



Elemental variation in prehistoric *Unionoida* shell: Implications for ceramic provenance



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ARTICLE INFO

Available online 11 September 2014

Keywords:

Neutron activation analysis
Shell temper
Calcium correction
Ceramic provenance

ABSTRACT

As part of ongoing efforts to refine ceramic-provenance methodology, we examine the universality of freshwater mussel shell chemistry as reported in an earlier study. We find that samples of prehistoric shell from several locations in eastern North America are chemically distinct from the modern sample of shell from Missouri that was previously used to develop a formula for adjusting elemental abundances in shell-tempered pottery. The observed elemental differences may be attributable to the use of prehistoric versus modern shell, or they may relate to watershed-specific geological chemistry. Regardless of the source of these differences, we suggest that future provenance studies involving shell-tempered wares would be well served by complementary analyses of locally derived prehistoric shells to better the effects of shell tempering on ceramic chemistry.

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1. Introduction

When present in a ceramic matrix, shell (and other Ca-rich materials such as bone, limestone, and calcite) creates a dilution effect that reduces the detected abundances of all other elements in the specimen (Cogswell et al., 1998; Steponaitis et al., 1996). The effects of Ca-rich tempers are analogous to those introduced by Si-rich tempers such as quartz (Perlman and Asaro, 1971; Sterba et al., 2009) except that unlike Si, Ca can be detected and quantified using neutron activation (NAA). The use of shell as a tempering agent significantly increases the amount of Ca in pottery, and it causes an apparent decrease in other elements present in the pottery. This is because elemental abundances determined by NAA are typically reported in a fixed-sum matrix (e.g., parts per million, weight percent). Thus, most elements are “diluted” by the large proportion of Ca present in the specimen. If a particular element is present in sufficient abundance within the shell, but absent or exceedingly low in the clay to which shell is added, this element will be enhanced in the resultant pottery. Thus, shell tempering has the potential to both dilute and enrich the bulk chemistry of pottery.

Cogswell et al. (1998) reported the results of a study directed at evaluating methods for correcting this well-documented elemental dilution/enrichment effect of shell as a tempering agent in prehistoric ceramics. The goals of their research were to (1) assess whether elemental differences significant to ceramic-provenance research exist among shells of freshwater mussel genera, (2) evaluate potential elemental contributions of freshwater mussel-shell temper to prehistoric

pottery, and (3) compare three proposed methods for eliminating or minimizing the dilution/enrichment effects of shell temper so as to more accurately estimate the chemical makeup of the clay. Here, we expand upon this research through an evaluation of whether the mean values reported by Cogswell et al. (1998) are universally precise estimates for freshwater mussel shells used as ceramic temper across a broad swath of eastern North America. The research reported here is part of larger efforts to refine protocol of the analysis of archaeological ceramics at the University of Missouri Research Reactor (Boulanger et al., 2013; Stoner et al., 2013).

2. Background

Five factors are known to influence the elemental composition of mussel shell: local geology, water temperature, water salinity, species of organism, and the calcite:aragonite ratio of the shell itself (Turekian and Armstrong, 1960; Crisp and Richardson, 1975). As discussed by Peacock et al. (2012), freshwater mussels are filter feeders that tend toward chemical equilibrium within their immediate environment. In the absence of foreign input from contaminants and pollution, stream and lake chemistry is largely dependent on local geology and climate. Particulates within a river system are most likely to derive from the immediate vicinity, and are thus determined by geology within a watershed. Miller (1980) observed that modern marine shell specimens exhibited elemental compositions that were significantly different than those of prehistoric shell specimens from the same localities. She attributed the differences to changes in land-use practices and the introduction of modern pollutants into the water system. Thus, within the context of archaeological and paleobiological studies, time may be considered a sixth factor influencing shell chemistry.

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Given that water chemistry is largely a result of local environment, and that mussels exist in equilibrium within the water, it stands to reason that the provenance postulate (Weigand et al., 1977) could be applied to shell chemistry (Claassen and Sigmann, 1993; Eerkens et al., 2007; Eerkens et al., 2009; Eerkens et al., 2010; Miller, 1980; Peacock et al., 2010). That is, we expect that freshwater mussel shells from different locations may be chemically distinct insofar as local geology varies chemically (Peacock et al., 2010; Peacock et al., 2012). If local habitat and geology does indeed strongly influence shell chemistry—and present evidence suggests that it does—this may have implications for the bulk analysis of shell-tempered pottery and the use of mathematical adjustments to compositions of shell-tempered ceramic specimens. In short, time-specific, region- or watershed-specific formulae may be necessary rather than a universal formula.

Cogswell et al. (1998) evaluated three methods for minimizing elemental dilution/enrichment effects of shell when used as a tempering material in ceramic production. Specifically, they analyzed a sample of modern freshwater mussel shells obtained from the Gasconade River, in Mt. Sterling, Missouri. Three genera (*Cumberlandia*, *Lampsilis*, and *Fusconaia*) were present in the sample, and the shells were analyzed both before and after burning in a laboratory furnace. Shells were burned in the furnace to simulate the effects of firing a ceramic vessel or the burning of shell by the potter to facilitate crushing for use as temper. No significant chemical differences resulting from burning were observed among any of the genera, leading the authors to conclude that burning of shell leads only to minor changes in the elemental composition (see also Collins, 2012). The authors also found that elemental compositions of shell did not vary significantly across taxa from the same river (Cogswell et al., 1998:66–71). Significantly, the authors also reported that concentrations of Sr, Na, and Mn in their shell sample were greater than those in the New Ohio Redart Clay (NOR, a commercially available clay used as a check-standard at MURR) and in several specimens of clays from various locations in the Mississippi River Valley. Thus, a novel observation of their study was that the concentrations of these elements in a ceramic body could potentially be influenced by the presence of shell tempering.

In order to correct for elemental dilution/enhancement caused by shell tempering, Cogswell and colleagues advocated the use of a mathematical correction formula first suggested by Steponaitis et al. (1996):

$$e' = \frac{10^6 \times e}{10^6 - 2.5c} \quad (1)$$

where e' is the adjusted concentration of a given element in ppm, e is the measured concentration of an element, and c is the measured concentration of Ca. The value 2.5 is a gravimetric factor compensating for the difference in the mass of Ca in ppm to the mass of calcium carbonate (CaCO_3) in shell. This formula normalizes abundances of all elements to the measured amount of Ca, and thus Ca is removed from the compositional data after applying the correction formula. Because Sr freely substitutes for Ca in shell, it too is removed from the compositional database.

As noted above, Cogswell and colleagues also found that the abundances of Na and Mn in their shell sample were greater than those in samples of commercial art clay and unrefined clays of the sort that may have been used prehistorically. In these instances, shell tempering would lead to an enrichment of Na and Mn, but the analyst is blind to which component of mixture (i.e., the pot sherd) is responsible for the elevated Na and Mn levels: the clay, the temper, or a combination of both. To correct for the added contribution of these two elements to a ceramic matrix, their concentrations are modified as follows:

$$e' = \frac{e - \frac{2.5c}{10^6} \times e_{\mu}}{1 - \frac{2.5c}{10^6}} \quad (2)$$

This is similar to the Ca-normalization formula presented above, but with the inclusion of the factor e_{μ} which is defined as the average

concentration of the particular element in shell. In practice, the shell-correction as historically implemented at MURR assumes concentrations of 1488 ppm for Na and 578 ppm for Mn (mean [average] values determined in the Cogswell et al. study), and adjusts the concentrations of those elements in the ceramic matrix accordingly.

As suggested above, the uncritical use of the chemical data reported by Cogswell and colleagues may present some problems in understanding the chemistry of shell-tempered wares. There is no a priori reason to assume that the chemical composition of shell from central Missouri is identical to, say, that of shell from east Texas, because of differences in geology and water chemistry. Further, there is no a priori reason to assume that the chemistry of *modern* shells from central Missouri is identical to *prehistoric* shells from central Missouri. Cogswell and colleagues had no intention of evaluating these aspects of shell chemistry; the goals of their study were to evaluate how best to minimize the dilution effects caused by Ca in a ceramic matrix. Our research is a logical extension of their earlier study. We note that our approach is purposefully pottery-centric, and aims to address only the potential confounding effects of shell temper in pottery; however, we point out that the implications of shell chemistry being influenced by land-use, local geology, and watershed chemistry are far ranging, including provenance studies of shell itself, conservation biology, and landscape ecology.

3. Methods

We are primarily interested in shell-tempered ceramics produced prior to the arrival of Europeans in the Western Hemisphere. Miller (1980) previously demonstrated that modern shell is an inappropriate analog for prehistoric shell because of the introduction of industrial pollution, increased sediment load from deforestation, changes in land use, and agricultural chemical use within a watershed. Therefore, all of the freshwater mussel shells analyzed in this study come from unequivocally prehistoric contexts (Table 1). We also adopt a broad-scale approach to evaluating variation in shell chemistry, and our sample reflects this. In total, 111 specimens of shell from several freshwater mussel genera were obtained from six archaeological sites from the Midwest, the Middle Atlantic, and New England (Table 2, Fig. 1). Because of differences in habitat, we were unable to ensure that all mussels from all sites were of the same species. All specimens were identified to at least the genus level. We compare our archaeological sample with the previously reported data of Cogswell et al. (1998).

All specimens were rinsed under water and scrubbed with a plastic-bristle brush to remove adhering soil, sediment, and organic materials. Once cleaned, specimens were rinsed in deionized water and placed under a heat lamp to dry. Cleaned specimens were fired in a laboratory furnace to a temperature exceeding 700 °C for 1 h. Once burned, each specimen was ground to a powder in an agate mortar and pestle. The pulverized specimens were placed into a glass vial and allowed to dry in a warming oven for at least 24 h.

Specimens were analyzed by neutron activation at the University of Missouri Research Reactor (MURR) following standardized protocols (Glascock, 1992; Glascock and Neff, 2003).

Table 1
Archaeological sites from which samples of mussel shell were obtained.

Site number	Site name	Location	Age
28SX17	Beisler	Walpack, NJ	Late Woodland
12HR11	Breeden	Mauckport, IN	Middle–Late Archaic
15OH1	Chiggerville Shell Mnd	Ohio Co., KY	Late Archaic
15BT6	DeWeese Shell Mnd	Highview, KY	Middle–Late Archaic
27CH85	Fort Hill	Hinsdale, NH	Contact
28WA392	Indian Hollow	Johnsonburg, NJ	Late Woodland
12HR12	Overflow Pond	Mauckport, IN	Late Archaic

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