

www.aaatec.org ISSN 2310-2144

# Social processes in ancient Europe and changes in the use of ore and alloys in metallurgical production

#### **Stanislav A. Grigoriev**

Institute of History and Archeology, Ural Branch of the Russian Academy of Sciences, Chelyabinsk, Russian Federation; E-Mail: stgrig@mail.ru

#### Abstract

In archeometallurgy, the main trends in the development of ancient technologies are well studied. And, usually they are considered as two principal trends. The first is associated with the type of minerals used: native copper — oxidized minerals — sulfide minerals. The second trend is associated with the types of metal used: pure copper – arsenic and antimony-arsenic copper – tin bronze. On the basis of materials from Northern Eurasia, we demonstrated that both these trends were interrelated (Grigoriev, 2017). The transition to new types of raw materials caused the transition to new types of copper alloys. This was caused, for example, as in the case of the transition from arsenic alloys to tin, by that in the production of arsenic alloy, ore with additions of arsenic minerals was smelted. But after the following transition to richer ores from quartz or to sulphides, conditions were created in the furnace when arsenic evaporated, which made it impossible to produce alloyed metal. This caused the transition to tin alloys, as tin was alloyed directly with copper. In the long run, this system depended on socio-economic processes, since they stimulated the growth of metal consumption and the need to use other types of ores. Tin, whose deposits are very rare, provoked prerequisites for creating a wide network of trade and exchange.

The task of this work was to study this system on the European material. The analysis showed that, in general, to Europe all the same regularities may be applied, which makes it possible to consider them as universal. There are some differences caused by the abundance of fahlores in Europe, which made it possible to produce antimony-arsenic alloys in some regions. Another feature is a higher level of economic development, compared with the Eurasian situation, and the proximity of the Eastern Mediterranean, where early civilizations arose rather early. As a result, a global network of trade and exchange was formed in Europe already by the Middle Bronze Age.

Keywords: Europe, Bronze Age, ore, types of alloys, socio-economic relations.

In archeology, the basic regularities in development of metallurgical technologies are well known. The general trend of this development is the following: 1) gathering native copper on the surface and its forging and casting; 2) smelting of oxidized copper ores; 3) smelting of sulphide ores (secondary sulphides and their mixture with oxides were initially smelted within this stage, and then the transition to smelting of primary copper-iron sulphides took place); 4) smelting of iron ore (Strahm, Hauptmann, 2009).

There is a clear understanding that these regularities are associated with two factors:

1) The sequence of occurrence of copper minerals in deposits, where the oxidized ores lie at the top, below is the zone of secondary sulphide enrichment with fahlores and other ores, and under it the main body of primary copper-iron sulphides. There are many deviations from this rule, but in this case we are discussing the main trends.

2) Technological development. Strictly in accordance with this geological sequence, the processing of underlying minerals required higher temperatures and new techniques in ore preparation, mining, etc. Therefore, when developing the upper zones of the deposits, pure or arsenic copper was produced, below it was possible to find fahlores and produce arsenic and antimony copper alloys. Antimony is not as volatile as arsenic, but deposits with copper-antimony minerals are less common, which led to the transition to tin alloys (Tylecote, 1976, pp. 7, 9).

And, there is an unconditional understanding that all this was connected by different channels with social processes, since it coincided with the growth of metal consumption. The peculiarity is that when smelting most part of oxidized ores, it is rather difficult to separate slag from metal. As a result, conglomerates were formed with many copper inclusions, which had to be removed mechanically, and scrupulously enough, and then it was necessary to re-melt them. All this limited the volumes of smelting (Strahm, Hauptmann, 2009, S. 123). In addition, the oxidized zone is poorer than the sulphide zones, and its ore volume is noticeably less.

There is another process – the development of alloying technologies: from pure copper to the use of arsenic copper and further to tin bronze. This is a fairly universal picture, which is often regarded as an independent process of technological development.

And there is an unconditional socio-economic component of the whole system. This is the role of metal in society, its place in formation of social hierarchy, in creation of exchange networks, etc. Geology is also an important factor in this case, namely, the irregular distribution of mineral resources.

## **Ores and alloys**

In the previous article I discussed the relationship between the ore base and the types of alloys in Northern Eurasia (Grigoriev, 2017). The general pattern is the following: at the first stages, when people used native copper or relatively pure copper carbonates, pure copper dominated. Then, with the beginning of the use of less pure ores (with gangue), some other minerals began to fall into the charge. In many areas of the world, copper mineralization is accompanied by copper-arsenic and arsenic mineralization, and if such minerals were taken into the furnace, it was possible to produce arsenic copper, which after forging is harder than pure copper. But, this required some different casting and forging technologies. We have already discussed the advantages of arsenic copper over pure copper (Grigoriev, 2017, pp. 19-21), but taking into account European perspective, let us touch upon the main aspects, since it is believed that the properties of low-arsenic and pure copper are not very different. Only since 4% arsenic content this metal shows sharp differences in hardness (Northover, 1989, p. 113).

Indeed, after cold-working (75% reduction) objects from pure copper can reach a hardness of 135 HV, and the hardness of 2.6% arsenic copper after cold-working increases from 65–70 HV to 150–160 HV. But even in the range of 0.5–2%, the toughness and strength of this metal is noticeably better than that of the pure copper, since arsenic also played a role of deoxidant (Scott, 1991, p. 82; Kienlin, 2008, S. 268; Budd, Ottaway 1990, p. 95).

The next transition to tin alloys is less understandable, since tin at low concentrations has insignificant advantages over arsenic (Kienlin, 2008, S. 180). But on the materials of Northern

Eurasia, the relationship between the use of low-melting ores and arsenic, and the use of refractory ores and tin is quite clearly visible (Grigoriev, 2017). This is explained by that in high-temperature smelting under oxidizing conditions the arsenic forms an oxide that volatilizes from the metal. This volatility and arsenic toxicity provoked even ideas about transition to tin alloys for medical reasons (Muhly, 1976, p. 90), and this idea is often present in the literature. But in antiquity before this, people used arsenic many years, and simply did not know about this problem. Moreover, in Northern Eurasia, ore with arsenic was smelted in dwellings, and only after abandoning these ores did the smelting in dwellings disappear, which was caused by the smell that occurs in process of smelting sulphide ore (Kienlin, 2008, S. 280; Grigoriev, 2017, p. 22). Therefore, it is more important to discuss the properties of these metals.

Of course, above all for ancient people the working properties of tools were important. And the advantages of high-alloyed metal are obvious. With a low tin content, there is no difference in the use of axes made of tin bronze and arsenic copper. But the high contents change the situation. An axe without tin impurities should be sharpened after 15–30 minutes of work, and an axe with 6% tin content should be sharpened after 250–310 minutes (Kienlin, 2008, S. 184, 247). At low concentrations, tin bronzes are only slightly harder than arsenic copper. Only after coldworking with 50% reduction the tin bronze is harder than arsenic copper. Its hardness rises to 200 HV only in objects containing over 6% tin (Kienlin, 2008, S. 176, 180, 274, 275). But even with the 8% alloying, after cold-working with 50% reduction, the hardness of tin bronze is less than a quarter higher than that of arsenic bronze (Scott, 1991, p. 83). However, the problem is that in antiquity it was difficult to create an arsenic alloy with content higher than 8% (Northover, 1989, p. 113). In contrast, tin makes it possible to increase its content regardless of the original ore, which ensured the stability of the result (Kienlin, 2008, S. 176).

But at the early stages of metallurgy there were no high-alloyed bronzes. In the LBA of Northern Eurasia and in the late EBA (Br A2) of Europe, the most objects have insufficient alloying for effective competition against arsenic and antimony-arsenic alloys. In addition, tin deposits are relatively rare; this metal had to be transported over long distances, so the tin alloys with a slight advantage over arsenic alloys could not be very profitable.

And we come back to the volatility of arsenic. Under oxidizing conditions it forms a trioxide which evaporates (McKerrell, Tylecote, 1972; Sabatini, 2015). This limited the re-melting and annealing of this metal. It was a reason why cold-working dominated in the Eneolithic Central Europe and throughout the Bronze Age in Iberia (Kienlin, 2008, S. 108, 109). The same situation was in the Urals: metallurgists annealed arsenic copper at low temperatures, the casting was simple and very limited (Degtyareva, 2010, pp. 121, 123, 134, 138). Nickel additions can partly solve this problem; they contribute to the preservation of arsenic in metal. Therefore, in many areas we see nickel in ancient arsenic copper (Tylecote, 1976, p. 9). Moreover, in some instances nickel was a deliberate addition (Ryndina, Ravich, Bystrov, 2008; Ryndina, Ravich, 2012, pp. 5-9). However, after some re-melts, the degree of alloying gradually decreases. This probably was a reason why tin, which does not have this drawback, contributed to the widespread introduction of casting technologies, and not because the tin bronze fills casting molds better. T. Kienlin showed that its composition does not play a decisive role in the successful casting. More important are other factors: the temperature of the metal and the degree of heating of the mold. And even against the background of pure copper, the melting point of metal with a low content of alloying agent decreases insignificantly: Cu - 1084° C, 5% Cu+As and Cu+Sn - 1000° C. A noticeable decrease below 1000° C begins only if the content of alloying components is 8-10% (Kienlin, 2008, S. 253, 262).

But in Northern Eurasia, in the context of this problem of arsenic evaporation, the transition from poor fusible ores to refractory ores from quartz and to sulphides was more important. After the beginning of their use, the possibility of producing arsenic copper disappeared (Grigoriev, 2017, pp. 23, 24). Smelting of chalcopyrite is a highly oxidizing process (Moesta, Rüffler, Schnau-Roth, 1989, S. 151), and under these conditions, arsenic would oxidize and evaporate. But in Eurasian metallurgy, the oxidized ore from quartz rocks was smelted also under oxidizing conditions.

And, in the context of this work it is important to note that the same pattern has been revealed in Central Europe. Sulphide inclusions were found in tin bronze axes, which indicate the use of sulphide ores. And initially it occurred in those areas where copper contains limited impurities of arsenic and antimony. In general, this process of transition from Altheim axes from arsenic copper to Neyruz axes with antimony and arsenic alloying from fahlores, and further to the late EBA Langquaid axes from tin bronze was directly related to the transition to new types of deposits and new ore smelting technologies, and to the development of the interregional exchange network. This provoked changes in the metalworking technique and also created conditions for the further dynamic development of this production (Kienlin, 2008, S. 235, 251, 276, 278).

There is another relationship: between the growth of metal consumption and the type of ore. Roughly speaking, refractory ores such as copper-iron sulphides or ores in quartz rocks are more widespread and rich. Therefore, this transition to the use of new types of ores was stimulated by socio-economic needs, and it also caused the change in the types of alloys. Changes in metalworking technologies and, in the long run, the morphology of metal objects were also associated with this. But after its appearance, the tin alloying contributed to the formation of a wide network of communications and exchange, since it was necessary to ensure its transportation over long distances, and all agents involved in this process should produce something for these exchange operations.

All together, this formed a very complex set of various relations that defined the originality of metallurgical production in different areas. And since geology of deposits and chemical processes were at the heart of this system, we should expect that it was quite universal. In principle, this is the case, and in most areas we see this sequence of using pure copper, arsenic copper and tin bronze. And the topic of this discussion is how this system worked in the European space.

#### European periodisation of the Eneolithic and Bronze Age

Some terms need to be clarified. There is some terminological diversity in the use of term for the first period, the Eneolithic. If this term was always accepted in southeastern Europe, in many other areas the period of early metal cultures is designated by the term "late Neolithic", and later the term "Copper Age" started to be used. But we will use here the single term "Eneolithic".

There are many systems of periodisation, which are very different, and which are aimed to identify regional specificities, but the main one is P. Reinecke's periodisation for Southern Germany, which is relevant for most areas of Central Europe, and to which other regional schemes are usually linked. This periodisation divides the Bronze Age into three periods: EBA (Br A), MBA (Br B, C) and LBA (Br D, and Ha A, B). The most interesting for us EBA is divided into two parts: Br A1 and Br A2. There is difference between this Central European and Eurasian periodisation: The first period EBA (2200-1500 BC) corresponds in Eurasia to the transitional period between the MBA and the LBA and partly to the developed LBA; the

European MBA, in general, corresponds to the developed LBA (Srubnaja-Alakul period), and the LBA corresponds to the period of the Final Bronze Age in the east.

## Ore base for ancient European metallurgy

To understand the specifics in the development of European metallurgy and its difference from metallurgy in Eurasia, it is necessary to touch upon the problem of the ore base in the regions discussed in this article (fig. 1).



**Figure 1.** Map of the main copper deposits in Europe mentioned in the text: 1 – Rudna Glava, 2 – Ždrelo, 3 – Majdanpek, 4 – Rudnik, 5 – Bor, 6 – Ai Bunar, 7 – Evzoni, 8 – Perivoli, 9 – Othris, 10 – Sithonia, 11 – Euboea, 12 – Cyprus, 13 – main areas of copper deposits in Iberia, 14 – Libiola, Monte Lareto, 15 – Poggio Malinverno, 16 – Grotta della Monaca, 17 – Cabriéres, 18 – Great Orme, 19 – Ross Island, 20 – Mount Gabriel, 21 – Staré Hory, Špania Dolina, 22 – St. Véran, 23 – Oberhalbstein, 24 – Acqua Fredda, 25 – Schwaz, 26 – Brixlegg, 27 – Mitterberg, 28 – Kitzbühel, Viehhofen, 29 – Eisenerzer Alps. Sn – tin deposits.

Romania, Serbia and Bulgaria have many copper deposits of rich oxidized ores (malachite and others) with many outcrops on the surface; the most famous are Rudna Glava, Ždrelo, Rudnik, Bor and Majdanpek in Serbia and Ai Bunar in Bulgaria (Chernykh, 1978, Pernicka et al., 1993, p. 25; Pernicka, Anthony, 2010, p. 170; Radivojević et al., 2010, p. 2777).

It is believed that in the Mainland Greece and on its islands the copper ores are rare and poor. Therefore, traces of their smelting have been rarely found (for the 3<sup>rd</sup> millennium BC, a small number of slag pieces were known on the settlements of Chrisokamino and Skouries) (Muhly, 2008, p. 72). But this opinion about the limited resources is true for the relatively late stages in

metallurgical development, but for its very early stages the situation with the provision of ore is different, since at that time small deposits on the surface were important. And in Greece, these small deposits are known, both on the islands and in the mainland. Above all, we note a series of deposits in ophiolitic complexes: Evzoni (near the border with Macedonia), Perivoli in the Pindos mountains, deposits in the Othris Mountains and on the Sithonia peninsula (Halkidika). Porphyry deposits are also known in northeastern Greece. There are mentions of Strabo about a large field on Euboea (Pernicka, 1987, S. 619–623, Abb. 5).

One of the richest areas in antiquity was Cyprus, where numerous and mighty deposits are situated in the ultrabasic and basic magmatites. There are practically no copper carbonates in these deposits; their main mineralization is represented by chalcopyrite with quartz (Hauptmann, 2008, S. 60; Kassianidou, 2009, p. 57). Therefore, the flourishing of the Cyprian metallurgy started relatively late, in the MBA, when tin alloys and mass production from sulphides appeared. But at the early stages, limited local production of arsenic copper was possible. In the Limassol area there is an arsenopyrite deposit. And nearby in Episkopi-Phaneromeni 1.5% of arsenic was found in a crucible. A piece of slag with arsenic was also found in Troulli. This production is dated to about 2000 BC (Zwicker, 1989, S. 196).

Another copper-rich region, Iberia, is located in the Western Mediterranean. Here, a huge amount of deposits with oxidized copper ores is known, and the main feature of the region is that most ores have arsenic impurities, deposits with antimony are less common, and as an exception (in the provinces of Avila and Toledo) there are copper ores with tin impurities. Only in the south some fahlore deposits are present (Montero-Ruiz, de la Esperanza, 2004, S. 54-59).

In the Central Mediterranean, copper ores used in antiquity are relatively rare. The main ore regions of Italy are in the Alps, western-central Italy and Sardinia, although there are small deposits in Calabria, in Northeastern Sicily and on Corsica. Mineralization is very different: oxides, fahlores, and copper-iron sulphides (Dolfini, 2013, p. 24). The Eneolithic mines in Eastern Liguria (Libiola and Monte Loreto), where the ore is situated on the contact of basalts and serpentines (Pearce, 2009, pp. 280, 281), have been studied better. In the north-west of Sardinia there are copper deposits in ophiolites (Hauptmann, 2009, p. 449), although there is no reliable information about their exploitation. In Etruria, rare outcrops of oxidized minerals have been found in limestone and quartz (for example, Poggio Malinverno), but there are no reliably documented pre-Roman exploitation. The situation with the mines in Calabria (Grotta della Monaca) is unclear (Dolfini, 2013, p. 24; Giardino et al., 2014, pp. 655-657).

In southern France, copper is represented by fahlores with antimony and arsenic, as well as ophiolitic sulphide mineralization with arsenic. For the Eneolithic, ancient mining was discovered in the Roque-Fenestre and Pioch-Farrus 4 mines in the Cabriéres massif (Mille, Carozza, 2009, pp. 148-151), as well as work in the Le Broum valley on small veins of tetrahedrite in association with chalcopyrite, malachite and azurite in dolomites, quartz and quartzite veins (Ambert et al., 2009, pp. 285, 289).

Some copper deposits are known on the British Isles, but they are absent in the most economically developed southern England. Ores are known only in Ireland, Scotland, Wales, Northern England and in Cornwall (Rowlands, 1976, pp. 3, 4). Moreover, in the latter region copper mineralization is associated with tin. One of the largest is the Great Orme mine in North Wales, where chalcopyrite, less commonly malachite, is situated in dolomites (Ixer, 2001). It is estimated that about 1,760 tons of copper were mined here. In Britain this is the earliest known mines, but mining here began after 1500 BC (Parker Pearson, 2009, p. 105). Before this, the metal was transported from Southwest Ireland from the Ross Island mine. Its mineralization is

represented by chalcopyrite, bornite, arsenopyrite and tennantite in limestone. About 1900 BC the mine was abandoned; and the mining was begun in the same area at the Mount Gabriel mine where the ore is represented mainly by copper-iron sulphides (O'Brien, 1994; 2004, pp. 40, 461-468, 572).

In some areas of Central and Western Germany, only small deposits are known, some with fahlores, but evidence on their exploitation in antiquity is not available (Krause, 2003, S. 32, 33). Lower Germany, the Netherlands and Scandinavia have no copper ores.

In the Western Carpathians, in Slovakia, there are deposits with malachite, tetrahedrite and chalcopyrite (Staré Hory, Špania Dolina), and in the Ore Mountains the Zipps-Gemer deposit with sulphide and oxidized ores, but there is no evidence of their use (Krause, 2003, S. 41, 42).

The most important European mining area was the Alps. Many years ago, their main characteristic was formulated: the mineralization of chalcopyrite is typical of the alpine deposits, and often it is associated with quartz (Pittioni, 1954, S. 524). But there are some regional differences. In the west, in the French Alps (St. Véran), the mineralization of bornite is present (Maas, 2004, S. 117-119). In the Western Swiss Alps (cantons of Bern and Valais), a series of chalcopyrite and fahlore deposits exploited in the LBA is known (Krause, 2003, S. 34). In the southeast, in the Oberhalbstein area (Graubünden), primary ores were mined from ophiolites, there are no fahlores (Fasnacht, S. 108, 109; Krause, 2009. S. 52; Naef, 2014, S. 78). In the adjacent area of Italian Trentino, exploitation of the LBA mines of Acqua Fredda with chalcopyrite in schists, quartz, and silicates has been discovered (Hohlmann u. a., 2004, S. 263). But the most numerous traces of ancient works are found in the Eastern Alps, in the North Tyrol, Salzburg and Styria. Most of these deposits are represented by chalcopyrite, but bornite and fahlores (tetrahedrite with antimony and arsenic) are also found. Ores are usually connected with dolomites and quartz (Martinek, Sydow, 2004, S. 201, 202; Goldenberg, Rieser, 2004, S. 39; Goldenberg, 2004, S. 167; Primas, 2008, S. 120). The most important were deposits in sedimentary rocks, Schwaz and Brixlegg in the lower flow of the Inn River with fahlores, and chalcopyrite deposits in the areas of Kitzbühel, Viehhofen in the Glemmtal, in the district of Mitterberg south of Salzburg, in the Eisenerzer Alps and Lower Austria (Lutz, Pernicka, 2013, p. 122, 123).

From this description some important *conclusions* follow. In the Northern Balkans, there was an ideal situation for smelting at the early stages, with the abundance of pure malachite. In the Aegean and Italy, where developed societies emerged early enough, copper mineralization is relatively poor and could not even provide production of arsenic copper. And the future development of civilization here largely depended on the import of raw materials. The exceptions are the ophiolitic deposits in Greece, and probably in Liguria and Sardinia, convenient for the production of arsenic copper. This was enough for the early stages, but it was insufficient after a sharp increase in metal consumption. Cyprian ores are very rich, but they need very developed metallurgical technologies, although at the early stages they could provide a limited amount of copper (including arsenic alloys) for local needs.

In the Western Mediterranean, in Iberia, the abundance of copper-arsenic ores made it possible to start producing high-quality metal early enough, but this provided the following larger proportion of this metal in the Eneolithic and EBA in the region in comparison with other areas.

Rich alpine ores are situated mainly in the east. The oxidized minerals in these deposits are poorly represented, but there is good mineralization of fahlores, which allowed ancient people to produce antimony-arsenic alloys. The main ores are copper-iron sulphides, and they are connected with the refractory rocks, which made it impossible to use them at the early stages. A similar situation is observed on smaller deposits in southern France, Ireland and Britain, where fahlores and copper-iron sulphides also prevail.

And many areas of Western and Northern Europe did not have significant copper sources at all, which, against the background of the early development of agricultural societies, necessitated these regions to be drawn into the exchange system.

## Base of the study

Currently, for Europe and some other regions, the so-called Stuttgart ancient metal database has been created, which includes 35,491 analyses (Krause, 2003). If we leave only the European analyses of the Eneolithic and Bronze Age in it, it will be 29,707 analyses (in some territories the later part of this period is considered as the Early Iron Age). This is a large amount of information that allows us to make statistically reliable conclusions.

German archaeometallurgist, when processing this database, usually refer the metal to more detailed groups based on a combination of different trace-elements, which makes it possible to connect these groups with specific ore types (eg. Junghans u.a., 1960; Krause, 2003). But types of alloys are more interesting for our subject now. There are some problems with their estimation. For tin alloys a conventional threshold of 1% has been taken. The threshold for arsenic and antimony alloys is 0.3% calculated for the Sintashta culture (Grigoriev, 2015, p. 153). But it is necessary to keep in mind the conventionality of this threshold. Copper-arsenic alloys could be obtained in various ways: by co-smelting of copper and arsenic minerals, by smelting specific ores, for example, fahlore, mixing (not always intentional) these ores with ordinary ores. In addition, arsenic evaporates in the processes of re-melting and ore smelting.

	Cu	Cu+As	Cu+Sb	Cu+As+Sb	Cu+Sn+As	Cu+Sn+Sb	Cu+Sn+As+Sb	Cu+Sn	Total
Eneolithic	2,265	1,473	127	91	73	1	17	57	4,104
%	55.19	35.89	3.09	2.22	1.78	0.02	0.41	1.39	100
EBA	1,877	2,060	1,144	6,407	2,401	412	2,844	3,022	20,262
%	9.26	10.17	5.65	31.62	11.85	2.5	14.04	14.91	100
MBA	62	14	5	11	792	67	234	648	1,833
%	3.38	0.76	0.27	0.6	43.21	3.66	12.77	35.35	100
LBA	87	25	13	47	280	193	817	619	2,081
%	4.18	1.2	0.62	2.26	13.46	9.27	39.26	29.75	100
1 <sup>st</sup> mill. BC	551	13	9	4	235	35	87	493	1,427
%	38.61	0.91	0.63	0.28	16.47	2.45	6.1	34.55	100

Table 1. Types of alloys in the Eneolithic and different periods of the Bronze Age in Europe.

Therefore, this threshold is conditional and not universal for each situation. It is taken only for initial calculations. Some conventionality is also present in the chronological grouping, since the unification of all European materials into one horizon reflects different historical processes. For example, metallurgy of the Iberian Peninsula due to geological and historical reasons was subject to regional regularities, which were sometimes subjected by impulses and processes of technological changes that occurred in the northern areas of Europe. In Greece, the Bronze Age started earlier, in the late 4<sup>th</sup> millennium BC, but this study is based on the Central European periodisation. As a result, the EBA materials from Greece fell into the group of Eneolithic

materials. In Bulgaria, the Proto-Bronze Age started in the first half of the 4<sup>th</sup> millennium BC. Therefore, the following text is a rather vague picture of the most general trends (Table 1).

#### European metallurgy in the Eneolithic

In the European Eneolithic, evidence of smelting process is limited. Small rare slag pieces are found in Selevac (Serbia), Zengövákony (Hungary), Brixlegg and Götschenberg (Austria), Monte Loreto (Italy), La Capitelle du Broum (France), Cerro-Virtud (Spain), and less reliable finds on Corsica and Sardinia. As a rule, it was the smelting of relatively pure copper oxides, less often fahlore, in small furnaces in the low-temperature and reducing conditions (Roberts, 2009, pp. 131-134).

The Northern Balkans was the most mighty mining area of the European Eneolithic. Previously, the absence of slag here created an impression that the objects were made from native copper. But the abundance of massive copper tools provoked doubts. Now on many settlements (Medvednjak, Selevac, Vinča, Anzabegovo IV, Stapari, Belovode) already for the first half of the 5<sup>th</sup> millennium BC, malachite smelting has been recognized, and even its connection with specific mines: malachite from the Selevac and Belovode settlements is identical to ores from Rudna Glava (Pernicka et al., 1993, pp. 3, 5, 37; Pernicka, Anthony, 2010, pp. 168, 170; Radivojević et al., 2010, p. 2778). The rarity of slag is explained by the fact that almost no slag is formed after smelting of pure malachite. Even if there is gangue in the ore, pieces of slag are small. For example, on the Belovode settlement, pieces of slag are about 4 gram and contain many inclusions of copper and cuprite (Radivojević et al., 2010, p. 2778). Soon, metallurgy began to spread from Southeastern Europe to Central Europe, where relatively pure copper was also originally used.

And, probably, from the Balkans, metallurgical production spread to the south, to Greece, where the earliest slag was found in the north, in Sitagroi (ca. 4000 BC), and later, about 3000 BC, ore was smelted on the Cyclades (Pernicka, 1987, S. 614). But some eastern impulses are not excluded. In the Copper Age of Greece, analyses show many objects with a high arsenic and nickel content (Pernicka, 1987, S. 695). They probably reflect the smelting of ores from ophiolites convenient for the production of arsenic copper.

Probably, there was a second way from the Near East through the Mediterranean. When this flow of influence reached Iberia and Italy<sup>1</sup>, the smelting of ores mixed with arsenic appeared there. These ores are widely distributed on the Iberian Peninsula. But the same tradition appeared in many other areas.

This picture corresponds to that we see in the table (Table 1; fig. 2). During this period, pure copper and copper-arsenic alloys clearly dominated. This database contains 148 objects with tin, which is about 3.6% of the total. Some of them have the tin content between 1.3 and 10.5%. However, some these objects were found in the context of the Corded Ware and Bell-Beaker cultures, and might be synchronous with the early EBA. Therefore, it is assumed that experiments with different metals took place, but the real bronze metallurgy did not happen (Krause, 2003, S. 210-212). And a significant part of these high-tin finds was found in Greece, i.e., they belong to the EBA too.

<sup>&</sup>lt;sup>1</sup> Perhaps in Italy, metallurgy spread from the Balkans through the Eastern Alps (Dolfini, 2013, p. 43).



Figure 2. The types of alloys in the Eneolithic Europe (%)

The nature of this metal was probably different. In Greece, it is certainly an artificial alloy. Probably, the finds of this time from the context of the Bulgarian Proto-Bronze Age are too. In Cornwall and Saxony, tin is associated with copper in the deposits, so accidentally their ores could be smelted together. Ancient slag from Ranis in Saxony contains 0.62% copper and 0.05% tin, but in copper particles the tin content reaches 1.2%. In the late (19<sup>th</sup> century AD) smelts of Cornwall ore, copper with 0.7% tin content was produced. One piece of ore contained 0.94% tin and 12.3% copper, which could give 7% tin in the metal. Therefore, the early tin bronzes could be a result of using such ores (Tylecote, 1976, p. 14). This is confirmed by finds from the Bronze Age context in Spain. Copper ore with an average tin content of 2.63% was found in the Avila province, on the Bronze Age settlements of Aldegordillo and Gravera del Puente Viejo, but some samples from the mine demonstrate up to 4.99% tin (Montero-Ruiz, de la Esperanza, 2004, S. 60). Therefore, in many instances, this was the result of random smelting.

But in the Northern Balkans, the situation was perhaps different. There are 15 Eneolithic objects from tin bronze with a tin content between 1 and 11%. Their examination shows that since the 5<sup>th</sup> millennium BC metallurgists knew properties of this metal and deliberately smelted it from different mixtures of tin sulphide (stannite) with fahlore, chalcopyrite and other ores, which is confirmed by the discovery of copper-tin slag on the Zengövákony settlement in Hungary. But this was not connected with the production of copper, these objects may be considered together with gold, since the color of tin bronze is similar to the color of gold, i.e., it was occasional production of a prestigious metal (Radivojević et al., 2014, pp. 237-247).

This situation is reflected in table 2. It needs only one comment. The relatively high proportion of tin bronzes in Northern Italy and France could be caused partly by a small number of samples, partly by an uncertain context of the finds. But the following relatively high proportion of bronzes in the Italian EBA allows us to admit the early exploitation of local tin sources.

In other areas, the use of fahlores was started in the Eneolithic, and metallurgists produced metal with a high content of arsenic, antimony and silver. The earliest place of mining and smelting of fahlore is Brixlegg in Tyrol, dated since the second half of the 5<sup>th</sup> millennium BC (Höppner et al., 2005). Later (the 3<sup>rd</sup> millennium) traces of fahlore smelting have been identified in La Capitelle du Broum in southern France, in the Cabriéres massif (Mille, Carozza, 2009, p. 151).

	Northern Balkans	Greece	Northern Italy	Southern Italy, Sardinia and Corsica	Iberia	Ireland	Britain and Ireland	Carpathian Basin	Southern France	France	sdIA	Southern Germany and Czech	Northern Germany, Low Lands, Poland and Scandinavia
Number of analyses	1058	233	35*	1*	952	7*	1*	1029	9*	19*	247	212	299
Proportion of bronzes (%)	4.73**	25.32**	8.57	0	1.47	0	0	0.49	0	10.53	0	1.89**	5.35**
Average tin content in bronzes (%)	5.31	6.24	6.07	-	7.55	-	-	5.68	-	4.5	-	4.8	7.49

**Table 2.** Tin bronzes in the European Eneolithic

\* Statistically invalid sampling

\*\* Part of the finds may relate chronologically to the EBA

Local ore contained up to 20% antimony, and during smelting, arsenic and a part of antimony evaporated from the ore, but the silver content increased. As a result, copper-antimony alloy was produced (Ambert et al., 2009, p. 289). This example is very indicative. Even in the process of smelting relatively pure ore, its alloying components reduce. And only due to their high content in the original ore it was possible to produce alloyed metal. Tylecote wrote about this problem. He pointed out that significant loss of arsenic occurred in case of smelting sulphide ores. Therefore, it was impossible to produce the same arsenic copper as smelting oxides. But sulphides in the zone of secondary enrichment contain more arsenic, and after all the losses, some arsenic remained in the metal (Tylecote, 1976, pp. 6, 7).

In the Eneolithic the metal from these ores was almost invisible in the total consumption (Table 1, fig. 2). However, the concentration of elements in the Eneolithic metal smelted from fahlores was lower than in the following periods (Krause, 2003, S. 133-137, 144, 145). Therefore, in this table a part of this metal might be included in the pure copper. Probably, the reason for this transition to fahlores was, in addition to their broad distribution in Europe, that, compared with arsenic, antimony is less volatile, and therefore is better preserved in the metal.

## European metallurgy in the EBA

In most of Europe, the so-called *Fahlerzmetal*, i.e. smelted from fahlores, is typical of the EBA. This is especially true for areas around the corresponding ore sources. In the Eastern Alps, most of the metal of this period was smelted from fahlores from the Schwaz and Brixlegg areas (Lutz, Pernicka, 2013, pp. 122, 126). This, of course, was combined with the use of oxidized ore, and in some instances on the same settlements. Thus, on the EBA settlement of Buchberg / Wiesing in Austria (ca. 19<sup>th</sup> century BC), both oxides and fahlores were found (Krause, 2003, S. 39). Unfortunately, in Central Europe there are only few smelting sites of this period (Primas, 2008, S. 120). Partly this may be explained by our insufficient knowledge. But it is possible that the small quantity of slag was caused by the careful ore selection and the removal of gangue. For example, ancient smelters in Wiesing used tetrahedrite from quartz and dolomites, and produced copper with high arsenic and antimony contents. At the same time, many iron oxides are present in the slag (56.9%), which does not correspond to the ore and gangue. Therefore, it is assumed that these oxides could be added to the charge (Martinek, Sydow, 2004, S. 204-209). But it is also possible that the ore was carefully selected, with the removal of refractory components of the gangue (see discussion of this subject based on Sintashta slag: Grigoriev, 2015, pp. 137, 137). Therefore, for the smelting they also tried to take pure ore pieces. However, analysis of slag from Brixlegg / Mariahilfbergl shows that some impurities of gangue (dolomite and quartz) fell into the charge (Huijsmanns, Krau $\beta$ , Stibich, 2004, S. 60–61). And, after careful studies of the mines and smelting sites on the Ross Island in Ireland, it was concluded that the gangue was removed from the ore as much as possible, smelting was carried out without fluxes, temperatures were low, and volumes of the ore were small. This caused the absence of large amounts of slag (O'Brien, 2004, p. 471).

In some areas, for example, in Iberia or the Aegean, arsenic copper is more typical. For the Aegean Basin, against the background of the absence of such mineralization, this is unclear, some supplies of alloying raw materials are admitted (Branigan, 1974, p. 58). But for Iberia, these copper-arsenic deposits are typical.

Metallurgical production penetrated into Ireland and Britain rather late, around the mid-3<sup>rd</sup> millennium BC, together with the continental Bell Beaker culture (O'Brien, 2004, p. 1; Parker Pearson, 2009, p. 103). However, there is no evidence about the copper mining of this period in Britain. Chemical analyses of local metal show that all the metal was brought here from Ireland, where it was mined from the Ross Island mines, and, like on the continent, it was fahlore (O'Brien, 2004; Needham, 2004, p. 235). The stage of primary use of copper carbonates was absent here, as in France. This is a reliable indicator that metallurgy was introduced here by migrating people, and was not formed independently.

The development of these processes in the EBA changed the situation: the proportions of pure copper and arsenic alloys fell, but the proportions of arsenic and antimony alloys grew (Table 1, fig. 3). Metalworking technologies corresponded to this use of fahlores. In most areas, small objects were produced, although their total number is large. A good example is the Unětice culture of Central Europe with a set of beautiful foil and wire ornaments, typical also of cultures in the adjacent regions (so called, *Blech- und Drahtindustrie*). There are hoards of massive ingots, indicating a broad metal circulation and the presence of exchange between the producing and consuming areas.



Figure 3. The types of alloys in the European EBA (%).

And the proportion of tin alloys increased noticeably (Table 1, fig. 3). Tin could be also added to the arsenic-antimony alloys. The total amount of tin alloys reached 43.3%, but a significant part of this metal belongs to the younger phase of the EBA (Br A2) (Krause, 1989, S. 27, 28). The situation with tin bronzes at the earlier stage Br A1 of Central Europe did not sufficiently changed. Only some objects allow us to talk about their intentional production. Three daggers with tin were found in the Singen cemetery, on the border of Germany and Switzerland. They have Atlantic parallels which led to a conclusion that they had been imported from Brittany or Britain (Krause, 1989, S. 27, 28). A wire fragment (8.65%), a dagger (5.75%) and a rivet (1.8%)

were found in the Remseck-Aldingen cemetery, and a dagger with tin content of 4.7% was found in the Raisting cemetery. But in other objects this admixture is lower than 1% (Krause, 2003, S. 213, 214). A situation in the Víčapy-Opatovce cemetery is indicative, where 112 metal analyses were done, and only two demonstrated a tin content above 1%, the rest showed a homogeneous dispersal between 0.1 and 1%. Only in the late EBA the situation changed, and objects with a stable high tin content appeared (Krause, 2003, pp. 216, 219, 220, Abb. 200; Kienlin, 2008, p. 184). Therefore, a part of the objects of the phase Br A1 with the high tin content can be considered as the import or the result of intentional production, but another part could be produced from Saxon copper-tin ores.

Imports from Britain in this period were quite possible. The first tin bronzes appeared there about 2300/2200 BC, and in Ireland later, about 2000 BC (O'Brien, 2004, p. 1; Parker Pearson, 2009, p. 103). Taking into account the presence of copper-tin ores in Cornwall, some of these objects could be smelted from them, but as early as 2200 / 2100-2000 BC in Britain, along with the bronze objects, metal tin is known: beads from Sutton Veny and Odoorn, pommel studs on the Bargerootsterveld knive, corroded tin in the rim of one of the Rameldry jet buttons, etc. (Needham, 2004, p. 217). Thus, this technology was known, but it did not become the basis for the metallurgical production.

	Northern Balkans	Greece	Northern Italy	Southern Italy, Sardinia and Corsica	Iberia	Ireland	Britain and Ireland	Carpathian Basin	Southern France	France	sdĮV	Southern Germany and Czech	Northern Germany, Low Lands, Poland and Scandinavia
Number of analyses	432	155	652	59*	744	1,158	312	3,732	525	419	2,885	4,659	4,488
Proportion of bronzes (%)	38.89	38.06	63.5	67.8	31.18	63.13	79.49	54.1	39.05	47.97	37.4	22.39	51.58
Average tin content in bronzes (%)	6.19	6.52	6.7	7.38	8.12	9.07	10.42	7.23	8.32	7.87	7.47	6.77	6.32

Table 3.	Tin	bronzes	in	the	European	EBA

\* Statistically invalid sampling

But in the EBA, the use of other tin sources is not excluded. The largest European tin deposits were situated in Cornwall, so it is not surprising that we see a high proportion of the EBA tin bronzes on the British Isles (Table 3). But beyond this, smaller sources of tin are known in Europe: in Tuscany and Sardinia, in Brittany and the Ore Mountains, in the Northern Balkans and Iberia (Garner, 2013, S. 16–18) (fig. 1). Therefore, relatively high proportions of these bronzes in Italy (63–68%) allow us to admit the exploitation of local sources in Tuscany and Sardinia. And, in this period, the smallest proportion of tin bronzes was on territories of Southern Germany and the Czech Republic (22.23%), although these areas are located directly to the south

of the Ore Mountains. However, the tin mines could be exploited there, since to the north we see a high proportion of these bronzes (51.58%). But in conditions of emerging trade relations, the distance to the sources of raw materials is not always equivalent to a final result. However, this picture does not make an impression of an established global trading system.

## Transformations in the late EBA and MBA

Significant changes started in the younger phase of EBA (Br A2), when we see a widespread transition to tin. Within the system of calibrated radiocarbon dates, the beginning of the period may be dated to about  $19^{\text{th}}/18^{\text{th}}$  centuries BC.

This is an interesting and complex problem. The nature of these processes in Europe is different than that in Northern Eurasia. At first glance, these changes were caused by penetration of eastern traditions. Reinecke's separation of phase Br A2 is based on the emergence of new types of metal, characterized by a series of hoards, above all the Langquaid hoard in Bavaria with a combination of objects made in the local Unětice tradition and a spearhead having eastern Seima-Turbino prototypes. But from table 1 we have seen that tin alloys appeared in Europe before the appearance of objects made in the Seima-Turbino tradition, but only after their appearance tin bronzes become the leading alloy, and with the high tin content.

R. Krause considers this period of the late EBA (18<sup>th</sup>-16<sup>th</sup> centuries BC) within the 4<sup>th</sup> stage of development of metallurgy in Central Europe, when the predominant smelting of chalcopyrite was accompanied by the widespread use of tin (Krause, 2003, S. 11, 84). This last stage (more precisely, phase A2b) corresponds to the penetration of Seima-Turbino bronzes (Grigoriev, 2018, p. 45). Therefore, the appearance of tin was not associated with these eastern impulses, but their effects on the wide distribution of this alloy cannot be excluded.

There is another distinct tendency throughout the Bronze Age: a steady decrease in the arsenic content (Table 4), which made this ligature less effective compared to tin bronzes.

Eneolithic	EBA-1	EBA-2	MBA	LBA	1 <sup>st</sup> mill. BC
1.58%	1.24%	1.08%	0.71%	0.65%	0.55%

**Table 4.** The average arsenic content in arsenic copper (with more than 0.3%).

What was the reason for this decline? In Northern Eurasia, it is clearly visible that during the transition to the LBA (which is close to the end of the EBA and the very early MBA in Europe) there was a transition to richer and widespread ores from acid rocks or sulphides (Grigoriev, 2017). And their smelting caused the evaporation of arsenic, which may explain the decrease in its content in the metal. Unfortunately, there is no way for Europe to investigate this process at a statistical level, since there is practically no slag on the settlements. But as a trend it is well demonstrated.

Already at the end of the EBA, mining activities was relocated from the fahlore deposits to the deposits of copper-iron sulphides. This was not unexpected. These technologies already existed in the EBA. In some areas of the Eastern Alps (East Trentino, Inn Valley) already ca. 2000 BC (Br A1b) chalcopyrite was mined (Stöllner u. a., 2016, S. 76). In the Hautes Alps (St. Véran), mines of the very early EBA have been identified, where bornite was mined and smelted (Maas, 2004, S. 117-119).

But an intensive transition to these ores occurred just at the end of the EBA. This took place in Tyrol, where furnaces and slag heaps of Mitterberg (Mühlbach am Hochkönig) are dated since the late EBA (Primas, 2008, S. 121). About 1900 BC the mining of fahlores on Ross Island in Ireland was stopped and the mining of chalcopyrite began on Mount Gabriel (O'Brien, 1994; 2004, pp. 40, 461-468, 572). Later, already in the MBA, the Great Orme mine in Wales began to be exploited (Parker Pearson, 2009, p. 105).

This transition to the copper-iron sulphides and the decrease in the extraction of fahlores took place everywhere, and it coincided with the transition to tin alloys (O'Brien, 2004, p. 561; 2013; Sperber, 2004, S. 304, 329; Pernicka, Lutz, 2015). There is an opinion that it was caused by tin, since this alloy must be free from impurities (Stöllner et al., 2016, p. 76), but metallographic studies show that additional alloying components do not harm the tin alloying (see Kienlin, 2008). Both in the MBA and LBA, many objects were alloyed with tin, and they have arsenic and antimony impurities (Table 1). It can be assumed that intensive use of metal within the first phase of the EBA led to the exhaustion of more limited (in comparing with the primary ores) fahlore deposits. But after the cessation of mining at the Ross Island mine in Ireland its exploitation was restarted in the 19<sup>th</sup> century AD, and 5,000 tons of ore were mined there (O'Brien, 2004, p. 572). In the Alps, we see a complete cessation of the extraction at the stage Br A2, and it was not re-started in the MBA. But in the LBA, about 1100 BC, people returned to the use of fahlore, along with the use of primary sulphides (Sperber, 2004, S. 329). This means that at least in some areas the cessation of mining was not caused by the exhaustion, but by some other processes.



Figure 4. The types of alloys in the MBA of Europe (%).

As a result, we see (Table 1, fig. 4) that, in general, there is almost no copper from fahlores having only impurities of antimony and arsenic. In some areas it was smelted, but then it was alloyed with tin. In areas close to the chalcopyrite mines in the Eastern Alps, copper from fahlores almost disappeared, and copper smelted from chalcopyrite dominated. Just in this period was a peak in the mining in the Mitterberg district (Lutz, Pernicka, 2013, pp. 122, 124, 126; Pernicka, Lutz, 2015, S. 107-111).

In the territorial distribution of tin bronzes there are distinct differences from the EBA: almost everywhere (where the sampling is statistically reliable) the majority of the metal is represented by tin alloys (Table 5). And, it almost does not depend on the distance to the tin sources. In Eurasia, we have seen this dependence very clearly (Grigoriev, 2017, p. 29, 30). The different situation in the European MBA demonstrates a well-organized trading system.

	Northern Balkans	Greece	Northern Italy	Southern Italy, Sardinia and Corsica	Iberia	Ireland	Britain and Ireland	Carpathian Basin	Southern France	France	sdlA	Southern Germany and Czech	Northern Germany, Low Lands, Poland and Scandinavia
Number of analyses	68	34*	96	97	74	15*	61*	78	79	488	262	54*	416
Proportion of bronzes (%)	97.06	17.65	96.87	80.41	100	100	100	93.59	97.47	99.59	96.18	100	94.95
Average tin content in bronzes (%)	7.49	6.5	7.5	7.22	11.59	8.77	14.1	7.81	9.76	12.49	8.07	7.5	8.32

**Table 5.** Tin bronzes in the European MBA.

\* Statistically invalid sampling.

#### European metallurgy in the LBA

Initially in the LBA the tendencies remained that had been formed in the MBA. The mining and smelting of copper-iron sulphides dominated in many areas: smelting of bornite in Saint-Véran (Hautes Alpes) (Maas, 2004, S. 117-119; Mille, Carozza, 2009, p. 158), exploitation of chalcopyrite in Mitterberg (Lutz, Pernicka, 2013). Some smelting sites are known where only chalcopyrite was used, for example, Versunkene Rirche, Trieben (about 1000 BC) (Preβlingen, Eibner, 1989, S. 238, 239). The mining and smelting of chalcopyrite also appeared in new areas: in Trentino (Acqua Fredda) in Northeastern Italy (Hohlmann u. a., 2004, S. 265, 266), Graubünden (Oberhalbstein) in Southeast Switzerland (Naef, 2014, S. 83). But in some places chalcopyrite was smelted together with fahlores, for example, in Paltental (Styria in the Eastern Alps) (Presslingen, Eibner, 2004, S. 66). And this was not an accidental episode, but a reflection of important patterns that help to understand mechanisms in the development of metallurgical production.

In the LBA and the Early Iron Age the metal from fahlores of the Schwaz and Brixlegg areas reappeared in the Eastern Alps, and only a quarter of the LBA metal was smelted from chalcopyrite, which was probably caused by the exhaustion of chalcopyrite close to the surface (Lutz, Pernicka, 2013, pp. 122, 126; Stöllner et al., 2016, p. 77). And many objects were melted from a mixture of metal from these two sources, possibly to reduce the content of arsenic and antimony and to improve forgeability (Lutz, Pernicka, 2013, p. 124). But, probably, it is a natural result of metal circulation and repeated re-melting of scrap.

The same processes occurred in Switzerland, where in the mid-11<sup>th</sup> century BC the fahlores were used again (Primas, 2008, S. 124).

Smelting sites reflecting these processes are also known in the Eastern Alps, for example, Mauk A (12<sup>th</sup>-11<sup>th</sup> centuries BC) located not far from the Brixlegg fahlore deposit. In this place fahlore was smelted. It was probably impossible to separate slag from metal. Therefore, the slag was grinded and then washed to extract copper particles. But in addition to fahlores, chalcopyrite was also smelted there (Goldenberg, Rieser, 2004, S. 45, 49; Töchter u. a., 2013, S. 4). Thus, at this time there was no complete transition to fahlores. The old technological tradition remained, and the metal from fahlores was alloyed with tin. Just the ratio of ore sources was changed (Table 1, fig. 5).



Figure 5. Types of alloys in the LBA of Europe (%)

Table 6. Tin bronzes in the European LBA.

	Northern Balkans	Greece	Northern Italy	Southern Italy, Sardinia and Corsica	Iberia	Ireland	Britain and Ireland	Carpathian Basin	Southern France	France	Alps	Southern Germany and Czech	Northern Germany, Low Lands, Poland and Scandinavia
Number of analyses	483	6*	48	9*	55*	9*	7*	442	134	8*	763	32*	85
Proportion of bronzes (%)	92.55	100	100	88.89	85.45	100	100	88.91	90.3	87.5	92.92	87.5	92.94
Average tin content in bronzes (%)	6.14	6.8	8.91	7.37	9.25	10.48	11.29	6.77	7.77	8.5	7.67	10.14	6.24

\* Statistically invalid sampling

And, the spatial situation with tin bronzes is similar to that in the MBA: they dominate in all regions, regardless of the distance to tin sources (Table 6).

To explain all these transitions to other types of raw materials, it is necessary to discuss the problem of metal consumption.

## Dynamics of change in metal consumption

#### Metal

Above all, it is necessary to pay attention to the growth of metal consumption. It is difficult to estimate this growth reliably. For example, in the Stuttgart database, there are 4,104 analyses of the Eneolithic and 20,262 of the EBA, i.e., an increase of almost 5 times (fig. 6, a). But it should be noted that, first of all, the earliest metal was analysed. For the Eneolithic, the proportion of the analysed metal is 90%, and for the EBA it is 70–80% (Krause, 2003, S. 52). In addition, the size of objects increased, therefore, the real growth was 10–15 times. It is also necessary to take into account that the Eneolithic period lasted for more than 2000 years, and the EBA did about 700 years, which increases this metal consumption by 2 times (fig. 6, b). And to an even greater extent this may be applied to all subsequent periods. In particular, about the sharp increase in metal consumption in the late  $3^{rd}$  and early  $2^{nd}$  millennium BC wrote M. Primas (1997).



**Figure 6.** The increase in the metal production in the Eneolithic and the EBA: a - the number of metal objects in the Stuttgart database; <math>b - a possible real ratio of metal production in the Eneolithic and the EBA.

But the most important changes took place ca. 1600/1500 BC (Radivojevic et al., 2018, p. 22), which is clearly visible at the regional level. In Ireland for the period of 2400-2100 BC about 700 copper objects are known, for the period of 2100-1500 BC – 1700 objects, and for the period of 1600-1300 BC – about 2500 bronze objects (O'Brien, 2004, pp. 1, 5) (fig. 7, a).





At the same time, taking into account that the range of 2100-1500 BC is twice as long as the other two, the real metal consumption during this period did not grow. The boom happened already in the next period (fig. 7, b).

In Scandinavia between 2350 and 1500 BC the metal consumption was constantly growing, but in the period of the Nordic Bronze Age IB (1600-1500 BC), this growth demonstrates explosive patterns (Table 7, fig. 8) (Vandkilde, 2010/11).

**Table 7.** The number of metal objects in Scandinavia from the Late Neolithic I (LN I) to the Nordic Bronze Age IB (NBA IB) (after Vandkilde, 2010/11, fig. 2A).

LN I	LN II	NBA IA	NBA IB
23	186	275	1,044



Figure 8. The growth of metal consumption in Scandinavia

#### Furnaces

This sharp increase in production may be demonstrated by furnaces. In the Eneolithic and the EBA their number is very limited. But for the MBA and LBA, many furnaces are known, especially in the Eastern Alps. They were excavated on the settlement of Mühlbach, Mitterberg (Herdits, Löcher, 2004, S. 188). In St. Johann in Pongau (Salzburg) 50 bowl furnaces with a diameter of 80–130 cm were excavated (Moosleitner, 2004, S. 213). And for the MBA and LBA of the Alpine region, we already know several smelting places with batteries of furnaces standing close to each other, which indicates a noticeable increase in production: Hechenberg (North Tyrol); Oberschwärzen and Eisenerzer Ramsau (Styria); Acqua Fredda (Trentino); Paltental (Obersteiermark); Kitzbühel (Tyrol), etc. (Preβlingen, Eibner, 1989, S. 236; Goldenberg, 2004, S. 168; Presslingen, Eibner, 2004, S. 68; Cierny u. a., 2004, S. 158-159; Klemm, 2004, S. 192, 193; Primas, 2008, S. 121).

## Slag and ore heaps

This growth may be also well traced by the study of ancient mining and slag heaps. According to estimations of Chernykh, 2,000-3,000 tons of ore were mined and about 500 tons of copper were smelted near the Eneolithic mine of Ai Bunar in Bulgaria (Chernykh, 1978; Pernicka, Anthony, 2010, p. 170). In Eastern Liguria, in Monte Loreto, about 2,250 tons of ore were mined in the Eneolithic, and probably it is true for the nearby Libiola mine. The average annual copper production was 515 kg (Pearce, 2009, p. 281).

In the Alps, 10–14 tons of copper were mined annually, but these figures are probably overestimated (Primas, 2008, S. 117, 120), although they still do not contrast as sharply as the next ones.

The situation radically changed in the late EBA and in the following periods. In Saint-Véran (Hautes Alpes), about 2,000 tons of ore were mined in the LBA (Mille, Carozza, 2009, p. 157). But an especial boom happened in the Eastern Alps. This time is marked by the appearance of colossal slag heaps, in large quantities and in various locations (Preβlingen, Eibner, 1989, p. 235; Goldenberg, 2004, p. 165; Fasnacht, 2004. 108; Wyss, 2004, p. 113-119; Herdits, Löcher, 2004, S. 188; Primas, 2008, S. 122; Krause, 2009, S. 52). This plenty is complemented by volumes. In Switzerland, in Oberhalbstein (Graubünden), the total weight of the LBA slag heaps is 76.5 tons (Naef, 2014, S. 80). In Acqua Fredda (Trentino), 800–1,000 tons of slag have been found (13<sup>th</sup>-11<sup>th</sup> centuries BC) (Cierny u. a., 2004, S, 158). At the Mauk A smelting site (12<sup>th</sup>-11<sup>th</sup> centuries BC), the weight of slag heaps is 100 tons, therefore, about 5-10 tons of copper were smelted (Töchter u. a., 2013, S. 4). It is believed that in Mitterberg between 1700 and 1000 BC about 7,000 tons of copper were produced, i.e., about 10 tons annually (Krause, 2003, S. 206 with reference to Eibner 1982). These facts led to a conclusion that the LBA production in the Eastern Alps reached an industrial scale (Presslingen, Eibner, 2004, S. 64).

Archaeological evidences demonstrate a sharp increasing in metal consumption. If in the EBA the most part of the metal was used by elites, in the LBA it had wider application. There are calculations that only to Denmark more than 1 ton of copper had being brought annually (Radivojevic et al., 2018, p. 32, 33).

But even against this background, the scale of smelting on Cyprus is impressive, where slag heaps are dated since the MBA. About 4 million tons of slag are found on the island (Hauptmann, 2008, S. 60). Probably its part was smelted during the Classical antiquity, but the volumes are amazing.

## Social processes

Thus, we see that, as well as in Eurasia, in Europe the types of ore were linked with the types of alloys. The use of pure copper is associated with relatively pure oxidized ores, primarily with malachite. The next stage of arsenic and antimony-arsenic alloys is associated with the use of the corresponding low-melting oxidized and sulphide ores. Finally, the widespread use of tin bronzes is associated with the smelting of copper-iron sulphides. Tin may be alloyed with copper smelted from any sources. But this transition to the smelting of chalcopyrite and tin alloys was accompanied by the cessation in smelting fahlores, for the reason that they could not provide large production volumes. Although the process of this smelting was easy, this was production without slag. Accordingly, the metal could not be completely separated, and crushing and sorting of the smelted mass was necessary, as is the case with oxidized ore. And each time the transition to new types of ore was accompanied by the increase in production volumes. But there is a very accurate correlation between the growth of metal consumption and tin alloys (O'Brien, 2011, p. 346; 2013, p. 433).

And we see that the roots of these processes were not in the development of metallurgical technologies. Tin was invented already in the Eneolithic. A part of the early tin alloys of the Eneolithic and early EBA could be a result of random smelting, but in some instances we see intentional production of this metal, but it was not fixed as a stable technological scheme. And the early smelting of copper-iron sulphides in the phase Br A1 did not lead to their widespread use. The rejection of fahlores and the transition to chalcopyrite were not everywhere caused by the exhaustion of resources; this was a rather sharp break with the previous technological tradition. Any technology could occasionally occur early enough, but they become sustainable only when they are needed on the market. Therefore, just the discussed metal consumption growth was at the heart of all these technological changes.

It is obvious that this coincided with significant social changes, but not metallurgy was their basis. In Iberia, throughout the Bronze Age, metal circulated within the region, there was no commodity production, and the volumes were limited. The commodity production appeared only with the penetration of the Phoenicians, and it was based on the silver production. Here, as elsewhere, the growth of metallurgy occurred only after the appearance of the market for its production (Bartelheim, 2007, S. 75, 81, 85, 174, 257).

In Central Europe, the appearance of rich burials in the developed phase of Unětice culture was not associated with the development of metallurgy; it was based on the development of agriculture and salt extraction. Only at the end of EBA significant changes in economy and social structures happened: centers of leaders, hierarchy of settlements, numerous traces of metalworking on settlements and tin bronzes, which were associated with Madjarovce culture and Boheimkirche group (Krause, 2003, S. 260, 261; 2009 S. 64). In the Alps, in conditions of deficiency of fertile land, permanent settlements arose mainly in the phase Br A2, since the 18<sup>th</sup>

century BC, when in other areas the metal consumption grew. Before that, the volume of production was small; there were only seasonal journeys for the ore (Kienlin, Stöllner, 2009, pp. 67, 82-89). The growing market at the end of EBA was supplied just by the smelting of copperiron sulphides. This production was based on separation of slag and metal, which made possible to increase its volumes.

The next boom in Central Europe in the period of Urnfield culture was caused by improved climate, increasing exploitation of agricultural resources and growing population. These reasons resulted in increasing consumption of metal (Bartelheim, 2007, S. 211, 213). Its deficit made it necessary to use all possible sources, including those that were abandoned earlier.

But there was another important factor.

#### Metal trading and circulation

The irregular distribution of copper deposits in Europe always resulted in some circulation of metal. Already in the Eneolithic and early EBA, copper of the Northern Alps penetrated into the barren regions of Northern Germany and Scandinavia. But a sharp increase in the flow of this metal occurred not when the growth of production in the Alps had begun; it started some later. In Scandinavia, a boom in the metal consumption occurred in the Nordic Bronze Age IB (1600-1500 BC) (Vandkilde, 2010/11). And it was caused not only by the possibility of supply, but by local needs. Since this time, Denmark (where the most Scandinavian metal objects are found) was being drawn into the world trade system, and amber even disappeared from the burials, because it was necessary for this trade and exchange (Kristiansen, Larsson, 2005, p. 122). These links between the world and regional exchange and metal are well described by Muhly (1973).

The same took place in Britain and Ireland. Ross Island ores (metal A, with a high content of arsenic and antimony) were used to produce 95% of metal objects in Ireland and 80% in Britain (O'Brien, 2004, pp. 5, 7, 546). But in the 19<sup>th</sup> century BC this mine was abandoned, and chalcopyrite started to be mined at Mount Gabriel. In Britain, the start of extraction of copperiron sulphides from the Great Orme mine occurred later (Wessex II), when the island was included in the broad trade relations extending to the Baltic and Eastern Mediterranean.

This is even more expressed in the most developed regions of the Eastern Mediterranean. On Crete, the metal consumption was already increasing in the MBA, caused by that society was getting richer and participated in exchange with the Levant and Italy (Branigan, 1974, p. 65). But the flourishing Minoan civilization needed more metal, which could not be provided by the poor Cretan ores. Therefore, already in the LM period, tin and copper ingots from Cyprus began to be brought here (Muhly, 2008, p. 72).

The boom of production on Cyprus is associated with these processes. It was also not caused by the internal technological development, but by the other markets. This metal trade began only in the Middle Cypriot III period (1700-1600 BC), but intensive production and trade are dated from the Late Cypriot I (1600-1450 / 1400 BC), when metal trade started to play a crucial role in the economy, and this had an impact on social structures (Bartelheim, 2007, S. 159, 162, 164, 248). Previously, the Cypriot ores were in low demand, as chalcopyrite requires more complex smelting operations. But it is convenient for mass production, and now Cyprian copper ingots spread throughout the Mediterranean (there is even one find in Germany), and their analyses show the presence of sulphide inclusions (Kassianidou, 2009, pp. 57, 58; Lo Schiavo, 2009, p. 170). And especially intensive were deliveries from Cyprus to the Levant and Egypt. Texts from Mari and Egypt make it possible to date the beginning of these supplies to the 18<sup>th</sup> century BC. The Amarna archives (14<sup>th</sup> century BC) contain information about 26 tons of copper brought

22

from Alashia (Cyprus) (Muhly, 1973, pp. 186, 194-196; Knapp, Kassianidou, 2008, p. 135; Kassianidou, 2009, p. 58).

In terms of this close relationship between the consumption and ore type, a very interesting example is Sardinia. A magnificent Nuragic civilization arose there, and there was a boom in metal production and consumption. About 100 hoards of metal are known. Artifacts demonstrate links with the Eastern Mediterranean (for example, tripods and figurines from Cyprus). Finds of many bronze objects and hundreds of Cypriot oxide ingots are known. The number of these Sardinian ingots is even more than in any other area, including Cyprus itself. Therefore, the researchers raise absolutely fair questions: why was it necessary to carry so many ingots to the island with its local ore? And where did so many tin bronzes come from? Does this indicate that Sardinia played a role of an intermediate link in this Mediterranean trade between the east and west? (Hauptmann, 2009, pp. 500, 501; Lo Schiavo, 2009). But above we discussed that the Sardinian ores are associated with ophiolites and comparatively poor. They could not provide mass production, although, judging from the slag in Genna Maria, some copper was produced from the local ore (Hauptmann, 2009, p. 509).

This was even more clearly manifested in the Eastern Mediterranean, where as a result of the massive copper supply to the Levant and Egypt in many old producing centers of the region, the production was stopped; it could not withstand the competition (Weisgeber, 2004, S. 23). It will be re-started later, in the Final Bronze Age, after a sharp change in the political situation in the Near East.

And, there is also the connection between the mass production of Cyprus copper from chalcopyrite and tin. Just the tin trade made it possible to reach the huge scale of extraction of the Cyprian ores, which previously were less interesting, since they were not capable to produce alloyed metal. An important role in this was played by the change in the political situation in the Near East, when in the Old Babylonian period (18<sup>th</sup> century BC) tin through a network of merchants began to flow via Sippar to Mari, and from there to the Levant (Muhly, 1973, pp. 293, 294, 301). We see a similar situation in Europe. For the Bronze Age, the exchange relations between the Atlantic complex and the northern Alps are well known when tin was delivered from the Atlantic zone towards the Alps and copper was flowing in the opposite direction (Brun, 1993, pp. 171, 173). And there was a sharp increase in the production and consumption about the 16<sup>th</sup> century BC, which was partly contributed by those extensive trade relations that the Mycenaeans established in this period.

Thus, the development of economy resulted in the increase of metal consumption, and this triggered mechanisms for the mass production of copper from sulphide ores and the tin trade. But we also see the reverse effect of tin on the choice of ore types (the ability to smelt ores from which it was impossible to produce alloyed metal earlier) and on social processes, as the emerging elites began to control trade (Kristiansen, Larsson, 2005, pp. 104, 112-114, 123; Bartelheim, 2007, S. 258). There is another illustrative example. Above, discussing the return to the fahlores in the LBA, we cited the opinion of the researchers about the need to compensate the lack of chalcopyrite in conditions of the growing consumption. It is possible that for some areas it is only a part of the truth. If to look at the table of the average tin content in tin bronzes (Table 8), we see its constant growth. But during this period there was a decline, which recovers then

slowly<sup>2</sup>. Some authors explain this by problems with tin (Sperber, 2004, S. 329). In addition to the general jump in the metal consumption, at this time in the Near East a weakening of the trade system happened, caused by destructive political events. And this affected partly even peripheral areas, although this process was largely stimulated by the increased consumption, as some decrease in the tin content could be caused by that in this period some of tin bronzes contained other alloying components: antimony and arsenic from fahlores.

Table 8. The average tin content in bronze (with a content of more than 1% Sn).

Eneolithic	EBA-1	EBA-2	MBA	LBA	1 <sup>st</sup> mill. BC
6.1%	6.6%	7.6%	9.7%	7.2%	8.7%

As a result of all these processes, extensive interconnected systems arose, which the authors describe in terms of the "world system" (Edens, Kohl, 1993) or "bronzization", as an equivalent of globalization caused by this introduction of tin alloys (Vandkilde, 2016).

## Conclusion

Thus, the scheme of development of ancient metallurgy, which we discussed for Northern Eurasia (Grigoriev, 2017), is relevant for Europe, but has some new interesting details. Here, the growth in metal needs was also stimulated by the socio-economic development (fig. 9). This caused the transition to smelting of richer and more common ores and technological innovations in their smelting, and then the transition to new types of alloys. As a result, we see the same fundamental correspondences: smelting of relatively pure oxidized ore – production of pure copper; smelting of fusible ores – production of arsenic and antimony-arsenic copper; smelting of copper-iron sulphides – production of tin bronzes. In some areas an important difference was the phase of use of antimony-arsenic alloys from fahlores, which was caused by two factors: the presence of large deposits of these ores and the lower volatility of antimony compared to arsenic.



Figure 9. Scheme of development of ancient metallurgy.

But in Europe, the dependence of metallurgical technologies on the social development and the metal market is clearly manifested. In Eurasia, this took place too; at the transition from the Abashevo-Sintashta period to the Srubnaja and Alakul cultures, the number of people using metal and the metal consumption increased dramatically. But in Europe, due to developed

 $<sup>^{2}</sup>$  These figures are for the whole European space. In some areas, the situation was even more indicative. For example, in North Tyrol, copper contains 1.5–6.5% tin, while in MBA the average tin content was between 8 and 8.6% (Sperber, 2004, S. 330).

agricultural economy and contacts with the Eastern Mediterranean, it is more expressed, and after the transition from the EBA to MBA, a broad market of metals appeared.

In Europe, we see also some other trends, which, being universal, are weakly expressed in Eurasia. Some innovations in metallurgical production might arise, but they are not perceived by the society, and cannot be fixed as a technological tradition. On the other hand, at later stages, this whole system of production began to have an opposite effect on the social development and social structures, as a wide network of regional and interregional exchanges is formed.

As a result, we see a rather complex system, in which many factors intertwine, constantly interacting with each other and differing in individual areas.

## Acknowledgments

This study has became possible thanks to the support of the Alexander von Humboldt Foundation, which provided an opportunity to work in libraries of the Göttingen University and the Free University (Berlin), the grant from the University of Sapienza (Rome), and the grant from the Slovak Academic Information Agency, which supported the work at the University of Bratislava, where this article has been written finally. And I am very grateful to the colleagues from these universities who contributed to the organization and provided excellent conditions for this work: Prof. Dr. Lorenz Rahmstorf (Göttingen University), Prof. Dr. Elke Kaiser (Free University), Prof. Giovanni Gigante (Sapienza University) and Prof. Jozef Bátora (Bratislava University).

#### References

- Ambert et al., 2009 Ambert P., Figueroa-Larre V., Guendon J.-L., Klemm V., Laroche M., Strahm Ch. The copper mines in Cabriéres (Hérault) in Southern France and the Chalcolithic metallurgy. In: Metals and societies. Studies in honour of Barbara S. Ottaway; R. Habelt: Bonn, Germany, 2009, 285-295.
- Bartelheim, 2007 Bartelheim M. Die Rolle der Metallurgie in vorgeschichtlichen Gesellschaften. Sozioökonomische und kulturhistorische Aspekte der Ressourcennutzung. Ein Vergleich zwischen Andalusien, Zypern und Nordalpenraum; Marie Leidorf Verlag: Rahden/Westf., Germany, 2007.
- Borić, 2009 Borić D. Absolute dating of metallurgical innovations in the Vinča culture of the Balkans. In: Metals and societies. Studies in honour of Barbara S. Ottaway; R. Habelt: Bonn, Germany, 2009, 191-245.
- Branigan, 1974 Branigan K. Aegean Metalwork of the Early and Middle Bronze Age; Clarendon Press: Oxford, United Kingdom, 1974.
- Brun, 1993 Brun P. East-West relations in the Paris Basin during the Late Bronze Age // Trade and exchange in prehistoric Europe; Oxford: Oxbow books, United Kingdom, 1993, 171-182.
- Budd, Ottaway, 2004 Budd P., Ottaway B. Eneolithic arsenical copper: chance or choice? In: Ancient mining and metallurgy in Southeast Europe. Archaeological Institute Beograd monographs, 27; Archaeological Institute: Belgrade, Yugoslavia, 1990, 95-101.
- Chernykh, 1978 Chernykh E.N. Gornoe delo i metallurgiya v drevneyshey Bolgarii [Mining and Metallurgy in Ancient Bulgaria]; Akademiya Nauk: Sofia, Bulgaria, 1978.

- Cierny u. a., 2004 Cierny J., Marzatico F., Perini R., Weisgeber G. Der Späpbronzezeitliche Kupferverhüttungsplatz Acqua Fredda am Passo Redebus (Trentino). In: Alpenkupfer – Rame delle Alpi. Der Anschnitt. Beiheft 17; Deutsche Bergbaumuseum: Bochum, Germany: 2004, 122, 155-164.
- Degtyareva, 2010 Degtyareva, A.D. Istoriya metalloproizvodstva Yuzhnogo Zaural'ya v epokhu bronzy [History of metalworking of the Southern Transurals in the Bronze Age]; Nauka: Novosibirsk, Russia, 2010.
- Dolfini, 2013 Dolfini, A. The Emergence of Metallurgy in the Central Mediterranean Region: A New Model. In: European Journal of Archaeology 2013, 16 (1), 21-62.
- Edens, Kohl, 1993 Edens Ch.M., Kohl Ph.L. Trade and world system in Early Bronze Age Western Asia. In: Trade and exchange in prehistoric Europe; Oxbow books: Oxford, United Kingdom, 1993, 17-34.
- Fasnacht, 2004 Fasnacht, W. Prähistorischer Kupferbergbau in der Schweizer Alpen I. In: Alpenkupfer – Rame delle Alpi. Der Anschnitt. Beiheft 17; Deutsche Bergbaumuseum: Bochum, Germany, 122, 2004, 107-111.
- Garner, 2013 Garner J. Das Zinn der Bronzezeit in Mittelasien II; Archäologisches Institut: Berlin, Deutsche Bergbaumuseum: Bochum, Germany, 2013.
- Giardino et al., 2014 Giardino C., Occhini G., Petitti P., Steiniger D. Ricerche archeominerarie in Etruria meridionale. In: Ricerche e scavi. Atti dell'undicesimo incontro di studi centro studi di prehistoria e archeologia; Milano, 2014, 653-666.
- Goldenberg, 2004 Goldenberg G. Ein Verhüttungsplatz der mittleren Bronzezeit bei Jochberg (Nordtirol). In: Alpenkupfer Rame delle Alpi. Der Anschnitt. Beiheft 17; Deutsche Bergbaumuseum: Bochum, Germany, *122*, 2004, 165-176.
- Goldenberg, Rieser, 2004 Goldenberg G., Rieser B. Die Fahlerzlagerstätten von Schwaz/Brixlegg (Nordtirol). In: Alpenkupfer Rame delle Alpi. Der Anschnitt. Beiheft 17; Deutsche Bergbaumuseum: Bochum, Germany, 122, 2004, 37-52.
- Grigoriev, 2015 Grigoriev S. *Metallurgical Production in Northern Eurasia in the Bronze Age;* Archaeopress Publishing Ltd: Oxford, United Kingdom, 2015.
- Grigoriev, 2017 Grigoriev S. Social processes in Ancient Eurasia and development of types of alloys in metallurgical production. In: Archaeoastronomy and Ancient Technologies 2017, 5 (2), 17-44.
- Grigoriev, 2018 Grigoriev S.A. Problema khronologii sintashtinskoy kul'tury. [Problems of chronology of Sintashta culture]. In: Stepnaya Evraziya v epokhu bronzy: kul'tury, idei, tekhnologii. [Steppe Eurasia in the Bronze Age: cultures, ideas, technologies]; Chelyabinsk Universitet, Chelyabinsk, Russia, 2018, 37-61.
- Hauptmann, 2008 Hauptmann A. Erzlagerstätten in östlichen Mittelmeerraum. In: Anatolian Metal IV. Der Anschnitt, Beiheft 21; Deutsche Bergbaumuseum: Bochum, Germany, 157, 2008, 55-66.
- Hauptmann, 2009 Hauptmann A. Lead isotope analysis and the origin of Sardinian metal objects. In: Oxide ingots in the Central Mediterranean; Roma: Instituto di studi sulle civilta dell'egeo e del vicino oriente, 2009, 449-514.

- Herdits, Löcher, 2004 Herdits H., Löcher K. Eine bronzezeitliche Kupferhütte Im Mitterberger Kupferkies-Revier (Salzburg). In: Alpenkupfer Rame delle Alpi. Der Anschnitt. Beiheft 17; Deutsche Bergbaumuseum: Bochum, Germany, 122, 2004, 177-188.
- Hohlmann, Hauptmann, Schröder, 2004 Hohlmann B., Hauptmann A., Schröder B. Der Spätbronzezeitliche Verhüttungsplatz von Acqua Fredda am Passo Redebus (Trentino). Rohstoffbasis, metallurgische Untersuchungen und Versuch einer Prozessrekonstruktion. In: Alpenkupfer – Rame delle Alpi. Der Anschnitt. Beiheft 17; Deutsche Bergbaumuseum: Bochum, Germany, 122, 2004, 261-267.
- Höppner u. a., 2005 Höppner B., Bartelheim M., Huijsmans M., Krauss R., Martinek K.-P., Pernicka E., Schwab R. Prehistoric copper production in the Inn valley (Austria), and the earliest copper in Central Europe. Archaeometry 2005, 47 (2), 293-315.
- Huijsmanns, Krauβ, Stibich, 2004 Huijsmanns M., Krauβ R., Stibich R. Prähistorischer Fahlerzbergbau in der Grauweckenzone. Neolithische und bronzezeitliche Besiedlungsgeschichte und Kupfermetallurgie im Raum Brixlegg (Nordtirol). In: Alpenkupfer – Rame delle Alpi. Der Anschnitt. Beiheft 17; Deutsche Bergbaumuseum: Bochum, Germany, 122, 2004, 53-62.
- Ixer, 2001 Ixer R. A. An assessment of copper mineralization from the Great Orme Mine, Llandudno, North Wales, as ore in the Bronze Age. Proceedings of the Yorkshire Geological Society 2001, 53, 213-219.
- Junghans, Sangmeister, Schröder, 1960 Junghans S., Sangmeister E., Schröder M. Metallanalysen kupferzeitlicher und frühbronzezeitlicher Bodenfunde aus Europa; Mann: Berlin, Germany, 1960.
- Kassianidou, 2009 Kassianidou V. Oxide ingots in Cyprus. In: Oxide ingots in the Central Mediterranean; Instituto di studi sulle civilta dell'egeo e del vicino oriente: Roma, Italy, 2009, 41-81.
- Kienlin, 2008 Kienlin T.L. Frühes Metall im nordalpinen Raum. Eine Untersuchung zu technologischen und kognitiven Aspekten früher Metallurgie anhand der Gefüge frühbronzezeitlicher Beilen. Vol. I. Rudolf Habelt Verlag: Bonn, Germany, 2008.
- Kienlin, Stöllner, 2009 Kienlin T.L., Stöllner Th. Singen copper, Alpine settlements and Early Bronze Age mining: Is there a need for elites and strongholds? In: Metals and societies. Studies in honour of Barbara S. Ottaway; Rudolf Habelt Verlag: Bonn, Germany, 2009, 67-104.
- Klemm, 2004 Klemm S. Der prähistorische Kupferschmelzplatz S1 in der Eisenerzer Ramsau (Steiermark). In: Alpenkupfer – Rame delle Alpi. Der Anschnitt. Beiheft 17; Deutsche Bergbaumuseum: Bochum, Germany, 122, 2004, 189-198.
- Knapp, Kassianidou, 2008 Knapp A.B., Kassianidou V. The archaeology of Late Bronze Age copper production. Politico Phorades on Cyprus. In: Anatolian Metal IV. Der Anschnitt, Beiheft 21; Deutsche Bergbaumuseum: Bochum, Germany, 2008, 157, 135-147.
- Krause, 1989 Krause R. Early tin and copper metallurgy in South-western Germany at the beginning of the Early Bronze Age. In: Old World Archaeometallurgy. Anschnitt. Beiheft 7; Deutsche Bergbaumuseum: Bochum, Germany, 1989, 44, 25-32.

- Krause, 2003 Krause R. Studien zur kupfer- und frühbronzezeitlichen Metallurgie zwischen Karpatenbecken und Ostsee; Marie Leidorf Verlag: Rahden/Westf., Germany, 2003.
- Krause, 2009 Krause R. Bronze Age copper production in the Alps: organization and social hierarchies in mining communities. In: Metals and societies. Studies in honour of Barbara S. Ottaway; R. Habelt: Bonn, Germany, 2009, 47-66.
- Kristiansen, Larsson, 2005 Kristiansen K., Larsson T.B. The rise of Bronze Age society; University Press: Cambridge, United Kingdom, 2005.
- Lo Schiavo, 2009 Lo Schiavo F. Oxide ingots hoarding and offering in Nuragic Sardinia. In: Oxide ingots in the Central Mediterranean; Instituto di studi sulle civilta dell'egeo e del vicino oriente: Roma, Italy, 2009, 391-407.
- Lo Schiavo, Albanese Procelli, Giumlia-Mrair, 2009 Lo Schiavo F., Albanese Procelli R.M., Giumlia-Mrair A. Oxide ingots in Sicily. In: Oxide ingots in the Central Mediterranean; Instituto di studi sulle civilta dell'egeo e del vicino oriente: Roma, Italy, 2009, 135-221.
- Lutz, Pernicka, 2013 Lutz J., Pernicka E. Prehistoric copper from the Eastern Alps. Archaeometry 2013, 1(1)e25, 122-127.
- Maas, 2004 Maas A. Zur chalkolithisch-frühbronzezeitlichen Kupferproduktion von St. Véran / Hautes Alpes. In: Alpenkupfer Rame delle Alpi. Der Anschnitt. Beiheft 17; Deutsche Bergbaumuseum: Bochum, Germany, 122, 2004, 117-119.
- Martinek, Sydow, 2004 Martinek K.-P., Sydow W. Frühbronzezeitliche Kupfermetallurgie im Unterinntal (Nordtirol). In: Alpenkupfer Rame delle Alpi. Der Anschnitt. Beiheft 17; Deutsche Bergbaumuseum: Bochum, Germany, 122, 2004, 199-211.
- McKerrell, Tylecote, 1972 McKerrell H., Tylecote R.F. The working of copper-arsenic alloys in the Early Bronze Age and the effect on the determination of provenance. Proceedings of the Prehistoric Society 1972, 38, 209-218.
- Mille, Carozza, 2009 Mille B., Carozza L. Moving into the Metal Ages: the social importans of metal at the end of the Neolithic period in France. In: Metals and societies. Studies in honour of Barbara S. Ottaway; R. Habelt: Bonn, Germany, 2009, 143-171.
- Moesta, Rüffler, Schnau-Roth, 1989 Moesta H., Rüffler R., Schnau-Roth G. Zur Verfahrenstechnik der bronzezeitlichen Kupferhütten am Mittelberg. In: Old World Archaeometallurgy. Anschnitt. Beiheft 7; Deutsche Bergbaumuseum: Bochum, Germany, 1989, 44, 141-153.
- Montero-Ruiz, de la Esperanza, 2004 Montero-Ruiz I., de la Esperanza M.J.R. Der prähistorische Kupferbergbau in Spanien. In: Anschnitt. Heft 2–3; Deutsche Bergbaumuseum: Bochum, Germany, 56, 2004, 54-63.
- Moosleitner, 2004 Moosleitner F. Bronzezeitliche Grubenöfen in St. Johann im Pongau (Salzburg). In: Alpenkupfer Rame delle Alpi. Der Anschnitt. Beiheft 17; Deutsche Bergbaumuseum: Bochum, Germany, 122, 2004, 213-221.
- Muhly, 1973 Muhly J.D. Copper and tin. The distribution of mineral resources and the nature of the metals trade in the Bronze Age; Archon Books: Hamden, 1973.

- Muhly, 2005 Muhly J.D. Cyprus and copper for the world. In: Anatolian Metal III. Abschnitt, Beiheft 18; Deutsche Bergbaumuseum: Bochum, Germany, 2005. S. 137-141.
- Muhly, 2008 Muhly J.D. Metal deposits in the Aegean Region. In: Anatolian Metal IV. Der Anschnitt, Beiheft 21; Deutsche Bergbaumuseum: Bochum, Germany, 2008, 157, 67-75.
- Naef, 2014 Naef L. Stierva Tiragn eine spätbronzezeitliche Schlackenhalde im Oberhalbstein (Graubunden, Schweiz). In: Anschnitt. Heft 2–3; Deutsche Bergbaumuseum: Bochum, Germany, 2014, 66, 78-88.
- Needham, 2004 Needham S. Migdale-Marnoch: sundurst of Scotland metallurgy. In: Scotland in Ancient Europe; Society of Antiquaries of Scotland: Edinburgh, 2004, 217-245.
- Northover, 1989 Northover J.P. Properties and use of arsenic-copper alloys. In Old World Archaeometallurgy. Anschnitt. Beiheft 7; Deutsche Bergbaumuseum: Bochum, Germany, 1989, 44, 111-118.
- O'Brien, 1994 O'Brien W. Mount Gabriel: Bronze Age mining in Ireland (Bronze Age studies); National University of Ireland: Galway, Ireland, 1994.
- O'Brien, 2004 O'Brien W. Ross Island. Mining, Metal and Society in Early Ireland; National University of Ireland: Galway, Ireland, 2004.
- O'Brien, 2011 O'Brien W. Prehistoric copper mining and metallurgical expertise in Ireland. In: Povoamento e Exploração dos Recursos Mineiros; Citcem: Braga, Portugal, 337-357.
- O'Brien, 2013 O'Brien, W. Bronze Age copper mining in Europe. In: Oxford Handbook of the Bronze Age; University Press: Oxford, United Kingdom, 2013, 433-449.
- Parker Pearson, 2009 Parker Pearson M. The Earlier Bronze Age. In: The Archaeology of Britain. An Introduction from Earliest time to the twenty-first century; Routledge: London– New York, United Kingdom–USA, 2009, 103-125.
- Pearce, 2009 Pearce M. How much metal was there in circulation in Copper Age Italy? In: Metals and societies. Studies in honour of Barbara S. Ottaway; R. Habelt: Bonn, Germany, 2009, 277-284.
- Pernicka et al., 1993 Pernicka E., Begemann F., Schmitt-Strecker S., Wagner G.A. Eneolithic and Bronze Age copper artefacts from the Balkans and their relations to Serbian copper ores. Praehistorische Zeitschrift 1993, 68 (1), 1-54.
- Pernicka, 1987 Pernicka E. Erzlagerstätten in Ägäis und ihre Ausbeutung im Altertum. Geochemische Untersuchungen zur Herkunftsbestimmung archäologischer Metallobjekte. In: Jahrbuch des Romisch-Germanischen Zentralmuseums; RGZM: Mainz, Germany, 1987, 34, 607-714.
- Pernicka, Anthony, 2010 Pernicka E., Anthony D.W. The Invention of Copper Metallurgy and the Copper Age of Old Europe. In: *The Lost World of Old Europe. The Danube Valley*, 5000–3500 bc; University Press: Princeton, USA, 2010, 162-177.
- Pernicka, Lutz, 2015 Pernicka E., Lutz J. Fahlerz- und Kupferkiesnutzung in der Bronze- und Eisenzeit. In: Bergauf Bergab – 10.000 Jahre Bergbau in den Ostalpen. Deutsche Bergbaumuseum: Bochum, Germany, 2015, 207, 107-111.

- Pittioni, 1954 Pittioni R. Urgeschichte der Österreichischen Raum; Franz Deuticke: Wien, Austria, 1954.
- Presslingen, Eibner, 2004 Presslingen H., Eibner C. Montanarchäologie im Paltental (Steiermark). Bergbau, Verhüttung, Verarbeitung und Siedlungstätigkeit in der Bronzezeit. In: Alpenkupfer – Rame delle Alpi. Der Anschnitt. Beiheft 17; Deutsche Bergbaumuseum: Bochum, Germany, 122, 2004, 63-74.
- Preβlingen, Eibner, 1989 Preβlingen H., Eibner C. Bronzezeitliche Kupferverhüttung im Paltental. In: Old World Archaeometallurgy. Anschnitt. Beiheft 7; Deutsche Bergbaumuseum: Bochum, Germany, 1989, 44, 235-240.
- Primas, 1997 Primas M. Bronze Age economy and ideology: central Europe in focus. In: Journal of European archaeology 1997, 5, 1-37.
- Primas, 2008 Primas M. Bronzezeit zwischen Elbe und Po. Strukturwandel in Zentraleuropa 2200–800 v. Chr.; Rudolf Habelt Verlag: Bonn, Germany, 2008.
- Radivojević et al., 2010 Radivojević M., Rehren Th., Pernicka E., Šljivar D., Brauns M., Borić D. On the origins of extractive metallurgy: new evidence from Europe. In: Journal of Archaeological Science 2010, 37, 2775-2787.
- Radivojević et al., 2014 Radivojević M., Rehren Th., Kuzmanović-Cvetković J., Jovanović M., Northover J.P. Why are there tin bronzes in the 5th mill BC Balkans? In: Archaeotechnology: Studying Technology from Prehistory to the Middle Ages; Prosveta: Belgrade, Serbia, 2014, 235-256.
- Radivojević et al., 2018 Radivojević M., Roberts B., Pernicka E., Stos-Gale Z., Martinon-Torres M., Rehren T., Bray, P., Brandherm D., Ling J., Mei J., Vandkilde H., Kristiansen K., Shennan S.J., Broodbank C. The Provenance, Use and Circulation of Metals in the European Bronze Age: The state of debate. In: Journal of Archaeological Research 2018, 27(2), 1-55.
- Roberts, 2009 Roberts B. Origins, transmission and traditions: analyzing early metal in Western Europe. In: Metals and societies. Studies in honour of Barbara S. Ottaway; R. Habelt: Bonn, Germany, 2009, 129-142.
- Rowlands, 1976 Rowlands M.J. The production and distribution of metalwork in the Middle Bronze Age in Southern Britain; British archaeological reports: Oxford, United Kingdom, 1976.
- Ryndina, Ravich, 2012 Ryndina N.V.; Ravich I.G. O metalloproizvodstve maykopskikh plemen Severnogo Kavkaza (po dannym khimiko-tekhnologicheskikh issledovaniy) [About metal production of Maikop tribes of the Northern Caucasus]. In: Vestnik arkheologii, antropologii i etnografii [Bulletin of archaeology, anthropology and ethnology] 2012, 2 (17), 4-20.
- Ryndina, Ravich, Bystrov, 2008 Ryndina N.V.; Ravich I.G.; Bystrov S.V. O proiskhozhdenii i svoystvakh mysh'yako-nikelevykh bronz maykopskoy kul'tury Severnogo Kavkaza (ranniy bronzovyy vek) [About origins and properties of arsenic-nickel bronzes of Maikop culture of the Northern Caucasus (Early Bronze Age)]. In: Arkheologiya Kavkaza i Blizhnego Vostoka [Archaeology of the Caucasus and the Near East]; Taus: Moscow, Russia, 2008, 196-221.

- Sabatini, 2015 Sabatini B.J. The As–Cu–Ni System: A Chemical Thermodynamic Model for Ancient Recycling. In: JOM: the journal of the Minerals, Metals & Materials Society 2015, 67, 2984-2992.
- Scott, 1991 Scott D.A. Metallography and microstructure of ancient and historic metals; The J. Paul Getty Trust: Los Angeles, USA, 1991.
- Sperber, 2004 Sperber L. Zur Bedeutung des nördlichen Alpenraumes für die spätbronzezeitliche Kupferversorgung in Mitteleuropa mit besonderer Berücksichtigung Nordtirols. In: Alpenkupfer – Rame delle Alpi. Der Anschnitt. Beiheft 17; Deutsche Bergbaumuseum: Bochum, Germany, 122, 2004, 303-345.
- Stöllner et al., 2016 Stöllner T., von Rüden C., Hanning E., Lutz J., Kluwe S. The enmeshment of Eastern Alpine mining communities in the Bronze Age. From economic networks to communities of practice. In: From bright ores to shiny metals. Anschnitt. Beiheft 29; Deutsche Bergbaumuseum: Bochum, Germany, 2016, S. 75-107.
- Strahm, Hauptmann, 2009 Strahm Ch., Hauptmann A. The metallurgical developmental phases in the Old World. In: Metals and societies. Studies in honour of Barbara S. Ottaway; R. Habelt: Bonn, Germany, 2009, 116-128.
- Töchter u. a., 2013 Töchter U., Goldenberg G., Schneider Ph., Tropper P. Spätbronzezeitliche Verhüttungsdüsen aus dem Bergbaurevier Mauken in Unterinntal. Nordtirol: Typologie, mineralogisch-petrographische Zusammensetzung und experimentelle Rekonstruktionsversuche. In: Anschnitt, 65, Heft 1–2, 2013, 2-19.
- Tylecote, 1976 Tylecote R.F. A history of metallurgy; The Metal Society: London, United Kingdom, 1976.
- Vandkilde, 2010/2011 Vandkilde H. Cultural Perspectives on the Beginnings of the Nordic Bronze Age. In: Offa. Berichte und Mitteilungen zur Urgeschichte, Frühgeschichte und Mittelalterarchäologie 2010/11, Band 67/68, 51-78.
- Vandkilde, 2016 Vandkilde, H. Bronzization: The Bronze Age as Pre-Modern Globalization. Praehistorische Zeitschrift 2016, 91(1), 103-123.
- Weisgeber, 2004 Weisgeber G. Schmelzanlagen früher Kupfergewinnung ein Blick über die Alpen. In: Alpenkupfer – Rame delle Alpi. Der Anschnitt. Beiheft 17; Deutsche Bergbaumuseum: Bochum, Germany, 122, 2004, 15-36.
- Wyss, 2004 Wyss R. Prähistorischer Kupferbergbau in den Schweizer Alpen II. In: Alpenkupfer – Rame delle Alpi. Der Anschnitt. Beiheft 17; Deutsche Bergbaumuseum: Bochum, Germany, 122, 2004, 113-119.
- Zwicker, 1989 Zwicker U. Untersuchungen zur Herstellung von Kupfer und Kupferlegierungen im Bereich des östlichen Mittelmeeres. In: Old World Archaeometallurgy. Anschnitt. Beiheft 7; Deutsche Bergbaumuseum: Bochum, Germany, 1989, 44, 191-201.

© This article is an open access article distributed under the terms and conditions of the Creative Commons by Attribution (CC-BY) license (http://creativecommons.org/licenses/by/4.0/).