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# ORIGINAL RESEARCH PAPER





# Tracing the road of elephant ivory at the end of Late Antiquity – Archaeometric analysis of ivory artefacts from the 6th–7th-century Carpathian Basin

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#### **ABSTRACT**

Elephant ivory, a prestigious and valuable raw material in the post-Roman West and Byzantium between the 5th and 7th centuries AD, may originate from various sources. While both written and art historical evidence suggests that in the case of early medieval artefacts, African provenance is more likely than Asian, no data at hand is conclusive. The present paper investigates, with the help of FTIR and Raman spectroscopy, carbon and nitrogen concentration and nitrogen isotope (8<sup>15</sup>N) analyses, the material resources of elephant ivory artefacts discovered in 6th- and 7th-century AD archaeological context in the Carpathian Basin to contribute to our understanding of late antique long-distance trade networks and economic relations.

#### **KEYWORDS**

elephant ivory, Late Antiquity, early Middle Ages, long-distance trade, vibrational spectroscopy

### INTRODUCTION

Ivory as raw material was considered exceptionally suitable for manufacturing prestige objects in the ancient and medieval world. This also holds true for the late antique Mediterranean, where archaeological research equated ivory mainly with elephant ivory for a long time. However, in other geographical regions of late antique and medieval Europe, "ivory", in fact, may equally or more likely mean tusks or teeth of other terrestrial and marine mammals, such as mammoths, hippopotami, warthogs, walruses, and different species of toothed whales. Therefore, and since published identifications (a great number of which are solely based on the untrained eyes of archaeologists) often contribute to the emergence of confusion in this regard, scientific analyses are inevitable for gaining a proper understanding of the use of different sorts of ivories as well as for mapping the related trade routes and networks. This is even more so in the cases of geographical regions situated at an almost

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<sup>1</sup>Lane (2015).

equal distance from the natural habitats of African elephants and hippopotami on the one hand and of northern walruses and whales on the other, as well as periods in which access to resources from one or another direction became increasingly limited or, conversely, entirely unrestricted. The present study focuses on such a period and region, the Carpathian Basin in the 6th and 7th centuries AD.

Although in Imperial Roman times and Late Antiquity, ivory artefacts reached fairly broad strata of Mediterranean society, the availability of ivory carvings seems to have dropped drastically by the early 7th century AD and, in Byzantium, became restricted to the highest echelons of the elite between the later 7th and the 9th centuries AD.2 A closely similar chronology is outlined in the post-Roman West, where artefacts manufactured from elephant ivory seem to have become widespread only from the late 6th century AD on, while a plummeting in their number is apparent from the late 7th century AD.<sup>3</sup> However, despite this chronological parallelism, there are surprisingly few common elements between the uses of ivory in the late Roman/early Byzantine world and large regions of the post-Roman West. While during Late Antiquity, Mediterranean craftsmen continued the Roman tradition of manufacturing ivory into elaborately carved prestige and everyday objects, in the overwhelming majority of the known cases, north of the Alps, ivory was employed after minimal treatment in the form of annular rings, often enclosing cast copper-alloy circular decorative rings used as components of pouches.4 As we shall see below, the Carpathian Basin, situated at the fringes of both regions, has yielded findings pertaining to the products of both traditions.

Research into late antique and early medieval ivory carvings has been largely dominated for a long time by a preeminent focus on elaborately carved figural pieces, especially diptychs and pyxides. Since the overwhelming majority of these artefacts have been discovered in secondary archaeological contexts, they are traditionally approached by art historical methods. Investigations of objects with a known find context gained prominence only slowly,5 while comprehensive regional surveys<sup>6</sup> are even scarcer. The recent years saw an increase of interest in the provenance of the raw material of these artefacts, including a differentiation between the possible main sources - namely African and Indian resources - of elephant ivory in Antiquity. Since both written and art historical evidence strongly suggests that Indian resources were available for and exploited by Mediterranean craftsmen and consumers during Late Antiquity, even if perhaps in lesser quantities than in Imperial Roman times,8 such an inquiry can significantly contribute to our

understanding of the late antique long-distance trade networks and economic relations. As archaeology alone is unable to provide answers in this case, the present study utilises FTIR and Raman spectroscopy, as well as carbon and nitrogen concentration and nitrogen isotope analyses to determine the possible origin of the ivory of the artefacts from the 6th- and 7th-century AD Carpathian Basin.

## HISTORICAL AND ARCHAEOLOGICAL CONTEXT

The Carpathian Basin of the 5th to 7th centuries AD provides an ideal field for regional study. The many peoples settling there from the 5th century AD onwards, including Huns, Goths, Skirs, Heruls, Langobards, Gepids, etc., maintained intimate political and economic contacts with both the Mediterranean world and the Roman and post-Roman West. After the arrival of the nomadic Avars in the later 6th century AD, both the importance of the late Roman economic and cultural structures and the nature of the long-distance connections with the Mediterranean world and the post-Roman West changed significantly. Yet, the influx of foreign products in the form of diplomatic gifts, booty, and commercial goods to the territories under their rule continued, and its importance increased even more.

In the study area, archaeological artefacts identified as ivory in previous publications came to light in at least six sites dated between the 5th and the 7th centuries AD (Map 1). Since a few years ago, we discussed these findings from five of the six sites in two in-depth studies, 10 while the sole remaining artefact is thoroughly investigated by Ágnes B. Tóth and her colleagues in the present volume, 11 we can be content with enlisting the sites and finds in question with their most basic data. The artefacts in question are:

- 1. Hauskirchen, Grave 8 (Austria):<sup>12</sup> Annular decorative ring, probably part of a purse, recovered from the burial of an adult woman; first half/mid-6th century AD.
- 2. Jánossomorja/Mosonszentjános, Grave 12 (Hungary):<sup>13</sup> Ten gaming pieces of similar design but in two different sizes from the burial of an adult man; second half of the 6th century AD. (Fig. 1)
- 3. Kölked-Feketekapu, Cemetery A, Grave 539 (Hungary): <sup>14</sup> Conical disc with a hole in its centre (probably a gaming piece of a spindle-whorl) from the burial of an adult; mid-7th century AD. (Fig. 2)



<sup>&</sup>lt;sup>2</sup>See the discussion in Bollók and Koncz (2020) with previous literature.

<sup>&</sup>lt;sup>3</sup>Drauschke (2011) 119–123.

<sup>&</sup>lt;sup>4</sup>Gaborit-Chopin (1978).

<sup>&</sup>lt;sup>5</sup>Cf., e.g., Cutler (1993); Drauschke (2011).

<sup>&</sup>lt;sup>6</sup>Drauschke (2011) 113–125; Bollók and Koncz (2020); Koncz and Bollók (2021).

<sup>&</sup>lt;sup>7</sup>For previous attempts, see the discussion below.

<sup>&</sup>lt;sup>8</sup>See the discussion of available evidence in Bollók and Koncz (2020).

<sup>&</sup>lt;sup>9</sup>See the latest in-depth synoptical discussions of the topic in Bálint (2019) and Bollók (2021).

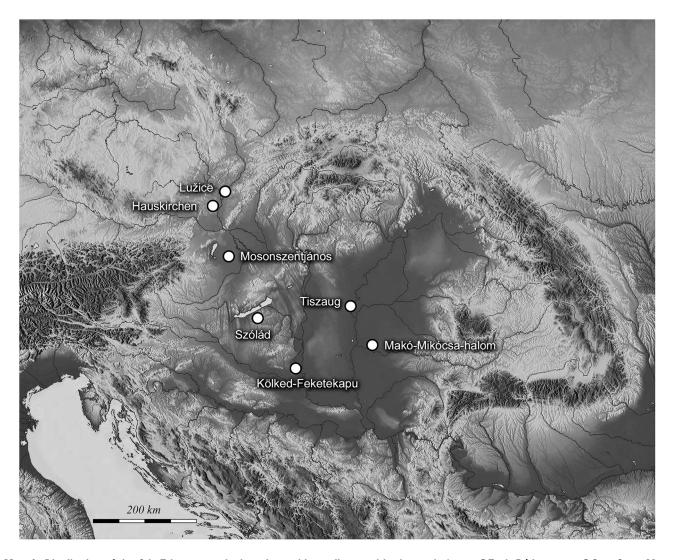
<sup>&</sup>lt;sup>10</sup>Bollók and Koncz (2020); Koncz and Bollók (2021).

<sup>&</sup>lt;sup>11</sup>B. Tóth et al. (2023).

<sup>&</sup>lt;sup>12</sup>Stadler (2008) 267–270; Koncz and Bollók (2021) 19–21.

<sup>&</sup>lt;sup>13</sup>Koncz and Tóth (2016); Koncz and Bollók (2021) 20-21.

<sup>&</sup>lt;sup>14</sup>Kiss (1996) 142–143, Taf. 95.11; Koncz and Bollók (2021) 21–22. The raw material was identified by István Vörös.



Map 1. Distribution of the 6th-7th-century elephant ivory objects discussed in the study (map: ©Zsolt Réti; source: ©OpenStreetMap, https://maps-for-free.com/#close, http://openstreetmap.org/)

- 4. Makó-Mikócsa halom, Grave 127 (Hungary):<sup>15</sup> Elongated bar of a toggle clasp-like purse lock from the horse burial of an adult man; beginning of the 7th century AD.
- 5. Szólád-Kertek mögött, Grave 38 (Hungary):<sup>16</sup> Annular decorative ring, probably part of a pouch, recovered from the burial of a young girl; mid-6th century AD. (Fig. 3)
- 6. Tiszaug-Országúti bevágás, Grave 301 (Hungary):<sup>17</sup> Conical object (possibly a gaming piece) from the burial of a 9–11-year-old child; mid to late 6th century AD. (Fig. 4)

Besides, two unpublished ivory finds (i.e., fragments of two unidentifiable artefacts) are known from an Early Avar Period burial at Bojt-Gulya legelő and a 6th-century AD settlement structure at Zamárdi-Kútvölgyi-dűlő, respectively. We shall also mention two additional annular decorative rings discovered in Graves 94 and 119 of the Lužice cemetery (Czech Republic), a Langobard Period burial site located on the north-western edge of the Carpathian Basin, which, strictly speaking, is outside our study area. Upon being examined under a light microscope, a yet another late 5th- to early 6th-century AD belt buckle from Hódmezővásárhely-Dilinka (Hungary), identified as ivory in previous publications, was proved to be made of soapstone, and, thus, excluded from our analysis.



<sup>&</sup>lt;sup>15</sup>The grave is still unpublished and we did not have the opportunity to examine the finds. The data listed here were kindly provided by the leader of the excavation, Csilla Balogh (İstanbul Medeniyet Üniversitesi Sanat Tarihi Bölümü), whom we wish to thank for sharing this information with us. Cf. Koncz and Bollók (2021) 21.

<sup>&</sup>lt;sup>16</sup>von Freeden (2008) 405-407; Koncz and Bollók (2021) 21, 24.

<sup>&</sup>lt;sup>17</sup>B. Tóth et al. (2023).

<sup>&</sup>lt;sup>18</sup>We are grateful to Márta Daróczi-Szabó for calling our attention to these two unpublished finds and sharing the above information with us.

<sup>&</sup>lt;sup>19</sup>Klanica and Klanicová (2011) 287-288, Taf. 72.15.

<sup>&</sup>lt;sup>20</sup>Nagy (2005) 102. Previously, Csallány (1961) 125, Pl.230.14 described the buckle as bronze.

<sup>&</sup>lt;sup>21</sup>Cf. Koncz and Bollók (2021) 21-23.





Fig. 1. Set of elephant ivory gaming pieces, Mosonszentjános, Grave 12 (after Koncz and Tóth, 2016, fig. 2)







Fig. 2. Elephant ivory spindle whorl or gaming piece, Kölked-Feketekapu A, Grave 539 (Photo: ©Hungarian National Museum, Budapest)

Except for the unpublished ivory fragments from Zamárdi, all finds listed above originate from a funerary context. The graves from Szólád, Hauskirchen, Mosonszentjános, Tiszaug,



Fig. 3. Elephant ivory pouch ring, Szólád, Grave 38 (Photo: ©Péter Skriba, ©Tivadar Vida, ©Rippl-Rónai Museum, Kaposvár)

and Lužice are dated to the mid and late 6th, whereas the graves from Makó-Mikócsa halom and Kölked-Feketekapu belong to the early and mid 7th century AD, respectively. The cemeteries of Hauskirchen, Lužice, Mosonszentjános, and Szólád are located in the presumed settlement area of the Langobards, while Tiszaug lies in the area of the Gepid Kingdom. Under scrutiny, the late 6th-century AD dating of some of the burials raises the strong possibility that the deceased were interred already during the earliest decades of the Avar rule of the Carpathian Basin. In contrast, the early Avar Period dating of the Makó and Kölked burials is evident. With the exception of Kölked, all burials can be considered rich or lavishly furnished in their respective contexts. Based on the other items in the related grave find assemblages, all six or, with the inclusion of Lužice, all seven - cemeteries show intensive long-distance connections with the Mediterranean and/or the post-Roman West.

# **AIMS**

The main aim of this paper is to trace the geographic origin of the raw materials of the fifteen elephant ivory artefacts known from the study area. This is interesting on several counts. Firstly, a survey of the available written and art historical evidence shows that – in view of the general image of the late antique ivory trade in the Mediterranean – while





Fig. 4. Conical object, Tiszaug-Országúti bevágás, Grave 301 (Photo: Szabolcs Dankó)

the employment of African elephant ivory for crafting our pieces is considerably more likely than that of Indian ones, the latter possibility can hardly be excluded.<sup>22</sup> Secondly, a formal analysis of the ivory artefacts under study yielded no definite conclusions on the precise place of manufacturing in most cases. Yet, what is certain is that the annular decorative rings were, in all probability, imported from the West. A similar - most likely Italian - origin can be assumed for the gaming set from Mosonszentjános, while formal analogies of the purse lock bar from Makó point towards the Central and Eastern Balkans and the more easterly regions of the Mediterranean.<sup>23</sup> Even less can be said of the Kölked and the Tiszaug pieces, the former of which might even be a reused object.<sup>24</sup> This diversity definitely raises the possibility that 6th- and 7th-century AD workshops in different regions had access to and used different raw materials. This is corroborated by the fact that the cultural and trade connections of the populations of the Gepid and Langobard Kingdoms differed significantly during their parallel existence in the first two-thirds of the 6th century AD, while the related networks underwent a fundamental restructuring with the arrival and settling in of the Avars and the emergence of the Avar Khaganate. This

circumstance clearly raises the possibility – at least on a theoretical level – that not all of the investigated 6th- and 7th-century AD elephant ivory finds originate from the same major (African or Indian) source(s).

For the above reasons, we attempted to analyse as many ivory samples in the present study as possible. The available finds included the gaming pieces from Mosonszentjános, the annular decorative ring from Szólád, the conical disc from Kölked, and the conical artefact from Tiszaug.

# MATERIALS AND METHODS

Altogether sixteen archaeological samples were analysed: thirteen, including three repairs, from ten gaming pieces from Mosonszentjános, and one from each piece from Szólád, Kölked, and Tiszaug, respectively (Table 1). For comparison, a series of recent elephant ivory objects from the Mammal Collection of the Hungarian Natural History Museum (Table 1) was studied under identical conditions and using the same analytical methods. Only ivories of proven African and Asian origin were selected for this comparative set. The African ivory pieces (seven samples, savannah elephant) were seized goods, whereas the single Asian ivory piece came from a private collection. Small amounts of material (up to a few tens of milligrams) were taken from all investigated ivory objects and powdered for further analysis.

In order to determine the African or Asian provenance of the ivory findings, a variety of analytical methods, namely



<sup>&</sup>lt;sup>22</sup>Bollók and Koncz (2020).

<sup>&</sup>lt;sup>23</sup>Cf. Koncz and Bollók (2021).

<sup>&</sup>lt;sup>24</sup>Cf. Koncz and Bollók (2021) 21.

Table 1. The archaeological ivory artefacts (repairs denoted as 'R' in the sample name) and modern ivory pieces analysed in this study

| Archaeological ivory |    |  |                    |                                      |  |
|----------------------|----|--|--------------------|--------------------------------------|--|
| Inv. no.             |    | Site   | Object             | Dating                               |  |
| 66.5.23.1            |    | Mosonszentjános (Hungary)                        | gaming piece       | second half of the 6th century AD    |  |
| 66.5.23.2            |    | Mosonszentjános (Hungary)                        | gaming piece       | second half of the 6th century AD    |  |
| 66.5.23.3            |    | Mosonszentjános (Hungary)                        | gaming piece       | second half of the 6th century AD    |  |
| 66.5.23.4            |    | Mosonszentjános (Hungary)                        | gaming piece       | second half of the 6th century AD    |  |
| 66.5.23.4R           |    | Mosonszentjános (Hungary)                        | gaming piece       | second half of the 6th century AD    |  |
| 66.5.23.5            |    | Mosonszentjános (Hungary)                        | gaming piece       | second half of the 6th century AD    |  |
| 66.5.23.5R           |    | Mosonszentjános (Hungary)                        | gaming piece       | second half of the 6th century AD    |  |
| 66.5.23.6            |    | Mosonszentjános (Hungary)                        | gaming piece       | second half of the 6th century AD    |  |
| 66.5.23.7            |    | Mosonszentjános (Hungary)                        | gaming piece       | second half of the 6th century AD    |  |
| 66.5.23.8            |    | Mosonszentjános (Hungary)                        | gaming piece       | second half of the 6th century AD    |  |
| 66.5.23.8R           |    | Mosonszentjános (Hungary)                        | gaming piece       | second half of the 6th century AD    |  |
| 66.5.24.1            |    | Mosonszentjános (Hungary)                        | gaming piece       | second half of the 6th century AD    |  |
| 66.5.24.2            |    | Mosonszentjános (Hungary)                        | gaming piece       | second half of the 6th century AD    |  |
| Szólád               |    | Szólád-Kertek mögött (Hungary)                   | pouch ring         | second half of the 6th century AD    |  |
| Kölked               | Kö | ked-Feketekapu, cemetery A (Hungary)             | conical disc       | 7th century AD                       |  |
| Tiszaug              | ,  | Tiszaug-Országúti bevágás, cemetery<br>(Hungary) | conical object     | second half of the 6th century AD    |  |
| Modern ivory         |    |  |                    |                                      |  |
| Inv. no./Sample no.  |    | Country of origin                                | Species            | Sampling site                        |  |
| 59.76.1              |    | Kenya  | Loxodonta africana | basal end of tusk                    |  |
| 59.77.1              |    | Tanzania   | Loxodonta africana | basal end of tusk                    |  |
| 59.78.1              |    | Uganda   | Loxodonta africana | top end of the pulp void of the tusk |  |
| 2001.1.10            |    | Cameroon   | Loxodonta africana | basal end of tusk                    |  |
| 2001.1.11            |    | Cameroon   | Loxodonta africana | end of cut tusk                      |  |
| 2016.4.5             |    | Nigeria  | Loxodonta africana | basal end of tusk                    |  |
| 2016.4.6             |    | Nigeria  | Loxodonta africana | basal end of tusk                    |  |
| Asian                |    | ivory from a private collection                  | Elephas maximus    | basal end of a cut tusk              |  |

FTIR and Raman spectroscopy, carbon and nitrogen concentration, and nitrogen isotope analyses, were applied.<sup>25</sup>

As a composite material, ivory consists of inorganic and organic constituents. The collagen protein matrix incorporates bioapatite, which is carbonate-substituted hydroxylapatite (dahllite,  $Ca_5[(PO_4,CO_3)_3]OH)$ , i.e., a carbonate- and hydroxyl-bearing phosphate. Vibrational spectroscopic methods, like FTIR and Raman spectroscopy,

are capable of identifying collagen, phosphate, and carbonate molecules as their diverse vibrations appear as specific bands in the spectra. Vibrational spectroscopy is widely used to identify the species in the case of modern ivory samples.<sup>27</sup> In contrast to modern ivory items, interred archaeological artefacts made from ivory are affected by degradation and physical and chemical alterations due to taphonomic processes. During the degradation of ivory, organic components decompose and, thus, vanish from the bone to various degrees. At the same time, apatite recrystallises, resulting in a higher degree of apatite crystallinity in ancient ivory. Additionally, either the amount of carbonate in the apatite tends

<sup>&</sup>lt;sup>27</sup>Edwards et al. (1997, 1998); Brody et al. (2001); Edwards (2004); Edwards et al. (2006b); Banerjee et al. (2008); Turner-Walker and Xu (2014).



<sup>&</sup>lt;sup>25</sup>Wang et al. (2007); Drauschke and Banerjee (2007); Schuhmacher et al. (2009); Edwards et al. (2006a); Long et al. (2008); Banerjee et al. (2011); Edwards and O'Connor (2012); Nocete et al. (2013); García Sanjuán et al. (2013); Rozalen and Ruiz Gutierrez (2015); Morillo León et al. (2018); Li et al. (2023).

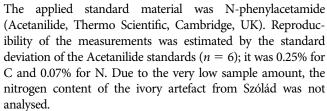
<sup>&</sup>lt;sup>26</sup>Weiner and Wagner (1998).

to decrease or carbonate becomes incorporated in the apatite from the soil or sediment while the object is interred. These processes advance the drying out of the bone, which, thus, becomes more and more brittle and friable over time. Therefore, the infrared and Raman spectra of archaeological ivory do not necessarily match the spectra of recent ones. For example, in the spectra of archaeological items, the intensity of the bands related to phosphate may be increased, whereas those linked to organic components decreased. Several papers have presented case studies where vibrational spectroscopy was used to determine the state of preservation of a sample or identify the species of the raw material of ivory findings, also discussing the limitations of the related methods in studying archaeological and fossil ivory. <sup>29</sup>

FTIR (Fourier-transformation infrared spectroscopy) analyses on powder samples were carried out using a Bruker Vertex 70 IR spectrometer in ATR (Attenuated Total Reflectance) mode at the Institute for Geological and Geochemical Research (RCAES, Budapest). For each sample, 32 scans were recorded in the 4,000–400 cm<sup>-1</sup> spectral range with a resolution of 4 cm<sup>-1</sup>. The spectra were processed using OPUS 7.2 software (Bruker Optik GmbH, Germany).

Raman microspectroscopy analysis of the same powder samples (except for the Kölked piece) was carried out using a HORIBA JobinYvon LabRAM HR (high resolution) 800 dispersive, edge-filter based confocal Raman spectrometer (focal length: 800 mm) equipped with an Olympus 21 BXFM microscope, at the Research and Instrument Core Facility (RICF, Faculty of Science, Eötvös Loránd University, Budapest). The spectra were collected using a Nd:YAG laser ( $\lambda=532$  nm) excitation, 1,800 grooves/mm optical grating, a 200 µm confocal hole, a 5–10 s acquisition time, and a 100 × (NA 0.9) objective. Data evaluation (background fitting and peak fitting using the Gaussian–Lorentzian sum profiles) was done using LabSpec 5.41.15 software (HORIBA Scientific).

Stable nitrogen isotope ratios and element concentrations (C, N) of the powder samples were measured at the Institute for Geological and Geochemical Research (RCAES, Budapest). Duplicate aliquots of 2.5–3 mg of the ivory samples were weighed and packed into tin capsules (IVA Analysetechnik e.K. Meerbusch, Germany) for C and N concentration analysis. The element-specific peak area was detected by the internal TCD detector of the Flash 2000 Organic Elemental Analyzer (Thermo Scientific, Rhodano, Italy). The element concentration was calculated as  $X\% = ((A_{spl}^X - A_{blk}^X)/K)/(W_{spl})^*100$ , where X stands for C or N,  $A_{spl}$  and  $A_{blk}$  stand for the peak area of the sample and the blank, respectively, and K is the average K-factor determined by a six-point calibration curve fitted with the K-factor function in the mass range of 1–3 mg.  $^{30}$ 



Aliquots of 2.5–3 mg of the ivory samples were weighed and packed into tin capsules for stable nitrogen isotope analysis. Samples were combusted using the Flash 2000 Organic Elemental Analyzer (mentioned above) under the following operational conditions: 1,000 °C combustion reactor, 640 °C reduction reactor, 75 °C GC temperature. Yielded gases were transferred via a Conflo III into an isotope ratio mass spectrometer (Delta V Advantage, Thermo Finnigan, Bremen, Germany) operating in continuous flow mode with a carrier gas (He + O<sub>2</sub>, Messer 4.6) with a 90-95 ml/min flow speed. Nitrogen stable isotope ratios of the combusted material were measured and reported per mil (‰) using the  $\delta$  notation. Two-point linear normalisation was applied for  $\delta^{15}N$  to convert raw isotope values into the internationally recognised AIR scale. Two laboratory standards (carbamide and gelatine) were used for the calibration of nitrogen. Carbamide and gelatine were calibrated independently to isotopic reference materials IAEA-N-1 ( $\delta^{15}$ N: +0.4‰) and IAEA-N-2 ( $\delta^{15}$ N: +20.3‰). The standard deviation of the reference standards analysed at every sixth place was <0.05% for  $\delta^{15}N$  (n = 6), indicating the reproducibility of the measurements. Due to sample inhomogeneity, duplicates were measured.

# RESULTS AND DISCUSSION

#### FTIR spectroscopy

All ivory artefacts from Mosonszentjános, including repairs, exhibit similar FTIR spectra (Fig. 5). The intense bands at 713, 874 and 1,416 cm<sup>-1</sup> in the spectrum of the Szólád piece mark the carbonate that was incorporated in the ivory while the item was underground.<sup>32</sup> In contrast to the gaming pieces from Mosonszentjános, the bands at ~1,240 and ~1,340 cm<sup>-1</sup>, marking organic substance, are absent from the spectra of the Kölked, Szólád, and Tiszaug



<sup>&</sup>lt;sup>28</sup>Wang et al. (2007); Long et al. (2008); Edwards and O'Connor (2012).

<sup>&</sup>lt;sup>29</sup>Edwards et al. (2006a); Wang et al. (2007); Drauschke and Banerjee (2007); Long et al. (2008); Schuhmacher et al. (2009); Edwards and O'Connor (2012); Nocete et al. (2013); Garcia Sanjuán et al. (2013); Rozalen and Ruiz Gutierrez (2015); Morillo León et al. (2018); Li et al. (2023)

<sup>&</sup>lt;sup>30</sup>Hedges and Stern (1984).

<sup>&</sup>lt;sup>31</sup>For the detailed assignments of the absorption bands, see Michel et al. (1995) and Wang et al. (2007).

<sup>&</sup>lt;sup>32</sup>Wang et al. (2007).

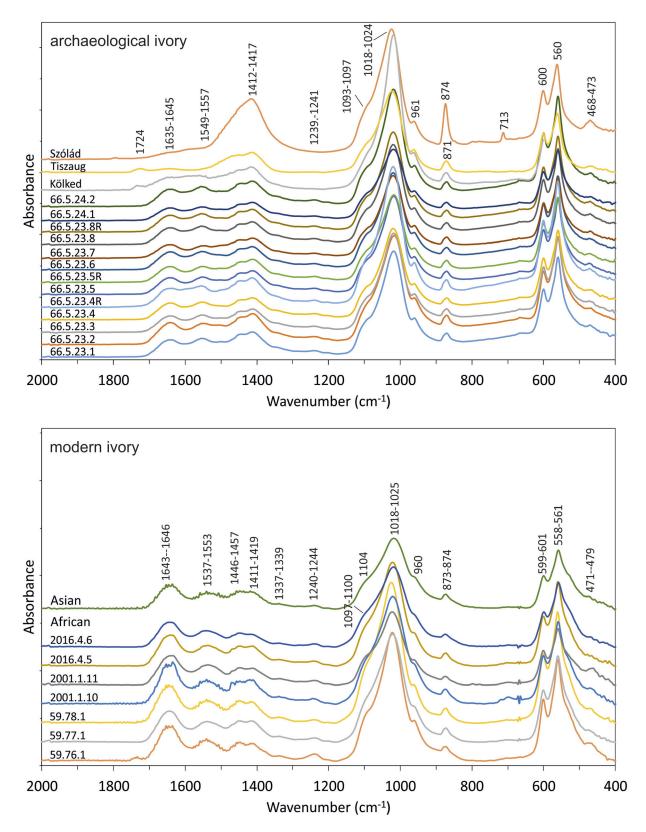


Fig. 5. Fourier-transform infrared spectra in the region 2,000-400 cm<sup>-1</sup> of the studied archaeological ivory findings from Moson-szentjános, Kölked, Szólád, and Tiszaug (Inv. numbers beginning with 66 indicate objects from Mosonszentjános, including repairs denoted as 'R'), as well as the studied modern African and Asian elephant ivory pieces from the Mammal Collection of the Hungarian Natural History Museum. The bands at 713 cm<sup>-1</sup>, 814 cm<sup>-1</sup>, and 1,416 cm<sup>-1</sup> in the spectrum of the sample from Szólád indicate the carbonate incorporated in the material possibly while the finding was underground

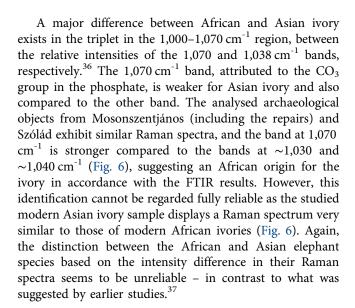


pieces. The bands at ~1,550 cm<sup>-1</sup> (related to both the carbonate group of the apatite and organic components) and ~1,640–1,650 cm<sup>-1</sup> (related to organic components) are weak or absent in the spectra of the Kölked, Szólád, and Tiszaug samples and stronger in the spectra of the finds from Mosonszentjános. Seemingly, the organic components in the material of the Kölked, Szólád, and Tiszaug pieces were highly degraded, while they were better preserved in the ivory finds from Mosonszentjános.

According to some studies, African and Asian elephant ivory can be distinguished via FTIR analysis. Hydroxylapatite crystals are smaller in Asian ivory than in African, which affects the shape and intensity of the absorption bands in their FTIR spectra, resulting in a more intense absorption between 1,500 and 500 cm<sup>-1</sup> for African ivory.<sup>33</sup> Additionally, there is a weak difference near the most intense peak at ~1,033 cm<sup>-1</sup>, related to the phosphate group of hydroxylapatite. The FTIR spectrum of African elephant ivory is characterised by a distinct shoulder near 1,100 cm<sup>-1</sup>, where Asian ivory only features a 'smooth slope'. 34 Based on the clear presence of the 1,100 cm<sup>-1</sup> (1,093-1,097 cm<sup>-1</sup>) band in the FTIR spectra of the Mosonszentjános, Szólád, and Tiszaug samples, these finds might have been made from African elephant ivory, whereas the slope at around 1,100 cm<sup>-1</sup> without a shoulder in the spectrum of the Kölked sample possibly indicates the Asian origin of the raw material of the finding (Fig. 5). However, the FTIR analysis of the modern African and Asian ivory samples from the Mammal Collection of the Hungarian Natural History Museum indicates that the shoulder at 1,100 cm<sup>-1</sup> appears not only in the spectra of the modern African but also of the modern Asian elephant (Fig. 5). Therefore, the distinction between the African and Asian elephant species is highly unreliable as it is only based on a very slight difference in their FTIR spectra, or, alternatively, the provenance of the modern Asian ivory has not been determined correctly (i.e., the ivory is of African rather than Asian origin).

#### Raman spectroscopy

As a complementary method to FTIR spectroscopy, Raman spectroscopic analysis was also performed on the archaeological and modern ivory samples. As our goal was to determine the provenance of the archaeological artefacts, we focused on the 800–1,200 cm<sup>-1</sup> region of their Raman spectra. The piece from Kölked was not analysed. The ivory artefact from Tiszaug was analysed but did not yield a relevant spectrum, most probably because it is in an advanced state of degradation. Only one identifiable band has appeared at 963 cm<sup>-1</sup>, i.e., the most intense band marking the phosphate (PO<sub>4</sub>) in animal and human bones, which, therefore, cannot be considered diagnostic.<sup>35</sup>



# Carbon and nitrogen concentrations and nitrogen isotope ratio ( $\delta^{15}N$ )

The archaeological ivory samples from Mosonszentjános exhibit narrow ranges for carbon and nitrogen contents (mostly 12.4–14.1% for carbon and 3.3–3.7% for nitrogen) and a broad range for  $\delta^{15}N$  (8.3–15.9‰) (Table 2, Fig. 7). Each of them exhibit, however, distinctly dissimilar values for C and N content and  $\delta^{15}$ N, which suggests that these artefacts might have been made from different tusks or at least from several pieces of ivory. The ivory used for repair is significantly different from the original material of two Mosonszentjános objects (66.5.23.4, 66.5.23.8) both in element concentration and nitrogen isotope ratio, implying that the repairs were made from ivory pieces different from the raw material of the original objects. The C and N contents of the third repair are similar to that of the original material of the repaired object from Mosonszentjános (66.5.23.5), but their  $\delta^{15}N$  values are distinct; therefore, we suspect that the ivory piece used for the repair might have been different from the original object in this case as well. One should keep in mind, however, that ivory may have intra-tusk variability in C and N content and δ<sup>15</sup>N value,<sup>38</sup> which may weaken our interpretation.

The Szólád, Kölked, and Tiszaug pieces exhibit lower C and N contents compared to most Mosonszentjános artefacts; only the artefact from Kölked has carbon and nitrogen contents similar to one of the Mosonszentjános repairs (66.5.23.8R) (Table 2, Fig. 7). The Tiszaug piece contains the lowest amount of carbon and nitrogen (5% C and 0.1% N) (Table 2, Fig. 7).

Except for two outliers (ivory pieces from Cameroon), most modern ivory samples analysed in this study have a carbon content between 14.6% and 16.8% and a nitrogen



<sup>&</sup>lt;sup>33</sup>Nocete et al. (2013).

<sup>&</sup>lt;sup>34</sup>Drauschke and Banerjee (2007); Banerjee et al. (2008); Nocete et al. (2013); Morillo León et al. (2018).

<sup>&</sup>lt;sup>35</sup>Edwards et al. (1997, 1998, 2006a, 2006b).

<sup>&</sup>lt;sup>36</sup>Edwards et al. (1997, 1998).

<sup>&</sup>lt;sup>37</sup>Edwards et al. (1997, 1998); Banerjee et al. (2008); Nocete et al. (2013).

 $<sup>^{38}</sup>$ For  $\delta^{15}$ N value see Ziegler et al. (2016).

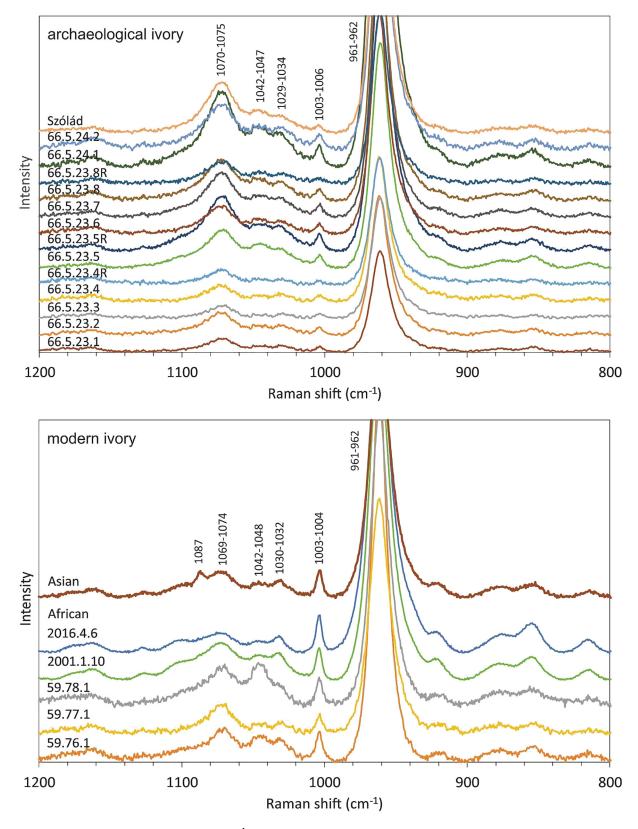


Fig. 6. Raman spectra in the region 1,200–400 cm<sup>-1</sup> of the studied archaeological ivory findings from Mosonszentjános and Szólád (Inv. numbers beginning with 66 indicate objects from Mosonszentjános, including repairs denoted as 'R' in the sample name), as well as the studied modern African and Asian elephant ivory pieces from the Mammal Collection of the Hungarian Natural History Museum



Table 2. Carbon and nitrogen content and nitrogen isotope ratio of the studied archaeological and modern ivory samples (including repairs denoted as 'R' in the sample name). Due to a very low sample amount, the Szólád piece could not be analysed for nitrogen content

| Sample               | C%    | N%   | δ <sup>15</sup> N (‰) |  |  |  |
|----------------------|-------|------|-----------------------|--|--|--|
| Archaeological ivory |       |      |                       |  |  |  |
| 66.5.23.1            | 14.08 | 3.57 | 11.26                 |  |  |  |
| 66.5.23.2            | 14.00 | 3.68 | 8.40                  |  |  |  |
| 66.5.23.3            | 13.30 | 3.47 | 10.88                 |  |  |  |
| 66.5.23.4            | 13.87 | 3.67 | 10.47                 |  |  |  |
| 66.5.23.4R           | 12.44 | 2.74 | 6.63                  |  |  |  |
| 66.5.23.5            | 13.45 | 3.58 | 7.31                  |  |  |  |
| 66.5.23.5R           | 13.72 | 3.43 | 9.69                  |  |  |  |
| 66.5.23.6            | 13.65 | 3.61 | 10.34                 |  |  |  |
| 66.5.23.7            | 12.35 | 3.33 | 15.88                 |  |  |  |
| 66.5.23.8            | 13.48 | 3.49 | 10.17                 |  |  |  |
| 66.5.23.8R           | 10.95 | 2.42 | 8.34                  |  |  |  |
| 66.5.24.1            | 12.87 | 3.28 | 11.71                 |  |  |  |
| 66.5.24.2            | 13.86 | 3.43 | 8.25                  |  |  |  |
| Szólád               | 8.27  | -    | 8.33                  |  |  |  |
| Kölked               | 11.23 | 2.59 | 14.17                 |  |  |  |
| Tiszaug              | 4.95  | 0.11 | 11.62                 |  |  |  |
| Modern ivory         |       |      |                       |  |  |  |
| 59.76.1              | 16.05 | 5.34 | 13.40                 |  |  |  |
| 59.77.1              | 15.84 | 5.12 | 11.97                 |  |  |  |
| 59.78.1              | 15.47 | 5.10 | 8.45                  |  |  |  |
| 2001.1.10            | 6.86  | 2.26 | 8.26                  |  |  |  |
| 2001.1.11            | 20.96 | 6.75 | 9.38                  |  |  |  |
| 2016.4.5             | 16.83 | 5.16 | 8.94                  |  |  |  |
| 2016.4.6             | 14.60 | 4.85 | 8.60                  |  |  |  |
| Asian                | 14.93 | 4.98 | 10.14                 |  |  |  |

content between 4.9% and 5.3%, respectively (Table 2, Fig. 7). These data are very close to and partly overlap with the C and N data of modern African forest elephant ivory, published earlier (mainly in the form of bivariate plots and not publicly available data sets),<sup>39</sup> although one of our modern African ivory samples shows values in the range characteristic of modern Asian (Indian) elephant ivory. Comparing the C and N content of the archaeological samples to those of modern African and Asian ivory measured in this study, it becomes evident that archaeological ivory samples have remarkably lower element contents (Table 2, Fig. 7). Very low carbon and nitrogen

contents – similar to the Tiszaug piece – were measured previously in archaeological ivory findings from Morocco and Spain (Fig. 7).<sup>40</sup> The decrease in carbon and nitrogen contents in archaeological finds is related to the degradation and loss of organic components in ivory with time. The highly degraded state of the material of the Tiszaug piece has been verified by optical microscopy<sup>41</sup> as well as FTIR analysis (discussed above). In conclusion, C and N contents themselves cannot be used for provenancing archaeological ivory.

Nitrogen isotope ratios of African ivory (bone collagen) are related to rainfall or water stress. Low  $\delta^{15}N$  values correspond with high rainfall conditions in forest habitats, whereas high  $\delta^{15}$ N values are characteristic of the more arid habitats of the savannah (areas with annual precipitation of <400 mm). 42 According to studies performed by van der Merwe et al. and Vogel et al., the ivory and bone of modern African elephants show a considerable variation in  $\delta^{15}$ N values from 2.4 to 14.6%, with isotopic differences for various elephants from different regions of Africa.<sup>43</sup> Based on the analysis of 31 tusks of Asian elephants, Singh et al. found that the δ<sup>15</sup>N values of Asian and African elephant ivory are significantly different. 44 As based on van der Merwe et al.'s data, the mean  $\delta^{15}N$  of African ivory (7.9  $\pm$ 0.59 %) is higher than that of Asian ivory (5.03  $\pm$  0.29 %), with no overlaps, Singh et al. suggested that  $\delta^{15}$ N could serve as a tool to distinguish between these elephant species. 45 The δ<sup>15</sup>N values of the modern African ivory samples measured in this study (8.6-13.4%) fall into the isotope range determined for African ivory by van der Merwe et al.; however, our single modern Asian ivory sample also shows a similar  $\delta^{15}$ N value (10.1‰). A recent study<sup>46</sup> has also indicated a considerable overlap of the nitrogen isotope ratios of African and Asian ivory (4.2-17.2 ‰ based on 487 African ivory samples from different parts of Africa and 5.9-11.2 ‰ based on 20 Asian ivory samples), corroborating our observation. The  $\delta^{15}$ N values of our archaeological ivory samples (8.3– 15.9‰) overlap with the  $\delta^{15}N$  ranges characteristic of modern African and Asian ivory. The degradation of organic components in archaeological ivory may again influence the nitrogen isotope ratio via fractionation of isotopes, and the measured δ<sup>15</sup>N values may not represent the original nitrogen isotope ratio of the ivory. Therefore, based on the considerations explicated above, the origin of the studied archaeological ivory samples, i.e. African vs Asian, cannot be determined with certainty based solely on the nitrogen isotope ratio.



<sup>&</sup>lt;sup>39</sup>Banerjee et al. (2008); Nocete et al. (2013).

<sup>&</sup>lt;sup>40</sup>Banerjee et al. (2011); García Sanjuán et al. (2013); Nocete et al. (2013).

<sup>&</sup>lt;sup>41</sup>B. Tóth et al. (2023).

<sup>&</sup>lt;sup>42</sup>van der Merwe et al. (1990).

<sup>&</sup>lt;sup>43</sup>van der Merwe et al. (1990); Vogel et al. (1990).

<sup>&</sup>lt;sup>44</sup>Singh et al. (2006).

<sup>&</sup>lt;sup>45</sup>Singh et al. (2006).

<sup>&</sup>lt;sup>46</sup>Ziegler et al. (2016).

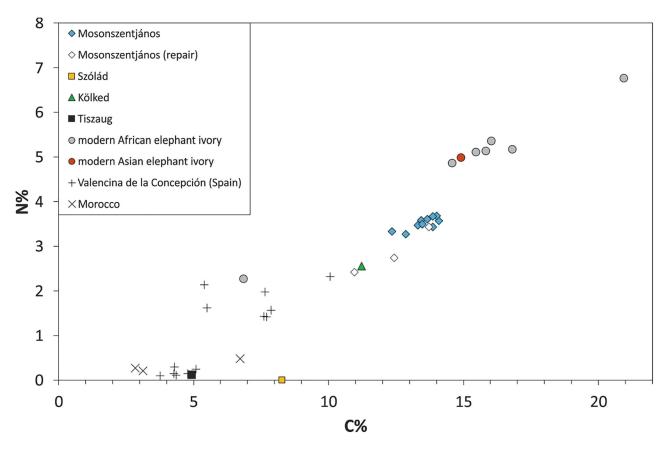


Fig. 7. Nitrogen vs. carbon content of the studied archaeological ivory findings from Mosonszentjános, Szólád, Kölked, and Tiszaug (Inv. numbers beginning with 66 indicate objects from Mosonszentjános, including repairs denoted as 'R' in the sample name). Archaeological ivory objects from Morocco (Banerjee et al., 2011) and Spain (Nocete et al., 2013; García Sanjuán et al., 2013), as well as modern African and Asian ivory pieces from the Mammal Collection of the Hungarian Natural History Museum are indicated for comparison. As the N content of the Szólád sample could not be determined due to small sample size, it is indicated as 0% N in the figure

In conclusion, while some of the used analytical methods, such as carbon and nitrogen concentration and nitrogen isotope analyses, provided some data, these are of no help in identifying the provenance of the raw material of the ivory findings from Mosonszentjános, Szólád, Kölked, and Tiszaug. Although compared to earlier ivory provenance studies, FTIR and Raman spectroscopy suggests a likely African origin for most of our findings, except probably for the Kölked piece, the distinction between the ivory of different elephant species based on these vibrational spectroscopic methods seems not to be robust enough to determine the African origin with all certainty.

# **CONCLUSIONS**

Based on the available written and art historical evidence, it seemed likely that, during the centuries of Late Antiquity, African elephant ivory resources were significantly more widely relied upon than Indian ones by the ivory workshops in the Mediterranean and north of the Alps. However, analysing historical sources alone could not provide

unquestionable evidence in this regard.<sup>47</sup> This is also true for the raw material of the ivory artefacts from archaeological contexts in the Carpathian Basin. Questions regarding the utilisation of multiple sources were also raised by the quite diverse character of the finds. Although they were all buried within about a century or only slightly more – approximately between the mid-6th and mid-7th centuries AD, with a noticeable concentration around the mid- and late 6th century AD, a transitional period leading from the last decades of the Langobard and Gepid rule to the time of the Early Avar Khaganate –, their formal analogies point toward both the post-Roman West, including Italy, and the Eastern Roman Empire.

Despite this heterogeneity, the results of the scientific analyses presented in the current paper point towards an African origin of the elephant tusks that became the raw material of most studied pieces. The single exception seems to be the conical object from Kölked, in the case of which the possibility of Asian origin could not be excluded. This

<sup>&</sup>lt;sup>47</sup>Bollók and Koncz (2020); Koncz and Bollók (2021).



artefact appears to be a possible outlier from the series in two other respects as well: the burial that contained it is the youngest amongst the graves discussed in this paper; furthermore, the object was discovered among a series of *archaicas*, which, alongside its fairly simple form that makes an exact dating quite difficult, raise the possibility that it was manufactured significantly earlier than interred.

Based on the above, the results of the studies exploring the historical and art historical sources and the scientific analyses complement each other neatly, all pointing to an overwhelmingly African origin of the tusks used by late antique craftsmen for making carvings in Europe and most of the Mediterranean. This holds true for the elephant ivory finds recovered from 6th-to 7th-century AD contexts in the Carpathian Basin in particular, but also for coeval Mediterranean and Western European workshops in general.

### **DECLARATION**

The chapters on scientific analysis were written by Bernadett Bajnóczi, while the historical and archaeological sections are the work of Ádám Bollók and István Koncz. The authors declare that they have contributed equally to the article and have no conflict of interest.

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<sup>&</sup>lt;sup>48</sup>A recent study addressing the same research question by using biomolecular methods was just published after our paper has been submitted for publication: Hemer et al. (2023). Despite the differences in the methods employed, the authors' results coincide with our conclusions.

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