PRELIMINARY HANDHELD XRF ANALYSIS OF LATE BRONZE AGE METAL FINDS FROM THE BUDAKESZI-ŐZVÖLGY-TETŐ SITE (PEST COUNTY, HUNGARY)

A BUDAKESZI-ŐZVÖLGY-TETŐRŐL (PEST MEGYE) SZÁRMAZÓ KÉSŐ BRONZKORI FÉMLELETEK ELŐZETES KÉZI XRF ELEMZÉSE•

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Abstract

This study discusses the chemical analysis results of Middle and Late Bronze Age stray finds and the Late Bronze Age hoards excavated at the recently discovered Budakeszi-Őzvölgy-tető site (Pest County, Hungary). This measurement series is the second stage in the scientific evaluation of the finds, which helps to characterize the elemental composition and raw material characteristics of the A and B hoards and the stray finds. Our aim is to obtain elemental composition information on the 103 objects, which can be used to identify the main alloy types within the hoards, as well as to establish a research strategy for the further examination of the metal finds from the Őzvölgy-tető with more complex and reliable methods. A handheld XRF device was used, which provides compositional information on the surface of the objects, but its main advantage is that it enables the analysis of a large number of samples within a short time. Here, we interpret the obtained data within the limits of the applied method. Based on our results, the majority of the artifacts can be identified as Cu-Sn alloys. Pb, Sb, Ni, Zn, Ag, Fe, Co and As are the main accompanying elements of the objects' patina from Budakeszi, their presence being indicative of the raw material and its source. The presence of certain elements at higher quantities may indicate specific technological phenomena. The high surface Pb content on the convex part of the ingots is probably due to the phenomenon of lead segregation. While the high Fe content could be interpreted as a technological defect and the result of an incomplete iron removal process during smelting. Handheld XRF measurements show a correlation between patina color and surface elemental composition in some cases (e.g., dark gray – high Pb, black – high Pb-Sb), but more typically, the same color can be associated with different elemental combinations (grayish-silver, high Pb-Sb, Pb-Sn, high Sn). Our results help to independently verify the relative dating of the hoards and the stray finds. The high Fe or Zn content, which is not typical of the Late Bronze Age, allows us to date some of the stray find lumps and one sheet of metal to the Medieval Period or Modern Ages. The high surface Sb and Pb contents observed in Late Bronze Age artifacts from Budakeszi are in good agreement with former quantitative elemental composition data from Transdanubian (e.g., Biatorbágy, Velem-Szent Vid), Austrian and Slovenian artifacts deposited in the Ha B1 period, which support the archaeological dating of the hoards and most stray finds.

Kivonat

A tanulmányban az újonnan felfedezett Budakeszi-Őzvölgy-tető (Magyarország, Pest megye) lelőhelyről származó, késő bronzkori kincsleletek, illetve középső- és késő bronzkori szórványok elemanalitikai vizsgálatának eredményeit tesszük közzé. A vizsgálatsor a tárgyak feldolgozásának második fázisa, amely segít jellemezni az A és B depók, illetve a szórványok elemösszetételét és nyersanyagának jellemzőit. Munkánk célja, hogy a 103 darab tárgyról elemösszetételi információt szerezzünk, mellyel azonosíthatók a főbb ötvözettípusok a kincsleleteken belül, valamint kutatási stratégiát állítsunk fel az Őzvölgy-tetőről származó fémleletek összetettebb módszerekkel való további vizsgálatára. Méréseinkhez kézi XRF készüléket használtuk, mely a tárgyak felszínéről ad összetételi információt, ám a módszer nagy előnye, hogy rövid idő alatt nagyszámú minta analízisét teszi lehetővé. Tanulmányunkban a vizsgálati módszer korlátainak figyelembevételével vonjunk le következtetéseinket. Eredményeink alapján, a tárgyak nagy részét Cu-Sn ötvözetként határozhatjuk meg. A

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budakeszi tárgyak patinájának fő mellékelemei a Pb, Sb, Ni, Zn, Ag, Fe, Co és As, melyek jelenléte a nyersanyagra és a nyersanyagforrásra utal. Egyes elemek nagyobb mennyiségben való jelenléte összefüggésbe hozható különleges technológiai jelenségekkel. A nagy felszíni Pb tartalom az öntecsek domború felén valószínűleg az ólom szegregáció jelenségével magyarázható. Míg a nagy Fe koncentráció technológiai hibaként értelmezhető, és a vas nem megfelelő eltávolításának eredménye a kohósítás során. A kézi XRF mérések alapján a patina színe és a felszíni elemösszetétel között néhány esetben megfigyelhetünk összefüggést (pl. sötétszürke – nagy Pb, fekete – nagy Pb-Sb), de jellemzőbb, hogy ugyanolyan színt különböző elemkombinációk alkotnak (szürkés-ezüst – Pb-Sb, Pb-Sn, nagy Sn). Az eredményeink segítenek tisztázni a depók és szórvány tárgyak keltezését. A késő bronzkorban nem jellemző nagy Fe vagy Zn tartalom alapján néhány szórvány rög és egy lemez korát a középkorra vagy a modern korra datáljuk. A késő bronzkori, budakeszi tárgyakban megfigyelhető nagy felszíni Sb és Pb tartalmak pedig jól illeszkednek a kvantitatív módszerekkel elvégzett, Ha B1 időszakban deponált, dunántúli (Biatorbágy, Velem-Szent Vid), osztrák és szlovén tárgyakon elvégzett eredményekhez, melyek alátámasztják a depó és a legtöbb szórvány régészeti keltezését.

KEYWORDS: HANDHELD XRF, TRANSDANUBIA, MIDDLE (BR A2) AND LATE BRONZE AGE (HA B1), CU-SN ALLOYS, HIGH PB CONTENT, METAL HOARDS

KULCSSZAVAK: KÉZI XRF, DUNÁNTÚL, KÖZÉPSŐ (BZ A2) ÉS KÉSŐ BRONZKOR (HA B1), CU-SN ÖTVÖZETEK, NAGY PB TARTALOM, FÉMDEPÓK

Introduction

In 2017, the Community Archaeological Program of the Hungarian National Museum excavated new hoards in close proximity to each other in the Budakeszi-Őzvölgy-tető site (Pest County, Hungary). The assemblages, named Hoard A and Hoard B, can be dated to the Ha B1 phase (ca. 1000–900 BC) of the Hungarian Late Bronze Age (**Fig. 1.**). Also, in the Őzvölgy-tető, several metal detecting campaigns have been carried out, which resulted in a handful of bronze stray finds from the Middle Bronze Age (Br A2) and the Late Bronze Age (Ha A1 and Ha B1).

The complete evaluation of these findings was part of a 2-year long research program between 2020 and 2022, which was funded by the János Bolyai Research Scholarship of the Hungarian Academy of Sciences. The archaeological results of the research project have been recently published in a book titled *"Twin Hoards. Metals and Deposition in the Buda Hills, the Pilis and the Visegrád Mountains during the Late Bronze Age"* (Tarbay 2022).

In this study, we are presenting the handheld XRF analysis results for the two hoards and the stray finds, a total of 103 objects. Our goal is not to over-interpret the results but to gain a general overview of a diverse body of archaeological material from a point of elemental composition, identifying the main constituent elements and alloying materials.

We would also like to demonstrate that the handheld XRF method is a useful tool to refine a research strategy before embarking on a complex archaeometallurgical analysis dealing with expensive and advanced methods.

Method and samples

A total of 103 objects from Hoard A (A1–68), Hoard B (B69–75), and stray finds mostly dated between the Middle Bronze Age (S101–103) and the Late Bronze Age or later periods (S76–100) (Tarbay 2022, 187–192, Pl. 1–14) have been analyzed using a handheld XRF. It is noteworthy that finds dated to the Medieval period, the Early Modern period, and World War II were also found at this site. These objects were not included in the present study (**Fig. 1.**). (As it will be explained in the *Discussion*, some stray finds may also belong to this group: S89–91, S93, S98.) All the analyzed objects have been cleaned by Eszter Tóth in the laboratory of the National Centre for Conservation and Conservation Training, Hungarian National Museum, who used only distilled water to remove soil residue from the objects' surfaces. The objects' patina was not altered during the cleaning process.

No chemicals or artificial coating materials were used during or after the cleaning process. The cleaned finds were later studied with respect to metalwork production and wear analysis performed by J. G. Tarbay, whose results have already been published in-depth elsewhere (Tarbay 2022, 55–71). For this study, new micrographs were taken by a Digi-Micro Mobile digital USB microscope-camera (Digital magnification: 20×–500×, Optical photographs: 2592×1944 Pixel) in order to visualize and document our macroscopic observations on the different color surfaces of the studied objects from Budakeszi-Őzvölgy-tető. During the study of the finds, five color variants were identified that seemed to be different from the "average look" of a prehistoric object (**Fig. 2.1**): 1.) pale gray (**Fig. 2.2**), 2.) grayish-silver (**Figs. 2.3–4, 8**), 3.) dark gray (**Fig. 2.5**), 4.) red (**Fig. 2.6**) and 5.) black (**Fig. 2.7**).

Fig. 1.: The Budakeszi A and B hoards (Hungarian National Museum, Photo: József Rosta)
1. ábra: A Budakeszi A és B depó (Magyar Nemzeti Múzeum, Fotó: Rosta József)

Shugar and Mass used X-ray fluorescence spectrometry as a well-established and widely used method in various fields of applications (Shugar & Mass 2013). Reproducible XRF measurements require sample preparation or, in the case of a homogeneous object, an even, flat surface to be measured. Surface morphology and the presence of corrosion products can affect the XRF results due to the limited information depth (Donais & George 2013; Šatović et al. 2013). Commercially available handheld XRF spectrometers are easy to operate and provide elemental concentration data quickly. Owing to their relatively small size, they are the perfect choice for on-site measurements. Though one needs to be aware of and consider its limitations, XRF can provide valuable information by posing pertinent questions. XRF spectrometry has proved to be an efficient tool in the non-destructive characterization and pre-screening of alloys, e.g., to identify the alloy-type (Maróti et al. 2018) and forgeries (Rózsa et al. 2019). Handheld XRF is also a powerful tool with which to examine composite objects and to draw conclusions on their production technology (Mozgai et al. 2021).

We are aware of the limitations of the various analytical techniques that were thoroughly detailed in handbooks and papers (Nørgaard 2017; Rózsa et

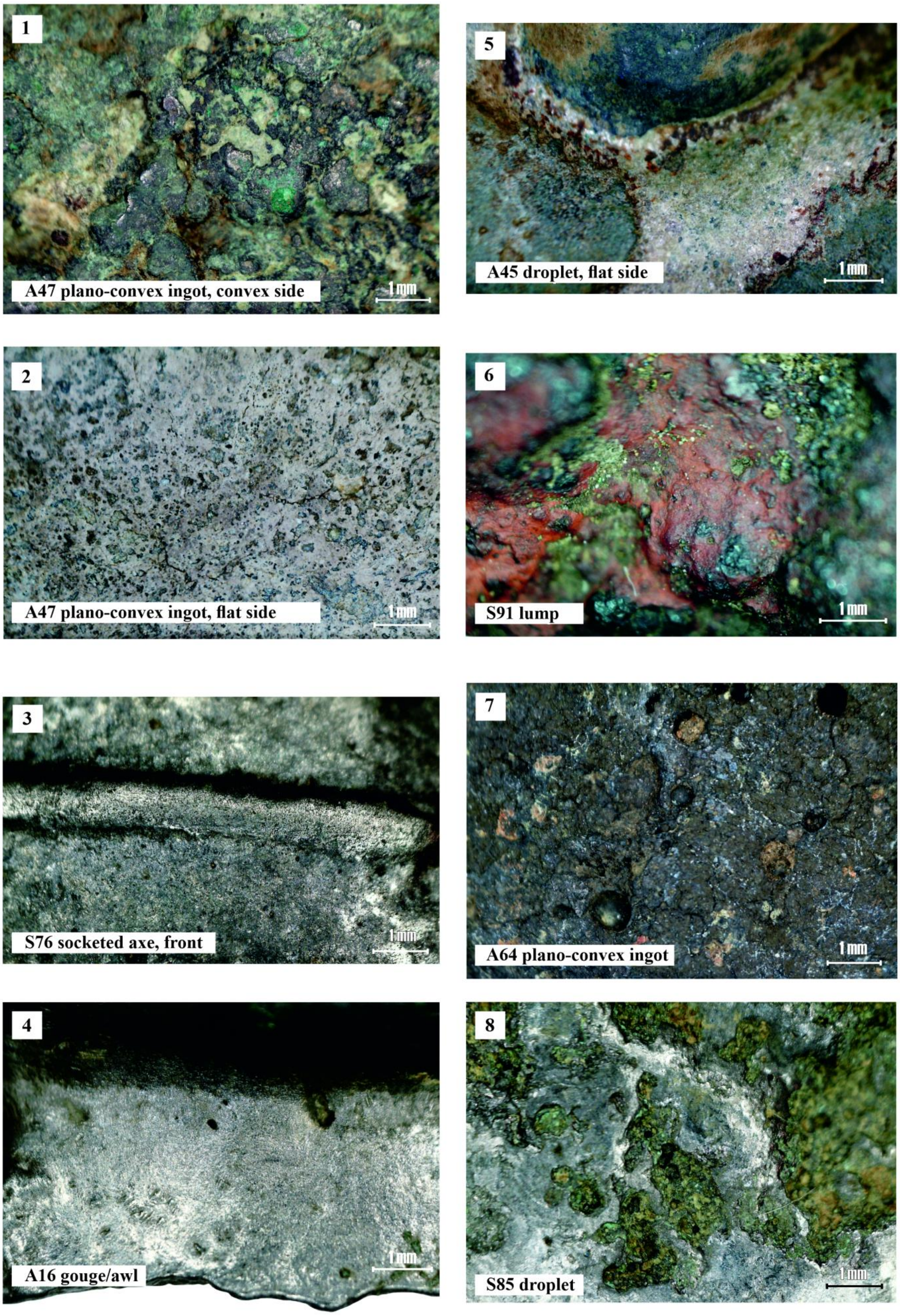

Fig. 2.: Different colored surfaces of the objects from the Budakeszi-Őzvölgy-tető hoard (Microscope-camera images: J. G. Tarbay).

2. ábra: A Budakeszi-Őzvölgy-tető lelőhelyről származó tárgyak különböző színű felszínei (Mikroszkóp-kamera felvételek: Tarbay J. G.)

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al. 2019; Szabó et al. 2019). As in our previous studies, the XRF method has been applied as a preliminary analytical method (Maróti et al. 2018; Maróti et al. 2020a; Tarbay et al. 2021). XRF is primarily used here to gain a general overview based on surface elemental composition data, to hypothesize patterns, to identify specific representative objects, and to strategize for further analysis. The non-destructive, semi-quantitative surface chemical composition analysis was done with an InnovX (now Olympus) Delta Premium type handheld XRF spectrometer (Maróti et al. 2020b). The power of its X-ray tube is 4 W, the anode material is Rh. The device is equipped with a Peltier-cooled silicon drift detector. The Alloy Plus calibration setting was used to detect the components of the copper alloys. The Alloy Plus mode operates with two different beam settings: 40 kV voltage with 100 µA current to determine Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, W, Hf, Ta, Re, Pb, Bi, Zr, Nb, Mo, Ag, Sn, Sb, and 8 kV / 200 µA to complement the set with lighter elements, e.g. Mg, Al, Si, P. The X-ray beam was 3 mm in diameter, while the total spectrum acquisition time was 40 second for each measured spot. Because the factory calibration of the device does not report the concentration of arsenic, an important ore-related element in ancient bronzes, BrightSpec bAxil spectrum analysis software was used to identify arsenic from the spectra (www.brightspec.be).

Results

Based on the handheld XRF analysis, many of the studied objects can be identified as binary Cu-Sn alloys. The main minor elements identified in the objects are: Pb, Sb, Ni, Zn, Ag, Fe, Co, and As. It can be observed in several cases that the Pb and Sb on the objects' surfaces appear as major components and are present in larger amounts than their Sn or Cu content. There are also objects with high Fe and high Zn content. **App. 1** summarizes the XRF results. In the object ID, A and B mark the artifacts of Hoard A and B, respectively, while S stands for the stray finds. More than one XRF measurement was made for some objects that are larger in size or look heterogeneous. Please note that the mass% concentration of an important accompanying element, arsenic, is not included in the factory calibration element suite of the XRF spectrometer, but its peaks are identified from the spectra. Therefore, the arsenic concentrations are listed in arbitrary units (a.u. (arbitrary unit) = As K_α - Cu K_α peak area ratios), while the other elements are listed in mass percent (m%).

Discussion

Color

During the analysis, we paid attention to the documentation of colored surfaces (**Fig. 2.**). Based on the color of the bronzes' surface and the elemental composition analysis results available at the time, Amália Mozsolics had already assumed that certain colors could indicate specific elemental composition: silverish-black (white bronze); blue (presence of Sb); light green (bell bronze, Zn around 20 m%) (Mozsolics 1984, 29–30; Mozsolics 1985, 94, 146). The pale gray color that was observed on several specimens in Hoard A (e.g., A3, A7–8, A12, A44, A47) and among the stray finds (e.g., S82), in some cases with corrosion flakes, can be associated with high Pb content based on the results of the handheld XRF analysis (**Fig. 2.2; App. 1.**). The dark gray patina observed on the droplet (A45) was also related to high Pb content (**Fig. 2.5**). Grayish-silver-colored surfaces, which may be identical to Amália Mozsolics' "white bronze patina" (Mozsolics 1984, 29), were observed on numerous objects from Hoard A and among the stray finds (A1–11, A14, A16, A42, S76, S83, S85) (**Fig. 2.3–4, 8**). The XRF analysis could not relate them to specific elements. These phenomena seem to be related to different element combinations observed in higher ratios on the surface, such as Pb-Sb, Pb-Sb-Sn or high Sn (S83 bar ingot). The black-colored A64 ingot can be associated with high Pb-Sb content (**Fig. 2.7**). It is worth noting that the destructive SEM-EDX analysis (energy dispersive X-ray analysis) of Maclaen and McDonnel revealed high Sb content along with 2 m% Pb on a "black ingot" from the Románd hoard (Ha B2, Győr-Moson-Sopron County) (Maclaen & McDonnel 1996, 81). The red-colored surface is a result of high Fe content (S89–91, S93), these objects were most likely made of iron (**Fig. 2.6**). Thus, our analysis supports the idea that there are some correlations between the composition and the color of the patina, but not in every case. The presence of different patina and colored surfaces on objects that were excavated from the same assemblage itself goes against the common mistaken belief that only artifacts with identical patina can compose a hoard, which was a key argument during the last century in Hungary to "reconstruct hoards" which were acquired by museums from uncertain archaeological contexts (see e.g., Kemenczei 1984, 147, 183; Mozsolics 1985, 24, 99, 119, 122, 134, 175, 190–191, 193, 201, 209, 211).

Tin

The surface Sn content provided by the pXRF measurements ranges up to 63 ± 1 m% (on line 43, in **Table 1**, measurement ID 41, object A23) in the studied finished products like tools and ornaments (**App. 1.**). Based on earlier publications, components above 2 m% are considered as intentional alloying elements (Figueiredo et al. 2007; Oudbashi & Davami 2014). However, it is also known from the literature that Sn and Pb are often present in elevated amounts in the corrosion layers, up to 3–4 times more than in the bulk (Figueiredo et al. 2007; Maróti & Káli 2021; Tarbay et al. 2021). Therefore, the elevated Sn (and Pb) content of the studied objects may be caused by this phenomenon (**App. 1.**). The measurements provided by the pXRF are not suitable to draw an exact conclusion on the ratio of alloying elements in the finished products.

Not all the finds were binary Cu-Sn alloys. Some objects' patina from the Hoard A lacks Sn completely or Sn is below the detection limit: A5 as-cast Palotabozsok-type socketed axe, A40 hammered sheet metal bracelet, A49, A52–56, A59–60, A63, A66 plano-convex ingots. Besides objects identified as non-prehistoric (S89–91, S93, S98), several Bronze Age stray finds fit into this pattern: S76 socketed axe, S79 and S81 plano-convex ingots (**App. 1.**). It is worth noting that all plano-convex ingots lacking Sn can be distinguished by a high surface Sb content and a lower ratio of different accompanying elements. On the other hand, the two socketed axes and the bracelets (e.g., A7, A12, A39–A40) can be characterized by high surface Pb content. It is important to note that most of the ingots (plano-convex, bar, and cuboid) do mainly contain Pb along with Sn in some cases (**App. 1/67–109, 129–137**). This result points out the fact that the Budakeszi-Őzvölgy-tető hoards may contain several ingots that were not made of primary Cu raw material, as was identified in the case of other Transdanubian hoards. Some of them are most likely made of materials that were left over after casting in the crucible and are cast in the form of portable ingots that can be further used. Another very plausible scenario proposed by Zoltán Czajlik is that they are intentionally alloyed ingots such as the Gór type, of which examples are known from Transdanubia, West Hungary and France (Loczka 1889, 282, No. 6; Lázár 1941, 377, Fig. 9; Czajlik, et al. 1999, Fig. 2.3, Fig. 2A, Fig. 3(A), Fig. 4, Fig. 11. M9.10–12, 10.3–4; Czajlik 2006, 58; Czajlik 2012, 70, Fig. 7, Pl. 36; Tarbay et al. 2021). Alloying with Pb in the case of the current XRF ingot series is likely for objects A46–67, S80, and S82 (**App. 1/67–109, 131–132, 135–136**), while alloying with Sn is likely for bar ingot No. S83 (**App. 1/137**).

Iron

Among the Budakeszi-Őzvölgy-tető finds, high Fe content is correlated with reddish surfaces. The presence of Fe in the Bronze Age objects may have had technological implications. László Paksy proposed that Late Bronze Age objects were intentionally alloyed with Fe because of early experiment with this metal (Paksy 1986, 15). Zoltán Hegedűs proposed four different scenarios how Fe could enter the bronze products, of which he finds most likely that this elemental composition pattern is more likely the result of the incomplete iron removal during the smelting process (Mozsolics & Hegedűs 1963, 259–262). Zoltán Czajlik arrived at a similar conclusion and proposed that some of the Fe remains in the raw material after the smelting of chalcopyrite, and iron oxide traces are indicators of technological errors that can be related to the incomplete iron removing process (Czajlik 2012, 96–97). The Fe surface content of stray find lumps Nos. 89–91 and 93 is greater than 97 m%, implying that these iron objects may represent the modern or medieval habitation phase of the Budakeszi-Őzvölgy-tető site.

Zinc

Zn is also considered a rare element in Late Bronze Age finds. It was present in five plano-convex ingots, of which three were part of hoard A (A49, A54, A59) and five were found as stray finds (S80 plano-convex ingot, S90, S91 and S96 lumps, and S98 cast fragment). Zn is a specific element that may refer to the copper ore used to produce the objects. It is important to note that lead ores are often associated with zinc ores due to their geochemical characteristics (Liversage & Pernicka 2002, 425; Czajlik 2012, 53; Gregurek, Peng & Wenzl 2015, 1986). According to Zoltán Czajlik, Late Bronze Age artifacts containing Zn (ca. 1-2 m%) are known from the Bohemian Massif, suggesting that local ores may contain this element. A fine example of such is the area of Přibram, where even Bronze Age copper ore extraction is known. Banská Štiavnica in Slovakia and Zletovo in Macedonia are also plausible sources for such material (Czajlik 2012, 45, 53–54, 98). One cast fragment (S98) has a high surface Zn content of 80.2 m%, which presumably applies to the bulk as well. This specimen may be of modern origin (see Tarbay & Maróti 2021) and its bulk composition should be further investigated in the future by Prompt Gamma Activation Analysis.

Pb, Sb and the Transdanubian Ha B1 period

Since handheld XRF analysis does not provide us with bulk-representative data, it is not possible to compare our results to previous measurements with any confidence. Keeping this caveat in mind, however, some things can be concluded. Previous elemental composition results made on Transdanubian Ha B1 (Ha A2/Ha B1) finds are the most essential to consider because fine typological analysis of the finds suggests that most of the objects from Budakeszi-Őzvölgy-tető represent this period (Tarbay 2022, 31–53).

The elemental composition data on Late Bronze Age finds from Transdanubia were first summarized by Baron Kálmán Miske (Miske 1907) and more recently by Zoltán Czajlik (Czajlik 2012) and Géza Szabó (Szabó 2013). A new synthesis of previous measurements was provided by the main author of this work in an open-access, updatable Mendeley Data set published in 2022 and updated in 2023 (Tarbay 2023). This new database not only contains a vast collection of raw data from publications, but it also contains revised relative chronological dating, additional literature information, and re-identification of objects whose elemental composition data was published without the basic archaeological context. Transdanubian, Ha A2/Ha B1 published elemental composition data is known from a total of 10 sites, and 70 objects: Biatorbágy-Herceghalom (Ha A2/Ha B1) (Liversage & Pernicka 2002, Tab. 2), Csabdi (Ha A2/Ha B1) (Czajlik 2012, Pl. 36), Gór-Kápolnadomb-Quadrant C-12, pit "c" (1032-928 cal. BC) (Ilon 1996, 184), Lovasberény (Ha B1) (Czajlik 2012, Pl. 36), Nagydém (Ha B1) (Ilon 1989, Tab. 2), Székesfehérvár (Ha A2/Ha B1) (Czajlik 2012, Pl. 36), Szombathely-Jáki út (Ha B1) (Ilon 2002, Fig. 10), Várvölgy-Nagyláz-hegy 7th and 10th Ha A2/Ha B1 hoards (Mödlinger et al. 2014, Tab. 2–4; unpublished results in Müller 2017a, 46–47), Velem-Szent Vid IA (Ha B1) (Bakos & Borszéki 1989, Tab. 1.29–30; Czajlik et al. 1995, Fig. 4; Költő 1996, Tab. 1–2. Velem 13–14; Maclaen & McDonnel 1996, 81, Fig. 1; Szabó 1999, 333–334, BRVEL80, 86–88; Költő, Kis Varga & Maclaen 2002, Tab. 2. Velem 13–14, 52–53, 55, 67, Tab. 3. Velem 14, Velem 55), Velem-Szent Vid (Ha B1) (Tarbay et al. 2021, Tab. 3–5). There are also a handful of stray finds which can be securely related to the Ha A2/Ha B1 and Ha B1 periods: e.g. the Budapest-Óbuda sword with cup-shaped pommel (Ha B1) (Loczka 1889, 208, No. 2; Kemenczei 1991, 50, Pl. 44.199), the Velem-Szent Vid socketed axe with pseudo-wings (Ha A2/Ha B1) (Miske 1907, Pl. 14.39; Költő, Kis Varga & Maclaen 2002, Tab. 2. Velem 71), and a chased arm ring (Ha B1) (Miske 1907, Pl. 34.37; Szabó 1999, 331, BRVEL72). The above findings were studied by different, incompatible invasive and non-invasive methods, such as AES, XRF, SEM-EDX, XES, laser micro-spectrochemical analysis, PGAA, TOF-ND, and wet chemical analysis.

The accompanying elements (Pb, Sb, Ni, Zn, Ag, Fe, Co, As) observed in the Budakeszi-Őzvölgy-tető Hoard A and Hoard B (**App. 1.**) as well as in the stray finds are also present in the Transdanubian bronzes deposited at the Ha A2/Ha B1 and Ha B1 (Tarbay 2023). Considering the Ha B1, the most important are the Pb and Sb, which seem to be high in the objects' patina according to the pXRF analysis. Although the present data is quantitatively unreliable, the possibility should not be excluded that at least Pb was high in several of the studied finished products, as-casts, and ingots. High lead content was detected in numerous cases on light gray colored surfaces and on the convex (rounded side) of the ingots (**Fig. 3.**). In contrast, the ingots' flat sides usually showed higher Cu content. During the casting process of plano-convex ingots, and ingots in general, the flat side can be identified as the top, while the convex side is the bottom (see Modl 2010; Modl 2019) (**Fig. 3.**).

This may refer to the phenomenon of *lead segregation*. Since Cu does not dissolve the lead, it forms small droplets in the bronze in both liquid and solid phases. Too much Pb may cause lead droplets to sink to the bottom of the melt, resulting in a lead segregation process (Tylecote 1962, 119; Harrison–Craddock & Hughes 1981, 159–160; Hughnes, Northover & Staniaszek 1982).

Ingots with lead segregation in Transdanubia are known from the Velem-Szent Vid site. Such a plano-convex ingot was studied by Zoltán Czajlik and Kamilla G. Sólymos. Based on their four measurements taken from the object's cross-section, they have observed a similar phenomenon in this non-homogenous object, since the Pb content tends to decrease towards the flat side (Czajlik et al. 1995; Czajlik & G. Sólymos 2002, 317–318, Fig. 1–2). Another scenario is that some of these objects may have been almost pure lead ingots, as turned out to be the case in the PGAA analysis of ingots from Velem-Szent Vid (see Tarbay et al. 2021, Tab. 3–5).

High Pb content was observed in other Ha A2/Ha B1 and Ha B1 material from Transdanubia. The new pXRF dataset quite strongly reminds us of the AES results of the Biatorbágy-Herceghalom hoard (Pest County), which was found ca. 20 km north of Budakeszi. The analyzed objects had a relatively high Sb content and a Pb content of between 1 and 86 m% (Liversage & Pernicka 2002, Tab. 2). High lead content objects and ingots were also present in the Velem 1a hoard, based on SEM-EDX and XRF analyses (Költő, Kis Varga & Maclaen 2002, Tab. 2; Czajlik et al. 1995). Recent PGAA, pXRF, and TOF-ND analyses of another possible hoard find from Velem-Szent Vid revealed quite high lead content in ingots, reaching as high as 94 m% (Tarbay et al. 2021, Tab. 3–5). Both analyses seem to fit well with the general characteristics of the Velem-Szent Vid site based on numerous measurements made on stray finds (See further results with citations in Tarbay 2023). It is also worth noting a bar ingot with high Pb content found in the Ha B1 hoard from Szombathely-Jáki út hoard (Ilon 2002, Fig. 8.2, Fig. 9, Fig. 10. Jakd010). The PGAA analysis of the greave from the 10th hoard from Várvölgy-Nagyláz-hegy has also revealed 2.5 m% Pb content (Mödlinger et al. 2014, Tab. 4). Based on the currently available data on Transdanubia, high Pb and Sb content is sporadic prior to the Ha A2/Ha B1 and Ha B1 in this region (Czajlik 2012, 94–98; Tarbay 2023). As earlier examples, SEM-EDX data (Pb: 25.25 m%) on a

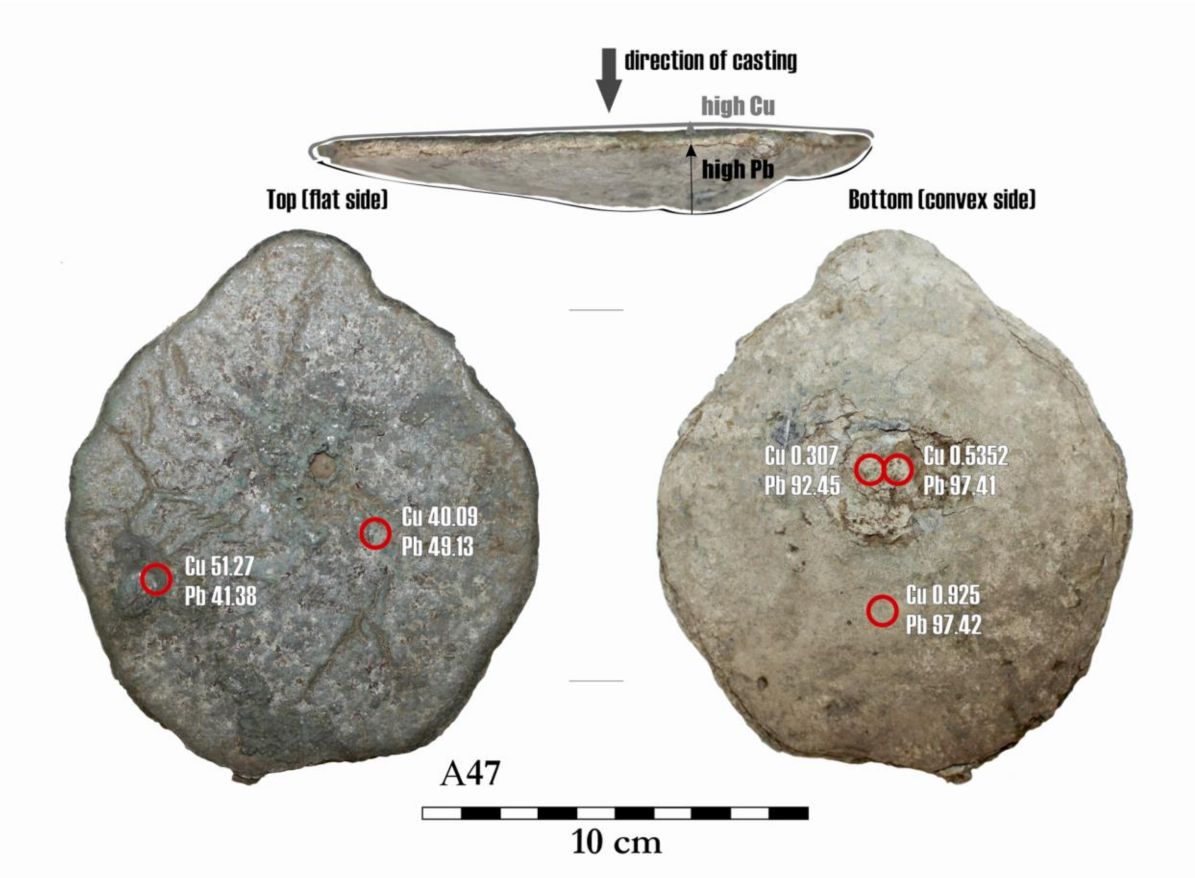

Fig. 3.: A47 plano-convex ingot (Inv. No. 2021.8.47) from the Budakeszi with possible lead segregation at the bottom. pXRF measurement spots are marked with red circles. (Hungarian National Museum, Photo: J. G. Tarbay).

3. ábra: Az A47-es Budakeszi öntőlepény (Ltsz. 2021.8.47), alján valószínű ólom szegregációval. A kézi XRF mérési pontok vörös körrel jelöltek (Magyar Nemzeti Múzeum, Fotó: Tarbay J. G.).

Regöly-Veravár fibula (Ha A1) (Szabó 1993, 179, Pl. 5.27; Szabó 1998, Fig. 4) and an AES data (Pb: 67.78 m%) on a Žatec-type sheet metal object from Várvölgy-Szebike-tető ("Ha A1") can be mentioned (Müller 2017b, 340, Fig. 9.4a, Tab. 1). Transdanubia is not the only region where high Pb and/or Sb content was observed in Ha A2/Ha B1 and Ha B1 objects. The sporadic appearance of high lead content objects before the Ha B1 was also observed in the Austrian and Slovenian Late Bronze Age material (Trampuž Orel 2004, 208–210). In the analysis of the finds from a potential hoard from Velem-Szent-Vid (see Tarbay et al. 2021), we have already discussed this trend with reference to literature dealing with the topic. Considering the Budakeszi-Őzvölgy-tető hoards, it is worth citing results from the adjacent territory. Important results were provided by coupled plasma atomic emission spectrometry, which also revealed high Pb and Sb content in Slovenian Ha B1 finds (see Trampuž Orel 1996, 208, 210; Trampuž Orel 1998). This trend can also be observed in Austrian hoards like Enzersdorf (Pernicka & Mehofer 2013, Tab. 2) and the Mahrersdorf hoard (Mödlinger & Trebsche 2020, Tab. 2). In brief, if the high Pb and Sb content of the studied finds from Budakeszi-Őzvölgy-tető are verified by quantitatively reliable methods, they can certainly be related to the above-discussed trend of leaded object horizons observed in Transdanubia and its adjacent territories during the Ha A2/Ha B1 and Ha B1 periods. The appearance of leaded objects, particularly semi-finished products such as hammer-shaped ingots, shaft-hole axe ingots, and plano-convex ingots, is part of a widespread pattern that can be observed in the whole Mediterranean area (Molnár et al. 2012, 263–264).

A strategy for future analysis

The elemental composition results provided by the handheld XRF instrument has provided us with several preliminary results which can be further investigated. Many objects were determined as Cu-Sn alloys; accompanying elements were detected; "time-marker" elements such as Pb and Sb were observed; and non-prehistoric objects were identified. The next step of our research will be the quantification of results by more precise methods and the investigation of specific technology-related questions and certain elements that play a key role in the archaeological interpretation of the studied finds.

1.) Determining the exact Sn ratio is an important question regarding the casting quality, durability and functional capabilities of prehistoric finished products. Therefore, the Sn ratio of these objects from the Őzvölgy-tető hoards, particularly the weapons and the tools, should be determined by a quantitatively reliable method. We support the use of Prompt Gamma Activation Analysis (PGAA) because it is non-invasive, and we intend to use this analysis method on these objects soon.

2.) Provenance of the finds is also essential, and it can also be achieved by invasive and destructive methods that are able to precisely characterize the objects' accompanying elements (*Laser Ablation Inductively Coupled Plasma Mass Spectrometry* – LA-ICP-MS) and determine the lead isotope ratios of the finds (*Multicollector Inductively Coupled Plasma Mass Spectrometry* – MC-ICP-MS). Among the many methods available for determining the accompanying elements (see Szabó et al. 2019, Tab. 1), XRF may be useful if the patina on the objects is removed (Radivojević et al. 2018). For this type of approach, we would first select plano-convex ingots from Hoard A, particularly those that seem to be lacking Sn and have low surface Sb and Pb content. In this way, we may be able to limit the possibility of analyzing alloyed objects made of a mixture of different raw materials. Since the presence of Zn is rare in the local Late Bronze Age material, ingots containing this element should also be studied. The Fuchsstadt-type can also be added to this list since it is a potential "import" find from the territory of Germany. It was found in 13 individual fragments, some of which are suitable for invasive analysis without substantially affecting the aesthetic appearance of the objects.

3.) Numerous objects showed "large" surface Pb and Sb content. Since Pb is an important element in the relative chronological dating of the assemblages, this phenomenon should be characterized by quantitatively reliable, preferably bulk methods. Our previous work has demonstrated that this goal is achievable by the combination of non-destructive and non-invasive analyses (PGAA, TOF-ND) that are able to identify the ratio of both Pb and Sb (Tarbay et al. 2021). In this case, the most suitable candidates for analysis are the ingots and the as-cast Palotabozsok-type socketed axes with defects, because the latter could be potential lead ingots cast in a specific shape.

Conclusions

Elemental composition pXRF results made by the handheld XRF instrument of 103 objects originating from the Budakeszi-Őzvölgy-tető site are presented in this study. Many of the studied Middle Bronze Age (Br A2) and Late Bronze Age (Ha A1, Ha B1) finds are Cu-Sn alloys. However, the pXRF results show that Sn is not present in the patina of every studied specimen. In almost all cases, the ratio of Sn in the patina is quantitatively not reliable enough to draw a conclusion about the bulk content of this element inside the casts. Some objects have high Zn content or consist almost completely of Fe, and therefore are probably dated to the Medieval, Early Modern Period or World War II eras of the Őzvölgy-tető. Pb, Sb, Ni, Ag, Fe, Co, and As were the most common accompanying elements found in the Middle and Late Bronze Age objects. Some specimens showed a detectable amount of Zn, a rare element in Carpathian prehistoric objects. The appearance of Fe could have production technological causes. It may enter the Cu during the smelting process. The rest of the accompanying elements, i.e., Ni, Sb, Pb, Ag, and Co, probably also refer to the copper ore. The presumably high ratio of Pb and Sb in some of the artifacts is important because these elements are "time markers" of the Ha B1 horizon, further supporting the relative archeological dating of Hoard A and Hoard B (Tarbay 2022, 19–53). Some objects, especially the ingots, showed significant compositional differences in Pb ratio between their top (flat) and bottom (convex) sides (**App. 1., Fig. 3.**). This phenomenon may be caused by the lead segregation effect also observed in Ha B material with high lead content in Transdanubia. The handheld XRF results support the hypothesis that Hoards A and B, as well as some of the stray finds, were made of the same raw material. This further supports the connection between the two hoards, and it is another argument to interpret them as one unit deposited separately in close proximity to each other in the framework of the hoarding ritual (Tarbay 2022).

Based on our results, we were able to provide a general overview of the finds from the Őzvölgy-tető and provide a basic characterization of the finds from an elemental composition point of view. This preliminary data set is essential for strategically planning the next research phase of the material, in which the main questions are focused on the reliable determination of the bulk alloy ratio (Sn) by PGAA, and the accompanying elements by LA-ICP-MS with particular attention to Pb and Sb, as well as the determination of the lead isotope ratio of the finds, which would allow us to formulate an opinion on the possible ore sources of the Bronze Age finds from the Őzvölgy-tető.

Contribution of authors

János Gábor Tarbay Conceptualization, Investigation, Formal analysis, Writing – Original Draft, Writing – Review and Editing, Visualization, Funding acquisition. **Boglárka Maróti** Investigation, Validation, Writing – Review and Editing, Data Curation.

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