treatments using organic residue analysis have shown that the organic material brought into contact with hot surfaces is not systematically completely degraded, and is absorbed within the ceramic wall (Diallo *et al.*, 1995; Drieu *et al.*, 2019). There is therefore a risk of confusion between the chemical signal of the surface treatment and of the actual uses of the containers. In addition, it has been proven on several occasions that some surface treatments, particularly high temperature treatments, only partially seal the surfaces (Drieu *et al.*, 2019; Gosselain, 2002, p. 183; Romanus *et al.*, 2009; Schiffer, 1990). The organic signal extracted from archaeological ceramics is therefore potentially the result of the accumulation of the surface treatment and the successive uses.

After cooling, the surfaces of the containers may also be treated with an organic material, to modify the physical properties (waterproofing, increasing mechanical strength) or the colour of the container. Resins, pitch, tar, beeswax, and bitumen are frequently mentioned (e.g. Connan *et al.*, 2004; Rice, 1987, p. 163; Rebert & Hart, 2008; Roffet-Salque *et al.*, 2015). The use of fatty substances such as dairy products, animal adipose fats, fish or vegetable oils, and blood is also documented (e.g. Gosselain, 2002; Harry *et al.*, 2009; Mahias, 1994). In some cases, the treatment of the internal surface is also the first use of the vessel (e.g. Gosselain, 2002, p. 191; Harry *et al.*, 2009).

In light of the wide distribution of surface treatments in ethnographic studies, it is very likely that such processes have also been used by ancient potters. This hypothesis should therefore be systematically considered when interpreting organic extracts. It is often mentioned when non-edible substances (pitch, resin, beeswax, etc.) are identified in archaeological vessels (e.g. Faraco *et al.*, 2016; Hayek *et al.*, 1991; Heron *et al.*, 1994; Reber & Hart, 2008; Regert *et al.*, 2001; Salque *et al.*, 2013). However, alternative hypotheses must be considered when interpreting the presence of these substances: ceramics may have been used to store these products, to mix them to produce adhesives, etc. An opposite scenario is that of edible substances, which, when identified, are immediately interpreted as remains of the ceramic content, very rarely mentioning the possibility that they may have been used as surface treatments. In particular, mid-chain ketones has proven to be formed during high-temperature surface treatments and should not be systematically considered as cooking pot evidence (Drieu *et al.*, 2019).

## Firing of the vessels

During the firing of the container, the fuel of vegetable or animal origin, is often in direct contact with the containers, which can result in the penetration of more or less degraded organic matter (resin, grease, etc.) into the walls (Gabasio *et al.*, 1986; Reber *et al.*, 2018). Smoke condensates are also likely to be absorbed in the ceramic matrix under certain firing conditions, as demonstrated by a recent study (Reber *et al.*, 2018). The presence of certain compounds, for example diterpenes (derived from coniferous resin and very often identified in archaeological pottery), must therefore be interpreted with caution.

## 3. PROPERTIES OF THE CERAMIC MATRIX AND THEIR IMPACT ON THE ABSORPTION OF ORGANIC MATTER THROUGHOUT THE LIFE CYCLE

### Mineralogical composition of the paste

The mineralogical composition of the paste plays a role in the formation of many thermal transformation markers, e.g. mid-chain ketones and  $\omega$ -(*o*-alkylphenyl)alkanoic acids. The presence of certain metal cations in the paste, by catalysing the reaction, promotes the formation of these markers (Evershed *et al.*, 2008; Raven *et al.*, 1997). The absence of these markers may therefore be related, not to the absence of heating of the content of the vessel, but to the composition of the paste itself.

### Porosity of the paste

The absorption of organic matter by the ceramic matrix is influenced by its porosity in general, and by its surface porosity in particular (Correa-Ascencio & Evershed, 2013; Evershed, 2008a; Stern *et al.*, 2000). Some authors suggest that surface burnishing may reduce the absorption of organic matter when the vessel is used (e.g. Correa-Ascencio & Evershed, 2013; Debono Spiteri, 2012). Indeed, during this type of treatment, the compaction of the clay material causes a migration of the finest particles towards the surface and sinks the largest inclusions (Echallier, 1984).

Preliminary experiments, consisting in impregnating with hot milk small tiles that have undergone various treatments of raw surfaces, have been set up in order to explore this question (Figure 2b and Table S1). Kneading clay paste seems to favour the absorption of organic matter in ceramics (Figure 2c and Table S1), with on average 70% more lipids absorbed in the experimental tiles of kneaded paste than in

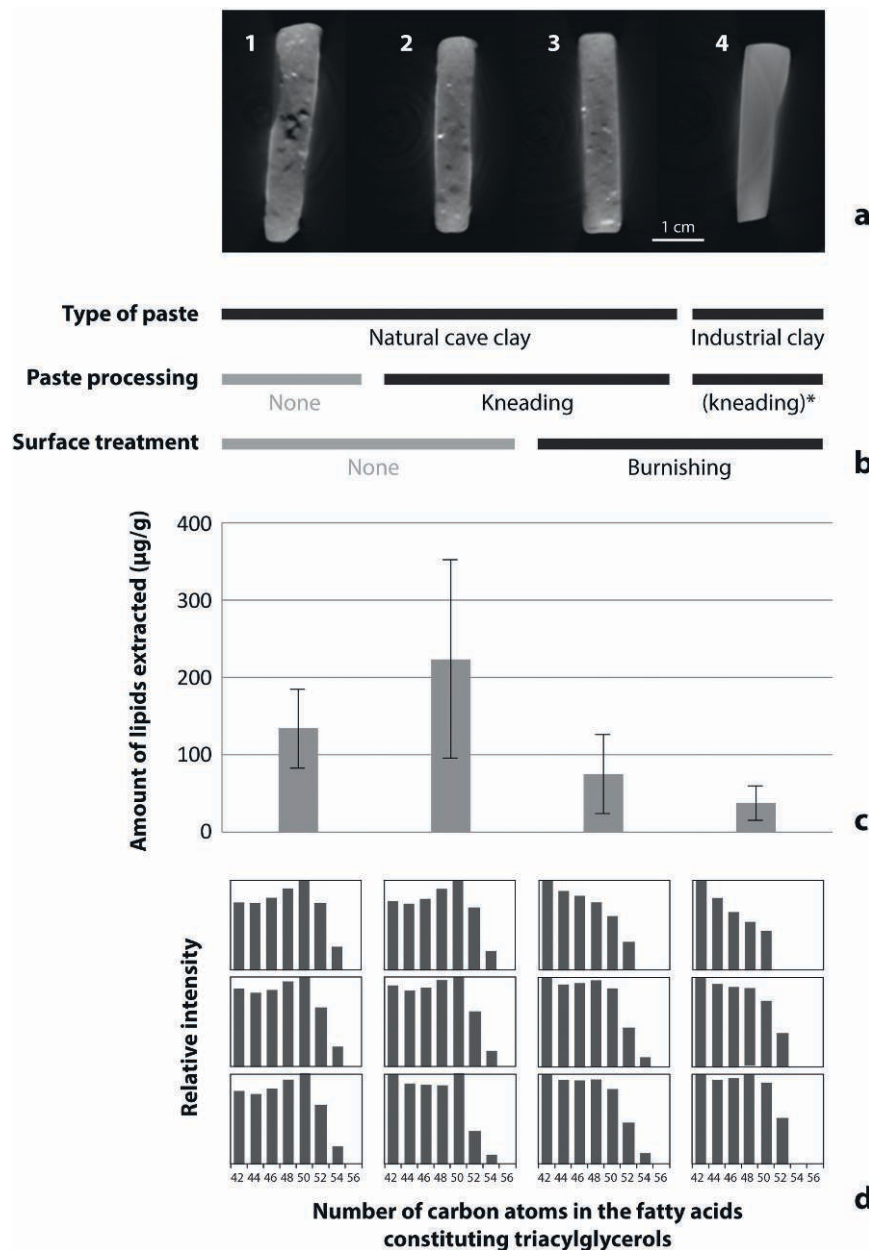


Figure 2 : Experimental tiles to test the effect of porosity on organic matter absorption. 1) cave clay, without paste processing or surface treatment; 2) cave clay, kneaded, without surface treatment; 3) cave clay, kneaded, burnished; 4) industrial clay, burnished. a) Examples of slices of the experimental tiles obtained by X-ray tomography (CT-Scan). Acquisitions in collaboration with D. Pisani (Institut Biologie Valrose, Nice); b) tile preparation methods; \*: the kneading having been carried out mechanically during the production of the clay blocks, no additional manual kneading has been carried out; c) average amounts of lipids extracted from the tiles. The error bars correspond to  $\pm$  the standard deviation on the three experimental replicas; d) triacylglycerol profiles in the tiles. The three graphs correspond to the tile triplicates. The quantitative data are available in the annexes (Table S1).

Figure 2 : Plaquettes expérimentales pour tester l'effet de la porosité sur l'absorption de la matière organique. 1) argile de grotte, sans travail de la pâte ni traitement de surface ; 2) argile de grotte, malaxée, pas de traitement de surface ; 3) argile de grotte, malaxée, brunie ; 4) argile industrielle, brunie. a) Exemples de tranches de plaquettes expérimentales obtenues par tomographie aux rayons X (CT-Scan). Acquisitions effectuées avec la collaboration de D. Pisani (Institut Biologie Valrose, Nice) ; b) méthode de préparation des plaquettes ; \* le malaxage ayant été effectué mécaniquement au moment de la fabrication des pains d'argile, aucun malaxage manuel supplémentaire n'a été effectué ; c) quantités moyennes de lipides extraites des plaquettes expérimentales. Les barres d'erreur correspondent à  $\pm$  l'écart type sur les trois répliques expérimentales ; d) profils de triglycérides dans les plaquettes expérimentales. Les trois graphiques correspondent aux trois répliques expérimentales. Les données numériques sont disponibles dans les annexes (Table S1).



those in which the paste has not been processed, all other parameters being equal. By eliminating air bubbles, kneading produces small and homogeneous pores (Rice, 1987, p. 119) that appear to be ideal for trapping organic matter, unlike the very large pores identified in unprocessed pastes (Figure 2a and Table S1). The burnishing of surfaces seems to restrict the absorption of organic matter in ceramics (66% less lipids absorbed on average in kneaded and burnished tiles than in simply kneaded tiles). Burnishing may also slightly modify the molecular profile of the heaviest molecules – the triacylglycerols in these experiments (Figure 2d and Table S1). The heaviest triacylglycerols ( $T_{50}$ ,  $T_{52}$  and  $T_{54}$ ) are extracted in smaller quantities from burnished tiles, compared to tiles that have not undergone surface treatment. Although it is unlikely that burnishing would have an effect on triacylglycerol-sized pores, this result can be explained by the presence in milk of triacylglycerol spherulites or globules that are several tens of microns in size (Goudédranche *et al.*, 2000; Sato & Ueno, 2011), and therefore too large to penetrate the porosity reduced by the surface treatment. These preliminary results will have to be further developed to try to understand what factors may constrain the absorption of high molecular weight compounds contained in fats (triglycerides, but also wax esters), as the distribution of these molecules, when preserved, is one of the main criteria for the characterisation of the natural origin of organic materials.

More generally, assessing with some certainty the effect of surface treatments on the porosity of the ceramic and on the absorption of organic matter remains essential, as a large number of pre and protohistoric ceramics show evidence of this type of treatment. The effect of slip, for example, has not, to the best of our knowledge, yet been investigated.

#### 4. THE MULTI-PARAMETER TRANSFORMATIONS OF THE ORGANIC MATTER ABSORBED IN POTTERY AFTER DISPOSAL OF THE POT

##### Influence of the paste on the degradation mechanisms

The mineralogical composition of the paste (clay minerals and non-plastic inclusions) determines the type of interactions that are formed with organic matter (e.g. hydrogen bonds, dipole-dipole interactions, ion-dipole or electrostatic interactions; Correa-Ascencio & Evershed, 2013; Craig *et al.*, 2004; Garnier & Valamoti, 2016; Goldenberg *et al.*, 2014; Matlova *et al.*, 2017) and the variable strength of these bonds influences the level of degradation of organic matter. It should be noted that the mineral composition

may be slightly modified during the use of the vessel or due to taphonomic processes (addition of certain cations, for example, via cooking water or stream water; Correa-Ascencio & Evershed, 2013).

However, the role of ceramic paste is rarely considered in the analysis of organic extracts from archaeological vessels. To the best of our knowledge, only one study has examined the preservation of organic matter according to the mineralogical nature of the paste (Goldenberg *et al.*, 2014), and has shown that unsaturated lipids preferentially bind to calcium-rich inclusions, such as calcite. However, the absorption of saturated lipids, much more commonly identified in archaeological pottery, according to the mineralogy of the paste, has not yet been investigated. In addition, very few publications in the field of organic residue analysis mention the mineralogical or chemical composition of the ceramic paste of the samples being studied, making it difficult to synthesize on a large scale its actual impact on the preservation of organic matter.

##### Influence of environmental context on the degradation of organic matter

Low temperatures in cold or frozen climates limit hydrolysis and microbial activity (Aillaud, 2001, p. 145; Eglinton *et al.*, 1991), resulting in very good preservation of organic matter in archaeological ceramics. Arid environments are also very favourable to the preservation of organic matter. In this type of context, organic matter is mainly subject to oxidation mechanisms because low hygrometry limits both hydrolysis reactions and the activity of microorganisms (Copley *et al.*, 2005; Eglinton *et al.*, 1991; Evershed, 2008b; Regert *et al.*, 1998). Conversely, Mediterranean-like climates, characterised by a hot and dry climate that alternates with periods of heavy rainfall, is thought to be favourable to the degradation of organic matter, since they favour both hydrolysis and oxidation mechanisms (Evershed, 2008b), but the preservation has proved to be very good at a number of sites (e.g. Drieu *et al.*, 2018; Spiteri *et al.*, 2017).

Underwater sites, as well as wetland and peatland environments, generally show excellent preservation of organic matter. Despite the occurrence of hydrolysis and reduction reactions, the low oxygen availability limits both degradation by microorganisms and oxidation (Berstan *et al.*, 2004; Den Dooren De Jong *et al.*, 1961; Eglinton *et al.*, 1991; Killops & Killops, 2009). It also seems that cave sites offer a much more favourable context for the preservation of organic matter than open-air sites, regardless of the type of substrate and soil, because they are less exposed to seasonal variations (temperature and hygrometry) and undergo

fewer leaching processes (Debono Spiteri, 2012, p. 267; Decavallas, 2011, p. 159).

Soil properties also have a significant impact on the degradation of organic matter. A number of data suggest that lipids absorbed in ceramics are better preserved in acidic soils. Indeed, experiments on the degradation of organic matter in sediment have shown that under aerobic conditions, the optimum degradation occurs at a neutral pH (pH 6.5; Delaune *et al.*, 1981; Moucawi *et al.*, 1981). When

the pH is more basic (pH 8) or more acidic (pH 4-5), the organic matter degrades much more slowly (Delaune *et al.*, 1981). This result is mainly to be explained because neutral soils are the most favourable to the development of microorganisms responsible for part of the degradation mechanisms (Hita *et al.*, 1996; Moucawi *et al.*, 1981). It should be noted that soil pH could also impact chemical degradation mechanisms: in basic sediments acids exist in the form of soluble salts, which are therefore more easily

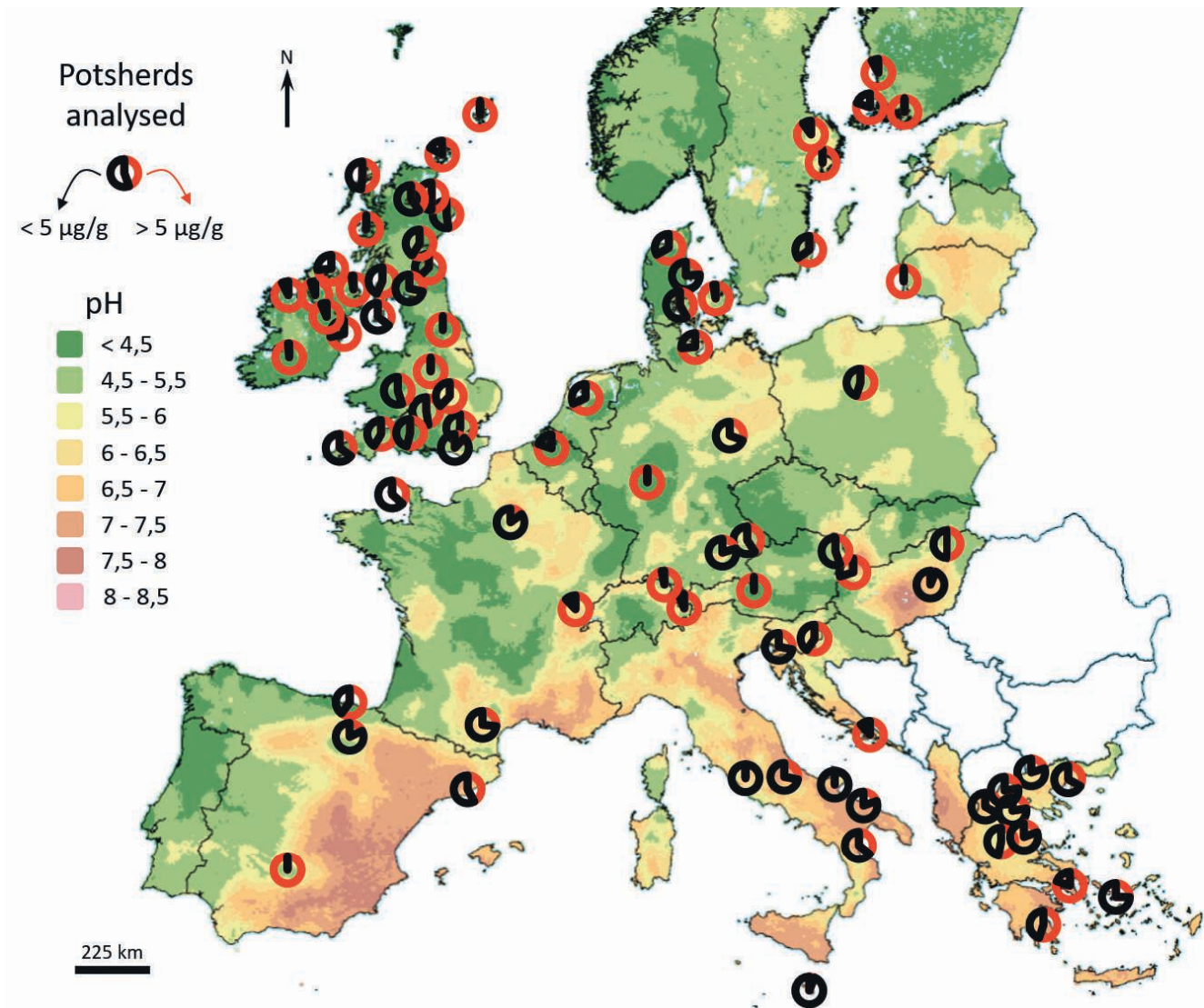


Figure 3 : Preservation of organic matter in ceramics from pre and protohistoric sites as a function of soil pH. Map based on data available on the website of the European Commission's Scientific Service [<http://esdac.jrc.ec.europa.eu/content/soil-ph-europe>]. Data on organic matter extracted based on a synthesis of published literature completed in June 2017 (Drieu, 2017).

Figure 3 : Préservation de la matière organique dans les céramiques de sites pré- et protohistoriques en fonction du pH du sol. Carte d'après les données disponibles sur le site du service scientifique de la Commission Européenne [<http://esdac.jrc.ec.europa.eu/content/soil-ph-europe>]. Les données sur les lipides extraits sont basées sur une synthèse de la littérature publiée, réalisée en juin 2017 (Drieu, 2017).

eliminated by leaching processes (Evershed *et al.*, 1997; Oudemans & Boon, 2007), while an acid sediment might promote the hydrolysis of triacylglycerols (Evershed, 1990; Evershed & Connolly, 1988).

In general, the influence of soil pH on the preservation of organic matter is reflected in the extraction yields of organic matter from pre and protohistoric European pottery (Figure 3), with higher yields in northern Europe where soils are very acidic. This map should, however, be taken with caution, on the one hand because anthropogenic factors can influence soil pH, particularly since the beginning of the industrial era (spreading fertilizers, pesticides, increasing rainfall acidity, etc.). On the other hand, the data are represented at the continent scale and soil pH can show significant variations at the local scale that cannot be reflected in this map (Fabian *et al.*, 2014). In order to investigate this question further and to study the correlation between soil pH and organic matter preservation in pottery, it would be valuable to measure the pH of the sediments surround-

ing the analysed potsherds whenever possible (Drieu *et al.*, 2018, 2019; Heron *et al.*, 1991).

### Lipid inputs by contamination of the surrounding sediment

One of the first steps in the development of biomolecular archaeology was to verify that the sediment surrounding the potsherds did not cause any contamination. Most of the combined analyses of potsherds and sediments that have been carried out have shown that this contamination is minimal: on the one hand, the quantity of lipid in potsherds is very often much higher than in sediments, and on the other hand, molecular profiles are generally very different from each other (Aillaud, 2001, p. 31-57; Evershed *et al.*, 1994; Heron *et al.*, 1991). This is mainly because the lipids generally present in sediments (linear long-chain fatty acids, alkanes and alcohols, derived from vegetable waxes or microbial activity) are highly hydrophobic, which limits their

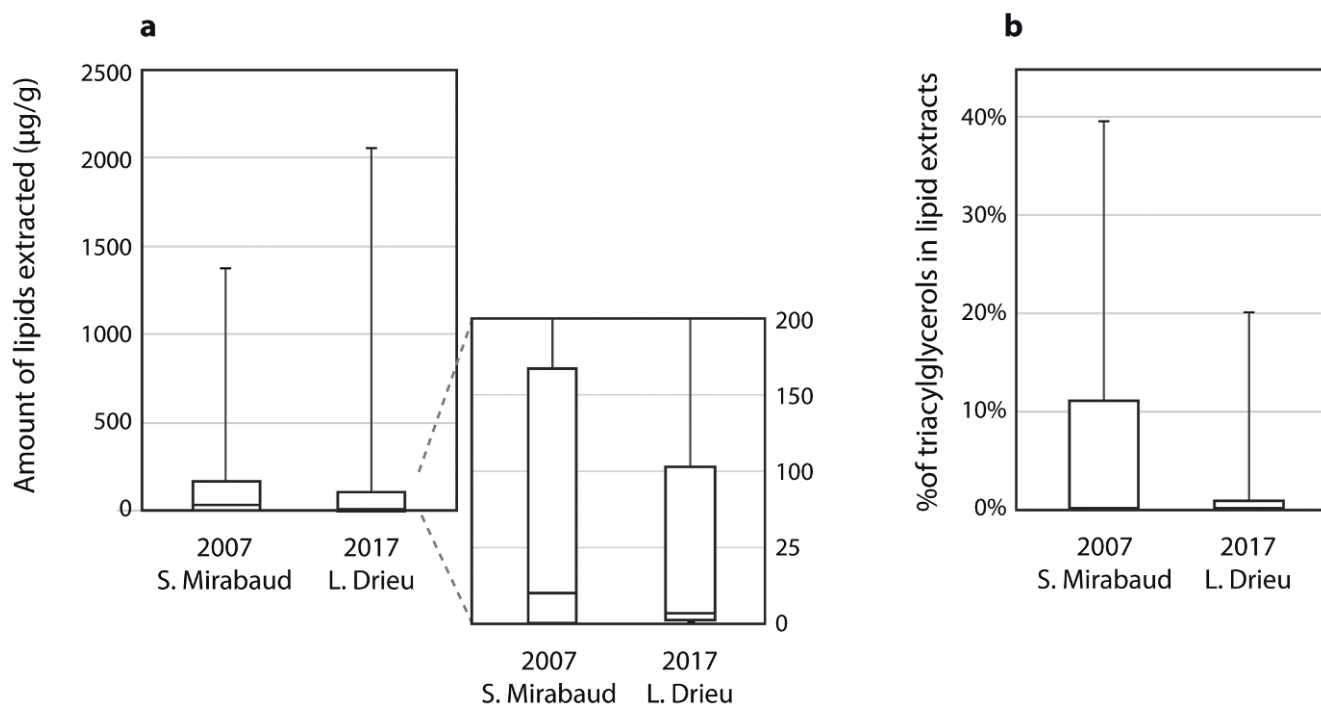


Figure 4: Comparison of lipid preservation in Clairvaux XIV ceramics according to the analysis campaigns: thesis by S. Mirabaud (Mirabaud, 2007) and L. Drieu (Drieu, 2017; Drieu *et al.*, 2020). a) Quantity of lipids extracted from ceramic vessels; b) percentage of triacylglycerols in each lipid extract. The box plots represent the minimum and maximum, as well as the median and the 1<sup>st</sup> and 3<sup>rd</sup> quartiles of the dataset.

Figure 4 : Comparaison de la préservation des lipides dans les céramiques de Clairvaux XIV selon les campagnes d'analyse : thèse de S. Mirabaud (Mirabaud, 2007) et L. Drieu (Drieu, 2017 ; Drieu *et al.*, 2020). a) quantité de lipides extraits des récipients céramiques ; b) pourcentage de triglycérides dans chaque extrait lipidique. Les diagrammes à moustache représentent les minimum et maximum, ainsi que la médiane et les 1<sup>er</sup> et 3<sup>e</sup> quartiles du jeu de données.

transfer from sediment to ceramic and vice versa (Aillaud, 2001, p. 31; Heron *et al.*, 1991).

Despite this, it is still important to remove the potentially contaminated surface of the potsherd (using a scalpel or microdrill) before analysis, and as much as possible, to analyse the sediments in which the potsherds have been found in order to minimise misinterpretation. Indeed, some short chain lipids, which are more soluble in water than their long chain homologues, are more likely to be carried by water. This is true of the linear alcohol  $C_{18}OH$ , which remained difficult to interpret in archaeological potsherds (Drieu *et al.*, 2021; Fanti *et al.*, 2018; Roumpou *et al.*, 2003). Its joint presence in potsherds and sediments of the Pendimoun rock-shelter suggests that it may be attributable to contamination from the sediment (Drieu *et al.*, unpublished data).

### Post-excavation degradation

The multiple analyses carried out since the early 1990s have shown that burial duration is not a critical parameter for organic matter degradation, as most organic matter degradation occurs in the first few years after disposal, before an equilibrium is reached (Aillaud, 2001; Stacey, 2009). The excavation breaks this equilibrium by exposing the potsherd again to light, oxygen, changes in temperature and hygrometry and the introduction of new microorganisms. For example, two analyses campaigns of the potsherds of Clairvaux XIV, separated by about ten years (Drieu, 2017; Drieu *et al.*, 2020; Mirabaud, 2007), showed a clear drop in the overall preservation of organic matter, both in terms of the quantity of lipids extracted and the preservation of the signal (especially that of triacylglycerols; Figure 4). When the analysis of organic matter cannot be carried out within a few years of the excavation, it is therefore recommended to store the archaeological material in controlled light, temperature and hygrometry conditions, ideally at  $-40\text{ }^{\circ}C$ .

### Synthesis and outlook

Archaeological pottery is a complex artefact, whose lifecycle is not limited to its usage. Recent studies on the parameters influencing the absorption and preservation of organic matter demonstrate that these are multifactorial mechanisms, occurring from the sourcing of raw material to the disposal of the pot, and that this complexity must be considered during sampling, analysis and interpretation. However, this does not call into question the informative potential of organic residue analyses. When extraction yields

are high, for example, the organic matter extracted is most likely related to the use of the pot because organic matter from manufacture or contamination, where it exists, is generally absorbed and/or preserved in small amounts (for example, 5 to 6 times more lipids are absorbed during use than during high temperature surface treatment; Drieu *et al.*, 2019).

In order to verify this on archaeological samples, the systematic analysis of control samples (sediments and pottery elements that have not contained organic matter such as handles, tiles, kiln wasters, etc.) is essential to verify the absence of contamination compounds, or to identify and exclude them from interpretation. Due to the impossibility, in the current state of knowledge, to distinguish between heating related to firing and heating related to use, the interpretation of molecular markers of thermal treatments (mid-chain ketones,  $\omega$ -(*o*-alkylphenyl)alkanoic acids, polyaromatic hydrocarbons, etc.) must be associated with an in-depth ceramological study to identify potential evidence of the use of organic matter during manufacture: organic temper (e.g. Jan & Savary, 2017; Kulkova & Kulkov, 2016; Sestier *et al.*, 2011), dark core due to incomplete carbonisation of the organic matter in the clay (London, 1981; Maritan *et al.*, 2006; Rice, 1987, p.88), or visual traces of surface treatments (Drieu *et al.*, 2019). Variations in organic extracts in the same vessel may indicate the presence of two distinct products: difference in stable carbon isotope values between thermal transformation markers and fatty acids, difference in molecular and/or isotopic composition between visible residues and extracts from the ceramic walls, or between samples located at different levels on the pot (e.g. Charters *et al.*, 1995). Two distinct products may be related to manufacture and use respectively, and this should be considered in the discussion, as should the assumption of successive uses or intentional mixing of products. The use of organic residue analyses as part of an in-depth functional approach, combining the study of vessel shapes and dimensions, physical properties of the ceramic paste, and use-wear analysis (Rice, 1987, p. 211-243; Skibo, 2013) is also effective. For example, it has been used to discuss possible surface treatments in a vessel that was not shaped for cooking and did not show traces of soot but delivered mid-chain ketones (Fanti *et al.*, 2018).

It is important to note that when extraction yields are low, there is a much greater risk that the extracted chemical signal is a mixture of use, manufacture and/or contamination, making all of these analytical and interpretative precautions mentioned above absolutely essential. However, such organic extracts should not be discarded, as they may offer better insight into the molecular markers related to

the manufacturing process. The systematic implementation of such approaches on large pottery assemblages (e.g. on samples with visible traces of high temperature surface treatments) could lead to the identification of practices related to the manufacture of pottery, opening up an unprecedented field of investigation for organic residues analysis: the study of the manufacturing *chaîne opératoire* (additions of organic material to modify the properties of the clay, selection of the firing fuel, treatment of surfaces with organic products, etc.).

It will be necessary to pursue these methodological studies, which has recently been attracting renewed interest, with approaches in experimental archaeology and ethnoarchaeology. Continued work on organic matter related to manufacture is essential in order to determine whether it can be differentiated from organic matter related to use. An interesting avenue to explore is that of a comparison between free and bound organic matter (accessible with different extraction methods), to determine whether the high temperatures associated with firing can result in stronger bonds between the organic matter and the ceramic (Drieu *et al.*, 2019).

In addition, there is a need to better understand the influence of clay mineralogy, porosity, and soil properties on the preservation of organic matter. These methodological developments will find a very concrete application by helping to identify contexts or pots that are favourable to the preservation of organic matter. It is only with such rational sampling that taphonomic hypotheses can be discarded to explain a low extraction yield, which can then be interpreted in terms of use (for solid or non-fatty substances, for example), broadening the possibilities of functional analyses of batteries of pottery vessels.

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## Annexes

Sample	Description	Lipids extracted from the tiles (µg/g)	Amount of TAGs (µg/g)						
			T42	T44	T46	T48	T50	T52	T54
LD10809	cave clay, without paste processing or surface treatment	174.31	9.29	9.18	9.96	11.17	12.34	9.22	3.23
LD10810	cave clay, without paste processing or surface treatment	76.40	3.98	3.76	3.94	4.36	4.58	3.02	0.96
LD10811	cave clay, without paste processing or surface treatment	151.88	8.76	8.42	9.04	10.11	10.75	7.11	2.11
LD10812	cave clay, kneaded, without surface treatment	202.90	11.44	10.96	11.81	13.42	14.81	10.39	3.32
LD10813	cave clay, kneaded, without surface treatment	361.41	22.01	20.60	21.56	23.57	24.19	14.89	4.05
LD10814	cave clay, kneaded, without surface treatment	108.05	6.46	5.81	5.72	5.66	4.87	2.38	0.62
LD10815	cave clay, kneaded, burnished	27.24	1.65	1.44	1.36	1.24	0.98	0.51	1.65
LD10816	cave clay, kneaded, burnished	129.12	7.84	7.23	7.33	7.59	6.88	3.43	0.84
LD10817	cave clay, kneaded, burnished	68.51	4.02	3.74	3.75	3.79	3.33	1.81	0.43
LD10818	industrial clay, burnished	11.51	0.65	0.52	0.42	0.35	0.28	0.00	0.00
LD10819	industrial clay, burnished	50.41	3.32	3.08	2.97	2.91	2.46	1.25	3.32
LD10820	industrial clay, burnished	50.96	2.94	2.78	2.85	2.95	2.68	1.51	0.00

Table S1: Quantities of lipids and triacylglycerols extracted from experimental tiles.  
 Tableau S1 : Quantités de lipides et de triglycérides extraits des plaquettes expérimentales.