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PALAEOMETALLURGICAL INVESTIGATIONS IN THE REGION BETWEEN THE RIVERS MOSEL AND WERRA (BUNDESREPUBLIK DEUTSCHLAND)

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Hoards of metal objects are very often the main source of information about the Bronze Age and especially about the period of the Urnfield Culture. The evaluation of these informations is performed using the traditional archaeological methods. For example the collection and presentation of big amounts of depot - findings in Southern Germany has the intention, to find out cultural or religious reasons for their concealing. An evaluation under economic historical point of view is known only in few cases up today (Rowlands, 1976). A special significance in this field has the research of the structure and organisation of production of these metallic materials. As pointed out in a detailed manner for settlements (Jockenhövel, 1985), the production was based on a very uniform scheme. It was possible to distinguish the following five steps:

1. Preparation of the ores
2. Smelting and casting
3. Manufacturing of goods using various techniques
4. Period of use
5. Recycling of metal.

A basic condition for the production was a sufficient supply of raw material

for smelting copper and its alloys (e.g. tin, arsenic, antimony and lead). Many regions, because of lack of those ores were obliged, to import them. In this way the beginning of the intentional production of copper alloys (arsenic - copper) created an interregional network for the exchange of the raw materials. All regions of Europe were integrated into this system. The long period of the Bronze Age in Europe (more than thousand years) demonstrates the stability of this network of economic and communicative connections. It was even able to overcome some serious difficulties at the end of the Bronze Age. Slowly the importance of this network decreased as the use of the bronze was replaced by that of iron. Iron ores were available almost everywhere.

The search for copper and tin mines is therefore of especial importance. In this field Austrian scientists were successful, discovering in the Alpine Region traces of mining from the Bronze Age and especially from the Urnfield Culture. These traces have been partially visible to the present days (Zschocke/Preuschen, 1932). Some surprising details of methods and organisation of prehistoric mining could be discovered in this way (Eibner, 1983). The "Wiener Arbeitsgruppe" (R.Pittioni and H.Neuninger) succeeded in making a further step investigating the relations among mines and manufactured products. By means of semiquantitative analysis (atomic - emission spectroscopy AES) they established "impurities patterns" for different mines. It was possible to distinguish several regions of mines. Their investigations were simplified by the fact, that samples of ores and slags could be obtained from mines that have been in operation from the Bronze Age up to the present days. This type of work is impossible for other traditional copper producing regions of Europe such as Mansfeld/Thuringia or Slovakia because the intensive and extensive mining in these regions during the later Middle Ages and in modern times (as described by G.Agricola) has destroyed all traces of prehistoric work. Therefore W.Witter and H.Otto in Halle/Germany tried by means of fully quantitative analysis (atomic emission spectroscopy) of manufactured materials and rawcastings to establish relations to the corresponding mines. The investigations concerned essentially the region of Middle Germany with its rich mines and copperslates. Witter and Otto could demonstrate that these mines delivered ores for coppersmelting since the late Neolithic (1952). The investigation of the propagation and development of metallurgy was the aim of the biggest project in the field of palaeometallurgical analysis. The title of this project of the "Stuttgart Group" was

"Studien zu den Anfängen der Metallurgie" (SAM). There was no attempt to find relations among manufactured materials and mines. The main subject of this work was the Copper and Early Bronze Age (Junghans, Sangmeister and Schröder 1960,1968,1974).

The critical discussion of the statistically formed material - groups, presented by the SAM scientists, is well known. A review over the situation shows a resignation concerning the answer of the question of supply of raw - materials during the Bronze Age.

According to that at least in Germany, one can see serious doubts of the value of the analysis of prehistorical metallic material (H.Härke, 1978, pp 165). Nevertheless in the neighbouring countries new research programms have been started in this field under the title "Palaeometallurgy". The French palaeometallurgy studies (Paléometallurgie, 1984) is a splendid example of this type of work. The aim of these investigations is the determination of the composition of the metallic findings, by means of using different analytical methods. Further there is great hope that this information will give them the opportunity to get a diachronic view of the variation of the concentrations of the different elements. This could be a source of informaton about the development of metallurgy, economy and culture during this period. Taking into account the investigations "Relations between copper-metal and original ore" (H.D.Schulz, 1983) and "Alloy or seam ore?" (Eibner, 1976) a research program was developed in Frankfurt with aim the study of the relations among seams of ore and manufactured products using more modern analytical methods. This work is restricted to a relatively small region and a limited period of time. The link between ore and manufactured product is the "primary metallurgy",that means the smelting of the raw-material and manufacturing of the final product. Information can be obtained analysing slags, raw castings, waste of casting and ingots. If the impurity pattern of these materials were known, it should be easier to look for related seams of copper-ores and correlations to manufactured products of this region. The region of interest is the region of the German Central Chain of mountains between the Rivers Mosel and Werra and the period chosen the late Bronze Age.

In the region of interest (betweenR.Moseland R.Werra)there are many seams of copper-ores related to different geological formations, that have not been mapped or described in review articles. All exploitable mines have been partially in operation to the present days. The most important districts are:

the middle and lower Hessen formations of Zechstein with the layer of slatelike copper-ore (Jockenhövel, 1983), the Donnersberg mountain in Rheinland-Pfalz (Geis, 1955) and the district of Wallerfangen in Saarland (Schindler, 1968). Beside these "classical districts" there is a big number of small seams of ore, that most probably have been exploited in prehistoric periods (Köbrich, 1936).

Concerning the period of the investigation many depot-findings are known from the period of the end of the later Bronze Age(Ha B3 to H. Müller-Karpe; 8^{th} century b.c.). They are considerably large and contain a high percentage of manufactured products, that are characteristic for this region (Jockenhövel, 1981). Findings of casting moulds prove the existing of a local metallurgy in this region. This is also confirmed by raw-castings or parts of raw-castings in the depots. In some cases depots pieces of resmolten metal were found. These findings may provide informations about recycling procedures.

The region of our investigation is part of the circle of the "bronzes of lake-dwellings". This is the name for bronze materials of similar types from the latest period of Bronze Age, originating from the region of Switzerland, Southwest of Germany and the Eastern France. Regardless of local variations, (e.g. "groupe lorrain" at Saar and Mosel) the province of origin is of extreme importance. Its strong influence on the North (during period V to O.Montelius) can be seen from the rich flux of metal from the Upper - Rhine through the territories of Hessen and Thuringia to the plains of Northern Germany and finally to Scandinavia (Sprockhoff, 1956; Thrane, 1975). Therefore it is very probable, that copper from the region of our interest had reached the North either as raw-copper or as manufactured products. Starting from the point of view, that the region of our interest belongstoa greater provinceof culture in the later Bronze Age possessing a lot of copper ore, we can assume that these ores had been used for the production of raw-copper etc.. Therefore the principal aims of our program are:

1. Search of sources of raw-material for smelting copper during the end of Bronze Age.
2. Circulation of metals.
3. Evaluation of the analytical data in order to establish relations among seams of ore and manufactured products.

There are no similar investigations done until now. In this way we intend to analyse only objects of metallurgical interest and the choice is limited by

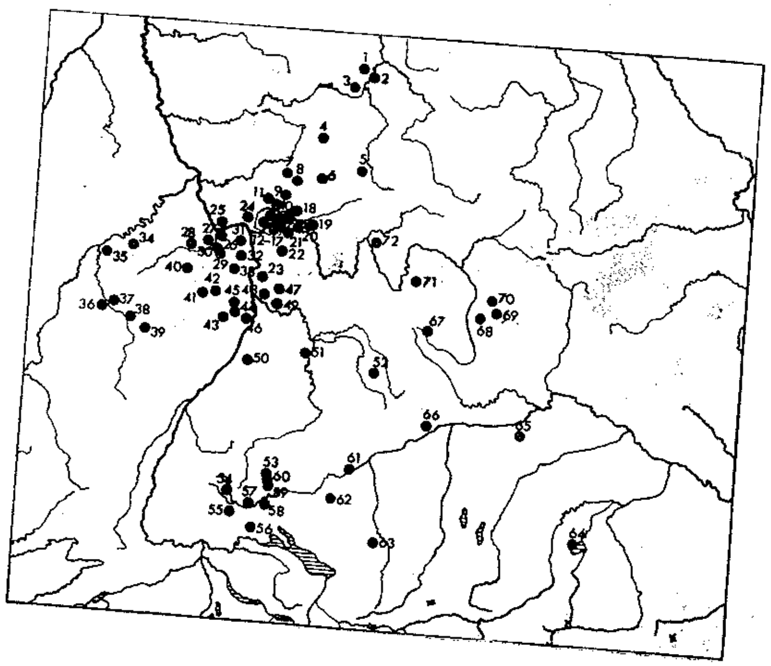

Fig. 1 Distribution of depot - finds from the late Bronze Age (Ha B3) in Southern Germany (A. Jockenhövel, 1975, p.55, fig.17).

a relatively short period and a restricted region. We hope, that the analysis of raw - castings will give information about the status of fabrication of fresh raw - copper at the end of the Bronze Age and about the amount of scrap-bronze recycled. It is often said, that the cycle of metals during the Bronze Age was in an increasing degree replaced by scrap and, at the end of the period only a small amount of fresh unalloyed copper was used.
Nevertheless we should notice, that big amount of metal was removed from the cycle because of religious reasons (depots, graves etc) had replaced.
A first result of our investigations is the demonstration, that at the end of the Bronze Age still considerable amounts of fresh raw - copper firstly entered the cycle of metal. These raw - castings that have been broken into pieces are constituent parts of depot - findings. On the contrary the alloyed raw - castings are intact and small. They have a spherical, oval or flat shape and some of them consist of several layers arising from repeated casting. A piece from the late period of the Urnfield Culture found in a depot - finding at Bad Homburg (Herrmann, 1966, fig. 186,16) has even the form of a molten thin

plate (see also for bronzecraps the remark of A. Mozsolics, 1981).
The copper mining in the region of interest and its surroundings is documented only since 1000 A.D.. No extended documentation can be found for the periods earlier than the 15th/16thcentury, especially for the region of Hessen and Thuringia. Traces of prehistorical mining such as patterns of working, tools places of smelting together with slags could not be detected until now. Therefore it was necessary to restrict to assumptions. The degree of probability of these was related to archeological history of colonization, the abundance and concentration and the character of the findings. Informations had been obtained in many cases by analysis of metallic objects. Those observations led to the well established assumption that in Eastern Thuringia existed small minings and well known fabrication workshops since the end of middle Bronze Age (K.Simon, 1982). As a result of studying the concentration of hoard - findings around fortifications from the Bronze Age, O.Kytlicová also assumed mining in the region of Příbram during the late period of Bronze Age (1982).
In the western parts of the region of our investigation it was succeeded to prove the existance of mining places originating from the Roman period. These traces of galleries were found there. Authenticated by inscriptions are the Marcus - gallery near Kordel - Butzweiler (south - western part of Eifel) and the Emilianus - gallery at Wallerfangen (Schindler, 1968), Saar district. In bothplaces copper ores had been probably found in quarries of sand - stone. Only the front part of "Pützlöcher" at Kordel - Butzweiler (fig.2) is antique. Beside the entry of the gallery there were two inscriptions with the name Marci (today only one of them exists). This name can also be found on several squared stones of the "Porta Nigra" in Treves. The Roman gallery has a length of 15m and branches out. From there six vertical excavations rise upward to the surface on proceed downwards. Besides the inscriptions Roman fragments of pottery and tile are suitable for dating. Later during the 18thcentury the antique gallery was enlarged.
A Roman inscription (CIL XIII,4238: INCEPTA OFFICINA EMILIA III X NONIS MART) lead to the investigation of the Emilianus - gallery of St.Barbara near Wallerfangen, Saar district, by R. Schindler from 1964 till 1965. This gallery is a two floor facility. The purpose of the lower gallery, which is connected with the upper one, seems to be the exploration. Layers of copper - ores were found in the upper gallery. The end of the gallery has not been reached yet. Lateral branches pits to the surface and a drainage are remarkable. Many small findings of the Roman period as well as residues of tools allow no doubt to

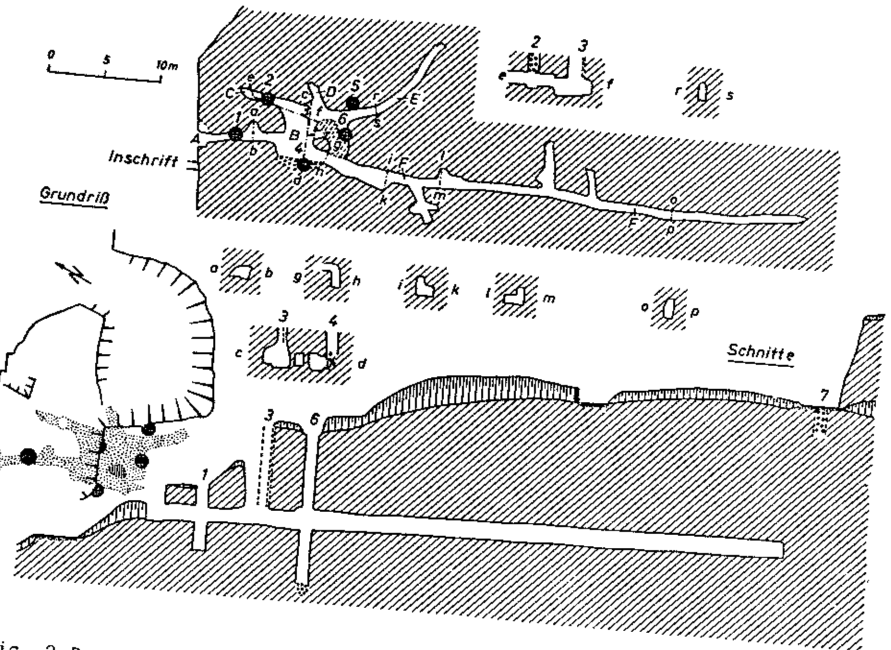

Fig. 2 Roman gallery for mining copper-ore "Pützlöcher" at Kordel-Butzweiler (R.Schindler, 1968, p.34, fig.6).

the Roman origin of that sophisticated mine.

The third place, where Roman copper - ores mining could be detected is the part of the Rhenish Palatinate, in the neighbourhood of the Donnersberg - mountain. This region consists of porphyry. On the top of the mountain a celtic oppidum was situated.

Furthermore Roman minings are supposed to be located at Dannenfels, Göllheim and Imsbach (Walling, 1977).

Surely the Roman mining with its sophisticated engeneering can not be compared with prehistorical local methods. An important remark is that the Roman mines were located in places with seams of ores existed on the surface or in the near surface layers. The prospector of the prehistorical period was also able to find such places. Appearing ores, changes in the colour of the surface, special vegetation or an earlier melting of the snow on the surface layers containing ores, enabled him to discover seams and to exploit them by his own method.

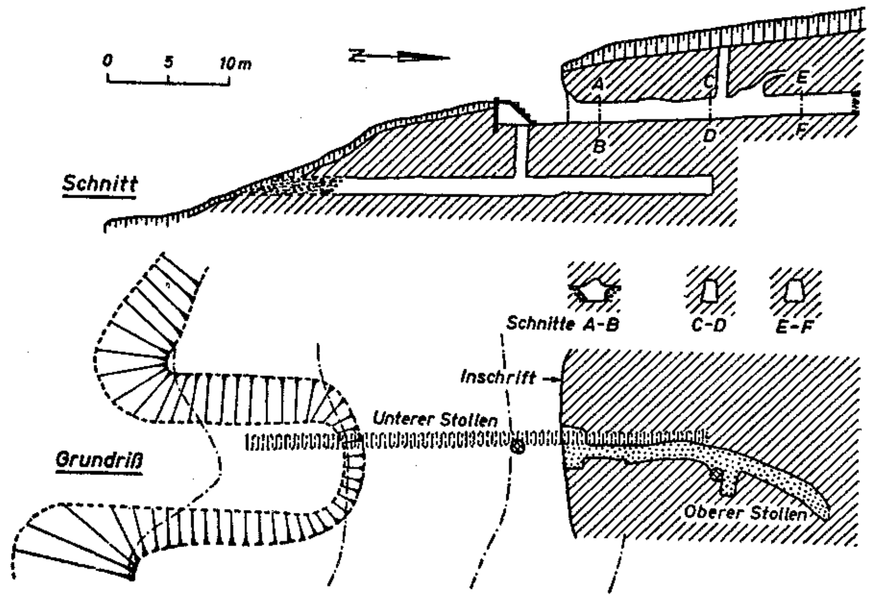

Fig. 3 Roman mine "Emilianus" at Wallerfangen, Saar district (R. Schindler, 1968, p.30, fig.4)

Although to the present days no traces pf prehistorical mining could be detected in our region, the investigation of R.Schindler (1968) and H.Conrad (1968) demonstrates with considerable probability, that Azurit from Wallerfangen had been smelted for copper already at the end of Bronze Age. R.Schindler as first interpretated from the point of view of the economic history and social aspects the depot - findings from the period of Urnfield Culture, graves from the Hallstatt - period and a fortification on the "Limberg" from the same period. The utilization of this place of refuge enabled the arising of the "dynysty of Wallerfangen". He connected that unusual concentration in one place and its internal structure causally with existance of copper mine in the neighbourhood.

H.Conrad made use of analysis of crumbs of ore from Wallerfangen and of a tintinnabulum from the period Ha B3 in order to prove the local route from the mine to the manufactured products. As significant elements were considered the concentrations of cobalt and nickel in the ore and the products.

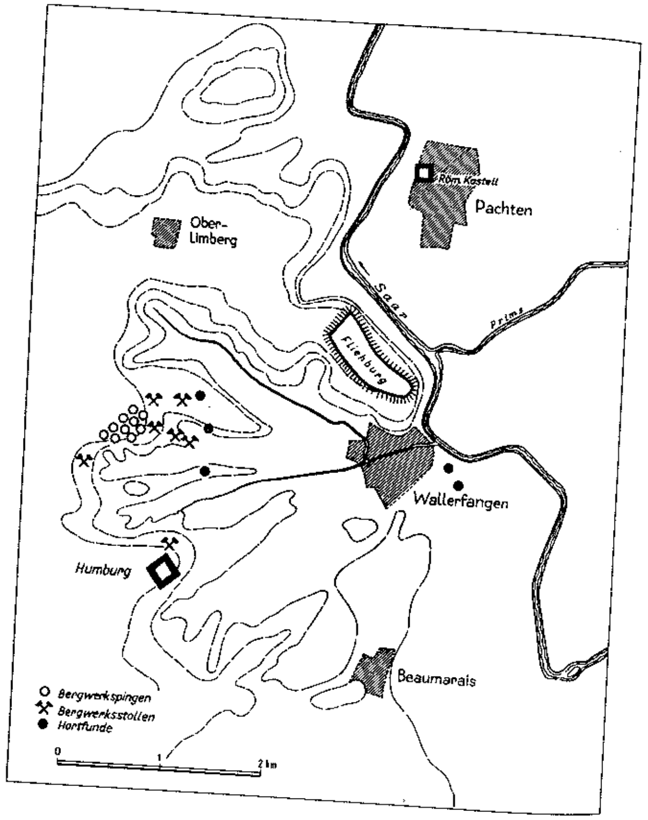

Fig. 4 Topography of the Wallerfangen - region, Saar district (R.Schindler, 1966, p.162)

In the surroundings of the Roman gallery at Kordel - Butzweiler, in a distance of 6 km western direction near Welschbillig - "Kunkelborn" a settlement from the period of Urnfield Culture has been discovered some time ago (fig. 5). (H.Löhr, private communication). Residues of metal manufacturing were found among other findings in this location. Of special importance is an ingot of raw - copper. In this case it is also necessary to perform comparative analysis of ore and casting.

From the region of the "Donnersberg" and its neighbourhood only few archeological findings are known. Nevertheless on the top of the mountain there is an asupmtion of facility from the period of Urnfield Culture. This shows

certain similarities to others in Southern Germany, that had been created by the metallurgical industry.

Taking into account the present results, it is possible to assume a local exploiting of seams of copper - ores in the region of our investigations during the Bronze Age. It is not possible for the moment to decide, where is the best place to start promising mining - archeological work. This may be possible after the analytical investigations of the raw - copper findings (especially raw - castings). The first task is to recognize, how many species of copper existed in the region. We can try with the knowledge of the "finger - prints" (patterns of the concentrations of different elements) to excavate residues of pings, places of smelting or heaps of slags. On the other hand it should be possible to recognize the way of the raw - copper to its alloyed end - products. H.D.Schulz (1983) demonstrated by analysis of copper from Helgoland that this is not obstructed by changed conditions of the smelting process. The success depends on the analytical method. Therefore we have chosen for our work the instrumental Neutron Activation Analysis (INNA).

Instrumental Neutron Activation Analysis (INNA) - analysis without chemical separation after irradiation - is an excellent tool for the investigation of copper and bronze.

Most of the components, to be determine, produce during neutronirradiation activities with much longer half - lives than that of the matrix. Therefore it is possible to wait for the decay of matrix activities before the actual measurement of the sampler. In this way the interference of backgound - radiation can be avoided. The isotopes, of tin either have only small neutroncapture crossections or the - ray emission probability of the resulting radioisotopes is small. That means, that a relative large content of tin in the samples does not reduce the effectivity of the measurement. Regardless of the common opinion INNA is not a non - destructive method, but only small amount of sample (10mg) is required for the analysis. Until now 60 objects meanly raw - castings and other semimanufactured material have been analysed.

From each object two analysis were made. In the first one, after two hours of irradiation, the short living radioactive Isotopes lives of the elements Indium (In), Manganese (Mn) and Arsenic (As) were determined. This analysis also supplied the first information about the content of Antimony (Sb) in the sample. The knowledge of this is important for the calculation of the time required for the second irradiation lasts normally three days. This time must

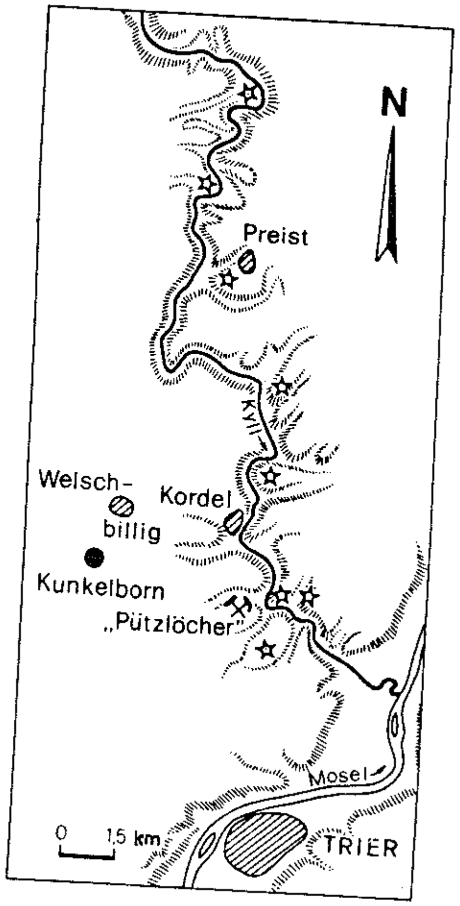

Fig. 5 Prehistorical fortifications at the lower part of the river Kyll together with the Roman copper mine "Pützlöcher" and the settlement from the period of Urnfield Culture "Kunkelborn" (Welschbillig) (stars: hillforts, mainly Iron Age)

be reduced if the content of Antimony is higher than one percent. Other ways, the activity of the irradiated sample would be very high and the sample cannot be handled without risk. The reduction of irradiation - time can in some cases be compensated by longer counting.
The measurement cannot begin before seven days after the end of irradiation, because the high activity of the matrix (owing to Cu-6^4) must decay at first.

In that second analysis the radioactive isotopes of the elements antimony (Sb), tin (Sn), iron (Fe), cobalt (Co), nickel (Ni), silver (Ag), gold (Au) and occasionally selenium (Se), chromium (Cr), rhenium (Re) and others are measured and the concentration of these elements in the sample is determined. The possible sensivity and accuracy of the determination of the elements depends very much on the content of antimony in the sample. That limits not only the irradiation - time, but also reduces the sensivity of the determination of As, Sn, Au, Cr and Re. The greatest sensivity is normally achieved for gold (10^{-6}%). Considering usual concentrations, the highest accuracy can be achieved for cobalt (±3%). The largest error arises in the determination of iron (up to 30% at concentrations lower than 0,05%). Longer counting - time can reduce this error, but from the other side of view the concentration of iron in copper or bronze is not significant for their evaluation. A serious disadvantage is the missing information for the content of lead (Pb), because INNA is not sensitive for this element. Therefore we intend to determine lead by Atomic Absorption Spectroscopie (AAS).
Samples were taken from the metallic objects after carefully cleaning the surface at a suitable place and removing the corroded surface by drilling with a borer of 3mm diameter. This treatment was continued till the blank metal appeared in the whole region. After that the whole was cleaned once again carefully and samples were taken boring with a drill of 1mm diameter. The weight of the borings was 30 to 50mg from each object.
The counting has been performed by means of Ge (Li) detector connected with a 4096 multi - channel analyzer.
The energy - resolution of the detector was about 2.2 keV for Co-60.
Fig. 6 presents a graphic survey of the concentrations of the different elements that had been measured (Copper has not been taken in account) the mean values for As and Sb are relatively extreme values up to 14% may exist. This is most probably due to the smelting of grey copper ore with a high percentage of antimony. Relatively high are also the mean - values for Co and Ni. This is not surprising, because it is well known, that the copper ores from the region of interest have a large content of these elements.
The values for silver are also larger than the average of analysis of prehistoric opper or copper - alloys.

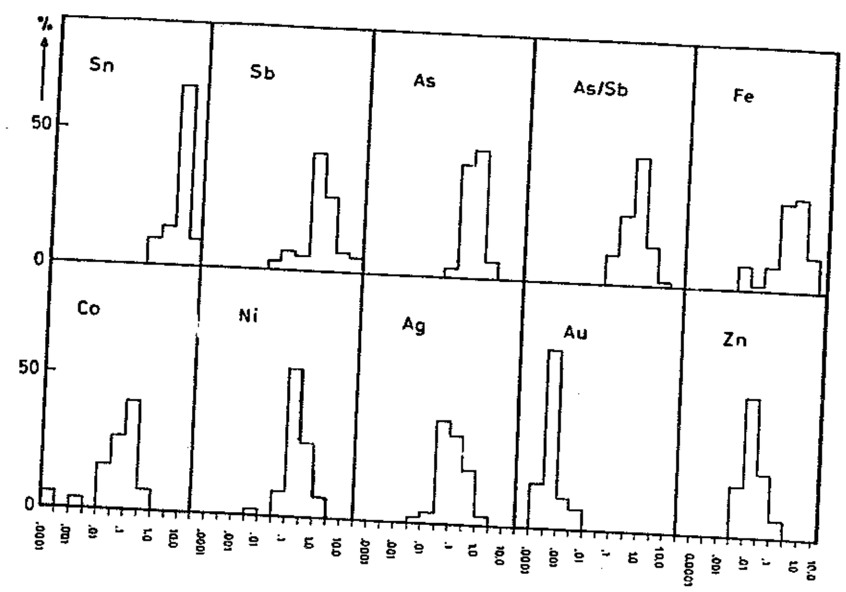

Fig. 6 Frequency of concentration of elements in analysed objects (Remark: PB is not determined)

The objects analysed allow to distinguish for groups:

1. Raw - copper:	No Sn, As and Sb less than 1%:	19 objects
2. Tin - bronzes:	Sn 0,25 - 13%, As and Sb less 1%:	11 objects
3. Antimony - bronzes:	No tin, Sb between 1 and 5%:	12 objects
4. Smelting products of Grey Copper Ores:	No tin, Sb more than 5%:	3 objects

The findings of this group are very brittle, that seems impossible to fabricate articles for practical use from this material without further purification or addition of relatively pure raw - copper.

5. Tin-antimony-bronzes: Sn 1,0 - 10,8%, Sb 1,0 - 2,6%: 9 objects

The objects of the different groups are mostly related to different places where they were found. In these places they appear as several specimen, but often not of a single category. The figure 7 presents the results of analysises of the objects found at Unadingen (Kr.Breisgau - Hochschwarzwald, Südbaden: Stein, 1979, p.121, Nr.293). The results seem to be especially in-

formative. Besides a smelting - product of a grey copper ore (51) four samples of raw - copper (52, 53, 54 and 56) and one tin - bronze (55) are registered. The four raw - coppers have - within the limit of error - identical contents of Co, Ni, Au and Zn. The value for Ag is nearly the same for the four samples. No In was found. The values for Fe, As and Sb are of the same order of magnitude for the four specimen but they differ distinctly. The agreement of the measured values for Co, Ni, Au and Zn is almost better than results of two samples taken from the same object. Therefore it can be supposed that the four fragments of raw - copper castings were smelted from the same using the same technique. Obviously the same technique results the same identical concentrations of Co, Ni, Zn, Ag and Au in the products, but a small change in the conditions of smelting (such as temperature) causes a difference in the concentration of As and Sb. (We do not believe in an intended addition of these elements in such small concentrations.) Nr. 55 is one of the above mentioned tin - bronze samples. Most probably it was fabricated by adding tin to raw - copper of the type described before. The concentrations of Co and Ni are reduced in comparison to the raw - copper because of the resulting dilution (Zn seems to be unchanged because of the greater uncertainty of this element). The contents of Ag and Au are considerably higher. The value of In is very high related to other tin - bronzes (52 ppm). This proves, that Ag, Au and In have been satelites of the added tin. On the other hand Co and Ni are supposed to be only contaminants of copper nad not of tin.

Similary it can be demonstrated, that two tin - bronzes from the depot - finding of Weinheim - Nächstenbach had been produced from the identical raw - copper with addition of tin, having at least in both cases a similar pattern of impurities (object 59 and 60). The concentrations of nickel and cobalt therefore are supposed to be suitable criteria to characterise raw - copper in bronzes. The observation of the results of analysis shows, that the concentrations of Co vary much more than that of Ni and the measurement of Co is more exact than that of Ni. Therefore we have chosen the Co - values of the samples for the classification of the groups.

We distinguish five groups of findings:

Group 1	Co 0.001%	2 objects
Group 2	Co 0.001% - 0.01%	2 objects
Group 3	Co 0.01% - 0.1%	22 objects

Group 4	Co 0.1% - 0.3%	24 objects
Group 5	Co 0.3%	4 objects

It is obvious, that we can find in group 3 mostly tin and tin - antimony - bronzes while in group 4 raw - copper and antimony - bronzes are dominating. Raw - copper of the type "Unadingen" can also be detected in objects found in other places (Niederrad, Bad Homburg and Gudensberg). Our investigations are not yet complete and therefore we avoid further discussion of the results and comparisons with other analytical methods.

UNADINGEN

Probe-Nr.	Sn	Sb	As	Fe	Co	Ni	Ag	Au	Zn	Mn	In
51	--	13.5	2.70	1.0	0.947	2.00	1.01	0.0002	?	0.0011	0.0013
52	--	0.27	0.16	0.3	0.109	0.18	0.047	0.0004	0.03	0.0023	--
53	--	0.33	0.23	0.4	0.109	0.18	0.050	0.0004	0.03	0.0014	--
54	--	0.72	0.35	0.6	0.110	0.18	0.043	0.0004	0.03	0.0014	--
55	5.0	0.44	0.25	0.4	0.100	0.15	0.118	0.0006	0.03	0.0011	0.0042
56	--	0.31	0.14	0.3	0.115	0.2	0.052	0.0004	0.03	0.0008	--

Fig. 7 Analysis of ingots from Unadingen (Remark Pb is not determined)

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