

# Testing technological practices: neutron activation analysis of neolithic ceramics from Valencia, Spain

Sarah B. McClure\*, Joan Bernabeu, Oreto García, Emili Aura, Lluís Molina, Christophe Descantes, Robert J. Speakman, Michael D. Glascock

*Department of Anthropology, University of Oregon, Eugene, OR 97402, USA*

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## Abstract

Using instrumental neutron activation analysis (INAA) of prehistoric pottery, daub, and modern clay samples from Valencia, Spain, we tested expectations on changes in raw material use with socio-economic shifts during the Neolithic (ca. 5600–2800 BC). Elemental analysis identified three distinctive clay source groups used by Neolithic potters. Contrary to expectations, a shift in raw material use was identified between the Early and Middle Neolithic despite general similarities in technological practices. In the Late Neolithic, pottery production became more specialized, but potters used the same range of clay sources documented earlier. This study illustrates the utility of INAA for testing hypotheses of prehistoric craft production.

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## 1. Introduction

Pottery was introduced into eastern Spain with the transition to subsistence economies during the 6th millennium BC. Regional developments of pottery styles and forms have provided the basis for a Neolithic chronology [5], and striking shifts in the quality of pottery through the course of the Neolithic mirror greater economic and social shifts visible in the archaeological record [5,6,34,35,37]. Recent technological analysis has shown that not only did pottery decorations change through time, but chronologically distinctive ceramic technologies were practiced by Neolithic potters as well [37]. It is within this context that a pilot study of the elemental composition of Neolithic ceramics from five sites in the Alcoi Basin, Valencia, Spain was conducted to test the hypothesis that changes in ceramic style and technological practices were linked with raw material availability and use.

## 2. Archaeological context

The introduction of pottery to eastern Spain is part of a larger economic and social phenomenon that occurred with the transition to agriculture in the western Mediterranean. By the 6th millennium BC domesticated plants (e.g., einkorn and emmer wheat, peas, lentils) and animals (sheep, goats, cattle and pigs) are documented at archaeological sites from Liguria, Italy to the Iberian Peninsula (see e.g. [4]). Pottery, specifically a distinctive shell-impressed style known as Cardial Ware, is found in concert with domesticates at sites throughout the Western Mediterranean.

Due to the effects of post-glacial sea level rise, modern coastal development, and archaeological traditions, the evidence for the earliest farming communities in the western Mediterranean tends to come from cave and rock shelter sites. Few open-air Early Neolithic villages are known to exist, but a limited number of sites in southern France and eastern Spain have been excavated (see e.g. [4]). Because of the scarce documentation of village sites, it has long been assumed that early farming communities were more ephemeral on the landscape than their counterparts in the eastern Mediterranean. However,

\* Corresponding author. Tel.: +1 541 346 5101.

*E-mail address:* [sbm@uoregon.edu](mailto:sbm@uoregon.edu) (S.B. McClure).

current research at the open-air village site of Mas d'Is in the Alcoi Basin in Valencia, Spain is challenging that assumption [11,12]. Evidence of household structures and monumental ditches indicate a significant degree of labor investment. In addition, subsistence data from the limited numbers of open-air villages in the western Mediterranean are in agreement with data from caves and rock shelters with Early Neolithic occupation in Valencia, suggesting a reliance on agricultural products that exceeds 50% of subsistence requirements (e.g., La Draga in Catalonia [16]). In contrast to other regions of the world, it appears that the early farming societies in eastern Spain were committed to agricultural production and cannot be considered low-level food producers (*sensu* Smith [52]).

By the 3rd millennium BC a different kind of Neolithic economy is documented. Greater numbers of villages are known to exist during this period and their locations in more agriculturally marginal areas suggest demographic expansion. Shifts in farming technologies include the introduction of the plow and a reorientation of animal management practices [6,13,38]. Long-distance exchange of polished stone axes is well documented [43], as is the specialized production of at least some stone tools, personal ornamentation, and bone idols [24,46]. The Early Neolithic burial custom of single inhumations in caves or rock shelters is replaced by collective burials of individuals in similar locations but with varied quantities of grave goods [9,44,45]. In short, Late Neolithic society is marked by increased population density, more intensive agriculture, long-distance regional exchange, semi-specialized craft production, and emergent social differentiation.

### 2.1. Neolithic ceramics

The introduction of pottery during the 6th millennium BC was not limited to a single pottery style or type. Despite the emblematic nature of Cardial Ware as a marker of the Early Neolithic throughout the western Mediterranean, it was just one of several distinctive decorative styles produced by potters during this period. Indeed, the diversity in decorative styles from the Early Neolithic onwards provides the basis for the regional chronology ([5]; Table 1). Phases of the Early Neolithic (Neolithic Ia and Ib) are determined by the relative percentage of decorative styles such as Cardial impressed, other impressed, incised, and undecorated wares. The Middle Neolithic (Neolithic Ic and IIa) witnessed the appearance of a distinctively carved pottery type known as *esgrafiada*, and the complete disappearance of Cardial Ware. Finally, the Late Neolithic (Neolithic IIb) is characterized by the decline of decorated wares. Only a few vessels uncovered from this period are decorated with incised lines. Furthermore, the Neolithic IIb witnessed a shift in the form of pottery. Whereas Neolithic I and IIa pottery consisted predominantly of closed shapes, Neolithic IIb pottery includes many open forms such as plates and platters. This shift has been likened to a change in pottery function from primarily storage to food preparation and serving [5,6].

Technologically, pottery production during the Neolithic has been shown to undergo numerous changes [37]. Based

Table 1  
Neolithic chronology in Valencia (after [11: 174])

Period	Phase	Characteristic ceramic type
Neolithic I: ca. 5600–4500 cal BC	NIa: ca. 5600–5300 cal BC	Cardial ceramics dominate assemblages
	NIb: ca. 5300–4900 cal BC	Incised and impressed ceramics (non-Cardial) dominate assemblages
	NIc: ca. 4900–4500 cal BC	Combed ceramics. Incised and relief decorations constitute <5% of assemblages
Neolithic II: ca. 4500–2800 cal BC	NIIa	Carved ( <i>esgrafiada</i> ) ceramics
	NIIb	Undecorated ceramics. Emergence of open forms (plates, platters)
Bell Beaker Horizon (Horizonte Campaniforme Trascional) after 2800 cal BC	HCT	Bell Beaker Ware. Copper metallurgy

on the analysis of over 500 Neolithic vessels from in and around the Alcoi Basin, Early Neolithic pottery was identified as not only stylistically diverse, but also made from a variety of different clay fabrics [37]. Grog, crushed pottery, was the primary tempering agent in Early Neolithic pottery (constituting ca. 70% of Neolithic Ia pottery), although calcite and quartz were also commonly used. A shift in the use of tempering agents is seen during the Middle Neolithic (NIIa), when the practice of tempering with grog disappeared, and all vessels, regardless of decorative style, were tempered with calcite. This practice remained intact during the Late Neolithic (NIIb) when, despite changes in pottery style and form, the only tempering agent used was calcite.

In addition to shifts in tempering agents, pottery production underwent other technological changes in the course of the Neolithic. By estimating the degree of labor and time input in pottery production based on the relative expediency of each step along the production sequence of a vessel (following [21,29]), distinct shifts between the Early and Late Neolithic have been documented [37]. A point system was used to characterize the relative labor investment in each of the Neolithic vessels analyzed in the technological study to compare vessels within and between sites as well as time periods [37]. Based on this system, each variable along the production chain received a point, and more labor-intensive links scored higher. For example, a smoothed exterior surface received 1 point, whereas a surface that was well smoothed (*espatulado*) received 2 points and a burnished or polished surface received 3 points. A similar point structure was used for all identifiable stages of the production sequence. This approach served only as a proxy of time or labor investment, since key factors, such as distance to clay source, vessel size, and inclusion source were not taken into account. However, qualitative differences in vessel treatments were quantitatively measured, and averages were established. By comparing vessels to these averages, a better sense of the impact of ceramic variability on time allocation and labor investment was made [37]. Based

on this approach, pottery from the Early and Middle Neolithic (Neolithic Ia–IIa) was relatively high in labor investment. In contrast, Neolithic Ib vessels were made much more expediently and they are more standardized within and between sites. It should be noted that the shift in labor investment follows the pattern of stylistic shifts and not changes in the tempering agents used in manufacture.

Pottery during the Early Neolithic is relatively homogenous in its production within households, but exhibits variability between households and sites, suggesting that potters during the Early Neolithic produced their wares within households following household traditions [37]. In contrast, Late Neolithic pottery production was very homogeneous within and between households as well as between sites, and suggests that it was more specialized, with fewer people within a village manufacturing pottery for the entire community [37]. Unfortunately, the organization of pottery production during the Middle Neolithic remains unclear, largely due to the general lack of excavated Middle Neolithic open-air sites in the region. The archaeological record clearly indicates significant social and cultural shifts during the course of the Neolithic and the co-occurrence of changes in pottery manufacture and the organization of production. However, it remains unclear how cultural and environmental factors intertwined and influenced technological practices. Are differences visible in Neolithic pottery due to shifts in storage or cooking technologies, symbolic or ritual value, in the materials potters had available, or a combination of factors? In the context of changing land use and increasing population densities during the Neolithic, the central question addressed in this paper is to what extent can shifts in pottery technology be attributed to shifts in raw material sources, specifically clay sources? Elemental analysis using instrumental neutron activation analysis (INAA) was employed to test this question.

## 2.2. The samples

A total of 56 samples of prehistoric pottery and daub from five Neolithic sites from the Alcoi Basin (Fig. 1, Table 2) were selected for geochemical analysis. An additional four samples

of clay from modern deposits in the Alcoi Basin were included in the analysis. Due to the destructive nature of INAA, only few samples of ceramic vessels were available for analysis. Samples were selected to represent the assemblage at each site; however, the sampling strategy was modified based on the availability of sherds of sufficient size to allow a sample to be taken.

The Alcoi Basin in the northern Alicante Province, Valencia, is characterized by five interconnected valleys (Vall de Penaguila, Polop Alto, Serpis, Vall de Alcalá, and Vall de Ceta) that are surrounded by mountains. The calcareous Sierra del Benicadell (1104 m) in the north is followed to the east by the Carrasqueta, Safor, Mustalla, Almirall and Gallinera mountains that continue to the coast. To the east is the Jurassic fold of the Sierra de Mariola (1390 m), while the Sierra de Aitana (1558 m) closes the Basin to the south. These mountains form the northern extent of the Baetic mountain system that spans the southeastern Iberian Peninsula.

The geological deposits are characterized by a succession of folds oriented ENE–WSW, often breaking inverse faults. Geomorphologically and topographically this situation results in parallel alternating mountains and valleys. The mountains are formed by outcrops of limestone and cretaceous dolomite, whereas the valleys are synclinal depressions filled with thick neogenic materials [22: 13]. Open-air sites are located on terraces in the valley bottoms, whereas cave and rock-shelter sites are situated on the mountainsides.

### 2.2.1. Early Neolithic samples

A total of 18 samples from three Neolithic Ia contexts were analyzed. Mas d'Is is the only open-air village dating to the Early Neolithic to be excavated in Valencia. The site is located on a strip of land at the headwaters of the Penaguila River, a tributary of the Serpis River. A number of structures were excavated dating to the Early Neolithic, including evidence for three houses. Of these, House 2 in Sector 80 and House 3 in Sector 52 are thought to be contemporaneous (i.e., NIa; [11: 178]). Eight samples, including Cardial impressed, incised, decorated cordon and undecorated vessels, were chosen from the Neolithic Ia phases of Sector 80.

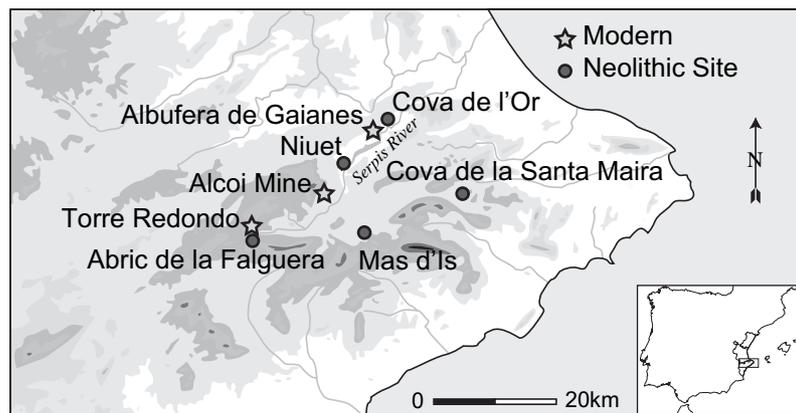


Fig. 1. Location of Neolithic sites and modern clay sources analyzed.

Table 2  
Summary of INAA samples by site

Site	Period	# Samples	Sample type
Cova de l'Or	NIa	3	Pottery
Mas d'Is	NIa	8	Pottery
	NIa	3	Daub
	NIa	4	Pottery
Abric de la Falguera	NIa	7	Pottery
	NIb	6	Pottery
	HCT	3	Pottery
Santa Maira	NIa	9	Pottery
	HCT	1	Pottery
Niuet	NIb	10	Pottery
	NIb	3	Daub
Torre Redondo	Modern	2	Clay
Albufera de Gaianes	Modern	1	Clay
Alcoi clay mine	Modern	1	Clay

The site of Cova de l'Or is inarguably one of the most important Early Neolithic sites on the Iberian Peninsula due to the large quantities of Cardial ceramics, bone tools, personal adornments and domestic plant and animal remains recovered (e.g. [4,32,33,35,36,39,57–60]). The cave is located at an elevation of 650 m on the northern edge of the Serpis valley on the south-east slope of the Sierra del Benicadell. The importance of the materials found at Or lies not only in the impressive quantity and quality of materials uncovered, but also in the significance of these materials for building regional chronologies, typologies, and models for the transition to agriculture (e.g. [5–9,35]). Due to the destructive nature of geochemical analyses, only three samples from the Neolithic Ia levels, all Cardial impressed, could be included in this study.

Finally, seven samples were analyzed from the Neolithic Ia levels at Abric de la Falguera, a small rock shelter located in the Serra Menetjador in the Polop Valley, one of the upland valleys of the Alcoi Basin. Located at ca. 800 m, the site affords a panoramic view of the higher elevations of the Polop Valley. Excavations in 1981 [50] and 1998–2001 [24–27] resulted in the documentation of human occupation at the site from the Mesolithic, Neolithic, Bronze Age, and Roman periods. The Neolithic levels consist of a series of phases that contained ash and charcoal lenses including hearths and larger burned areas. The larger areas of fire use are interpreted as animal-corrals, fires, used to clean corrals after animals had been penned for a prolonged period [17,26]. The samples consisted of vessels with decorated cordons, incisions, as well as undecorated wares.

### 2.2.2. Middle Neolithic samples

A total of 13 samples dating to the Neolithic Ic/IIa were analyzed from Mas d'Is and Cova de la Santa Maira. In addition to the house structures mentioned above, several trenches were found during excavation at Mas d'Is. A Middle Neolithic V-shaped trench, Foso 4, was uncovered [11], and four samples consisting of vessels with a plain and a decorated cordon and combed decorations were obtained from the mid-fill levels of Foso 4 dating to the Middle Neolithic (NIc/NIa) [37].

Cova de Santa Maira is located at an elevation of 600 m in the Sierra d'Alfaro. It is a sub-triangular cave with an interior gallery and with three exterior openings and evidence for human occupation spanning the Upper Palaeolithic to Bronze Age [2,20]. The east opening currently has dry stone walls, testifying to its historic use as a corral. In the Neolithic levels, typical Middle Neolithic decorated ceramics (incised, applied, *esgrafiada* and combed) and a small number of lithics were found associated with domestic sheep and goat bones. The presence of characteristic charcoal lenses in the two sectors may be evidence of use as corral during the Middle Neolithic [2: 80,3,55]. A total of nine samples of Neolithic IIa pottery were analyzed and consisted of three plain cordons, three *esgrafiada*, two combed and two undecorated vessels. One Bell Beaker vessel (HCT) was included in the sample for comparative purposes.

### 2.2.3. Late Neolithic samples

Sixteen samples from two Neolithic IIb sites were included in the analysis. Niuet is situated on a fluvial terrace between the Río Serpis and the Barranc de la Querola, in the mid-valley by the modern town of L'Alqueria d'Asnar. A large part of the terrace has been eroded by the Serpis River and pebble mining since the 1960s [23]. Consequently, only parts of the original site were preserved in three areas, and archaeologists from the Universitat de València conducted a salvage excavation from 1988 to 1993. One large sector (Sector A) was excavated along with a number of structures visible on the surface or in erosion cuts [47]. Features, such as pits, ditches, and a house floor on the uppermost level were recorded [14]. Numerous pottery fragments were found in excavation units and on the surface and analyzed typologically by Bernabeu and Orozco [10]. The ceramic assemblage was very similar to material found at other Late Neolithic sites in the region and consisted primarily of undecorated ware. In general, simple forms of open or slightly closed vessels, especially plates and shallow dishes, dominate the deposits. Ten samples of undecorated Neolithic IIb pottery were analyzed from Niuet. Abric de la Falguera also contained several levels of Late Neolithic occupation, and five samples of undecorated ware and one of an incised vessel were analyzed from the Neolithic IIb contexts. In addition, three samples from the Bell Beaker Transition (HCT) were included for comparative purposes.

### 2.2.4. Daub and modern clay

Given that ceramic production necessitates the selection and procurement of raw materials, modern clay and samples of pre-historic construction material (daub) were analyzed to determine, if possible, the sources of raw materials used in the manufacture of pottery. In the Alcoi Basin, human-induced alterations of the landscape through time have had serious environmental impacts such as the down cutting of riverbeds, erosion, and infilling of lakes. For this reason, a large-scale collection of samples of modern clay sources is not possible. Four clay samples were taken from historic clay mines in the Alcoi valley (Fig. 1; Torre Redondo by Abric de la Falguera in the Polop Alto Valley and Alcoi clay mine outside the city of Alcoi in the upper Serpis Valley) and from a known former lakebed

(Albufera de Gaianes in the lower Serpis Valley). Where applicable, prehistoric clay building material (daub), likely procured in proximity to the village site, provides a proxy for locally available clay. Three daub samples were analyzed from Neolithic Ia levels at Mas d'Is and Neolithic IIB levels at Niuët, respectively.

### 3. Instrumental neutron activation analysis (INAA)

Geochemical analysis of archaeological ceramics provides a means to assess the use of raw materials through time as well as local and long-distance interaction. Studies of this type have been conducted in a range of cultural contexts and geographic regions (e.g. [19,30,31,42,48,53,54]), and have resulted in a greater understanding of both technological practice and human interaction. Geochemical analysis was chosen here to help illuminate the following question: Did potters choose different raw material sources during the course of the Neolithic mirroring shifts in settlement locations? If so, the chemical characterization of ceramic samples should reveal differences in pottery composition between villages in the Late Neolithic as a consequence of the use of different clay sources.

The use of chemical composition for identifying the source areas of clays used in prehistoric pottery production is not unproblematic. In general, the basic, underlying proposition is that different sources will have distinct chemical compositions (also known as the 'provenience postulate' [56: 24,41]). To counter these issues, many archaeologists have approached the sourcing of prehistoric pottery in a different manner (e.g. [40,51]). With the analysis of large samples of pottery, compositional or reference groups of chemically similar potsherds are created. Also known as the 'criterion of abundance' [15], it is assumed that a homogeneous compositional group of unknown derivation was produced at the site or region where it is most represented archaeologically [41]. If modern clays are available, researchers only attempt to assign the pottery compositional groups to geographical locations based on compositional similarities (as opposed to exact matches) to clays that have been collected from the study area [15]. Due to the problems with assigning compositional groups to geographical areas (see [1,40,41,49,53]), a cautious approach in the interpretation of compositional groups should be taken. In addition, potters may select chemically distinct clays from the same areas because of their differences in functional properties. In this case, pottery from chemically distinct compositional groups may still be locally derived. For these reasons, the present analysis focused on the prehistoric pottery sherds. Compositional groups were then compared to the prehistoric daub and modern clay samples.

### 4. Methods

Samples were prepared for INAA using standard practices at the Missouri Research Reactor (MURR) (see [28,41] for detailed discussions of sample preparation, irradiation, and data analysis). Sub-samples were taken for long and short radiations along with reference standards, and the clay samples were fired in a kiln at 700 °C for 1 h. The short irradiation

is conducted through the pneumatic tube irradiation system, and the 720-s count generates gamma spectra with peaks for the short-lived elements aluminum (Al), barium (Ba), calcium (Ca), dysprosium (Dy), potassium (K), manganese (Mn), sodium (Na), titanium (Ti), and vanadium (V). The second sub-sample is subjected to a 24-h irradiation. Subsequently, these samples decay for 7 days and are then counted for 2000 s, also known as the 'middle count'. This yields determinations of seven medium half-life elements: arsenic (As), lanthanum (La), lutetium (Lu), neodymium (Nd), samarium (Sm), uranium (U), and ytterbium (Yb). Finally, after an additional 3- or 4-week decay, an 8500-s count is conducted. This last count yields the 17 long half-life elements cerium (Ce), cobalt (Co), chromium (Cr), cesium (Cs), europium (Eu), iron (Fe), hafnium (Hf), nickel (Ni), rubidium (Rb), antimony (Sb), scandium (Sc), strontium (Sr), tantalum (Ta), terbium (Tb), thorium (Th), zinc (Zn), and zirconium (Zr). For each sample analyzed, the density of an element in parts per million (ppm) was calculated. A correction was applied to sherds with calcium concentrations greater than 1% (see [18]) and subsequently the elemental concentrations of calcium (Ca), manganese (Mn), and sodium (Na) were eliminated. Furthermore, nickel (Ni) was eliminated because of its low detection limits. In total, 29 elements were used to identify compositional groups.

### 5. Results

The majority of pottery samples (73%; 36 out of 50) were assigned to three discrete compositional groups (Groups ES1, ES2, and ES3) consisting of 14, 15, and 7 samples, respectively. A total of 27% of the samples was unassigned. Pottery assigned to Group ES1 ceramics is characterized by higher arsenic (As) and rubidium (Rb), whereas Group ES3 expresses samples enriched in strontium (Sr) and depleted in arsenic and rubidium. Group ES2 members have concentrations that lie intermediate between Groups ES1 and ES3 (Table 3). The groups are represented graphically in Fig. 2 by bivariate plots of (a) principal components 1 and 2, and (b) principal components 1 and 6. A similar separation of the groups is shown in Fig. 3A, a bivariate plot of strontium and lanthanum concentrations. In the following, the significance of the INAA results is discussed by time period.

#### 5.1. Neolithic Ia pottery

A total of 18 samples from three Neolithic Ia contexts were analyzed using INAA. Samples from Mas d'Is ( $n = 8$ ) were obtained from Sector 80. The cave sites represented in the analysis are Abric de la Falguera ( $n = 7$ ) and Cova de l'Or ( $n = 3$ ). Despite the small sample size, it is important to note that only 27% ( $n = 5$ ) of the Neolithic Ia sample was not attributed to an identified source group. Table 3 summarizes the distribution of the INAA groups by site. As is readily apparent, none of the Neolithic Ia ceramics fall into Group ES3, with the majority of vessels in Group ES2. There is little difference between the rock shelter of Falguera and the open-air site of Mas d'Is. The higher representation of ES1 at Cova

Table 3  
Descriptive statistics of INAA data for three groups of Neolithic ceramics identified in this study

Element	ES1 (N = 14)			ES2 (N = 15)			ES3 (N = 7)		
	Mean (ppm)	Std. Dev. (ppm)	%CV	Mean (ppm)	Std. Dev. (ppm)	%CV	Mean (ppm)	Std. Dev. (ppm)	%CV
As	6.25	3.52	56.35	4.93	3.40	68.80	3.11	2.97	95.55
La	25.57	7.33	28.66	20.04	6.24	31.15	17.30	4.34	25.09
Lu	0.29	0.10	33.17	0.23	0.08	34.33	0.19	0.05	27.21
Nd	22.97	6.23	27.12	18.11	6.04	33.34	16.32	5.08	31.13
Sm	4.66	1.19	25.58	3.60	1.21	33.69	3.31	1.20	36.20
U	2.07	0.50	23.99	1.77	0.67	37.67	1.54	0.33	21.75
Yb	2.07	0.68	32.72	1.65	0.58	35.06	1.32	0.37	28.15
Ce	52.93	14.45	27.29	40.04	14.58	36.43	36.29	10.78	29.63
Co	8.06	1.59	19.75	7.74	3.26	42.10	6.11	1.84	30.11
Cr	62.24	19.77	31.77	58.24	19.53	33.53	42.10	7.73	18.35
Cs	7.08	1.67	23.57	4.60	2.18	47.27	4.98	2.03	40.79
Eu	0.88	0.21	23.77	0.72	0.24	33.78	0.64	0.24	37.09
Fe (%)	2.56	0.74	29.28	2.44	0.99	40.81	1.96	0.54	27.92
Hf	4.38	2.04	46.56	3.06	1.38	45.18	2.44	0.60	24.63
Ni	7.22	18.76	259.98	15.41	29.69	192.61	13.47	17.63	130.90
Rb	95.25	21.34	22.40	67.25	29.62	44.04	69.69	20.92	30.02
Sb	0.72	0.27	37.34	0.52	0.26	50.64	0.57	0.37	65.03
Sc	9.74	2.66	27.35	8.81	3.48	39.53	6.63	1.32	19.93
Sr	141.88	34.97	24.68	459.79	132.15	28.74	1246.11	191.09	15.33
Ta	0.98	0.34	35.16	0.77	0.33	43.37	0.63	0.14	21.59
Tb	0.63	0.20	30.82	0.50	0.18	35.83	0.44	0.13	30.18
Th	9.42	3.09	32.82	7.10	3.09	43.53	6.04	0.98	16.27
Zn	56.06	19.73	35.21	56.14	9.32	16.60	42.04	12.29	29.24
Zr	107.71	61.68	57.26	79.03	28.71	36.33	74.15	20.02	27.00
Al (%)	5.85	1.39	23.89	5.16	2.33	45.12	4.14	1.12	0.05
Ba	336.41	202.43	60.17	379.13	242.50	63.96	370.17	241.91	65.35
Ca (%)	16.66	5.52	33.12	18.91	7.98	42.21	22.44	3.02	13.47
Dy	3.60	1.02	28.42	2.89	1.09	58.15	2.37	0.79	33.42
K (%)	2.35	0.71	30.09	1.54	0.89	43.67	1.48	0.61	41.24
Mn	181.90	87.38	48.04	275.17	120.16	43.67	209.16	34.48	16.49
Na (%)	0.18	0.11	65.16	0.13	0.07	52.79	0.09	0.03	37.85
Ti (%)	0.33	0.11	35.42	0.27	0.10	40.60	0.19	0.03	17.95
V	73.71	22.01	29.87	66.27	24.06	36.31	49.69	12.14	24.42

de l'Or is potentially inflated due to the small sample size from that site. A number of vessels from both Mas d'Is and Falguera were not classifiable (UNC), suggesting that part of the pottery found at these sites was made with a variety of distinct clay raw materials. Finally, six of the Neolithic Ia vessels analyzed were Cardial Ware, and these fell into all three groups. Cardial vessels from Cova de l'Or are members of ES1 and ES2. In the case of Mas d'Is, Cardial vessels are members of ES2 and unclassified. Increased elemental analysis of Neolithic Ia vessels from these sites is necessary to test these patterns.

### 5.2. Neolithic Ia pottery

In contrast to the Neolithic Ia vessels analyzed, samples from Mas d'Is Foso 4 ( $n = 4$ ) and Santa Maira ( $n = 9$ ), dating to the Neolithic Ia, show a very different distribution between INAA groups (Table 4). Santa Maira has vessels that fall within the Groups ES1, ES3, and unclassified (UNC). The majority of Mas d'Is Foso 4 vessels are members of ES3. The presence of INAA Group ES3 in these samples is a shift from the Neolithic Ia material, where ES3 is not represented. It appears, therefore, that ES3 is a clay source that was first used during the Neolithic Ia. In addition, it is important to note that Santa Maira, a cave site, appears to have a greater diversity of

clay sources represented in the sample than the open-air site of Mas d'Is. This is not surprising given the use of Santa Maira as an animal corral, likely by a range of people over time. What is surprising, however, is that the Mas d'Is material from the Neolithic Ia is strikingly distinct from the Neolithic Ia material from the same site.

### 5.3. Neolithic Ib pottery

The Neolithic Ib sample analyzed by INAA consists of 16 vessels from Abric de la Falguera ( $n = 6$ ) and Niuet ( $n = 10$ ). All three groups are represented in the sample, although the sample from Abric de la Falguera does not include ES3 (Table 4). Indeed, in the case of Falguera it is important to note a shift in INAA group representation between the Neolithic Ia and Ib. Although both groups are present in both periods, the proportion shifts from a majority of ES2 vessels during the Neolithic Ia to ES1 during the Neolithic Ib. In addition, it is noteworthy that within the Neolithic Ib assemblage studied here, the open-air site of Niuet has a greater variety of clay sources than the rock shelter of Falguera. This may suggest that the use of Falguera as an animal corral was limited to a certain group or groups of people during the Neolithic Ib.

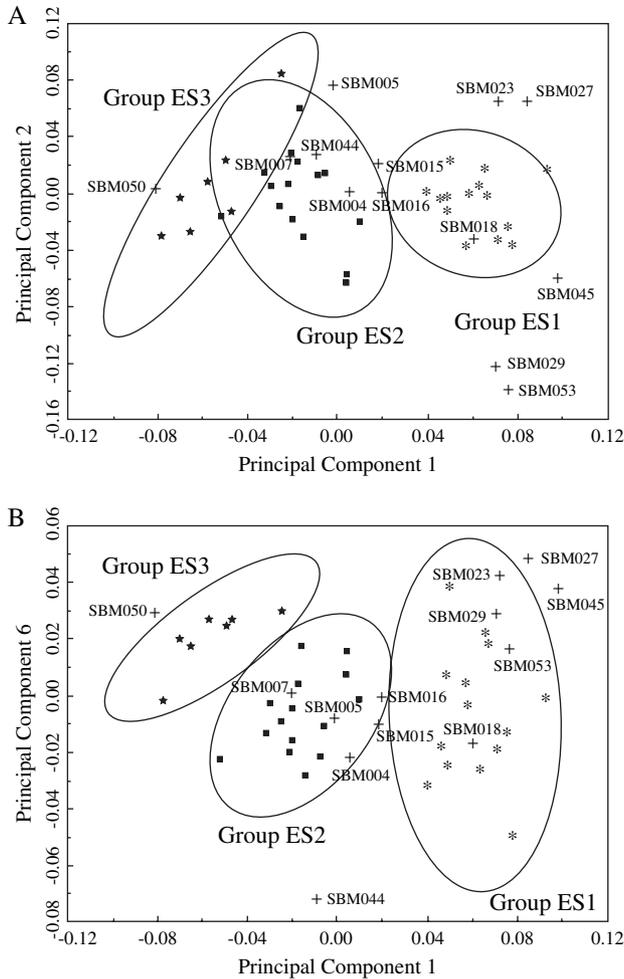


Fig. 2. Bivariate plot of (A) principal components 1 and 2, and (B) principal components 1 and 6 for Neolithic pottery (three groups) and unassigned samples (+). Ellipses represent 90% confidence level for group membership.

#### 5.4. Modern clay and prehistoric daub

A small sample of prehistoric daub and modern clay were also analyzed (Table 5). The elevated strontium concentrations found in the clay and daub samples resemble ES2 and ES3 members (see also Figs. 3B and 4). The enrichment of strontium in historic clay samples suggests that this is likely a characteristic of the region's clays. Due to the low levels of strontium in the ES1 group, it is therefore possible that ES1 may be a non-local group. Further samples of modern clays from the region will permit testing of this idea.

Table 5 presents the possible attribution of clay and daub samples to the chemically distinct groups found for the prehistoric pottery. The samples here are not part of these groups, however the position in the bivariate plot of the 1st and 6th principal component were used as a proxy for comparison to the elemental groups established for the pottery (Fig. 4). The daub samples from Mas d'Is Sector 80 (SBM058 and SBM060) are both outside of the 90% confidence levels for each group. In contrast, the sample from Mas d'Is Foso 4 (SBM059) falls into the ES3 realm. This difference in

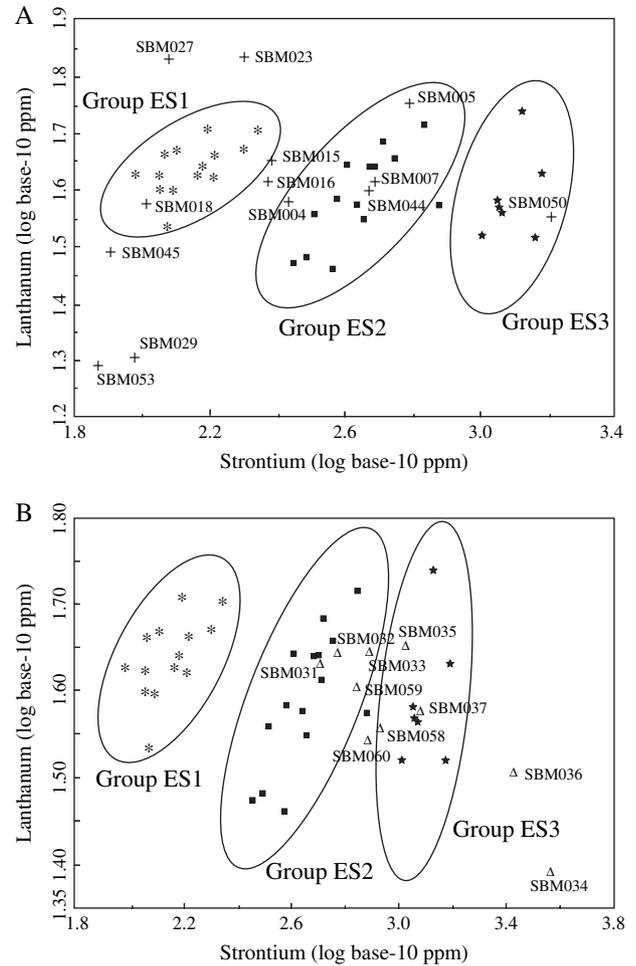


Fig. 3. Bivariate plot of strontium and lanthanum concentrations for neolithic Spanish ceramics (three groups) and (A) unassigned samples (+) and (B) clay and daub samples ( $\Delta$ ). Ellipses represent 90% confidence level for group membership.

construction material is particularly interesting considering the sudden appearance of the ES3 group in Neolithic IIA pottery. A problem arises, however, when the bivariate plot of strontium and lanthanum (Fig. 3B) is used to assess group proximity. Instead of following the pattern found among the prehistoric pottery, the daub is taken from Group ES3 in the Neolithic Ia and from ES2 in the Neolithic IIA. If correct, this pattern would also be interesting, in that it suggests that regardless of time period, people living at Mas d'Is made a distinction between clay for potting and clay for house construction, since in neither case are both groups in use for potting and daub at the same time. It is possible, therefore, that there was a shift in clay source preference (if both were readily and locally available). Testing these two possibilities can only be done in the future if a greater number of samples (pottery and daub) from these periods are analyzed.

Finally, it should be noted that the clay samples from Torre Redondo (SBM034 and SBM036) lie well outside of any identified compositional group (Figs. 3B and 4). Torre Redondo is a historic clay mine in the Polop Alto Valley with the earliest documented use during the Roman period and it served as

Table 4  
Results of the INAA analysis by time period and site (Cardial vessels in **bold**)

Period	Site	Clay Group			
		ES1	ES2	ES3	UNC
Neolithic Ia	Mas d'Is	1	4/1	0	3/2
	Abric de la Falguera	1	4	0	2
	Cova de l'Or	<b>1</b>	<b>2</b>	0	0
Neolithic Ic/Iia	Mas d'Is	0	0	3	1
	Foso 4				
	Santa Maira	4	0	3	2
Neolithic Iib	Niuet	3	3	1	3
	Abric de la Falguera	3	1	0	2

a source of clay for local roof tile production until the 1950s (Ortiz i Gisbert, personal communication to S.B.M., 2000). Samples from this site were included in the analysis because of its proximity to Abric de la Falguera. It is therefore interesting to note that the material from Falguera falls mostly into the compositional Groups ES1 and ES2, with seemingly no relationship to the elemental composition of the Torre Redondo samples.

## 6. Conclusions

The INAA analysis of Neolithic pottery from the Alcoi Basin was successful in identifying three distinct chemical compositions of pottery. In the Early Neolithic (NIa), potters used clay Groups ES1 and ES2 to make their pottery. A shift is visible in the Middle Neolithic (NIc/Iia), when data indicate the use of ES1 and ES3 in pottery production, but there is no evidence of ES2 in the sample. This pattern in clay source from the Neolithic Ia to the Neolithic Iia is intriguing. Can it be understood by changes in land use during the period? Unfortunately, our knowledge of Middle Neolithic land use and village settlement is limited, so it is unclear to what extent it may have differed from the Early Neolithic. An interesting similarity, however, lies in the identification of ES1 in both time periods. If indeed this clay group is non-local, as the modern clay composition suggests, it indicates that the use of this clay source continued through time despite changes in other clay procurement activities. It should be noted, however, that no systematic characterization of the natural variation in clays within the region has been conducted. It is therefore possible that less calcareous,

Table 5  
Modern clay and daub samples with group proximity based on the bivariate plot of principal components 1 and 6 and strontium-lanthanum concentrations (in **bold**)

Sample location	Type	Clay group			
		ES1	ES2	ES3	UNC
Mas d'Is Sec. 80	Daub			<b>1</b>	2/1
Mas d'Is F4	Daub		<b>1</b>	1	
Niuet	Daub		3/3		
Torre Redondo	Clay				2/2
Albufera de Gaianes	Clay		1		<b>1</b>
Alcoi Clay Mine	Clay			1/1	

Note the differences in the material from Mas d'Is and Albufera de Gaianes.

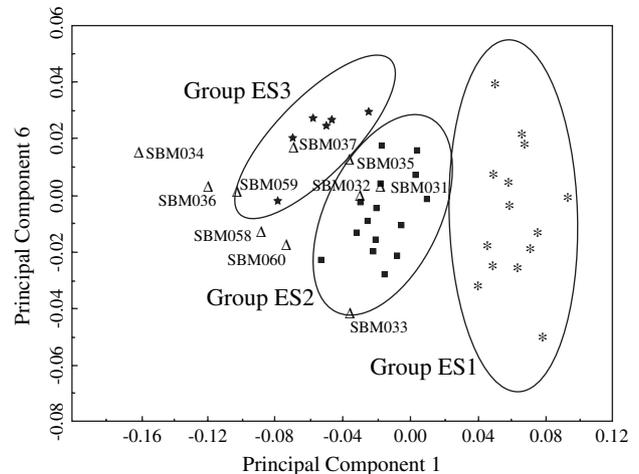


Fig. 4. Bivariate plot of principal components 1 and 6 for Neolithic ceramics (three groups) and clay and daub samples ( $\Delta$ ). Unassigned samples are not shown. Ellipses represent 90% confidence level for group membership.

low-strontium clays may be available locally. Alternatively, since the differences between the three groups are quite subtle, they could be the result of some clay-processing effect such as enrichment through tempering or clay refinement. In other words, differences in clay groups identified through INAA may not be source-related. These hypotheses can and should be tested with expanded samples both of the prehistoric pottery and modern clay deposits.

In contrast to the Early and Middle Neolithic samples, the data from the Late Neolithic (NIib) show the use of all three clay sources for the production of pottery. This finding was unexpected. It was hypothesized that changes in land use, organization of ceramic production, and increased social differentiation should be visible in the raw materials used to produce pottery (see above). Elemental analysis has shown, however, that the same raw materials were used as in earlier periods. This suggests that the typological changes, including more open forms and predominance of undecorated wares, have little to do with raw material selection. Rather, this striking shift must be understood primarily in cultural terms. Given that similar clay sources were used in both the Early and Middle Neolithic, Late Neolithic potters were making decisions on pottery styles without any clear constraints based on the raw materials available. Therefore, we must look to other factors of change during the Neolithic to help explain the technological and stylistic shifts visible in pottery. Future research can focus more explicitly on shifts in the symbolic nature of pottery, its relationship to other types of artifacts, and its primary role in daily technologies such as cooking, storage, and serving.

With these kinds of preliminary data, more detailed studies of provenance and resource use through time can be conducted. It is our hope that samples will be available to extend this study in the future. Results of the analysis of prehistoric daub and modern clay sample is a taste of the potential for INAA in this area. A systematic collection of modern clays in the region would benefit the interpretations of prehistoric patterns and provide a better understanding of chemical variability in clays

across this region. Furthermore, building materials at Neolithic sites generally have received little attention from archaeologists. If the INAA results discussed above are any indication, much can be gleaned about Neolithic resource use and general clay technologies from this class of artifact. Pottery production has long been studied independently from house construction material; however, an approach that focuses on the modification of clay regardless of end product is an interesting avenue for more insights on clay technologies. In any event, INAA has proven a useful method in distinguishing discrete elemental compositions of pottery in the Alcoi Basin, and future research will illuminate this aspect of ceramic production further.

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