CONCLUSIONS TO THE GEOCHEMICAL ANALYSES OF SOME COPPER SOURCES AND OBJECTS

Introduction

The copper objects analysis, as well as the identification of copper sources and of other metals used during prehistory and the ancient times are part of an interdisciplinary reseach. They were also the subject of several research works *(Deshayes 1960)* and of the expeditions organized both in countries neighbouring ours and in some remote ones: in Yugoslavia *(Jovanović 1971; 1971a; 1975; 1975a; 1978; 1978a; 1979; 1979a; 1982; 1985; 1989; 1990; 1991; Jovanović—Ottoway 1985—1986; Milojčić 1986)*, Greece *(Pernicka 1987; Hauptmann 1988)*, Bulgaria *(Mikov 1961; Todorova 1975*, p. 5; *Cernîh 1965*, p. 127; *Georgiev 1961*, p. 75), Eastern Mediterranean *(Wagner 1986; Zwicker 1989; Gale 1989; Z. A. Gale 1989)*, Anatolia *(Pernicka 1984; Seeliger 1985; Wagner 1986; Wagner & 1986; Wagner 1989; Otzunali 1989; Muhly 1989*, p. 3—4). The Near East *(Hauptmann 1989; Muhly 1989*, p. 2, 4), Czechoslovakia *(Novotna 1970*, p. 3; *Točik-Zebrak 1989)*, Austria *(Eibner 1982; Presslinger — Eibner 1989; Ottoway 1976; 1989)*, South Germany *(Krause & 1988*, p. 181—245; *Presslinger — Eibner 1989)* s.a.

In Romania ancient metallurgy and especially that of copper was the object of a constant preocupation for archaeologists *(Nestor 1933; 1944; 1945; 1955; Popescu 1951; Rusu 1971; 1975; 1975a; 1977;* Diss. doc. and bibl.; *Vulpe 1970; 1975* and bibl.; *Petrescu — Dâmbovița 1977* and bibl.;) as well as geologists *(Niculescu — Otin* 1913; *Coghilan 1962; Stoicovici 1965; 1967; 1977; Stoicovici — Stoici 1971)*.

In Transylvania, owing to its richness in copper minerals and native copper, copper and bronze metallurgy is characterized by a briliant evolution nowhere surpassed in Europe *(Fig. 1) (Gaal 1908; Roska 1941; 1942; Vlassa 1966; 1967; Vulpe 1970; 1975* and bibl.; *Petrescu — Dâmbovița 1977* and bibl.; *Rusu 1971; 1975; 1975a; 1977;* Diss. doct. and bibl.; *Bader 1978*, p. 81—108).

The present analyses are based on the project initiated in the 1980's by prof. Călin Beșliu from the University of Bucharest, the Nuclear Physics Chair, in collaboration with some researchers from the National Museum. Collaborations were extended to the Commission for Archeometry initiated by the Ministry of Culture, the Department for Museums and Collections (project 1990—1991), the Transylvanian History Museum, ITIM Cluj (CA project — SCIMETRIX 1990 S/04/A), IFA Bucharest. Other museums from Transylvania and Banat (Satu Mare, Reșița) initiated a project (1991—1992 program) for copper objects, coordinated by the Transylvanian History Museum and supported by the Romanian Institute of Thracology (project 1992—1993), the

Mineralogical Museum and Department of Mineralogy-Petrometallogeny from the „Babeş-Bolyai“ University of Cluj-Napoca *(Fig. 1—2)* (project 1991—1993), the Reşiţa Museum (project 1992) and other institutions (Department of Civil Engineering from the Polytechnic Institute of Timişoara, project 1992) etc.

The object analysis *(Fig. 1—2)* was extended to copper analysis by analyzing some copper samples displayed at the Mineralogical Museum of the „Babeş-Bolyai“ University (project 1991). The analyses of bronze objects (project CA-SCIMETRIX 1990 S/04/A) and of gold objects are still to be developed.

1. Theoretical aspects of analysis methods

1.1. Neutron activation analysis (NAA)

The methods of analysis consist of thermic neutron activation and X-ray fluorescence. For the neutron activation analysis about 10 mg sample have been introduced in small polyethilene bags and irradiated in the UVR-S atomic pile from the Nuclear Physics Institute, Bucharest — Măgurele. The irradiating process took place at an average influx of 10.12 neutrons/cm p.sec. in a time interval of an hour. After 4—5 days desactivation period, the induced radioactivity was measured with a spectrometer consisting of a Ge (Li) detector and a microcomputer M 118 B with a nuclear interface.

As a result of the characteristic gamma-ray spectra analysis, the following elements were identified: copper, stibnite, arsenite, iron, gold, silver, mercury.

1.2. X-ray fluorescence (XRF)

On the same occasion, native copper objects and several mineral samples (marked UC 51—81), consisting mainly of native cooper, chalcopyrite, sometimes associated with other minerals (covellite, quartz, pyrite, „limonite“, malachite, sphalerite and azurite) were analyzed.

The native copper samples, as well as chalcopyrite samples were investigated by X-ray fluorescence in order to render more accurate the neutron activation technique. Using a Pu 238 activation source of 3×33 miliCurie, the X ray fluorescence was detected by means of a hyperpure germanium detector, data analysis was obtained on a M 118 B microcomputer.

X spectra characteristics for the chemical components of some native copper samples are illustrated in the figures below *(Fig. 10)*.

2. Theoretical issues concerning the objects

(See ActaMP, 1992)

The archaeological literature mentions the use of the following copper minerals: malachite, cuprite and azurite (Fafos, Vinča: *Jovanović 1971*, p. 22; *1987*, p. 31—34), Cu + As minerals in Austrian Alps

(Jovanović 1987, p. 47; *Pittioni 1951*, p. 34; *Witter 1952)* and Slovakia *(Novotna 1970*, p. 3; *Jovanović 1987*, p. 47). other localities from which sulphides were described are Gornja Tuzla *(Jovanović 1987*, p. 47) with which some sulphides *(Jovanović 1987*, p. 47) or carbonates (in Divostin, G. Tuzla, Fafos) *(Jovanović 1987*, p. 52, 54), appear.

Traces of dump slag were identified on several archaeological sites: Iernut *(Vlassa 1966*, p. 11; *Horedt 1976*, p. 175; denied by *Roman 1969*, p. 68); in Cuptoare — *Sfogea* (1980 excavation campaign: S3—1, c. a2a, —0,50—0,60 m) in Sălcuţa culture, phase IIb-IIc), Băile Herculane — *Grota Hoţilor*, Moldova Veche — *Humka (Roman 1976*, p. 16) and Cornea-Cornereva — *Piatra Ilişovii* in late Coţofeni culture.

Copper usage is historically dated in the East aceramic neolithic, from Tell Maghzaliyeh (VII-th milenium BC: *Muhly 1989*, p. 2—5, a copper-silver alloy), pearls made up of native cooper in Tell Halaf Horizon *(Childe 1953*, p. 149) and other objects within Hassuna Horizon and later ones *(Schmidt 1943*, p. 119—120; *Perkins 1963*, p. 8; *Muhly 1989*, p. 4), a horizon contemporany with the CBA phenomen (Chalkolithikum Balcano-Anatolian) *(Lazarovici 1979*, p. 131—132; *1991a; 1991b; 1991c)* in the late Starčevo-Criş, Vinča A and Policromy horizons.

Several small copper objects from these horizons are already known in Trestiana (awl/needle, inf. Eugenia Popuşoi, in the Starčevo-Criş IIIB culture, 1984), Leţ *(Vlassa 1966*, p. 11; *Horedt 1976, 175; Lazarovici 1979*, p. 32, N. 142), Sf. Gheorghe — *Bedehaza* (*Vlassa 1966*, p. 11; *Horedt 1976*, p. 175; *Lazarovici 1979*, p. 32, N. 142), Glăvăneşti Vechi *(Vlassa 1966*, p. 11; *Horedt 1976, 175; Lazarovici 1979*, p. 32, N. 142), Iernut — *Bideşcutul Mare (Vlassa 1967*, p. 3, 2, 407; *Horedt 1976*, p. 175; *Roman*, p. 111, 68; *Lazarovici 1979*, p. 32, N. 142) Dubova — *Cuina Turcului (Nicolaescu-Plopşor 1968*, p. 24; *Horedt 1976*, p. 175; *Lazarovici 1979*, p. 32, N. 142), Balomir *(Vlassa 1966*, p. 11; *1967*, p. 406—407; *Jovanović 1987*, p. 41; *Horedt 1976*, p. 175) and Verbicioara *(Berciu 1961*, p. 38—39; *Comşa 1974*, p. 59, 82; *Lazarovici 1979*, p. 87, N. 127). In the Late Neolithic horizons there are several other discoveries in: Ovčarovo *(Todorova 1975*, p. 5; *Jovanović 1987*, p. 41); Parţa, in Banater Culture (excavation 1991) and in Boian culture *(Comşa 1974*, p. 81) and Liubcova-Orniţa, in Vinča C culture *(Comşa 1974*, p. 82).

Copper metallurgy is an extremely vast theme of the Eneolithic Period for each historical province, its analysis requiring a much larger survey.

The discovery from Ciumăfaia *(Vlassa 1966*, p. 11; *Horedt 1976*, p. 175; *Lazarovici 1979*, p. 32, N. 142) is difficult to assign culturally.

The short presentation from above reveals a permanent interest of prehistoric communities from the Romanian territory for copper metallurgy. Our knowledge is still clearly determined by the field research level in our country. The permanent contact between the Balcanic world and the Anatolian one must be analized from this point of view, otherwise the copper metallurgy expansion in our eneolithic age can not be fully understood.

The results concerning copper objects and sources were submitted to a subsequent cluster analysis. We used two possibilities: first, all the chemical elements were taken into consideration, then a few of the

Genetic Classification of Romanian

Copper Ores and their Minor Elements Associations

(modified, after Giuşcă et al 1969, Petrulian 1972; Mârza 1983, Cheşu 1983, Popescu 1986

1. A. Preneogene ores

Metamorphosed volcano-sedimentary ores	Magmatic ores			Genetic type and main minerals / Metallogenetic age	
	Skarn associated ores	Hydrothermal			
		Veins	Porphyry-copper		
cupriferous pyrites, pyrite, chalcopyrite, galenite, sphalerite	chalcopyrite, bornite, pyrite, pyrhotite, magnetite, molibdenite, tetrahedrite, bismutite	pyrite, chalcocite, digenite, tennantite, bournonite	chalcopyrite, chalcocite, bornite, molibdenite		
				Pre-balkalian	
BURLOAIA (Bi) GURA BĂII (Sc) [In] FUNDU MOLDOVEI (Te) LEŞU URSULUI (Tl) [Sn] BĂLAN (SÂNDOMINIC) (Sn) FAGU CETĂŢII				Lower Cambrian	Paleozoic
MUNCELU MIC VEŢEL				Hercinic	
		HIGHIŞ MTS. BAIA DE ARAMĂ DROCEA MTS.: - ALMĂŞELULUI VALLEY - ROŞIA MONTANĂ - CĂZĂNEŞTI		Paleoalpine	Mezozoic
	BĂIŢA-BIHOR, BĂIŞOARA [Bi] △Ga SASCA MONTANĂ [Co] △Ge STÂNĂPARI-CĂRBUNARI [Ni] △Sn ORAVIŢA-CICLOVA M.-MĂIDAN △In OCNA DE FIER (Cd) DOGNECEA, BOCŞA △Se RUŞCHIŢA △Te		MOLDOVA NOUĂ	Laramic (Banatitic)	Neozoic

I. B. Neogene Ores

Magmatic ores (Hydrothermal)						Genetic type and major elements / Metallogenetic age	
Veins					Porphyry-copper		
Baia Mare-Ţibeş-Rodna Area			Metalliferous Mts.		Cu, Mo (chalcopyrite, chalcocite, bornite, molibdenite)		
Pb ± Cu	Au-Ag-Pb-Zn ± Cu	Cupriferous pyrites	Cu - As (Enargite)	Au-Ag-Te ± Pb ± Zn ± Cu			
CAVNIC BĂIUŢ ŢIBLEŞ x Tl TOROIOGA x Tl RODNA ○Ga	ILBA x Tl BAIA SPRIE ○Ga ○Co ○Ni	ILBA x Tl NISTRU JEREAPĂN	PÂRÂUL LUI AVRAM xCd xIn ○Se ○Te ○Ge BUCIUM ○Ge (ARAMA VEIN)	ZLATINA SLĂNIJA MUŞCA SĂCĂRÂMB BAIA DE ARIEŞ	DEVA ROŞIA POIENI MUSARIU VALEA MORII ROVINA BOLCANA □Mo □Re xAs xSb xIn xGe xcR	Neogene **	Neozoic
□Cd	□Se □Te □Tl ○Ge ○In	□Sn □Bi	○Sn ○Tl △In △Bi	○In ○Ni △Cd △Co			

Legend 1 A ** Including the ores from Germany:
- HERZDORF
- BETZDORF GRUBE

(no genetic information)

Metallogenetic province = = each field of the tabel

Legend 1B ** including the foreign ores:
- GÖMÖR, RECSK (MATRA Mts) = Hungary SZOMOLNOK (= Smolnik, Slovakia)
- BANSKA STIAVNICA (Slovakia)

□ = Minor element with maximum content in Romanian ores

○ = Minor element with maximum values and constant presence in the metallogenetic province

△ = Minor element with high values and frequent presence in the metallogenetic province

x = Absent minor element within the province, ore more frequent chemical elements allowed some further classifications. In *Fig. 9* two of the main characteristics are represented (for classification methods see: *Lazarovici 1990* and bibl.), in order to notice the assignement of certain items to different copper sources or source groups and to identify their possible remelting. In using the hierarchical analysis, fuzzy samples were classified according to all their characteristics. In order to present the situation in certain clusters much more clearly, only those clusters were submitted to a detailed analysis.

As the above presentation reveals, copper occurs in association with several elements in various frequencies. From some late deposits (Uioara) we learn about the existence of some mineral and metal sources used for obtaining the alloys *(Stoicovici 1965*, p. 471—474; *Rusu 1975*, p. 26—29).

3. Theoretical issues concerning sources

3.1. The Romanian copper ores

It is a well known fact that all the ores and copper accumulations consisting of native copper sulfides (chalcopyrite, chalcocite, covellite, bornite, cubanite) and sulphosalts (bournonite, enargite, tetrahedrite) lead to important concentrations of oxides (cuprite, tenorite), carbonates (malachite, azurite), sulphates (chalcanthite) on their upper side, within the oxidation zone, respectively to native copper, sulfides (chalcocite, covellite, bornite) and sullphosalts right under the hydrostatic level, within the cementation (reduction) zone. The native copper amount generated in this supergeneous process is cleary superior to that of hypogeneous origin, occuring during the process of magmatic differentiation and therefore represented the main and most available source for primitive people.

As far as the hypogeneous copper ores from our country are concerned, a relative abundance and a diversified metallogeny can be noticed, however characterized by the preponderence and the economic importance of magmatic origin sources and subordinately of the volcano-sedimentary metamorphosed ones.

Regional distribution is closely dependent on the orogenetic (montaneous) areas, more specifically the Apuseni Mountains, the northern border of the Eastern Carpathians, Banat, Poiana Ruscă Mountains, with a single notable exception, that of Altân-Tepe ore (Dobrogea). From this point of view Transylvania was the most favoured area, as it is practically surrounded by a „belt" of virtual copper sources (see Fig. 3—4, drown up with a few changes according to that of *Giuşcă & 1969; Petrulian 1972; Mârza, 1982; Cheşu 1983)*.

The geochemical association of major elements in the copper ores is slightly different *(Fig. 3—4)*, however, their paragenesis (i.e. the mineralogical association) is, to a great extent, very similar (pyrite, chalcopyrite, spholerite, galena), which implies a metallogenetic frame in interpreting data resulting from geochemical analysis.

According to Pelissonnier's classification (in *Dănilă-Dănilă, 1982)*, which is taking into account the mineralogical association of the copper-bearing rocks as well as the local structure, the Romanian copper ores and accumulations may be assigned to the following categories (groups) *(Fig. 5)*:

Gr. no. Genetical type of ore	Frequent chemical elements	Romanian ores
1 Copper-pyrites associated with volcanic rocks	Zn, Pb, Ag, Au, Se, Te, Ba	Bălan
5 Copper/tin ores	Bi, W, Ag	Băiţa (BH)
6 Porphyry-copper ores with molibden	Au, Re	Roşia—Poieni
7 Enargite containing ores (Cu-S-As)	Ag, Pb, Zn	
9 Basic and ultrabasic rock associated ores, highly nickeliferous	Pt, Co	Ciungani—Căzăneşti
11 Siderite containing ores	Ni, Ba, Ag, Bi, Hg	
12 Arsenopyrite or pyrrhotite and gold ores	Au, Ag, Bi	

Fig. 5. Genetical types of Romanian copper ores (after Pelissonnier, 1972)

3.2. The sample of sources

The samples analyzed in order to identify the copper sources belong to the Mineralogical Museum of the University of Cluj-Napoca and consist of 22 samples represented by dendritic or compact native copper (12 samples from Romania and 10 samples from adiacent territories, respectively) and other 10 samples consisting of chalcopyrite (all these samples belong to foreign occurrences).

Owing to the fact that the samples existing in our museum are quite old, some errors regarding sample localization may have occured and also it is possible that these samples, though mineralogically interesting and having a value of their own, represent only some isolated, unminable copper occurrences. The samples from thes sources unfortunately do not provide a characteristic image for the metallogenetic possibilities of copper accumulation in Romania and the surrounding areas *(Fig. 2)*.

Our selection of native copper samples and copper minerals is just a preliminary stage of the final study, in order to appreciate the accuracy of the analytical and statistical methods employed.

3.3. Determinant factors in the geochemistry of copper sources

We consider that these factors may be grouped into three categories:

1. Geological factors
2. Analytical factors
3. Statistical factors

1. Geological foctors:

a) The geological age of the ore, that is its metallogenetic age, taking into consideration that evolution at global scale of the main geostructural units also influenced the geochemical content of the tectono-magmatic cycles.

b) Genetic type of the ore, the stage reached by the magmatic differentiation process being significant for the accumulation and, respectively, for the dispersion of certain chemical elements.

c) Evaluating, as accurately as possible, the sample position within the succession of crystallisation, taking into account the polyphasic and pulsating character of the mineralized hydrothermal solutions. This process determines a certain vertical and horizontal zonality of magmatic ores, with a content of minor elements slightly differentiated. Owing to this fact, relatively significant different geochemical contents may occur within the same ore between different veins, levels, bodies.

d) The mineral's primary or secondary nature.

2. Analytical factors:

a) The purity degree of the analyzed mineral, in order to avoid the influence of other minerals in intimate growth with the metallic minerals. The monomineral separation methodology implies the chalcographic study, separation under microscope and then some further physical and chemical separation methods.

b) Applying the same method in determining minor elements both for sources and for the archaeological items. This was realised for the 73 samples in the same laboratory and by the same group of researchers. The spectrographic "semi-quantitative" analysis, as well as the quantitative ones (colorimetry, spectro-photometry) mentionned in the geological references being different from those used in the present paper, we had to compare the analythical data used by the respective authors with the present ones with great circumspection. Further classifications and comparisions have to be attempted in future.

3. Statistical factors:

a) The low number of source-samples (22 were relevant) *(Fig. 1)*.

b) The scarcity of geological knowledge concerning certain (foreign) source occurrences.

c) The acceptance of isolated occurrences irrelevant as virtual paleomining centres, as well as of the samples from minor copper accumulation areas;

d) Lack of analyses concerning all the copper minerals which were available as sources.

e) The emphasis, in the future, on the study of supergeneous (secondary) copper minerals which present a greater probability of having served as sources in the past.

f) Future correlation of samples from the Mineralogical Museum with field samples, according to archaeological information regarding paleomining activities, in order to create a data base as complete as possible.

3.4. Theoretical considerations concerning the amount of trace elements in the Romanian copper ores

The metallogenetic provinces of great interest in this case are characterized by the trace elements associations mentioned in *Fig. 3—4 (Cheşu, 1983).*

Within these provinces, certain ores or genetical ore types have an "exotic" geochemical position. We have to mention porphoyry-copper ores, with the presence of Mo, Re *(Vlad 1983)* and the absence of As, Sb, In, Ge, Cr, *(Petrulian & 1965);* Băiţa (BH) characterized by the trace elements W, Re, *(Stoicovici — Stoici 1977);* the Ţibleş Mountains for the lack of Te and the presence of Ga; Ilba and Toroioaga for the lack of Ti; Baia Sprie for the presence of Co and Ni; Herja and Ilba for the presence of Ga; Pârâul lui Avram, for the lack of Cd, In and the presence of Se, Te, Ge; Bucium for the presence of Ge; Valea Lita for the presence of Tl and Tincova, for the presence of Ga *(Gheşu 1983) (Fig. 3—4).*

4. Cluster structure and organization

Data concerning the results of geochemical analyses were dealt with by computer (as far as the method is concerned: *Dumitrescu 1990:* with some improvements by Tarcea 1992), thus the cluster distribution of copper objects and source samples was obtained. The position of each cluster is given by the number of indices that form their name; within each cluster we represented the percentage of copper objects, and of various copper sources (the latter given according to the metallogenetic province) from the total number of samples contained in the cluster. The different number of analyzed samples from sources belonging to a certain metallogenetic province (marked with the same sign) may lead to a slightly altered image concerning the extent to which various metallogenetic types are associated with certain object categories.

Fuzzy coefficient = 2.00 Number of points = 73 Number of characteristics = 6 Hicrarhical cluster classification:

```
--+--+--+-------------- 45-46 50  56  59-60  64  71  -72
  |  |  |
  |  |  +-------------- 1-9, 11-13, 17-21, 23-36, 40 47 49 54-55 57
  |  |                        62  63  65  66  67
  |  |
  |  +----------------- 10 14 15 16 22 37-39, 41 42-43 48 73
  |
  +--+----------------- 44() 51  52  53  58  68  69
     |
     +--+-------------- 61 (Szomolnok)
        |
        +-------------- 70 (Alghero-Sassari, Italia)
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Fig. 6. Clusters of analysed copper objects and sources.

5. Conclusions and suggestions

The present geochemical study dealt with 14 chemical elements identified by the analythical method we used. However, the geological references mention other associations of trace elements present in the metallic sulfides from Romania, slightly different from the "traditional" ones. Cheşu *(1983)* separated the following groups of chemical elements, genetically associated with the metallic sulfides from ores occuring in our country *(Fig. 3—4)*:

a) Major elments: Cu, Pb, Zn, Au, Ag.

b) "Companion" elements (which usually occur in large amounts in sulfides, but in our country they constantly occur as minerals proper.

c) Trace elements, which usually form minerals and ores but in our country they are to be found mostly in sulfides: Bi, Sn, Co, Ni.

d) Trace elements which form quite accidentally and in small amounts their own minerals ("proper" trace elements): Cd, In, Ga, Ge, Tl, Se, Te.

As far as the copper objects, respctively bronze objects, are concerned, based on the results of quantitative spectral analyses, Stoicovici *(Stoicovici 1967)* divides chemical elements — present especially in bronze objects — into the following categories:

— major elements: Sn, Cu;

— minor elements: Pb, Ag, Zn;

— impurities: Fe, Mn, Al, Mg, Ca, Na, N (representing an alumosilicate contribution from rocks is which ore has been melted by primitive men and therefore can provide information about paleominning and paleometallurgical activities regarding the respective objects).

Our next purpose is to include within the analythical interest sphere the trace elements already mentionned in the Romanian geological literature and in the current geochemical production analysis: Cd, In, Ga, Ge, Tl, Se, Te, Bi, Sn, Co, Ni, in order to have the possibility to evaluate correlations qualitatively and quantitatively.

As far as the statistical data analysis is concerned, we intend, in case of having access to geochemical information about the vaste possibilities for copper accumulations from Romania and the surrounding territories, to build some geochemical matrix. They could represent the preponderence/absence of specific trace elements structured in mineralogical, metallogenetical and geochemical categories, also submitted to a factorial analysis which makes possible a rapid comparision between the total virtual copper sources and the archaeological objects.

Copper sources from Romania (Fig. 2—4)

The specification of possible copper sources in Banat and Transylvania is a difficult problem because of the large distribution of copper deposits and of some minerals with copper content that are distributed on large areas. From the study of some specialized works results that this metal appears in a large variety of minerals in our country.

The native copper was found in more than 20 places across the country *(Rădulescu—Dumitrescu 1966*, p. 119—120) *(Fig. 2)*. In some minerals copper appears to be associated with lead or arsenic; algodonite Cu+As; domeykite (Cu+As); pseudomalachite (P); eukairite (Cu2Se. Ag2Se). In other situations copper appears as:

— carbonates (auricalcite; azurite; malachite);

— sulfides (bornite; bournonite; brochantite; chalcophyllite; chalcopyrite; chalcocite; caledonite; cyanotrichite; covelliite; cubanite (microscopic); dognacskaite (where bismuth prevails); emplectite (where bismuth prevails); enargite (+As); jalpaite (Cu2S.3AgS); linarite (+Pb); polibazite (Ag,Cu) 2S.Sb2S3; stromeyerite (Cu2S.Ag2S); tetrahedrite (Cu3SbS3—4); valerite (Cu3Fe4S7);

— silicates; (crisocola);

— oxides: cuprite; dioptase; tenorite (CuO); tyrolite; veszelyte (Cu+Zn).

*

From the above results that copper appears associated with many elements in different frequencies. From some late deposits, like Uioara case, we find out about the existence of some different minerals and metals used for alloys obtaining *(Stoicovici 1965*, p. 471—474; ***Rusu 1975*,** p. 26—29). Some chemical analyses are well known *(Rădulescu—Dumitrescu 1966*, p. 51, 75 and others). The absence of corellations between objects and sources was the main restriction for research development.

The first statistical classification of objects and sources analyses gives us the classification from *Figure 6*. This classification reveals the hierarchic correlation between Bălan mine and the pieces from Caţa and Beşeneu and on the other side between the same zone and those from Turdaş deposit. All objects belongs to Petreşti culture, so we can determine clearly that this civilization discovered and used this source which has southern, balcano-anatholian origin *(Lazarovici 1992 — Draşovean 1992; 1993; 1994)*.

From the same classification results also that a hachet — pick-axe from Turda neighborhood belongs to sources from Criscior and Băiţa vecinity (*Fig. 6*, N. 8, 43, 49). The cluster analysis with fuzzy sets (see *Dumitrescu — Lazarovici 1990*, for method and applications in archaeology) gives us the following classification *(Fig. 7)*:

```
*                                   22 Unknown, chisel

*                                   10 Unknown, flat hachet

*                                   14 Dragu, flat hachet

*                                   15 z. Dej, flat hachet, 41 Ariuşd

*    *                            * 16 Hoghiz, 70 flat hachet, 44 Băiţa,
**    ***    *     *                1—9, 11—13, 17—21, 23—40, 42—43, 58, 68,
                                    53, 52, 51, 45—50, 54—57, 59—60, 62—67, 9,
                                    61, 71—73
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Fig. 7. Objects and sources classification with all elements.
The clusters of objects analyses (L1-41) and sources analyses (UC 42—73).

From the classification of the same data but using this time minor elements and 6 characteristics we obtain the folowing classification *(Fig. 8)*:

```
  *                                         53 Cavnic, nativ
                                         *  68 Betzdorf, native+cuprit
  *                                 *       Matra native/malahit, 70 Sasari native,
      *          *                          52 Bucium native, 44 Băiţa native/malahit'
*                                           L 14=Dragu, flat hachet
            *                               51 M. Nouă native+cuprit+malahit

            *                               69 Hetzdorf native
 * *                                        15 (z.   cj, flat), 61 (Szomolnok native),
*                                           41 (Ariuşd, dagger)

*                                           16 (Hoghiz, flat),

 *                                         37—38 (Cheile Aiudului), 48 UC 7,

*                                           10 (Unknown hachet), 22 (Unknown chisé= *
*                                           10 (Unknown hachet), 22 (Unknown chisel),
                                            42 UC 1,
*                                           L=1—9, 11—13, 17—21, 23—36, 39—40, 43
                                            UC 3, 45—47 (UC 4—6), 49—50 (UC 8—9),
                                            54—57 (UC 13—16), 59—60 (UC 18—19),
                                            62—67, 71—73,
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Fig. 8. The clusters of objects analyses (L1—41) and sources analyses (UC 42—73)

By making a refined classification of the 73 samples with the following technical data: number of points = 73; number of characteristics = 14; maximum number of levels = 7, Fuzzy coefficient = 2.00, maximum admited error = 0.0001, threshold value of the polarization index = 0.750, weight of the linear prototypes = 0.900, we obtain the following clusters (see also fig. 8 with clusters of sources and archaological objects):

The 1st cluster (1.1.1) contains the points: L14 = Dragu, flat hachet, 15 Dej zone, flat hachet, 41 (Ariuşd, dagger). This reveals a grouping of flat hachets and a dagger from Ariuşd.

The 2nd cluster (1.1.2.1.) contains the X's points with the following set: 10 unknown hachet 16 Hoghiz, flat; 22 unknown chisel; 37—38 Cheile Aiudului; 42 Băiţa, native copper, 48 Sântimbru Băiuţului, native copper. This group seems to suggest the possibility that the above objects are made up from any of the two sources.

The 3rd cluster (1.1.2.2.1) contains the X's points with the following set: 45—46 Sândominic, Bălan, native; 50 Banat, native + quartz; 56 Recsk, Matra, native + malachite, 59 Gömor, Ungaria, native copper + malahite, 71—72 Urals, 73 Siberia; a grouping of sources only.

The 4th cluster (1.1.2.2.2.) contains the X's points with the following set: 1—9, 11—13, 17—21, 23—36, 39—40, 43, 47, 49, 54—55, 57, 60, 62—67. This group has to be reanalysed.

The 5th cluster (1.2.1.) contains the X's points with the following set: 51 Moldova Nouă, native copper + cuprite + malachite, 69 Hetzdorf, native copper; a grouping of sources.

The 6th cluster contains the X's points with the following set: 52 Bucium, native; 53, Cavnic, native; 58 Recsk, Matra, native copper + malachite; 61 Szomolnok (= Smolnik) Slovakia, native copper; a grouping of sources also.

The 7th cluster (2.1.) contains the X's points with the following set: 44 Băița native copper + malachite; the group includes only the source from Băița.

The 8th cluster (2.2.1.) contains the X's points with the following set: 68 Betzdorf, source, native copper + cuprite.

The 9th cluster (2.2.2) contains the X's points with the following set: 70 Sassari, Italy, native copper.

From the above classification results that the first cluster contains more sources and all objects analyses from MIT. The subcluster 1.1.1. includes the hachet from Dragu which contains 2.61% arsen. so the piece can be included in the group of arsenic copper. Next subcluster 1.1.2.1. includes L22 samples, a chisel and the pieces from Cheile Aiudului L37—38, the last ones linked to the beginning of arsenic-copper metallurgy and to earlier bronze (due to the chronological position). The same subcluster contains native copper sources from Băița (UC 42) and Sântimbru Băiuțului (UC 48). Another subcluster 1.1.2.2.1. contains sources only (UC 45, 46, 50, 56, 59, 71—72, 73). Another subcluster 1.1.2.2.2., contains many samples of objects and sources. A more refined classification was necesary, so we resumed the classification using only the samples of this cluster.

The 1.1.2.2.2. cluster:
number of objects = 36; number of characteristics = 6;
abbreviations: br = bracelet, h = hachet, fh = flat hachet, hpa = hachet — pick-axe, c = chisel, bs = brilenspirale, hh = hachet — hammer, a = axe.

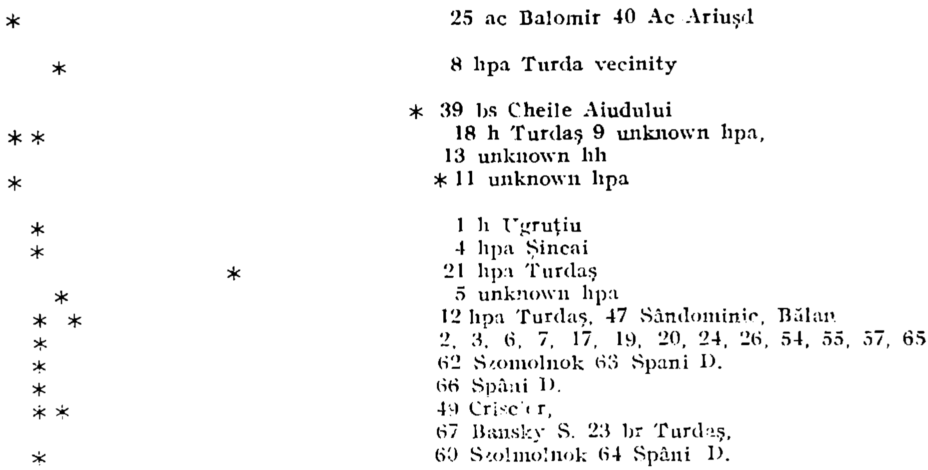

Fig. 9. The new classification of 1.1.2.2.2. cluster

The above image is suggestive, because it gives a refined grouping of samples by two main characteristics. From the above class results that the closest source for the discovered objets is the Bălan mine and the Sândominic zone followed by Recs, Matra and Spani Dolina zones. The bracelet from Turdaş (P 23) is closest to the cluster containing Crişcior (P 49) and Banska Stiavniča (Slovakia) (P 67).

Some of the samples were analysed using X-rays fluorescence with the limits imposed by method (see the diagrams, *Fig. 10*).

In former investigations of copper sources from Transylvania we usualy used to argue the classification diagrams in two dimensions, where Ag/Au diagrams previal and distances are represented by real values (*Fig. 11*).

For better understanding we realised a cumulative diagram of proportions of the same elements. This diagram reveals the reduced measure in which the copper of ores from Transylvania was used locally for artifacts manufacturing (*Fig. 11*).

GH. LAZAROVICI — DANA POP —
CĂLIN BEŞLIU — AGATA OLARIU

ABBREVIATIONS AND REFERENCES

BEŞLIU — LAZAROVICI — OLARIU 1992 C. BEŞLIU, GH. LAZAROVICI, AGATHA OLARIU, *O piesă de cupru din Sălaj şi câteva probleme teoretice privind analizele de cupru preistoric aflate în Muzeul din Cluj*, în *ActaMP*, XVI, 1992, p. 97—128.

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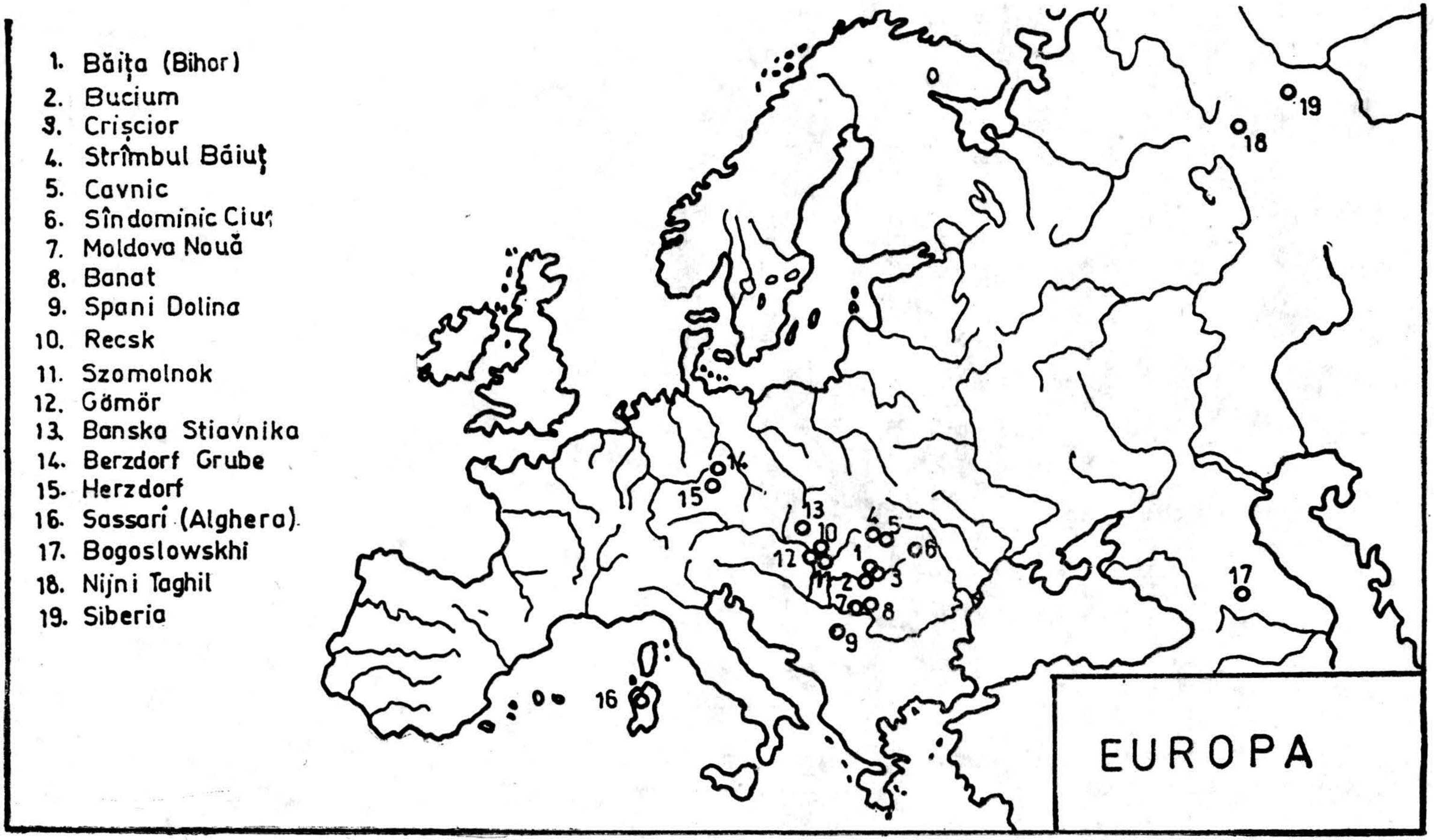

Fig. 1. Geographical setting of the native copper and chalcopyrite samples under study.

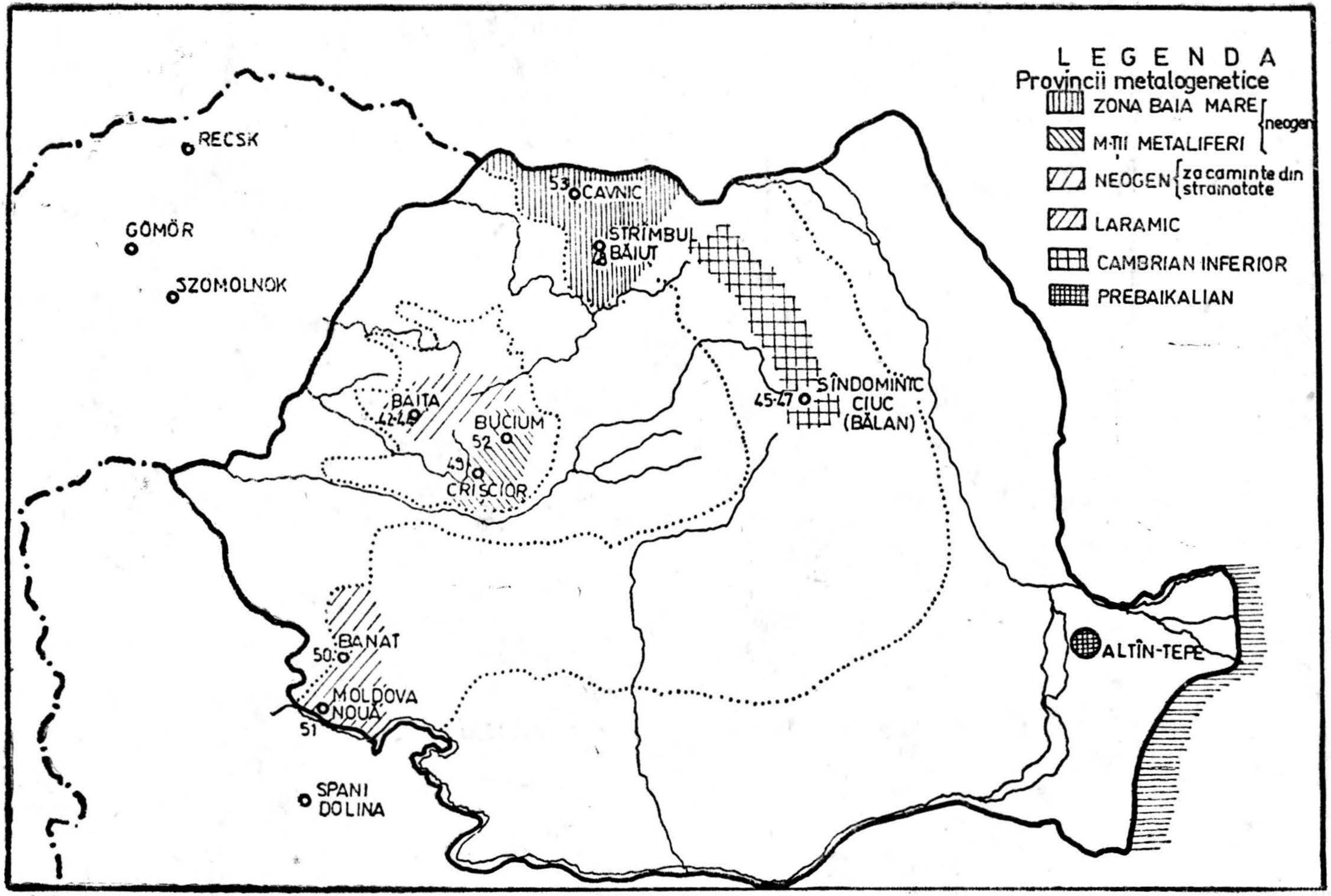

Fig. 2. Metallogenetic frame of copper source samples

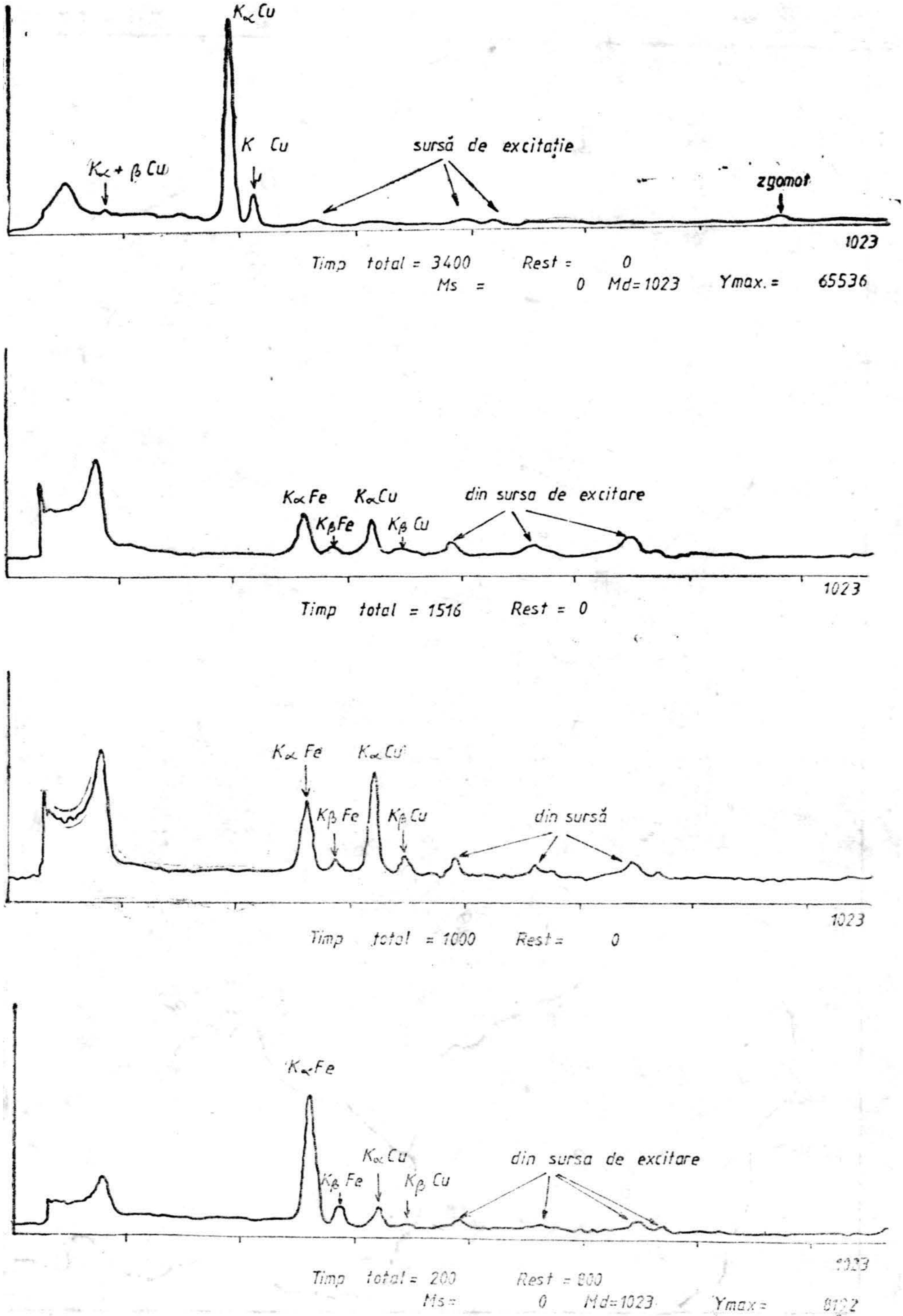

Fig. 10. X-ray fluorescence spectra of some native copper samples (sources).

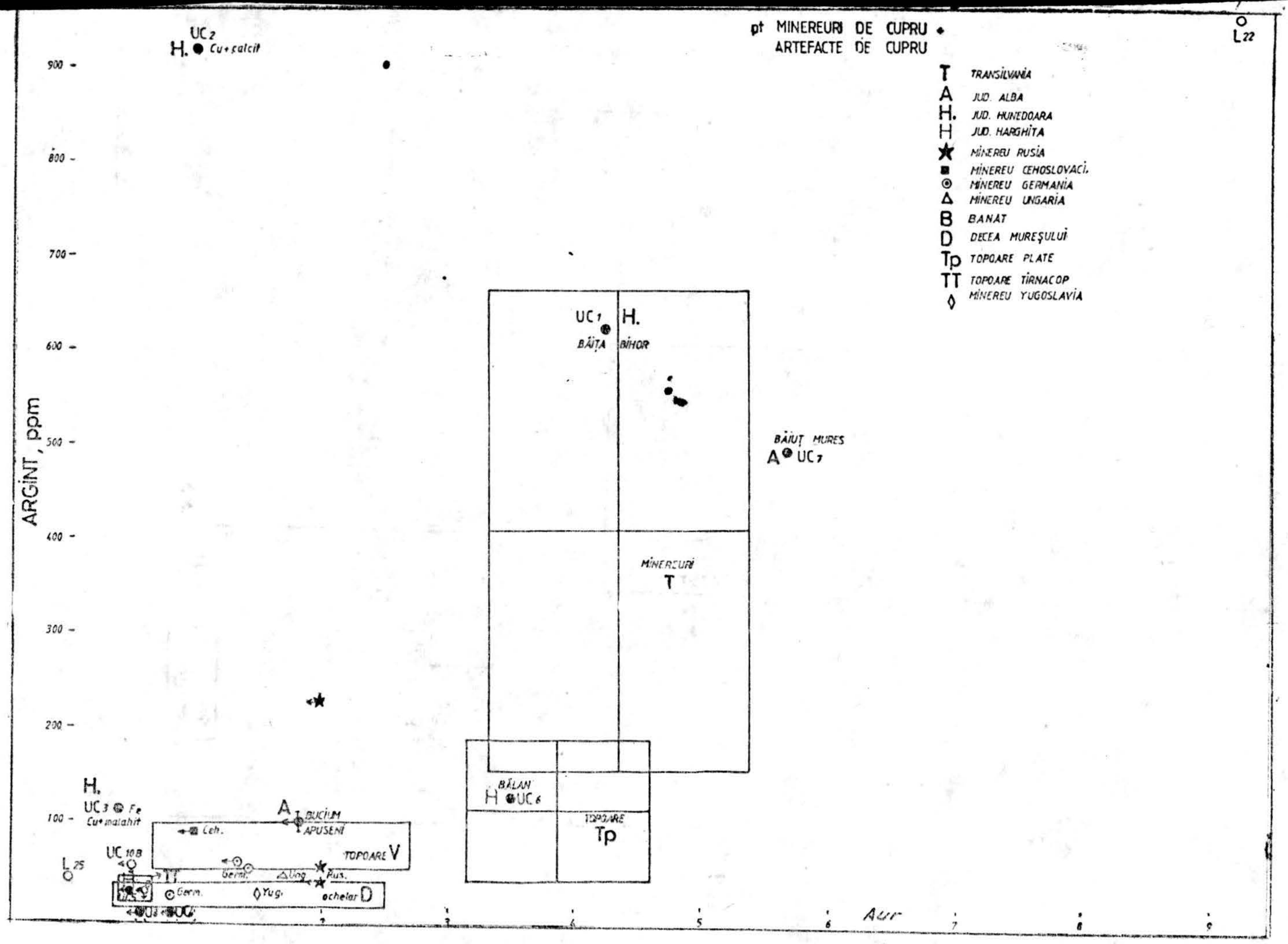

Fig. 11. Ag/Au classification for copper objects and sources.

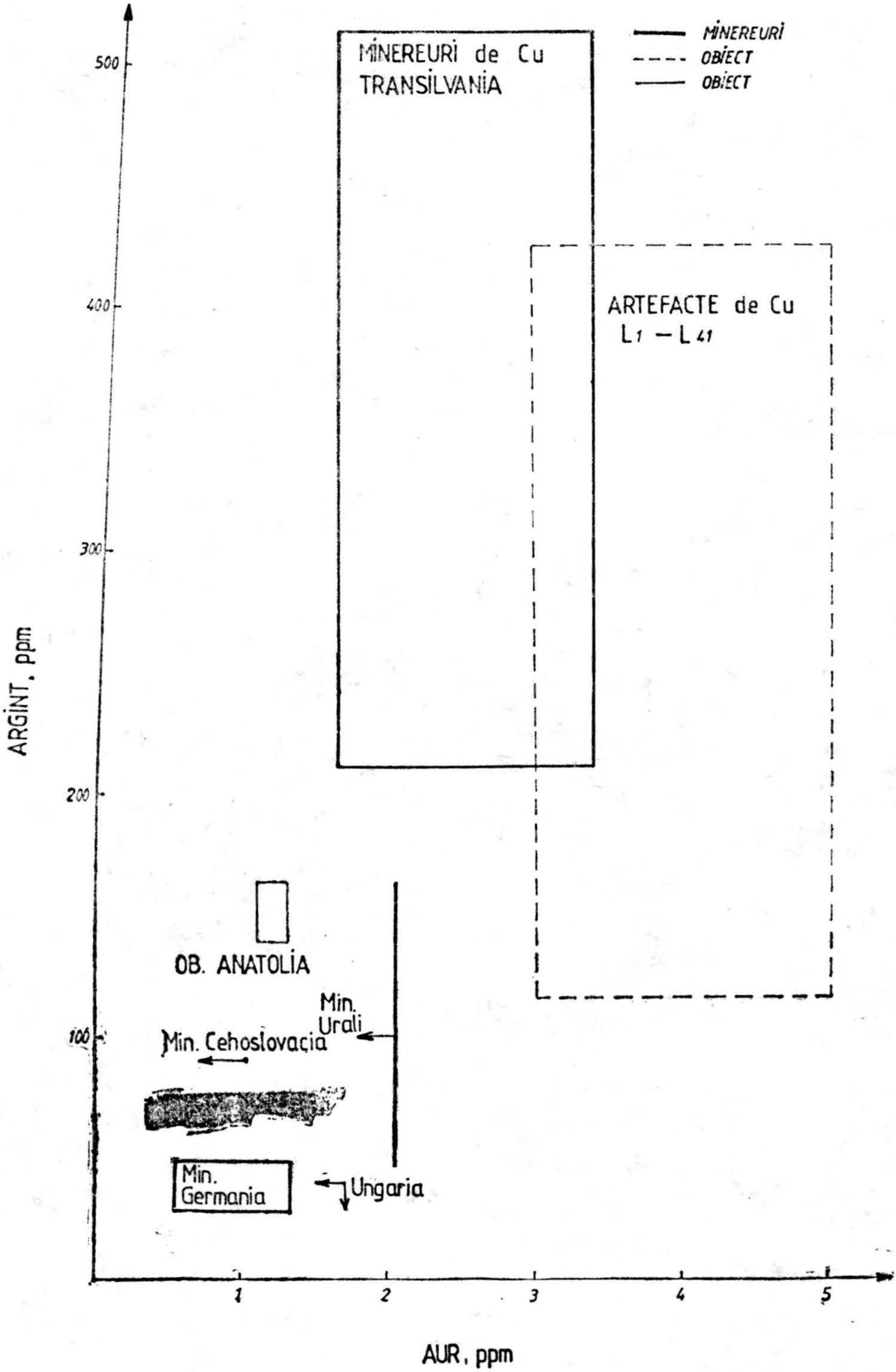

Fig. 12. Ag/Au diagram of copper objects and sources from Transylvania and adjacent areas.