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Moving metals III: Possible origins for copper in Bronze Age Denmark based on lead isotopes and geochemistry

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ABSTRACT

This article presents the results of a comprehensive provenance study based on a combined geochemical-isotopic and archaeological approach, comprising 98 analyses of 97 copper-alloy objects from the Danish Bronze Age. When it comes to the question of the origin of the metal, our interpretations diverge somewhat from earlier established theories about the origin of copper imported to Denmark, which mainly pointed to Central and Eastern Europe. Clear geochronological patterns in the Danish dataset are interpreted as being due to shifts in ore sources; reflecting varying areas of origin as well as the utilization of varying ore types. This again relates to shifting trade networks/suppliers and shifting technological trends. Plausible sources for Danish copper-alloys identified in the current study are ore regions in the *British Isles, Alpine ore districts* in Italy and Austria, as well as ore regions in the *western part of the Mediterranean* and to some degree *the Slovak Carpathians*. The comparison includes hundreds of recently published lead isotope data for ores in Slovakia, the Iberian Peninsula and the Italian and Austrian Alps.

1. Introduction

This article presents the results of a provenance study of 97 Cu-based objects from Denmark, aiming, among other things, at understanding Scandinavia's role in the European exchange networks (Kristiansen, 2013). The study combines trace element analysis with lead isotope analysis. A combined approach is currently the most useful tool when trying to identify copper ore sources. While lead isotope ratios may indicate the age and origin of the ore, element composition characterizes which *type* of ore the copper derived from. Apart from limited lead isotope studies of singular objects (Klassen and Stürup, 2001; Kresten, 2005; Pernicka, 2010; Schwab et al., 2010), the methodology has only recently been applied more widely in analyses of Scandinavian prehistoric metalwork. While the provenance ascriptions of the current study largely overlap with recent studies of Swedish bronzes conducted by Ling et al. (2013, 2014) and Vandkilde (2017), differences between these two regions of Scandinavia may also be noted.

Given that the total number of Bronze Age metal finds in Denmark amounts to an unspecified 5-digit number, it is clear that the current study can only represent a point of departure for more targeted chronological and regional studies. Despite this, the results can be used to go more in-depth on certain aspects of Danish metalwork and metal consumption, since included in the study are some of the finest bronze finds from Denmark. Moreover, the broad chronological scope represents a new approach compared to previous provenance studies carried out in Scandinavia, focusing first and foremost on early metal-using periods (Cullberg, 1968; Junghans et al., 1968, 1974; Klassen, 2000; Krause, 2003; Vandkilde, 1996, 1998; for exceptions, see Liversage and Northover, 1998; Liversage, 2000).

2. Background for the study

Denmark is one of the richest regions in Bronze Age Europe in terms of retrieved metalwork. This remains a paradox, since apart from Greenland, present-day Denmark is completely lacking natural copper-

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bearing ore deposits and would have had to depend totally on metal imported from other areas throughout the Bronze Age. The origin of the metals used to produce the distinguished Nordic metalwork has been an unresolved problem. In the formative years of scientific archaeology, it was presupposed that all Bronze Age copper had been imported to Scandinavia from Continental Europe (e.g. Montelius, 1885), and during the following century provenance ascriptions were biased toward Central and Eastern Europe (e.g. Kristiansen, 1998; Thrane, 1975). Interpretations have been circumstantial, resting in the final analysis on typology, artefact distribution patterns and hypothetical matches with Bronze Age mines. A theory of indigenous mining on the Scandinavian Peninsula (e.g. Melheim, 2009, 2015; Prescott, 2006), suggested that populations in Denmark might have depended on copper from ore sources in Norway or Sweden. However, in the recently published studies of c. 70 Swedish metal objects and casting debris (Ling et al., 2013, 2014), all analyses (with one exception – a slag) were interpreted as being of non-local origin. [1]

3. Lead isotopes as provenance indicators

The applied methodological approach broadly follows Ling et al. (2014). The fundamental aspects of lead isotope (LI) tracing are well-known and we shall only briefly address some source-critical factors of relevance for this study (for a broader discussion of the method's applicability in archaeology, see e.g. Albarède et al., 2012; Baron et al., 2013; Gale, 2001; Hauptmann, 2009; Northover et al., 2001; Rehren and Pernicka, 2008; Stos-Gale and Gale, 2009). The use of LI for provenancing is based on the assumption that the lead component in bronze (usually < 1%), originates from the copper ore used in production. Occasionally, lead was added during the metallurgical processes, and when added, generally in concentrations well above 1%. Tin, the other major component of bronze, is usually not associated with appreciable amounts of lead in nature, and the number of known ore sources of Sn is limited in relation to the number of Pb-bearing copper ores known to have been exploited during the Bronze Age in Europe.

Several factors may complicate the interpretation of the source of lead, and the foremost are the wide ranges of isotopic compositions often noted within a certain ore district and the isotopic overlap between data from different districts. It must also be noted that if ores formed at the same time but acquired lead from source rocks having contrasting isotopic characteristics, then this may result in misleading ore lead model ages. Further complexities during the stage of interpretation of provenance areas may arise if manual mixing of ores from different districts took place, or if previously cast objects were re-melted. Linear patterns do occasionally show up when data are plotted in Pb-Pb diagrams. Such patterns could be explained in two different ways; either as a result of physical mixing of isotopically different source material at the time of smelting or casting, or, *in situ* decay of uranium and thorium which took place between the time of ore formation and mining (producing additional amounts of ^{206}Pb, ^{207}Pb and ^{208}Pb).

4. Material and methods

The artefacts sampled for the current data-set cover the entire Nordic Bronze Age and most parts of Denmark, and also include two Late Neolithic axes (Figs. 1–2, for details see Table 1, Supplementary Table 1). The periodization builds on well-established typo-chronologies (Montelius, 1885), and is anchored in absolute chronology (Olsen et al., 2011; Vandkilde et al., 1996) (Table 2). The original assemblage of 77 samples comprised high-quality objects from hoard finds like Late Neolithic (2350-1700 BCE) Store Heddinge, Early Bronze Age (1700-1100 BCE) Dystrupgård, Torsted, Bagterp, Vognserup Enge and Røjlemose, and Late Bronze Age (1100-500 BCE) Ejsdrup, Kostræde, Fløe, Lysemosegård and Antvortskov. Also included were iconic finds like the period II octagonal-hilted sword from Ramløse. The remaining 21 analyses come from a targeted regional study from Thisted and Randers in northern Jutland, focusing on swords and bracelets from Early Bronze Age barrow burials.

The 77 samples reused for LI analysis were kindly provided by Peter Northover, responsible for the analytical undertakings and the preparation of the samples for the earlier electron microprobe analysis (EPMA). Samples of millimetre-size mounted several together in resin were ground and polished. The original analyses were made using a CAMEBAX instrument at the Department of Materials, Oxford University. Wave length dispersive (WDS) analyses were made at three different points measuring 50 by 50 μm at an accelerating voltage of 25 kV and an absorbed current of 30 nA. Pure element standards were used and data were ZAF corrected.

A few of the originally mounted samples had not been analysed in the first study. These samples were included in the present study following the same analytical procedure, along with the 21 new samples from northern Jutland. Subsequently, an optical microscope with polarised reflected light was used in order to define the structure and texture and prepare for succeeding EPMA using the JEOL JXA-8530 F at the Centre for Experimental Mineralogy, Petrology and Geochemistry at Uppsala University. WDS analysis was made as point analyses in separate phases as well as in area scans (50 by 50 μm). The scans were distributed along traverses to attain the bulk chemical composition of the alloy. Due to the common heterogeneity of copper alloys, exhibiting dendritic textures, multiple area scans were made and mean values calculated. Operating conditions during runs involved an acceleration voltage of 20 kV and an electron beam current of 20 nA. The obtained analytical data were related to standards (oxides, sulphides, metals) and ZAF corrected. Furthermore, the detection limits for most elements (see Supplementary Table 2) were improved with the currently used instrument; in particular for As, avoiding the interference with Pb (see also Northover, 1996b).

Since two types of analytical instruments (CAMEBAX and JEOL JXA-8530 F) were involved in producing the element composition data-set, some samples for which results were initially presented by Northover and co-workers (Northover, 1996a; Liversage and Northover, 1998; Liversage, 2000) were re-analysed by the current team in order to be able to compare the results. Generally, the agreement between the old and the new analyses was good (Supplementary Table 2), but in a few cases discrepancies were larger. This is potentially due to heterogeneous material in some samples. The new analyses form the basis for interpretations (Supplementary Table 1).

Following the microprobe analyses, suitable bronze pieces were digested in hot 6 M HNO_3 in order to prepare them for LI analysis. Prior to dissolution they were leached for a few minutes in HNO_3 at room temperature in order to remove possible surface contamination and some of the alteration products such as malachite. After dissolution the lead was extracted by means of anion exchange column chemistry. The isotope measurements were performed at the Department of Geosciences at the Swedish Museum of Natural History in Stockholm with a Multi-Collector Inductively Coupled Plasma Mass Spectrometer (MC-ICP-MS) of the type Micromass Isoprobe. The mass bias correction was made by adding thallium to the sample. Details are described in e.g. de Ignacio et al. (2006). The total error (external reproducibility) for reported Pb isotope compositions of unknowns is estimated to be ± 0.10% or lower.

[1] While copper ores on the European Continent were dominantly formed in the 400-100 million years (Ma) interval, many of the Cu ores in Sweden have ages around 1900 Ma, belonging to the Precambrian basement. A large age contrast means that lead isotope ratios are very different from each other, and it is easy to discriminate between e.g. hypothetical origins in Europe and Bergslagen in Sweden. Other copper-bearing ore deposits in Scandinavia with younger ages exist; e.g. c. 1200 Ma old deposits in Dalsland in southwestern Sweden (Alm et al., 1997), c. 500-400 Ma old deposits in the Caledonian mountain range that straddles the border between Norway and Sweden (Grenne et al., 1999), and c. 300 Ma old deposits in the Oslo field (Grenne et al., 1999). No clear matches with any of these ores were, however, obtained in the Swedish study.