

CLAY RESOURCES AND TECHNICAL CHOICES FOR NEOLITHIC POTTERY (CHALAIN, JURA, FRANCE): CHEMICAL, MINERALOGICAL AND GRAIN-SIZE ANALYSES*

R. MARTINEAU§

*Laboratory of Archaeology, Cultures and Societies, UMR 5594, CNRS/University of Burgundy, Earth Sciences
Centre, 6 bd Gabriel, 21000 Dijon, France and Department of Geosciences, University of Fribourg,
Switzerland*

A.-V. WALTER-SIMONNET

*Laboratory of Chrono-écologie, UMR 6565, CNRS/University of Franche-Comté, 16 route de Gray,
25030 Besançon cedex, France*

B. GROBÉTY

*Department of Geosciences, Petrography and Mineralogy, University of Fribourg, chemin du musée 6,
1700 Fribourg, Switzerland*

and M. BUATIER

Laboratory of Geosciences, University of Franche-Comté, 16 route de Gray, 25030 Besançon cedex, France

Many authors have considered pottery manufacturing constraints and sociocultural elements as factors in change in past civilizations over time. The main issue of this research is to better understand the reasons for changes, or choices, in pottery raw materials. The very precise and detailed stratigraphy and cultural succession of occupations is based on dendrochronological data from the lake-dwelling sites of Chalain (Jura, France). Petrographic, palaeontological and chemical analyses were used to determine the nature and origins of the raw materials used by the Neolithic potters. Stratigraphy and dendrochronological data were used to reconstruct in detail the evolution dynamics of fabric changes. Several raw material sources were identified for many of the pottery groups. Each of them was sampled for qualitative experimental tests of pottery forming. The experimental results show a high variability between the sediments tested. This variability was quantitatively estimated by XRF, XRD, the Rietveld method, calcium carbonate quantification and laser grain-size analyses of matrices, indirect measures of plasticity. These analytical results allow a better understanding of the differences observed in the experimental tests. On the basis of these experimental and analytical results, changing parameters such as pottery manufacturing constraints, mineralogical characteristics of raw materials and sociocultural factors are considered. In conclusion, all the social and technical parameters, in each archaeological context, must be taken into account for a better understanding of the changes occurring throughout the chronological sequence.

KEYWORDS: NEOLITHIC, CERAMIC TECHNOLOGY, JURA, HORGEN, CLAIRVAUX CULTURE, LAKE OF CHALAIN, PETROGRAPHY, CLAY, LASER GRAIN-SIZE ANALYSIS, X-RAY DIFFRACTION, RIETVELD METHOD, TECHNICAL CHOICES

INTRODUCTION

Chemical and mineralogical investigations of Neolithic pottery clearly reveal that the wide variety of raw materials used is related to the natural diversity of the environment. The idea behind this paper stems from the observation that some of the raw materials used by the Neolithic potters would be deemed of poor quality by potters today. Moreover, sudden changes from one raw material source to another were common. Such changes have been observed in various archaeological sites belonging to different Neolithic or protohistoric contexts (Balfet 1962; Courtois 1971; Hultén 1977; Steponaitis 1984; Constantin 1985; Constantin and Courtois 1985; Allen 1991; Echallier and Courtin 1994; Molinas 1995; Morzadec 1996; Colas 2000; Martineau 2000a,b; Sénépart and Convertini 2000). Within the framework of a general study of raw materials and techniques of Late Neolithic pottery from lake dwellings in Chalain and Clairvaux (Jura, France) (Martineau 2000a), the origin of several raw materials has been identified (Martineau *et al.* 2000 and in press). From a modern ceramic manufacturing point of view, some of the clays processed by the Neolithic potters are of poor quality; that is, with low plasticity and a high carbonate content.

The transition period from one culture to the next is not only accompanied by changes in pottery style, but also in the raw material source exploited by potters. Several observations contradict the well-accepted notions that ancient potters predominantly used the nearest outcrops as their raw material source, and that Neolithic people habitually settled where clay and other resources were readily accessible. Ethnographic evidence has clearly established that clay and temper are mainly exploited within a radius of 7 km (86% for clay and 91% for temper; Arnold 1985, 2005). But these results do not signify that raw material sources are always located at the archaeological (or ethnographic) site. Only in 37% and 49% of cases, for clay and temper, respectively, are sources found at less than 1 km from the production site (Arnold 1985, 2005).

In the region studied, the nearest clay outcrops (located under the dwelling sites) seem to have been mainly exploited for manufacturing the floor structure beneath the fireplaces in houses, but generally not for pottery manufacture. Similar dichotomous situations have been observed at Swiss Neolithic sites (Di Pierro 2002, 2003; Bonzon 2003). Ceramic artefacts can be divided into two categories: the first is pottery, while the second is composed of fireplace floor structures and building materials (such as cobs and loom-weights). Each category corresponds to one specific type of raw material.

Fundamental questions arise concerning the interrelations between raw material constraints and their selection, which may depend on natural diversity and accessibility. One of the main aims of this paper is to suggest reasons for the sudden change in the choice of materials, which forced potters to introduce the transport of raw materials as a new step in the 'chaîne opératoire'. Behind this question stands the theoretical debate on ceramic ecology (Matson 1965; Kolb 1989) and cultural identity (Constantin and Courtois 1985; Gosselain 1995; Gosselain and Livingstone Smith 1995). This is one facet of the eternal debate on culture and nature. But this paper relies rather on an ethnological theory of material culture, which considers that technical aspects, such as the choice of raw materials, are an integral part of any culture (Lemonnier 1986, 1993; Latour and Lemonnier 1994).

What was the relationship of the Neolithic potters to their environment? Did they exploit environmental resources based on technical choices or not? What were the main factors responsible for technical changes and choices? Recently, these issues have given rise to substantial debate (Sillar and Tite 2000; Cumberpatch *et al.* 2001; Sillar *et al.* 2001). Some studies concentrate on the cultural peculiarities of ceramic techniques, while others focus on the properties and functionality of the final products as a driving force for the choice of some

raw materials. However, the influence of the quality of raw materials on the manufacturing process is an aspect that is rarely considered. The analyses presented here indicate that the arrival of a new population with new techniques, habits, know-how or environmental knowledge, in a new territory, could be one of the main arguments that may have led the Neolithic potters of Chalain and Clairvaux to change the raw material sources.

Sillar and Tite (2000), Cumberpatch *et al.* (2001) and Sillar *et al.* (2001) have criticized the systematic opposition of technical and social aspects in archaeological contexts. The choice of a technology responds to cultural, functional and production objectives, at one and the same time. Therefore, material science aspects (experimental, chemical and physical data) have to be considered and also included in archaeological or ethno-archaeological studies. The morphological and decorative variations in pottery styles are thought to respond to fashion changes. Techniques respond to different mechanisms, related to apprenticeships of technical knowledge and know-how. The point of view of the techno-functionalists is very different. Today, in our consumer society, maximum yield and performance are the main objectives. It is difficult to imagine that these objectives existed as such in the past, particularly during the Neolithic period. Nevertheless, there are constraints in the use of any raw material, related to its physical and mechanical properties. The choice of raw materials also imposes constraints on the different stages in the pottery 'chaîne opératoire'.

THE ARCHAEOLOGICAL CONTEXT

Ceramic types and dating

This study covers a chronological sequence from *c.* 3169 BC to 2980 BC (calendar years; Lavier 1996) and includes 604 pots of Chalain 2 (layers C and A//), Chalain 3 (layers VIII, VI and IV) and Chalain 4 (phases 1–4).

Two main types of pottery are compared in this study. The first type belongs to the Horgen culture (*c.* 3169–3080 BC) and is characterized by flat-based pots (Table 1). Some of these ceramics were produced using marly Dryas limestones found in the vicinity of the lake dwellings. Horgen pottery was formed by joining internal rings, and quickly smoothing the outer and inner surfaces (Martineau 2000a; see also Table 1). The second type belongs to the Early Clairvaux culture (*c.* 3025–2980 BC) and is characterized by pottery with rounded bases. This pottery was produced by alternating internal and external rings (Table 1), which were joined using a bone tool (Martineau 2002; Martineau and Maigrot 2004). These technical and morphological changes from one culture to the other reflect a more general change that affected all aspects of material culture at that moment (Pétrequin 1997a).

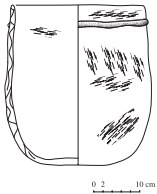
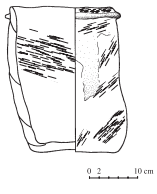
Both types of pottery were fired in open fires (Martineau and Pétrequin 2000). The large carbonate inclusions of the raw materials required thick walls and quick firing to prevent cracking. Pétrequin *et al.* (1994) indicate that pottery production was probably domestic. Organo-chemical analysis of residues found on the pottery walls demonstrated that all of them served for cooking purposes (Regert *et al.* 1999), indicating domestic usage.

PETROGRAPHIC AND CHEMICAL CHARACTERIZATION OF CERAMIC FABRICS

Petrographic and palaeontological characterizations

For the period studied, 604 pottery samples have been petrographically and palaeontologically analysed.

Table 1 *The archaeological context; the morphological, technical and raw material data of the pottery of the Early Clairvaux and Horgen cultures*

Archaeological culture	Type and techniques of pots	Raw materials	Primary forming	Secondary forming	Firing	Context of production and use	Function
Early Clairvaux culture, c. 3025–2980 BC		<i>Acuminata</i> marls with fossilized oysters (Upper Bajocian)	Internal and external alternating rings	Junction of rings with a bone spatula; then smoothing with the hands	Open firing	Domestic	Cooking pots
Horgen culture, c. 3169–3080 BC		Marly limestones with rounded oolitic limestones (Dryas) plus marls and limestones (Oxfordian)	Only internal rings	Smoothing with the hands	Open firing	Domestic	Cooking pots

The Acuminata group (groups 11 and 12)

The pottery in this group contains a large quantity of natural fossil inclusions (Martineau *et al.* 2000). Most of them are oysters (*Praeexogyra acuminata*), which are attributable to the Upper Bajocian period (J1b) (Contini 1970; Dumanois 1982) and are associated with bryozoans, crinoids and urchin spines (Fig. 1). This fossil assemblage is characteristic of the *Acuminata* marls (Contini 1970), corresponding to the Plasne and Châtillon marls of Girardot (1890–6). Identification leaves no doubt about the origin of the raw materials (Martineau *et al.* 2000). In these marls, *Acuminata* shells are naturally included in the argillaceous matrix. They were not added to the clay by the potters. *Acuminata* marls are sediments that form a natural ready-to-use pottery paste.

The rounded oolitic limestone group (group 19)

The pottery from this group is characterized by inclusions of rounded oolitic limestone fragments, rounded quartz and some mica crystals (Fig. 1). Occasionally, pieces of micritic limestone, crinoids and some shell fragments occur. Macroscopically, the matrix consists of white, fine-grained carbonates. The rounded shape of the inclusions suggests that they are of natural origin. This sediment, like the *Acuminata* marls, is also a natural paste.

Chemical characterization of fabrics

Chemical characterization was carried out on 20 samples, principally to define the variability of the ceramic composition, and to compare it with the natural sediment composition (12 samples). The CaO composition depends largely on the quantity of carbonate and fossil inclusions in the ceramics. Two chemical groups of ceramic fabrics are distinguishable on the basis of CaO,

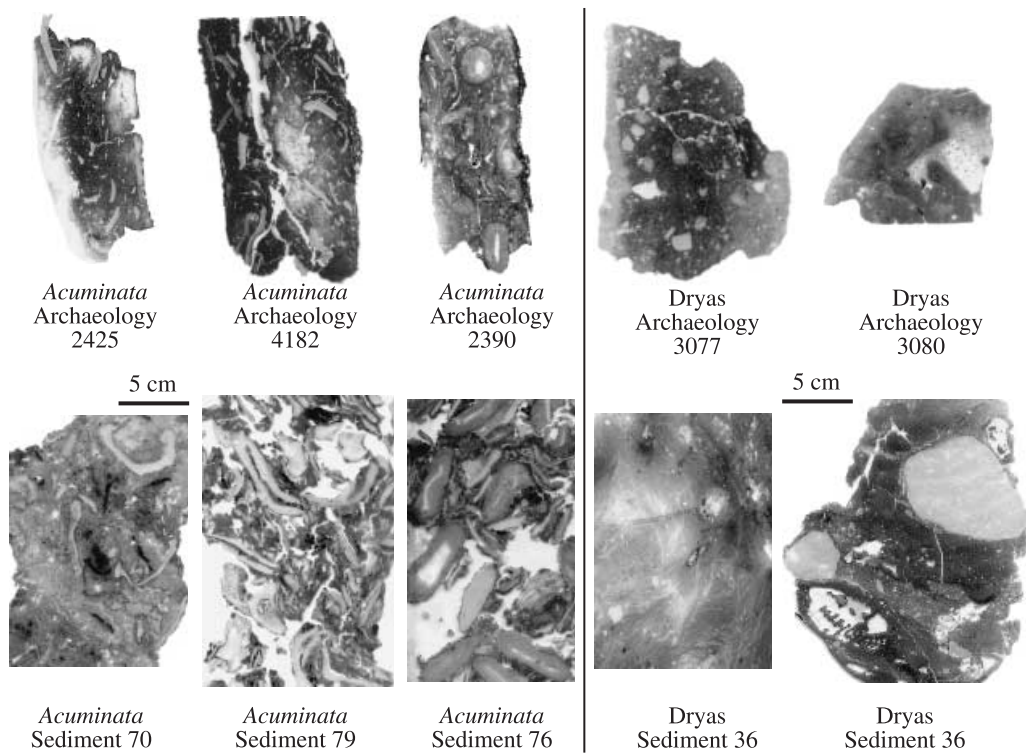


Figure 1 On the left, thin sections of *Acuminata* marls (M2): the main inclusions are natural fossil oysters from the Upper Bajocian (magnification $\times 2$). Samples 70 and 76 are the most similar to the archaeological pottery. On the right, thin sections of Dryas sediment from the western shore of the lake of Chalain (D6): the rare inclusions are detrital rounded oolitic limestone, micritic limestones and quartz (magnification $\times 2$). The two natural sediments show the high variability in the Dryas outcrops, as do the archaeological samples. Archaeological numbers refer to Appendix A and sediment numbers to Table 2.

SiO_2 , K_2O , Al_2O_3 , Fe_2O_3 , Ni, Cr and Ba (Figs 2 and 3; Appendix A). The ternary diagram cumulates between 84.9% and 97.5% of the elements. One group (group A) concerns all pots with *Acuminata* inclusions (groups 11 and 12). The range of CaO in group A is between 15.7% and 46.6%. The second chemical group, group B, corresponds to ceramics with white matrix and rounded oolitic limestone and quartz (group 19; Fig. 2). The range of CaO in group B is between 51.7% and 70.5%. The two groups are very rich in CaO, but CaO is higher in group B than in the *Acuminata* group. In addition, SiO_2 , Al_2O_3 , K_2O and Fe_2O_3 are lower in group B than in group A (Figs 2 and 3; Appendix A). However, the chemical composition in the triangular diagram (Fig. 2) clearly shows two groups corresponding to petrographic groups 11 and 12 (*Acuminata* fabric) and chemical group A, on the one hand, and to petrographic group 19 (rounded quartz and Dryas limestones) and chemical group B, on the other. These results confirm the petrographic and palaeontological characterizations and distinctions between *Acuminata* marls and rounded oolitic limestone sediments. The most important result here is that the chemical composition of some of the natural sediments corresponds to that of the archaeological pottery, as we shall see below in detail.

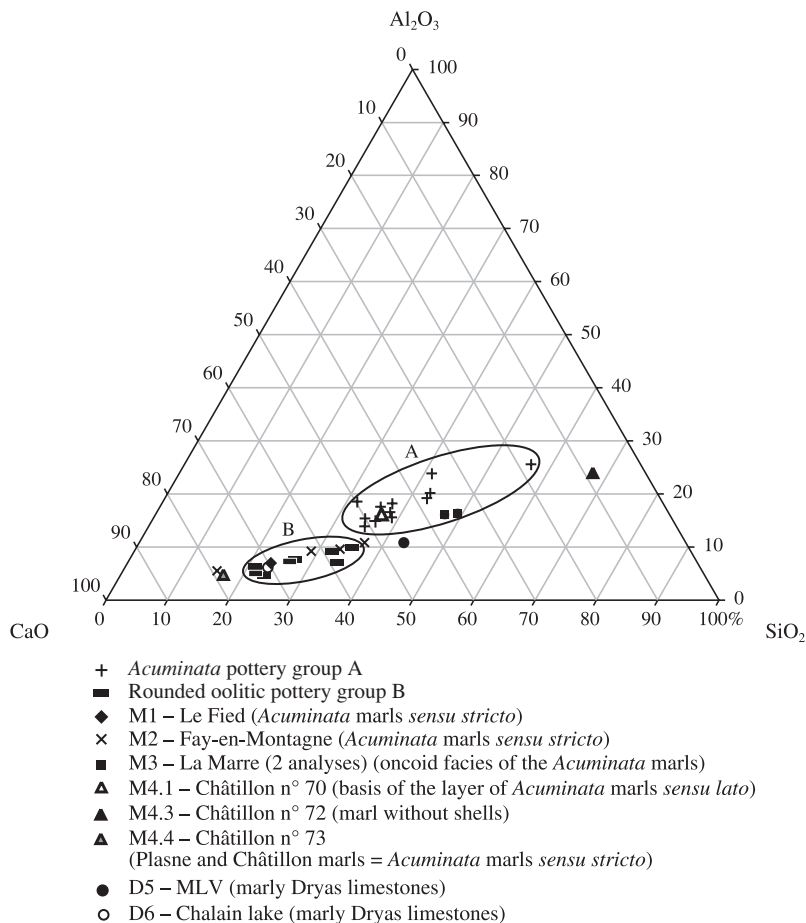


Figure 2 A ternary diagram comprising $CaO\%$, $SiO_2\%$ and $Al_2O_3\%$, showing two pottery groups and their relations with the natural sediment compositions. A, *Acuminata* marls; B, rounded oolitic pottery group. The natural sample best correlated with pottery group A is M4.1. The natural sample best correlated with pottery group B is D6.

THE GEOLOGICAL CONTEXT

The lake of Chalain (Jura, France) is located in the Combe d'Ain, on the eastern side of the French Jura. The geological environment of the lake dwellings of Chalain is dominated by carbonate-rich rocks of Jurassic age (Guillaume and Guillaume 1965; and see Fig. 4). In the Combe d'Ain, the bedrock is covered by quaternary clay sediments deposited during Würmian glaciation and several cold phases during the Late Glacial period (Campy 1982). Due to their high carbonate, low clay content, the rocks and soils of the Combe d'Ain area are 'poor pottery raw materials'.

Previous studies and raw material analyses have allowed the geological provenance of several raw materials to be identified (Martineau *et al.* 2000 and in press). The raw materials used by the Neolithic potters from the lake of Chalain are summarized on the map shown in Figure 4. The identification of four geological formations used by the Neolithic potters allows comparison

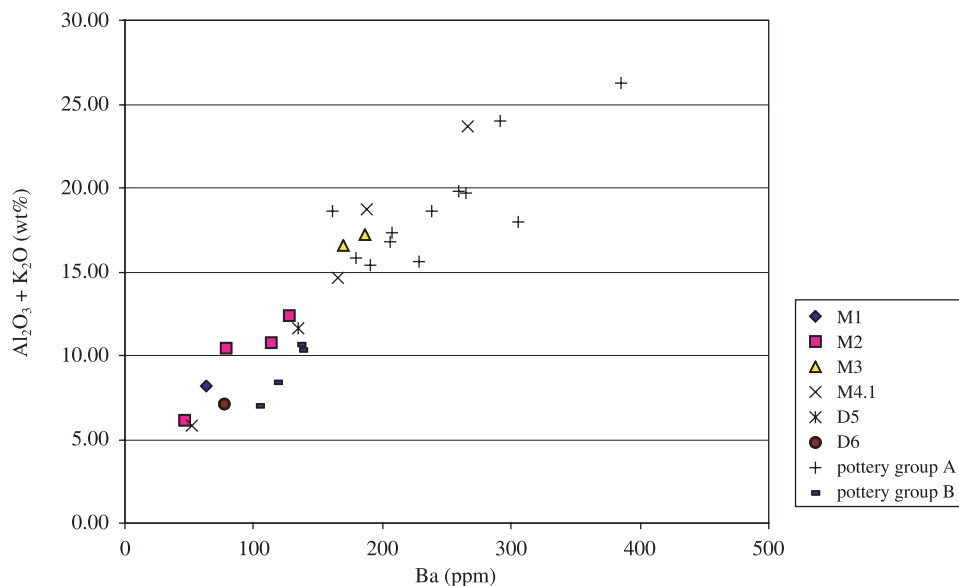


Figure 3 The distribution of $Al_2O_3 + K_2O/Ba$ clearly confirms the two chemical groups corresponding to the petrographic groups A and B.

with the general geological context that has previously been presented (Fig. 4). In the area studied, only a small part of the available raw materials has been exploited by the potters' community (Fig. 4). We do not know the reasons for these 'choices'. We do not know whether it is even possible to speak of 'choice'. We shall focus our attention on two main raw materials: the *Acuminata* marls and the marly Dryas limestones.

Clayey sediment evidence

The Acuminata marls from the Upper Bajocian The outcrops of *Acuminata* marls are rare and difficult to localize. They are located on the Lons-le-Saunier plateau, at 5–12 km from the lake dwellings (Fig. 4). Extensive prospecting revealed only four accessible outcrops. The more detailed stratigraphic section of Châtillon 'La Percée' shows three distinct layers (Girardot 1890–6, Contini 1970), summarized in the stratigraphic chart shown in Figure 5. At the basic level are the Lower Jurassic marly limestones of Courbouzon (Girardot 1890–6) with rare *Acuminata* shells and brachiopods (*Parkinsonia subarietis* Wetzel and *Terebratula cf. masticonensis* Liss), corresponding to the base of *Acuminata* marls *s.l.* of Contini (1970) (Fig. 5). The second layer is composed of *Acuminata* marls (the Plasne and Châtillon marls of Girardot 1890–6), referred to as '*Acuminata* marls *s.s.*' by Contini (1970). In this layer we have distinguished a base level of marl without shells and an upper level of marl with many *Acuminata* shells. Plasne oolitic alternated marls and limestones form the third upper layer (Fig. 5). In this level, the shells are coated in oncoïds and associated with *Parkinsonia parkinsoni* Sow (Contini 1970). This type of level is called 'oncoïd facies of the *Acuminata* marls' (the Plasne pisolitic marls of Girardot 1890–6). These three facies of Girardot have been regrouped by Contini (1970) in the same formation: the marls of Plasne. The systematic presence of abundant *Acuminata* shells in this formation has led to the name '*Acuminata* marls'.

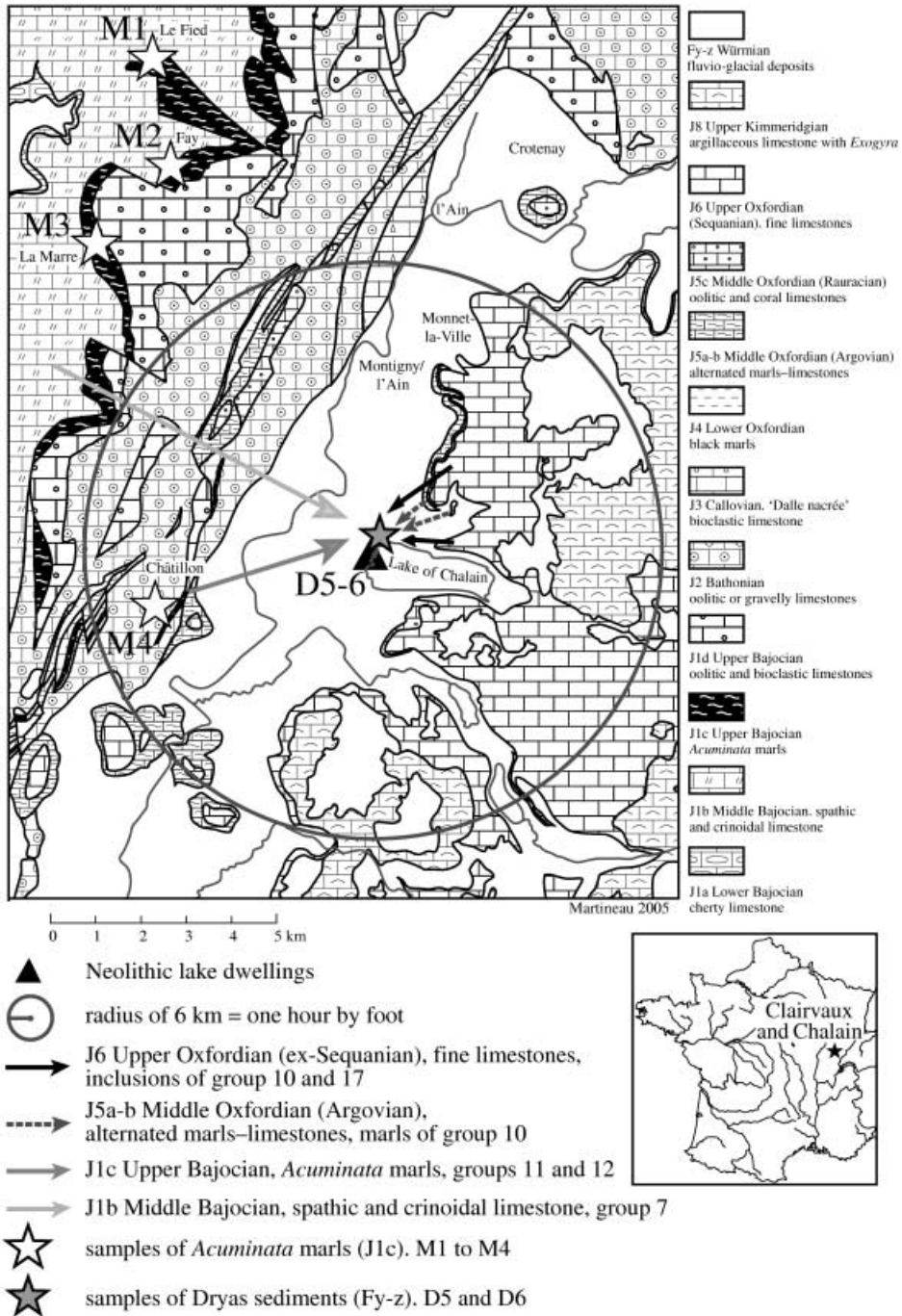


Figure 4 The geological context of the Neolithic sites of the lake of Chalain. In this area, all the sediments are carbonate rich. The map shows the locations of the sediments used by the Neolithic potters between c. 3169 bc and 2950 bc. The Dryas sediments are located at the Neolithic sites. The Oxfordian marls are located 1.5 km from the lake-dwelling sites. The *Acuminata* marls are situated 5–12 km from the lake-dwelling sites.

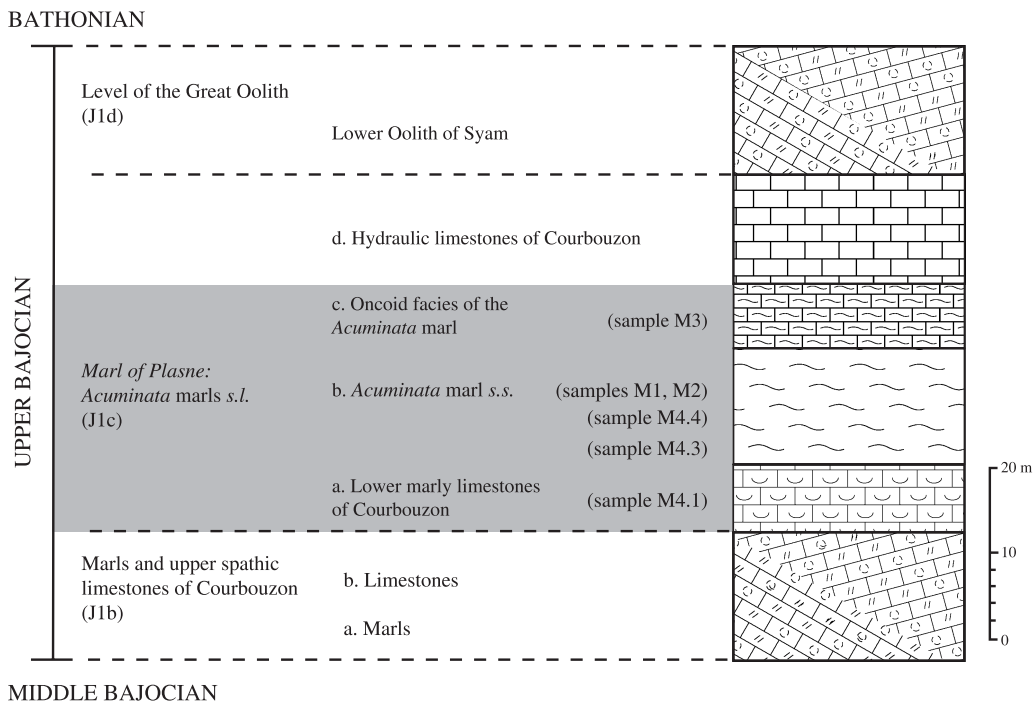


Figure 5 A stratigraphic log of the Upper Bajocian in the Lons-le-Saunier area (Contini 1970). The analysed samples have been placed in the corresponding levels.

Samples M1 and M2 were taken from the *Acuminata marls sensu stricto*, whereas sample M3 represents the oncoïd facies of the *Acuminata marls s.l.* (Table 2; Figs 4 and 5). M4.1 is a sample of the Lower Jurassic marly Courbouzon limestones of Girardot (*Acuminata marls sensu lato*). Sample M4.4 corresponds to Plasne marls (*Acuminata marls s.s.* of Contini 1970). M4.3 comes from a little clayey level without fossils, in *Acuminata marls s.s.*, located between M4.1 and M4.4. This type of level corresponds to lateral facies variations, frequently found in these formations, and related to variability in sedimentation. On the geological map, no distinction has been made between the *Acuminata marls sensu stricto*, and the oncoïd facies of *Acuminata marls* (Chauve *et al.* 1993).

All of these sediments are very rich in CaO (Contini 1970, Dumanois 1982), but some variability exists. Such variations will be very useful for further comparison between archaeological pottery and these natural sediments.

The marly Dryas limestones Two samples (D5 and D6) of marly Dryas limestones from outcrops on the western shore of the lake of Chalain (Fig. 4 and Table 2) were analysed. The inclusions of D5 and D6 are characteristic of a detrital terrigenous allochthonous sediment (noted as 'Fy-z' on the geological map) deposited during the cold phases of the Late Glacial (Old and Younger Dryas), between 15 000 and 10 000 BP (Magny 1991, 1992; Bossuet *et al.* 1996, 1997). These sediments are often covered by lacustrine chalk, which constitutes the substratum of the lake (Campy 1982), but several outcrops of the underlying Dryas sediments are accessible on the western shore of the lake of Chalain (Fig. 4).

Table 2 *Natural sediment references studied by chemical, mineralogical and grain-size analyses*

<i>Sample</i>	<i>Location</i>	<i>Geological attribution</i>	<i>Number of thin section</i>	<i>Lambert co-ordinates</i>
M1	Le Fied	<i>Acuminata</i> marl s.s. Upper Bajocian (J1c)	52	$x = 858.4, y = 184.5$
M2	Fay-en-Montagne	<i>Acuminata</i> marl s.s. Upper Bajocian (J1c)	79	$x = 859.8, y = 182.5$
M3	La Marre	Oncoid facies of <i>Acuminata</i> marl s.l. Upper Bajocian (J1c)	77	$x = 857.5, y = 181$
M4.1	Chatillon 'La Percée'	<i>Acuminata</i> marl s.l. Upper Bajocian (J1c)	70	$x = 859, y = 188$
M4.3	Chatillon 'La Percée'	<i>Acuminata</i> marl s.s. Upper Bajocian (J1c)	72	$x = 859, y = 189$
M4.4	Chatillon 'La Percée'	<i>Acuminata</i> marl s.s. Upper Bajocian (J1c)	73	$x = 859, y = 190$
D5	Marigny-les-Vernois (MLV)	Old and Younger Dryas, Late glacial (Fy-z)	49	$x = 863.5, y = 181.6$
D6	Station 2 of lake of Chalain	Old and Younger Dryas, Late glacial (Fy-z)	36	$x = 863, y = 182$

Mineralogically, D5 and D6 are identical, but the proportions of the quartz and carbonate natural inclusions are different, which may explain the chemical differences between them. In thin sections from D5, the quantity of quartz is higher than in sample D6, where quartz is rare. Moreover, quartz morphologies of the samples are different. In D5, the morphology of quartz is angular, whereas quartz is rounded in sample D6. After firing, the matrix of D5 is red, whereas the D6 matrix remains white, certainly because of its lower FeO content (Appendix A).

These Dryas sediments are very rich in CaO, whereas the CaO content of D6 (66.06%) is higher than the CaO content (42.18%) of D5 (Fig. 2, Appendix A). In the same way, Al₂O₃ and SiO₂ are almost twice as high in D5 (9.85% and 39.45%, respectively) as in D6 (5.91% and 21.69%, respectively) (Fig. 2, Appendix A). Samples D5 and D6 can be identified and distinguished by their chemical and petrographic composition.

COMPARISONS BETWEEN POTTERY AND SEDIMENTS

The chemical discussion is based on a ternary diagram depicting Al₂O₃, SiO₂ and CaO, regrouping between 84.89% and 97.53% of the bulk chemical composition of the samples.

The marly Dryas limestones in sample D5 have a higher amount of quartz inclusions than pottery group B. The chemical composition of D5 is higher in SiO₂ and lower in CaO than pottery fabric group B (Fig. 2 and Appendix A). After firing, the colour of the D5 matrix is red, probably because of FeO discrepancies (Appendix A), whereas the group B pottery fabric matrix is white. However, textural analysis reveals a higher percentage of inclusions in D5 than in D6, which underlines the natural variability of these sediments. The percentage of quartz inclusions in D6 corresponds well with that found in pottery group B. The chemical

composition of D6 is similar to that of pottery group B. After firing, the D6 matrix remains white, as found in pottery group B. Thus, the archaeological thin sections for pottery group B show the same matrix colour and mineralogical inclusions as the marly Dryas limestones in sample D6 (Martineau 2000a). Analyses of all available sediment evidence confirm that sample D6 is far more compatible with pottery group B than sample D5 (Fig. 2).

Analyses show clearly that *Acuminata* marls have high carbonate variability. Sample M4.3 is an example of *Acuminata* marls without shells. It is low in CaO, but rich in SiO₂ and Al₂O₃, because of the clay concentration and the absence of shells (Fig. 2). Several samples of *Acuminata* marl sediments (M1, M2 and M4.4) have CaO contents higher than that of pottery group A. Their chemical composition corresponds to pottery group B but, as we have seen before, they are petrographically and palaeontologically incompatible with this fabric. On the contrary, sample M4.1 (the Lower Jurassic marly limestones of Courbouzon, corresponding to *Acuminata* marls *sensu lato*) from Châtillon coincides exactly with pottery group A (Fig. 2). The bulk chemical composition of this sample is compatible with that of pottery group A (Fig. 2, Appendix A). Sample M3, corresponding to the oncoid facies of *Acuminata* marls, is close to pottery group A in our diagram, but it differs in the amount of SiO₂, which is slightly higher (Fig. 2, Appendix A). These two samples confirm chemically the observed petrographic and palaeontological compatibility of *Acuminata* marls with pottery group A, notably concerning samples M4.1 and, to a lesser extent, M3.

Textural comparisons of sediments and matrix (Figs 1 and 2) show a trend that confirms the chemical observations. The pottery in group A contains a lower quantity of fossil inclusions than some sediments. There is, however, great natural lateral variability in the sediment composition. The Châtillon M4.1 sample has the lowest percentage of fossil inclusions of the *Acuminata* samples, which explains its lower percentage in CaO (Fig. 2, Appendix A). In sample M4.1, the lower percentage of CaO shows a very good correlation with archaeological pottery group A.

THE ORIGINS OF THE POTTERY RAW MATERIALS

It is surprising to note that sample M4 from Châtillon (Fig. 4) is the nearest *Acuminata* marl outcrop to the Chalain dwellings, 5 km from the lake of Chalain. Until about 30 years ago, this outcrop was far more visible in the landscape than today, because the exposed stratigraphic cross-section was 14 m high (Girardot 1890–6). Twentieth-century roadworks have raised the level of the road until only a 5 m difference remains (D. Contini pers.comm.).

It is interesting to observe that the situation is comparable for the marly Dryas limestones, the nearest clayey outcrop to the Chalain lake-dwelling sites (Fig. 4). For the two fabric groups A and B, the inclusions are naturally mixed with the matrices. The Neolithic potters may have looked specifically for this type of sediment.

The bone tools used for the surface treatment of the green bodies found within the site of the lake dwellings are strong indications that the pottery was not manufactured close to the raw material source, but that the production sites were in the dwellings themselves. This required transport of heavy raw materials over a distance of at least 5 km to the Neolithic sites (Martineau and Maigrot 2004). This kind of transport would represent about an hour's journey each way.

POTTERY FABRICS: ORIGINS AND TECHNOLOGICAL CHANGE

The bar diagram of Figure 6 shows the different raw materials within the chronological sequence of the archaeological cultures studied. The more siliceous paste groups are placed on

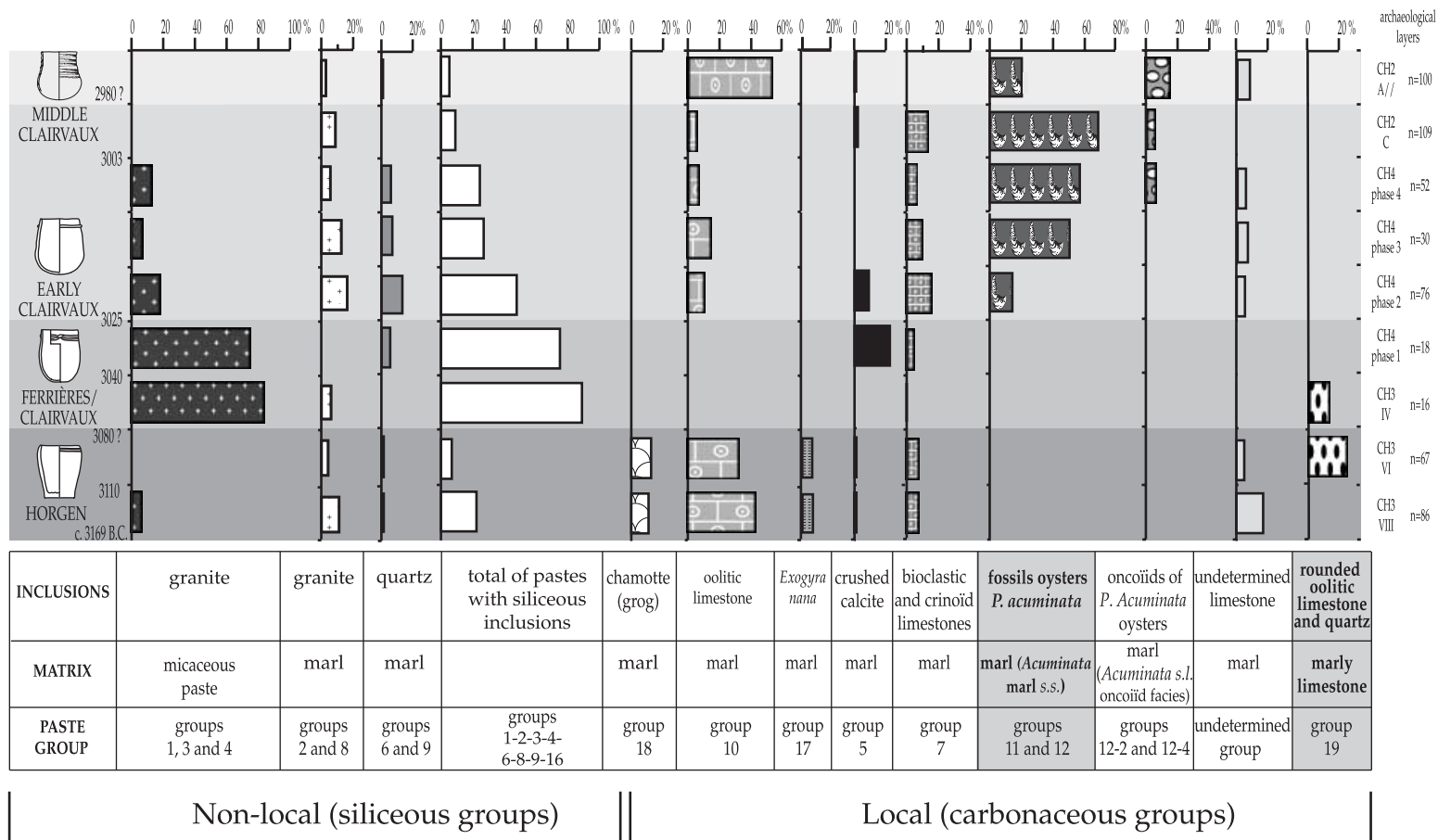


Figure 6 The chronological sequence and distribution of pottery paste groups. The carbonate proportion increases from left to right in the figure. Several raw materials changes have occurred during this two-century sequence.

the left, whereas the more carbonate-rich ones are on the right of the figure. The pottery raw materials from Chalain can be subdivided into two categories and 12 main groups of pastes (Martineau 2000a,b; and see Fig. 6).

The first category is characterized by siliceous inclusions. The main group of this category is composed of granite rock fragments in a micaceous paste (Martineau *et al.* 2000; Di Pierro and Martineau 2002a,b). The second category is characterized by several types of carbonate-rich (or very rich) matrices with a large variety of inclusions, such as limestone fragments or fossils (Fig. 6).

During the Horgen period, potters used several paste groups (groups 7, 10, 18 and 19)—notably group 10—with the highest frequency (Martineau 2000a). Crushed added oolitic limestone inclusions constitute the main type of inclusion in this pottery fabric. On the basis of angularity and sedimentological associations, we know that these inclusions were crushed and added to a marl. The inclusions from group 10 correspond to the ‘Couches du Morillon’ of the Upper Oxfordian (J6). The geological attribution of this fabric is attested by the presence of foraminifers, *Lituolides Pseudocyclamina s.s.* (formerly *Alveosepta jaccardi* from Sequanian), determined by A. Boullier (Geosciences Department, Besançon, France; see also Enay 1966; Guillaume 1964; Guillaume and Guillaume 1965; Gaillard 1983), in oolitic fragments (Martineau *et al.* in press). The nearest outcrop of this formation is located at ‘sur la Côte’, in Marigny/Ain (Jura), 1.5 km from the Neolithic lake-dwelling sites of Chalain (Fig. 4). The marls used for this paste group come from the Middle Oxfordian (J5a-b), and are located nearer than inclusions of the same group.

All of the carbonate groups have a local source (outcrops at a distance closer than 7 km). However, while a potential source for the petrographic pottery groups 1, 3 and 4 (micaceous pastes with granite inclusions; Fig. 6) may be located 20 km away from the lake dwellings of Chalain, a more likely site is known to exist 50–60 km away (Campy 1982; Martineau 2000a; Martineau *et al.* 2000). These pottery groups, represented by 46 pots belonging to the Ferrières/Clairvaux culture (c. 3045–3030 BC), were most probably not locally produced, but imported by an immigrant population, as proposed in the hypothesis of P. Pétrequin (Pétrequin 1993, 1997a,b, 1999; Pétrequin *et al.* 1998), or exchanged with another cultural group. After this period, pastes with siliceous inclusions were gradually replaced by carbonate pastes, notably the *Acuminata* marls *s.l.* (Fig. 6), which have already been characterized and localized.

In this paper, we focus on marls with fossilized oyster shells (*Acuminata* marls *s.l.*) and on marly Dryas limestones, with rare fragments of rounded oolitic limestone. The marly limestones of the Dryas sediments (group 19) were exploited in the Horgen culture, whereas the *Acuminata* marls were only used for pottery production during the Clairvaux culture (Table 1 and Fig. 6). Pottery made with marly Dryas limestone is only found in two layers of the chronological sequence. The respective percentages of pottery produced with this raw material are 26.9% from layer VI (18 pots) and 12.5% of pottery from layer IV (two pots) of Chalain 3 (Fig. 6).

The use of the *Acuminata* marls (groups 11 and 12) covers the period between c. 3025 BC and 2980 BC, corresponding to the Early Clairvaux culture. They were never employed during the Horgen culture or the Ferrières/Clairvaux culture (Chalain 3, layer IV), but became the main sediment exploited by the potters of Chalain during the Early Clairvaux culture (163 pots; Fig. 6). From 11% at the beginning, the proportion of *Acuminata* marls *s.s.* increased to 63.5% of the pastes used during this period (Fig. 6). From the end of the Early Clairvaux culture to the Middle Clairvaux culture, potters also used the oncoid facies of the *Acuminata* marls *s.l.* (between 5.5% and 15%) (Fig. 6). At the peak of the *Acuminata* marl *s.l.* exploitation, nearly 80% of the pottery from the Chalain sites was made using this raw material.

Such a diversity of raw materials used in manufacture is a good archaeological context in which to seek possible simultaneous changes in sociocultural factors, and parameters for pottery raw materials, such as their physicochemical composition and mechanical properties.

AN EXPERIMENTAL APPROACH TO SEDIMENT QUALITY

Questions about the nature of raw materials

Acuminata marls and marly Dryas limestones are sediments that are very rich in CaO. Moreover, *Acuminata* marls contain a large quantity of fossil inclusions. The marly Dryas limestones seem to have a very low plasticity. All of these observations highlight a surprising use of raw materials. This situation spurs us on to try to understand the reasons behind their use as pottery clays. Answers to this question require good knowledge and thorough analyses of these raw materials.

Paste preparation and technical choice

We performed a comparative experimental study between three kinds of paste preparation: exploitation of local sediments such as the Dryas sediments (natural mixing between clay and natural inclusions), utilization of local clay and the addition of local limestone inclusions, and utilization of *Acuminata* marl, found 5 km from the dwelling sites. The first method (Dryas sediments) requires only the extraction of the sediment, while for the second one (local clay and added inclusions) the limestone must be crushed, and then added to local clay and mixed in. The third method (the *Acuminata* marls) already involves a natural mixture of clay and inclusions, like the first one, but requires the transport of the sediment over 5 km to the dwelling sites. For the second type of paste preparation, we measured the time needed to crush limestone fragments to obtain a grain size from 2 mm to 7 mm, like the fragments added to the Clairvaux pottery. About 1 kg of limestone can be crushed in 22 min (Martineau 1995; see also Table 3).

A comparative table shows the differences between these three modalities of paste preparation (Table 3). The Dryas sediments require no temper crushing and only a very short transport. The use of local clay requires local limestone to be crushed before it can be added. *Acuminata* marls with natural fossil inclusions require a lengthier transport of raw materials. The quantitative results show that the amount of time spent on the preparation of local clay with local limestone is higher than that spent on the acquisition of *Acuminata* marls, which is twice as high as for the Dryas sediments. Crushing and adding a local limestone to a local clay takes three times longer than using the Dryas sediments.

Forming the body

Our experimental approach to pottery techniques is based on approximately 10 years of training and practice, which represents the production of over three hundred pots (Martineau 1995, 2000a). Experimental pottery manufacturing used all six different sediment samples (Table 2). Pottery was produced with the same morphology and using the same techniques as in the past (Martineau 2000a, 2002; Martineau and Maigrot 2004: see also Table 1). From the potter's point of view, both the marly Dryas limestones and the *Acuminata* marl are 'poor pottery raw materials'.

Table 3 A comparative table of time measurements of three kinds of paste preparations; the use of Dryas sediments should have been the most cost-effective choice in terms of time spent

	<i>Dryas sediment (D5 or D6)</i>				<i>Local clay + local limestone</i>				<i>Acuminata marl from Chatillon (M1 to M4)</i>			
	<i>Clay + natural inclusions extraction</i>	<i>Clay transport (0.5 km * 2)</i>	<i>Adding and mixing water</i>	<i>Total</i>	<i>Clay extraction</i>	<i>Limestone crushing</i>	<i>Adding water and mixing temper</i>	<i>Total</i>	<i>Clay + natural inclusions extraction</i>	<i>Clay transport (5 km * 2)</i>	<i>Adding and mixing water</i>	<i>Total</i>
Weight measured	21	21	24	24	14.4	6	3.6	24	21	21	24	24
Weight %	90		10	100	60	30	10	100	90		10	100
Time spend (min)	30	10	50	90	24	132	90	246	30	100	50	180
		3.75 min for 1 kg				10.25 min for 1 kg				7.5 min for 1 kg		

The quantity of carbonate in the matrix and the high proportion of inclusions (natural or added) are largely responsible for the low plasticity of these raw materials. As a result, these raw materials are difficult to use for pottery manufacture. The high proportion of inclusions may cause difficulty during forming, notably when joining rings.

Textural characteristics are heterogeneous in the different samples of *Acuminata* marls (Martineau 2000a). Samples such as M2 and M4.1, with few inclusions, allowed the production of pottery without too many problems. Samples M2 and M4.1 have a better workability than the other *Acuminata* marl samples.

The forming experiments highlighted differences between the Dryas sediments and the *Acuminata* marls. It was significantly more difficult to produce pottery using the Dryas sediments, notably with sample D6. The shaping process had to be completed very quickly because, for this sample, the drying process of the surface junction was very fast. This sediment has a thixotropic behaviour. The plasticity of the paste is increased by adding water, but if too much water is added, this paste loses its coherence and collapses. This problem is less often encountered with the *Acuminata* raw material.

The mineralogical analyses of the raw materials will allow the quantification and qualification of the differences between the two pastes in the forming process. It could be interesting to know why these raw materials, in particular, were used and perhaps even selected. There are many reasons for such choices: time spent, transport distance, the weight of heavy raw materials, cultural and pottery traditions and know-how, mechanical forming constraints such as workability, and firing or functional constraints. Such parameters should be analysed and quantified. Here, we propose to explore some of them, in particular the mineralogical, petrographic, textural and chemical characteristics of raw materials. These analyses were carried out on the natural samples (M1, M2, M3, M4.1, M4.3, M4.4, D5 and D6; Table 2, Fig. 4) as well as on the archaeological ones.

METHODOLOGICAL AND ANALYTICAL CHOICES

The methodological techniques are detailed in Appendix B. One of the key parameters for shaping a pottery body is the plasticity of the paste. Therefore, paste viscosity measurements seem to be a straightforward way of comparing the quality of different sediments for their use as pottery raw materials. The viscosity of a paste, however, is not only dependent on the physical and chemical properties of the solid, but also on the water/solid ratio, the water chemistry, the curing conditions and so on, parameters that remain unknown for the pastes used by the Neolithic potters. Therefore, the experiments only approximate the archaeological conditions. As a consequence, we have focused our analyses on the properties of the solid raw materials that may influence plasticity; for example, the sediment mineralogical composition (type and quantity of clay minerals) and its grain-size distribution.

Plasticity—the key parameter for the shaping of ceramic bodies—is, among others, a function of the mineralogical composition of the raw material. It is roughly proportional to the amount of clay minerals, especially of swelling types, in the sub-micron fraction. Therefore, physical characterization of the raw materials by X-ray diffraction (XRD), the Rietveld method and grain-size analysis should give independent, quantitative proof of the raw materials' plasticity and help to reinforce the qualitative results obtained from the manufacturing experiments.

To avoid confusion in the terminology, we would like to recall the distinction between the mineralogical and size-related use of the word 'clay'. In the classification scheme for grain

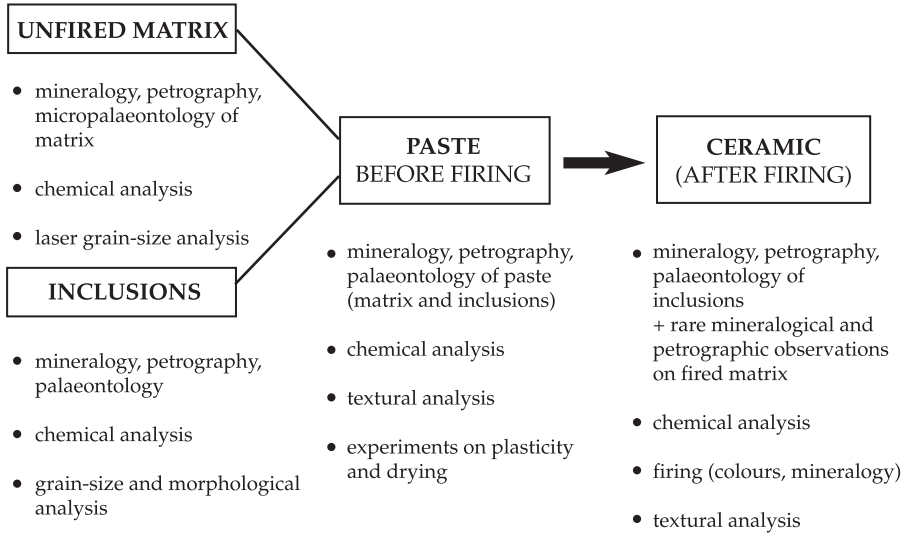


Figure 7 The correspondence between analyses applied to matrix and to inclusions, and the related steps of the 'chaîne opératoire'. Laser grain-size analyses cannot be performed after firing. Firing modifies the mineralogical information; for example, some clay minerals are altered.

size, the word 'clay' is used for the grain-size fraction $< 2 \mu\text{m}$ of sediment, independently of the nature of the minerals. Mineralogically, clay minerals (illite, kaolinite, chlorite and smectite, among others) are a subgroup of the phyllosilicate group. Although clay minerals have often grain sizes $< 2 \mu\text{m}$, their size is not limited to this fraction. Moreover, many minerals can have a size $< 2 \mu\text{m}$ without being clays.

Light optical, X-ray diffraction, grain-size distribution and calcimetric analyses were performed on the raw sediments. Figure 7 summarizes the different techniques applied, and their relations with the stages of the pottery production 'chaîne opératoire'. It is obvious that the laser-based grain-size analyses can only be used for the loose unfired raw materials but not for the fired clays.

RESULTS OF NATURAL SEDIMENT ANALYSES

Grain-size analyses

Samples were sieved into two fractions, with a cut-off point at 2 mm. Analyses were performed on the fraction below 2 mm. Table 4 gives percentages for the different grain-size classes. The *Acuminata* marls contain from 2.3 to 3.3 vol% clay (0–2 μm ; samples M1–M4). The Dryas sediments show a higher proportion of clay fraction (7.1 vol% for sample D5 and 19.6 vol% for sample D6). In all samples, the fine silt (2–20 μm) proportion is weak, except in sample D6 (main grain-size fraction with 39.9 vol%).

In sample D5, gravel ($> 2 \text{ mm}$) represents the main grain-size fraction (25.2 vol%). In the four *Acuminata* marl samples (M1–M4), the main grain size is gravel ($> 2 \text{ mm}$; 38.9–43.4 vol%), except in sample M2, where the main fraction is coarse sand (200–2000 μm ; 30.3 vol%).

In short, global grain-size analyses show that the amount of the finest fraction is higher in the Dryas sediments than in the *Acuminata* marls (Tables 4 and 5). Moreover, a lot of natural

Table 4 The bulk distribution of grain-size fractions in vol%

Sample	Clay, 0–2 μm (%)	Fine silt, 2–20 μm (%)	Coarse silt, 20–50 μm (%)	Fine sand, 50–200 μm (%)	Coarse sand, 200–2000 μm (%)	Gravels > 2000 μm (%)
M1	2.3	10.3	12.2	15.6	18.6	40.8
M2	2.7	13.3	9.8	19.3	30.3	24.5
M3	3.2	13.1	12.4	10.5	21.8	38.9
M4	2.5	10.7	9.6	9.9	23.8	43.4
D5	7.1	19.5	18.8	25.1	4.2	25.2
D6	19.6	39.9	27.4	8.9	0.2	3.9

Table 5 Laser grain-size analysis results; the vol% of the fraction < 2 μm and grain-size populations in vol%

Sample	Laser grain size											
	wt% > 2 mm	vol% < 2 μm	Granulometric populations < 2 mm (vol%)									
			0.15	0.23–0.26	0.4	2–5	7.5	10–30	40	90–115	230–250	770–890
M1	62.9	4	0	1	0	10	0	14	25	15	10	25
M2	37.7	4	0	1	0	9	0	15	11	17	16	31
M3	59.9	5	0	2	0	9	0	28	12	9	12	27
M4	66.9	4	0	1	0	7	0	22	12	10	15	34
D5	38.8	9	0	2	0	30	0	13	21	28	6	0
D6	6.0	20	1	0	4	40	2	33	12	7	0	0

inclusions (gravels, 24.5–43.4 vol%) are present in the *Acuminata* marls and in sample D5 (Dryas sediments), whereas they are rare in sample D6, which can be found close to the lake dwellings.

Grain-size distribution curves

The grain-size distribution curves are all of polymodal type. Samples M1–M4 show similar curves, characterized by the occurrence of a grain-size population whose mode is between 770 and 890 μm , not found in samples D5 and D6 (Table 5). These two samples also show higher proportions of particles < 2 μm , with a maximum of 20% in volume for sample D6 (Dryas sediment from the lake of Chalain), which is the sample with the lowest proportion of particles > 2 mm (Table 5).

After analysis by a smoothed second derivative method, 10 well-differentiated grain-size populations can be distinguished: 0.15 μm , 0.23–0.24 μm , 0.4 μm , 2–5 μm , 7.5 μm , 10–30 μm , 40 μm , 90–115 μm , 230–250 μm and 770–890 μm . Table 5 shows the proportions of these 10 populations in samples M1–D6. These results show that samples D5 and D6 clearly differ from samples M1–M4 as the 2–5 μm grain-size population prevails, and the 230–250 μm and 770–890 μm grain-size populations are either weakly represented or absent in these two samples. Moreover, the data in Table 5 also show that sample D5 differs from sample D6,

as the 0.15 μm , 2–5 μm and 7.5 μm grain-size populations only occur in sample D6, where the 0.23–0.26 μm grain-size population does not occur.

Mineralogical composition (XRD)

The mineralogical assemblages found in the two raw materials are similar, but the individual phases occur in different proportions. The main minerals are calcite and quartz, whereas ankerite ($\text{CaFe}(\text{CO}_3)_2$) is only present in three samples (M1, M2 and D6; see Table 6). Clay minerals were identified in the patterns obtained from the bulk analysis.

The XRD patterns from the clay fraction allowed a better identification and quantification of the clay minerals present. Illite and expandable clay minerals were identified in all samples from their (001) reflections at 10 \AA and about 12–13 \AA , respectively, in XRD patterns obtained from the air-dried samples. The shift of the basal reflections of the expandable fraction to 13.7 \AA and 17 \AA after glycolation of the sample, and to 10 \AA after heating, may indicate the presence of irregularly interstratified mixed layers, such as smectite and smectite–illite. Chlorite is present in samples M3, D5 and D6—that is, in oncoid facies of the *Acuminata* marls (M3) and in the Dryas sediments—and in a smaller proportion in sample M2 (Table 6). It is characterized by the (002) reflection at 7 \AA in patterns from air-dried, glycolated and heated samples. The (001) reflection at 14 \AA is difficult to characterize because of the presence of expandable clay. The hydrate–hydrazine treatment performed on samples D5 and D6 confirms that the 7 \AA reflection can be attributed to the presence of chlorite, and that these samples contain no kaolinite.

The relative abundance of each clay mineral in the clay fraction was semi-quantitatively estimated, using peak intensity ratios (Table 6). The expandable clay mineral (smectite and mixed layer) content in *Acuminata* marls (M1, M2 and M4) is relatively high, representing between 34% and 48% of the total clay minerals. The exception is sample M3 (oncoid facies), with only 19% of expanding clay minerals, but with 30% chlorite. The marly Dryas limestone from the lake of Chalain (D6) is characterized by 90% non-expandable clays, such as illite and chlorite (Table 6).

Mineralogical quantification (Rietveld method and calcimetry)

As the chemical results for the sediments have already been discussed and developed above, we will simply recall their main characteristics here. The *Acuminata* marls and Dryas sediments are both very rich in CaO (Fig. 2, Appendix A).

All Rietveld refinements converge and the residuals are in the expected range, as indicated by the χ^2 value [(measured residual/expected residual)²], which is between 1.5 and 1.8 (Table 6). The calcite content in all samples is high, between 68.4 and 88.5 wt% in the *Acuminata* marl samples (M1–M4) and between 57.6 and 81.7 wt% in the two Dryas samples (D5 and D6). The ankerite contents are between 4 and 7 wt%. The sum of calcite and ankerite obtained from the Rietveld refinement is within 10 rel.% of the carbonate content determined by calcimetry, with the exception of sample D5. The discrepancies may be explained by the heterogeneous nature of the samples, the fact that all CO_2 from the calcimetry analyses is attributed to calcite, and errors related to the refinement process (5 rel.%).

The illite concentration for sample M2 is 15 wt% (Table 6), but the finest grain-size fraction in the same sample represents only 2.7 vol% of the whole sample (Table 4). The weight and volume percentages are comparable in these samples, because all phases have nearly the same density. This is a clear indication that most of the illite minerals and white mica crystals in this

Table 6 *Qualitative XRD results and mineralogical quantitative measurements by the Rietveld method and calcimetry*

Sample	XRD bulk data				XRD clay fraction (%)				Rietveld method										
	Calcite	Ankerite	Quartz	Clays	Illite	Chlorite	Smectite	Illite/smectite mixed layer	Calcimetry		Calcite	Error	Illite	Error	Quartz	Error	Ankéríte	Error	Chi-squared
									CaCO ₃ (wt%)	(wt%)									
M1	xxx	×	×	×	51	0	24	24	81.3	77	1.11	5.4	1.5	10.6	0.34	7	0.78	1.73	
M2	xxx	×	×	×	59	7	8	26	68.4	59.4	1.04	15	1.19	20.8	0.5	4.8	0.72	1.77	
M3	xxx		×	×	51	30	5	14	81.5	89	1.1	5.4	1.13	5.4	0.29	–	–	1.6	
M4	xxx		×	×	58	2	14	26	88.5	88.9	0.78	6.1	0.8	4.9	0.18	–	–	1.52	
D5	xxx		xx	×	30	41	25	5	57.6	66.4	0.81	12.5	1.01	21.1	0.34	–	–	1.78	
D6	xxx	×	xxx	×	48	42	7	3	81.7	83.6	0.76	3.7	0.67	8.6	0.18	4	0.53	1.55	

Table 7 *A comparative table of the main characteristic parameters of the Dryas sediments and Acuminata marls: the grain size of the Dryas sediments is smaller, but most of this fraction is composed of quartz and calcite; natural oyster inclusions are present in Acuminata marls*

	Dryas sediments (D5 and D6)	Acuminata marls (M1–M4)
Main minerals	Calcite and quartz	Calcite
Main mineral clays	Illite and chlorite	Illite, smectite and illite/smectite mixed layer
Main granulometric fractions	Clay and silt	Coarse sand and gravels
Main minerals in matrix	Illite > 2 mm for D5; calcite and quartz < 2 mm for D6	Illite > 2 mm for M2
Coarse aplastic inclusions	Rare or few natural inclusions	The main granulometric fraction consists of natural inclusions

sample are larger than 2 μm , which is confirmed by thin-section analysis, where mica crystals of up to 150 μm were observed. The same observation is valid for sample D5. This sample contains 12.5 wt% illite (Table 6), but its finest grain-size fraction ($< 2 \mu\text{m}$) represents only 7.1 vol% of the bulk (Table 4). In this case, petrographic observations reveal the presence of large white mica crystals ($< 100 \mu\text{m}$). In sample D6, the situation is exactly the opposite: the illite concentration is only 3.7 wt% (Table 6), but the finest grain-size fraction represents 19.6 vol% of the bulk (Table 4). The difference may be explained by the contribution of non-clay minerals, such as quartz and carbonates, to the clay fraction. This result could very well explain the differential plastic behaviours of pastes during the forming process.

DISCUSSION

Table 7 summarizes the main characteristics of the marly Dryas limestones and the *Acuminata* marl sediments. Expanded clay minerals are only present in *Acuminata* marls. The grain size is lower in the marly Dryas limestones than in the *Acuminata* marls, but the fraction $< 2 \mu\text{m}$ is mainly represented by quartz and calcite. Natural aplastic inclusions are very abundant in *Acuminata* marls, whereas they are rare in Dryas sediments (Table 7).

Surprisingly, for both the *Acuminata* marls and the Dryas sediments, analyses show that the outcrops exploited by the Neolithic potters were not the best adapted for pottery making. Experiments and analyses suggest that D5 would be more suitable than D6, yet pottery fabric group B corresponds better, petrographically and chemically, with D6 (Fig. 2). In the same way, analyses show that M4.1 has the same chemical, palaeontological and petrographic characteristics as pottery fabric group A, whereas other samples share only the same petrographic and palaeontological characterization. Analyses and experiments suggest that M2 is a more suitable outcrop than M4.1 for pottery making, yet the Neolithic potters did not exploit it. One possible reason is that outcrop M2 is almost 10 km from lake-dwelling sites.

The lake of Chalain is located in an area where argillaceous sediments are rarely of good quality for pottery making (due to the high percentage of CaO and the low percentage of mineralogical clay). In this geological context, the sediments used by the potters of the lake dwellings represent extreme raw materials due to their very high percentage of calcium carbonate, their high inclusion content and their very low proportion of clay minerals. The CaO is higher in Dryas group B than in *Acuminata* group A, and SiO₂, Al₂O₃, K₂O and Fe₂O₃ are lower in group B than in group A. High amounts of CaO and discrepancies in the SiO₂, Al₂O₃ and K₂O compositions could explain the differential plastic behaviours observed between the two sediment groups.

High lateral facies variability of clay mineral proportions was observed in the *Acuminata* formation in Burgundy (Dumanois 1982) and in our Jura samples. Such high variability might explain the difference in plasticity and better workability of M2 and M4.1.

Archaeological experiments may be considered to be subjective. Nevertheless, our analytical results confirm the experiments using these raw materials in pottery forming. The sediment parameters influencing the plasticity and drying behaviour of the paste are better when using the *Acuminata* marls than the Dryas sediments. The D6 fraction $< 2 \mu\text{m}$ is mainly constituted of quartz and calcite. The high drying speed observed in experiments with Dryas sediments is a consequence of the high proportion of carbonate and quartz and the low amount of expandable clays in the finest grain-size fraction ($< 2 \mu\text{m}$). In marls, the presence of detrital elements plays a role in the plasticity: the higher the proportion of detrital fragments, the lower is the plasticity (Rémont 1967).

The use of Dryas sediments for the manufacture of earth structures used as fireplaces in houses was not a problem, because they do not need to be fired. Moreover, their forming is not comparable to that of pottery. The principal technical advantage of using *Acuminata* marls may be the presence of natural fossil oyster inclusions in the sediment, which represent natural inclusions in the matrix (Table 7). Can time be saved by using a more distant sediment with natural inclusions rather than a nearer clay without inclusions? Effectively, the time saved in paste preparation could be another reason to explain the transport of the *Acuminata* marls. The amount of time required when using a local clay, mixed with a local hand-crushed limestone, is a third higher than that required, including transport, when using *Acuminata* marls with natural inclusions. The Neolithic potters of Chalain would have had the choice between these two paste preparations. Nevertheless, the Dryas sediments are located immediately around the lake dwellings, at a shorter distance than the *Acuminata* marls (Fig. 4), and they contain natural inclusions. The time spent on the acquisition of this sediment is half that required for the *Acuminata* marls, and three times less than that needed for local clay with added limestone (Table 3). Nevertheless, the *Acuminata* marls, which are located 5 km from the dwellings, were used more often in two successive periods (Figs 4 and 6). Could this be a strong indication that the oyster-bearing raw material had some technical advantage that made the longer transport distance worthwhile?

Evolution in the use of raw materials obviously led to technical improvements in the production process. However, social and cultural aspects could also have played a role. During this period, potters also tried out Dryas sediments and made 26.9% of their pottery with this raw material over a short period of time (Fig. 6). Then, an immigrant population arrived at the lake of Chalain (Pétrequin 1993, 1997a,b, 1999; Arbogast *et al.* 1995, 1996; Pétrequin *et al.* 1998). This new population, from the Ferrières/Clairvaux culture (Giligny 1994, 1997), brought with them their own pottery, manufactured using a raw material consisting of a micaceous matrix containing granite rock fragments (petrographic groups 1, 3 and 4; Fig. 6). This kind of raw material is not present in the surrounding area and is only found at a distance of at least 50–60 km or more (Martineau 2000a; Martineau *et al.* 2000; Di Pierro and Martineau 2002a,b). The matrix of this pottery is clay rich, and was certainly better suited to pottery making than all the carbonate raw materials found around the lake of Chalain. The Neolithic potters must have known what ‘good pottery raw materials’ look and feel like. This technocultural heritage may have been the driving force that encouraged the Ferrières/Clairvaux potters to look for better raw materials in their new environment. For a short time, this new population used, in small proportions (12.5%), the same Dryas sediments as the Horgen culture, before turning their attention to new raw materials, such as the *Acuminata* marls (Fig. 6).

It is somewhat surprising that the search did not end at the Dryas sediment outcrop represented by sample D5, which is clearly better suited for pottery making than D6 and lies closer to the dwellings than the *Acuminata* marls (Fig. 4). Similar observations can be made for the Oxfordian sediments (group 10, Fig. 6), found 1.5 km from the Neolithic sites (Fig. 4), which were often used during the Horgen and Middle Clairvaux cultures (Fig. 6), but not during the Ferrières/Clairvaux and Early Clairvaux cultures.

When discussing this complex question, all parameters have to be taken into account. In the case studied here, it seems that the low proportion of clay minerals in the Dryas sediment matrix and the natural oyster inclusions in the *Acuminata* marls could be among the main reasons for their selection.

The complexity of the archaeological situation suggests that a combination of social, cultural and also technical factors may well have motivated the potters’ behaviour at Chalain, between

c. 3169 BC and 2980 BC (calendar years). Changes in pottery raw materials and style may sometimes be induced by cultural factors, with or without technical advantages. In this case, both technical data and sociocultural factors, simultaneously, are the driving forces for change (Martineau 2000a). As explained by Sillar and Tite (2000), interpretation of technical results cannot be dissociated from the study of social context. Our research shows the complexity of the problem and demonstrates the need for a series of analyses and experiments, which must be embedded in the archaeological and socio-economic context.

CONCLUSIONS AND PERSPECTIVES

We have shown that differences in the workability of two pastes during experimental pottery making can be quantitatively explained by chemical and mineralogical analyses of the raw materials. We focused our study on the raw material factors affecting paste plasticity. Experiments also show differences in other properties, such as the drying behaviour of the paste. The factor that most affects the drying behaviour is the clay mineral content, but pore-size distribution in the paste must also be taken into account. Oyster shells are known to have a high pore volume. Paste porosity measurements should provide more quantitative proof of the technical advantage of the *Acuminata* marls over the Dryas sediments. In the less clayey matrices, the increase in bulk porosity due to the porosity of oyster inclusions could be another important technical parameter.

We have focused our attention on factors influencing the production of pottery more than on those concerning its use and functional characteristics. It has been argued that shell inclusions, due to their platy shape, improve thermal shock resistance (Steponaitis 1984). Many other parameters, such as the increase in workability using shell or limestone, the geological environment, social factors and even the nature of the matrix (calcareous or non-calcareous), should be taken into account in discussing this question (Tite and Kilikoglou 2002). Tests of mechanical strength and thermal shock resistance should be performed on pottery from Chalain.

The results obtained in this study should be confirmed by further analysis. In order to characterize and quantify the nature of each grain size, it will be necessary to sieve sediments and extract each grain-size fraction for XRD and geochemical determinations. The objective would be to estimate the proportion of each mineral in each fraction.

Moreover, textural analyses of archaeological pottery will allow us to evaluate precisely the proportion of inclusions, and compare this amount with that found in natural sediments. The Neolithic potters could have sieved the sediments, or chosen outcrops containing a lower proportion of inclusions (Martineau 2000a). It will also be very interesting to measure mechanical properties, such as plasticity and drying speed, in order to quantify the differences in workability observed experimentally between the Dryas sediments and the *Acuminata* marls. The comparison of these mechanical properties for the two types of sediments will allow us to understand better the differences observed through experimental practice and mineralogical analyses.

To understand changes in pottery technique, elements such as the environmental context, sociocultural factors, pottery forming, past technical knowledge and raw materials used must all be taken into account by an experimental approach, and by physical and mineralogical analyses. All of these aspects must be embedded in well-known and well-studied archaeological contexts. In short, it is necessary to multiply observations and results before drawing conclusions about tendencies or theories. The best method would be to obtain enough data from

a series of case studies covering different periods and geographical areas, in order to construct new theories of technical, historical and social changes and material culture evolution. In this paper, we have documented the adaptations of the potter communities to their environment. The carbonate-rich geological context limited their raw material choices. Nevertheless, the archaeological data show clearly a succession of changes during the two centuries studied. We have documented these changes and discussed their implications. Further studies in other contexts will enrich our discussion of the underlying reasons for these raw material changes or choices.

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APPENDIX A

Chemical data of Dryas and Acuminata pottery and sediments: FeO_{tot} as Fe₂O₃; oxides in wt%; trace elements in ppm

Sample	Location	Pottery group	SiO ₂	TiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MnO	MgO	CaO	Na ₂ O	K ₂ O	P ₂ O ₅	Ba	Cr	Cu	Nb	Ni	Pb	Rb	Sr	V	Y	Zn	Zr	S _{UM}
M1	Le Fied 52		21.32	0.21	6.44	3.03	0.02	3.26	63.8	0.01	1.55	0.08	60	60	36	8	28	9	61	610	59	18	18	80	99.72
M2	Fay en Montagne 79		32.7	0.37	9.79	5.3	0.2	2.44	46.7	0.01	2.31	0.27	124	83	13	11	42	15	119	451	119	40	39	138	100.09
M2B	Fay en Montagne 79		29.72	0.33	8.49	4.66	0.15	3.00	51.54	0.01	2.04	0.25	110	78	12	11	36	14	106	474	93	37	33	131	100.19
M2C	Fay en Montagne 79		25.8	0.28	8.19	3.81	0.11	3.56	55.63	0.01	2.06	0.15	75	65	13	10	37	7	93	510	86	26	29	102	99.6
M2D	Fay en Montagne 79		14.36	0.15	4.95	4.02	0.04	1.65	73.66	0.01	1.00	0.18	44	54	15	6	31	12	36	451	79	30	28	70	100.02
M3	La Marre 76–77		40.24	0.52	13.47	7.5	0.35	2.26	31.18	0.01	2.89	0.32	167	115	11	13	61	19	169	334	167	55	48	157	98.74
M3 bis	La Marre 76–77		42.14	0.54	13.98	7.82	0.36	2.33	29.55	0.01	3.02	0.33	184	116	14	13	62	18	173	319	168	57	56	164	100.08
M4.1	Chatillon 70		31.03	0.4	12.65	10.39	0.08	2.38	41.22	0.01	1.72	0.29	163	115	12	13	59	25	111	233	176	76	408	125	100.17
M4.3	Chatillon 72		59.01	1.03	20.04	6.3	0.03	1.99	7.82	0.01	3.4	0.15	264	170	9	30	89	18	213	135	208	37	913	244	99.78
M4.4	Chatillon 73		15.19	0.14	4.69	3.41	0.02	1.5	74.2	0.01	0.86	0.13	49	50	5	8	35	8	27	554	57	19	35	64	100.15
D5	MLV 33 and 34		39.45	0.48	9.85	4.57	0.05	1.4	42.18	0.01	1.59	0.13	132	85	10	16	40	19	96	245	108	38	70	200	99.71
D6	Lake of Chalain 36		21.69	0.23	5.91	2.57	0.04	2.63	66.06	0.01	0.99	0.09	74	51	8	10	27	7	45	472	68	16	35	102	100.22
RM 2408		Group 12-1	32.34	0.47	13.86	3.23	0.08	1.54	46.1	0.01	1.69	0.3	177	86	16	14	51	12	72	431	101	29	91	124	99.62
RM 2210		Group 12-2	33.92	0.6	16.38	4.05	0.09	1.71	39.81	0.01	2.03	1.33	235	103	40	15	65	18	78	522	135	42	184	147	99.93
RM 2219		Group 12	49.45	0.91	22.34	5.22	0.08	1.94	15.66	0.12	3.71	0.89	382	135	43	21	88	27	149	239	153	41	295	182	100.32
RM 2253		Group 12-2	29.62	0.62	17.06	2.58	0.06	1.21	46.24	0.01	1.34	1.26	158	101	23	18	55	17	49	372	112	39	142	156	100
RM 2271		Group 12	32.7	0.46	13.34	3.2	0.07	1.65	45.99	0.01	1.77	0.49	187	80	36	13	47	7	80	466	104	31	99	122	99.68
RM 2290		Group 12	37.55	0.82	21.56	3.58	0.05	1.21	31.77	0.01	2.19	1.03	289	126	35	22	89	28	88	340	144	40	141	184	99.77
RM 3005		Group 12-1	38.59	0.63	17.48	4.67	0.13	1.68	34.13	0.01	2.1	0.6	257	108	28	17	66	18	95	583	127	36	178	148	100.02
RM 3017		Group 12	38.58	0.63	17.39	4.65	0.13	1.67	34.26	0.01	2.08	0.59	262	105	28	15	65	13	94	588	130	34	177	144	99.99
RM 4081		Group 12-3	32.35	0.45	13.19	2.61	0.06	1.62	46.63	0.01	2.17	0.95	225	87	25	15	54	7	96	500	96	30	92	121	100.04
RM 4119		Group 12-1	34.31	0.54	15.13	4.09	0.09	1.71	41.62	0.01	2.01	0.82	205	95	31	17	57	14	83	581	111	32	94	134	100.33
RM 4172		Group 12-3	32.74	0.55	15.78	3.14	0.09	1.5	42.29	0.01	2.00	2.16	302	91	25	15	60	10	75	791	99	32	85	139	100.26
RM 4182		Group 11	35.34	0.5	14.28	2.95	0.1	1.69	42.28	0.01	2.29	0.47	203	93	41	14	56	13	102	666	97	28	91	124	99.91
RM 3060		Group 19	25.61	0.3	7.2	1.98	0.03	1.44	61.97	0.01	1.02	0.37	113	59	29	9	33	12	45	357	75	25	442	127	99.93
RM 3068		Group 19	30.58	0.39	8.85	2.02	0.03	1.26	55.11	0.01	1.26	0.36	133	80	10	12	51	10	58	317	74	26	96	147	99.87
RM 3069bis		Group 19	20.69	0.25	5.98	1.53	0.03	1.36	68.43	0.01	0.75	0.62	99	55	32	11	33	12	34	398	58	20	195	113	99.65
RM 3115		Group 19	33.18	0.42	9.23	2.47	0.03	1.55	51.68	0.01	1.19	0.27	131	73	18	13	32	12	66	296	94	25	167	151	100.03
RM 3053		Group 19	31.76	0.37	7.28	1.8	0.03	1.35	57.52	0.1	1.1	0.16	58	58	38	10	27	12	55	334	68	18	103	136	101.47
RM 3061		Group 19	24.77	0.31	7.06	1.63	0.03	2.29	64.14	0.11	0.93	0.29	58	53	19	9	30	13	48	335	61	18	71	102	101.56
RM 3077		Group 19	22.53	0.24	5.26	1.56	0.03	1.31	69.45	0.12	0.83	0.36	21	43	13	9	26	10	35	343	49	18	82	110	101.69
RM 3080		Group 19	21.61	0.25	5.42	1.3	0.04	1.39	70.5	0.06	0.65	0.38	21	40	10	8	24	10	31	403	42	17	86	105	101.6

Petrographic and palaeontological analyses

The sediments and fired pottery pastes were observed under binocular and optical microscopes, using polarized light (Fig. 7). Binocular observation was used for the identification of large fossil inclusions. The determination of the oyster species allows the geological age of the sediments to be dated. The provenance of the raw materials was determined by comparison of archaeological ceramic samples with sediments from natural outcrops, following the method proposed by Tite (1999). About 1 kg of each sediment type, from 30 to 50 cm below the surface, was sampled.

Chemical analyses (XRF)

Ten major elements and 12 trace elements have been chemically analysed by XRF (Philips PW 2400). Oxides are in weight percent and trace elements in ppm. The error margin is 1% for major elements and 10% for trace elements.

FeO wt% was measured with dipyrilidil protocol, using the Philips Pye Unicam PU 8650 spectrophotometer.

Calcimetry

The percentage of CaCO₃ is deduced from the measure of CO₂ volume produced by destruction of sediment with HCl solution (30%). Quantities measured for the samples are related to a reference obtained on pure CaCO₃.

Grain-size analyses

Grain-size analyses of the matrix are rarely performed on archaeological clayey artefacts, because the technique is only valid before clay firing (Fig. 7). It is important to not confuse grain-size analysis of inclusions (natural or added) and grain-size analysis of matrix (clay, silt or sand, in the granulometric sense). This second type of analysis, here using the laser method, is rarely performed in archaeometry because of clay firing. Some textural or grain-size analyses have been performed in ethno-archaeological contexts (Gosselain 1994; Livingstone Smith 2000; Mercader *et al.* 2000).

Grain-size analyses (from 0.5 µm to 2 mm) were performed after extraction of the > 2 µm fraction, using a Laser Sizer Coulter LS32, after sieving at 2 mm. A few milligrams of each sample were placed in the LS32 vat, and dispersed by ultrasonic waves. During analysis, the laser ray is diffracted by particles and focused in a detector. An overestimate of the grain size of particles smaller than 2 µm may occur. Therefore, results obtained on the clay fraction (< 2 µm) cannot be considered as absolute values, but only as a way of comparing the different sediments studied.

In order to obtain detailed information on grain-size distribution, grain-size data were treated according to the method described by Wang *et al.* (1999), Lacroix *et al.* (2000) and Lesourd (2000). We considered the grain-size frequency spectra for the volume for a range of particle sizes from 0.5 µm to 2 mm. The populations of particles can be distributed over a number of lognormal curves, which allows the determination of each individual constituent.

Therefore, grain-size frequency spectra are sums of grain-size populations distinguished by their mode, and were characterized by the relative area of these populations, related to the bulk area of the grain-size distribution curve, and then compared one to another. A smoothed second derivative method was used in order to distinguish the grain-size populations and determine their mode.

X-ray diffraction

Bulk sediment and clay fractions were analysed by XRD. For the bulk sediment analyses, each powdered sample was analysed using a Philips PW 1710 diffractometer running between 3° and 65° 2θ at 40 kV and 20 mA, using Cu- K_α radiation, a Ni filter and a scan speed of 1° min^{-1} . In preparation for the XRD of the clay fractions, the samples were dispersed on de-ionized water, disintegrated, decarbonated with HCl (0.2 M) solution and washed several times before separation of the $< 2 \mu\text{m}$ fraction by settling in a water column. A preparation of oriented aggregates of clays was then collected on a glass slide. The samples were sequentially air-dried, glycolated and heated for 4 h at 490°C for identification of the clays; that is, the (001) basal reflections. Two samples were treated with hydrazine in order to identify the presence of kaolinite. The analyses were performed using the same conditions as for the bulk samples, but between 2° and 32° 2θ .

The Rietveld method

Rietveld refinements of the powder X-ray patterns allow quantitative information to be obtained about the mineralogical composition of the samples (Bish and Post 1993). The method consists of comparing the measured pattern with a calculated pattern that is based on a model provided by the user. The input model parameters—for example, the phases present, the proportion of each phase, the structure data, the instrumental parameters, the peak profile parameters and so on—are given approximate starting values that are varied in a least-squares procedure until the calculated pattern best matches the observed pattern. The SIROQUANT program (Taylor 1991) was used for the least-squares refinements. Zero shift, sample displacement and peak shape, modelled with a Pearson VII profile function, were the instrumental parameters, which were refined. The structure information for the present phases was taken from the SIROQUANT data bank, but only the unit cell parameters were refined. The atomic positions and the site occupancy were kept fixed during the refinement. This is acceptable, considering the usually small deviation from endmember stoichiometry observed for most of the present phases (quartz, calcite). The composition of the ankerite phase was estimated using the dependency of the lattice parameter on the iron content. The refined lattice parameters ($a = 4.830 \text{ \AA}$ and $c = 16.167 \text{ \AA}$) are compatible with the following stoichiometry: $\text{CaMg}_{0.5}\text{Fe}_{0.5}(\text{CO}_3)_2$. The phyllosilicate with 10 \AA layer spacing was modelled with an illite structure. The analysis of the clay fractions separated from the bulk sediment shows that the situation is more complex in the presence of smectite and mixed-layer illite-smectites. The peaks associated with these phases, however, do not show up in the diffractogram of the bulk material, indicating that they are present only in minor concentrations.