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Tartalom / Contents

Archeometriai Műhely / Archaeometry Workshop No. 2025/3.

Scientific meeting on the occasion of the 20th anniversary of the journal "Archeometriai Műhely"

Az Archeometriai Műhely megalapításának 20. évfordulóját ünneplő előadói nap

29th October 2024, Ceremonial Hall, Hungarian National Museum

Szervezők / Organizers: Hungarian Society of Archaeology and Art History, Hungarian National Museum, Subcommittee on Archaeometry, Committee on Geochemistry, Mineralogy and Petrology, Xth Section of Earth Sciences, Hungarian Academy of Sciences

A kötet szerkesztői / Eds.: Szilágyi Veronika, Bajnóczi Bernadett

Cikkek / Articles

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Közlemények / Other communications

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A rendezvény szervezési és a 2025/3. szám nyomtatási költségeit az Nemzeti Kulturális Alap A2034/N0078 sz. pályázata és a Magyar Régészeti és Művészettörténeti Társulat biztosította.

AZ ARCHEOMETRIAI MŰHELY 20 ÉVE: MÚLT, JELEN S JÖVŐ...

20 YEARS OF ARCHEOMETRY WORKSHOP: PAST, PRESENT AND FUTURE... •

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Az Archeometriai Műhely 2024-ben ünnepelte fennállásának 20. évfordulóját, ami különleges alkalmat nyújt arra, hogy visszatekintsünk a folyóirat történetére, fejlődésére és elért eredményeire. A Műhely a régészettudományhoz kapcsolódó természettudományos kutatás hazai szinten meghatározó, nemzetközileg is jegyzett szaklapjává nőtte ki magát, amely magyar és angol nyelven biztosít publikációs lehetőséget a tudományos közösség számára.

Az Archeometriai Műhely 2004-ben indult útjára, egy olyan korszakban, amikor a digitális publikációs formák még gyerekcipőben jártak Magyarországon. A folyóirat alapítását az archeometria tudományának hazai és nemzetközi előmozdítása iránti igény hívta életre. Bár a Műhely elsősorban a magyar archeometriai kutatások főbb eredményeinek bemutatására alakult, már a kezdetektől fogva nyitott a nemzetközi közösség felé. A szerkesztőbizottság fő célja az volt, hogy a tudományos eredményeket gyorsan és széles körben elérhetővé tegye. A szakmai minőség biztosítása érdekében szigorú lektorálási folyamatot vezettünk be, amely máig a folyóirat egyik erőssége.

A folyóirat fejlődésében fontos állomás volt, amikor bekerült a Scopus nemzetközi tudományos adatbázisba. Ez az eredmény nemcsak a folyóirat ismertségét növelte, hanem jelentősen hozzájárult a benne megjelent cikkek tudományos értékének elismeréséhez is (**1. ábra**). A Scopusba való bekerülés szigorú minőségi követelmények teljesítését feltételezte, amelyek magukban foglalták a publikációs normák betartását, a nemzetközi szerzőgárda bevonását, valamint a cikkek szélesebb körű, angol nyelvű közzétételét.

Az Archeometriai Műhely nemzetközi elismertségét tükrözi az is, hogy ma már a Q2-es minősítésű folyóiratok közé tartozik a Scimago Journal Rank (SJR) szerint. Ez azt jelenti, hogy a Műhelyt nemzetközi szinten a középkategóriába tartozó, de kiemelkedő minőségű folyóiratként tartják nyilván.

Archaeometry Workshop (Archeometriai Műhely) celebrated its 20th anniversary in 2024, which provides a special opportunity to look back on the history, development and achievements of the journal. It has grown into a nationally leading, internationally recognized journal in archaeological science, providing publication opportunities for the scientific community in Hungarian and English.

Archaeometry Workshop was launched in 2004, at a time when digital publication forms were still in their infancy in Hungary. The journal was founded by the need to promote the science of archaeometry both nationally and internationally. Although the Workshop was primarily established to present the main results of Hungarian archaeometric research, it has been opened to the international community from the very beginning. The main goal of the editorial board was to make scientific results quickly and widely available. To ensure professional quality, a rigorous peer review process was implemented, which has remained one of the journal's strengths to this day.

An important milestone in the development of the journal was its inclusion in the international academic database, the Scopus. This achievement increased the visibility of the journal and significantly contributed to the recognition of the scientific value of the published articles (**Fig. 1**). Inclusion in Scopus claimed the fulfilment of strict quality requirements, which included the compliance with publication standards, the involvement of international authorship, and the increasing publication of articles in English.

Archaeometry Workshop's international recognition is also reflected in the fact that it is now a Q2-rated journal according to Scimago Journal Rank (SJR). This means that it is registered as an intermediate level, high-quality journal. Reaching this rank, the Workshop is a defining communication platform for heritage science in Central Europe.

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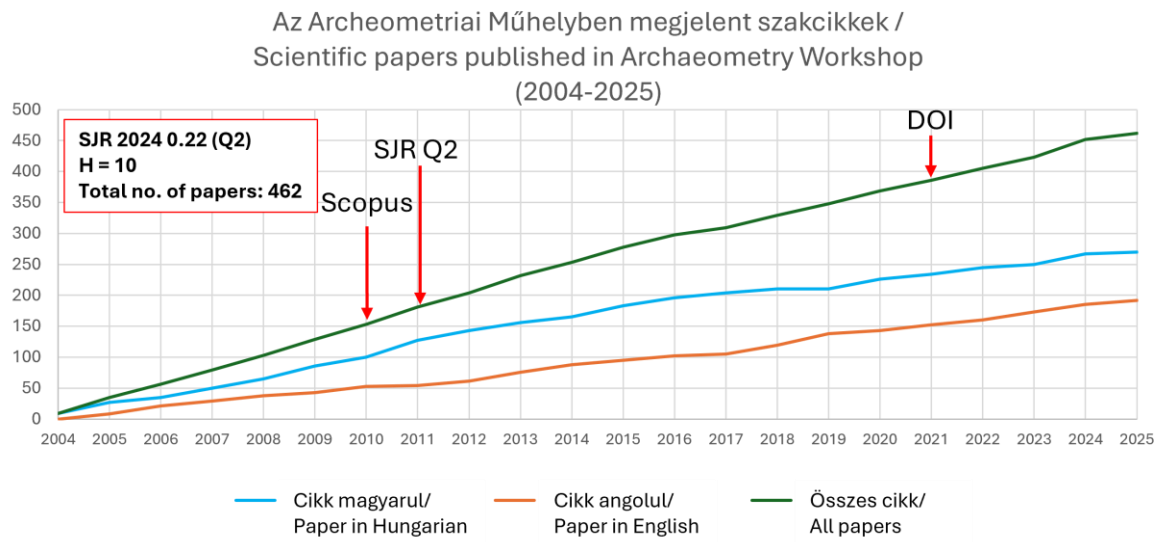

1. ábra: Az Archeometriai Műhelyben megjelent szakcikkek számának alakulása az idő függvényében (2004-2025), néhány folyóiratmetrikai szempontból fontos esemény jelölésével

Fig. 1.: The number of scientific papers published in Archaeometry Workshop by time (2004-2025), with the indication of some issues that are important from the point of view of journal metrics

Így Közép-Európában az örökségtudományi kutatások egyik meghatározó közlési platformja.

A szerkesztőbizottság az elmúlt 20 év során számos változáson ment keresztül, de mindig sikerült megőriznie azt a szakmai színvonalat, amelyet az induláskor kitűzött. Az egyes tagok hozzájárulása jelentős mértékben formálta a folyóirat arculatát és irányvonalát. Az alapítók közül sokan máig aktív résztvevői a folyóirat munkájának, miközben az újabb generációk is bekapcsolódtak, friss szemlélettel gazdagítva a kiadványt. A szerkesztőbizottság különösen nagy figyelmet fordít a fiatal kutatók bevonására, ezzel is támogatva a tudományos közösség folyamatos megújulását. A folyóirat mára egy olyan platformmá vált, ahol a kezdő és tapasztalt kutatók egyaránt bemutathatják eredményeiket, biztosítva a tudásmegosztás és a szakmai fejlődés lehetőségét.

A folyóirat örök motorja az alapító főszerkesztő, T. Biró Katalin. Kati álma és szakmai igénye valósult meg a Műhely létrejöttével és azóta is fáradhatatlanul tevékenykedik a zökkenőmentes fenntartás és fennmaradás nem mindig egyszerű folyamatában. Hálásak vagyunk, hogy a teremtő gondolat mellett támogató társakat, intézményi és időnként anyagi hátteret is szerzett, és a tudományterületek kutatóinak, szerzőinknek lelkes érdeklődésének fenntartásával immár 20 éve tördeli cikkeinket. Az Ő közbenjárásával válhatott a Műhely a Magyar Régészeti és Művészettörténeti Társulat szakosztályává.

Ünnepelni sokféle módon lehet. Az Archeometriai Műhely megalapításának 20. évfordulójáról a magyarországi archeometriai kutatás ünneplésével

The editorial board has undergone many changes over the past 20 years, but has always managed to maintain the professional standards set at the time of its launch. The contributions of individual members have significantly shaped the image and direction of the journal. Many of the founders are still active participants in the journal's work, while members of the younger generation have also joined in, enriching the publication with a fresh perspective. The editorial board pays particular attention to involving young researchers, thus supporting the continuous renewal of the scientific community. The journal has now become a platform where both early-career and experienced researchers can present their results, providing opportunities for knowledge sharing and professional development.

The eternal driving force of the journal is its founding editor-in-chief, Katalin T. Biró. Kati's dream and professional aspirations were realized with the establishment of the Workshop, and she has been tirelessly working ever since in the not always easy process of its smooth maintenance and survival. We are grateful that in addition to her creative idea, she has also gained supportive colleagues, institutional and sometimes financial support, and by maintaining the enthusiastic interest of researchers, our authors, in the fields of science, she has been editing our articles for 20 years. Due to her intercession, the Workshop became a department of the Hungarian Society of Archaeology and Art History.

There are many ways to celebrate. We commemorated the 20th anniversary of the founding of the

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emlékeztünk meg. Ahogyan tíz évvel korábban (Ilon 2014), úgy most is elismert külföldi kutatók meghívásával emeltük az ünnep fényét. Az előadók – részben e kötet cikkeinek szerzői – olyan kutatási eredményeket mutattak be, amely valamilyen módon – témájában, a vizsgált leletanyag, az alkalmazott kutatóberendezések vagy laboratóriumok révén, magyar szakértők által – a hazai örökségtudományhoz kapcsolódnak.

Thilo Rehren (The Cyprus Institute, Science and Technology in Archaeology and Culture Research Center) és magyar kutatótársai (Maróti Boglárka, Kasztovszky Zsolt, Len Adél, Kis Zoltán, Káli György, Füzi János†, Kovács Imre, Szentmiklósi László, Szőkefalvi-Nagy Zoltán, Rosta László) az anyagvizsgáló módszerek egy speciális és csak hazánkban megtalálható kombinációját, a HUN-REN EK Budapest Neutron Központ neutronos berendezéseit alkalmazták 19. századi orosz platinaérmék anyagvizsgálatára (Maróti et al. 2025, ebben a kötetben). *Žiga Šmit* (Univerza v Ljubljani, Fakulteta za matematiko in fiziko; Institut "Jožef Stefan") szerzőtársával (Šmit & Šemrov 2025, ebben a kötetben) a Szlovén Nemzeti Múzeum gyűjteményében fellelhető, 11–13. századi magyar pénzérmék anyagvizsgálatával kapcsolódott a hazai kutatáshoz. *Wayne Powell* (Brooklyn College, CUNY Department of Earth and Environmental Sciences) az Alföld közösségeinek az európai késő bronzkori önkereskedelemben betöltött szerepéről számolt be az ónizotóp adatok tükrében. *Alessandra Giumlia-Mair* (AGM Archeoanalisi, Merano) és Bartus Dávid (ELTE BTK Régészettudományi Intézet) a Magyar Nemzeti Múzeum római bronzszobrainak hosszú múltú konzerválási tapasztalatait és következményeit kutatja (Giumlia-Mair & Bartus 2025, ebben a kötetben). *Beate Maria Pomberger* (Naturhistorisches Museum Wien, Prähistorische Abteilung; Universität Wien, Institut für Urgeschichte und Historische Archäologie) és kutatótársai a kaposvári múzeum gyűjteményében található avar kori csörgők és csengők átfogó (tipológiai, anyagösszetételi, akusztikai) vizsgálatával foglalkozott. A nemzetközi kutatógárda magyar résztvevői (Mozgai Viktória, Bajnóczi Bernadett) révén ez a publikáció (Pomberger et al. 2025, ebben a kötetben) nemcsak témájában, hanem a szakértelem és infrastruktúra tekintetében is képviseli a hazai archeometriai kutatást. *M. Isabel Dias* (Universidade de Lisboa, Instituto Superior Tecnico, Campus Tecnológico e Nuclear) a nukleáris analitikai módszerek örökségtudományi alkalmazásairól tartott előadást, amely témakörben a HUN-REN EK és HUN-REN Wigner Kutatóközponttal egy évtizede fennálló együttműködés keretében számos lelettípus eredetvizsgálatával foglalkozott. *A. J. Timothy Jull* (University of Arizona és HUN-REN Atommagkutató Intézet) a radiokarbon kormeghatározás

Archaeometry Workshop by celebrating archaeometric research in Hungary. As ten years earlier (Ilon 2014), we also celebrated the occasion by inviting renowned foreign researchers. The speakers – some of whom are the authors of the articles in this volume – presented research results that are somehow related to Hungarian heritage science – in terms of their topic, the investigated finds, the research infrastructure or laboratories used, or the Hungarian experts involved.

Thilo Rehren (The Cyprus Institute, Science and Technology in Archaeology and Culture Research Center) and his Hungarian research colleagues (Boglárka Maróti, Zsolt Kasztovszky, Adél Len, Zoltán Kis, György Káli, János Füzi†, Imre Kovács, László Szentmiklósi, Zoltán Szőkefalvi-Nagy, László Rosta) used a special and exclusively Hungary-related combination of material testing methods, the neutron-based facilities of the HUN-REN EK Budapest Neutron Center, to investigate 19th-century Russian platinum coins (Maróti et al. 2025, in this volume). *Žiga Šmit* (University of Ljubljana, Faculty of Mathematics and Physics; Jožef Stefan Institute) and his co-author (Šmit & Šemrov 2025, in this volume) have joined the research by examining the material of 11th–13th century Hungarian coins kept in the National Museum of Slovenia. *Wayne Powell* (Department of Earth and Environmental Sciences, Brooklyn College, CUNY) reported on the role of Hungarian Great Plain's communities in the Late Bronze Age European tin trade in light of tin isotope data. *Alessandra Giumlia-Mair* (AGM Archeoanalisi, Merano) and Dávid Bartus (ELTE BTK Institute of Archaeology) research the long-standing conservation experiences and consequences of Roman bronze statues of the Hungarian National Museum (Giumlia-Mair & Bartus 2025, in this volume). *Beate Maria Pomberger* (Natural History Museum Vienna, Prehistoric Department; University of Vienna, Institute for Prehistory and Historic Archaeology) and her fellow researchers are engaged in a comprehensive (typological, material compositional, acoustic) examination of Avar period bells and pellet bells found in the museum at Kaposvár. Thanks to the Hungarian participants of the international research team (Viktória Mozgai, Bernadett Bajnóczi); this publication (Pomberger et al. 2025, in this volume) represents Hungarian archaeometric research not only in terms of its topic, but also in terms of expertise and infrastructure. *M. Isabel Dias* (University of Lisbon, Instituto Superior Tecnico, Campus Tecnológico e Nuclear) gave a lecture on the application of nuclear analytical methods in heritage science, on which she dealt with the provenance analysis of numerous types of artifacts within the framework of a decade-long collaboration with HUN-REN EK and HUN-REN Wigner Research Center. *A. J. Timothy Jull* (University of

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múltjáról, jelenéről és jövőjéről adott összefoglalót. *Elisabetta Starnini* (Università di Pisa, Dipartimento di Civiltà e Forme del Sapere) évtizedek óta dolgozik együtt magyar kutatókkal (Szakmány György, Horváth Ferenc) magyarországi, a neolitikumra koncentráló régészeti problémákon. Ennek a több évtizedes ismeretszerzésnek egy adott nézőpontú összegzését adja cikkében (Starnini 2025, ebben a kötetben). *Antonín Přichystal* (Masarykova Univerzita, Brno, Ústav geologických věd) szintén több évtizede működik együtt hazai kutatókkal őskori kőeszközök anyagvizsgálatában. Ez alkalommal a Csehországban fellelhető magyarországi kőnyersanyag típusokkal foglalkozott. *Sztáncsuj Sándor* (sepsiszentgyörgyi Székely Nemzeti Múzeum, Régészeti Osztály) a délkelet-erdélyi rézkori közösségek pattintott kőeszközein hazai kutatókkal végzett nyersanyageredet vizsgálatok eredményeiről számolt be.

A rendezvény szervezési és az Archeometriai Műhely 2025/XXII./3 számának nyomtatási költségeit a Nemzeti Kulturális Alap (NKA) A2034/N0078 sz. pályázata és a Magyar Régészeti és Művészettörténeti Társulat biztosította. A helyszín az MNM KK Magyar Nemzeti Múzeum támogatásának köszönhető.

A 20 éves jubileum nemcsak a múlt sikereinek és mérföldköveinek megünneplésére ad lehetőséget, hanem arra is, hogy a jövőbe tekintsünk. Az Archeometriai Műhely továbbra is előtérbe helyezi a tudományos minőséget és a nemzetközi közösséggel való aktív kapcsolattartást. A digitalizációs trendek, az open access publikációs formák előretörése, valamint az újabb kutatási területek integrálása mind-mind hozzájárulhatnak ahhoz, hogy a folyóirat a jövőben is meghatározó szerepet játsszon az örökségtudományban.

Ezúton is köszönetet mondunk minden szerzőnek, lektornak, olvasónak és támogatónak, akik részesei voltak az Archeometriai Műhely 20 éves sikertörténetének. A következő évtizedekben is számítunk az Önök közreműködésére és támogatására!

Irodalom

ILON, G. (2014): 10 éves az Archeometriai Műhely (AM). *Archeometriai Műhely* **XI/1** 77–80.

Arizona and HUN-REN Institute for Nuclear Research) presented a summary of the past, present and future of radiocarbon dating. *Elisabetta Starnini* (University of Pisa, Department of Civilizations and Forms of Knowledge) has been working with Hungarian researchers (György Szakmány, Ferenc Horváth) for decades on Hungarian archaeological problems focusing on the Neolithic. The author provided a summary of this decades-long knowledge acquisition from a specific perspective in her article (Starnini 2025, in this volume). *Antonín Přichystal* (Masaryk University in Brno, Department of Geological Sciences) has also been collaborating with Hungarian researchers for decades on the material analysis of prehistoric stone tools. This time, he focused on Hungarian lithic raw materials on prehistoric sites in the Czech Republic. *Sándor Sztáncsuj* (Székely National Museum at Sfântu Gheorghe, Department of Archaeology) reported on provenance studies of Copper Age lithic industry from Southeastern Transylvania fulfilled in collaboration with Hungarian researchers.

The organizing costs the event and printing the 2025/XXII./3 issue of the Archaeometry Workshop were covered by the National Cultural Fund of Hungary (application No. A2034/N0078) and the Hungarian Society of Archaeology and Art History. The venue was provided by the MNM KK Hungarian National Museum.

The 20th anniversary is not only an opportunity to celebrate the successes and milestones of the past, but also to look ahead to the future. The Archaeometry Workshop continues to prioritize scientific quality and active engagement with the international community. Digitalization trends, the advancement of open access publication formats, and the integration of new research areas can all contribute to the decisive role of our journal in heritage science in the future.

We would like to thank all our authors, reviewers, readers, and supporters who have been part of the 20-year success story of the Archaeometry Workshop. We look forward to your collaboration and support in the coming decades!

Literature

ILON, G. (2014): 10 éves az Archeometriai Műhely (AM). *Archeometriai Műhely* **XI/1** 77–80.

INTERDISCIPLINARY RESEARCH INTO NEOLITHIC CULTURAL CONNECTIONS AND EXCHANGE NETWORKS IN THE CARPATHIAN BASIN: WHAT STONE ASSEMBLAGES CAN REVEAL

INTERDISZCIPLINÁRIS KUTATÁS A KÁRPÁT-MEDENCEI NEOLITIKUM KULTURÁLIS KAPCSOLATAI ÉS CSEREHÁLÓZATAI TÁRGYÁBAN: MIT TÁRHATNAK FEL A KŐESZKÖZ LELETEGYÜTTESEK?•

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Abstract

This paper summarizes the topics, steps, methods and results of an interdisciplinary research project started in Hungary in the 1990s. Its aim was to investigate the use, provenance and circulation of raw materials in the Carpathian basin during the Neolithic to obtain data to be compared with those of the Po River basin in northern Italy. The hypothesis was to test whether comparable geographic conditions could give rise to similar patterns of exploitation, use and circulation of raw materials during the Neolithic, since they both are large alluvial plains surrounded by high mountain ranges.

Archaeometry has proven to be a powerful means for highlighting the existence of complex interaction networks between even very distant areas, such as the Alpine arc and the Hungarian plain or the Carpathian Basin and the southern Balkans. These networks of exchange are interpreted as multi-level, nested networks of peoples, similar to peer polity interaction spheres.

Kivonat

Jelen írás egy Magyarországon a múlt század kilencvenes éveiben indult interdiszciplináris kutatási projekt témáit, lépéseit, módszereit és eredményeit foglalja össze. A cél az volt, hogy megvizsgáljuk a Kárpát-medencében a neolitikum idején a nyersanyagok felhasználását, eredetét és forgalmát, és az így nyert adatokat összehasonlítsuk a Pó folyó vízgyűjtő területével Észak-Olaszországban. Előzetes célunk annak vizsgálata volt, vajon a hasonló földrajzi viszonyok hasonló mintákat eredményeznek-e a nyersanyagok kitermelésében, használatában és forgalmában a neolitikum során, mivel mindkét terület nagy kiterjedésű alluviális síkság, amelyet magas hegyláncok vesznek körül.

Az archeometria hatékony eszköznek bizonyult arra, hogy még nagyon távoli területek, például az Alpok és az Alföld, vagy a Kárpát-medence és a dél-balkáni területek közötti komplex kölcsönhatási hálózatok létezését is megvilágítsa. Ezek a cserehálózatok az "egyenrangú politikai interakciós szférákhoz" hasonló, különböző népek többszintű, egymásba ágyazott hálózataiként értelmezhetők.

KEYWORDS: ARCHAEOMETRIC ANALYSES, RAW MATERIALS, SOCIAL NETWORKS, OBSIDIAN, CARPATHIAN BASIN

KULCSSZAVAK: ARCHEOMETRIAI ELEMZÉSEK, NYERSANYAGOK, SZOCIÁLIS HÁLÓZATOK, OBSZIDIÁN, KÁRPÁT-MEDENCE

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Introduction

This paper summarizes the topics and steps of an interdisciplinary research started in the ‘90s of the last century by the author and her Hungarian colleagues. The aim of the research was to investigate the circulation of raw materials in the Carpathian basin in order to obtain data comparable with the Po River basin, in northern Italy. In fact, these are two large alluvial plains, crossed by important river routes and surrounded by mountain ranges. The hypothesis was to test whether comparable geographical conditions could give rise to similar patterns of use and circulation of raw materials during the Neolithic.

Featured topics at the beginning of the research were Neolithic stone raw materials and early ceramic production traditions (Szakmány & Starnini 1996; 2002; 2007; Szakmány et al. 2005; 2006). This paper will summarize in brief only the main results obtained in the last decades from the archaeometric study of lithic raw materials employed during the Neolithic period in Hungary.

The polished stone tools

The scientific study of raw materials employed for the production of prehistoric polished and ground stone tools has been performed pursuing the use of non-destructive or mini-invasive archaeometric analyses (Biró 2024). The analytical methods involved macroscopic and stereomicroscopic observations (magnification range ×10–100), petrographic thin sections sampling small parts in the fractures of broken artefacts, Magnetic susceptibility (MS) (Bradák et al. 2009), SEM-EDX in a non-destructive mode (detailed mineral chemical analyses and textural investigation) (Bendő et al. 2013; 2014), PGAA (Szakmány et al. 2011; determine the average bulk concentrations of major and some minor elements) as the various methods were tested and developed by archaeometrists. More recently, an important analytical method has been developed for the polished stone tools for non-destructive SEM-EDX analysis on the original surface of the artefacts (Bendő et al. 2019). The sample is prepared for investigation by wrapping in aluminium foil. The latter has a window, and the sample is carbon-coated for the investigation only on the small area of interest, thus limiting the surface to be treated. Systematic analysis of several polished stone tool collections from Hungarian Neolithic sites revealed the presence of mixed jadeitite and Fe-mixed jadeitite artefacts of northwestern Italian Alpine origin in several sites, such as Alsónyék, Zengővárkony, Zirc, Hódmezővásárhely-Gorzsa, Szombathely-Olad plateau (Biró et al. 2003; Biró et al. 2017; Starnini et al. 2015; Váczi et al. 2017). The distance between the sources of origin and the discovery sites is over 1200 km as the crow flies (Bendő et al. 2019: Fig. 9). Medium-distance connections at the beginning of the V Millennium BC have been revealed with the archaeometric analyses of several early, middle and late Neolithic polished stone tools from important sites such as Bicske-Galagonyás, Méhtelek-Nádas, Ecsegfalva 23, Szarvas, Endrőd, Tápé-Lebő, Hódmezővásárhely-Gorzsa, and Pitvaros (Starnini & Szakmány 2000; Starnini et al. 2007). Archaeometric analyses coupled with geological surveys eventually identified sources of hornfels located in the Rusca and Apuşeni Mts. (Szakmány et al. 2016). The analyses of artefacts from Hungarian sites revealed also the presence of polished stone tools made of the so-called “white stones” (magnesite bearing siliceous rocks and others: Starnini & Szakmány 2021; Szilágyi et al. 2025) whose sources are to be found to the south, beyond the Carpathians (Antonović 1997, Fig. 1). In summary, the study of raw materials employed for the production of polished and ground stone tools revealed the existence of complex connecting networks in the Carpathian Basin during the whole Neolithic period.

Siliceous rocks

Siliceous rock sources employed for making knapped stone tools in the Carpathian Basin and surrounding regions have been investigated since long by K.T. Biró (2024 and literature therein), leading to the creation of an important raw material collection stored at the Hungarian National Museum, Lithotheca (Biró & Dobosi 1991; Biró et al. 2000). One of the most important siliceous rock sources are the Bakony Mountains, located north of Lake Balaton where extensive outcrops of radiolarites or radiolarian cherts are present (Szilasi 2017). Radiolarite of every colour, from brick-red to mustard yellow and deep brown, has been observed in almost all the territory extending from Bakonybél to Szentgál and the environs of Bakonycsernye or Sümeg. The raw material can be found all over a huge area of the Bakony Mountains since the radiolarian chert platforms are not impounded into micro-areas. Thick chert seams extend from Bakonycsernye up to the Zala Basin. All the radiolarian chert blocks, platforms or nodules can be easily located on the eroded surface and could have been easily mined in prehistory. Raw material can be collected also from the eroded slopes or on the banks of seasonal streams. No deeper mining knowledge was required to dig out these radiolarite blocks. The study of the variations in radiolarian cherts from the Bakony Mountain have been the object of several studies. The first attempts at fingerprinting the different variants of these rocks have been made at the end of the last century. However, these attempts were sporadic and destructive employing different methods: OES (Kozłowski et al. 1981), WS, OER, IR, XRD and NAA in the ‘80s (Biró & Pálosi 1986), NAA in the

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‘90s (Biró & Dobosi 1991; Varga 1991). Later, at the beginning of this century, with the improvements of the archaeometric techniques, attempts with non-destructive methods have been made by PIGE-PIXE (Biró et al. 2002), and within the frame of a TéT Project experimental examination with the non-destructive PGAA method have been performed (Biró et al. 2009). More recently a new study employing CRF X-ray analytical field instrument has been published (Szilasi 2017). This device is equipped with an analogue X-ray tube which bombs the surface with high intensity beams with the help of an optimized drift detector. The length of the reflected waves shows the presence and the quantity of the different elements. The range of the impacts gives the rate between the elements. The results showed that radiolarian cherts from the Jurassic Period are almost identical throughout Transdanubia, from Gerecse Mountain to Zala Basin, thus making their separation a challenge. This new evidence suggests that it is better to use the general name of Bakony radiolarites for all the raw materials from this area (Szilasi 2017, Fig. 9). The same author suggests that designations of smaller groups like Szentgál-type, Úrkút-Eplény-type, Hárskút-type, etc., which were identified mostly by their colours, should no longer be used. In fact, all colour variations can be found in each and every geological source and it is impossible to define them even by geochemical examinations.

The source of another important siliceous raw material circulating at long distance in the Carpathian Basin during the early Neolithic, the Balkan Flint, was identified. It was not located in the territory of Romania as originally supposed, but along the right bank of the Danube River in the Pre-Balkan platform chalky limestone formations in present Bulgaria (Biagi & Starnini 2012).

The study of the distribution of Carpathian Obsidian

Carpathian obsidian sources and distribution have been characterized and investigated since long (Biró 1984; 2018; Williams-Thorpe et al. 1984). However, due to the invasive character of the earlier analytical method and their rather expensive costs, rarely the whole obsidian artefact assemblage from a site has been sourced. Recently, the introduction of pXRF devices (Frahm 2014) allows the non-destructive analysis of a large numbers of obsidian samples and facilitate the analyses of the assemblages since it is no longer necessary to move the artefacts from the Museums to the laboratories (Glascock et al. 2015; Tykot 2018; Riebe 2019; Bonsall et al. 2024; Starnini et al. 2024). Moreover, the new holistic approach is to publish not only the provenance result of each single piece, but also its typological and technological description, possibly accompanied by photographs and use wear analysis. In this case we are also able to evaluate the mode of distribution and circulation of this raw material and the way it was employed.

Merging the study of raw materials for polished, ground and knapped stone tools

A multidisciplinary, raw material sourcing approach to the whole stone assemblage, i.e. knapped, polished and ground tools, has been applied in the study-case of the Late Neolithic tell settlement of Hódmezővásárhely-Gorzsa (Szakmány et al. 2009; Starnini et al. 2015; Miklós et al. 2024), employing different archaeometric technologies, including new non-destructive chemical analyses for the raw material analyses.

The results achieved till now showed that a complex network of territorial, cultural and social interactions was active at Gorzsa and in the Carpathian Basin during the entire Late Neolithic period (**Fig. 1.**), and that the river courses probably acted as main routes for these connections.

For the siliceous rocks, the study of the distribution of Carpathian obsidian and Balkan flint revealed long-distance networks connecting the northern part of the Carpathian Basin, the southern Balkans and the north Aegean basin (**Fig. 2.**) starting from the early Neolithic (Kilikoglou et al. 1996; Özbek & Erdoğu 2014; Milić 2016; Biagi et al. 2023).

Discussion and conclusions

In recent decades, giant strides have been made in the analysis of the provenance of prehistoric artefacts. It is now possible to attempt to make a synthesis and provide a social meaning to the information and results so far obtained, in the attempt at overpassing the simple archaeometric result. The patterns we can observe in the archaeological record revealed by the raw material provenance studies may be seen as a testimony to exchanges that are essentially cultural and social rather than economic. The exchange-networks involved the transfer of materials, artefacts and goods, though their values were as information, shared ideas, common beliefs and converging symbolic representation. In the Carpathian Basin, during the Neolithic period we have notable evidence of a wide-area cultural networking through the exchange of goods and materials and the sharing of cultural behaviours. The late Colin Renfrew (1986) described the phenomenon as "*peer polity interaction*". Renfrew argued that ‘polities’, representing socio-political realities, rather than ‘cultures’, should be the basic units with which we investigate prehistoric archaeology, and tell the story of prehistory. Moreover, we have to think about the exchange also of peoples among different communities, since artefacts, goods and materials did not travel by themselves. Ancient DNA (aDNA)

Fig. 1.: Long and medium-distance connections revealed by lithic raw material sourcing in the Carpathian Basin and at Gorzsa during the Late Neolithic, letters: A= Monviso and Western Alps; B=Voltri Massif; C=Oligocene Conglomerate series and redeposited gravels/pebbles (maps elaborated from Bendő et al. 2019, left, and Szakmány et al. 2008, right).

1. ábra: Kőzetnyersanyagok forrásterületeinek meghatározásával feltárt nagy- és közepes távolságú kapcsolatok a Kárpát-medencében és Gorzsán a késői neolitikumban (betűjelek: A= Monviso és Nyugati-Alpok; B= Voltri Masszívum; C= oligocén konglomerátumösszlet és áthalmozott kavics). Térképek átdolgozva Bendő et al. 2019 (bal oldal) és Szakmány et al. 2008 (jobb oldal) alapján.

and isotopic analyses are now proving the mobility of individuals during the prehistoric period (Haak et al. 2010; Depaermentier et al. 2020). T. Watkins explaining his social network theory (Watkins 2003; 2008) underlined the expansion and intensification of a socio-cultural interaction sphere, with more and more communities "buying into" the networks with the passage of time. He put two questions: Why did communities join such networks of interaction? What were the perceived advantages? The answers may relate to socio-political needs, cultural imperatives, social relationships implying exchange/transfer of peoples. When possible, aDNA and isotopes analyses can demonstrate these transfers directly. However, raw materials identification, sourcing and the study of their distribution in the territory may reveal indirectly people's transfers and mobility as well. And in any case are important complementary data. For early prehistory, specialists on lithic industries usually refer to knapped stone traditions, as well as styles of formal tools such as projectile points, often with little consideration given to what a shared chipped stone industrial tradition might mean in social and cultural terms, how small groups spread thinly over a landscape maintain such common traditions, or indeed why. In the Neolithic, we find implicit reference to the communities that are represented by the settlement sites that we have excavated, and we continue to find references to technological traditions of knapped stone and ceramic manufacture, or ceramic decoration. However, in constructing narratives that synthesize, integrate and make comprehensible the archaeological data across individual sites, and at a higher level than shared chipped stone and ceramic traditions, we have continued to use models derived from the notion of the 'cultural group' defined nearly 100 years ago by V. G. Childe (1929). Childe's idea that there were blocks of time and definable areas on the map within which everyone shared common cultural beliefs, knowledge and practice worked neither in our own experience of the modern western world, nor in the simple, small-scale societies explored by ethnographers and ethno-archaeologists (Renfrew 1977; Shennan 1978; Hodder 1982).

T. Watkins (2008) proposed that, instead of cultural groups, we should think of these networks of exchange as multi-level, nested networks, similar to peer polity interaction spheres. The larger communities would certainly have needed another layer of structuring between the household and the overall community of several thousand people. Thus, we may observe that there are one or two layers of social organization to be inferred below the level of the co-resident community represented in the excavated settlements. According to Watkins (2008) these constitute nested networks of cultural, social and economic interaction. In the negotiation of our personal interactions and relations with one another, we have emotional, material and symbolic resources with which to operate. Turner and Maryanski (1991) identify three circles of personal networks, the most basic of which is the intimate network, represented by our close relatives, those

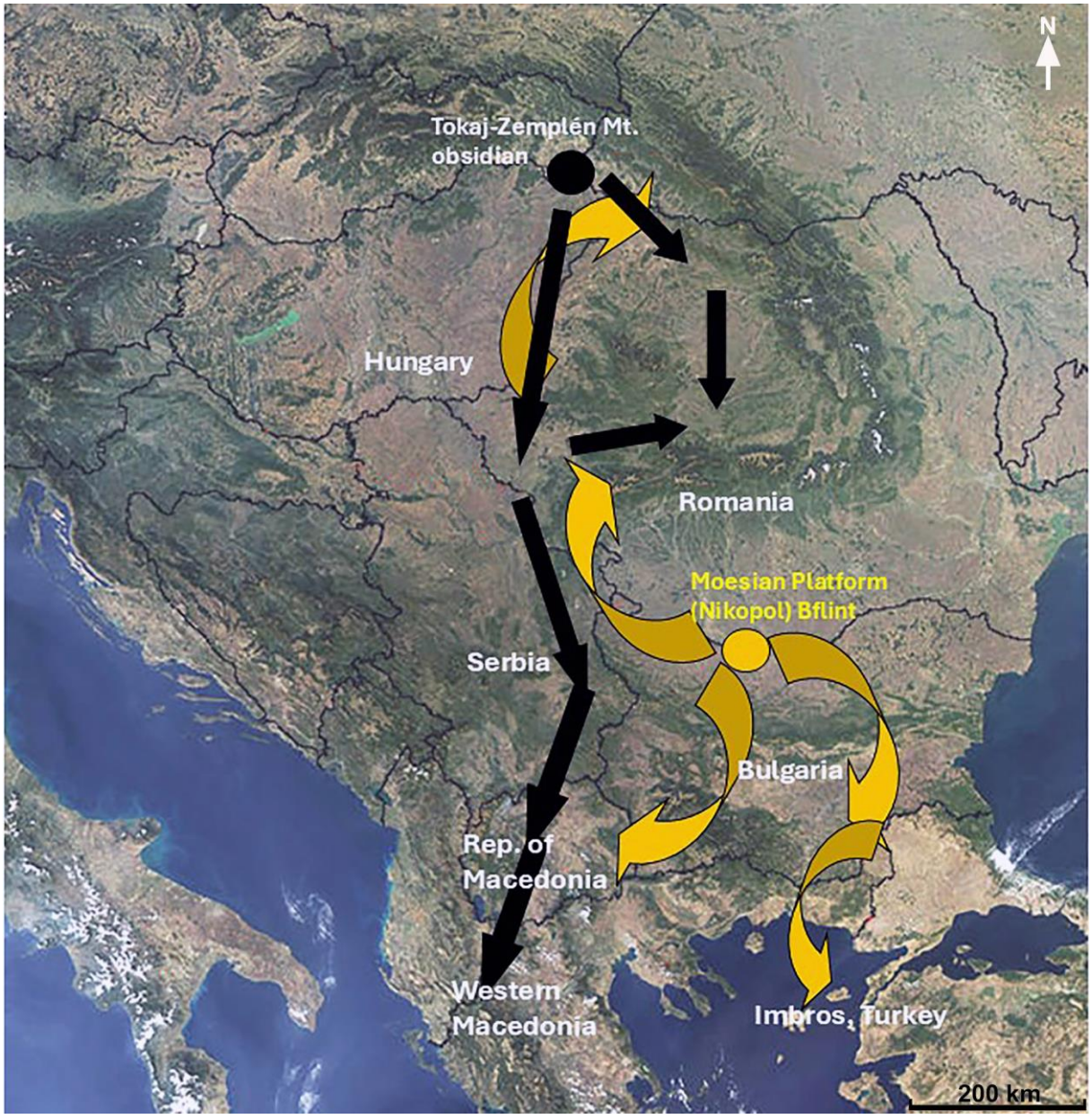

Fig. 2.: Long-distance connections revealed by obsidian and Balkan Flint distribution (map by the Author)
2. ábra: Távolsági kapcsolatok obszidián és balkáni kova régészeti elterjedése alapján (a szerző térképe)

with whom we have day-to-day contact, which they quantify in a group of about 20 people. Instead, an extended network consists of '*acquaintances and friends-of-friends*' (Gamble 1998, 435), a kind of social relationship involving up to 200 people. Beyond the concentric circles of personal networks, there is a global network, people who are 'them' rather than 'us', but with whom an occasional transaction may need to be negotiated, whether at first hand or through an intermediary. This last type of relationship can involve up to over 1000 people. C. Gamble (1999) using network theory to investigate Palaeolithic societies concluded that early Palaeolithic groups operated intimate and effective networks, and that only in the Upper Palaeolithic there is evidence for the operation of extended networks of hundreds of kilometres. What changed in the Neolithic is that the trend towards larger, permanently co-resident settlement communities meant that the intimate networks of each of the members (essentially the close kin group), the effective networks (those 25–40 people with whom each member comes into regular contact), and the extended network (of several hundred people, with whom each member may need to deal from time to time), all came together. According to this model, each individual would have a day-to-day contact with any number of the extended network, and the geographical spread of the extended network was reduced to a few hundred metres rather than tens or

hundreds of kilometres. Actually, the Neolithic settlements were the first modern communities, since they correspond to the anthropologist Anthony Cohen's description of the characteristics of a community (Cohen 1985, 15):

> *Community is that entity to which one belongs, greater than kinship but more immediate than the abstraction we call ''society''. It is the arena in which people acquire their most fundamental and most substantial experience of social life outside the confines of the home ... At the risk of substituting one indefinable category for another, we could say it is where one acquires ''culture''.*

In this perspective, the more sites we have, and the more detail we are given from those sites, the more difficult it becomes to sustain the notion of traditional 'culture-groups'.

In conclusion, thanks to decades of archaeometric research, there are more and more robust archaeological indicators of exchange of goods and materials, and of the sharing of cultural practices in the Neolithic. Beyond thinking of these phenomena as evidence of trade or social exchange, we can see them as surviving indicators of extensive net-working. One of the best documented examples of networking is indeed the exchange of obsidian that we can consider as part of a system of cultural relations (Riebe 2019).

Contribution of authors

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METALLIC IDIOPHONES OF THE AVAR PERIOD FROM SOMOGY COUNTY IN THE COLLECTION OF THE RIPPL-RÓNAI MUSEUM IN KAPOSVÁR

AVAR KORI FÉMIDIOFONOK SOMOGY MEGYÉBŐL A KAPOSVÁRI RIPPL-RÓNAI MÚZEUM GYŰJTEMÉNYÉBEN •

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Abstract

A total of more than 50 pellet bells and bells dating to the Avar period belong to the archaeological collection of the Rippl-Rónai Museum in Kaposvár. They were investigated within the frame of the research project "Metallic Idiophones between 800 BC and 800 AD in Central Europe", in an interdisciplinary and cross-national co-operation between Austria and Hungary. The results, gained through archaeological, archaeometric, acoustic, psychoacoustic methods and experimental textile archaeology are presented here.

Kivonat

A kaposvári Rippl-Rónai Múzeum régészeti gyűjteménye több mint 50 darab avar kori csörgőt és csengőt őriz, amelyeket a "Metallic Idiophones between 800 BC and 800 AD in Central Europe" kutatási projekthez kapcsolódva, Ausztria és Magyarország közötti interdiszciplináris együttműködés keretében vizsgáltunk. Jelen tanulmányban a régészeti, archeometriai, akusztikai, pszichoakusztikai és kísérleti textilrégészeti módszerekkel kapott eredményeket mutatjuk be.

KEYWORDS: BELLS, PELLET BELLS, AVAR KHAGANATE, MUSIC ARCHAEOLOGY, ACOUSTICS, PSYCHOACOUSTIC, ARCHAEOMETRY, TEXTILE ARCHAEOLOGY, EXPERIMENTAL ARCHAEOLOGY

KULCSSZAVAK: CSENGŐK, CSÖRGŐK, AVAR KAGANÁTUS, ZENERÉGÉSZET, AKUSZTIKA, PSZICHOAKUSZTIKA, ARCHEOMETRIA, TEXTILRÉGÉSZET, KÍSÉRLETI RÉGÉSZET

Introduction

The collection of the Rippl-Rónai Museum in Kaposvár keeps approximately 50 pellet bells and bells from the Early Middle Ages. They originate from the Avar cemeteries Kaposvár-Toponár 40-es őrház, Kaposvár-Toponár-Fészerlak-puszta, Kaposvár 61. sz. út, Kaposmérő-Agyagbánya, Zamárdi-Rétiföldek, Vörs-Papkert B, Vörs-Battyáni Disznólegelő, and Kereki-Homokbánya (**Fig. 1.**). These idiophones were investigated in the research project "Metallic Idiophones between 800 BC and 800 AD in Central Europe", which is funded by the Austrian Science Funds FWF and supported by the Natural History Museum Vienna. The scientists' team already investigated idiophones from the Hungarian National Museum Budapest (Pomberger et al. 2022b), the Savaria Museum in Szombathely (Pomberger et al. 2021b)

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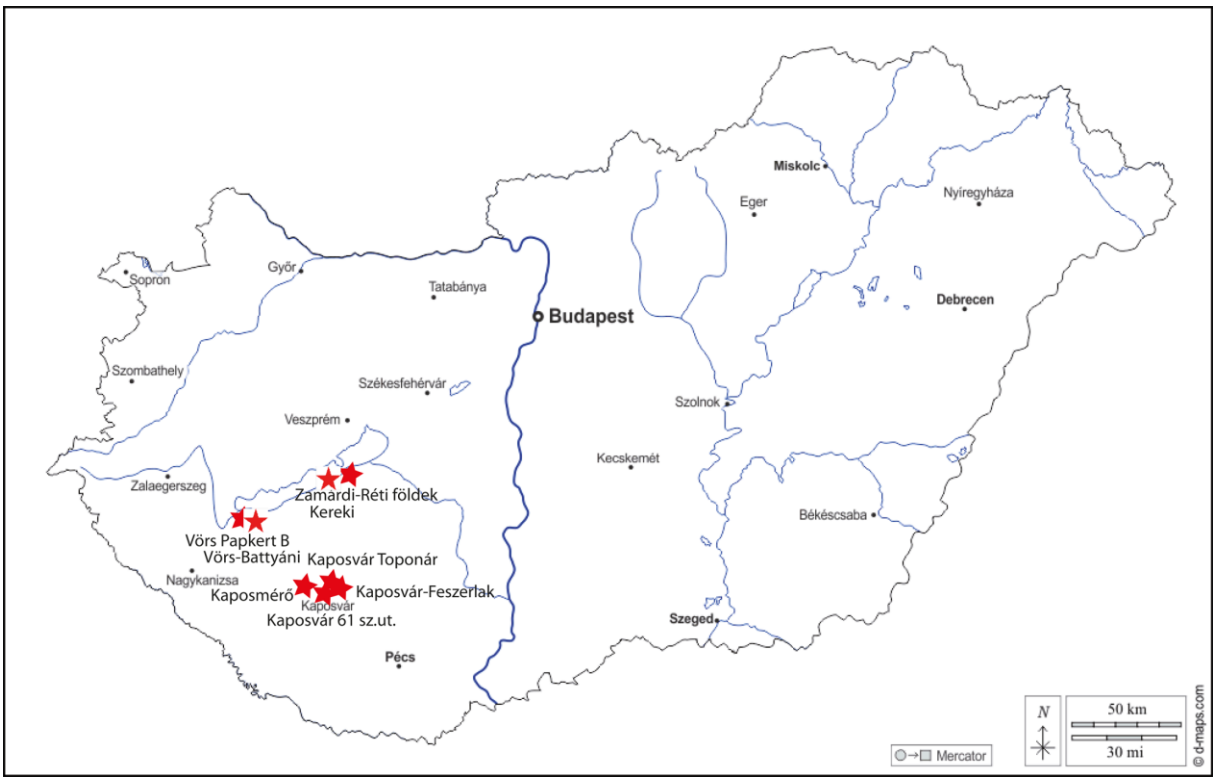

Fig. 1.: Cemeteries with pellet bells from the Avar period in the Somogy county (Graphic: B. M. Pomberger, base map: www.d-maps.com)

1. ábra: Avar kori temetők csörgőkkel Somogy megyében (rajz: B. M. Pomberger, alaptérkép: www.d-maps.com)

and the Balaton Museum in Keszthely (Pomberger et al 2023).

The sites

Only 57 burials in all eight cemeteries together contained idiophones, such as pellet bells and bells, most of them dating to the late Avar period (**Appendix 1**).

2.1. Kaposvár-Toponár, 40-es őrház (Kaposvár FO 33, 40/a), cemetery

The excavations of the cemetery were carried out with interruptions from 1968 to 1981 (Szentpéteri 2002) and in 2003. So long 205 graves, dating to the late Avar period, could be saved (status 2002). (Hungarian National Museum Archaeology Database, 21 June 2023, https://archeodatabase.hnm.hu/en/node/4753)

Ten pellet bells originate from six burials (**Fig. 2.**) (Bárdos 1978). One pellet bell was excavated in each of the girls' graves 18 (cat. 1) and 36 (cat. 2). Another one was found in the child grave 52 (cat. 6) and two in the girl's grave 43 (cat. 3 and 5). One originates from the woman's grave 48 (cat. 5). All these pellet bells lay near the left femurs and are cast in copper alloys except the pellet bell from grave 52, which is made from sheet metal. The other four pellet bells (cat. 7–10) were found in grave 57, where a man was buried with his horse. These pellet bells belong to the horse bridle (**Plate 1/1–10**).

2.2. Kaposvár-Toponár-Fészerlak-puszta, cemetery

The cemetery was excavated in several campaigns in 1968, 1970 (Szimonova 1972), from 1974 till 1981 with interruptions, in 2004 and in 2016 (Hungarian National Museum Archaeology Database, 21 June 2023, https://archeodatabase.hnm.hu/en/node/44847).

218 graves are excavated so far (Évinger 2003) and dates to the late Avar period. Two pellet bells are known, one from grave 47 (cat. 11) and a stray find (cat. 12) (**Plate 2/2–3**).

2.3. Kaposvár-61. sz. út

First excavations of the cemetery were carried out in 1979 and 1980. During this campaign 87 burials were unearthed, dating to the 9th century (**Fig. 4.**) (Bárdos 1985), but only grave 82, burial of a child, contained a pellet bell (cat. 13, **Plate 2/1**). The position of this pellet bell in the grave is uncertain.

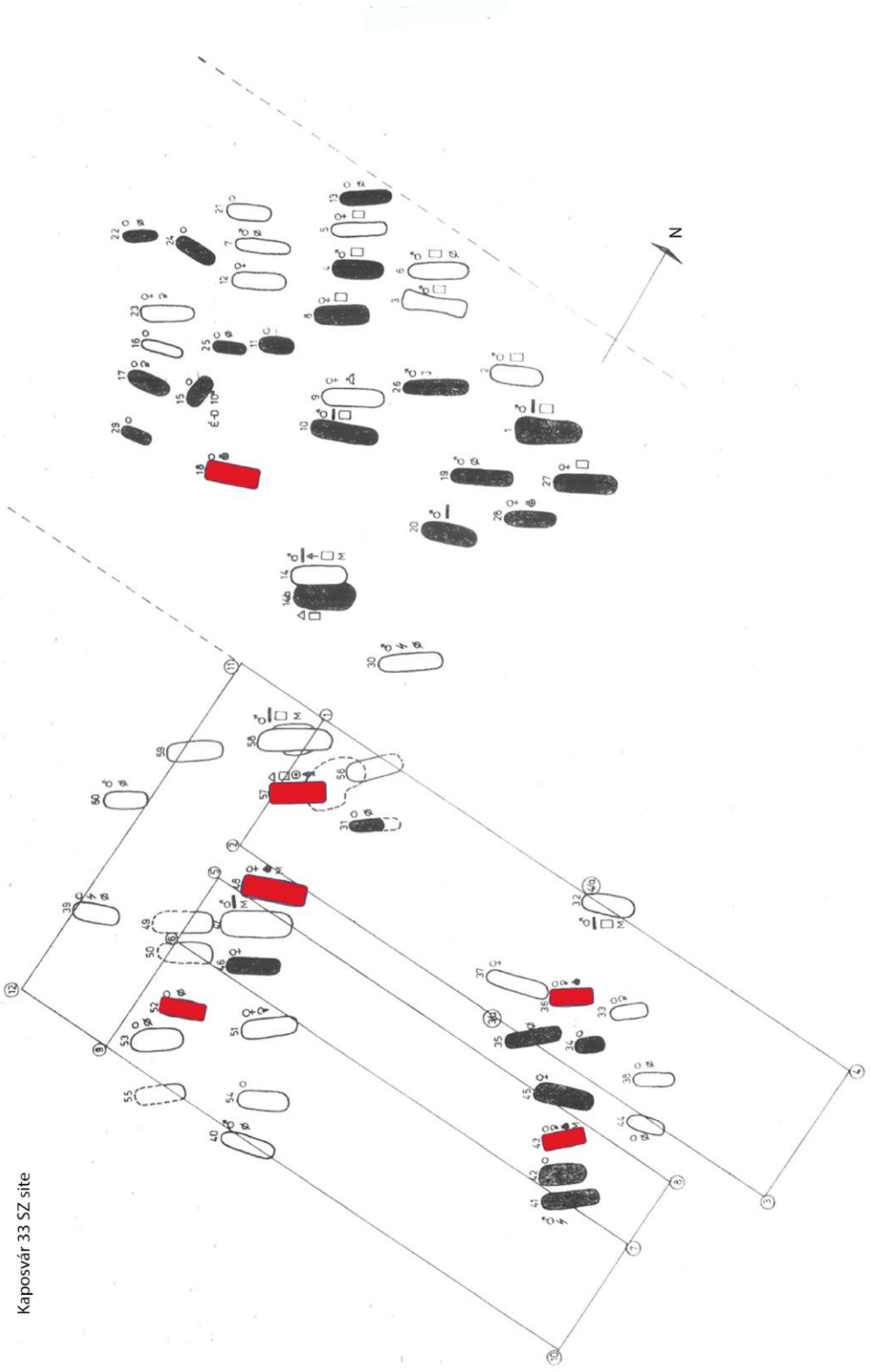

Fig. 2.: Graves with pellet bells from the cemetery Kaposvár Toponár 40-es őrház (according to Bárdos 1978, 30, Fig. 4; Graphic: B. M. Pomberger).

2. ábra: Csörgőket tartalmazó sírok a Kaposvár Toponár 40-es őrház temetőben (Bárdos 1978, 30, 4. ábra nyomán, rajz: B. M. Pomberger)

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2.4. Kaposmérő-Agyagbánya

The cemetery dates to the late Avar period and contained 254 graves, which were excavated in 1980, 1985 and 1989–90 (Szentpéteri 2002). Six pellet bells and one bell were found. One pellet bell (cat. 51) originates from grave 80 (Varga 2016, pl. 6/32), where a horseman with a horse is buried. The pellet bell seems to be part of the horse bridle. Grave 48 contained a double burial – an adult and child. This child was buried with three pellet bells (Varga 2016, 173, pl. 3/2). Another child in grave 231 had one pellet bell (Varga 2016, 168, pl. 3/2). One pellet bell belonged to a woman in grave 19 and a cone shaped bell to the woman in grave 120 (cat. 52) (Varga 2016, 168, pl. 8/10) (see **Plate 2/5–6**).

2.5. Zamárdi-Rétiföldek

The cemetery in Zamárdi-Rétiföldek dates from the 6th to the 9th century CE. So far, 2700 graves are excavated (Balogh 2019). First finds were reported in 1963. During excavations in 1972 and 1987–1998 with interruptions 2368 burials could be saved (Bárdos & Garam 2009, 7–8). 2334 graves contain humans' remains (97%) and 70 (3%) are horse burials (**Fig. 3.**) and those among them containing bells and pellet bells are subject to our investigations. The attires of the buried individuals resemble closely to those of the Merovingian culture, which could be observed in several burials (Garam 2011; Balogh 2019). We also observed that around 1.3% of the burials (35) contained idiophones, divided between 30 individuals' and 5 horses' burials (**Fig. 3.**). In the new-found graves 37 pellet bells, 5 bells and 4 tutuli were excavated. Tutuli are cone-shaped pendant or objects cast on bronze or forged from sheet metal. Pellet bells were found in fourteen women's, nine children's burials (four girls', one boy's, four children's) and one man's burial. The bells originate from two women's, one young girl's (?) and two undefined individuals' burials. Four horse burials contained pellet bells. Four tutuli were unearthed from one burial (**Fig. 4.**).

Pellet bells were part of nine child burials (cat. 16/burial 498; cat. 19/burial 702; cat. 21/burial 1172; cat. 22/burial 1273; cat. 25/burial 1685; cat. 26/burial 1688; cat. 27/burial 1689; cat. 33/burial 1878; cat. 37/burial 1904). Thirteen women burials contained pellet bells as part of their belt ensembles (cat. 14/burial 97a; cat. 23/burial 1304; cat. 28/burial 1711; cat. 30/burial 1874; cat. 31/burial 1832; cat. 34/burial 1885; cat. 41/burial 2099; cat. 42/burial 2125; cat. 43/burial 2275; cat. 45/burial 2308; cat. 46/burial 2349; cat. 47/burial 2352; cat. 48/burial 2357). Three of the bells belonged to women burials (cat. 15/burial 407; cat. 17/burial 517; cat. 32/burial 1872).

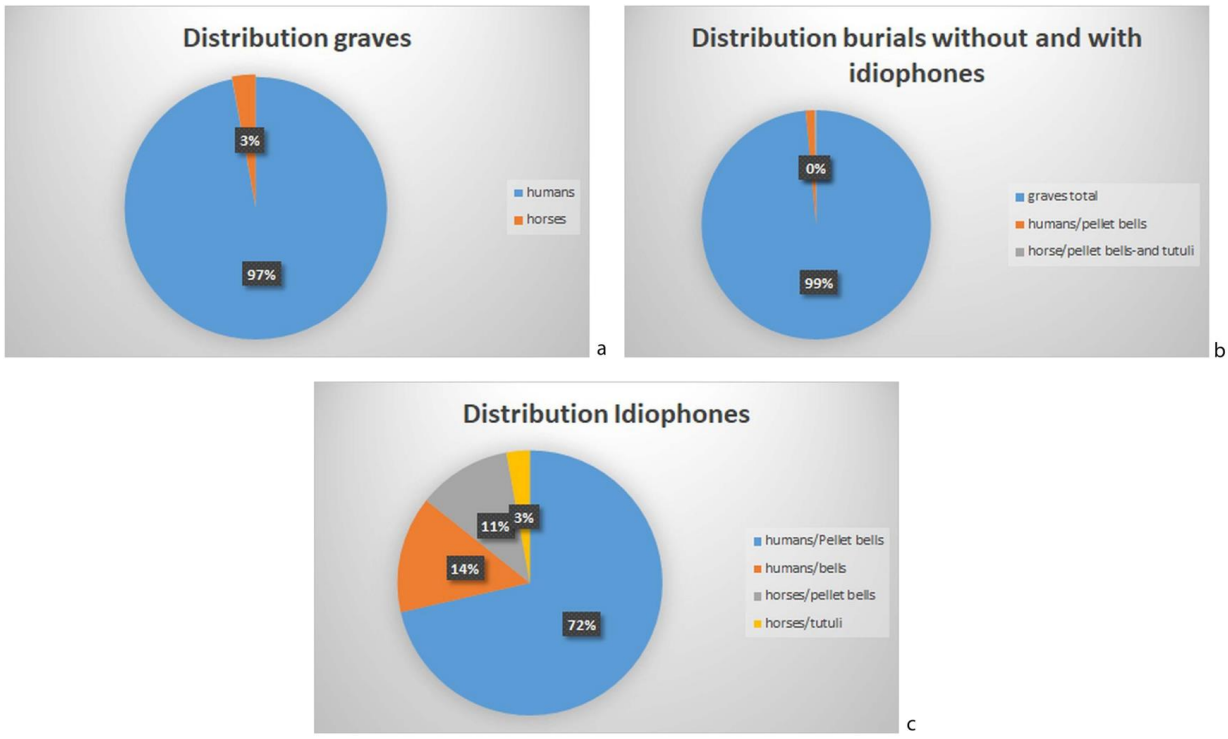

Fig. 3.: Distribution of idiophones in the graves from the cemetery Zamárdi-Rétiföldek (Graphic: B. M. Pomberger).

3. ábra: Az idiofonok eloszlása a Zamárdi-Rétiföldek temető sírjaiban (rajz: B. M. Pomberger)

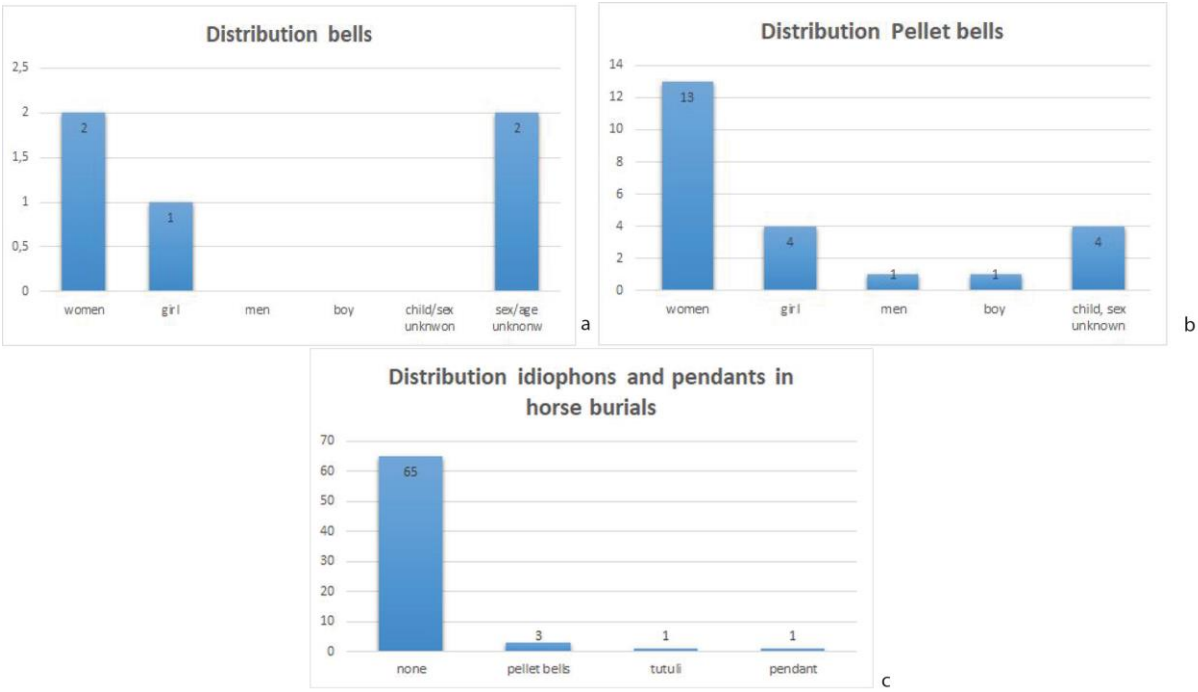

Fig. 4.: Distribution of bells and pellet bells in human and horse graves from the cemetery Zamárdi-Rétiföldek (Graphic: B. M. Pomberger).

4. ábra: A csengők és a csörgők eloszlása az ember- és lósírokban a Zamárdi-Rétiföldek temetőben (rajz: B. M. Pomberger)

The other two bells were found in burial 614 (cat. 18) and burial 960 (cat. 20). Sex and age of the deceased individuals in these graves are unknown. Only one masculine burial contained a pellet bell (cat. 39/burial 2088) (see **Table 1.**). The horse burial 1900 (cat. 24) outstands with five pellet bells. The grave was disturbed and therefore the original position of the pellet bells is unknown. The three pellet bells of the horse in grave 1653 (cat. 24) were near the neck. Probably the two pellet bells in grave 2283 (cat. 29) were located within the horse's neck area. The one pellet bell from grave 2091 (cat. 40) was found next to the horse's breast. The horse burial 1900 unfortunately is disturbed, but stands out with five pellet bells (cat. 35). Grave 1903 (cat. 36) contained four tutuli made from sheet metal, which were found near the breast of the horse skeleton (see **Plates 3–7**).

2.6. Vörs-Papkert B

The cemetery in Vörs-Papkert B consists of 716 graves. 167 of them can surely be dated to the late Avar period. The rest are from the Early Bronze Age (Kisapostag culture), the 9th-century Carolingian fringe culture, the period of the conquest, as well as archaeologically indeterminate age and empty burial pits. They were unearthed during rescue excavations in 1983–85 and 1987–93 (Szentpéteri 2002). One pellet bell was found in grave 321 (cat. 50; **Plate 2/4**).

2.7. Vörs-Battyáni Disznólegelő

The site contained one grave with pellet bells – so far known to the authors –, dating to the late Avar period. In grave 634 three pellet bells cast in copper alloy and one forged from iron sheet found near the left knee, organic material underneath. Since the pellet bells were below each other with their eyelets upwards, they may have been suspended from a ribbon (László 2012; Költő 2016) (cat. 53; **Plate 9.**). The pellet bells were not available for investigations and sound recordings.

2.8. Kereki-Homokbánya

The cemetery contained 151 graves from the late Avar period. Excavations in Kereki-Homokbánya were carried out in 1973, 1987, 1988 and 2004 (Költő 2005; Költő & László 2013; László 2012) (**Fig. 5.**) (Hungarian National Museum Archaeology Database, 10 June, 2024, https://archeodatabase.hnm.hu/en/node/55885).

Each one bell was found in grave 53 and 104. Pellet bells were excavated in six graves. Two of them were children's burials (48, 115), graves 27, 63, 104/A and grave 120 women's and 109 a man's burial. In grave 120, the burial of a young woman the pellet bell shows displays a face-like decoration (cat. 54–61; **Plates 10 and 11)**. This pellet bell was not available for investigations and sound recordings.

Table 1.: Graves of Zamárdi-Rétiföldek cemetery with bells and pellet bells and their positions (by B. M. Pomberger). RRM = Rippl-Rónai Museum

1. táblázat: A Zamárdi-Rétiföldek temető sírjainak csengői és csörgői, valamint ezek helyzete (a táblázatot összeállította: B. M. Pomberger). RRM = Rippl-Rónai Múzeum

Cat. nr.	Idiophone	Inventory nr.	Site	Grave nr.	Sex/Age	Position
15	bell	RRM 84.224.8	Zamárdi-Rétiföldek	407	woman	in earthfilling
17	bell	RRM---	Zamárdi-Rétiföldek	517	woman?	bell on chain fixed on ornamental disc
32	bell	RRM 247.1.1872.1	Zamárdi-Rétiföldek	1872	young girl	near left knee
18	bell?	RRM 247.1.614.1	Zamárdi-Rétiföldek	614	human	disturbed
20	bell	RRM 247.1.960.3	Zamárdi-Rétiföldek	960	human	disturbed
14	pellet bell	RRM---	Zamárdi-Rétiföldek	97/a	woman	left knee
23	pellet bell	RRM 247.1.1304.3	Zamárdi-Rétiföldek	1304	woman	in earthfilling
28	pellet bell	RRM 247.1.1711.8	Zamárdi-Rétiföldek	1711	woman	near left femur
29	pellet bell	RRM 247.1.1728.12	Zamárdi-Rétiföldek	1728	woman	near left knee
30	pellet bell	RRM---	Zamárdi-Rétiföldek	1874	woman	under left femur
31	pellet bell	RRM 247.1.1832.4	Zamárdi-Rétiföldek	1832	woman	near left knee
41	pellet bell	RRM 247.1.2099.1	Zamárdi-Rétiföldek	2099	woman	right leg
42	pellet bell	RRM ---	Zamárdi-Rétiföldek	2125	woman	near left femur
43	pellet bell	RRM ---	Zamárdi-Rétiföldek	2275	woman	near left knee
45	pellet bell	RRM ---	Zamárdi-Rétiföldek	2308	woman	left femur
46	pellet bell	RRM ---	Zamárdi-Rétiföldek	2349	woman	near left femur
47	pellet bell	RRM 247.1.2352.2	Zamárdi-Rétiföldek	2352	woman	near femurhead
48	pellet bell	RRM 247.1.2357.9	Zamárdi-Rétiföldek	2357	woman	woman, near left knee
33	pellet bell	RRM 247.1.1878.3=247.1.1877.3	Zamárdi-Rétiföldek	1878	girl	near pelvis
34	pellet bell	RRM 247.1.1885.1	Zamárdi-Rétiföldek	1885	girl	near left femur
37	pellet bell	RRM 247.1.1905.4 => burial 1904?	Zamárdi-Rétiföldek	1904	girl	left femur
26	pellet bell	RRM 247.1.1688.9	Zamárdi-Rétiföldek	1688	boy	in earthfilling
16	pellet bell	RRM---	Zamárdi-Rétiföldek	498	child	between femurs
19	pellet bell	RRM 247.1.792.10	Zamárdi-Rétiföldek	792	child	near left femur
21	pellet bell	RRM 247.1.1172.1	Zamárdi-Rétiföldek	1172	child	pelvis/chest
22	pellet bell	RRM ---	Zamárdi-Rétiföldek	1273	child	right leg
25	pellet bell	RRM 247.1.1685.3	Zamárdi-Rétiföldek	1685	child	on femurs
39	pellet bell	RRM---	Zamárdi-Rétiföldek	2088	man	near right femur
27	pellet bell	RRM 247.1.1689.1	Zamárdi-Rétiföldek	1689	youth	near femurs
38	pellet bell	RRM---	Zamárdi-Rétiföldek	1937	empty grave	empty grave
24	3 pellet bells	?	Zamárdi-Rétiföldek	1653	horse	neck
35	5 pellet bells	?	Zamárdi-Rétiföldek	1900	horse	disturbed
40	pellet bell	RRM---	Zamárdi-Rétiföldek	2091	horse	breast chest
44	2 pellet bells?	RRM ---	Zamárdi-Rétiföldek	2283	horse	neck of horse?
36	3 tutuli		Zamárdi-Rétiföldek	1903	horse	on breast strap?

Morphological analyses, decorations and measurements

The bells excavated from early Avar period burials are very similar to Roman bell types und thus reinforce the assumption that people just used ancient Roman bells. Reusing ancient bells was also a common practice in the Merovingian Empire (Quast & Wolf 2010, 171). Three Roman bell types appear in the cemetery of Zamárdi-Rétiföldek.

The copper alloy bell type 1/var. B was found in grave 960, bell type 5/var. C in graves 407 and 614 and from grave 1872 an iron bell type 3 was unearthed. The bell from burial 517 contained a cast bell with an unusual form. Its base is rectangular, and the mantel is similar to bell type 1, but the crown is topped with aspherical ball, from which a hook is suspended. This detail is not typical for Roman bells, but artful mouldings of the bell crown and the handle were developed in Egypt during the Roman period (Hickmann 1951; Hickmann 1956). A figure of the Egyptian god Bes tops the bells UC33262 and UC52168 that can be seen in the Petrie Museum of Egyptian Archaeology in London.

The cone shaped bell made of sheet metal from burial 120, cemetery Kaposmérő-Agyagbánya, is a rarity and cannot be classified to any Roman bell type. It shows similarities with a Western Iranian bell type, called 'zang' (https://en.wikipedia.org/wiki/Zang_%28bell%29#/media/File:Bell_LACMA_M.76.97.894.jpg; https://www.britishmuseum.org/collection/object/W_1975-0301-3). The two bells from Kereki-Homokbánya (grave 53 and 104A) correlate with the Roman types 1 and 5.

Small cone shaped bells from the Avar period are only known from Kiskőrös-Vágóhídi-dűlő, graves 3, 8, 17, 41, 67 and date to the late Avar period (Garam 1993). Furthermore, truncated shaped pendants – tutuli – were found in the horse burial 1903, Zamárdi-Rétiföldek and thus correlate with the horse burial of Wien-Liesing, Carlbergergasse, grave 3 (Moßler 1948; Pomberger et al. 2022a) (**Fig. 6.**).

Pellet bells are represented in the shapes I, II, IV, V, VII and VIII according to the typology of Avar pellet bells (Pomberger 2020). They are cast in various copper alloys and forged from iron sheet as well as from copper alloy sheet. Sounds slots are simple or cruciform shaped. Pellet bells composed of two vertical halves made from sheet metal show no sound slots and no sound holes. The number of sound holes varies between two and four with different positions. The surface of pellet bells shows decorations like vertical grooves, vertical and radial-horizontal grooves, oblique line bundles and vertical grooves, and circumferential-vertical grooves. One pellet bell from the burial 48, Kaposvár-Toponár, 40-es őrház (cat. 5) has circumferential banding pattern with dots. This kind of decoration appears on three pellet bells, which were found in burial 50 in the Szent Márton Church, Szombathely, dating to the 11th century CE (Kiss 2000, Fig. 91/10; pl. 81/50/2–4).

Very rare are pellet bells with faces, in our case known from burial 2308, Zamárdi-Rétiföldek (cat. 45) (**Fig. 7.**) and Kereki-Homokbánya, grave 120 (cat. 61). Analogies are known for example from grave 11, Komárno IV (Čilinská 1982, pl. 4), grave 107, Komárno IX (Trugly 1993, pl. 12) and Keszthely-Városi temető (Lipp 1885, 116/326). Decorations of vertical grooves on the lower part of the pellet bell appear on pellet bells in grave 660, Wien-Csokorgasse (Pomberger et al. 2022a), in grave 104, Gyenesdiás, in graves 40 and 42 from Esztergályhorváti-Alsóbárándpuszta. The two latter graves date to the Carolingian period (Pomberger et al. 2023) as well as the grave 34 of the cemetery Bratislava-Rusovce (Pomberger et al. 2022c). An analogy to the pellet bell with vertical and radial-horizontal grooves from grave 1711, Zamárdi-Rétiföldek (cat. 28) is known from grave 660, Wien-Csokorgasse (Pomberger et al. 2022a). One of the three pellet bells from grave 634, Vörs-Battyáni Disznólegelő, is decorated with an x-shaped sign (cat. 53).

The sizes of the pellet bells vary between 1.4–3.8 cm without handle and 1.8–4.6 cm with handle. The weight of complete conserved pellet bells is between 9.67–24.56 g.

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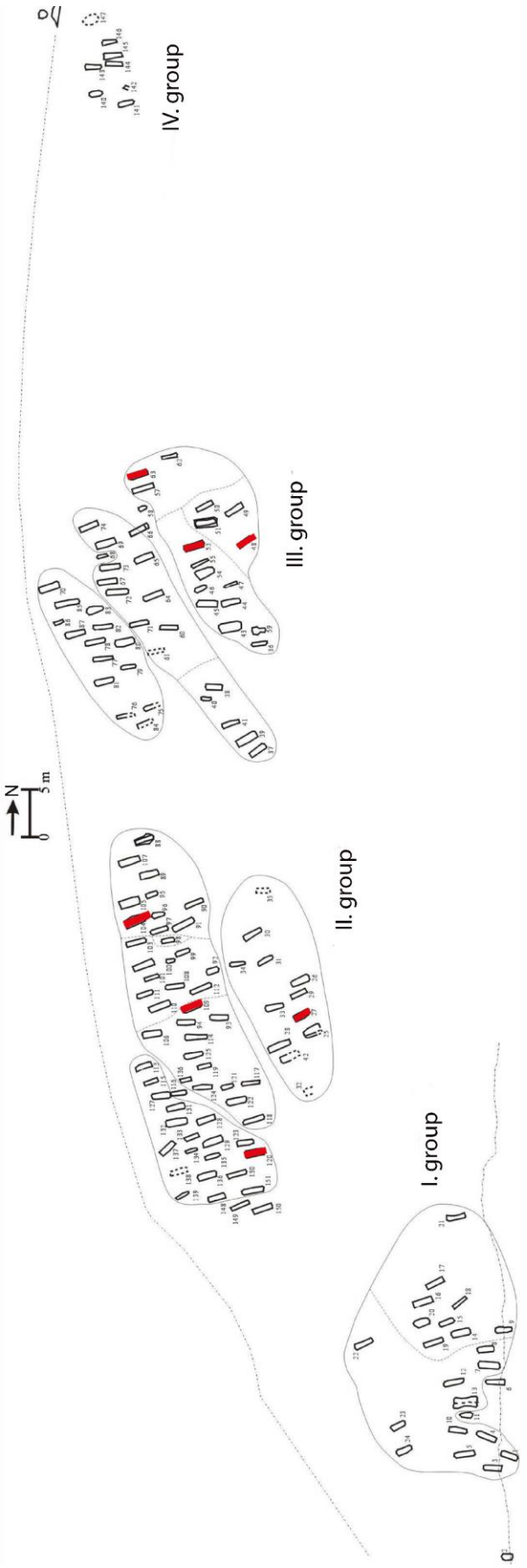

Fig. 5.: Graves with bells and pellet bells from the cemetery of Kereki-Homokbánya (Graphic: B. M. Pomberger, according to László 2012, map 4).

5. ábra: Csengőket és csörgőket tartalmazó sírok a Kereki-Homokbánya temetőben (rajz: B. M. Pomberger, László 2012, 4. térkép nyomán).

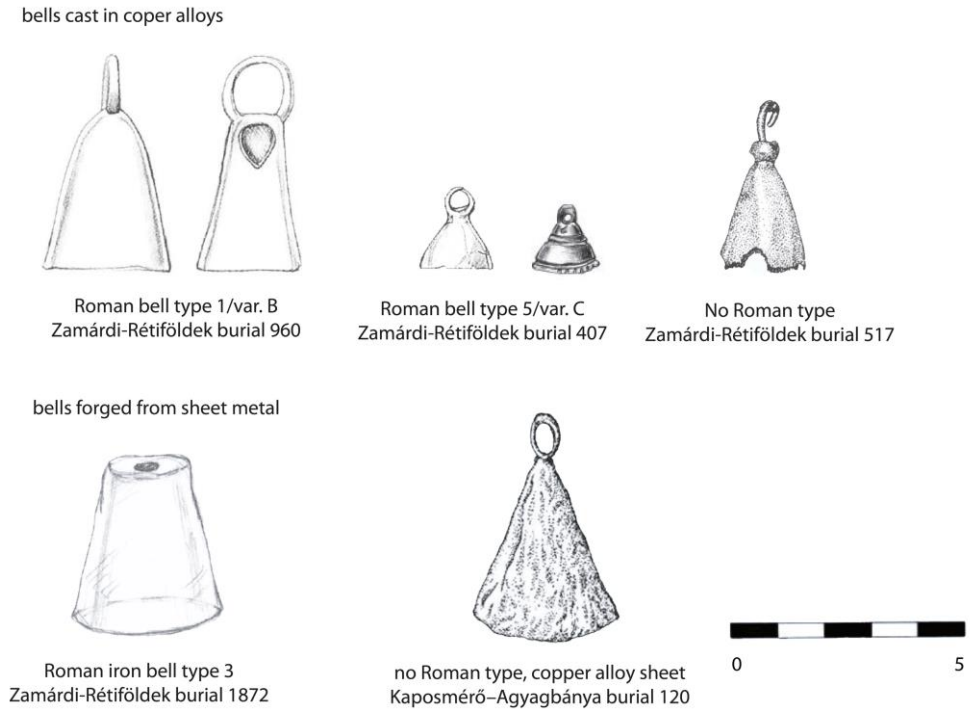

Fig. 6.: Bell types from Avar burials (Graphic: B. M. Pomberger according to Bárdos & Garam 2009, pl. 47, 67, 112; Bárdos & Garam 2014, pl. 196; Varga 2016, pl. 8/9; László 2012, pl. 70/4).

6. ábra: Csengőtípusok avar temetkezésekben (rajz: B. M. Pomberger, Bárdos & Garam 2009, pl. 47, 67, 112; Bárdos & Garam 2014, pl. 196; Varga 2016, pl. 8/9; László 2012, pl. 70/4 nyomán)

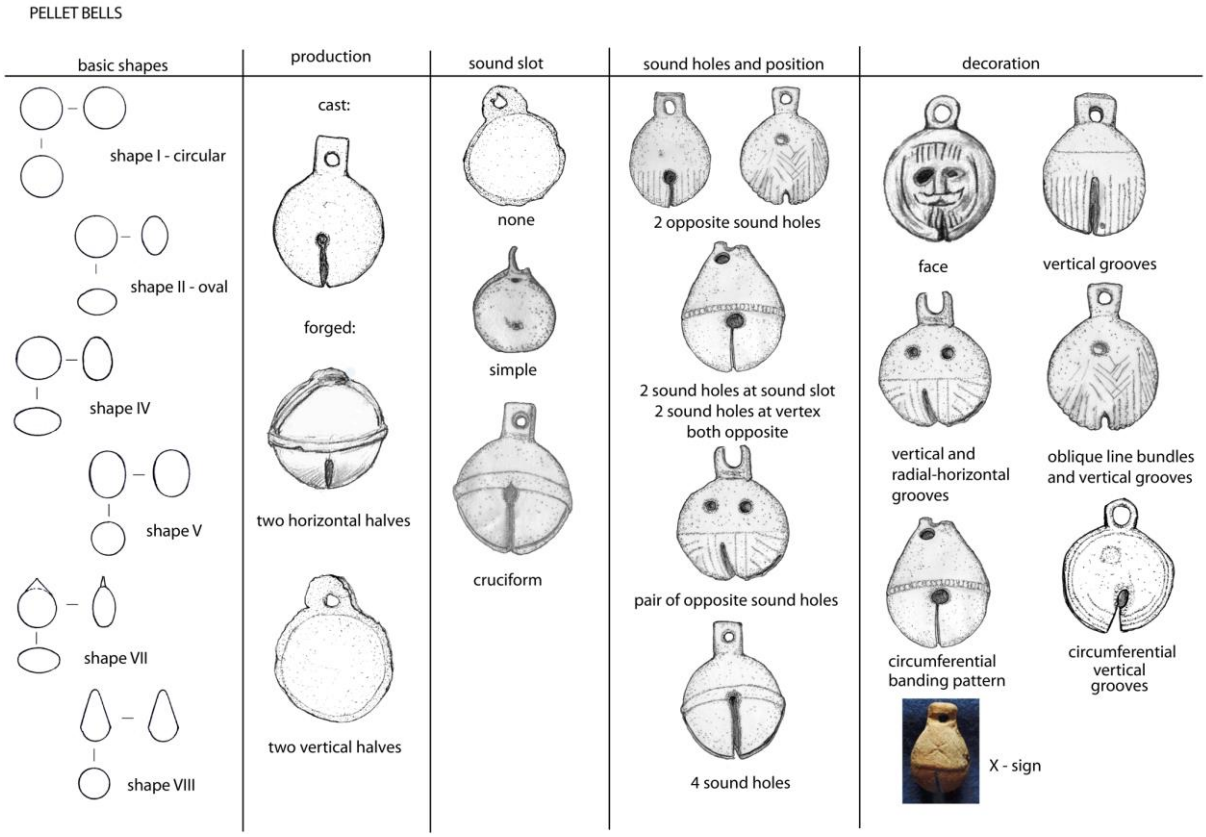

Fig. 7.: Shapes and decorations of pellet bells (Graphic: B. M. Pomberger).

7. ábra: A csörgők alakja és díszítései (rajz: B. M. Pomberger)

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Chemical analyses

31 metallic idiophones were analysed non-destructively by using a SPECTRO xSORT Combi handheld XRF spectrometer (15–50 kV, 30–120 µA, Rh anode, SDD detector, 'Alloy Plus including Light Elements' built-in calibration, measurement area 3 mm in diameter, 60 sec measurement time).

The analysed metallic idiophones were manufactured from different types of copper alloys: mainly bronze, leaded bronze, and gunmetal (**Figs. 8–9; Table 2.**). The composition of the alloys is very heterogeneous, generally the measured concentration of the alloying elements (Pb, Zn, Sn) is very high. It can be due to corrosion processes (especially in case of Pb and Sn, which can be enriched in the surface layer of the objects), or manufacturing processes (e.g., to enhance sound-ing).

The heavily corroded objects are the following: RRM 247.1.2352.2; pellet bells 2,3,4; RRM 247.1.1885.1; RRM 247.1.2099.1; RRM 247.1.1711.8; RRM 247.1.1905.4 and RRM 247.1.960.3. Data of these objects can be taken into consideration only qualitatively.

RRM 76.625.11 and RRM 77.11.31-32-33 pellet bells were presumed to be gilded, however, the measurements disproved gilding.

Elevated silver content (4.1 wt%) was measured in case of RRM 78.31.7 pellet bell, but no sign of silvering is seen by naked eye.

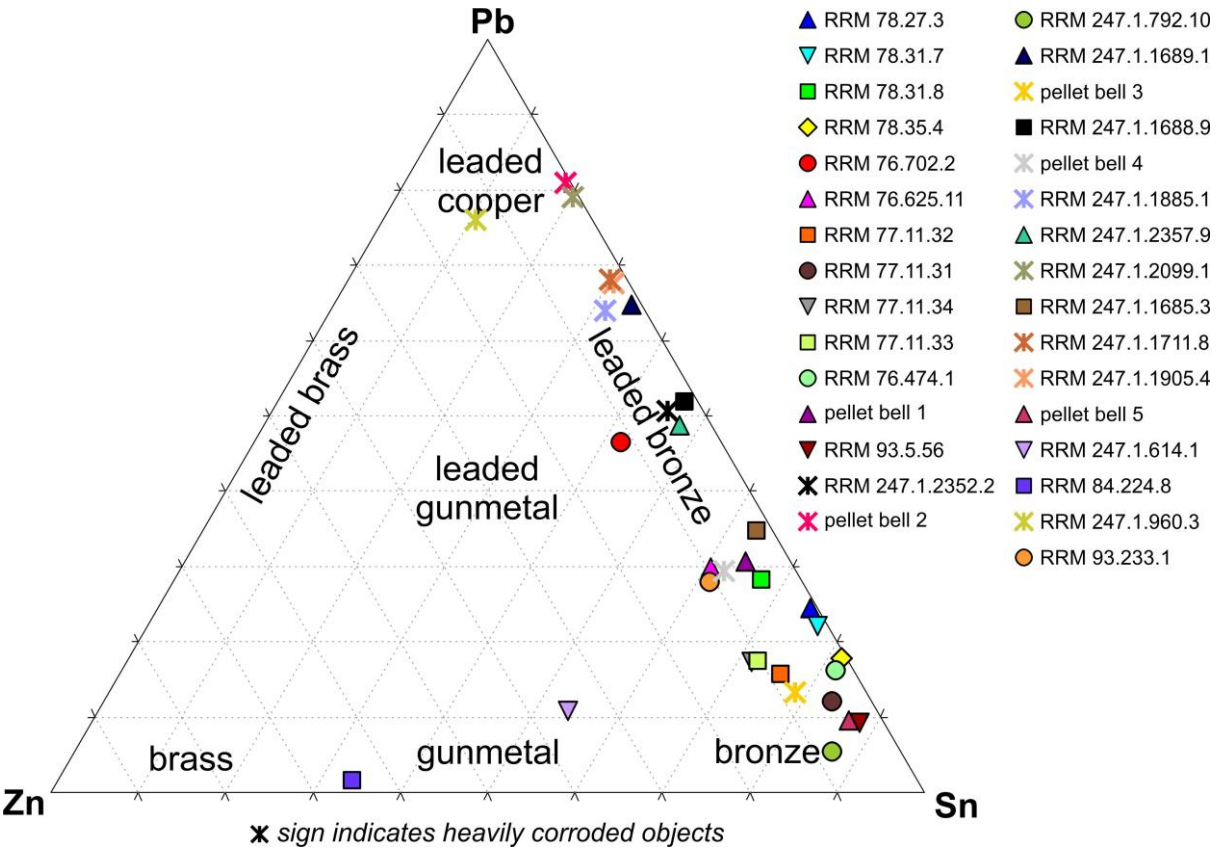

Fig. 8.: Chemical composition of the metallic idiophones from the Rippl-Rónai Museum (Kaposvár) plotted on the Sn-Pb-Zn ternary diagram (after Bayley 1989). During corrosion processes of copper-based alloys lead and tin contents increase, whereas zinc content decreases towards the surface (Graphic: V. Mozgai).

8. ábra: A kaposvári Rippl-Rónai Múzeum fémidiofonjainak kémiai összetétele az Sn-Pb-Zn háromszög-diagramon (Bayley 1989 nyomán). Rézalapú ötvözetből készült tárgyak korróziója során az ólom- és az óntartalom növekszik, míg a cinktartalom csökken a felszín felé (rajz: Mozgai V.).

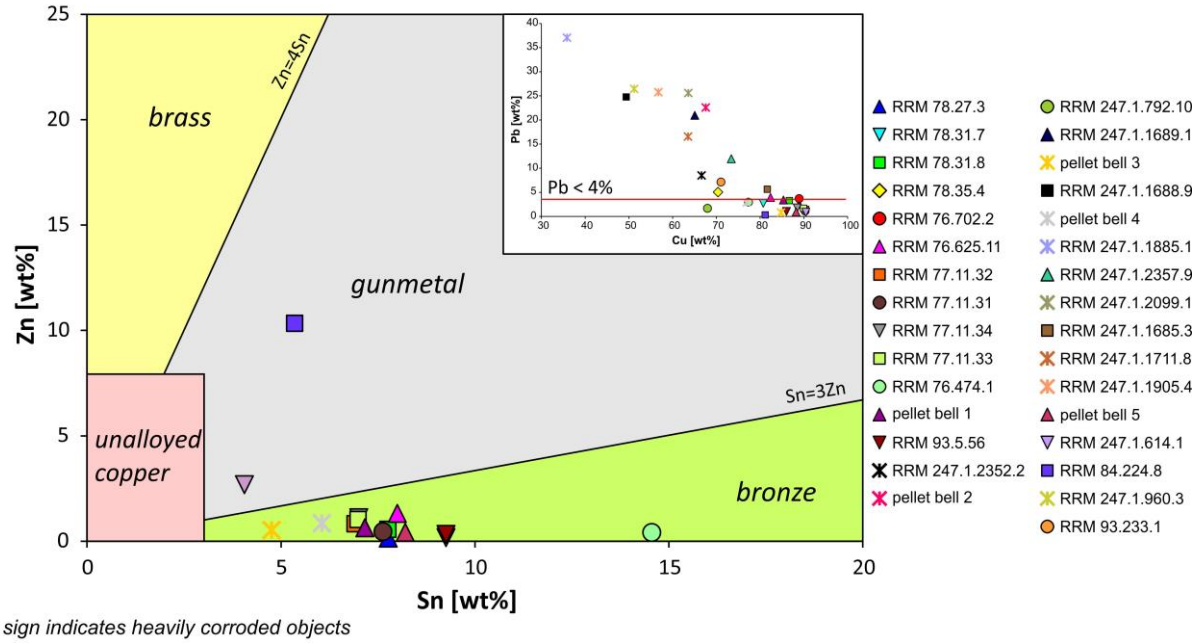

Fig. 9.: Chemical composition of the metallic idiophones from the Rippl-Rónai Museum (Kaposvár) plotted on the Zn-Sn and Pb-Cu binary diagrams (after Bayley 1989). The Pb-Cu binary plot is used to differentiate leaded and non-leaded alloys (Graphic: V. Mozgai).

9. ábra: A kaposvári Rippl-Rónai Múzeum fémidiofonjainak kémiai összetétele az Sn-Zn és a Cu-Pb diagramon (Bayley 1989 nyomán). A A Cu-Pb diagramot az ólmozott és nem ólmozott ötvözetek elkülönítésre alkalmazzuk (rajz: Mozgai V.).

(Psycho-) Acoustical Measurements

The acoustics of idiophones is complex and depends on a variety of factors, most of which are connected to the physical properties of the object itself, like material/alloy, weight/size, shape or wall thickness, but also the mode of excitation plays a crucial role. The sound and timbre of pellet bells consists of a series of single excitations over time, that occur when an encapsulated smaller object bounces against the inner walls, forcing the 'mantle' of the pellet bell to oscillate. When such an idiophone is excited constantly (e.g., by shaking it), typically up to 25 single hits per second are created. Those single impulses have a very short decay time of roughly 20–150 milliseconds. Bells are usually larger and create less hits per seconds, because the clapper is mounted to the inner surface. They can oscillate more freely with less dampening, and typically have significantly longer decay times of a few hundred milliseconds to sometimes even some seconds.

How long an idiophone can oscillate mainly depends on the physical properties, if an object is solid but does not dampen the oscillation too much, a lot of so-called 'modes' can form. In idiophones (and also membranophones) the sound does not consist of just one fundamental frequency (f0) with overtones (harmonics) being integer multiples, as is the case in aerophones and chordophones, but rather a complex combination of single oscillations that create a number of partial frequencies or 'overtones' (Hall 1980, 156–168).

Every instrument, that is excited impulsively, has a very short attack time of just a few milliseconds, within that timespan the maximum amplitude is reached and the oscillation immediately starts to decay. The moment of impact is high in spectral energy, and partial frequencies can be seen after a few milliseconds. In the spectrogram, the impact resembles a vertical line with dense colouring and the decay of partials can be seen as horizontal lines. Decay time is different for all the partials, but typically low frequency oscillations are dampened less (Hall 1980, 168), thus have a longer decay time than higher ones. **Fig. 10a–b** illustrates the spectral differences in excitation and decay time between pellet bells (a) and smaller bells (b).

In the time and frequency domain, bells and pellet bells typically overlap, depending on the size and weight of the objects. Smaller ones create more single impulses than larger ones in both groups and also the frequency range is higher in smaller ones. Larger pellet bells and smaller bells, however, might be equal in both parameters. But overall, bells produce more pronounced partials in the spectrum, thus are more tonal and evoke a clearer sensation of pitch in listeners, while the pellet bells sound is rather noisy and can evoke only little pitch perception (Schneider 2000).

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Table 2.: Chemical composition of the metallic idiophones from the Rippl-Rónai Museum (Kaposvár) measured by hXRF. The results are given in wt%. LOD = limit of detection. The elevated Fe, Al, Si, S and P content is due to corrosion processes and soil contamination. Red: gilding presumed but not proved; purple: silvering(?). (by V. Mozgai).

2. táblázat: A kaposvári Rippl-Rónai Múzeum fémidiofonjainak kézi XRF-fel mért kémiai összetétele. Az adatokat tömeg%-ban adtuk meg. LOD = kimutatási határ. A megnövekedett Fe-, Al-, Si-, P- és S-tartalom a korróziós folyamatok és talajszennyeződés eredménye. Piros számok: feltételezett, de nem igazolt aranyozás, lila számok: ezüstözés (?) (a táblázatot összeállította: Mozgai V.).

Cat.	Inventory Nr.	Object	Cu	Zn	Sn	Pb	Sb	As	Au	Ag	Fe	Al	Si	P
2	RRM 78.27.3	pellet bell	88.9	0.1	7.8	2.5	0.1	<LOD	<LOD	0.6	0.02	<LOD	<LOD	<LOD
3	RRM 78.31.7	pellet bell	80.6	0.1	9.3	2.7	0.1	0.1	0.1	4.1	0.03	1.6	<LOD	0.3
4	RRM 78.31.8	pellet bell	86.4	0.5	7.7	3.3	0.7	0.1	<LOD	0.4	0.1	<LOD	<LOD	0.4
5	RRM 78.35.4	pellet bell	70.1	0.2	22.6	5.0	0.2	0.3	0.4	0.1	0.2	<LOD	<LOD	0.5
1	RRM 76.702.2	pellet bell	88.8	0.9	3.3	3.7	0.1	0.2	0.1	0.6	0.1	1.5	<LOD	0.2
6	RRM 76.625.11	pellet bell	82.2	1.3	8.0	3.9	0.1	0.2	0.2	0.2	0.7	<LOD	<LOD	2.9
6	RRM 76.625.11	pellet bell	86.4	1.2	6.2	3.0	0.1	0.1	0.2	0.1	0.5	<LOD	0.2	1.7
7	RRM 77.11.32	pellet bell	90.3	0.8	6.9	1.5	0.1	0.03	0.1	0.1	0.1	<LOD	<LOD	<LOD
7	RRM 77.11.32	pellet bell	87.2	2.3	6.7	0.7	<LOD	0.1	<LOD	<LOD	0.1	0.5	1.6	0.1
8	RRM 77.11.31	pellet bell	90.2	0.4	7.6	1.1	0.1	0.1	0.1	0.2	0.1	<LOD	<LOD	<LOD
8	RRM 77.11.31	pellet bell	88.1	5.5	4.0	0.1	<LOD	<LOD	<LOD	<LOD	0.1	<LOD	<LOD	0.1
9	RRM 77.11.34	pellet bell	88.4	1.1	7.0	1.8	0.1	0.03	<LOD	0.1	0.2	<LOD	1.1	0.1
10	RRM 77.11.33	pellet bell	89.8	1.0	7.0	1.7	0.1	<LOD	<LOD	0.1	0.1	<LOD	<LOD	0.04
10	RRM 77.11.33	pellet bell	88.4	2.9	6.7	1.2	<LOD	0.1	<LOD	<LOD	0.1	<LOD	0.1	0.1
11	RRM 76.474.1	pellet bell	77.2	0.4	14.6	3.0	0.2	0.04	0.2	0.2	0.4	<LOD	1.0	1.7
12	pellet bell 1	pellet bell	85.3	0.6	7.2	3.4	0.2	0.1	<LOD	1.8	0.3	0.5	0.5	0.1
13	RRM 93.5.65	pellet bell	85.9	0.3	9.3	1.0	0.3	0.1	0.2	0.4	0.2	<LOD	<LOD	1.6
47	RRM 247.1.2352.2	pellet bell	66.5	0.7	7.5	8.5	0.1	0.1	<LOD	0.04	0.4	2.9	11.4	1.8
42	pellet bell 2	pellet bell	67.5	0.2	5.1	22.6	0.1	0.6	0.1	0.03	0.4	<LOD	0.7	2.6
19	RRM 247.1.792.10	pellet bell	67.9	2.3	25.1	1.7	0.1	0.6	<LOD	<LOD	0.2	<LOD	1.3	0.8
27	RRM 247.1.1689.1	pellet bell	65.0	0.5	11.0	21.0	0.3	0.3	<LOD	0.1	0.1	<LOD	<LOD	1.8
46	pellet bell 3	pellet bell	84.7	0.5	4.8	0.8	<LOD	<LOD	<LOD	<LOD	0.2	2.8	6.0	0.2
26	RRM 247.1.1688.9	pellet bell	49.2	0.8	21.9	24.7	0.2	0.8	<LOD	0.7	0.7	0.5	0.2	0.1
43	pellet bell 4	pellet bell	76.9	0.8	6.1	2.9	<LOD	<LOD	<LOD	0.04	0.2	2.7	4.7	5.7
34	RRM 247.1.1885.1	pellet bell	35.7	2.7	18.0	36.9	0.3	<LOD	<LOD	0.2	1.3	<LOD	1.0	3.5
48	RRM 247.1.2357.9	pellet bell	73.1	1.0	11.6	11.9	0.3	0.4	<LOD	0.5	0.1	0.3	0.4	<LOD
41	RRM 247.1.2099.1	pellet bell	63.5	0.2	6.4	25.6	0.2	0.3	<LOD	0.4	0.1	0.8	0.2	2.1
25	RRM 247.1.1685.3	pellet bell	81.6	0.3	10.1	5.6	0.1	0.1	<LOD	0.1	1.7	<LOD	<LOD	0.2
28	RRM 247.1.1711.8	pellet bell	63.4	0.5	7.2	16.6	0.3	0.6	<LOD	<LOD	0.6	1.9	5.9	2.9
37	RRM 247.1.1905.4	pellet bell	56.6	0.7	11.6	25.7	0.5	0.6	<LOD	0.1	0.2	<LOD	<LOD	3.9
49	pellet bell 5	pellet bell	88.2	0.4	8.2	0.9	0.1	0.1	<LOD	0.8	0.1	<LOD	0.8	0.2
18	RRM 247.1.614.1	bell	90.4	2.7	4.1	0.9	0.04	0.1	<LOD	<LOD	0.3	0.9	0.4	0.2
15	RRM 84.224.8	bell	81.1	10.4	5.3	0.3	0.1	0.3	<LOD	<LOD	0.5	0.7	<LOD	0.1
20	RRM 247.1.960.3	bell	51.1	4.6	3.6	26.4	0.03	0.4	0.2	<LOD	6.2	0.2	<LOD	7.0
50	RRM 93.233.1	pellet bell	70.8	2.7	15.4	7.1	0.1	0.3	<LOD	0.6	0.2	<LOD	<LOD	1.3

One of the most important acoustic parameters is the sound pressure level (SPL) of the objects, given in dB re p0 (= 0.00002 Pa), which is calculated from the root mean square (RMS) value of the calibrated recordings. Since the SPL decreases by 6 dB per doubling of the distance to the source (Attenborough 2014, 119), the maximum audible distance for a sound object can be calculated for a given background noise. Another key parameter is the frequency range covered by the sound from the lowest (f_{min}) to the highest (f_{max}) partial frequency. The overall SPL of small idiophones is not high, however, the frequency range covered typically overlaps with the most sensitive area of the human hearing, which is roughly between 1.5–6 kHz (Moore 2003, 57). In most cases, one of the partial frequencies is particularly high in amplitude, which is called the 'peak frequency' that matches the '*eigenfrequency*' of the idiophone and also plays a key role in pitch perception.

A different approach to describe sounds is psychoacoustic parameters that do not directly reflect physical properties of sounds, but rather model subjective human perception thereof. Psychoacoustics as part of psychophysics seeks to find the correlations between the physical (objective) qualities of a sound and the perception (subjective) of it. In psychoacoustics, the measure for the intensity sensation is loudness, which can be given either in '*phon*' or '*sone*'. They are defined with a reference sound of 1 kHz at 40 dB SPL equalling 40 phon or 1 sone (Fastl & Zwicker 2007, 203–204). The unit phon is interval-scaled, while the sone is ratio-scaled, which means that 50 phon is twice as loud as 40, and 2 sone is twice as loud as 1. Sharpness is a parameter that describes a sensation resulting from the spectral shape and density, where the humans' most sensitive hearing area also plays a crucial role. It is measured in '*acum*', which is also interval-scaled and defined as one critical band wide noise at 60 dB with a center frequency of 1 kHz (Fastl & Zwicker 2007, 241). Roughness is created by a modulation of the amplitude (AM), which starts around 15 Hz of modulation frequency and reaches its maximum in 70 Hz, but starts to disappear at a modulation frequency higher than 150 Hz. The unit to measure the parameter is '*asper*', which can be a value between 0 (not rough at all) and 1 (maximum roughness at 70 Hz AM at 1 kHz (Fastl & Zwicker 2007, 257). Tonality as a model predictor is represented by the tone-noise-ratio (TNR), given in dB that compares the tonal part of a sound (partial frequencies) to the noise part based on a human hearing-model (Becker et al. 2019, 5820). Those four parameters are often used to quantify a concept called the 'sensory pleasantness' of sound (Fastl & Zwicker 2007, 243).

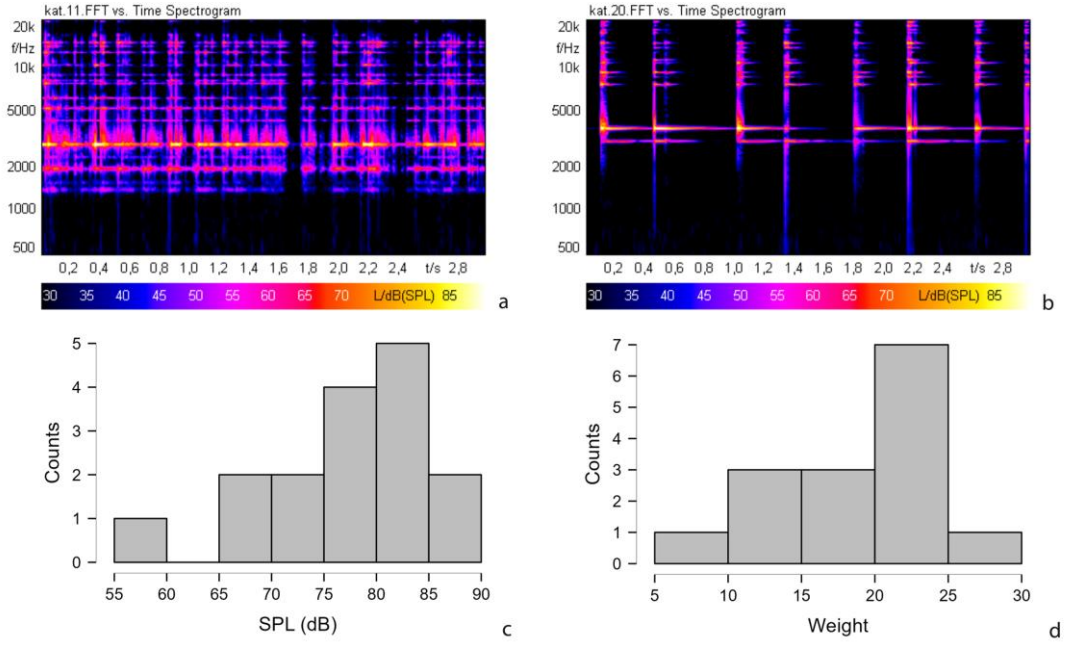

Fig. 10.: a-b - Spectrogram (HAN, 1024) of pellet bell cat. 11 (left) and bell cat. 20 (right). Yellow/red colour indicates high amplitudes in the respective frequency range (y-axis) over time (x-axis). **c–d** - Histograms for the distribution of sound pressure level (left) and weight (right, one value unknown and missing in the graph) of the 16 analysed objects (Graphic: J. Mühlhans).

10. ábra: a-b - A kat. 11 csörgő (balra) és a kat. 20 csengő (jobbra) spektrogramja (HAN, 1024). A sárga/vörös színek a nagy amplitúdókat jelzik az adott frekvenciatartományban (y tengely) az idő függvényében (x tengely). **c–d** - A 16 elemzett tárgy hangnyomásszintjének (balra) és tömegének (jobbra, egy hiányzó adat kivételével) eloszlási hisztogramja (rajz: J. Mühlhans).

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More commonly used adjectives to describe the prominent timbral feature of brightness are high/low or dark/brilliant. This parameter can be estimated using the spectral centre of gravity or '*spectral centroid*' (SC) in Hz (Saitis & Siedenburg 2020, 2256). Last but not least, the impulsiveness of the sounds is measured, that quantifies the degree of impulsive content perceived in a sound. This property of sound is often described as 'rattles' or 'clicks' (Willemsen & Rao 2010, 2), in the non-standardized '*impulsiveness unit*' (iu).

The (psycho-) acoustic parameters mentioned above have been calculated using Audition CC (Adobe Audio Team 2024), ArtemiS Suite 9.3 (HEADAcoustics 2024) and Praat (Boersma & Weenink 2024) and additional statistical calculations have been done using JASP (JASP Team 2024).

16 objects, 15 pellet bells and one bell, have been recorded and analysed. While **Fig. 10a-b** already highlighted the differences in excitation between bells and pellet bells, there are still notable differences between the pellet bells as well.

The histograms (**Fig. 10c–d**) show that, with one exception (cat. 29, 56.1 dB), the objects lie in the range between 65–85 dB. The distribution also has a tendency towards louder objects. The weight distribution shows a concentration in the 20–25-gram range. However, the sound pressure level is not correlated with the weight, so larger objects are not louder in this case, as one would expect.

Apart from this exception (cat. 29), the objects are within 20 dB of each other, i.e. physically speaking, the SPL of the loudest object is about 8 times as high as that of the quietest, while subjectively speaking, as the width of the loudness measurement (14.8–50.6 sone) shows, the loudest object is almost four times as loud as the quietest. The differences in frequency range can be mainly seen in the lower end (f_{min}=1.0–3.9 kHz), because human hearing is rather sensitive in this area. In the upper end, where hearing gets increasingly insensitive, the range gets larger (f_{max}=13.5–21.8 kHz), but will less impact on sound perception.

The peak frequency, however, is highly correlated with the spectral centroid (r=0.716, p=.002), both being strong indicators of the perception of both pitch and brightness.

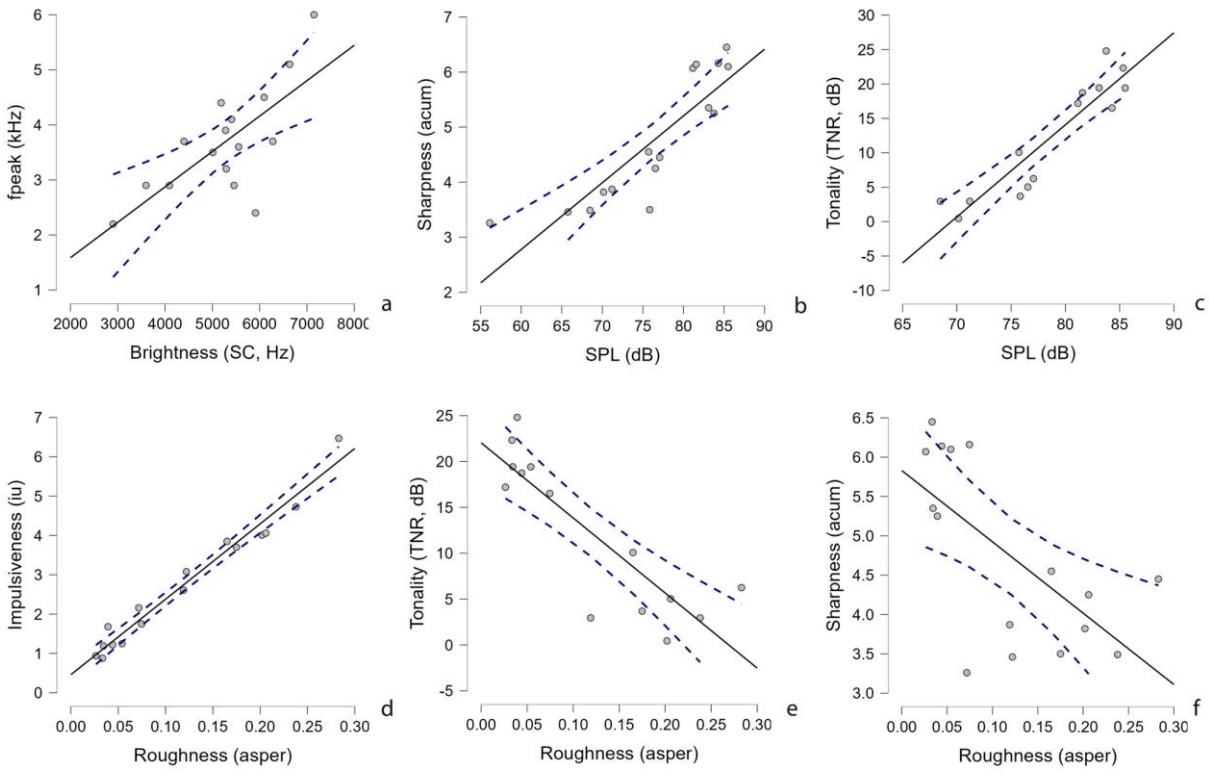

Fig. 11.: Correlation scatterplots with trend lines and 95% confidence interval for acoustical and psychoacoustic parameters (Graphic: J. Mühlhans).

11. ábra: Az akusztikus és pszichoakusztikus paraméterek korrelációs szórásdiagramjai trendvonalakkal és 95%-os konfidencia intervallummal (rajz: J. Mühlhans).

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SPL is – despite the very obvious loudness – also highly correlated with sharpness (r=0.869, p<.001) and tonality (r=0.922, p<.001). In simple words, the louder an object, the sharper and more tonal, i.e. less noisy. Roughness is correlated positively with impulsiveness (r=0.984, p<.001) and negatively to tonality (r=-0.859, p<.001) and sharpness (r=-0.654, p=.006).

Overall, the objects can be described as sharp (3.26–6.45 acum) and hardly to mildly rough (0.026–0.283 asper). In tonality, quite a large margin could be observed, ranging from 0.45 dB, which can be described as equally tonal and noisy, up to 28.8 dB (cat. 20, the bell), that has almost no noise components. Also, the objects are bright on average, with an SC ranging between 2.9–7.1 kHz and a peak frequency between 2.4–6.0 kHz (see **Fig. 11a–f**).

For the material, there was only one significant result – a higher amount of copper is associated with a lower value in f_{min} (r=-0.532, p=.041). Controlled experiments with replicas in many different alloys have shown that the influence on acoustic parameters is very large, but not in a predictable or linear way, that would result in statistical correlations (Mühlhans & Pomberger 2023, 165).

In the spectral properties, the objects can be roughly divided into three groups, one being sounds with very pronounced partial frequencies (cat. 1,3, 4, 5, 11, 20, 50), one being the less tonal sounds with less and broader partials (cat. 12, 41, 42, 46, 47) and one with hardly any partials at all (cat. 27, 28, 29, 49). **Fig. 12.** illustrates this with two examples for each of those ‘groups’.

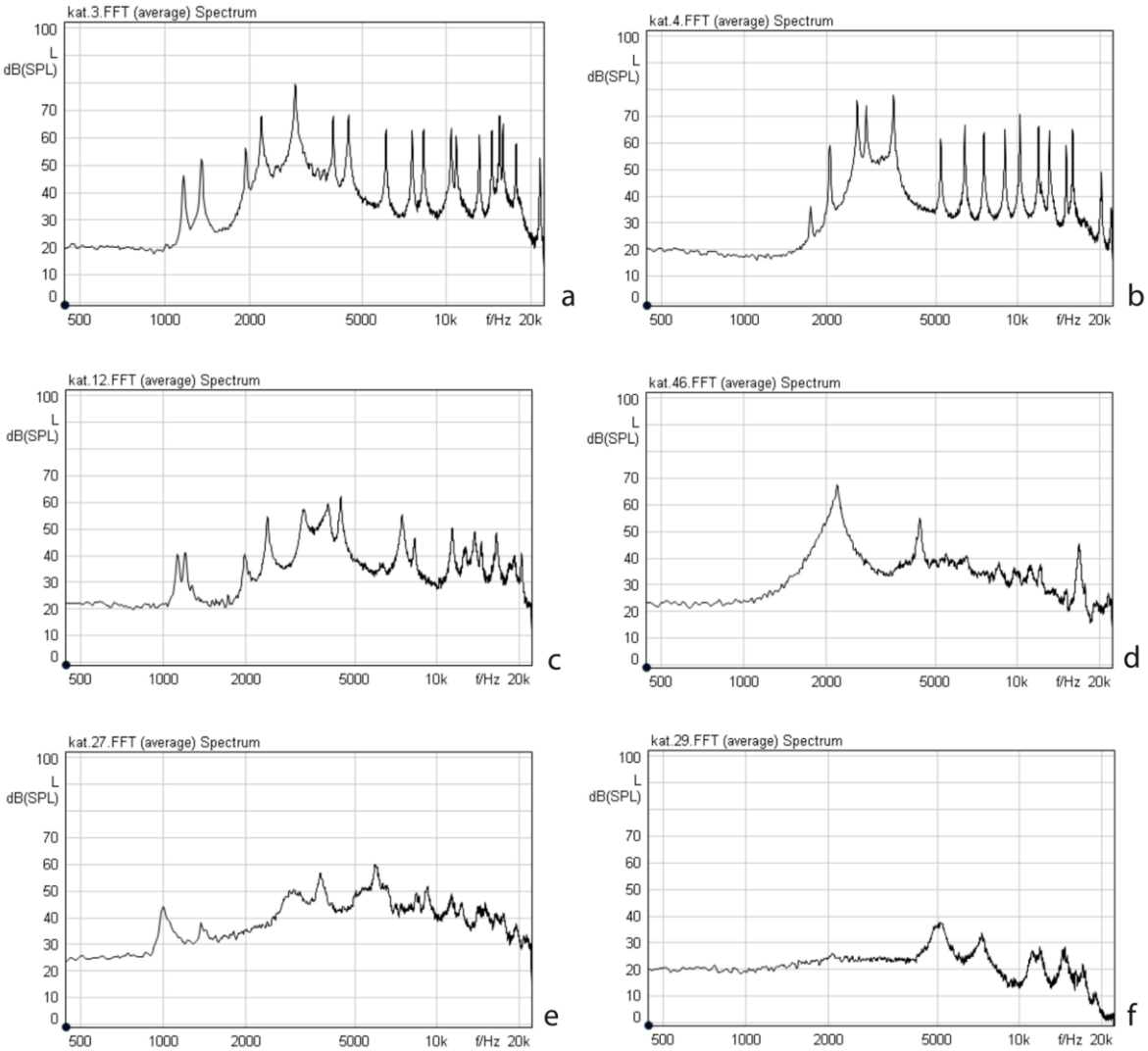

Fig. 12.: Spectra (HAN, 4096) of sounds (**a–b**) with very strong and pronounced partial frequencies, (**c–d**) less pronounced and weaker ones and (**e–f**) sounds with hardly any partials at all (Graphic: J. Mühlhans).

12. ábra: (**a–b**) Nagyon erős és hangsúlyos parciális frekvenciájú, (**c–d**) kevésbé hangsúlyos és gyengébb, és (**e–f**) parciális frekvenciát alig tartalmazó hangok spektrumai (HAN, 4096) (rajz: J. Mühlhans).

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The previously mentioned negative correlation between impulsiveness and tonality can be seen in the spectrogram. **Fig. 13.** shows two examples each, high in impulsiveness and low in tonality, and vice versa. Tonal sounds clearly have a decent amount of decay time and also visible spectral energy in the partials.

Conclusively, we can say that the idiophones from Kaposvár cover a wide range of all (psycho-) acoustic parameters, thus single objects are more or less typical for pellet bells. The only bell stands out in its temporal features but not quite in spectral ones, except for being the most tonal object. The statistical analyses show that objects high in loudness and SPL are also higher in sharpness and tonality, but lower in roughness and impulsiveness. These analysis results reflect the current condition of the objects, some of which are less and some are more corroded. In the uncorroded original condition, it can be assumed that SPL/loudness, sharpness, tonality and brightness would be considerably higher, while the values for roughness in particular would be lower.

Function and use of pellet bells

We have evidence that the pellet bells and bells were fixed to the belt with ribbons, strings or chains because of the position in which they were found. In the graves 18, 36 and 43 from the site Kaposvár-Toponár, 40-es őrház, as well as in grave 82 from site Kaposvár-61. sz. út the deceased have their pellet bells on their left sides near the thighbones.

The same find position is known from sixteen graves from the cemetery Zamárdi-Rétiföldek, all burials of female individuals. In most of these cases it seems that the idiophones hang from long belt straps. Some women and girls wore them hooked into their ornamental discs which were fixed on long hanging straps (graves 517, 97a, 1685, 1711, 1904, 2099, 2275, 2308, 2357). This kind of wearing belts and belt assemblages is typical for women of the Eastern Pannonian communities with Germanic culture (Garam 2011; Balogh 2019). On the other hand, the man interred in grave 2088 wore his pellet bell on the right side (Bárdos & Garam 2014). The pellet bells of the Kereki-Homokbánya cemetery were also found near the left hips, femurs/legs and knees. Interesting is the position of a small bell from burial 53, which was found on the skull. It was probably fastened on a necklace. In grave 634 from Vörs-Battyáni Disznólegelő the three pellet bells lay near the left leg. It might have been the custom for women to wear their bells and pellet bells on the left side and for men on the right. However, we have too few examples of pellet bells found in male graves to be able to derive a rule.

Both pellet bells and bells are made from metal. Pellet bells imitate vascular fruits and thus are

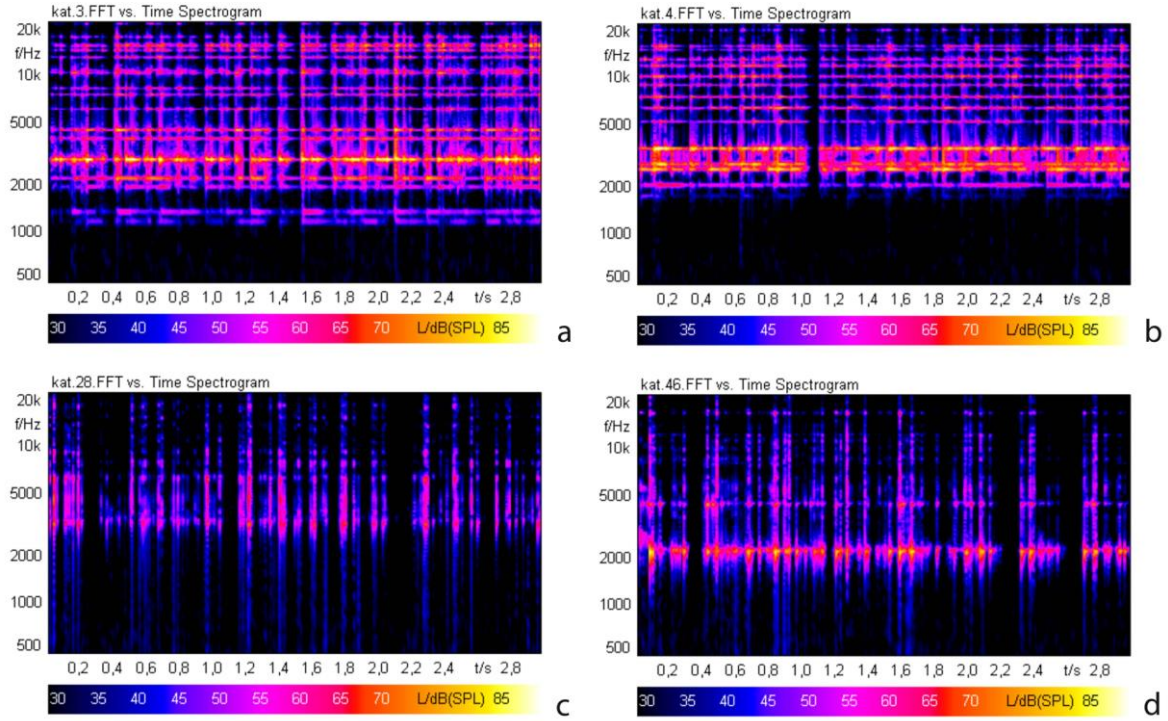

Fig. 13.: Spectrograms (HAN, 1024) of sounds (**a–b**) high in tonality and low in impulsiveness (cat. 3 & 4) and (**c–d**) vice versa (cat. 28 & 46) Graphic: J. Mühlhans).

13. ábra: (**a–b**) Nagy tonalitású és kis impulzivitású (kat. 3 & 4), illetve (**c–d**) fordított jellemzőjű (kat. 28 & 46) hangok spektrogramja (HAN, 1024) (rajz: J. Mühlhans).

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symbols for fertility, abundance and life after death (Pomberger in print). Bells developed from sounding stones, clay bells, fruit bowls and sounding bowls and imitate upturned bowls (Kramer 2015, 13). Both idiophones produce noises and sounds when being shaken respectively struck. They should be understood as sounding amulets that fulfil their duty as long as the owner believes in its supernatural properties (Hirschberg 1988, 23–24). Since there are only less than 1.17% of graves with idiophones, this custom and belief seems to be practised by only few Avar people.

Horses with pellet bells as parts of the horse gear were found in the graves 1653, 1900 and 2019 from the cemetery in Zamárdi-Rétiföldek (cat. 24, 44, 35, 40) as well as from grave 57, Kaposvár-Toponár, 40-es őrház (cat. 7–10). The five pellet bells from grave 1900 and the three ones from grave 1653, Zamárdi-Rétiföldek, are forged from iron sheet, probably composed of two horizontal halves (The authors could not investigate these items). The pellet bell from grave 2091, Zamárdi-Rétiföldek is cast in copper alloy. Interesting are the four pellet bells from grave 57, Kaposvár-Toponár, 40-es őrház, which are forged from sheet metal (bronze) and composed of two vertical halves in shape VII (**Fig. 7.**). This type of pellet bell mostly appears in horse burials as parts of the bridle. Analogies are known from Avar cemeteries in Slovakia, Hungary and Austria. Mostly this type of pellet bells is part of the horse head harness. But in a few cases, this type pellet bells were found in the context with human skeletons like in grave 276, Szebény I, and Kaposvár-Toponár, 40-es őrház, grave 52, Hungary. Interesting is the pellet bell from grave 525, Vösendorf-Laxenburgerstraße, Austria, in which a man was interred. Among his grave goods was the head harness sleeve of a horse – as a symbol for a horse – as well a sheet metal pellet bell, which could be a further symbolic attribute of a horse or it simply belonged to the man (Tarcsay 2013; Pomberger & Stadler 2018) (see **Table 3.**, graves containing pellet bells of copper alloy sheet metal). Pellet bells became parts of the Avar horse gear in the second half of the late Avar period (8th/9th century CE) with a few exceptions, namely grave 79, Bratislava-Devínska Nová Ves (cemetery I), dating to MAP II, grave 401 from the same cemetery, dating to LAP I and grave 107, Komárno (cemetery IX), dating to LAP I (Pomberger & Stadler 2018; Csuthy 2019; Pomberger et al 2022c). Avar people have been wearing bells since the middle of the 7th century (Pomberger & Stadler 2018).

Table 3.: Graves containing pellet bells of copper alloy sheet metal (by B. M. Pomberger).

3. táblázat: Rézötvözet fémlemezből készült csörgőket tartalmazó sírok (a táblázatot összeállította: B. M. Pomberger)

Pellet bells	Site	Grave nr.	Belongs to	Literature
15	Radvaň nad Dunajom, Žitava I, SK	10	horse	Budinský-Krička 1956
4	Radvaň nad Dunajom, Žitava I, SK	31	horse	Budinský-Krička 1956
1	Komárno IV, SK	25	horse or man, unclear	Čilinská 1982
1	Komárno VIII, SK	24	horse	Čilinská 1982
1	Komárno IX, SK	36	horse	Trugly 1987
1	Komárno IX, SK	71	horse or man, unclear	Trugly 1987
2	Komárno IX, SK	79	horse	Trugly 1987
1	Komárno IX, SK	101	horse	Trugly 1993
2	Komárno IX, SK	121	horse	Trugly 1993
1	Komárno IX, SK	153	horse	Trugly 1993
3	Valaliky-Všechsvätých, SK	98/84	horse	Zábojník & Béreš 2016
1	Wien-Csokorgasse, AT	650	horse	Pomberger et al. 2022a
12	Pitvaros-Viztározó, HU	51	horse	Bende 1998
1	Vösendorf/Laxenburgerstraße, AT	525	symbolic attribute for horse?	Tarcsay 2013; Pomberger & Stadler 2018
1	Szebény, HU	276	child	Garam 1975
1	Kaposvár-Toponár, 40-es őrház, HU	52	child	Bárdos 1978

Visuality and sound

Though there is surely some symbolism involved in the use of idiophones, they can also be seen as accessories at the same time. Just as bright colours, jewellery, and decorations of clothing attract attention visually, so do sounds – without the listener needing to even look in the wearer's direction. Thus, idiophones can be visually appealing, for example decorated, shining pellet bells, or dangling small metal elements, but also acoustically. One specific clothing composition from the Early Medieval period demonstrates this well: women's costumes with long hanging straps on their belts (German: "*Gehängegürteltracht*") (Garam 2011). This costume, which can be found throughout burials of various Early Medieval cultures from Western to Eastern Europe, is characterised by a belt that was worn on the outside that was often decorated with many fittings (otherwise rather untypical for Avar women) and had multiple objects attached to it – some of them clapping and rattling, such as pellet bells and disc pendants.

Practical everyday objects could also be attached, such as spindles, knives, keys, needle cases, and perhaps also small bags. These hung quite low, in the area of the knee or even lower, thus they must have dangled a lot while walking, drawing the eye to the area due to movement and also creating sounds with the objects clapping against each other. Examples for these ensembles are the burials 82, 91, 97a, 517, 646, 1055, 1321 (Bárdos & Garam 2009), 1887, 1918, 1921, 2125, 2308 and 2357 (Bárdos & Garam 2014) from Zamárdi-Rétiföldek.

In order to get an idea of how such a costume could have appeared, we decided to recreate the costume of the woman buried in Zamárdi-Rétiföldek grave 2357, dating to the first half of the eighth century CE (**Fig. 14.**). The metal objects and the glass beads were all replicated from the grave goods. The organic objects were of course no longer preserved – and there are in fact no complete garments known from the Avar period. For this reason, the textile garments and the belt were recreated using the closest sources available, while at the same time incorporating organic fragments we have from the period. This means that all the fabrics were woven in a typical tabby weave and the underdress was made of linen, the most common fibre of the Avar period (Pomberger et al. 2021a, 124–125).

For the dresses, the garments from Moshchevaja Balka in the Northern Caucasus (Ierusalimskaja 1996), associated with the Alanic period, were used as a basis for the cuts, as these are excellently preserved and date to the 8th to 10th century CE. Furthermore, they exhibit characteristics of steppe cultures as well, such as caftans for clothing, which

Fig. 14.: Recreation of a woman's sounding costume based on grave 2357 of Zamárdi-Rétiföldek (photo: Simon Dupper).

14. ábra: Hangot adó női ruházat rekonstrukciója a Zamárdi-Rétiföldek 2357. sírja alapján (fotó: Simon Dupper)

are also known from the few Avar period pictorial sources (e.g., Vida 2017, Fig. 67.3). The overdress included some western elements, while still incorporating a Moshchevaja Balka cut. For example, the sleeves were shorter and wider, based on a depiction of a woman's dress in the 9th century Stuttgart Psalter (fol. 83v top).

Recreating the belt brought forward some interesting new insights. For example, the two fittings that were attached onto the horizontal part of the belt were necessary for practical rather than decorative purposes, since they helped stabilise the belt from the heavy weight of the attachments on the hanging strap. The strap end clapped against one of the fittings when in movement, also acting as an idiophone. Due to the different shapes and materials of the attachments – two bronze discs, two iron keys, and a bronze pellet bell – along with the

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clapping strap end, this belt creates many different types of sounds when in movement. Furthermore, this costume is to be used in science communication, showing that people of proto-history did not just wear simple rags, but instead liked to adorn themselves and had a complex visual and audial coding system using clothing. In addition, this costume was made to fit many sizes in order to allow people to experience wearing the garment. Though very subjective, it will be interesting to see how people feel wearing such noisy clothing, especially comparing introverted with more extroverted people.

Summary and conclusion

Pellet bells and bells (both belong to the group of idiophones) excavated from the Avar-age cemeteries Kaposvár-Toponár 40-es őrház, Kaposvár-Toponár-Fészerlak-puszta, Kaposvár-61. sz. út, Kaposmérő-Agyagbánya, Zamárdi-Rétiföldek and Vörs-Papkert B were investigated. Adding all excavated graves of all those cemeteries together the result is 4180 graves. Only 48 of them contained idiophones, which is 1.17%. Mainly women, followed by children, horses and men wear pellet bells or bells. Only 4.6% of the graves from the cemetery Kereki-Homokbánya contained idiophones and the distribution shows that women followed by children had pellet bells/bells and on the last position is only one man's burial. Three pellet bells from grave 634 of the cemetery Vörs-Battyáni Disznólegelő complete the study. It is a woman's grave. Most of the bells are very similar to Roman bell types und thus reinforce the assumption that people just used ancient Roman bells. They can be classified into three types. One conical shaped bell from Kaposmérő-Agyagbánya has its roots in Western Iran. Other cone-shaped bells and truncated shaped tutuli have pendants in Kiskőrös-Vágóhídi-dűlő, Hungary and Wien-Liesing/Carlbergergasse, Austria. The pellet bells are represented in shapes I, II, IV, V, VII and VIII with six different decorations. Their sizes are between 1.8–4.6 cm and with weights from 9.67–24.56 g. 31 pellet bells were analysed by using a handheld XRF spectrometer. They are forged from iron sheet and copper alloy sheet and cast in bronze, leaded bronze and gunmetal. The compositions are very heterogeneous. Pebbles, bronze balls and lumps of cinder serve as rattle bodies. Sound recordings were made of 15 bells and one bell. Their frequencies range from 0.8 up to 20 kHz. Their most pronounced partials are in the range between 1.6–6 kHz and thus in the sensitive range of human hearing. Their sound pressure level is between 65–85 dB, with one exception. Louder objects are sharper and more tonal than quieter ones. All in all, the idiophones can be described as sharp, hardly to mildly rough and bright. Some are equally tonal and noisy, but there are also some that have almost no noise components. There was one significant result: a higher amount of copper is associated with a lower value in frequency range f_{min}. Concerning spectral properties, the objects can be roughly divided into three groups, one being sounds with very pronounced partial frequencies, one being the less tonal sounds with less and broader partials and one with hardly any partials at all. The statistical analyses show that objects high in loudness and SPL are also higher in sharpness and tonality, but lower in roughness and impulsiveness. These analysis results reflect the current condition of the objects, some of which are less and some are more corroded. There is evidence that pellet bells and bells were attached to the belts with chains, ribbons ore strings. Especially in the graves from Zamárdi-Rétiföldek women wore a typical belt assemblage, known from Western to Eastern Europe, with ornamental discs, pellet bells or bells, spindles, knives, keys, needle cases and sometimes small bags. Hanging quite low, in the area of the knee or even lower, thus they must have dangled a lot while walking, drawing the eye to the area due to movement and also creating sounds with the objects clapping against each other. A recreation of the costume with the belt of grave 2357 brought forward that the two fittings that were attached onto the horizontal part of the belt were necessary for practical rather than decorative purposes, since they helped stabilise the belt from the heavy weight of the attachments on the hanging strap. The strap end clapped against one of the fittings when in movement, also acting as an idiophone. Pellet bells, imitating vascular fruits, originally were symbols for fertility, abundance and everlasting life. In the Avar period they "worked" as apotropaic amulets. Bells seem to have the same function. People liked to adorn themselves and had a complex visual and audial coding system using clothing. Pellet bells and tutuli in horse burials belong to the bridle. Mainly they were fixed on the head stall, in some exceptions also on the saddle or on the breast strap.

Contribution of authors

Beate Maria Pomberger Conceptualisation, Investigation, Visualization, Writing – Original Draft, Writing – Review and Editing. **Jörg Mühlhans** Investigation, Visualization. **Kayleigh Saunderson** Investigation, Visualization. **Viktória Mozgai** Investigation, Visualization, Writing – Review and Editing. **Bernadett Bajnóczi** Investigation, Writing – Review and Editing.

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ANALYSIS OF HUNGARIAN SILVER COINAGE OF THE 11TH-13TH CENTURIES, KEPT IN THE NATIONAL MUSEUM OF SLOVENIA

A SZLOVÉN NEMZETI MÚZEUM 11–13. SZÁZADI MAGYAR EZÜSTÉRMÉINEK VIZSGÁLATA •

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Abstract

A series of 24 Hungarian silver coins dated between the 11th and 13th century kept in the National Museum of Slovenia was analyzed by the method of PIXE using in-air proton beam. The analysis showed that most of the coins were made of high-quality silver except for the coins of anonymous issues during the 12th century. The analysis did not detect degradation during the period of Mongolic attacks in the mid-13th century, which points to the strict monetary politics of Béla IV, contrary to the issues in debased metal during the previous century. The analysis also did not detect silver marked with high levels of bismuth that might have reached Hungary from the west, but the slightly increased bismuth levels in certain coins suggested that this type of silver was diluted with silver from other sources. According to the admixture of gold, the silver metal was principally obtained from cerussite and oxidized ores.

Kivonat

A Szlovén Nemzeti Múzeum 24 példányból álló, 11–13. századi magyar ezüstérme gyűjteményét kivezetett protonnyalábos PIXE módszerrel vizsgáltuk. Kimutattuk, hogy az érmék többsége kiváló minőségű ezüstből készült, csupán néhány 12. századra datált, nem azonosítható példány bizonyult eltérő összetételűnek. A vizsgált leletegyüttes alapján a 13. század közepén nem mutatható ki a tatárjáráshoz köthető értékromlás, ami jelzi – a 12. századi csökkent értékű érmeösszetételekhez képesti – a IV. Béla uralkodása alatt fennálló stabil gazdasági helyzetet. A Magyar Királyságba ez idő tájt nyugat felől érkező, bizmuttartalmú ezüstöt nem azonosítottunk, azonban a több érmében érzékelhető, kismértékű bizmutdúsulás jelezheti a nyugati és más eredetű ezüstércek szándékos ötvözését. Az érmék aranytartalma az ezüst cerusszitos és más oxidos érc eredetre utal.

KEYWORDS: MEDIEVAL SILVER COINS, HUNGARY, PIXE

KULCSSZAVAK: KÖZÉPKORI EZÜSTÉRMÉK, MAGYARORSZÁG, PIXE

Introduction

The numismatic cabinet of the National Museum of Slovenia also keeps a series of Hungarian medieval silver coins. This enabled us to study their composition from the early issues on, yet for practical reasons and historical arguments we limited our study to the first three centuries, i.e. from the 11th to the 13th century. The main historical boundary in the 13th century is invasion of Mongols, who in 1241 inflicted a devastating defeat to the Hungarian knight army, led by their king Béla IV and his brother duke Coloman (Kálmán).

The defeat annihilated the armed forces, and the following raids caused numerous victims among the civilians.

Between 50 to 80 percent of the settlements were destroyed in the plains, and about 20–35 percent of population was lost in the woodlands (Makkai 1994a). According to a more conservative estimate, the total loss of population was between 15–25% (Laszlovszky 2018). We were interested in whether such fatal events were also reflected in the quality of contemporary coin issues.

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Fig. 1a: The analysed coins kept in the Numismatic Cabinet of the National Museum of Slovenia. The numbers as in **Table 1.**

1a ábra: A Szlovén Nemzeti Múzeum Numizmatikai Gyűjteményéből elemzett érmék. A számozás feloldása az **1. táblázat**ban.

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Fig. 1b: The analysed coins kept in the Numismatic Cabinet of the National Museum of Slovenia. The numbers as in **Table 1.**

1b ábra: A Szlovén Nemzeti Múzeum Numizmatikai Gyűjteményéből elemzett érmék. A számozás feloldása az **1. táblázat**ban.

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Our second motivation was based on our earlier study of the medieval silver coinage on the territory of the present-day Slovenia and the neighboring countries (Šmit & Šemrov 2006). In this study we characterized the quality of silver by predominant gold or bismuth impurities. Coins marked with bismuth concentrations up to 1.2 mass% were identified in the mints of Carinthia in Austria and in the mints on the Slovenian eastern border. The activity of these mints vanquished in the 13th century. We assumed that the Carinthian silver was traded to the eastern mints, which further supplied silver to the Hungarian kingdom on the east. The events of the 13th century, besides the Mongolian invasion also the ascendancy of Habsburgs, reduced the supply of silver to the east and thus caused the eastern mints to decline. The most current currency then became the Viennese pfennig. We were thus interested if the bismuth-primed silver can be traced in the Hungarian silver coins as well.

For comparison with our data, we used a comprehensive set of measurements of (Rácz et al. 2013) executed by X-ray fluorescence, though these data include a different set of elements and extend over a broader historical range up to the 15th century.

Methods

The analysis involved 24 coins (**Table 1.** and **Fig. 1.**) extending from the first Hungarian king István I (997–1038) to Béla IV (1235–1272) (Huszár 1979). For our study we distributed the coins into 5 specific groups: coins of the 11th century (István I and László I), coins of the 12th century (Kálmán I, Béla II and Béla III), anonymous coinage of the 12th century, coins of the 13th century (among them we included three bracteates that can be ascribed either to Béla III or Béla IV), while the fifth group included Slavonic banal denars from the king István V to Gutkeled Joakim. Beside the three bracteates and one obolus (András II), all coins were denars.

The coins were in different states of patination. For the measurement we selected the cleanest part of the coin, which was washed with alcohol. Measurements were taken on both sides of the coin, and the mean value of the results was calculated.

The analytical technique was in-air PIXE, which is frequently used for the analysis of historical silver coins (Daccà et al. 2000, Flament & Marchetti 2004, Rautray et. al 2011, Beck et al. 2017). The analytical procedures were applied at the Tandetron accelerator of the Jožef Stefan Institute in Ljubljana. The beam of 3 MeV nominal energy entered air through a Si_3N_4 window of 200 nm thickness. After passing about 1 cm air gap, the energy on the target was about 2.91 MeV. The proton current was a few tenths of nA and the measurements in particular point lasted about 500 seconds. The induced X-rays were detected by a Si(Li) detector of 140 eV resolution at 5.89 keV. The detector was positioned at 45º with respect to the target surface normal and 4.5 cm from the target. The detector was equipped with an aluminium absorber of 0.1 mm thickness; another absorber was an ice layer on the detector crystal which we presently estimate to 0.0025 g/cm^2. The X-ray spectra were fitted by the Xantho code (Šmit 2023), while the elemental concentrations were calculated according to the code presented in (Šmit et al. 2005), with an assumption that the mass percentages of individual elements sum to 100%. The accuracy of major concentrations was estimated to be within ±5% and those of minor concentrations within ±10%. The uncertainties are also larger for Zn in the presence of strong copper lines, as Zn Kα line is superimposed on the low-energy tail of Cu Kβ line. This also implied the detection limit for zinc about 0.05 mass%. The detection limits for other minor elements were 10 µg/g for Ni and 50 µg/g for Fe and the L-lines based elements (Au, Pb, Bi). The detection limits for the elements heavier than silver (Sn, Sb) were 100 µg/g.

As the effects of silver surface enrichment are known, it is necessary to check the measurements by a method that is bulk sensitive. Here we chose a simple density measurement, but which also represented a challenge as several coins have mass below 0.2 g. For the first attempt we chose the heaviest coins and performed weighting with a handy digital scale with a precision of 0.01 g. For determining the density of a bracteate that only weighted 0.18 g, we applied a scale with a precision of 0.001 g and designed a special hook for immersing the coin into water. The optimal solution was a hook of silvered copper wire of 0.2 mm thickness. The silvered surface eliminated the surface tension effects, while the known dimensions of the wire allowed to include the volume correction of the immersed part of the wire numerically. The density at the silver level (10.5 g/cm^3) was then measured with an accuracy of 0.4 g/cm^3.

Results

The analytical results are given in **Table 1.**; beside silver and copper, the trace elements detected were nickel, zinc, tin, antimony, gold and bismuth, while iron may be surface impurity from the earth or from the iron die. At one coin (anonymous of the 12th c.), a variable mercury content was also detected, signifying surface silvering.

The silver content for the coins, arranged in decreasing silver content, are shown on **Fig. 2**.

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Table 1.: Elemental concentrations in mass%. Dashes (-) denote the element was not detected. The colour scale denotes distribution into specific groups: 11th century (yellow, 1-2), 12th century (dark blue, 3-6), anonymous coinage of the 12th century (light blue, 7-13), 13th century (red, 14-18) and Slavonic bans of the 13th century (magenta, 19-24).

1. táblázat: Elemi összetétel adatok tömeg%-ban megadva. A nem kimutatott elemeket negatív jel (-) jelöli. Színkód: 11. sz. (sárga, 1-2), 12. sz. (sötétkék, 3-6), 12. sz.-i ismeretlen eredetű (világoskék, 7-13), 13. sz. (piros, 14-18) és 13. sz.-i szlavón bánsági (magenta, 19-24).

No.	Cat.No.		Fe	Ni	Cu	Zn	Au	Hg	Pb	Bi	Ag	Sn	Sb
1	45087	István I (997–1038)	0.22	-	2.9	0.04	0.12	-	0.56	0.04	96.2	-	-
2	45088	László I (1077–1095)	0.03	-	10.3	0.06	0.36	-	0.21	0.04	89.0	-	-
3	76289	Kálmán (1095–1116)	0.04	-	1.7	0.06	1.09	-	0.43	0.06	95.9	0.7	-
4	76293	Béla II (1131–1141)	0.06	-	16.2	0.25	1.18	-	0.54	0.02	81.7	-	-
5	14080	"	0.09	0.052	19.4	0.39	1.23	-	0.83	0.05	77.4	0.66	-
6	76288	Béla III (1172–1196)	0.13	0.012	6.49	0.04	0.56	-	0.83	0.15	91.8	-	-
7	51730	Anonymous 12th c.	0.04	-	5.49	0.13	1.78	-	0.68	0.04	91.4	0.45	-
8	45089	"	0.04	-	21.3	-	1.34	-	1.06	0.08	76.2	-	0.07
9	76292	"	0.04	-	49.3	-	0.77	-	1.67	0.07	48.1	-	0.07
10	51731	"	0.11	-	59.0	-	0.75	-	1.48	0.12	38.6	-	-
11	76287	"	1.31	-	91.5	-	0.45	0.23	0.78	0.03	5.44	-	0.26
12	51729	"	0.08	-	74.5	-	0.37	-	0.64	0.1	24.2	-	0.16
13	204173	"	0.86	-	72.0	0.40	1.53	-	0.85	0.01	24.2	-	0.23
14	204172	Béla III (1172–1196)-Béla IV (1235–1270)	0.02	-	2.24	-	0.11	-	0.25	0.01	97.4	-	-
15	76291	"	0.03	-	4.92	-	0.52	-	0.81	0.19	93.6	-	-
16	14081	"	0.02	-	8.17	-	0.51	-	0.17	-	91.1	-	-
17	51728	András II (1205–1235)	0.03	-	1.55	-	0.82	-	0.41	0.1	97.1	-	-
18	16087	Béla IV (1235–1270)	0.06	-	1.47	-	0.89	-	0.23	-	97.4	-	-
19	50858	Slavonic bans (1270–1272)	0.03	-	3.15	-	0.67	-	0.31	0.07	95.8	-	-
20	50855	"	0.03	-	5.11	-	0.27	-	0.36	0.14	94.1	-	-
21	79172	"	0.05	-	10.3	-	0.78	-	1.29	0.14	87.2	-	0.21
22	79173	"	0.02	-	8.98	-	0.87	-	0.3	0.03	89.8	-	0.04
23	26889	"	0.02	0.021	3.03	-	0.24	-	0.34	0.07	96.3	-	0.04
24	88323	"	0.02	-	1.48	-	0.32	-	0.13	0.05	98.0	-	-

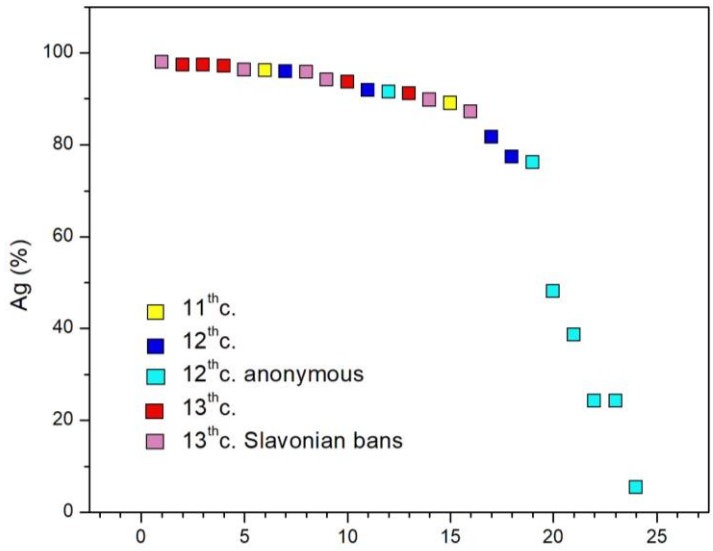

Fig. 2.:
Silver content (mass%) in the coins, re-arranged in decreasing silver concentrations

2. ábra:
Az érmék ezüsttartalma (tömeg%), csökkenő koncentrációérték szerint rendezve

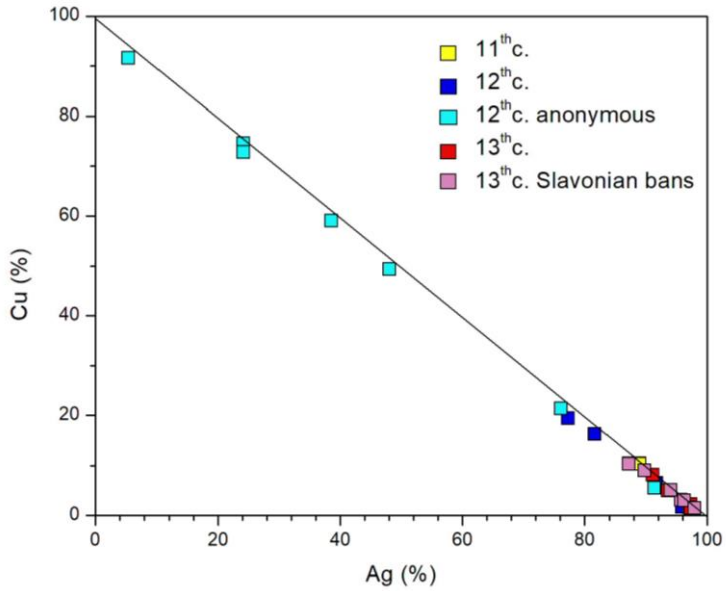

Fig. 3.:
Copper content (mass%) in the coins as a function of the silver content. The solid line corresponds to the binary copper-silver alloy.

3. ábra:
Az érmék réz- és ezüsttartalmának (tömeg%) viszonya. Az egyenes a réz-ezüst kétfázisú rendszert jelöli.

We can see that most coins are made of high-quality silver, exceeding 90 mass% Ag. Markedly different are coins of the 12th century, namely two coins with king names (while two are among the high-Ag group) and all anonymous issues. In the study of (Rácz et al. 2013), a compositional difference was detected before and after the reign of András II (1205-1235). We comment that this difference reflects the occurrence of low-grade coins during the 12th century.

In **Fig. 3.**, a relation between copper and silver is shown. One can easily observe that all measured points lay on a straight line, characteristic for the binary copper-silver alloy. The density measurements for the heaviest two coins (László I and Béla IV) showed the densities of 10.1 ± 0.5 g/cm^3 and 10.4 ± 0.5 g/cm^3. For the lighter bracteate of Béla III/Béla IV, a measurement with a more precise scale gave 10.4 ±0.4 g/cm^3. These three values confirm that the high-silver values measured at the surface extend into the bulk region. This is not obvious, as silver enrichment of the surface could have been present (Beck et al. 2004, Beck et al. 2008, Hrnjić et al. 2020). Small Celtic silver coins from Slovenia, showing uniform silver content on the surface, in total contain much less silver, pointing to a two-head distribution (Šmit et al. 2020). Medieval coins may even be made by hammering together brass and silver foils (Trela et al. 2025). Surface enrichment is less prominent at high-grade silver, besides several trace elements, such as gold, bind to silver; these effects were exploited in the study of small silver coins from the classical Greece (Šmit &Šemrov 2018).

As for the bracteate coin it is also tempting to compare the measured density to the silver content,

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as calculated from the semi-empirical relation that connects the alloy density with silver percentage in binary and ternary alloys (Kraut & Stern 2000). The density of 10.4 g/cm^3 corresponds to 94 mass% Ag in the silver-copper alloy, which is close to our surface values.

Discussion

Further properties of the coins were studied from the admixtures of traces elements, notably bismuth and gold.

As several coins (of the 12th century) contain lesser quantities of silver, we normalized the contents of Au and Bi with respect to that of silver; for low Au and Bi concentrations, these ratios correspond to mass percentages in the bulk silver. **Fig. 4.** shows that Bi/Ag ratios are smaller than 0.2%, and Au/Ag ratios smaller than 1%. Marked exceptions are again coins of the 12th century. Bi/Ag ratios for three coins are between 0.3 and 0.6%, while Au/Ag ratios exceed 1%, with the maximum values for two anonymous coins between 6 and 8%; however, these two high values are consequence of normalization to very low silver concentrations.

For further comparison with the silver coins from Slovenian mints we rely on the plots of kernel density estimate (KDE) (Šmit &Šemrov 2006). The results are shown in **Fig. 5**. The Hungarian coins peak at around 0.1% Bi/Ag, with a small contribution towards 0.2%, while the values above this value correspond to the three coins that excel in **Fig. 4.** as well. On the other hand, the mints from the territory of the present-day Slovenia can reach higher values, up to 1.2 mass% Bi (these values were not normalized to the silver content as it normally exceeded 85 mass%, Šmit & Šemrov 2006). These results suggest that the Bi-marked silver was used in Hungarian coins on a rather small scale and was probably diluted by silver from other sources. In a much larger set of data that involves coins up to the 15th century (Rácz et al. 2013), 38 coins out of 304 exceed the level of 0.2 mass% Sb (a fraction of 12.5%), and only six (2%) are in the range of 0.5–0.97 mass% Sb.

The gold content may to some extent reveal the source or the type of silver ore. In Antiquity and Middle Ages, silver was mainly extracted from lead-silver ores by the method of cupellation, which is based on the oxidation of metal silver until more noble silver remains.

In the study of (Wood et al. 2017), the distinction between the silver obtained from galena (PbS) or cerussite $PbCO_3$ (including oxidized ore) was determined from the gold and iridium concentrations. Though the study was based on Byzantine and Sassanian silver, the results can be deduced to our study as well; however, we can only rely on the gold values as the low levels of iridium are undeterminable by X-ray methods. The critical value is 0.1 mass% Au – concentration below this level signify galena as silver source, higher gold concentrations are significant for cerussite or oxidized ore.

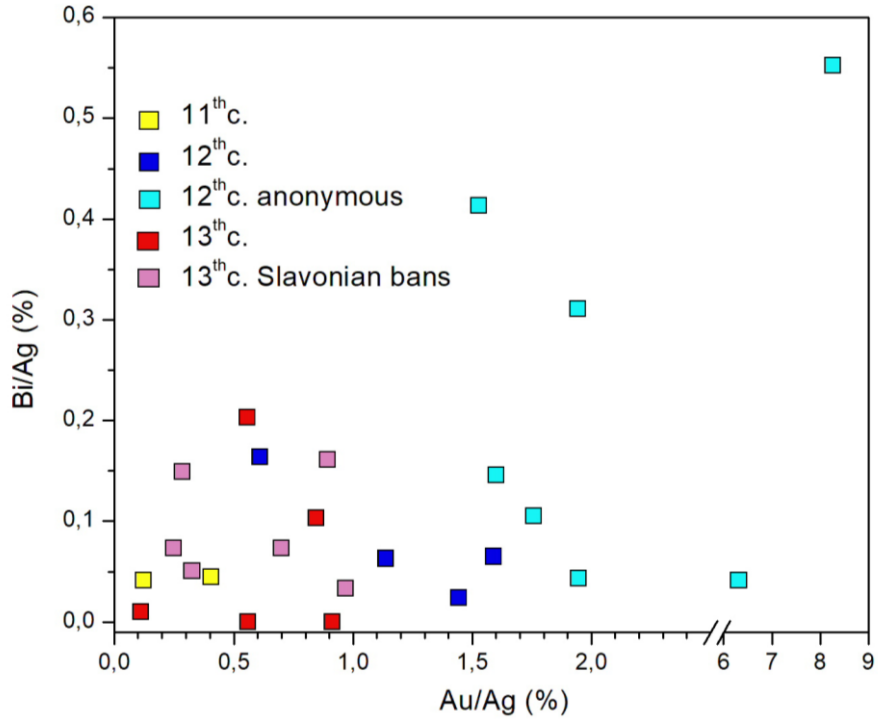

Fig. 4.: Relative bismuth and gold concentrations, normalized to the percentage of silver

4. ábra: Relatív bizmut- és aranytartalom, az ezüsttartalomra normálva

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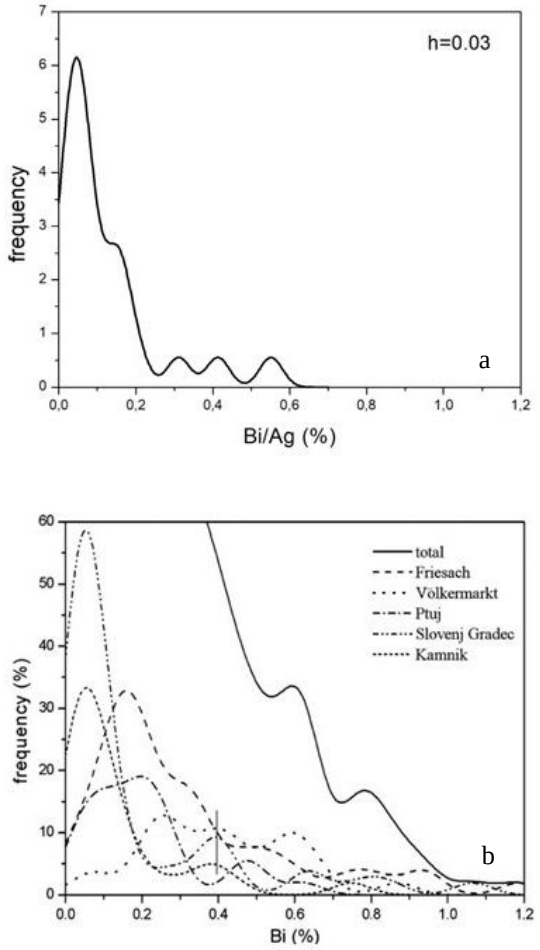

Fig. 5.: Kernel-density-estimate for the bismuth content. **a** – Hungarian coins; **b** – coins from the selected mints given in (Šmit & Šemrov 2006).

5. ábra: A bizmuttartalom magfüggvényes sűrűségbecslése (kernel density estimate, KDE) **a** – magyar ezüstérmékre és **b** – ismert verdék érméire (Šmit & Šemrov 2006).

Fig. 6. shows comparison of KDE for gold in Hungarian coins and the contemporary coins from the mints given in (Šmit & Šemrov 2006). In this case we calculated KDE for the Au/Ag ratio and the net Au content. Both distributions are similar, the difference is for the high-Au side. The distributions seem to be two-fold, suggesting two distinct silver sources.

Absolute Au concentrations extend up to 2 mass%, which is higher than for the mints in (Šmit & Šemrov 2006). It is easily observable that the fraction of the coins with less than 0.1 mass% Au is negligible (only two coins slightly exceed this limit), which excludes galena as an important source of metal silver.

The quality of the cupellation procedure is studied in the binary diagram (**Fig. 7.**) which shows the ratio of Pb/Ag with respect to Au/Ag. **Fig. 7.** shows approximate linear correlation. The values of Pb/Ag

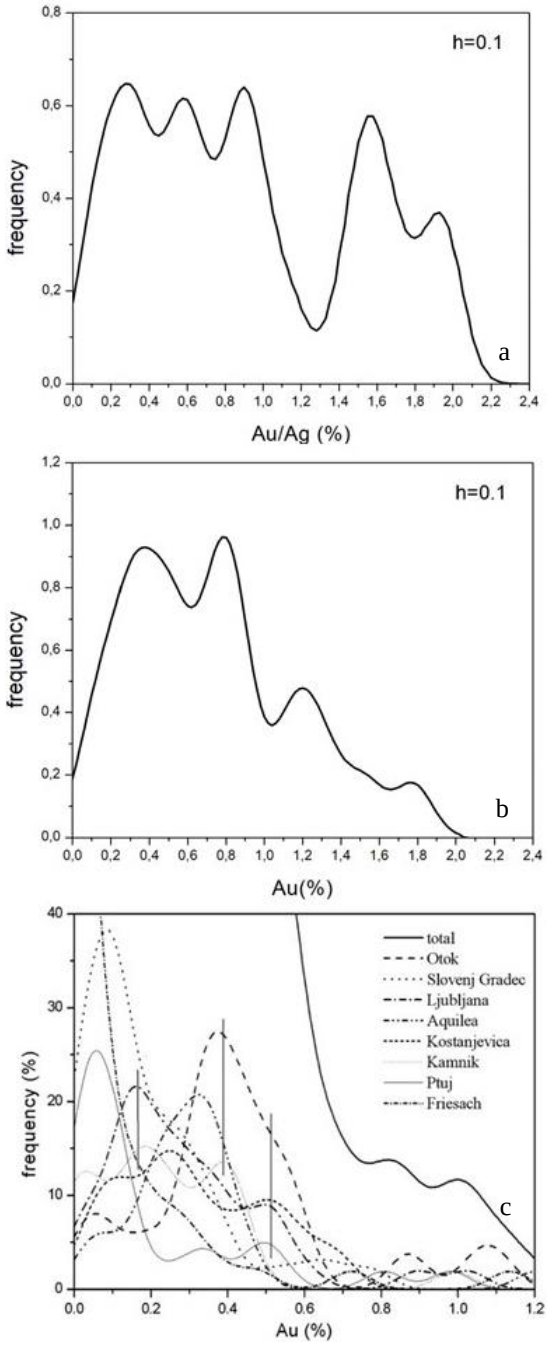

Fig. 6.: Kernel density for the gold content. **a** – relative gold content (with respect to Ag) for Hungarian coins; **b** – absolute gold content for Hungarian coins; **c** – absolute gold content for the mints in (Šmit & Šemrov 2006).

6. ábra: Az aranytartalom magfüggvényes sűrűségbecslése (KDE) **a** – magyar ezüstérmék relatív aranykoncentrációjára, **b** – magyar ezüstérmék abszolút aranykoncentrációjára és **c** – ismert verdék érméinek abszolút aranykoncentrációjára (Šmit & Šemrov 2006)

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above 1.5% up to 15% are again characteristic of the anonymous coins of the 12th century. We may guess that the highest lead concentrations were not consequence of ineffective cupellation process, but lead was added intentionally to dilute the much more costly silver.

Coins of the 11th–12th century contain detectable amounts of zinc, with the highest values for the two coins of Béla II and two anonymous coins of the 12th century. Zinc may denote the presence of brass in the alloying copper.

Coins of the 12th century further contain small admixtures of tin and antimony. **Figs. 8** and **9** show the concentrations of both elements as a function of silver content. It is interesting to note that the higher content of tin (around 0.6 mass%) appears in two coins with names and in one anonymous coin (the only one with a high Ag content). Higher tin concentrations are then characteristic for the coins of high silver concentration. The reason for this is unknown. High tin concentrations were observed in small Celtic coins (Šmit & Kos 1984) and in Roman *victoriati* (Laharnar et al. 2017); the reason for the former was probably alloying silver with bronze instead of pure copper, while for the latter it was diluting of silver with cheaper tin. We may conjecture that the employment of bronze was also the source of tin in our case. An opposite behavior is observed for antimony. In this case the highest antimony concentrations (up to 0.25 mass%) are observed in the coins with the lowest silver concentrations. We expect that antimony in this case was admixture of the added lead, probably not so much from the ore, but likely by using the metal from some lead objects, which were made harder by addition of antimony.

At last, we check if the distinction of the 12th century coins can be stated as a collective property. For this we employ the methods of principal component analysis (PCA), and, since our five groups were pre-selected, the method of linear discrimination (LDA), using the concentrations of Sn, Sb, Au, Pb and Bi. **Fig. 10.** (for PCA) confirms that the coins of the 12th century (except for one coin with a name and two anonymous ones) form a separate group, located at the right side of **Fig. 10**. Among them is also one coin of the Slavonic bans. LDA (**Fig. 11.**) shows a similar result: while most of the coins with a high silver content are closely grouped, there is a diffuse group on the right side of **Fig.11.** that contains all 12th c. anonymous coins and the already mentioned coin of the Slavonian bans (no. 21). The reason for its position is relatively high concentrations of lead (1.29 mass%) and antimony (0.21 mass%). The two coins with names of the 12th c. are located between the two groups.

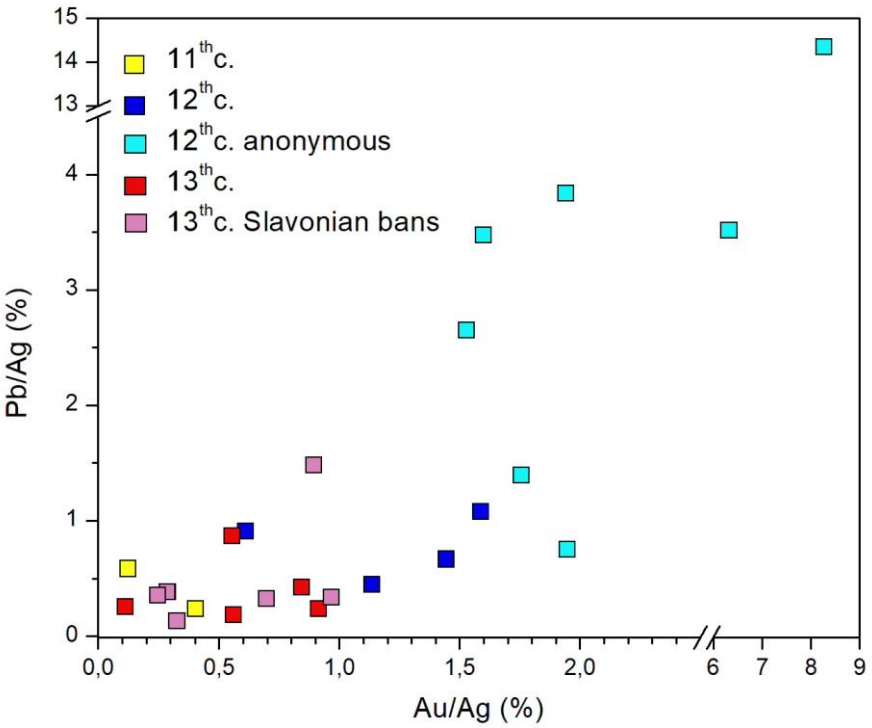

Fig. 7.: Relative lead content as a function of the relative gold content

7. ábra: Relatív ólom- és aranytartalom az ezüstkoncentrációhoz viszonyítva

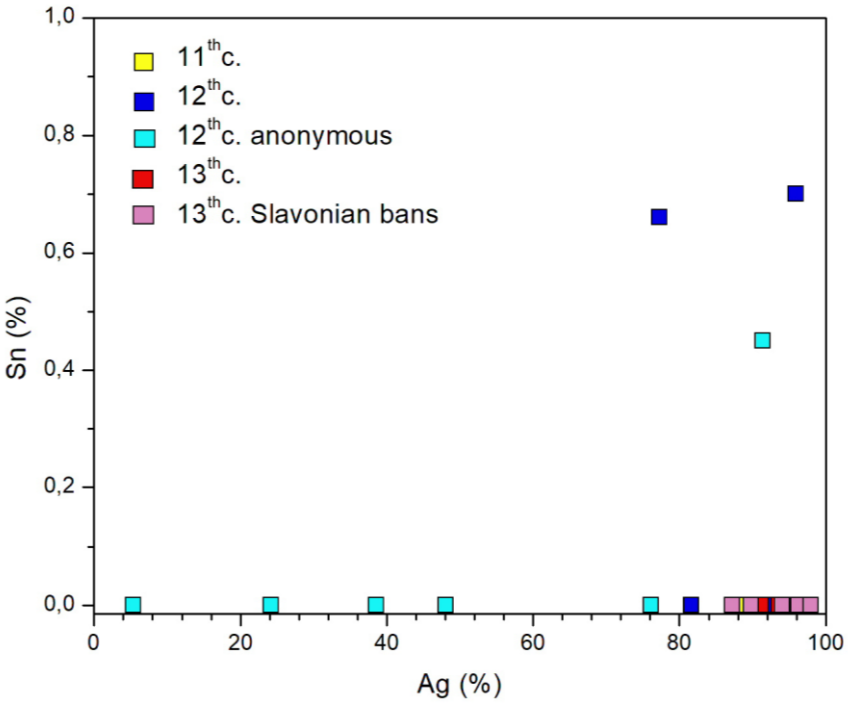

Fig. 8.:
Tin content (mass%) as a function of silver content (mass%)

8. ábra:
Az érmék ón- és ezüstkoncentrációja (tömeg%)

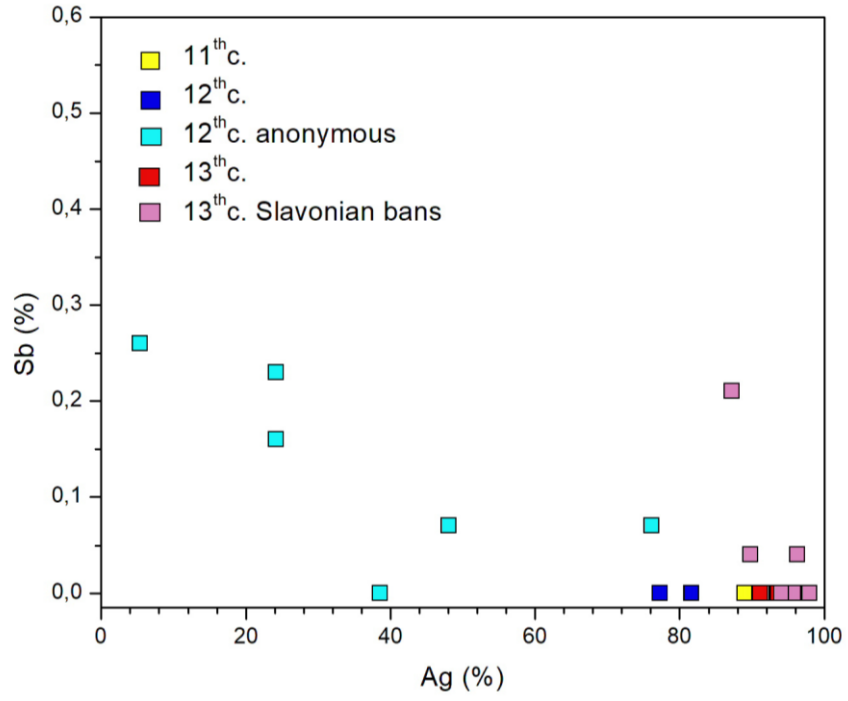

Fig. 9.:
Antimony content (mass%) as a function of silver content (mass%)

9. ábra:
Az érmék antimon- és ezüstkoncentrációja (tömeg%)

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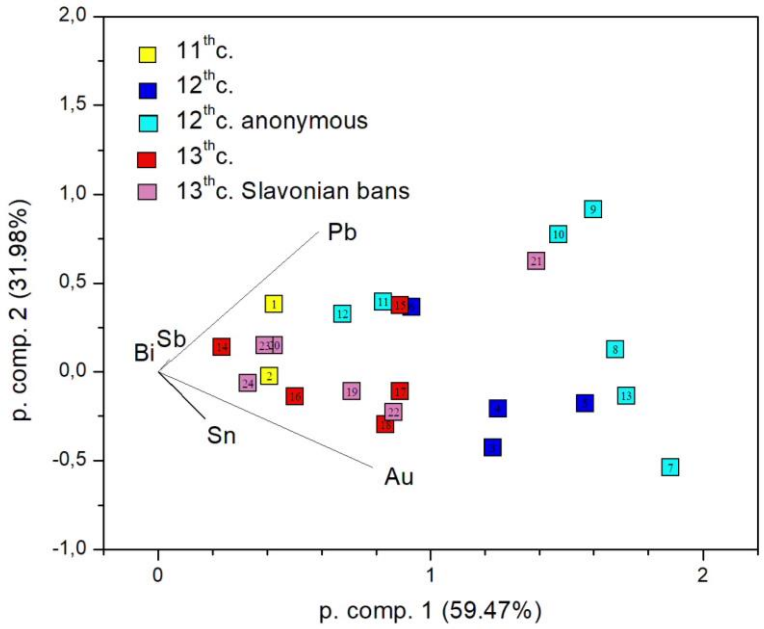

Fig. 10.: Principle-component analysis of the coins concerning the contents of Sn, Sb, Au, Pb and Bi. The first two components retain 91.45% of the total variation

10. ábra: Az érmék Sn, Sb, Au, Pb és Bi koncentráció-adatainak főkomponens analízise. Az első két komponens a változékonyság 91.45%-át fedi le.

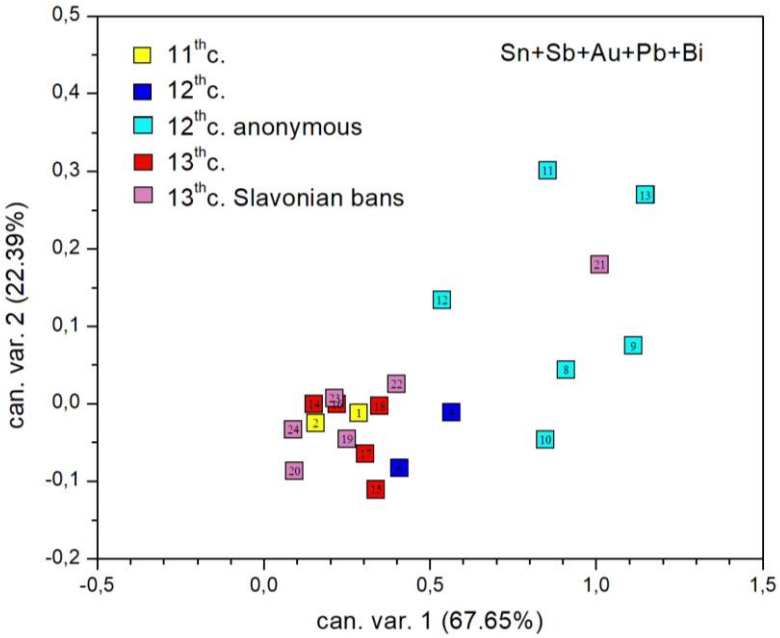

Fig. 11.: Linear discriminant analysis of the coins, concerning the contents of Sn, Sb, Au, Pb and Bi, and relying on the distribution of five distinct groups

11. ábra: Az érmék Sn, Sb, Au, Pb és Bi koncentráció-adatainak lineáris diszkriminancia analízise az öt régészeti csoport jelölésével

Conclusions

The analyzed coins belonging to the period of the 11th–13th century are produced of high-quality silver, with the silver content exceeding 90%. Exceptions are the coins of the 12th century, which are produced of low-quality silver, with a higher percentage of impurities of lead, tin and antimony. On one coin, the presence of mercury signifies silver plating with silver-amalgam or later processing with liquid mercury with an aim to improve the 'silver' look. This coin may be regarded as forgery.

There is no debasement in the 13th century, which confirms the stable monetary politics of Béla IV during the Mongolian attack. This complies with the explanation that several, mostly wooded western areas of Hungary remained unaffected by the invasion. The king, after a short flight to Dalmatia, was able to raise a military campaign for recapturing the territories of Friedrich II Babenberg of Austria just two years after the unexpected Mongol withdrawal in 1242 (Makkai 1994a).

The bismuth-primed silver was not directly detected. Three coins with an increased bismuth concentration do not meet the highest level of 1.2 mass% Sb detected in the coins minted beyond Hungarian western borders (Šmit &Šemrov 2006), so we may conclude that this type of silver was only traced in the recycled material, circulating

during the 12th century. High gold concentrations above 0.1 mass% exclude exploitation of galena lead ore as a source of silver but point to cerussite or oxidized ore.

It is certainly challenging to discuss why the low-quality silver coins and forged coins did not appear during the politically and economically critical period, but during the century when the economy was prospering (Makkai 1994b). We may conjecture that the expanding economy was requiring large quantities of currency but there was simply not enough silver on the market, which was then characteristic for the whole continent (Laszlovszky 2021). This had stimulated anonymous issues of the coins, while the lust for profit fostered minting coins with fraudulently lower silver content.

Contribution of authors

Žiga Šmit Investigation – Formal analysis – Data curation – Writing – Original Draft, Review & Editing. **Andrej Šemrov** Resources – Writing – Original Draft, Review & Editing.

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BULK PHASE AND CHEMICAL COMPOSITION ANALYSIS OF TWO 19TH CENTURY RUSSIAN PLATINUM COINS BY NEUTRON AND COMPLEMENTARY METHODS

19. SZÁZADI OROSZ PLATINAÉRMÉK TÉRFOGATI FÁZIS ÉS KÉMIAI ÖSSZETÉTEL VIZSGÁLATA NEUTRONOS ÉS KIEGÉSZÍTŐ MÓDSZEREKKEL•

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Abstract

The composition of early 19th century platinum coins reflects the advances of platinum metallurgy from laboratory-scale refining to industrial-scale processing. An array of neutron methods was used to determine for the first time the bulk phase and elemental composition of two coins, dated 1837 and 1838. Neutron imaging confirmed the homogenous internal structure of the metal at the micrometer scale, with further support from small-angle neutron scattering at the nanometre scale. Prompt gamma activation analysis showed the coins to consist of technically pure platinum, appropriate for the time of their production with iridium and iron between 1 and 2 weight percent. Lower amounts of copper, rhodium, manganese and gold were also detected in the range of tens to thousands of μg/g. Overall, the investigation demonstrated the suitability of neutron methods as non-invasive and reliable bulk means to determine the composition of early industrial platinum to track the development of refining processes and metallurgical processing.

Kivonat

A 19. század elejéről származó platinaérmék összetétele tükrözi a platinakohászat fejlődését a laboratóriumi méretű finomítástól az ipari méretű feldolgozásig. Tanulmányunkban két, 1837-ből és 1838-ból származó érmén végzett első neutronos vizsgálatok térfogati fázis- és elemi összetétel eredményeit mutatjuk be. A neutronos képalkotás megerősítette, hogy a fém belső szerkezete mikrométeres skálán homogénnek tekinthető. Ezt az eredményt tovább erősítették a kisszögű neutronszórás eredményei, melyek nanométeres léptékben igazolták ugyanezt. Prompt gamma aktivációs analízissel meghatároztuk, hogy az érmék tiszta platinából állnak 1–2 tömegszázalékos iridium és vastartalommal, amely jellemző a gyártásuk időszakára. Az érmékben ezen kívül kisebb, tíztől ezer μg/g-os mennyiségben mutattunk ki rezet, ródiumot, mangánt és aranyat. Vizsgálataink igazolták, hogy a neutronos módszerek teljesen roncsolásmentes módon adnak megbízható eredményeket a korai ipari platina térfogati összetételéről, ezen túlmenően a finomítás és a kohászati eljárások fejlődésének nyomon követésére is alkalmasak.

KEYWORDS: NON-DESTRUCTIVE ANALYSIS, PLATINUM COIN, SANS, PGAA, ToF-ND, PIXE

KULCSSZAVAK: RONCSOLÁSMENTES VIZSGÁLAT, PLATINA ÉRME, SANS, PGAA, ToF-ND, PIXE

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Introduction

During the first half of the 19th century, platinum refining and processing made major progress and evolved from a small-scale though profitable laboratory-based niche technology (Wollaston 1829) to a scale sufficient to sustain nearly two decades of routine coin production (Sobolewsky 1835), totalling nearly 50 tons of metal (McDonald & Hunt 1982). In a climate of intense academic competition in fundamental research into platinum chemistry and the associated commercialisation of platinum metal production few contemporary reports exist detailing the research and development conducted at the different research centres of the time (Schneider 1868). In particular, issues of quality control and consistency of operation are not documented on a level sufficient to trace incremental progress in the operations, or the implementation of specific innovations even where they are mentioned in the literature. A series of papers using primarily X-ray methods on a set of seven Russian 3-Rouble coins dated from 1828 through to 1842 provided an initial framework to identify these well-dated issues as the most promising and unbiased material archive of the development of platinum metallurgy during the critical period in the first half of the 19th century AD (Auer et al. 1998; Rehren 2006). To trace the technical development on a scale sufficient to be representative for the metal production for each year, and to identify the potential effects of the reported modifications in the production process would require a much larger set of coins to be studied, to understand the consistency in composition within each year's issue, and then among the different issues as time progressed. Also, the analyses need to be non-invasive and without the distorting effects of intentional surface modification, such as stamping and pickling as part of the production process, and the subsequent alterations through wear and tear.

Neutron methods are uniquely suited to determine key physical and chemical aspects of objects without the need for invasive sampling, and without being limited to the surface only, even for such challenging materials as metallic platinum. This pilot study examined the utility of a range of neutron methods to address research questions related to the development of platinum metallurgy at the Royal Mint of St Petersburg in Russia, which spearheaded the development of platinum refining and processing from the laboratory scale of western chemists to a routine large-scale operation. Targeted research used prompt gamma activation analysis (PGAA) to determine the bulk composition of two coins, complemented by particle-induced X-ray emission (PIXE) to correlate surface composition to bulk composition. High resolution time-of-flight neutron diffraction (ToF-ND) was used to determine the phase composition of the coins, focussing on the presence of a few volume percent of iron oxides and possible shifts in the cell parameters of the platinum matrix as a result of the presence of alloying components in the metal. Neutron imaging was performed to ascertain the coarse-grain (>100 μm) homogeneity of the coins' interior, and small-angle neutron scattering (SANS) was used to explore the homogeneity of the metal matrix on the nanoscale, confirming or contradicting the existence of nanometric-size second phases or precipitates, as well as the texture orientation of the coins.

Two coins of the original set studied in the 1990s and early 2000s were available for this pilot study. Issued in 1837 and 1838, respectively, they weighed just over 10 grams each with a density of just over 20 g/cm^3 and showed a noticeable magnetic response to a hand-held magnet, indicating the presence of a significant iron compound (Auer et al. 1998). They had been thoroughly studied before using primarily X-ray based methods (wavelength-dispersive and energy-dispersive X-ray Fluorescence spectrometry, X-ray Diffraction). These methods indicated that the coins consisted of technically pure platinum metal (96 to 98 wt%) with impurities of iron and iridium in the order of 1 to 2 wt% each, and lower impurities of a wide range of PGEs and transition metals (Rehren et al. 2012). This was further supported by invasive metallography for a single coin (1837), identifying the presence of various iron oxide inclusions throughout the matrix of that coin but not near the surface, probably due to the pickling of the metal discs prior to striking (Weerd et al. 2004). These investigations demonstrated the real potential of compositional and phase analysis of these coins, while emphasising the necessity to apply whole-body analyses rather than being restricted to surface-only approaches.

Materials and methods

PGAA and off-line counting measurements

Prompt gamma activation analysis (PGAA, Révay & Belgya 2004) (**Fig. 1.**) is a non-destructive nuclear analytical technique to determine elemental compositions (Révay 2009). It has been previously applied mostly in provenance or technological studies to characterise heritage objects made of stone (Kasztovszky et al. 2022) and glass (Moropoulou et al. 2016). PGAA proved to be successful in the analysis of alloys (Kiss et al. 2015) even with micro- and macroscopic heterogeneities (Tarbay et al. 2021). The main benefit of PGAA is that due to the highly penetrating properties of neutrons and gamma photons, its results represent the entire irradiated volume instead of only the near-surface volume. Therefore, it was chosen to determine the true bulk chemical composition of the coins unaffected by surface treatments.

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During analysis, the whole coin was irradiated in a cold (i.e. low energy) neutron beam and the gamma-rays from the radiative capture were detected. In most cases, major components and a few significant trace elements can be quantified from one spectrum. Elements have fingerprint-like prompt gamma spectra with gamma peaks of 50 keV–10 MeV energy. In principle every chemical element (except He) can be detected with the standard PGAA set-up that includes a high-purity germanium detector. Due to their more complicated spectra, elements with higher atomic number (Z>30, except for Sn and Pb) can be analyzed more precisely using a low-energy germanium detector (LEGe) with superior energy resolution (Maróti et al. 2016), since these elements have their strongest peaks in the low energy region. During the neutron irradiation of the prompt gamma measurements, short and medium half-life radioactive nuclides might form. Thus, PGAA can be complemented with a subsequent series of off-line counting measurements to improve detection of minor impurities within the platinum matrix – if these have short or medium half-life nuclides. This method is called in-beam activation analysis (Szentmiklósi et al. 2008; Révay et al. 2015). The PGAA measurements were carried out at the NIPS-NORMA station (**Fig. 1c.**) of the Budapest Neutron Centre (Szentmiklósi et al. 2010; Szentmiklósi et al. 2013; Kis et al. 2015) using a Compton-suppressed LEGe detector (Maróti et al. 2016), which was successfully applied earlier on a large set of silver coins (Šmit et al. 2020). The intensity of the neutron beam, characterized by the thermal equivalent flux, was 2.7×10^7 cm^{-2} s^{-1}, the cross section of the neutron beam was set to 10×13 mm, thus, almost the entire coin volumes were irradiated.

The gamma-ray spectra were evaluated using the Hypermet-PC program. The spectroscopic data library used in the analysis was established earlier at the Centre for Energy Research (Révay & Molnár 2003; Révay et al. 2004). As the data library contains the first 42 most intense platinum prompt gamma peaks down to about 1% relative intensity, a pure (99.97) platinum foil was analysed immediately after the two coins to detect its weaker prompt gamma peaks. For the detection of elements with lower cross section in the platinum matrix, these peaks need to be adequately corrected for. The compositions of the analysed samples were determined using the methods described in Révay (2009), while the uncertainties of the concentrations were calculated according to Révay (2006).

The off-line counting measurements were carried out in a low-background chamber (Kis et al. 2013) using an unsuppressed 13% HPGe detector (**Fig. 1d.**). The calculation of concentrations relied on the existing k_0 database (Jaćimović et al. 2014; https://www.kayzero.com/k0naa/k0naaorg/k0-ISC.html), a different set of nuclear data (Farina Arboccó et al. 2014) as well as on the general formulae of INAA simplified to subthermal activetion, containing the saturation, decay and counting factors, described in more detail in Szentmiklósi et al. (2008) and Révay et al. (2015).

PIXE

External milli-beam particle induced X-ray emission spectroscopy (PIXE) is successfully used for non-invasive elemental surface analysis (Gyódi et al. 1999). The characteristic X-rays produced by the interaction of the incident proton beam with the material at the selected surface area are used for quantitative analysis of the irradiated volume. Due to the deceleration of the incident protons in the sample and the attenuation of the out-coming X-rays the method is inherently sensitive only for a near-surface region of depths up to some tens of micrometres, depending on the composition of the sample, the primary proton energy and the energy of the characteristic X-rays. The ultra-heavy matrix of the coins analysed here limited the analytical depth to a few micrometers. We chose to use this method in order to compare the surface composition to the bulk composition; any systematic differences are likely the result of the intentional surface pickling of the coin blanks and thus bear information on the final step of the coin production. Furthermore, the comparison between bulk and surface composition would indicate whether neutron methods are really necessary for the research question in hand.

In standard detection arrangement elements from Al to U can be detected simultaneously, in favourable conditions down to mg/g sensitivities (Johansson et al. 1995). Our PIXE measurements were performed at the 5 MV Van de Graaf accelerator of the HUN-REN Wigner Research Centre, Institute of Particle and Nuclear Physics. A closely collimated proton beam of 2.5 MeV energy was extracted from the evacuated beam pipe to air through a 7.5 μm thick Kapton foil. A target-window distance of 10 mm was chosen for the measurements at which point the beam diameter was found to be about 1.5 mm, using an external beam current in the range of 1–10 nA.

The PIXE measurements were done on the reverse side of the coin as well as on the cross section of the cut rubel (see **Fig. 1a-b.**). The characteristic X-ray spectra were recorded by a computer controlled Amptek X-123 spectrometer with an SDD type detector of 25 mm^2 × 500 μm active volume positioned at 135° to the beam direction. The energy resolution was 130 eV for the Mn Ka line. The net X-ray peak intensities were calculated into element concentrations using the off-line GUPIX program package (Campbell et al. 2000).

Fig. 1: Experiment photos. **a-b** Using PIXE the middle part of both platinum coins and the cut inner surface was analysed with a 1.5 mm diameter beam. **c** During the PGAA measurements, nearly the entire volume was irradiated in the NIPS-NORMA sample chamber. **d** The off-line counting measurements after the neutron irradiation took place at the low-background chamber.

1. ábra: Az alkalmazott kísérleti berendezések. **a-b** PIXE módszerrel az érmék külső felszínének közepén, illetve az érmék vágott keresztmetszetén végeztünk méréseket. **c** A NIPS-NORMA mintakamrában elvégzett PGAA mérések során az érmék közel teljes térfogatát besugaraztuk. **d** A méréseket követő off-line gamma spektrometriára a mérőhely mellett található alacsony hátterű mérőkamrában került sor.

ToF-ND

The ToF-ND at the Budapest Neutron Centre is a high-resolution time-of-flight neutron diffractometer capable of analyzing lattice parameters within the bulk of a heavy metal object. We used this specifically to determine the mineralogical nature of the iron oxide inclusions, and to determine potential cell parameter shifts of the platinum matrix as a result of the unintentional alloying of the metal with residual impurities from the refining process.

The fast double choppers of the ToF-ND instrument can produce neutron pulses as short as 10 ms; the total flight path of neutrons to the detectors is 25 m. In the highest resolution mode and back scattering geometry diffraction spectra with peak widths of 1.5×10^{-3} Å can be collected. The data acquisition consists of an event recorder registering all events (neutron capture, chopper signs and any external signal) together with a time stamp to a file. This method facilitated recording Bragg-diffraction peaks, i.e. the angular distributions of grains' orientations, thus separating larger crystallites from powder-like phases. The instrument is generally used to investigate much larger samples; to increase sensitivity in this case we applied very long measuring times, so that the detectable absolute mass of iron oxides was as low as about 10 mg, i.e. less than 0.1% of the measured sample mass.

SANS

Small-angle neutron scattering was used for the nanoscale structural analysis of the coins. The SANS method provides information about the nanoscale inhomogeneities, averaged over the whole measured volume. From the neutron scattering perspective, an inhomogeneity (also referred to

as a scattering domain) is a nanosized region of the material whose neutron scattering length density differs from that of its surrounding matrix. SANS yields insight into the size, shape, and surface characteristics of these scattering domains in a non-destructive way.

In a SANS experiment the neutrons scattered elastically and coherently in angles smaller than 10 degrees are collected by a two-dimensional (2D) neutron detector.

The neutron intensity is recorded as a function of the scattering vector Q, which is defined as the difference between the wave vectors of the incident and scattered neutrons, and is given by Equation 1

$$Q = \frac{4\pi}{\lambda} sin \frac{\theta}{2} \qquad \text{Eq. 1}$$

where λ is the wavelength of the monochromatic neutron beam, and Θ is the scattering angle.

The 2D scattering patterns recorded by the detector may appear symmetric around the center of the neutron beam (isotropic scattering) or exhibit asymmetry, indicating that the structural properties of the sample vary along different directions (anisotropy) (see section 'Nanostructure').

In isotropic cases, the recorded neutron intensity is radially averaged, and after the appropriate calibration routine, an intensity versus Q curve is generated (see section 'Nanostructure'). The shape of this curve will provide adequate information about the nanoscale structure of the sample. The quantitative evaluation of the one-dimensional SANS curves is made by the least-square model fitting method (Len et al. 2022).

The measurements were performed at the YS-SANS instrument of the Budapest Neutron Centre (Len & Almásy 2019). The used sample-to-detector distances were 1.13 m and 5.25 m, and the used wavelengths were 4.4 Å and 8.6 Å. The samples were placed in the beam as received.

Both coins showed anisotropy at the largest measured Q range (see section 'Nanostructure'). As the anisotropy was only perceived at the high Q range, the overall data evaluation was done in 1D. The radially averaged curve's middle and high Q range parts were model fitted with the aid of the power-law model, which is used for shape-independent scattering domains (Len et al. 2022).

Neutron Radiography

Neutron radiography (NR) is based on the attenuation of a neutron beam prior to its recording on a 2-dimensional screen. It is a direct imaging technique, where the visual representation of an object is obtained by detecting the modification of the incident beam as it passes through the matter (Banhart 2008; Anderson et al. 2009). The interactions between the radiation and the object determine the contrast, revealing the internal structure of the sample. Here, we used NR in order to test whether there are compositionally distinct particles trapped within the matrix of the coins which would indicate a potential by-passing of the laborious refining process for some of the raw platinum ore, or other fraudulent adulteration of the metal.

A setup called NORMA was installed at the Budapest Neutron Centre as a part of the NIPS experimental station in 2011 (Szentmiklósi et al. 2013; Kis et al. 2015), where the thermal equivalent flux of the guided cold neutron beam is about 2.7×10^7 cm^{-2} s^{-1} and the cross-sectional area of the neutron beam, i.e. the maximum area to capture an image is 43×43 mm^2. The coins were positioned immediately in front of the screen downstream of the neutron collimators, and the transmitted neutrons created signals in a two-dimensional position sensitive detector, i.e. a ^{6}LiF/ZnS(Cu) scintillator coupled with a Peltier-cooled Andor CCD camera, located behind the sample. The spatial resolution (about 330 μm) of the imaging system was limited by the divergence of the neutron beam (L/D = 233). To compensate for the spatial inhomogeneity of the beam and internal scintillation effects, the raw two-dimensional digital image was normalized (see **Fig. 2.**) both with the open beam profile recorded in the absence of the sample and with the dark image recorded with closed neutron beam.

Results and discussion

Bulk and surface composition

PGAA results verified that the coins contain 96 weight% platinum, minor amounts of Ir and Fe and further Cu and Rh content below 1 weight% quantity. With the off-line counting measurements, besides the decay gamma-lines of ^{199}Pt ($T_{1/2}$=31 minutes) and ^{194}Ir ($T_{1/2}$=19.3 hours), the off-line counting measurements detected ^{198}Au ($T_{1/2}$=2.7 days) and ^{56}Mn (2.6 hours), as well as ^{104}Rh ($T_{1/2}$=42 seconds). The amount of the elements could be determined relative to the calculated mass of Pt. Concentration of iridium was determined with 2.3–2.8 weight% uncertainty at the most precise level, while gold and manganese were of higher uncertainty due to their low quantity. Though the presence of iridium was also determined with the PGAA method, the uncertainty of these results was rather high, thus we listed the concentration values of iridium based on the off-line counting measurements. Because of the short half-life of the ^{104}Rh radionuclide, the exact quantity of Rh could not be determined from the off-line counting measurement, however its presence was indicated. **Table 1** summarizes the PGAA results. Note that the Pt and Fe results of coin 1837 and 1838 agree within (1σ) uncertainty

margin. The iridium content in the coin issued in 1838 is 15 weight% higher than in the coin issued in 1837, while the amount of the other elements is slightly less.

With PIXE, the following elements were detected: Pt, Fe, Ir, Cu, Mn, Au, V and Ni (**Table 2.**). The results indicated 5–10% and 10–30% relative internal concentration difference among the multiple analysed surface areas for each coin in the Ir and Cu content, respectively. The concentration difference in case of the other detected elements exceeds 50% relative of the analysed values, at several parts. These results are in agreement with the previous findings, i.e. the coins are spatially heterogeneous on the scale of 1–2 mm diameter areas (Rehren et al. 2012; Weerd et al. 2004).

Both the previous study (Rehren et al. 2012) and one of the PIXE results indicated the presence of elevated gold content on the reverse side of the Pt 3-Rubel coin from 1838. Using bulk PGAA and off-line counting measurement, more than fifty times less gold content (70–100 µg/g) was detected than the highest local concentration identified with PIXE (5300 µg/g). This confirms that the gold specks/inclusions (if present) are rare and unevenly distributed within the coins. This is even more characteristic of the amount of iron on the surface, which varies by up to ten times between the different measurement points with PIXE.

Based on earlier studies, the platinum content in its native ores rarely exceeds 80 weight% (Rehren et al. 2012). In a recent study Kutyrev et al. (2021) examined platinum-group minerals in Ural-Alaskan type complexes. They found that the Matysken complex contains the most common Pt-bearing mineral, isoferroplatinum (Fe_3Pt) in mm-scale nuggets, as well as a wide range of PGE, Fe and Cu alloys, sulfarsenides and antimonides, which formed in serpentine veinlets together with awaruite (Ni_3Fe) and base metal sulfides. In their Supplementary Table S4 (Kutyrev et al. 2021) the authors summarised the typical chemical compositions of different isoferroplatinum minerals and Ni_2FePt analyzed by inductively coupled plasma mass spectrometry (ICP-MS) and atom-emission spectroscopy combined with wet chemistry.

Table 1.: Bulk elemental composition of the platinum coins obtained using PGAA and off-line counting measurements. The concentrations are listed in weight percent unit together with their relative uncertainty (Rel. unc. (%)). The elements marked with asterisk were determined from the off-line counting measurements.

1. táblázat: A platina érmék PGAA módszerrel és off-line gamma spektrometriával meghatározott térfogati átlagösszetétele. A koncentrációkat tömegszázalékban adtuk meg, mérési bizonytalanságukkal együtt (Rel. unc. (%)). A csillaggal jelölt elemeket az off-line gamma spektroszkópiai mérések segítségével sikerült meghatározni.

	Coin 1837		Coin 1838	
Element	**Weight %**	**Rel.unc. (%)**	**Weight %**	**Rel.unc. (%)**
Pt	96	0.5	96	0.5
Fe	1.3	8	1.4	8
Ir*	1.17	2.3	1.35	2.8
Cu	0.49	3.8	0.19	3.8
Mn*	0.0014	12	0.0008	17
Au*	0.010	16	0.007	15
Rh	0.14	6	0.12	6

Table 2: PIXE results in weight percent concentration

2. táblázat: PIXE módszerrel meghatározott tömegszázalékos összetétel

			Elements in weight percent concentration							
	PIXE measurement	**filter**	**Pt**	**Fe**	**Ir**	**Cu**	**Mn**	**Au**	**Ni**	**V (ppm)**
Coin 1837	*#1 reverse*	60 µm PC	96.7	1.12	1.61	0.49	0.024		0.044	
	#2 reverse	100 µm Al	97.6	0.18	1.81	0.38			0.030	26
	#3 cross section	60 µm PC	96.5	1.47	1.49	0.47			0.077	85
	#4 cross section	100 µm Al	95.6	1.13	1.65	0.41			0.084	
Coin 1838	*#1 reverse*	100 µm Al	97.0	0.94	1.32	0.38	0.035		0.064	
	#2 reverse	60 µm PC	97.0	1.30	1.29	0.36	0.024		0.039	177
	#3 reverse	100 µm Al	97.7	0.14	1.42	0.22	0.0017	0.53	0.021	

According to their data, the typical Pt content of isoferroplatinum is 62–90 weight%, Fe varies between 8 and 30 weight%, Cu varies between 0.2–3 weight%, and Ir ranges from 0.07–3.5 weight%, while the maximum detected amounts of Ni and Rh were 3 and 1.7 weight%, respectively.

In Ni_2FePt the following concentrations were detected: 11–32 weight% Pt, 24–34 weight% Fe, 39–53 weight% Ni, 0.1–4.6 weight% Cu, and 0.02–0.15 weight% Rh. Although osmium, ruthenium and palladium were also detected in some instances, these are not discussed here, because their quantities were under the detection limit of our applied analytical techniques, PGAA and PIXE. Manganese and gold were detected with PGAA and PIXE, too, but we have no comparative data for them from the geological study. Both elements show high concentration differences in the surface and in the bulk.

In **Table 3.** the PGAA and PIXE data are compared to draw conclusions on the heterogeneity of the coins. Apart from platinum, all elements are unevenly distributed within the coin volumes. Significantly, the iron concentration measured with PIXE on the cross section of the cut coin shows good agreement with its bulk PGAA data. On the surface, however, its quantity is often only one tenth of that measured in the total volume. The iridium concentration of coin 1837 measured with PIXE is 30–50% relative higher than that obtained with PGAA, suggesting that iridium is enriched on the surface, while the surface-to-bulk iridium content in coin 1838 agrees within 2% relative of the absolute value, suggesting a more uniform distribution in the whole volume.

The almost completely depleted amount of Fe and Ni in the coins compared to the composition of the native Pt-containing minerals proves the mastery of large-scale routine platinum refining and the development in the first decades of the nineteenth century. The reduction in the contents of Ir and Ru in the coins, to around 1 weight% and 0.5 weight%, respectively, is less pronounced when using the raw mineral data from Kutyrev et al. (2021) for pure isoferroplatinum nuggets as a baseline; however, bulk Russian platinum ore concentrate data provided by Rainer (1902, 15) indicate an iridium content of more than 4 weight%, but only 0.3 weight% ruthenium. As it is likely that the large-scale processing in St Petersburg would have been based on panned ore concentrates of mixed mineralogy rather than pure isoferroplatinum, it is reasonable to conclude that also the separation of platinum from its main PGE companion iridium had made good progress by the time the coins were produced, despite the significant residual amounts left in the coins analysed here.

Coin 1837	**Pt**	**Fe**	**Ir**	**Cu**	**Mn**
PGAA min	95.5	1.2	1.14	0.47	0.0012
PIXE min	95.6	0.18	1.49	0.38	0
PGAA max	96.5	1.4	1.20	0.51	0.0016
PIXE max	96.7	1.47	1.81	0.49	0.024
PIXE/PGAA min	*1.00*	*0.15*	*1.31*	*0.81*	*0.00*
PIXE/PGAA max	*1.00*	*1.05*	*1.51*	*0.96*	*15.00*
Coin 1838	**Pt**	**Fe**	**Ir**	**Cu**	**Mn**
PGAA min	95.5	1.3	1.31	0.183	0.00066
PIXE min	97.0	0.14	1.29	0.22	0.0017
PGAA max	96.5	1.5	1.39	0.197	0.00074
PIXE max	97.7	1.3	1.42	0.38	0.035
PIXE/PGAA min	*1.02*	*0.11*	*0.98*	*1.20*	*2.58*
PIXE/PGAA max	*1.01*	*0.87*	*1.02*	*1.93*	*47.30*

Table 3.: Comparison of surface and bulk compositions, in weight percent

3. táblázat: Felszíni és térfogati összetétel eredmények összehasonlítása, tömegszázalékban megadva

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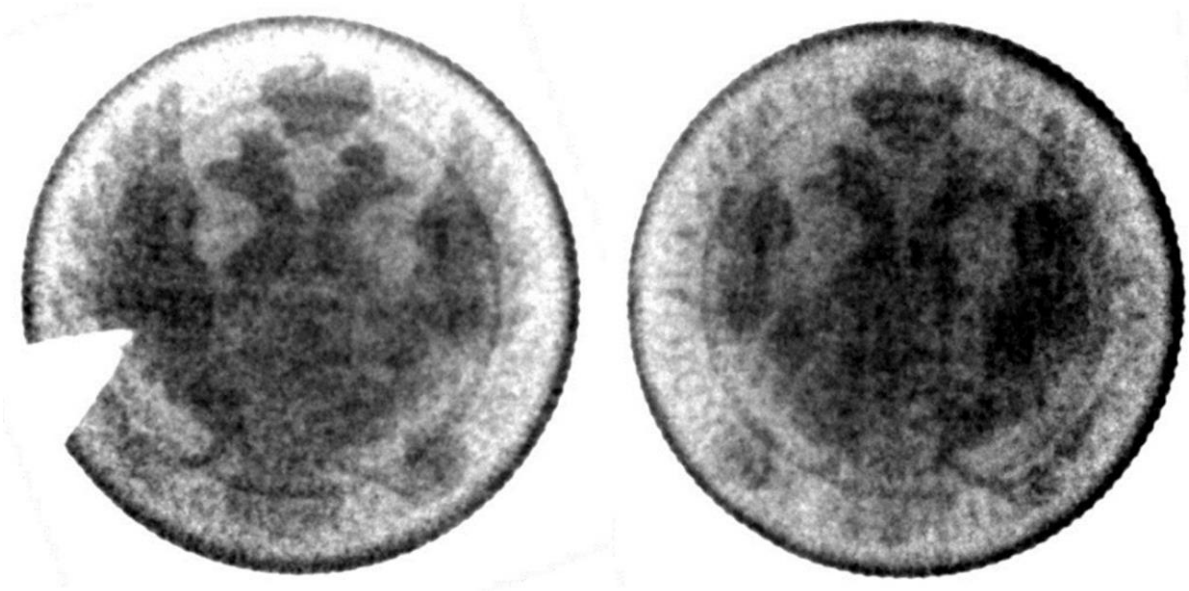

Fig. 2.: Neutron radiography of the platinum coins issued in 1837 (left) and 1838 (right)
2. ábra: 1837-es (bal) és 1838-as évből (jobb) származó platina érmékről készült neutron radiográfia

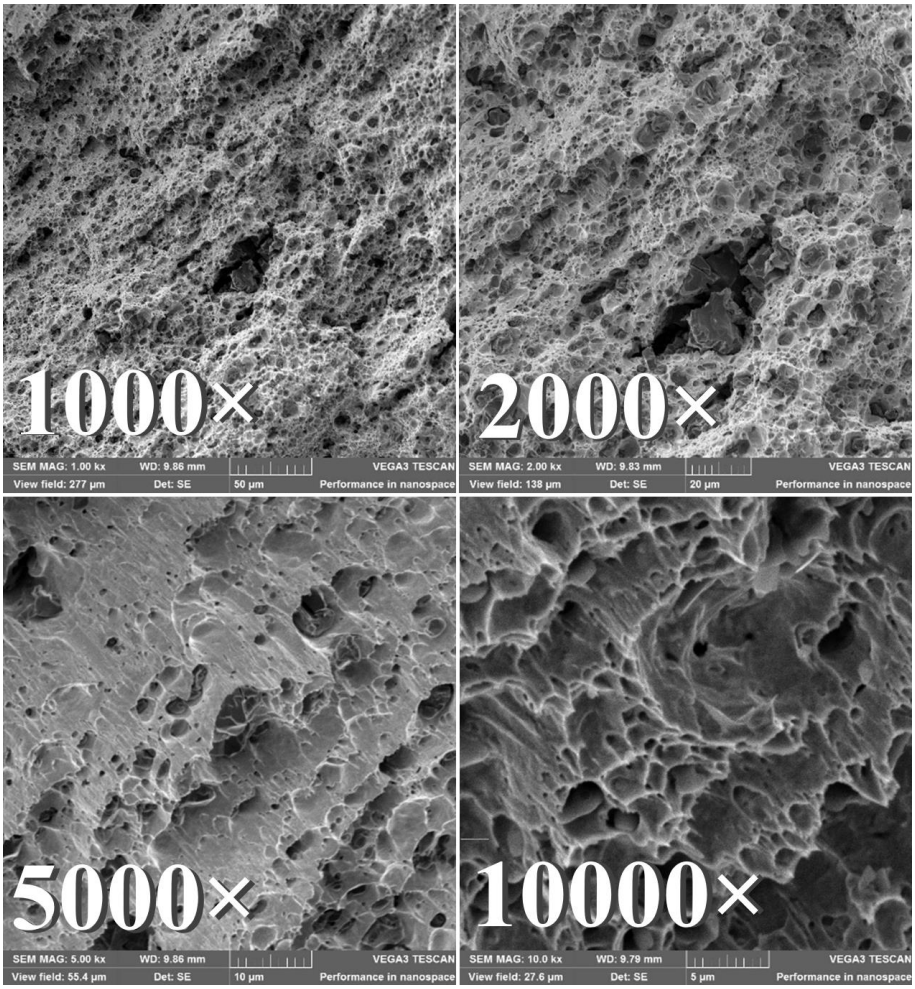

Fig. 3:
SEM pictures for a small piece fracture surface from the Pt coin issued in 1837

3. ábra:
Az 1837-es platina érméből kivágott darab törési felszínének pásztázó elektronmikroszkópos képe

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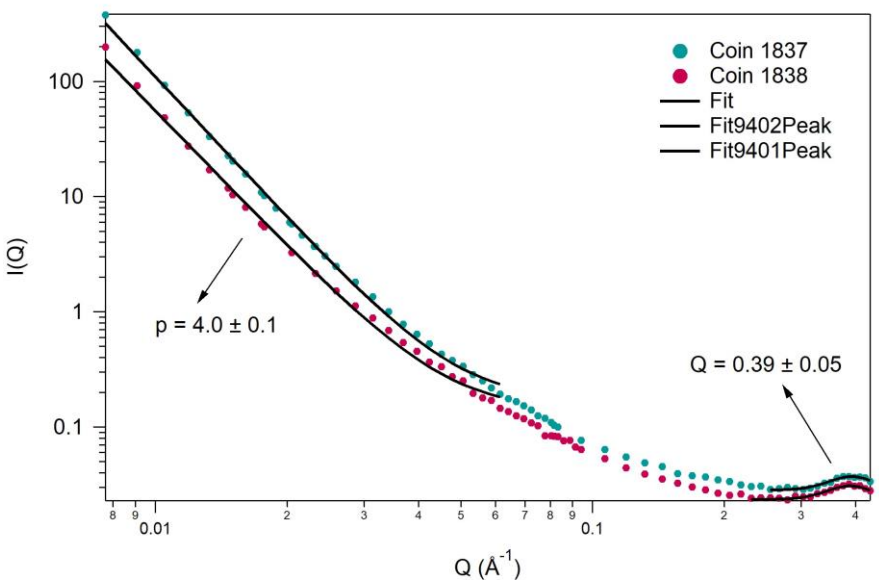

Fig. 4.: Radially averaged intensities as a function of the scattering vector, with the fitting curves and parameters

4. ábra: Radiálisan átlagolt intenzitások a szórásvektor függvényében, az illesztési görbékkel és paraméterekkel

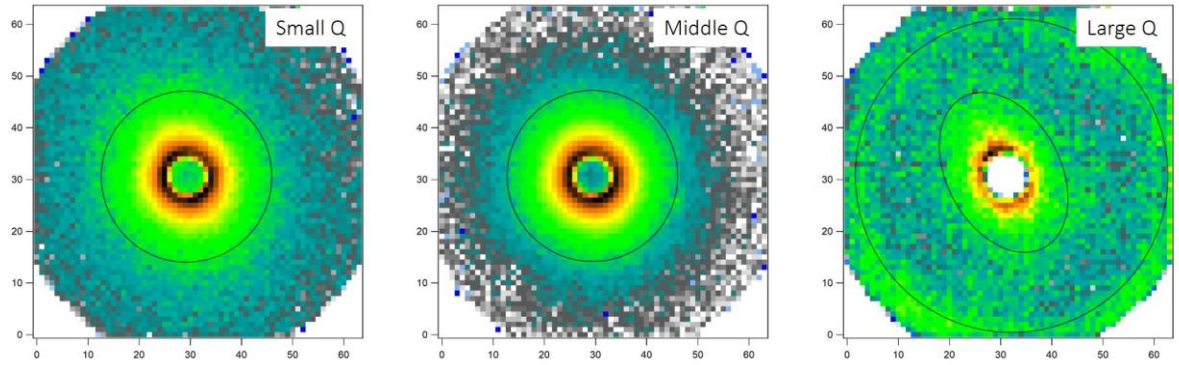

Fig. 5: 2D scattering images of the coin issued in 1837 measured at different scattering vector ranges

5. ábra: Az 1837-ben kiadott pénzérme 2D képei szórásképei különböző szórásvektor tartományokon mérve

Macro- and microscopic structure

The neutron radiography revealed no macroscopic heterogeneity. The darker areas correspond to thicker parts of the coins (see **Fig. 2.**), reflecting the engravings in the dies used for striking the coins. No other heterogeneities in NR transparency were observed.

SEM images were taken from a fractured piece of the coin issued in 1837. The used SEM instrument was a VEGA3 TESCAN scanning electron microscope. The images show generally homogeneous structure with inclusions already visible by 1000× magnification (**Fig. 3.**).

Nanostructure

The nanostructure of both studied coins showed similar SANS curve shapes, and the model fitting parameters were also similar (**Fig. 4.**). From the scattering in the high- and middle-Q range (**Fig. 5.**), power-law exponents of 4.06 ± 0.05 and 3.91 ± 0.05 were calculated, which are characteristic of smooth interfaces. In the present case, the interfaces are attributed to grain boundaries with sizes larger than 100 nm (100 nm being the upper size limit of the YS-SANS). Smooth grain boundaries indicate a homogeneous microstructure, as precipitates or other second phase separations would reduce the value of the power-law exponent.

At the Q value of 0.39 $Å^{-1}$, a small peak was observed, corresponding to a real-space size of $Q/2\pi$ = 16 Å. This peak is believed to originate from very small scattering domains (pores, cracks) whose small-angle scattering is at the detection limit of the instrument. Additionally, these domains exhibit a certain directionality, which cannot be detected in the middle and high Q ranges. This suggests that a possible directional fingerprint of the production remains in the sample at the level of the smallest entities.

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TOF-ND data did not provide clear results on the presence of iron-oxides, therefore it is not reported here in more detail.

Conclusion

In this study the suitability and potential of neutron-based methods were explored for the analysis of minor and trace impurities as well as structural features at several size scales within two Russian platinum coins. The surface and bulk compositions were determined using PIXE and PGAA techniques, respectively. By comparing the results of the surface and bulk analytical techniques, it can be concluded that the impurities, primarily iron, iridium, gold, copper, and manganese are unevenly distributed within the platinum coins. Particularly for iron and copper a systematic higher concentration was found in the body of the coins compared to their surfaces, which likely reflects the bleaching of the coins after their hot treatment to remove surface contaminants, such as iron oxide scales or inclusions that would diminish the silvery shine of the fresh coins. However, in the case of the coin issued in 1838 the surface and bulk iridium concentrations showed good agreement, implying its more homogeneous distribution compared to the coin from 1837. Small-angle neutron scattering studies confirmed the homogeneous microstructure of the coins, while a possible residual directional fingerprint of the production technique was observed in the smallest size range.

The underlying historical research question that triggered this study relates to the potential of these coins to document the development of platinum metallurgy in the first half of the 19th century. During this period, significant advances were made in the metallurgy and chemistry of the platinum group elements, and in the metal refining and processing at the mint in St. Petersburg. Some of these advances and changes in process may be reflected in the coins' composition, issued over 14 years, from 1828 to 1842. Based on the research presented here, PGAA analysis together with off-line counting of a large set of coins, including multiple coins from each year, is the most promising approach to unlock this archive of this specific chapter in the history of science and technology related to platinum metallurgy.

Contribution of authors

Maróti Boglárka Validation, Investigation, Visualization, Writing – Review & Editing. **Kasztovszky Zsolt** Investigation, Writing – Review & Editing, Project administration. **Len Adél** Validation, Investigation, Visualization, Writing – Review & Editing. **Kis Zoltán** Investigation, Visualization, Writing – Review & Editing. **Káli György** Investigation. **Füzi János**† Validation, Investigation. **Kovács Imre** Validation, Investigation, Visualization, Writing – Review & Editing. **Szentmiklósi László** Validation, Investigation. **Szőkefalvi-Nagy Zoltán** Validation, Investigation, Writing – Review & Editing. **Rosta László** Funding acquisition. **Thilo Rehren** Conceptualization, Investigation, Supervision, Resources, Writing – Original Draft, Review & Editing.

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TWO CENTURIES OF CONSERVATION TREATMENTS ON ROMAN BRONZE STATUETTES FROM THE HUNGARIAN NATIONAL MUSEUM. HOW DO THEY AFFECT SURFACE ANALYSES?

A MAGYAR NEMZETI MÚZEUM RÓMAI BRONZSZOBRAINAK KONZERVÁLÁSA AZ ELMÚLT KÉT ÉVSZÁZADBAN. HOGYAN BEFOLYÁSOLJÁK A FELÜLETI VIZSGÁLATOKAT?•

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Abstract

As a part of a larger project on Roman statuettes in Hungary we have also investigated the "restorations" and "improvements", carried out over the past centuries on most of the earlier pieces we have analysed. The statuettes were acquired from excavations, donated by private collectors or possibly bought from other countries and underwent various treatments.

An artificial patina, in the form of a black lacquer, was applied to most of the figurines. Our examination showed that it is possible to distinguish the ones employed in the 19th century – or even earlier – from the reddish-brown ones employed in the early 20th century. Interestingly, these treatments show notable similarities to the artificial patinations applied to Roman objects recovered in the same periods discovered at Pompeii and elsewhere in the area. Interventions with electrolysis can also interfere with surface analysis of older museum pieces. Another noteworthy aspect is the range of "improvements", additions and "repairs" performed on some of the statuettes. In several cases, missing limbs, body parts or attributes were reconstructed by the "restorers" who tried to "improve" the objects by coating them with some kind of artificial patina to homogenize their appearance. This is a relatively common phenomenon that can be observed in most of the collections acquired by larger museums in different ways in the 18th and the 19th centuries.

The primary aim of this paper is to highlight the impact that older "restorations" of various kind can have on surface analyses of the artifacts. This paper presents some case studies from the Hungarian National Museum in Budapest and compares them to artefacts from other Roman sites in Italy.

Kivonat

A Magyarországról előkerült római kori bronzszobrokkal foglalkozó OTKA-projekt kapcsán azokat a „restaurálásokat" és „javításokat" is vizsgáltuk, amelyeket az elmúlt évszázadokban végeztek az általunk elemzett darabok nagy részén. Ezek a tárgyak ásatásokból származnak, magánszemélyek adományai, esetenként más országokból vásárolták őket, és különböző felszíni kezeléseken estek át.

A legtöbb tárgyon mesterséges patinát alkalmaztak fekete lakkréteg formájában, és az elemzések kimutatták, hogy a 19. században – vagy még korábban – alkalmazott eljárásokat meg lehet különböztetni a 20. század elején alkalmazott vörösesbarna patinától. Érdekes eredmény, hogy ezek hasonlóságot mutatnak a Pompeiiből előkerült római kori tárgyakon ugyanebben az időszakban alkalmazott mesterséges patinákkal. Az elektrolízises beavatkozások szintén gátolják a régebbi múzeumi darabok felszíni vizsgálatait. Fontos vizsgálati téma az egyes szobrokon végzett „javítások" és kiegészítések technikája. Egyes szobroknak például hiányoztak a végtagjai, egyéb testrészei vagy attribútumai, így a hiányzó részek hozzáadásával és valamilyen mesterséges patinával való bevonással igyekeztek javítani a megjelenésükön. Ez a gyakori jelenség a legtöbb, nagyobb múzeumhoz tartozó, régebbi, a 18–19. században szerzett gyűjtemény leletanyagában megfigyelhető.

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A jelen tanulmány fő célja a különböző típusú régi "restaurálások" felszíni vizsgálatokra gyakorolt hatásának bemutatása a Magyar Nemzeti Múzeumból származó esettanulmányok és olaszországi lelőhelyekről származó párhuzamok bemutatásával.

KEYWORDS: ROMAN BRONZE STATUETTES; HUNGARIAN NATIONAL MUSEUM; XRF; SEM-EDS; ICP-OES; ARTIFICIAL PATINA

KULCSSZAVAK: RÓMAI KORI BRONZSZOBROK; MAGYAR NEMZETI MÚZEUM; XRF; SEM-EDS; ICP-OES; MESTERSÉGES PATINA

Introduction

Archaeological bronze artifacts from historical collections – such as the Roman statuettes in the collections of the Hungarian National Museum (HNM) in Budapest, which represent the focus of this paper – but also objects in many more museums around the world – underwent cleaning and restoration in the past, sometimes multiple times. Regrettably, in most cases, detailed documentation on the earlier restorations, reconstructions or additions is lacking. The absence of records poses significant challenges for today's scholars analysing the artifacts, as well as for conservation specialists, who need to update the conservation treatments. Not many studies have been carried out on how previous treatments might influence surface analyses, such as X-ray fluorescence (henceforth XRF), the method employed in this research or Scanning Electron Microscopy with Energy Dispersive Spectrometry (henceforth SEM-EDS).

Understanding past restoration procedures is crucial to avoid inaccuracies or misinterpretations, especially in cases when artifacts have been treated with acids, subjected to electrolysis and then covered with various kinds of artificial patinae produced by applying a coating or by using chemicals. In most cases, the lack of archive documentation represents a problem, and a scientific investigation is required to identify historical conservation treatments and evaluate the current condition of the objects. Studying the techniques and materials employed in past interventions can help researchers determine the appropriate approach to the study of these pieces. This paper does not aim to present a history of conservation – which would be out of place in this context and beyond the scope of this study - but rather to explore how past treatments, such as the coatings, artificial patination with acids and cleaning through acids and electrolysis may affect surface analyses. The paper is based on data collected from a small group of statuettes analysed in 2023 in the National Museum Budapest with comparisons from the National Archaeological Museum in Naples. Additional interesting aspects include reconstructions, additions and repairs observed on the statuettes.

Analytical methods

All Roman copper-based statuettes from the collections of the Hungarian National Museum studied for this project (250 pieces in total) were first autoptically examined with various magnification devices and viewed under a microscope to determine the conservation condition. The primary objective of this examination was to identify the most suitable area for performing the measurements. As a non-destructive method was required, we selected X-ray fluorescence analysis. XRF is a well-established and widely used method in archaeometry (Lutz et al. 1996; Helmig et al. 1989; Longoni et al. 1998; Mendoza Cuevas & Perez Gravie 2011; Soles & Giumlia-Mair 2018; Giumlia-Mair et al. 2015). The equipment employed for the analysis in the museum is a transportable X-ray fluorescence analyser consisting of various parts that can be mounted wherever necessary, even on excavations. It has been specifically developed for the analysis of cultural heritage objects by the company Assing in collaboration with experts of the Centro Nazionale di Ricerca (CNR) in Rome. The system comprises a modular setup suitable for both laboratory use and on-site applications. The head of the system integrates an X-ray source and a Si(Li) detector (~8 μm) with a beryllium window, along with a Class2 laser pointer (695 nm) for precise targeting of the measurement area (collimator, diam. 1 to 4 mm). The head is mounted on an adjustable tripod and emits acoustic feedback to ensure the correct focal distance (within +/- 0.1 mm). Additional built-in devices monitor position and stability during measurements. The spectrometer operates at a max. voltage of 50 kV and a max. current of 0.35 mA. Its energy resolution is <145–160 eV at Mn Kα (5.9 keV). The system is supported by a transformer, a voltage stabilizer and a laptop computer running dedicated software developed by Assing for the analysis of ancient metals (Assing 2020, 15-16). The measurements were conducted at a fixed angle and controlled distance, exclusively on clean areas, corrosion-free and mostly free of patina. Exceptions were made for a few measurements on flat areas with a thin, compact and stable noble patina, which has been demonstrated to have minimal impact on analytical results, (Lutz et al. 1996; Helmig et al.

1989; Longoni et al. 1998; Robotti et al. 2018; Giumlia-Mair 2022a, 89-90). Measurements on rough or porous patina were avoided. A minimum of three XRF measurements were carried out on each object, with additional readings performed in the case of ambiguous results, due for instance to complex geometries of items with an awkward shape or to possible vibrations during acquisitions. Elemental concentrations under 0.2 weight% have been considered as traces. The quoted detection limits (in ppm) were as follows: (in ppm) Pb 5–20; Sn 10–30; Fe 10–30; Co 10–30; Ni 10–30; As 20–50; Sb 15–40; Ag 5–15; Zn 10–30; Mn 15–40. For copper, results are expressed in weight percentage with a typical accuracy in the range of +/- 0.1–0.3%; The calculated precision is approximately +/- 1% for copper, +/- 2% for elements present in concentrations above 2 weight% and gradually and proportionally decreases to +/- 30% for elements near the detection threshold. To enhance data quality, relatively long acquisition times (200–300 seconds) were used.

Prior to each measurement session and whenever the equipment was switched off, standards with known composition were run to ensure reliable results. The standards, produced by AGM-Archeoanalisi, consist of polished specimens with compositions as similar as possible to ancient copper-based and silver-based alloys. They were analysed by ICP-OES after casting to obtain a precise composition. In this way drift, matrix and interference effects could be precisely monitored and taken into consideration when calculating the results with the dedicated software (see e.g. Soles & Giumlia-Mair 2018; Giumlia-Mair et al. 2023, 14).

For this paper we selected 5 representative pieces, as examples of different conservation treatments conducted in past centuries. The results of the XRF analyses are reported in **Table 1**.

Results

In the following text the term “patina” refers specifically to a natural patina, while “artificial patina” is employed as general term to describe either coatings made from various materials imitating a natural patina, or artificially induced corrosion layers produced with acidic copper salts, intended to imitate a natural patina. The specific methods and materials employed for individual items are discussed in detail for each piece.

As XRF is not suitable for analysing organic materials – such as the coatings on the statuettes – but can determine inorganic pigments mixed with the material of the coatings, the results must be regarded as semiquantitative. For this reason, the data from the coatings are expressed with “+++” for high concentrations, “++”, for medium levels, and “+” for low levels (see **Table 1**).

A very evident and recurring treatment on many of the objects is the application of an artificial patina, in the form of a thick, black lacquer. An example is a Lar statuette (HNM Inv. no. 54.18.17; **Fig. 1a**) cast in common leaded bronze, containing approx. 10 weight% Sn and 7.9 weight% Pb, completely coated with a black and thick lacquer. The XRF measurements revealed elevated levels of iron and calcium in the coating, while these elements were lower or absent on areas where the black lacquer was worn.

Table 1.: X-ray fluorescence results (in weight%) obtained from the measurements on the five statuettes discussed in the present paper. Results from measurements on the coating are only expressed qualitatively with +++; ++; and +.

1. táblázat: A jelen tanulmányban tárgyalt öt szobron végzett XRF-mérések eredményei, tömeg%-ban megadva. A bevonaton végzett mérések eredményei csak minőségileg jelezve (+++; ++; +).

Nr.	Object	Inv.nr.	Part	Cu	Sn	Pb	As	Sb	Fe	Ni	Ag	Zn	Mn
52	Lar	54.18.17	body	80	10.1	7.9			1.9				
52			coating	+++	++	++			++	+			
19	Fortuna	100.1895.38	body	78	7.8	10.2		tr.	0.4			3.7	
19			coating	+++	++	++			++	+			+
252	Venus	129.1895	body	89	8.4	2.5			0.2				
42	Bacchus	3.1944	head	75	6.7	18			0.2				
43			repair	86	6.4	7.6			tr.				
44			plate	75	7.5	16			1.5				
03	Minerva	105.1895.4	body	86	8.9	4.3			0.7				
04			arm fracture	73	7.4	3.4			0.3			16	
05			left arm	68	1.7	3.2			0.2			27	
06			shield	tr.					tr.				
07			shield	65	2	3.8			0.2			29	

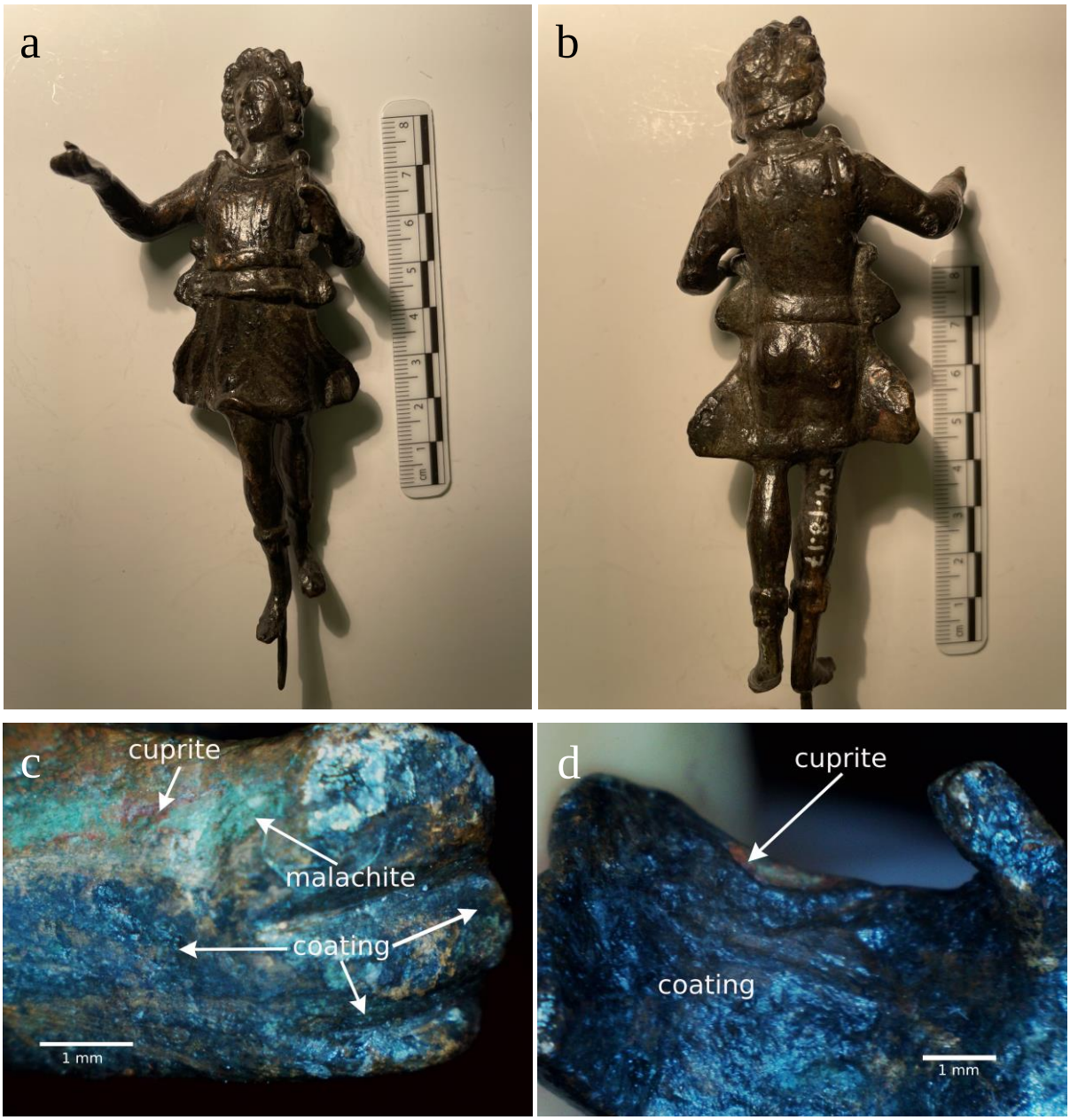

Fig. 1.: Lar statuette (HNM Inv. no. 54.18.17), made of leaded bronze (80 weight% Cu, 10 weight% Sn, 7.9 weight% Pb). **a –** Front: The statuette is covered by a thick black lacquer. **b** – Back: The figurine was produced by taking a cast from a naked statuette and adding the flowing Lar garment at the front, without bothering to add it to the back. **c** – Micrograph of left-hand detail showing that the original patina was removed down to the metal. Some remains consisting of a layer of red cuprite under green malachite are still visible at the top, while the black lacquer partly covers the rest. 50 X magnification. **d** – Detail of right hand, covered by a thickly applied black lacquer. On the side some cuprite and possibly malachite are still visible. 50 X magnification. (Photos A. Giumlia-Mair)

1. ábra: Lar szobor (MNM Ltsz. 54.18.17), ólmozott bronz (80 tömeg% Cu, 10 tömeg% Sn, 7.9 tömeg% Pb). **a –** Elölnézet: A szobor vastag fekete lakkréteggel van bevonva. **b** – Hátsó nézet. A szobrot egy meztelen szobor leformázásával készítették, az elülső felén elkészítették Lar ruházatát, a hátsó rész kialakításával nem foglalkoztak. **c** – Mikroszkópfelvétel a bal kéz részletéről. Az eredeti patina a fémig eltávolításra került. A felső részen még láthatóak egy zöld malachit alatti vörös kupritréteg maradványai, a többi részen a fekete lakkréteg borítja. 50-szeres nagyítás. **d** – A jobb kéz részlete, vastagon bevonva fekete lakkréteggel. Az oldalán egy kevés kuprit és talán malachit még látható. 50-szeres nagyítás. (Fotók: A. Giumlia-Mair)

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This Lar statuette is rather interesting, because the figurine was produced by taking a cast from a naked male statuette, possibly a Mercury or Apollon, as can be clearly seen on the shape at its back side (**Fig. 1b**). The arms were repositioned to hold a patera and a cornucopia. Finally, the typical flowing garment of a Lar was added on the front, while the back remained unchanged. This kind of black coating needs to be discussed in more detail.

18th–19th century: black coatings

Items covered by an artificial patina, consisting of a black, thickly applied lacquer (see **Fig. 1c-d**), are found in the older collections of many European museums (see e.g. Peltz 2021, 128–132). A good example from a different context is a lamp-tintinnabulum element representing an ithyphallic dwarf statuette from Pompeii (1st century AD), an old find, now in the National Museum in Naples (inv. no. MANN 27873; Pfisterer-Haas, 2023, 317-318, fig. 42; Kat. 94), like the Lar, covered by the same kind of black patina. The analyses on both objects revealed comparable results: enhanced calcium and iron levels on the coating and only traces of these elements on blank metal (Giumlia-Mair 2022a).

Anne-Claude Philippe de Tubières, in general just known as count of Caylus (1692–1765) a prominent antiquarian and collector, member of the Académie des Inscriptions et Belles Lettres and of many more cultural institutions, believed that Roman bronzes were black, because of a passage in the Natural History of Plinius the older, who mentions statues painted with bitumen (Pliny, Nat. Hist., XXXIV, 15). Caylus was a well-known and appreciated scholar, he and his publications were influential among people who at the time collected and studied ancient objects. Caylus repeatedly claimed that his Roman pieces found at Chalon-sur-Saône bore originally a black coating (Caylus 1752–1755). However, there is no documented archaeological evidence of bronzes with black coating, and no lacquer would survive a prolonged burial.

Similarly, the 19th century British collector Richard Payne-Knight had artificial black patinae applied to the objects in his collection. His blackened pieces are still now easily recognizable in the showcases of the British Museum (Craddock & Giumlia-Mair 1993, 31). This idea influenced many museums, dealers and collectors throughout Europe, and in the 19th century many pieces belonging to European collections were painted black.

An illustrative case is a lamp from Pompeii, decorated with a 21.6 cm high statuette of a dancer, excavated in the 18th or 19th century, now in the National Archaeological Museum in Naples (inv. no. MANN 72254, **Fig. 2**). Analyzed by one of the authors in 2019 (Giumlia-Mair 2022a), this piece underwent an analysis by XRF and was sampled with a drill, to be analysed by ICP-OES. All parts of the lamp, lamp's body, reflector, statuette, handle, chains and lock have been analysed, but here we only give the most important data: the lamp consists of an alloy with 75 weight% Cu, 11 weight% Sn and 17 weight% Pb (ICP-OES). The dancer boy statuette contains 74 weight% Cu; 7.4 weight% Sn and 21 weight% Pb. The ICP-OES analyses were done at the Ludwig-Maximilian University in Munich at the Department for Chemistry, Section for Central Analytics by J. Obel. The equipment was a sequential ICP-OES, so, the reason why the results do not sum up to 100% are small instrumental errors due to the fact that the elements were analysed one by one. The XRF analysis performed on the same parts of the objects gave the following results: for the lamp 74 weight% Cu; 10 weight% Sn; 15 weight% Pb. For the statuette the results were: 72 weight% Cu; 7.2 weight% Sn; 20 weight% Pb. The XRF analyses determined results similar to those of ICP-OES, but also high calcium, iron and nickel content, clearly coming from the material of the black coating applied in modern times.

Fig. 2.: Dancer statuette, decoration on lamp from Pompeii, now in the National Archaeological Museum in Naples (Inv. no. MANN 72254). The black coating applied in modern times contains iron and nickel. (Photo A. Giumlia-Mair)

2. ábra: Táncos szobra Pompeiiből, Nápolyi Régészeti Múzeum (Inv. no. MANN 72254). A modern fekete bevonat vasat és nikkelt tartalmaz. (Fotó: A. Giumlia-Mair)

The large lamp's reflector with double volutes and a central palmette was originally tinned (Giumlia-Mair 2022a, 98–99, Fig. 11.11).

The situation is slightly different with some finds from the Vesuvian area, which were blackened by the eruption. Items from Herculaneum in particular appear blackened due to thermal alteration and exposure to sulphur vapours, while some bronzes from Pompeii and other sites around the Vesuvius did not show any such discoloration. For instance, a lamp-tintinnabulum in shape of an ithyphallic dwarf (inv. no. MANN 27870; Pfisterer-Haas 2022, 318–320, Kat. 96, 420, Fig. 42.7) was already black when it was recovered from the soil. It was subsequently cleaned with part of the corrosion removed, before the usual black lacquer was applied on the surface and then placed on a base of some lost statuette (**Fig. 3**). We could analyse a small sample of the coating by SEM-EDX, but it only confirmed the presence of calcium, iron and nickel in the artificial patina. Apparently, it consists of an organic basis of some kind, possibly wax or some natural resins, mixed with a black pigment containing iron and nickel salts (Giumlia-Mair 2022a). Similar mixtures have been employed on many finds in large museums, including the National Museum in Budapest, in the 18th–19th centuries and possibly even later.

A report in the archives of the Römisch-Germanisches Zentralmuseum at Mainz without a date, but certainly written before 1887, describes this procedure: «The bronze is (mechanically) cleaned like iron, washed, heated and coated with pure wax. When it penetrates (the metal) and dries, the object is brushed with a brush made of goat hair until a matte sheen, similar to that of a patina, is created. If later some mushroom-shaped verdigris growth appears, it has to be cut away with a sharp instrument and the exposed spot can be dabbed with a «gum solution» (see Peltz 2021, 82).

It is plausible that in Budapest and Naples similar procedures were adopted.

In the 18th–19th century additional materials, such as paraffin, linseed oil, nitrocellulose (cellulose nitrate) and celluloid varnish, were applied on bronzes, both as protection and to homogenize the colour of the surface. Quite often, before the application of these materials, the objects were also exposed to high temperatures or treated with acids to eliminate the corrosion layers, quite often with detrimental results.

19th–20th century treatments

In the 19th–20th centuries and in some cases until today, collodion, a solution of nitrocellulose in ether and alcohol, was employed to consolidate corroded bronzes. Nitrocellulose (also known as cellulose nitrate) is a nitrated polysaccharide derived from cellulose, widely utilized in materials science for its film-forming properties. Although the terms "cellulose nitrate" and "nitrocellulose" are used interchangeably in the literature, this paper will refer to the material as "nitrocellulose" for consistency, in line with the prevailing scientific terminology. This material is initially colourless, can be coloured with pigments but it discolours over time. It cannot be identified with XRF, but the pigments added introduce elemental signals that will be determined, and this interferes with the analysis results of the underlaying metal. This material was widely used, and it is important to mention it.

A further material that was employed in this period is zaponlack, a commercial solvent-based varnish made with nitrocellulose, the only one available in the early 20th century in Hungary.

Fig. 3.: Statuette of an ithyphallic dwarf, originally from a hanging tintinnabulum-lamp from Pompeii, as indicated by eyelets on the phallus from which bells used to hang. The figurine was mounted on the base of a lost statuette after it was found. Its surface was blackened by sulfur vapors during the eruption of the Vesuvius and then covered by a black coating containing iron and nickel salts. (Photo A. Giumlia-Mair)

3. ábra: Ithyphallikus törpe szobra, eredetileg egy függesztett tintinnabulum lámpáról Pompeiiből, amit a falloszon lévő akasztók is jeleznek. A szobrot a megtalálás után egy már elveszett szobor bázisára erősítették. A felszínét a Vezúv kitörésekor kéngáz feketítette be, majd vas és nikkelsókat tartalmazó fekete bevonattal vonták be. (Fotó: A. Giumlia-Mair)

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It was also employed for the consolidation of corroded bronzes (Maier & Peltz 2013). This material has a high surface resistance and can be removed with amylacetate, ethanol, acetone or ethylacetate. Nowadays it is still, in some cases, used as a protection for metals and gilding on metals. Like collodion, it cannot be identified with XRF, but it can be also coloured with pigments to achieve the desired nuance for the artificial patina, again potentially interfering with the analyses. As already noted, the taste and the ideas on the patination colour of copper-based items evolved and changed with time.

Reduction by electrolysis

In the 20th century, with the beginning of "scientific" attempts at restoration and conservation, practices changed. In 1898 Rathgen, the chemist at the Museum in Berlin, published his «Handbuch» and described the "new process of conservation by reduction": electrolysis (**Fig. 4.**), one of the most common methods employed in restoration in this period (Rathgen 1898; 1905, 125–144). From then on in all museums in Europe and around the world, copper-based items were electrolytically cleaned, regrettably quite often with disastrous outcomes and only sometimes with "less-than-optimal" results.

The Isis Fortuna statuette from Brigetio (**Fig. 5a**), belonging to the collection of the Hungarian National Museum (inv. no. 105.1895.38.), may also have undergone electrolytic cleaning and re-patination. Alternatively, an older, even worse and harsher method might have been employed: a "cleaning" treatment with strong acids, such as sulphuric (H_2SO_4), hydrochloric (HCl) or nitric acid (HNO_3). This method is aggressive, difficult to control and can dissolve both, corrosion products and underlying metal, which is destructive and irreversible. Wherever acids were used in the past, they can significantly alter surface chemistry, removing or redistributing corrosion stratigraphy, and biasing surface analysis results, like those from XRF. As no documentation exists, electrolysis was widespread as cleaning method and was indeed in use in the National Museum both hypotheses are plausible.

Microscopy on the Isis Fortuna shows cuprite covered by a dark artificial patina, making an electrolytic treatment more probable, because a treatment with acids would be less well controlled, and this piece still shows a cuprite layer. The detail of the face shows that the corrosion was removed by the treatment and left some voids and pitting. Some remains of the green patina are still recognisable on top of the red cuprite layer on the face and in the hair (**Fig. 5b**). In some places, for example on the folded garment at the belt, the wear exposed the core metal (yellow spots) and cuprite (red spots) under the black lacquer (**Fig. 5c**), applied on the surface after the treatment: The colour is not as black as on the previous examples but rather reddish-brown, indicating thus the use of different coatings, possibly just the same kind of base prepared with different pigments. The XRF analysis on the coating indicated again an enhanced presence of iron and nickel.

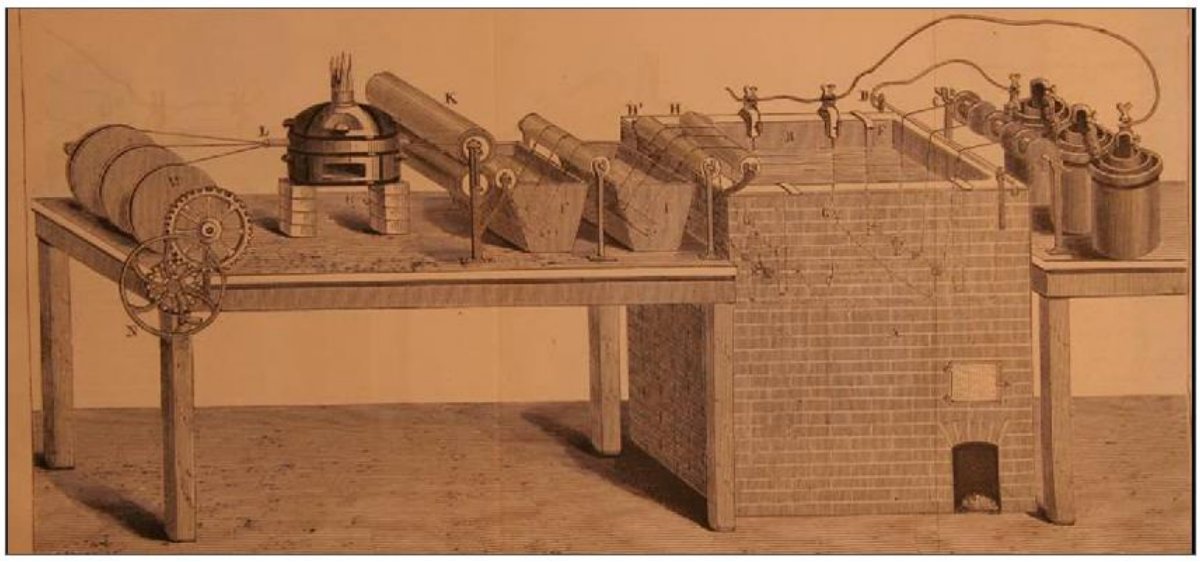

Fig. 4.: 19th century representation of the system for electrolysis and galvanoplastic. In 1924 Rathgen published his «Handbuch» describing the new process of «conservation by reduction». (Photo: A. Giumlia-Mair with permission of Museo Civico di Storia Naturale di Trieste, Italy)

4. ábra: Az elektrolízis és galvanoplasztika rendszerének 19. századi ábrázolása. 1924-ben Rathgen "Handbuch"-jában publikálta a "redukciós konzerválás" új módszerét. (A fotót A. Giumlia-Mair készítette a Museo Civico di Storia Naturale di Trieste engedélyével.)

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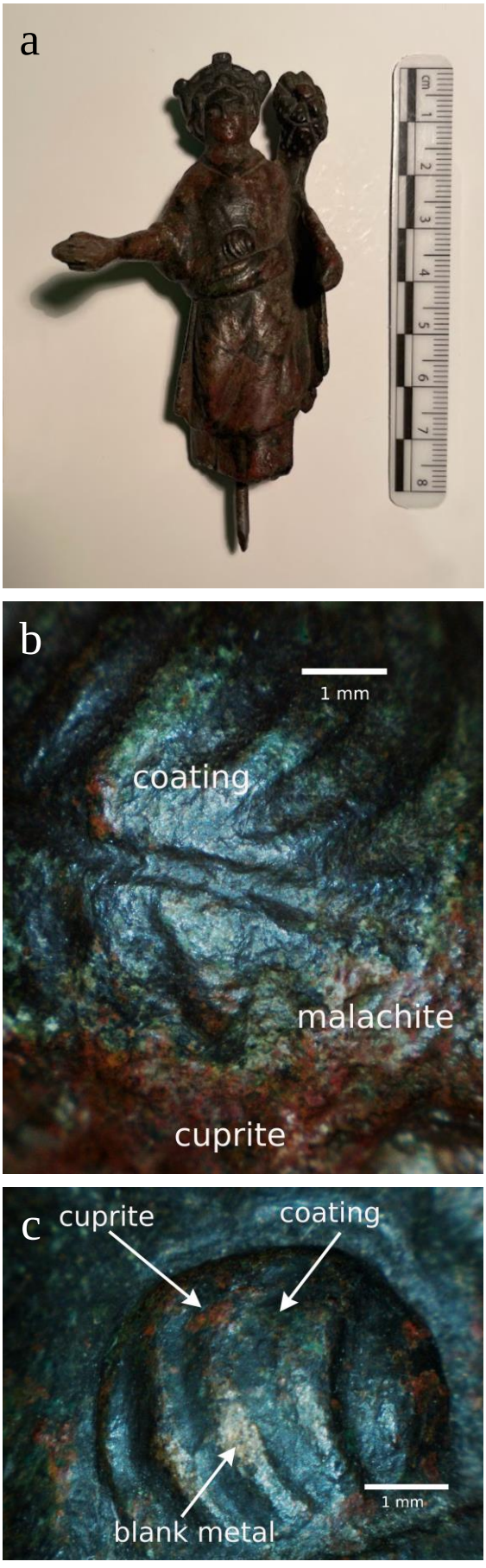

Fig. 5.: Isis Fortuna (HNM Inv. no. 100.1895.38). **a** – Presumably electrolytically cleaned or, less likely, treated with acids and then repatinated. **b** – Some remains of the green patina are still recognizable on the red cuprite layer in the hair at the top of the head and under the black lacquer. **c** – Detail of the folded garment at the belt. Microscopy shows some cuprite covered by a dark artificial layer. The light color is the blank metal. 50 X magnification. (Photo A. Giumlia-Mair)

5. ábra: Isis Fortuna (MNM Ltsz. 100.1895.38). **a** – Valószínűleg elektrolízissel tisztítva vagy, kisebb valószínűséggel savakkal kezelve és újra-patinázva.
b – A zöld patina maradványai még felfedezhetőek a vörös kupritrétegen a hajban a fej tetején és a fekete lakkréteg alatt.
c – A derékra csavart ruha részlete. A mikroszkópos vizsgálat fekete, mesterséges réteggel bevont kupritot mutatott ki. A világos szín a nyersfém. 50-szeres nagyítás. (Fotó: A. Giumlia-Mair)

Having analyzed around 250 pieces, we have been able to notice that pieces with older inventory numbers in the Hungarian National Museum seem to be predominantly covered with black lacquer, while those with later inventory numbers mostly show a reddish-brown coating. The analyses showed 7.8 weight% Sn; 10.2 weight% Pb; 3.7 weight% Zn. Both Sn and Pb concentrations were possibly higher in origin but disappeared because of selective corrosion of elements higher than copper in the activity series of metals, and the treatment for the removal of the corrosion layers.

It is important to note that the traces of Zn come most probably from the electrolytic cleaning process. The 3.7 weight% of zinc determined by the surface analysis, must be a residue left by the treatment rather than being an alloying element. As a piece of zinc was commonly employed as electrode in the electrolysis, most objects that have been electrolytically treated show zinc enrichment, sometimes up to 7–8 weight%, but only on the surface (Xu et al. 2017; Jansen et al. 2023, 21-23; for more insights into unwanted zinc deposition mechanisms see also Vijiaratnam et al. 2017; Popov et al. 1978). It is obviously important to be aware of this issue.

As demonstrated by many thousands of analyses of these items all over Europe, Roman statuettes generally show only very low zinc contents at trace level that usually can only be detected with precise destructive methods such as Atomic Absorption Spectrometry or Inductively Coupled Plasma Optical Emission Spectrometry, but not with XRF. As already shown early in the last century, by the analyses of Davies (1935, 60) and Caley (1964, 13) copper-zinc alloys circulated as precious and rare

materials in the 5th–4th centuries BC in Greece and presumably came from the area of Balya Maden in Anatolia (Caley 1964, 18-25; Craddock 1998, 3; Craddock & Eckstein 2003). This alloy, called oreichalkos (ὀρείχαλκος, copper of the mountains) in Greek, was highly valued (Homer, Hymn to Aphrodite, 6, 9; Hesiod, Shield of Heracles, 122). According to Strabo (Geographica,13, 56), it was produced in Andeira (Balya Maden) by mixing with copper the rare metal "mock-silver" i.e. zinc, found as silvery droplets coming from the upper, cooler part of the local furnaces in which silver was reduced from mixed ores (galena, sphalerite and pyrite). The zinc present in the sphalerite evaporated during smelting and condensed in the upper part of the furnace and, added to copper, was used only for jewellery and small, precious, decorative objects. Later, the copper-zinc alloy brass was produced by cementation process in a crucible and has a characteristic composition that permits to distinguish it from modern brasses (Bayley 1998, 9).

Brass, leaded brass and quaternary alloys were introduced in the Roman world in the 1st century BC (Craddock & Eckstein 2003; Istenič & Šmit 2007), apparently by metalworkers belonging to the Roman army, notably by Caesar's legions, who had learnt how to use zinc ores and cementation in Anatolia and Gallia (Giumlia-Mair 2025). Analyses indicate that freshly produced brass was only employed for coins and military equipment, suggesting that it was a state monopoly. Ordinary people had instead coins melted and mixed with copper or bronze to obtain shiny and almost golden-coloured personal ornaments (Giumlia-Mair 2025, 359-363). However, with few exceptions from France or the Rhine area (and fakes and imitation figurines produced – especially, but not exclusively - in the 19th century), statuettes were mostly made of leaded bronze and did not contain zinc in significant amounts. The possibility that the presence of zinc in some figurine is due to the use of scrap metal containing fragments of small decorative objects, like fibulae, cannot be ruled out, however, analyses of Roman objects clearly illustrate that, especially in the 1st–2nd centuries AD, and also later, the metalworkers usually kept carefully separated from bronze or leaded bronze the damaged items containing zinc (brass or leaded brass), to reuse them for similar types of objects (see e.g. Craddock 1985; Picon et al. 1966; 1967; 1968; Antonacci Sanpaolo et al. 1992; 1993; Giumlia-Mair 1993; 1996; Riederer 2002a, 286; 2002b; Thomas 2002, 302).

A second example of a potentially electrolytically treated piece is a Venus statuette (HNM Inv. no. 129.1895, **Fig. 6a-b**). This item was evidently corroded, and the electrolysis or, alternatively, a chemical treatment by acids removed the patina quite completely. The removal of corrosion layers produced holes in the areas, more affected by corrosion, in particular the details of the face. Even the edges of the repair at the back, where the patch is lost are blurred and rounded (**Fig. 6b**). The shape of the hand is now angular, and the fingertips are partly missing. **Fig. 6c** shows the detail of the left eye. The eye socket is much larger than it was originally, all details of eyelids and eyeball are lost, with part of the nose missing. The same happened with lips and chin (**Fig. 6d**). Part of the green crusts in the eye sockets and the crater of the mouth might be residues of natural patina. The green artificial patina directly applied on blank metal instead was produced with some kind of acidic solution of copper salts, as its typical runny appearance and the lack of underlaying cuprite suggest (see **Fig. 6e**). Of these solutions there are many recipes that cannot be detailed here because this discussion would transcend the scope of the paper (Buchner 1914; Hughes & Rowe 2009). The examination under the microscope showed that the treatment dissolved the tin-richer dendrites on the surface, leaving the more corrosion resistant phase alpha of the metallographic structure. This can be clearly seen from the dendrite ghosts visible e.g. on the buttock of the statuette (**Fig. 6e**). The rounded, smaller black dots are the places where the now missing lead globules were positioned in the metal, while the larger ones come from corrosion pitting. As it is well known, lead is not soluble in copper and builds small or larger globules (depending on the amount of lead present in the alloy) that are normally diffused in the metallographic structure, but can concentrate in thicker parts of the castings as happened in this statuette, or in peripherical parts. The segregation phenomenon illustrated by this statuette might possibly be due to inverse segregation (i.e. lead pushed to the surface, forced by shrinkage) or, if the mould was left laying during the cooling phase, it could be the result of gravity segregation (i.e. the lead would move to the bottom instead of being dispersed in the alloy). This also means that the elements tin and lead might be higher in the core metal of the statuette. The XRF measurement identified copper containing 8.4 weight% Sn and 2.5 weight% Pb. In particular, the lead seems rather low, thinking of the many typical voids seen by microscopy on the surface of the statuette.

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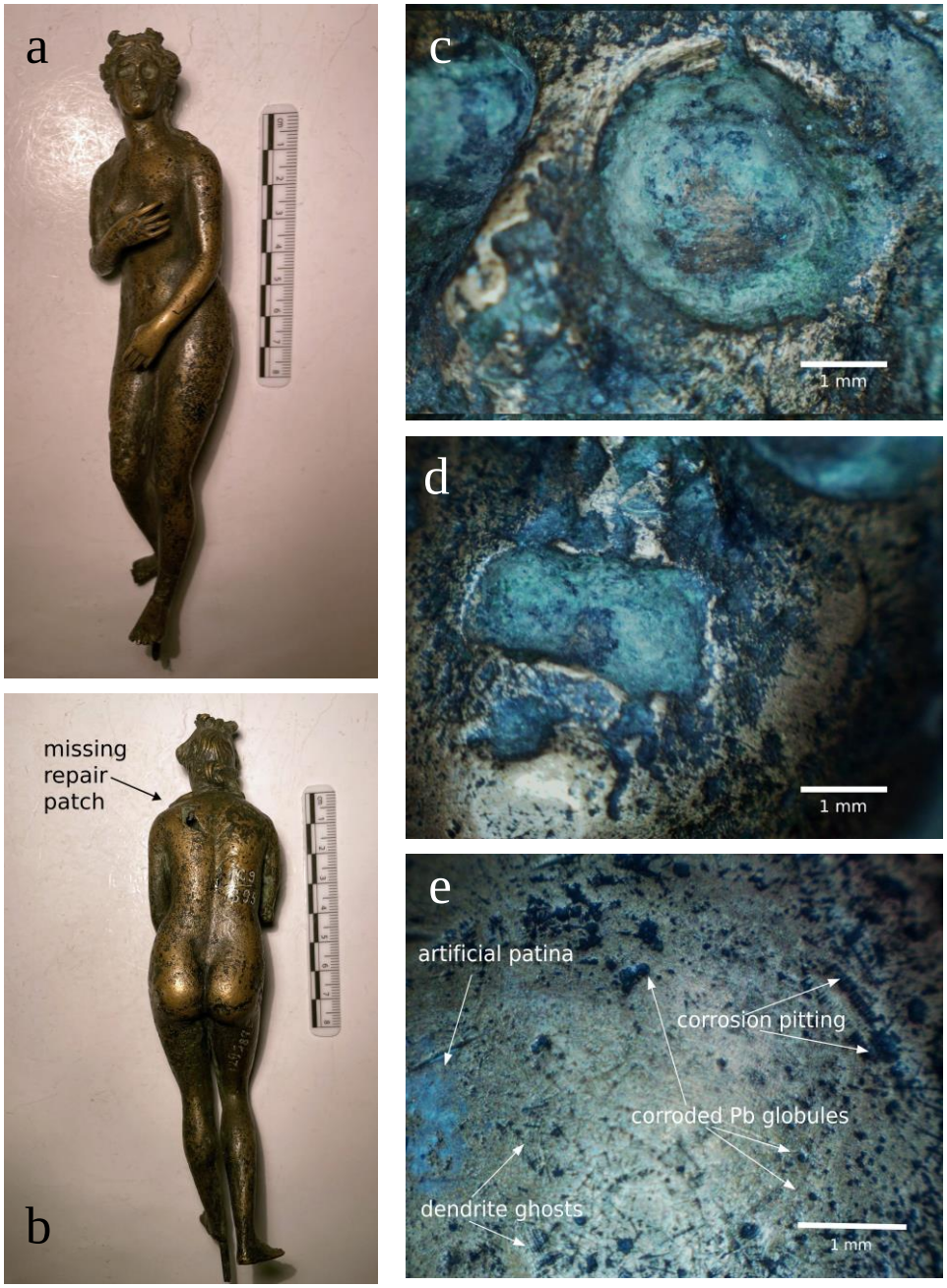

Fig. 6.: Venus (HNM Inv. no. 129.1895).
a – Front. The statuette was electrolytically stripped or treated with acids. **b** – Back side. On the right shoulder a repair patch was lost, and its contours are blurred by the treatment. **c** – Detail of eyes without any remains of lids and iris and eroded nose. The removal of corrosion by electrolysis or acids produced large holes and destroyed the facial features. X 50 magnification. **d** – Detail of eroded mouth and chin. The thicker green crusts in eye sockets and mouth are residues of the original patina, while the thinner green layers on blank metal without any underlaying traces of cuprite (see fig. 6e) come from an attempt to artificial patination. X 50 magnification. **e** – The electrolytic treatment evidenced the shape of dendrites on the buttock. The smaller round holes were left by now missing lead globules, the larger craters come from corrosion pitting. On the left some green artificial patina was likely produced with an acidic solution of copper salts. X 50 magnification. (Photos A. Giumlia-Mair)

6. ábra: Venus (MNM Ltsz. 129.1895).
a – Elölnézet. A szobor elektrolízissel tisztított vagy savakkal kezelt. **b** – A jobb vállról a javítás hiányzik, a javítás körvonalai elmosódottak. **c** – A szemek részletei a szemhéj és írisz nélkül és az erodált orr. A korrózió elektrolízises eltávolítása miatt a felületen nagy lyukak keletkeztek, az arc részletei sérültek. 50-szeres nagyítás. **d** – A sérült száj és áll részletei. A vastagabb zöld lerakódások a szem-gödörben és a szájon az eredeti patina maradványai, a vékonyabb, kupritnyomok nélküli zöld rétegek a nyers fémen (lásd 6e. ábra) a mesterséges patinázás eredményei. 50-szeres nagyítás. **e** – Dendritek, a hátsó részen végzett elektrolízis nyomai. A kisebb lyukakat a már hiányzó ólomgömbök okozták, a nagyobb mélyedések korróziós lyukak. 50-szeres nagyítás. (Fotók: A. Giumlia-Mair)

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The Bacchus bust from the legionary fortress of Brigetio (HNM Inv. no. 3.1944, **Fig. 7a**) highlights a different difficulty that is often encountered when studying or analysing pieces from older collections. Repairs, additions and amendments represent a problem if the reconstructed parts cannot be distinguished from the original alloy, either because they were coated or repatinated with acidic solutions of copper salt solutions, or simply – in the case of ancient repairs – because the surface is covered by a natural patina that disguises the patch. In this case, a raking light had to be employed to evidence on the photos of this Bacchus bust the repair covered by both a natural patina and a layer of conservation resin (**Fig. 7b**). The analysis showed that the composition of the bust and that of the repair are different, with the bust being of copper with 6.7 weight% tin and 18 weight% lead, while the patch only contained 6.2 weight% tin and 7.6 weight% lead. Nevertheless, as no difference in the patina can be detected, except for a slightly more polished aspect, this repair seems to be ancient. Had only the area on the breast been analysed, it would have given wrong results.

In the case of a Minerva statuette in the Hungarian National Museum (inv.no.105.1895.4, **Fig. 8a**) the situation is again different. The right arm holding the spear is missing and the statuette is rather corroded and covered with an artificial dark patina, i.e. a coating containing iron and nickel, possibly applied twice: the first time as black layer and then a brown layer on top (**Fig. 8b**). The body is leaded bronze with 8.9 weight% Sn and 4.3 weight% Pb, but the disproportionately thin left arm contains high zinc and is definitely a modern reconstruction made of gunmetal. The shield held by the left hand was completely re-done with resin and made of a modern gunmetal or quaternary alloy: copper containing tin, lead and zinc.

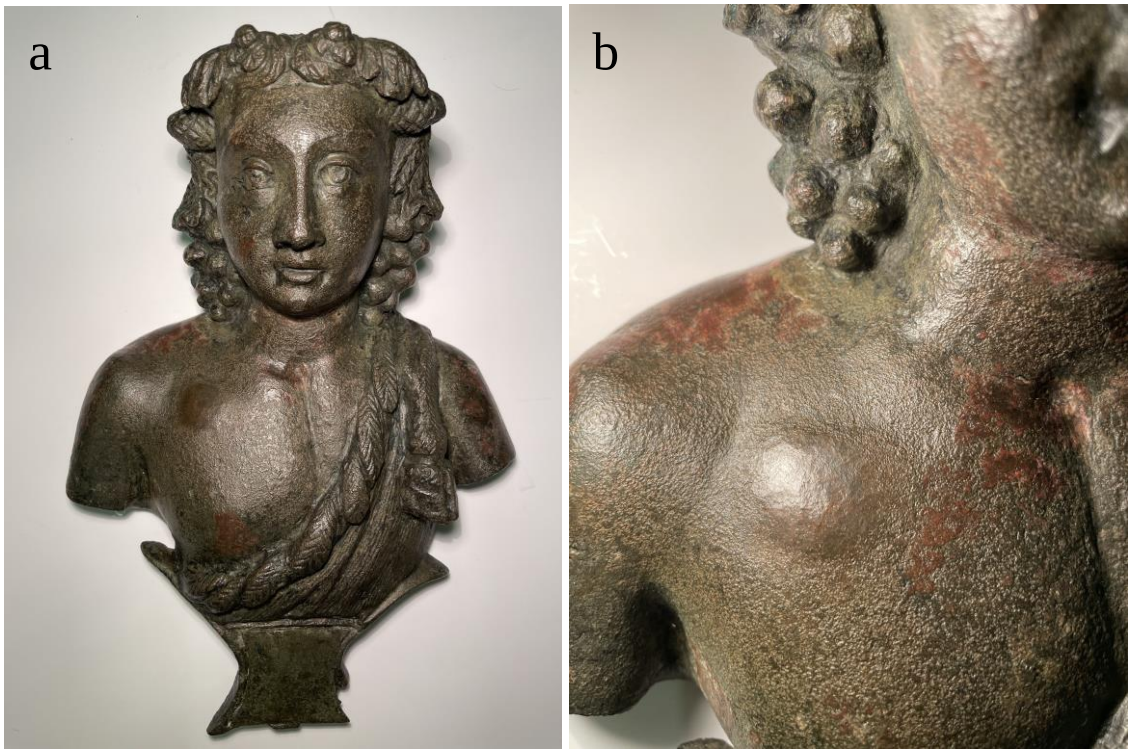

Fig. 7.: Bacchus bust (HNM n. 3.1944.) **a** – The alloy is Cu with 6,7 weight% Sn and 18 weight% Pb. The bust still retains most of the natural patina, covered by conservation resin. **b** – The ancient repair (Cu with 6.2 weight% Sn and 7.6 weight% Pb) on the right side of the bust, under the natural patina, was evidenced with raking light for the photo. (Photo A. Giumlia-Mair)

7. ábra: Bacchus büsztje (MNM Ltsz. 3.1944). **a** – A bronzötvözet 6,7 tömeg% ónt és 18 tömeg% ólmot tartalmaz. A patina nagy része természetes, a restaurálás során gyantával borítva. **b** – A büszt jobb oldalán súrlófényben jól látszik az ókori javítás (réz 6,2 tömeg% ónnal és 7,6 tömeg% ólommal) a természetes patina alatt. (Fotó: A. Giumlia-Mair)

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Fig. 8.: Minerva statuette HNM n. 105.1895.4. **a** – Front (right) and back (left). Right arm and spear missing, left arm and shield reconstructed with a quaternary alloy and conservation resins, while only the left hand and perhaps the top of the shield belong to the original casting.
b – Detail of the left hand, which lost part of its patina and coating, showing various layers: red cuprite, green malachite, a layer of black coating and the last brown coating applied on the entire surface of the statuette. X 50 magnification. (Photo A. Giumlia-Mair)

8. ábra: Minerva szobor (MNM Ltsz. 105.1895.4) **a** – Elölnézet (jobbra) és hátulnézet (balra). A jobb kar és a lándzsa hiányzik, a bal kar és a pajzs négykomponensű ötvözetből rekonstruálva, gyantanyomokkal, csak a bal kéz és esetleg a pajzs teteje tartozik az eredeti szoborhoz.
b – A bal kéz részlete. A patina és a bevonat nagy része már eltűnt, különböző rétegek láthatók: vörös kuprit, zöld malachite, a fekete bevonat egy része és a végső barna bevonat, amit a szobor teljes felületén alkalmaztak. (Fotó: A. Giumlia-Mair)

Formation of corrosion compounds from restoration materials

A last point to be touched upon in this paper is that often materials employed in the conservation of artifacts can alter and produce corrosion compounds. This phenomenon has not been observed on the pieces examined for this research, but it has been widely studied and it is important to mention this problem as well. For instance, compounds containing copper acetate have been identified on archaeological copper-based items. The sources of this alteration are not only the ammonium acetate, widely employed for artificial patination on stripped objects, but also acetic acid used for cleaning, cellulose acetate resins used in the past as adhesives or in lacquer, and vinyl acetate and polyvinyl acetate as well. Even Paraloid, commonly used since the 50ies of the last century, can promote corrosion that penetrates deeply into the metal, if it is applied over acidic remains from previous treatments or on small copper chloride formations caused by other factors (Paterakis 1999). Numerous materials used in the past can contribute to the formation of corrosion products and this should also be kept in mind when studying older collections.

Conclusions

This paper addressed only a few of the various problems which analysts face when working on old collections, and not only on statuettes. As discussed above, analysing statuettes coated with 19th century lacquer without recognizing the presence of iron and nickel-based pigments – or (in some other cases) iron-manganese-based pigments – employed for the colouring of the artificial patina would lead to enhanced results of these elements. This, in turn, might create the false impression that the copper employed for these items contains distinct trace elements, possibly suggesting a different provenance for this group of objects.

The electrolytic treatments, extensively employed in the 19th and in some places even into the 1970s, would on their side result in enhanced zinc results on the surface of the treated objects. Similarly, treatments involving strong acids would remove from the surface alloying elements more reactive than copper in the electrochemical series, such as tin and lead, leaving voids, compromising surface integrity and causing too low analytical results for these elements.

Between the 17th and the 19th centuries many more materials were used to cover the surface and imitate a natural patina. In the 20th century, electrolytic treatments used tin and aluminium, not only zinc, also affecting the data, in this case by enhancing the tin results.

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It is also important to mention that patina imitations and coatings often consist of several layers of different materials applied at different times. Good examples are famous statues like the Idolino (Iozzo 1999) or the Chimaera from Arezzo (Nicosia & Diana 1992; Iozzo 2009; Siano et al. 2012, 202-204), as well as several the statuettes from Pompeii (Giumlia-Mair 2022a; 2022b). This means that the different coatings applied on the surface might contain a variety of elements that might have a significant impact on surface analyses. Unfortunately, in recent years, the diffusion of portable XRF models, the famigerate "pistols", used by people with no experience and without any knowledge of ancient metallurgy, produced a plethora of patently wrong data in the archaeological literature.

It is important for both, restorers and analysts, to be aware of the many pitfalls possible with pieces from ancient collections. Understanding the conservation processes applied in the past centuries represents a pivotal point for the studies of ancient metallurgy, but also for evaluation of the preservation conditions of bronze statuettes and copper-based objects in general, and it greatly helps by providing scientific information when deciding the most suitable conservation procedures of collection pieces. Furthermore, it assists archaeologists in understanding the real significance of the analytical data and in the interpretation of the items. Often determining whether an attribute was replaced or repaired in modern times or in antiquity can be quite a challenge.

John Ruskin, the prominent 19th-century English art critic, writer, and thinker profoundly influenced ideas on art, architecture, and preservation and believed that the original material of an artwork, even if decayed, should be respected, because alteration is part of its history. So, in the case of bronze sculptures, patina and corrosion were not blemishes to be "cleaned up" but markers of age and authenticity that should be preserved, not removed. In his influential book The Seven Lamps of Architecture he wrote a section titled the "Lamp of Memory," and argued that signs of time should be retained, and integrity, authenticity, and memory in objects and art should be valued. Living in the Romantic period, Ruskin considered time itself as the artist (Ruskin 1849).

Considering the outcome of some "restorations" of the last few centuries, one feels that Ruskin was indeed right and can only agree with him. In his book he was mainly referring to architecture, but his idea stretched to all historic objects. He stated, "*Do not restore. Restore is to destroy. That spirit which should have preserved the work in the first place, should continue to watch over its decay.*" and also: "*It is impossible, as impossible as to raise the dead, to restore anything that has ever been great or beautiful…*" (Ruskin 1880, Chapter IV, 242).

Luckily, over the last half century at least, conservation has increasingly shifted to favouring minimal intervention, and preservation more than removal.

Contribution of authors

Alessandra Giumlia-Mair Investigation, Methodology, Formal analysis, Writing – Original Draft, Review & Editing. **Bartus Dávid** Investigation, Funding acquisition, Project administration, Writing – Original Draft, Review & Editing.

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Közlemények

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