Tin Provenance and Raw Material Supply – Considerations about the Spread of Bronze Metallurgy in Europe

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Keywords

Tin isotopy, Bronze Age, Europe, Mesopotamia, Cornwall, Saxon-Bohemian Ore Mountains

Abstract

The paper focuses on isotopic data of bronzes from the 3rd and 2nd millennium BC. The sample sets comprise bronzes from hoards, graves, and settlements from Central and Southeastern Europe as well as the Aegean and Mesopotamia. The analytical determination of tin isotopic compositions and a possible use of tin from different ore sources between the Carpathian Basin, the Aegeo-Balkan-Complex and tin bearing regions in Central and Western Europe will be discussed. Since the 2nd millennium bronzes show in general a different isotopic composition than those of the 3rd millennium, the presented analyses indicate a possible reorientation of exchange routes in Europe during the 2nd millennium BC. This is supported by the composition of a few Aegean samples from the turn of the millennia, which have heavier tin isotopic compositions than all other sample sets. This suggests that different tin sources might have been used to manufacture these bronzes.

Introduction

Recent research makes it more and more likely that tin sources in Western and Central Europe supplied large parts of continental Europe with tin (Nessel, Brügmann and Pernicka, 2015). Unfortunately, the provenance of this important raw material cannot be determined through archaeological research alone, which would be essential in particular for the understanding of the manufacture of the earliest bronzes before and during the Early Bronze Age in Europe. This unsatisfying situation led to the establishment of the multidisciplinary project "BRONZEAGETIN-Tin Isotopes and Sources of Bronze Age Tin in the Old World" funded by the European research Council (ERC), whose general aim is to investigate the isotopic composition of tin ores in the Old World and to determine the exploitation of specific tin deposits or provinces. The chemical and isotopic composition of tin ores and prehistoric bronzes from Europe and the Near East is investigated using XRF, NAA (Hauptmann and Pernicka, 2004) and MC-ICP-MS (Brügmann, Berger and Pernicka, 2017).

This paper provides an overview of some results of the analytical determination of tin isotopic compositions, and discusses a possible spreading of tin-bronze technology can be identified between the Carpathian Basin, the Aegeo-Balkan-Complex and tin-bearing regions in Central and Western Europe.

The analytical focus was on cassiterite. It is a hard, dense, weathering-resistant mineral, which is deposited during the erosion of granite and concentrated in fluvial placer deposits. These placers are considered as major tin sources in prehistory, because cassiterite could be obtained with comparatively little effort and high purity (Nessel, Brügmann and Pernicka, 2015). The cassiterite samples analysed in this study derive from deposits in southern England (Cornwall and Devon), Germany and the Czech Republic (the Fichtelgebirge, the Saxon-Bohemian Ore Mountains, the Vogtland, the Kaiserwald (Haustein, Gillis and Pernicka, 2010; Haustein, 2014; Marahrens, 2016) and Western Asia. The ore samples from European tin provinces show a large range of isotopic variation, with the $\delta^{124/120}$ Sn-ratios ranging from -0.28 to 0.85 ‰. The average $\delta^{124/120}$ Sn-values for Cornwall and Devon $(0.07 \pm 0.57\%)$ and the Saxon-Bohemian Ore Mountains (0.12 ± 0.38 ‰) indicate that the isotopic composition of cassiterite from the latter is on average lighter than that of southern England. This is reflected in a higher proportion of low $\delta^{124/120} \text{Sn-values}$ (<0.05‰) in samples from the Saxon-Bohemian Ore Mountains and a higher proportion of heavy isotope compositions in cassiterites from

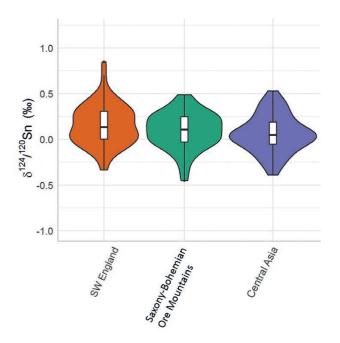


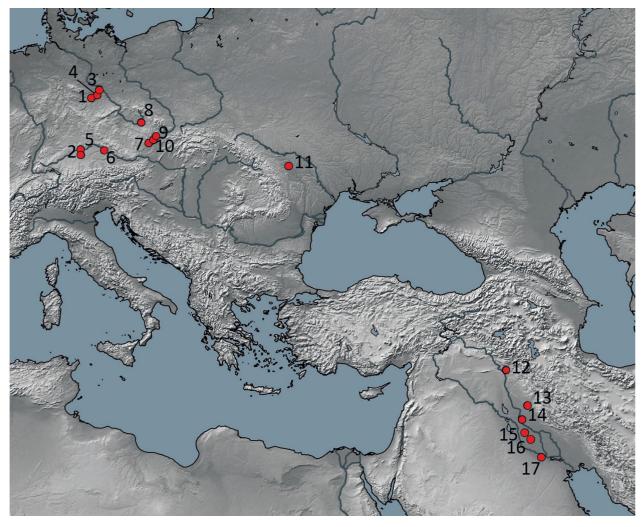
Figure 1. The variation of the $\delta^{124/120} Sn$ ratio in cassiterite samples of southern England, the Saxon-Bohemian Ore Mountains and Central Asia (created by Carolin Frank).

Cornwall (>0.55 ‰) (Brügmann, et al., 2017). The violin-plot in Figure 1 shows the variation of the $\delta^{124/120}$ Sn ratio in cassiterite samples from southwest England, the Saxon-Bohemian Ore Mountains and Central Asia. The isotopic ratios of Asian cassiterites substantially overlap with those of central and western European cassiterite. Although this tendency defines measurable differences between the two major European tin deposits and those of Central Asia, it is currently not possible to distinguish between the three large provinces by an analysis of tin isotope ratios because of the significant data overlap.

European bronzes of the 3rd millennium BC

The beginning of the Bronze Age in Southeastern Europe is currently dated at around 3000 BC (Boroffka, 2013; Băjenaru, 2014), long before tin bronze is used in the broader region. To find indications for the rise of tinbronze metallurgy in Europe, it seems plausible to com-

Figure 2. The sites mentioned in the text: 1 Allenstedt, 2 Augsburg-Haunstetten, 3 Gnetsch, 4 Salzmünde, 5 Biberach-Markt, 6 Osterhofen-Altenmarkt, 7 Smolín, 8 Bylany, 9 Ledce, 10 Bohdalice, 11 Glăvănești Veche, 12 Tepe Gawra, 13 Tell-es Suleimeh, 14 Tell Asmar, 15 Kish, 16 Nippur, 17 Ur.



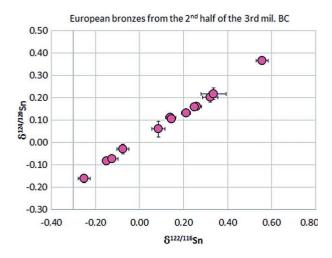


Figure 3. The variation of the $\delta^{124/120} Sn$ to $\delta^{122/116} Sn$ ratio in pre-Bronze Age artefacts of Central and Southeastern Europe belonging to the Corded Ware, Bell Beaker and Ochre Grave cultures.

pare in a first step the isotopic composition of the first known European bronzes from the second half of the 3rd millennium BC with contemporary bronzes from the Near East, in order to assess possible commonly used tin resources in one of the regions.

The bronzes in the European sample set mostly date to the second half of the 3rd millennium BC and belong to various contexts of the Corded Ware-, Bell Beakerand Ochre Grave cultures in the Czech Republic: Smolín (Novotný, 1958, pp.310-311), Bylany (Hájek, 1968, p.11), Ledce (Hájek, 1957, pp.392), Bohdalice (Kaloušek, 1956, pp.93), Germany: Biberach-Markt (Mahnkopf, 2007/2008), Augsburg-Haunstetten (Massy, et al., 2017, pp.248-249), Osterhofen-Altenmarkt (Schmotz, 1990), Allstedt (Bruchhaus and Holtfreter, 1984, pp.215), Gnetsch (Müller, 2001, p.412), Salzmünde (Schlette, 1948, p.36), and Romania: Glăvănești Veche (Comşa, 1987, pp.372-373). Only the bronze object from Gnetsch is dated to the last quarter of the 4th millennium BC (Figure 2). Most bronzes are comparably low in tin, which varies between 1.55 and 6.8 wt. %. These low tin bronzes are almost exclusively found in Bell Beaker and Corded Ware burials. However, two artefacts can be associated with burial of the Ochre Grave culture. Generally, diverse groups of objects with different functions, ranging from weapons and tools to jewellery, consist of tin bronze. Their $\delta^{124/120}$ Sn to $\delta^{122/116}$ Sn ratios vary between -0.28 and 0.55 ‰, which can be considered very large and may indicate the use of several different tin sources, because otherwise the variation would be smaller.

Interestingly, the tin isotopic composition of one item differs significantly from all others. The $\delta^{124/120}$ Sn ratio of a bronze tutulus from a large burial mound in Glavanești Veche (Junghans, Sangmeister and Schröder, 1968, no. 6567, 8568; Comşa, 1987, pp.372-373, fig. 6, fig. 7, fig. 11, 2-3, fig. 12, 1-3; Motzoi-Chicideanu, 2012, p.106), which belongs to the Ochre Grave culture, is much higher than those of all other bronzes in the sample set (Figure 3). It has even higher isotope ratios than the average of the southern European sample set and the averages of sampled ores from the tin province in southern England or the Saxon-Bohemian Ore Mountains. This indicates the use of a different tin source to manufacture the item. Even though currently that source cannot be identified, this is particularly interesting since the tutulus belongs to the oldest artefacts and is furthermore the only realy early tin bronze from the lower Danube region in the sample set. Thus, it is of interest to compare the results with those of the other sample sets.

Mesopotamian bronzes of the 3rd millennium BC

In the second half of the 3rd millennium BC the number of tin bronzes began to increase significantly in Mesopotamia and the Near East. There tin bronze was used to manufacture vessels, daggers, axes and bracelets. Typological similarities among the majority of these early bronzes indicate a Mesopotamian influence on Anatolian metalwork. Almost all tin bronzes around 2500 BC occur in burials and hoards. The Mesopotamian sample set includes Early Dynastic III and early Akkadian bronzes (2600 to 2200/2150 BC), which represent different objects groups and were found predominantly in the rich graves of Ur and Kish (Hauptmann and Pernicka, 2004, pp.7-8). In addition, bronzes from well investigated tell settlements like Tell es-Suleimeh, Tell Asmar (Müller-Karpe, 2004, p.5), Tepe Gawra (Moorey and Schweizer, 1972, p.186) and Nippur are part of the sampled items (Figure 2). Their tin content varies between 3.5 and 17.2 wt. %, which indicates yet a lack of standardisation in the manufacturing process of tin bronzes. A large isotopic variation of the $\delta^{124/120}$ Sn ratios between -0.18 ‰ and 0.44 ‰ (Figure 4) is comparable with that of artefacts from the European sample set. Again, the use of different tin sources could explain these results. Besides this, the isotopic composition excludes a regular blending of different tin ores before or during the manufacturing process, because this would lead to homogenisation of the metal inventory and thus to a low variation in isotopic ratios.

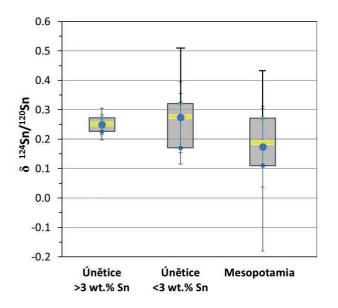


Figure 4. The variation of the $\delta^{124/120}$ Sn ratio in Early Bronze Age bronze objects belonging to the Únětice culture in Central Germany and Mesopotamia; Yellow lines = median value; large blue dots = average of the isotope ratio distributions (created by Gerhard Brügmann).

Central European bronzes of the 2nd millennium BC

Since the isotope ratios of bronzes dating to the 3rd millennium are diverse, it seems reasonable to compare them with those of bronzes of the 2nd millennium BC, so as to investigate whether any changes can be identified. Two sample sets were included in this study: the first set contains bronzes from Central Europe, and the second set from Southeastern Europe.

The sampled central European bronzes belong to different hoards of the Únětice Culture, which date between 2100 and 1700 BC (Rassmann, 2010, pp.809-812). The bronzes represent different artefact types such as weapons, tools and jewellery. A systematic study of their chemical composition established that most of the artefacts consist of fahlore copper (Lutz, Pernicka and Pils, 2010; Lutz and Pernicka, 2015). Like the Mesopotamian objects, the bronzes have highly variable tin contents, which ranges from 0.11 to 14.4 wt. %. This reflects different chronological positions of the finds. There is a tendency recognizable that younger hoards have higher and less variable values. A large variation is seen in the $\delta^{124/120}$ Sn ratios of the Únětice bronzes, ranging from 0.12-0.51 ‰. This is mainly due to the bronze objects with tin contents below 3 wt. % (Nessel, Brügmann and Pernicka, 2015). Apparently, no standard alloy composition was yet established and in some objects tin may even be an unintentional component. Tin bronzes having more than 3 wt. % tin have on average a much smaller

range between 0.2 and 0.31 ‰. The rather uniform isotopic ratios of the high-tin bronze artefacts imply that the tin added to the copper had also a rather homogeneous composition.

Southeastern European bronzes of the 2nd millennium BC

The beginning of the 2nd millennium marks the beginning of the Middle Bronze Age according to southern European terminology. At present 64 bronze artefacts were isotopically investigated, which are geographically fairly widely distributed between the Aegean and the upper Danube region. The sample set includes finds from the Carpathian Basin, Oltenia, Muntenia, Moldova and Crete, which date between 2100 and 1600 BC. In addition, some Aegean bronzes are slightly younger and date between 1700–1450 BC.

Again, objects of different types like swords, axes and bracelets were sampled and their chemical composition determined. They also consist of fahlore copper, which derived from the eastern Alpine region and the Slovak Ore Mountains (Pernicka, 2013; Pernicka, et al., 2016).

The tin contents vary from 3.1 to 11 wt. %, which is similar to the range observed in contemporary bronzes of the younger phase of the Early Bronze Age in Central Europe. Yet, the differences in the $\delta^{124/120}$ Sn relation in bronzes with higher tin contents (2-7 wt. %) are less obvious than in bronzes from Central Germany. The $\delta^{124/120} Sn$ ratios in the southeastern European artefacts vary from 0.06-0.35 ‰ (Figure 5). This smaller isotope variation indicates the exploitation of only a few, if not just one tin source. Since the isotope ratios of the bronzes overlap with the isotopic data from the tin provinces of southern England and the Saxo-Bohemian Ore Mountains, these two source regions cannot be distinguished. Only two early objects from Crete, which date around 2000 BC, have higher $\delta^{124/120}$ Sn ratios than the average of the southern European sample set and the ore samples from the Cornwall and Devon tin province and the Saxon-Bohemian Ore Mountains. This again suggests the use of a different tin source for the manufacture of the bronzes from Crete and the remaining bronzes from the sample set.

Discussion

European and Mesopotamian bronzes of the 2nd half of the 3rd millennium BC show a great variation of tin isotope ratios, which seems to indicate the use of several tin

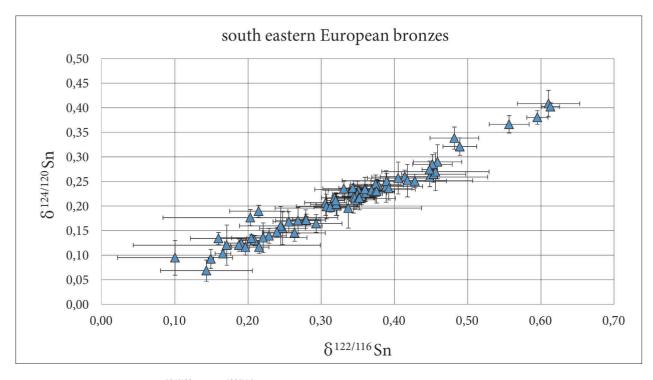


Figure 5. The variation in the $\delta^{124/120}$ Sn to $\delta^{122/116}$ Sn ratio in late Early and Middle Bronze Age artefacts of Southeast Europe found in hoards and settlements or separate finds.

sources. This is particularly interesting when recalling that at this time tin bronze metallurgy was already fully developed in the Near East, whereas it seems to have been in a more experimental stage in Central and Southeastern Europe. There the reproduction of manufacturing processes was still challenging, which basically did not change until 1900 BC.

At the beginning of the second millennium BC only a small number of technologically more developed bronzes are known from Early Bronze Age burials in southern Germany (Krause, 1988, p.191). However, a fully developed and widespread bronze technology in continental Europe is only recognizable from about 1700 BC onwards, which means in the younger, more developed phase of the early European Bronze Age (A2 in the Reinecke system).

In Central Europe this period is dominated by the Únětice culture and in Southeastern Europe by several different archaeological groups. The bronzes of these cultural phenomena show a significantly smaller variation in their tin isotope ratios than those of the 3rd millennium BC. This suggests a more homogeneous composition of the tin used for the production of bronzes of the 2nd millennium BC than for older bronze artefacts. The data suggest that probably fewer different tin sources were used to produce the 2nd millennium BC bronzes. Considering the distribution area of the Únětice culture directly next to the Saxo-Bohemian Ore Mountains and the strong typological relationship between the bronzes of both regions in 1800–1600 BC, it is even possible that only a few sources, if not just one particular source, were used to produce the sampled bronzes. Even if the source(s) are currently difficult to identify exactly through tin isotope analyses with the current database, the results indicate at least one important change in raw material supply.

This hypothesis is supported by the isotopic ratios of three bronzes from the lower Danube region and the Aegean, which date to the turn of the 3rd to the 2nd millennium: They have higher $\delta^{124/120}$ Sn ratios than the southeastern and central European sample sets and also the average values of the mentioned ore samples. This might suggest that they were probably manufactured using one or more different tin sources than exploited for the production of the other, in part much younger analysed bronzes. The data also indicate a reorientation of exchange routes and suppliers at least one time at the beginning of the 2nd millennium BC. Besides this, a development towards more standardised production processes resulting in a regular tin content of more than 3 wt. %, between 2000 and 1600 BC, is indicated by the analyses results.

Conclusions

Traditionally tin-bronze metallurgy was seen as having been invented in the Near East and spread through Europe either via its Southeastern Europe or via the Mediterranean Sea. Present discussions favour a distinctive adoption and establishment of tin-bronze technology in several different regions in Europe and the Near East (Pare, 2000; Radivojević, 2013; Yener, et al., 2015; Nessel, et al., 2018). A comparison of the tin isotopic ratios of cassiterite and bronze artefacts from Europe and the Near East supports the latter hypothesis. Considering the large tin isotope variation in bronzes of the 3rd millennium, it is probable that tin from several different deposits was used to manufacture them. In contrast, the tin isotopic ratios of European bronzes of the 2nd millennium BC show a significantly smaller variation.

Although an exploitation of at least one of the major European tin sources in Cornwall and Devon or the Saxo-Bohemian Ore Mountains is highly likely, it turned out that it is difficult to distinguish them isotopically, due to a significant data overlap. This is unfortunate as most of the isotopic values of central and southeastern European bronzes plot into this overlapping data range as well. However, this might also suggest the simultaneous use of both occurrences at least in the 2nd millennium BC.

Altogether, 95 % of the tin isotopic ratios of southeastern European bronzes do not differ from those of the central European sample set, which makes it likely that the tin used to manufacture them came from a very similar European tin source, probably west of the Tisza River region. A stylistic and typological comparison of the bronzes reveals that particularly most early bronzes in southeastern Europe show strong typological connections to Central European artefacts (e.g. Bátora, 2000). Therefore, a trade of finished and later also semi-finished bronze objects between central European communities and those of the Carpathian Basin needs to be considered. Bronzes with a regular tin content above 3 wt. % and a technologically advanced mode of production appear considerably earlier in Central Europe than in southeastern European contexts. The same is observed for the distribution of tin bronzes in general. An intensive use of tin bronze cannot be observed in Southeastern Europe before 1850 BC, because only very few bronzes date before 1900 BC. Therefore, the rise of tin-bronze metallurgy in the last quarter of the 3rd and the 2nd millennium BC in the Carpathian Basin and perhaps even the Balkans seem to have been influenced by the central European Únětice culture and related groups (Nessel and Pernicka, 2017; Nessel, et al., 2018, rather than by a transfer from the Aegean. A transfer of knowledge and technology via the Danube, Tisza and other great rivers seems reasonable.

The tin isotopic ratios of two 2nd millennium BC bronzes from Crete and one item from the lower Danube, which dates to the last third of the 3rd millennium BC, had higher tin isotope ratios than all other objects and the average of all ore samples from southern England and Central Germany/Czech Republic. These bronzes mark a change in raw material supply at the turn of the millennia. The tin sources used for 3rd millennium bronzes are different from those of the 2nd millennium. This is particularly indicated by the tin isotope composition of younger Aegean bronzes from the 15th century BC, which do not differ from those of the central European bronzes and ores anymore. Furthermore, it should be emphasised that no tin isotopic ratios of bronzes found between the Thracian plain and the lower Danube region plot close to the Cretan finds. This might indicate that tin-bronze metallurgy did not spread from the Aegean to the North at the turn of millennia. Instead, it seems more likely that communities between the Thracian plain and the southern Carpathian Mountains obtained raw material to produce tin bronze also via the Danube from Western and Central Europe.

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