Journal of Archaeological Science 40 (2013) 874–882

Contents lists available at SciVerse ScienceDirect

Journal of Archaeological Science

journal homepage: http://www.elsevier.com/locate/jas

Tracing the source of Late Neolithic *Spondylus* shell ornaments by stable isotope geochemistry and cathodoluminescence microscopy

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ARTICLE INFO

Article history:
Received 16 November 2011
Received in revised form
3 September 2012
Accepted 20 September 2012

Keywords:
Spondylus
Late Neolithic ornaments
Stable isotope
Cathodoluminescence
Provenance analysis

ABSTRACT

Determination of the source of *Spondylus* objects is essential for the interpretation of Late Neolithic exchange systems and the social role of shell ornaments. We performed stable isotope analysis combined with cathodoluminescence microscopy study on ornaments (beads, bracelets) made of *Spondylus* shells excavated at the Aszód-Papi földek archaeological site in Hungary, to define their origin. For comparison *Spondylus* finds from Neolithic sites of Greece, modern *Spondylus* shells from the Aegean and the Adriatic, as well as fossil *Spondylus* and *Ostrea* shells from the Carpathian Basin were also examined. Oxygen isotope composition of *Spondylus* finds from Aszód ranges between −1.9 and 2.1‰ and overlaps with the oxygen isotope range of shell objects from other Neolithic sites. Modern *Spondylus* shells from the Aegean and the Adriatic show overlapping $\delta^{18}O$ values with one another and with the Neolithic objects; while recent shells of the Black Sea clearly are separate isotopically from the Mediterranean ones and most of archaeological artefacts. *Spondylus* shells from the Aszód site have Mediterranean origin; their source can be the Aegean or the Adriatic. Based on a former strontium isotope study the use of fossil *Spondylus* shells is excluded as raw material used for ornaments, however, in recent years the use of fossil shells was reintroduced. The shell ornaments from Aszód-Papi földek and the fossil oyster shells collected from the Carpathian Basin exhibit some overlapping oxygen isotope values; however, cathodoluminescence microscopy indicates that the *Spondylus* objects retained their original aragonite material. Diagenetic calcite, which occurs typically in the fossil shells, was not detected in the ornaments suggesting that the studied objects were made of recent shells. Calcitic parts observed in some *Spondylus* objects are not related to fossilisation.

1. Introduction

Spondylus gaederopus, or thorny oyster, is a bivalve mollusc preferring warm-water seas. *S. gaederopus* species currently live in the Mediterranean – mainly in its eastern part, the Aegean and the Adriatic – cementing themselves to rocks or coral in a depth of 6–30 m. The width of a mature shell varies between 6 and 12 cm, recent shells are usually smaller than those known from prehistory. There are considerable differences in shape and size of the lower and the upper valves. The lower valve is oval or round, definitively thick, while the upper valve is round, thinner, thorny and purple on its outer surface. These characteristics determined the shape and size of the ornaments made from them (Tsuneki, 1987; Miller, 2003; Chapman and Gaydarska, 2007; Pappa and Veropoulidou, 2011; Siklósi and Csengeri, 2011).

One of relevant issues regarding Neolithic *Spondylus* ornaments is the origin, i.e. provenance of the shell raw material used for preparing artefacts. Based on stable isotope geochemical analyses

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http://dx.doi.org/10.1016/j.jas.2012.09.022

Shackleton and Renfrew (1970) proposed that the likely source of the Neolithic *Spondylus* shells was the Mediterranean, specifically the Aegean. *Spondylus* in recent years does not live in the Black Sea, however, Todorova supposed on the basis of the great amount of Neolithic and Copper Age *Spondylus* finds from the Black Sea coast of Bulgaria that *Spondylus* could live not only in the Mediterranean, but also in the Black Sea in this period. Todorova (2000, 2002) proposed that during the "climate optimum" the Black Sea could have been a habitat for *Spondylus*, therefore she did not exclude the Black Sea as a possible provenance.

Researchers unanimously excluded the possibility of using fossil *Spondylus* shells based on the results of strontium isotope analysis performed by Shackleton and Elderfield (1990). The overwhelming majority of *Spondylus* shell raw material used for preparing ornaments was supposedly to be contemporary to the Neolithic causing several researchers to reconstruct a long-distance exchange system from the Aegean through the Balkan and Central Europe to the Paris Basin (Séfériadès, 1995a, 1995b, 2000; Müller, 1997; Kalicz and Szénászky, 2001; Dimitrijević and Tripković, 2003; Siklósi, 2004). In the last few years the idea of using fossil shells for making ornaments during the Neolithic has risen again (Dimitrijević and Tripković, 2006; Sümegi, 2009). There are fossil *Spondylus* sites for example in the territory of Eastern Bulgaria (Chapman and Gaydarska, 2007). This fact is thought-provoking as this is the same area where the usage of *Spondylus* ornaments has the longest duration (see later) (Müller, 1997; Todorova, 2000, 2002).

Despite seemingly successful application of the isotope methods, studies of Shackleton and Renfrew (1970) and Shackleton and Elderfield (1990) are not "without potential problems" (Douka, 2011). The amount of analysed samples is relatively small, e.g. nine *Spondylus* artefacts from four archaeological sites were analysed for stable isotope composition, while empirical dating by comparison of $^{87}Sr/^{86}Sr$ ratio of shells to the Sr isotope evolution curve of seawater was performed only on three Neolithic *Spondylus* artefacts and one fossil *Spondylus crassicosta*. In addition, these studies did not check in detail the signs of postdepositional processes or diagenetic alteration (recrystallization) of artefacts, which can modify the shell mineralogy as well as the original isotope values (Douka, 2011). After the above-mentioned two publications application of isotope geochemical methods was not widely used in the research of Neolithic *Spondylus* ornaments. Later only Todorova (2002) published stable isotope data of *Spondylus* artefacts (17 objects) and interpreted them as indicating two biotopes.

During stable isotope analysis ratios of oxygen and carbon isotopes ($^{18}O/^{16}O$, $^{13}C/^{12}C$) of carbonate, the mineral constituent of shell, are measured. It is generally agreed that carbonate of bivalve shells is precipitated in equilibrium with ambient seawater (Epstein et al., 1953), this approach is also proven for *S. gaederopus* (Maier and Titschack, 2010). The oxygen isotope composition of shell carbonate depends on the oxygen isotope composition of seawater, which covaries linearly with salinity, and the temperature of carbonate precipitation (Hoefs, 2009). Different temperature and/or oxygen isotope composition of seawater in different habitats provides opportunity for distinguishing shells from different sources based on the oxygen isotope composition. During their early study Shackleton and Renfrew (1970) used this assumption and expected that Black Sea is isotopically more negative than the Mediterranean due to its water budget. Isotope analyses of recent *Nassa reticulata* and *Chamelia gallina* species from the Black Sea seemed to confirm their hypothesis and isotope values of recent shells did not overlap with the values of *Spondylus* artefacts from Bulgarian, Greek and Serbian archaeological sites, therefore artefacts were supposed to be originated from the Aegean rather than the Black Sea.

The assumption of Shackleton and Renfrew (1970) regarding the difference in stable isotope composition of the Black Sea and the Mediterranean seems to be valid. Surface waters of the eastern Mediterranean exhibit $\delta^{18}O$ values between 1.30 and 1.66‰ (Pierre, 1999) or may have even more positive values up to 2.2‰ (Gat et al., 1996), while the surface waters of present-day Black Sea have more negative $\delta^{18}O$ values (Swart, 1991), −2.8‰ in average with standard deviation of 0.34‰ (Ranke et al., 1999). Therefore basic prerequisite that the basins are different in isotope composition can be assumed and successful provenance study of shells can be expected provided that the calcium carbonate of the shells is deposited under isotopic equilibrium conditions or very close to equilibrium with environmental waters.

While the local, i.e. Aegean source can unequivocally be assumed for the *Spondylus* ornaments in the find material of Greek archaeological sites, the Aegean origin is not evident in case of artefacts found in the continental European sites. The possibility of Adriatic origin has also to be taken into account.

Our study intends to extend the number of archaeological samples analysed by using the stable isotope method combined with non-destructive cathodoluminescence microscopy, the latter is used to check whether shell artefacts preserved their original mineralogy. We performed analysis on *Spondylus* ornaments found at the Late Neolithic Aszód-Papi földek site (Hungary) and examined *Spondylus* finds from Neolithic sites in Greece, which are roughly contemporaneous with the Aszód settlement. The aim of our research is to determine the provenance of the raw material of *Spondylus* ornaments, i.e. whether the shells were recent ones originating from the Mediterranean or Black Sea or were fossil specimens. Recent *Spondylus* shells from the Adriatic and the Aegean were analysed as a comparative material. There are several geological sites in the Carpathian Basin where fossil *Ostrea* shells can be collected in great amount and fossil *Spondylus* can be found within cca. 50 km vicinity of Aszód (Csepreghyné Meznerics, 1954), therefore we cannot reject the possibility of using fossil shells without further research. Miocene *Spondylus* fossils and fossils of a related species, *Ostrea* from the vicinity of the site and from a distant locality in Romania were studied both for mineralogy and geochemistry in order to identify possible fossil shells in the find material.

2. Archaeological context

Spondylus ornaments as prestige goods played an important role in the expression of social inequality and prestige manipulation all through the Neolithic of the Carpathian Basin, but their usage changed considerably during this period (Kalicz and Szénászky, 2001; Siklósi, 2004; Siklósi and Csengeri, 2011).

The first sporadic *Spondylus* ornaments – exclusively bracelets – appeared during the Early Neolithic (cca. 6000–5500 cal BC) in the Carpathian Basin (Kalicz and Szénászky, 2001; Siklósi, 2004).

From the beginning of the Middle Neolithic (cca. 5500–5000 cal BC) the usage of *Spondylus* ornaments suddenly dispersed in the Carpathian Basin. During this period *Spondylus* ornaments reached even the area of present-day Poland, Germany and France (Willms, 1985; Müller, 1997; Séfériadès, 1995a, 1995b; 2000).

In the Middle Neolithic mainly large, heavy ornaments were used with high raw material requirement and were prepared individually. The characteristic Middle Neolithic types were large pendants made of a whole shell valve, *V-Klappen*, massive bracelets and large beads found mainly in graves and hoards, very rarely in settlement features (Kalicz and Szénászky, 2001). In this period male, female as well as children's graves contained *Spondylus* ornaments, although there are local differences (Siklósi, 2004).