

Analysis of the Elemental Composition of Four Bronze Sickles Dated at the Late Bronze Age

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to Carol Kacsó at his 80th anniversary

Abstract. *The present study presents the analysis of the elemental composition of four bronze sickles, three with hooked handle and one with holes on the handle, two from the deposit at Valea lui Darie, Vaslui county, one from the bronze deposit at Ciorani, and one discovered in a dwelling from the settlement belonging to the Noua culture from Dodești, following systematic archaeological research. The sickles from the Valea lui Darie deposit belong to the Ghermănești type, the Ghermănești variant, the Ciorani sickle to the Ilișești variant of the Ghermănești type, and the Dodești sickle to the Heleşteni type. These types of sickles are characteristic for the Late Bronze Age east of the Carpathians, being chronologically placed between XII – X BC. The elemental composition of the four pieces was identified by employing non-invasive analysis with a mobile spectrometer, Thermo Niton XL3, resulting in a composition of a binary copper-tin alloy (Cu-Sn), to which a number of secondary elements are added: arsenic, antimony, iron, nickel, lead, titanium, and sulphur. In the case of the sickle from Dodești, the percentage of lead is 1.593% which could come from the composition of the copper or tin ores used, given that the percentage should be higher than 2-3% to be considered an intentional addition. Arsenic was also identified in the composition of the four sickles, but in small percentages, > 1%, most likely coming from the composition of the copper ore used. The elemental composition of ancient artefacts enables the establishment of correlations between different types of objects, production areas, raw material resource areas, and distribution patterns. Based on the elemental composition, the four sickles fall into the group of copper objects with arsenic, antimony/antimony, and nickel as the main secondary elements.*

Rezumat. *Studiul de față prezintă analiza compoziției elementale pentru patru seceri din bronz, trei cu cârlig și una cu găuri pe mânerul de prindere, două provenind din depozitul de la Valea lui Darie, județul Vaslui, una din depozitul de bronzuri de la Ciorani și una descoperită într-o locuință din așezarea aparținând culturii Noua de la Dodești, descoperită în urma cercetărilor arheologice sistematice. Secerile din depozitul de la Valea lui Darie aparțin tipului Ghermănești, varianta Ghermănești, seceră de la Ciorani variantei Ilișești a tipului Ghermănești și seceră de la Dodești tipului Heleşteni. Aceste tipuri de seceri sunt caracteristice pentru perioada târzie a epocii bronzului de la est de Carpați, fiind încadrate cronologic între sec. XII – X a. Chr. Pentru identificarea compoziției elementale a celor patru piese a fost utilizată analiza non-invazivă cu un spectrometru mobil, Thermo Niton XL3, în urma căreia a rezultat o compoziție de aliaj binar cupru-staniu (Cu-Sn), la care se adaugă un număr de elemente secundare: arseniu, stibiu/antimoniu, fier, nichel, plumb, titanu și sulf. În cazul secerii de la Dodești, procentul de plumb este de 1,593% care ar putea să provină din compoziția minereurilor de cupru sau staniu utilizate, în condițiile în care pentru a fi considerată adăugare intenționată procentul trebuind să fie de 2-3%. În compoziția celor patru seceri a*

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foșt identificat și arseniul, dar în procente mici, > 1%, provenind cel mai probabil din compoziția minereului de cupru utilizat. Compoziția elementală a artefactelor antice permite realizarea unor corelații între diferite tipuri de obiecte, zone de producere, zone de resurse ale materiei prime și a unor tipare de distribuție. Cele patru seceri, pe baza compoziției elementare, se încadrează în grupa obiectelor de cupru cu arseniu, antimoniu/stibiu și nichel ca principale elemente secundare.

Keywords: *sickles, elemental compositional, XRF, Late Bronze Age, Moldova*

Introduction

In this study we propose to discuss four bronze objects, namely four sickles, belonging to the collections of the Ștefan cel Mare Vaslui County Museum (Fig. 1). Although they have been discussed in various studies on either bronze sickles or bronze deposits, the paper herein approaches a new angle of investigation by presenting the results of elemental composition analysis. The analysis of metallographic data of different types of bronze objects, including but not restricted to sickles, is a key factor in understanding the metallurgical phenomenon at the End of the Bronze Age. Our results extend a very small collection of reported data recorded on objects from the area between the Carpathians and the Prut, and originating from the end of the Bronze Age, that have been analyzed metallographically².

Of the four sickles, three come from hoards and one was discovered isolated. Three of them are sickles with their handle ending in an all-cast hook and are from the Ghermănești type, Ghermănești and Ilieșeni variants, and the fourth with holes on handle, from the Heleşteni type.

Description of the sickles

The first two sickles from Valea lui Darie, Roșiești commune, Vaslui county, come from a bronze hoard discovered accidentally in 1981 in the vicinity of a Noua settlement³.

The first sickle is complete (Fig.2), the blade is wide and strongly arched, with rounded tip, the handle is short and ends in a short one-part hook, and the edge presents a prominent line from the extremity of the hook to the tip of the blade, with the role of strengthening the sickle. It was cast in a closed monovalve mould from the handle joining side of the blade, with the casting stub visible, and it was processed after casting by hammering and sharpening. The blade shows usage marks and it is covered with dark green

² NICOLESCU-OTIN 1913, 404-405, 416-417, 422-423, 425-426; PETRESCU-DÎMBOVIȚA 1977, 18, note 58, 78, where are included *Studien zu den Anfängen der Metallurgie* project dates, Stuttgart; NICULICĂ 1999, 215-220; POPESCU 2009, 272-278; BOROFFKA, MANTU-LAZAROVICI 2011, 148-165; VASILACHE *et alii* 2015, 633-642; DIACONU 2016, 99; LAZANU 2020, 579-592; VASILACHE *et alii* 2021, 1811.

³MAXIM-ALAIBA 1983-1984, 381, fig. 1/1-2.

patina. Dimensions: length - 234 mm, blade width - 35 mm, height of the arch - 125 mm, and weight - 241 g. Inventory no 15331⁴.

The second sickle presents some differences compared to the first one (Fig. 3). It has a wide, knee-shaped bent blade with rounded tip and the handle is long and ends in a short one-part hook. The edge presents a prominent line from the extremity of the hook to the tip and a second line extending from the base of the hook to the tip, going through the middle of the handle and the blade. It was cast in a closed monovalve mould from the handle joining side of the blade, with the casting stub visible. It was processed after casting by hammering and sharpening. A dark and light green patina covers the entire surface of the object. Dimensions: length - 240 mm, blade width - 40 mm, height of arch - 139 mm, and weight - 236 g. Inventory no. 15332⁵.

The third sickle is from Ciorani, Pufești commune, Vrancea county (Fig.4), and belongs to a bronze houred consisting of eight pieces: six hooked sickles, one sickle with simple handle and with two holes (the Heleşteni type), and one socketed axe⁶. The blade is narrow and slightly arched, with tapered tip, and the handle is short and ends with a long hook. The object has a prominent line from the extremity of the hook to the tip of the blade on both edges, making a complete contour of the sickle which was never processed after casting. It was cast in a closed monovalve mould from the handle joining side of the blade, with the casting stub visible. The entire surface is covered with brownish green patina. Dimensions: length - 150 mm, blade width - 25 mm, height of arch - 84 mm, and weight - 70 g. Inventory no. 9.

The fourth sickle (Fig.5) is from Dodești, Dodești commune, Vaslui County, and it was discovered in a settlement belonging to Noua culture⁷. The blade is strongly arched and wide, and the handle has two holes for fixing the handle. The edge has a prominent line from the handle to the tip of the blade which has the role to strengthen the sickle. It was cast from the handle joining side of the blade in a one-part closed mould, with the casting stub visible. It was processed after casting by hammering and sharpening, and the blade shows usage marks. The sickle is covered with dark green patina on the entire surface. Dimensions: length - 170 mm, blade width - 35 mm, height of arch - 95 mm, and weight - 132 g. Inventory no. 1757.

⁴ДЕРГАЧЕВ, БОЧКАРЕВ 2002, 200, таб. 64/827; DERGAČEV 2002, 149; DERGAČEV, BOČKAREV 2006, 246, pct. 827, pl. 64/827.

⁵MAXIM-ALAIWA 1983-1984, 381, fig. 1/2; ДЕРГАЧЕВ, БОЧКАРЕВ 2002, 200, таб. 64/826; DERGAČEV 2002, 149; DERGAČEV, BOČKAREV 2006, 246, pct. 826, pl. 64/826.

⁶PETRESCU-DÎMBOVIȚA 1964, 255, fig. 1/20, 3/2-7; PETRESCU-DÎMBOVIȚA 1977, 74, pl. 78/19, 79/1-4; UDRESCU 1973-1974, 35, fig. 7/3; ДЕРГАЧЕВ., БОЧКАРЕВ 2002, 245, таб. 83.1147; DERGAČEV 2002, 151; DERGAČEV, BOČKAREV 2006, 261, pct. 926, pl. 72/926.

⁷FLORESCU 1991, 60, fig. 106A/9; ДЕРГАЧЕВ., БОЧКАРЕВ 2006, 211, таб. 72.926; DERGAČEV 2002, 147; DERGAČEV, BOČKAREV 2006, 297, pct. 1148, pl. 84/1148.

The four sickles are of two different types: three are hooked sickles and one with holes on the handle. The typological and chronological aspects of these types of sickles were discussed in previous studies. Petrescu-Dîmbovița made the first typological classification of the sickles and divided the hooked sickles into seven types⁸. Another typological classification was made by Dergačev, considering the metric parameters in particular⁹. The sickles from Valea lui Darie belong to the Ghermănești type, Ghermănești variant, after Dergačev typology¹⁰, to the Ghermănești-Ruginoasa type¹¹ after Petrescu-Dîmbovița typology, and to the C 24 and C 26 types after Cernych typology, respectively, all of them belonging to the hooked sickles from the North-Pontic region¹². The sickle from Ciorani is of Ghermănești type, Ilieșeni variant, after Dergačev typology¹³, and of Cristian-Drajna 2 type after Petrescu-Dîmbovița typology¹⁴, respectively. Ghermănești type, Ilieșeni variant is characterized by small size and is from the same typological range as Ghermănești and Ruginoasa variant of Ghermănești type.

The sickle from Dodești belongs to the Heleşteni type, Heleşteni variant, after Dergačev typology¹⁵, and type C 2/4, after Cernych typology¹⁶. The Ghermănești type sickles are common in Moldova in the Râșești-Băleni hoard series, in Transylvania in the Uriu-Domănești hoard series¹⁷, while in the early Hallstatt period this type of sickle declines in number and it can be found only during Ha A1 period¹⁸. The Ghermănești and Ilieșeni variants emerged at the end of the Late Bronze Age, in Br D (XIIth century BC) and developed until the Ha A1 period (Xth century BC).

During the Br D period, hooked sickles were concentrated at the East of Carpathians, with a lower presence in Transylvania, a situation that reverses during the Ha A and Ha B, suggesting that these sickles appeared east of the Carpathians and then spread to the neighboring areas. However, there are also hypothesis supporting the appearance of these sickles in Transylvania¹⁹. Metrical analysis of the three types shows that they are evolutionarily related, evolving from large, large-handled, heavy sickles, to lighter-weight, smaller-handled sickles that required less raw material for casting²⁰.

⁸PETRESCU-DÎMBOVIȚA 1978, 57-72.

⁹DERGAČEV, BOČKAREV 2006, 229-304.

¹⁰DERGAČEV 2002, 149-150; DERGAČEV, BOČKAREV 2006, 241.

¹¹PETRESCU-DÎMBOVIȚA 1978, 63.

¹²ЧЕРНЫХ 1976, 96-97, рис. 44.

¹³DERGAČEV 2002, 151-152; DERGAČEV, BOČKAREV 2006, 257-259.

¹⁴PETRESCU-DÎMBOVIȚA 1978, 60-61.

¹⁵DERGAČEV 2002, 146-148; DERGAČEV, BOČKAREV 2006, 286-291.

¹⁶ЧЕРНЫХ 1976, 89, рис. 40.

¹⁷PETRESCU-DÎMBOVIȚA 1977, 73-77, 51-72.

¹⁸PETRESCU-DÎMBOVIȚA 1977, 80-120.

¹⁹PETRESCU-DÎMBOVIȚA 1953, 475; PETRESCU-DÎMBOVIȚA 1964, 264; PETRESCU-DÎMBOVIȚA 1966, 348.

²⁰DERGAČEV, BOČKAREV 2006, 279.

The Heleşteni type, Heleşteni variant has no prototype and emerged in the Noua culture area, being produced in the Prut-Dniester interfluvium from where it expanded to the East in the Sabatinovka culture. The Heleşteni sickles are dated to the Br D - Ha A1 (XIIth century BC - Xth century BC).

Methods of Analysis

The four sickles were investigated through nondestructive X-ray fluorescence analysis with a portable spectrometer, Thermo Niton XL3, using a Si-PIN diode and a 50 kV voltage for X-ray excitation, 40 μ A, 2 Watt max. In order to perform the measurements, a surface of 1 square cm of patina was cleaned. The depth of the analysis was 0,01 mm, therefore the recorded data show the elemental composition status of the surface layer of the pieces only. Measurement data were processed with Thermo Nito, a program developed by the device manufacturer. The spectrometer can identify a number of 30 chemical elements, but some elements were identified below the limit of detection and their concentration could not be quantified. Thus, the elements indicated as <LOD> by the software were not included in our analysis.

For each element identified, the software adds to the database the analysis error, which is helpful when the data is centralized, analyzed, and corroborated with other data.

Results and Discussions

Analysis of the four sickles has shown that the objects are made of bronze, binary copper – tin alloy, with values of tin ranging between 0,179% and 12,01%. The sickles from Ciorani and Dodeşti have values of tin of 10,806% and 12,01%, respectively, which is within the normal ratio of copper and tin for bronze (90+10%). In the case of the sickles from Valea lui Darie, the percentage of Sn is extremely small, 1,068% and 0,179%, which is unusual but not unique. In the elemental analysis made by E. N. Cernych, there are twelve bronze sickles with tin concentrations between 0,015% and 2%, all of them from bronze hoards: Knjaze-Grigorovka, Becilovo, Rajgorodka, Orekhovo (Ukraine)²¹. From the Ulmi-Liteni hoard there is a hooked sickle with 0,35% tin in composition²². Usual content of tin in copper-tin alloys is around 10%, which could be an indication that the artefact is not recycled but at the primary alloy. When tin is found in alloys in small percentages, around 2% and even below 1%, it could be a sign of recycling of bronzes with different tin levels²³. The most important source of tin is cassiterite (SnO₂), and the closest source appears at Băile Borşa, Maramureş county, at *Burloaia*, *Măgura*, *Gura Băii*, *Izvoru Ursului*, and *Toroiağa* veins²⁴. Another source that could have been used is stannite (Cu₂FeSnS₄) which is a sulfide mineral of copper, iron, tin, and sulphur (28%

²¹ ЧЕРНЫХ 1976, таб 3,4, 8.

²² PETRESCU-DÎMBOVIŢA 1977, 78, inv. no. 893.

²³ BRAY *et alii* 2015, 207.

²⁴ RĂDULESCU, DIMITRESCU 1966, 94; KACSÓ 2011, 269-270.

tin, 12% iron, 30% copper, 30% sulphur) and occurs with chalcopyrite, tetrahedrite, and could be used to obtain a copper – tin alloy. This mineral is rare and has been identified at Băile Borșa, *Burloaia* and *Izvoru Ursului* veins²⁵, Valea lui Stan, Vâlcea county, Hondol, *Pârâul lui Avram – Livia* veins, Hunedoara county, here being associated with chalcopyrite and tetrahedrite²⁶. It is hard to believe that upon obtaining this type of *alloy*, the ancient metallurgists would have left it at this stage to make the two sickles.

All four sickles contain arsenic as a trace element and is difficult to say whether they come from recycling arsenical bronze objects or that minerals containing copper and arsenic were used. It is well known that arsenic losses under oxidizing condition, which occurs during solidification and in smaller scale during melting and purging in moulds²⁷. The sickle from Dodești (read no 1971) contains a small percentage of Pb, 1,593%, that could come from lead impurities in copper or tin ore residues, since an intentional alloy of lead should be considered only when it is above 2%.

Correlation between chemical composition of ancient bronze artefacts and geological sources of metal through trace elements is difficult to establish since it requires a large number of analysis from all known copper deposits. Furthermore, re/melting of metals makes it difficult to relate an object to a specific ore source.

The copper ores that could have been used by metallurgists from the region between Carpathians and Pruth at the Late Bronze Age are in the Carpathian Mountains and Măcinului Mountains, where accumulations of copper sulphosalts, sulphates, carbonates and oxides were present²⁸. To carry out an analysis of the link between the bronze artefacts and resource areas, two minimum conditions must be met: the existence of elemental composition analysis of bronze artifacts and analysis of the chemical composition of copper ores. An increase in the number of elemental composition analyses for artefacts is ongoing. However, analyses on ores are very rare²⁹ and thus, the two conditions are not met.

One useful tool to be used in provenancing copper ores is Pb isotopic analysis, but this method has its limitations since some objects have lower levels of Pb than required to obtain isotopic analysis results³⁰. Using Cu and Sn isotopic information is also a possibility to trace a bronze artifact to the original ore source³¹, but there is no absolute technique of direct provenance in bronze metallurgy. An ancient bronze artefact has a long *journey* which starts with extracting copper and tin ores, casting the material in different forms of ingots later transformed into various objects which are being used, re/melted, repurposed, and finally

²⁵ KACSÓ 2011, 269.

²⁶ RĂDULESCU, DIMITRESCU 1966, 276; SZAKÁLL 2002, 178.

²⁷ MÖDLINGER *et alii* 2019, 133-140.

²⁸ For a complete list of copper ores and there occurs see RĂDULESCU, DIMITRESCU 1966.

²⁹ RĂDULESCU, DIMITRESCU 1966, 51, 75.

³⁰ GALE 1997, 71-82.

³¹ BALLIANA *et alii* 2013, 2973-2986.

deposited. The time between origin and deposition of the object is dependent on the social and economic context³².

Bray considers that interpreting trace elements of copper could be done through the presence/absence of four elements: arsenic, antimony, silver, and nickel, which are the most common in ores. Using this method ancient bronze objects could be included in so-called *copper groups* which are not necessarily related to a specific ore source but rather to the geographical distribution of these artifacts over a certain period³³. Using this method, the four sickles can be included in group 14, copper with arsenic (As), antimony (Sb) and nickel (Ni). In the future, using more data, a statistical analysis regarding the distribution and connections between the copper groups in a certain area can be made and, if chemical analyses of copper ores exist, relations between ores centres and copper groups could be established.

Conclusions

This paper analyzes four sickles, three with hooked handle and one with holes on the handle, whose elemental composition was determined using a portable X-ray fluorescence spectrometer (XRF). This noninvasive technique yields information about the elemental composition of the objects, but only at the level of the surface layer, where patina was removed, and enables rapid and inexpensive analysis of multiple objects. Three of the four sickles have wear traces, except the one from Ciorani that was never used, and all four were discovered in so-called *closed complexes*.

Elemental composition revealed that all four sickles have arsenic traces which could be explained by the use of sulphidic copper ores, possibly tetraehidrite. The recycling of bronze artifacts is attested in the sickles from Valea lui Darie, being the most plausible explanation for the small percentage of tin in their composition.

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³² BRAY *et alii* 2015, 204.

³³ BRAY *et alii* 2015, 205.

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- Figure. 3. The sickle form Valea lui Darie (no. inv. 15332). (Photo and drawn by Lazanu Ciprian)
- Figure. 4. The sickle form Ciorani (no. inv. 9). (Photo and drawn by Lazanu Ciprian)
- Figure. 5. The sickle from Dodești (no. inv. 1757). (Photo and drawn by Lazanu Ciprian)

No Inv.	Reading No	Cu	Cu Error	Sn	Sn Error	As	As Error	Fe	Fe Error	P	P Error	S	S Error	Ti	Ti Error	Ni	Ni Error	Sb	Sb Error	Pb	Pb Error
9	1974	87,568	0,245	10,806	0,066	0,251	0,019	< LOD	0,017	0,106	0,019	< LOD	0,083	0,1	0,006	0,458	0,022	0,222	0,014	0,247	0,016
1757	1971	81,171	0,331	12,01	0,085	0,169	0,035	0,186	0,014	0,554	0,028	< LOD	0,124	0,158	0,007	0,293	0,02	0,17	0,014	1,593	0,037
15331	1953	97,115	0,212	1,068	0,021	0,415	0,021	0,026	0,007	0,077	0,014	< LOD	0,093	0,041	0,004	0,283	0,016	0,111	0,011	0,095	0,011
15332	1951	97,755	0,208	0,179	0,011	0,751	0,031	0,201	0,011	< LOD	0,024	0,2	0,041	0,018	0,003	0,333	0,018	0,116	0,011	0,323	0,02

Table 1. Elemental composition of the sickles from Ciorani (no. inv. 9), Dodești (no. inv. 1757), and Valea lui Darie (no. inv. 15331, 15332).

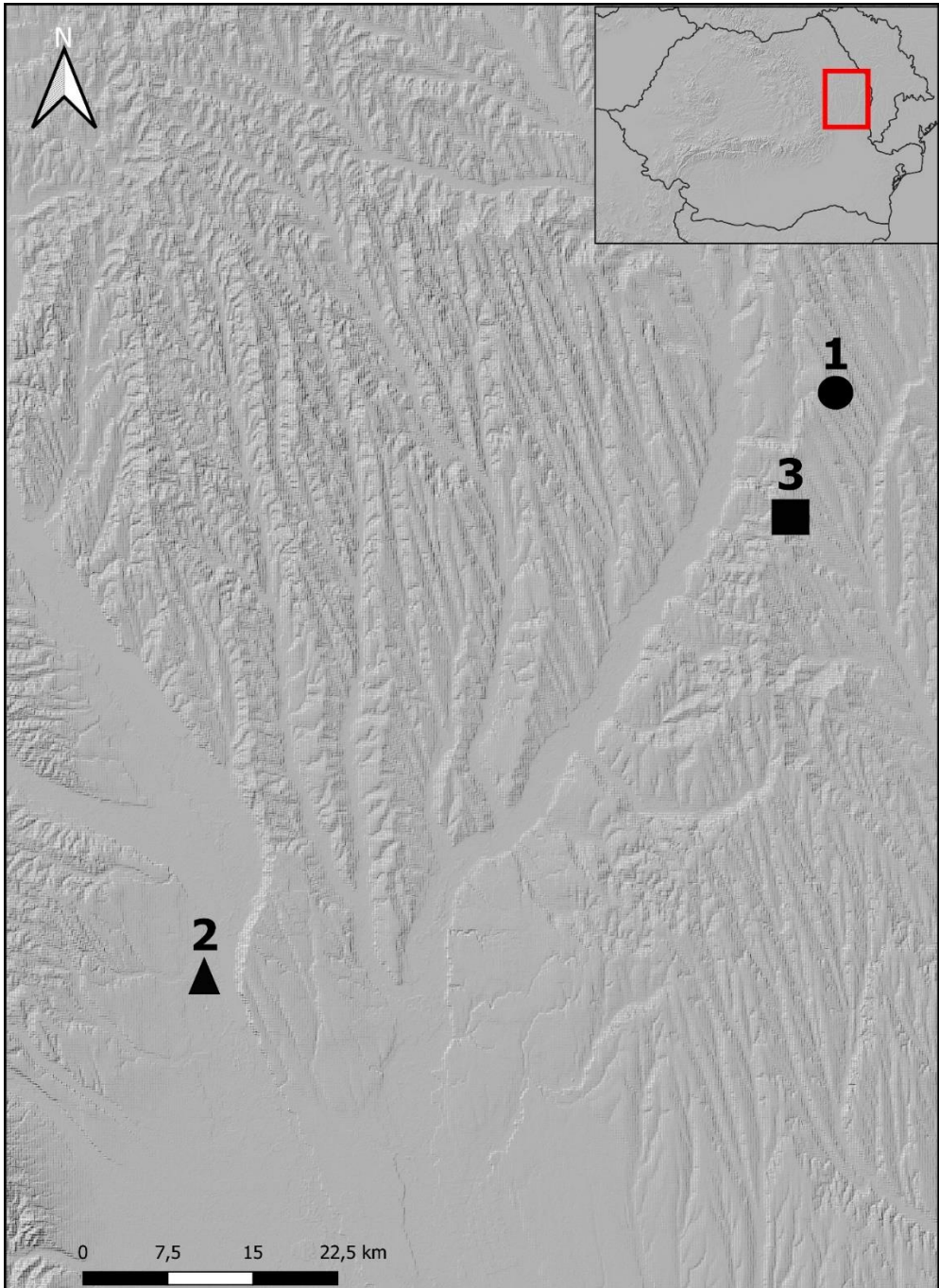


Figure. 1. The map with the sickles discussed in this paper. 1. Valea lui Darie; 2. Ciorani; 3. Dodești. (drawn in QGIS v. 3.34.1 by Lazanu Ciprian)



Figure. 2. The sickle from Valea lui Darie (no. inv. 15331). (Photo and drawn by Lazanu Ciprian)

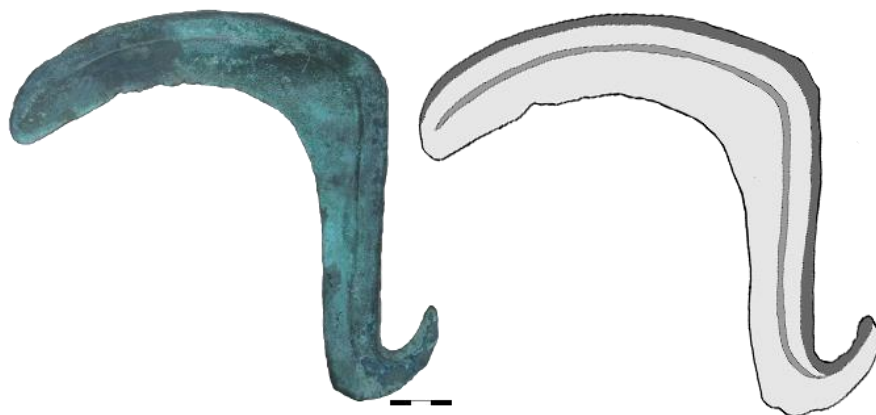


Figure. 3. The sickle form Valea lui Darie (no. inv. 15332). (Photo and drawn by Lazanu Ciprian)



Figure. 4. The sickle form Ciorani (no. inv. 9). (Photo and drawn by Lazanu Ciprian)



Figure. 5. The sickle from Dodești (no. inv. 1757). (Photo and drawn by Lazanu Ciprian)



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