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Lead isotope and geochemical analyses of copper-based metal artefacts from the Iron Age water burial in Levänluhta, Western Finland

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ABSTRACT

This article presents the results of the first-ever lead isotope (LI) analysis of copper-based archaeological artefacts found in the region of Finland. Eight metal objects recovered from the Iron Age water burial site of Levänluhta in western Finland were analysed via multi collector inductively coupled plasma mass spectrometry (MC-ICP-MS) and portable X-ray fluorescence spectrometry (pXRF) in order to attain geochemical and LI data. The majority of the objects are Merovingian period (ca. 550–800 CE) jewellery, displaying domestic Iron Age artefact styles, and were probably cast by local workshops in Finland. Until recently, the copper exploited in Bronze and Iron Age metallurgy in Finland had been linked to Scandinavian ores. However, this provenance scenario seems implausible in the light of recent LI studies demonstrating that Scandinavian Bronze Age artisans in fact relied on long-distance metal transport. Comparisons between the LI data of the analysed objects and published ore databases exclude the possibility of a domestic or Scandinavian copper source for the metals. Instead, it appears likely that the copper originated from southern European ores. The low arsenic and antimony levels in the copper alloys provide indication of long recycling patterns of the metals used in the Iron Age workshops in Finland. It is possible that the Iron Age artefacts contain recycled copper-alloys already acquired in the Bronze Age. The metals were transported for long distances, and it appears that the pan-European metal circulation network also crossed the Baltic Sea to reach coastal Finland.

1. Introduction

This article presents the results of a pilot study carried out at the University of Helsinki and the Geological Survey of Finland, in which copper alloy metal artefacts recovered from the Iron Age water burial site of Levänluhta in western Finland were analysed via multi collector inductively coupled plasma mass spectrometry (MC-ICP-MS) and portable energy-dispersive X-ray fluorescence spectrometry (pXRF) to acquire geochemical and lead isotope (LI) data of the objects. This artefact study was part of the broader research project of the Levänluhta burial site materials (Wessman et al., 2018), and is the first trial in the Finnish context of provenancing archaeological metal artefacts based on lead isotope (LI) data (see e.g., Gale and Stos-Gale, 2000; Niederschlag et al., 2003; Albaréde et al., 2012; Artioli et al., 2016; Radivojevic et al., 2018, and references therein).

The Levänluhta site is located in Isokyrö in Ostrobothnia, western Finland (Fig. 1). The site is the most extensive wetland cemetery in Finland, with recovered remains of at least 98 human individuals. The site is currently located in a wetland area (Fig. 2), including a couple of visible springs, but during the Iron Age, ca. 300–800 CE, it was probably a small pond. During that period, cremation was the prevailing burial practice in Finland, and water burials of unburned human remains were extremely rare. Furthermore, the gender-bias of the deceased and the lack of weapons and everyday objects as grave goods also make Levänluhta an unusual burial site (see Wessman, 2009, 2010; Wessman et al., 2018). Based on the osteological analysis of the human bone assemblage, most of the buried individuals were women and children (Formisto, 1993: 103; Niskanen, 2006: 29–30). Earlier interpretations of the site have varied from a sacrificial site to a mass grave for the victims of a famine, plague, or battle (Hackman, 1906; Leppäaho, 1949; Seger, 1982; Niskanen, 2006), however, no signs of trauma on the bones or other evidence supporting these speculations have yet been found (Wessman, 2009; Wessman et al., 2018). Previously, it was thought that the site was used only for a few decades

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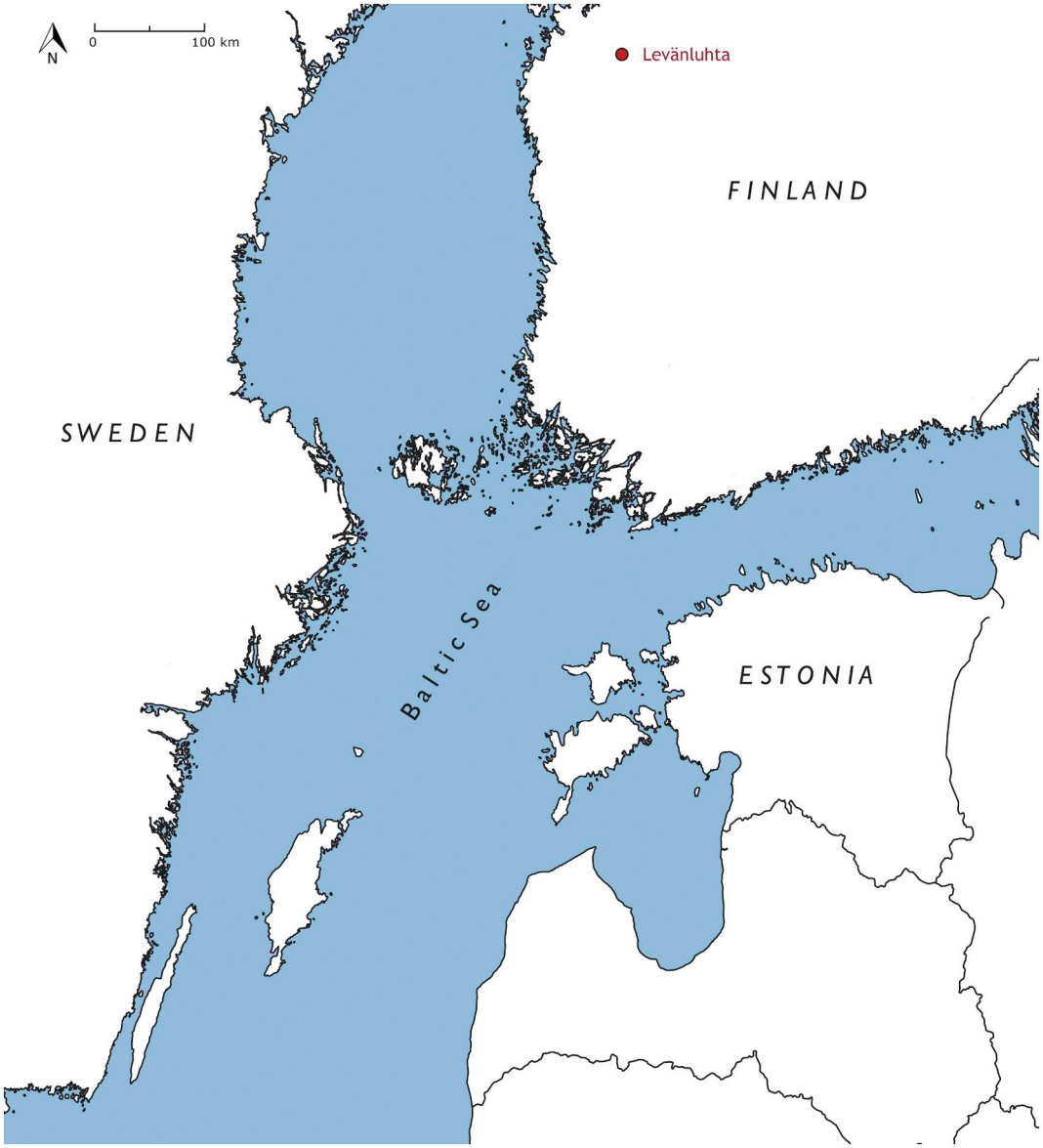

Fig. 1. Map of the Baltic Sea region showing the location of the Levänluhta site in Isokyrö in Ostrobothnia, western Finland (Map: E. Holmqvist).

during the Merovingian period (ca. 600–650 CE; e.g. Kivikoski, 1961; Seger, 1982; Lehtosalo-Hilander, 1984; Formisto, 1993; Niskanen, 2006), but new data from recent studies have indicated a longer time-span for the cemetery, ca. 300–800 CE (Wessman, 2009; Wessman et al., 2018). The first archaeological excavations at the site took place already in 1880s and 1910s, and fieldwork was continued in the 1980s (Meinander, 1950: 136; Meinander, 1977; Wessman, 2009: 82–84; Wessman et al., 2018).

In addition to the human and animal bones, altogether 22 metal artefacts (finger rings, arm and neck rings, brooches, a chain ornament, a Westland type bronze cauldron), unidentified metal rods, pieces of wood, burnt clay, and clay daub were found at the site, but notably no ceramic objects (Wessman, 2009, 2010; Wessman et al., 2018). The metal artefacts from the site are on display in the National Museum of Finland (Fig. 3). Based on the stylistic characteristics, it is probable that at least the jewellery recovered in Levänluhta were cast by local workshops in Finland. At the present time, however, no settlements or workshops have been identified in the immediate vicinity of the Levänluhta site, despite of recent intensive survey in the area (Wessman et al., 2018). The entire metal artefact assemblage was published in Wessman et al. (2018), including the data from the non-invasive pXRF analysis. The artefacts were manufactured from copper or a copper-based alloy (bronze, brass), and represent a variety of Iron Age object forms. In this article, we report the results of the ICP-MS analysis of eight of these artefacts (Fig. 4) sampled for lead isotope (^{204}Pb, ^{206}Pb, ^{207}Pb and ^{208}Pb) and geochemical analyses. Our aim was to determine the lead isotope ratios and identify possible geochemical signals in the objects deriving from the copper ore geology, in order to investigate the artefact provenance and possible raw material origins, i.e. trace the source area of the base metal used to forge these artefacts. Furthermore, our aim was to see if the compositional results acquired by the non-invasive pXRF analyses could be verified by the ICP-MS data, as well as to address possible inter-method data compatibility issues.

Until recently, the copper alloys exploited in the Bronze and Iron Age metallurgy in Finland have been associated with Scandinavian ores (see, e.g. Nordqvist and Herva, 2013: 418), based on the assumption that the prehistoric inhabitants of Finland were unfamiliar with domestic copper and tin resources. Accordingly, our hypothesis was that either the artefacts or the raw materials used in their manufacture would be of imported origin. The acquisition of Scandinavian metals, however, now seems unlikely in the light of recent LI studies revealing that southern Scandinavian Bronze Age artisans relied on long-distance

Fig. 2. The wetland context of the Levänluhta site (Photo: Tarja Sundell).

metal transport (Ling et al., 2013, 2014; Melheim et al., 2018). In this study, the LI and geochemical characteristics of the analysed objects were compared to domestic, Scandinavian, and European copper and lead ore databases.

2. Materials and methods

2.1. Analysed copper-based artefacts

The assemblage of copper-based metal artefacts recovered from Levänluhta consists of different artefact types, mainly Iron Age jewellery and personal adornments, including concave-convex and multi-zoned arm-rings, loop-shaped dragon brooches and equal armed brooches, neck-rings and finger rings, and one Westland-type cauldron (Wessman et al., 2018: Table 1). This paper concentrates on eight of these artefacts (Fig. 4, Table 1; NM = National Museum of Finland collection ID), which were subjected to pXRF and ICP-MS analyses. The analysed artefacts include two concave-convex arm-rings (sample A = NM2440:2, B = NM2440:3), a convex arm-ring (C = NM2440:5), the Westland type cauldron (D = NM2441:1), a neck-ring (E = NM2441:2), a small arm-ring made of a plain rod with a flattened end made of brass (F = NM6373:2), a ca. 15 cm long brass chain fragment, part of a dress ornament (G = NM6373:5), and a fragment of a neck ring with saddle-shaped ends (H = NM6373:6).

Of these artefacts, the arm-rings in particular represent artefact designs favoured in the region of Finland at the time. Hence, typologically speaking, these artefacts appear to be regional products. The Westland cauldron, on the other hand, is a high-status elite object, which implies Scandinavian, possibly Norwegian contacts (Ørsnes, 1966: 105–106; Oestigaard, 1999: 357; Dahlin Hauken, 2005: 46–63). Westland cauldrons are associated with a Provincial Roman origin, and are named after the site in western Norway where they were first

Fig. 3. Levänluhta collection on display in the National Museum of Finland (Photo: E. Holmqvist).

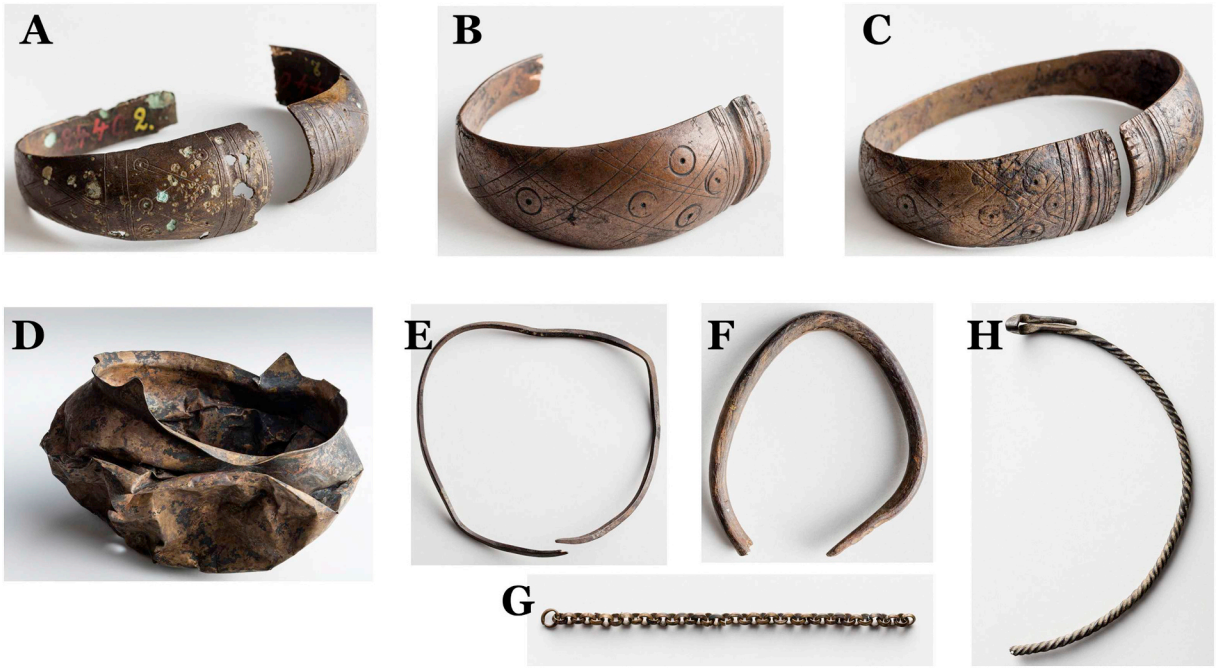

Fig. 4. Metal objects from the Levänluhta burial site analysed with ICP-MS and pXRF: two concave-convex arm-rings (A = NM2440:2, B = NM2440:3), a convex arm-ring (C = NM2440:5), a Westland type cauldron (D = NM2441:1), a neck-ring (E = NM2441:2), a small arm-ring made of a plain rod with a flattened end (F = NM6373:2), a chain fragment belonging to a dress ornament (G = NM6373:5), and a fragment of a neck ring with saddle-shaped ends (H = NM6373:6) (Photo: Finnish Heritage Agency).

identified. Apparently, the cauldrons were transported to Norway along a long-distance trade network (Oestigaard, 1999: 357) or exchanged as elite gifts between allied communities (Dahlin Hauken, 2005: 61–63). The majority of the Scandinavian cauldrons derive from burial contexts and date from the Early Roman Iron Age to the end of Migration period (Dahlin Hauken, 2005). The original form and type of the Levänluhta cauldron is difficult to distinguish due to its poor state of preservation and its damaged and, subsequently repaired profile (Wessman et al., 2018: Fig. 3). However, its triangular ears suggest that it is of a type produced sometime between 300 and 575 CE (Dahlin Hauken, 2005: 28, 45), although it is also differentiated from typical Westland cauldrons by its ears, which are missing holes for a handle.

Table 1
List of the analysed artefacts, the National Museum (NM) of Finland catalogue numbers, descriptions and typological dates.

Levänluhta sample	National Museum Catalogue no	Artefact description	Date
A	NM2440:2	Concave-convex arm-ring	550–800 CE
B	NM2440:3	Fragment of a concave-convex arm-ring	550–800 CE
C	NM2440:5	Convex arm-ring	550–800 CE
D	NM2441:1	Cauldron of Westland type	300–575 CE
E	NM2441:2	Neck-ring	550–800 CE
F	NM6373:2	Small arm-ring made of a plain rod with a flattened end	100 CE–
G	NM6373:5	Piece of chain (ca. 15 cm)	400 CE–
H	NM6373:6	Fragment of a neck-ring with saddle-shaped ends	550–800 CE

2.2. Sample preparation for the ICP-MS analysis

For the ICP-MS analysis, microdrilling was used as a sampling technique in order to minimise the damage to the artefact and allow the collection of pristine metal under the corrosion. The material collected from the surface layer was excluded from the analyses.

Approximately 7–10 mg of metal was drilled from each of the artefacts (see Craddock, 1976: 97 and Pernicka, 1986). A few milligrams of each sample of drilled metal was weighed for the concentration and Pb-isotope analyses. Powdered samples were washed with dilute HNO_3. Afterwards, samples were rinsed several times with deionised water with a resistivity of ≥18.2 MΩ·cm (Milli-Q). The washed samples were dissolved in a 1:1 6 N HCl −7 N HNO_3 acid mixture. After evaporation, the samples were re-dissolved in 1 ml of 1 N HBr. Using an ion-exchange chromatography, other elements were washed down with 2 × 1 ml of 1NHBr and 2 × 1 ml of 0.1 N HBr, and Pb was eluted using 2 ml of 6.2 N HCl. Each evaporated sample was diluted in 1 ml of 2% HNO_3. An aliquot from the sample solution was taken for the concentration analyses. The other aliquot was further on diluted to 30 ppb Pb in 1.5 ml of 2% HNO_3. The samples were then spiked with 20 ppb of Tl for mass bias correction. The Pb isotopic measurements were performed on Faraday detectors, using 6 blocks of 10 integrations of approximately 5 s each. Mercury (^{202}Hg) interferences on Pb were monitored during acquisition. The mass bias was corrected using an exponential law and a $^{205}Tl/^{203}Tl$ ratio of 2.3875. A standard reference material (NIST 981) was used to monitor the precision and accuracy of the measurements, every five samples, over the whole period of analysis. The obtained average accuracy is estimated to be below 1‰ (2σ) for $^{208}Pb/^{204}Pb$, 0.5‰ (2σ) $^{207}Pb/^{204}Pb$ and for $^{206}Pb/^{204}Pb$, compared with the certified value of Todt et al. (1996).

The lead isotopic ratios were measured via MC-ICP-MS at the Geological Survey of Finland, and the elemental analyses of the diluted samples were carried out at the ICP-MS laboratory of the Eurofins Labtium Oy.

2.3. Portable X-ray fluorescence spectrometry (pXRF)

The non-invasive pXRF data for the complete Levänluhta metal artefact assemblage was reported in Wessman et al. (2018: Table 1), and here in Table 2 for the objects selected for the ICP-MS analysis, to allow data comparison. The invasive sampling of the entire Levänluhta metal assemblage was not possible due to the high antiquarian value of the objects. The pXRF analysis was carried out at the National Museum exhibition hall (Fig. 5), with the aim to identify the alloy types of the Levänluhta metal artefacts; however, surface analyses of copper-based artefacts by pXRF are prone to surface corrosion and patina related problems, e.g. enrichment of certain elements, such as lead and iron (i.a. Lutz and Pernicka, 1996; Smith, 2012; Charalambous et al., 2014; Orfanou and Rehren, 2014; Dussubieux and Walder, 2015; Holmqvist, 2017). Therefore, in some cases it was not possible to identify the original alloy composition based on the surface data. The pXRF instrument was a University of Helsinki owned Bruker S1 Titan portable energy-dispersive X-ray fluorescence spectrometer, equipped with a silicon drift detector (SDD). The instrument has an Rh-target X-ray tube, and was operated using a copper alloy calibration mode, with a total acquisition time of 120 s. The quantitative results were calculated by the instrument's software. The reported result for each artefact is a mean value of 3–5 measurements conducted on seemingly corrosion free, even areas on unprepared artefact surfaces (spot size 8 mm).

3. Results and discussion

3.1. Geochemical characterisation via invasive ICP-MS vs non-invasive pXRF

The geochemical compositions of the artefacts measured by ICP-MS and unprepared artefact surfaces by pXRF are reported in Table 2 (normalised results; see Supplementary Information Table 1 for the ICP-MS measured geochemical raw data). The artefacts are made of different copper-based metals, with the assemblage including copper, bronze, and brass objects (Fig. 6). The apparent disparities between the pXRF and ICP-MS datasets most likely derive from surface issues affecting the pXRF data accuracy. The main components, copper (Cu) and tin (Sn), show fairly good correlation between the two datasets in most cases, but the pXRF results are clearly affected by surface processes, for example iron enrichment introduced by corrosion (Fig. 7a–c). Trace elements nickel (Ni), cobalt (Co), zinc (Zn), arsenic (As), silver (Ag), antimony (Sb), and lead (Pb) were detected by the ICP-MS.

The three typologically related concave-convex/convex arm-rings (samples A–C, NM2440: 2–3, 5) are made of different alloys (Table 2; Fig. 6). Sample A is made of bronze with ca. 6.6 wt% of tin (12.7 wt% of Sn was measured by pXRF, Fig. 7b) and sample B (NM2440:3) of copper (Cu 99.4 wt%). Sample C (NM2440:5) presents a high copper content by both methods, but the ICP-MS data indicated a higher Sn concentration at 2.6 wt% (instead of 0.4 wt% measured by non-invasive pXRF, Fig. 7b). Sample D, the cauldron (NM6373:5), was also made of bronze (Cu 91.2 wt%, Sn ca. 8 wt%).

The pXRF analysis of sample E (neck-ring, NM2441:2) was unsuccessful due to patina-related lead and iron enrichment (pXRF measured Pb 5.14 wt% and Fe 27.6 wt%, Fig. 7c) on the artefact surface, however, it was confirmed as to be made of bronze in the ICP-MS analysis. Sample F (NM6373:2), a small arm-ring made of a plain rod with a flattened end, is confirmed to be made of brass by the ICP-MS data, nevertheless, significant iron enrichment (Fe ca. 16 wt%) was detected by the pXRF on the artefact surface, compared to only 1.2 wt% of Fe measured by ICP-MS from the drilled sample (Fig. 7c). The chain fragment (sample G, NM6373:5) is also made of brass, with ca. 10 wt% of Zn. Sample H, a neck-ring fragment with saddle-shaped ends (NM6373:6), catalogued as a “silver neck-ring”, appears to be, in fact, made of bronze (Cu at ca. 92%; Sn at 4.6 wt% by the ICP-MS) and only coated with silver. Its surface analysis by pXRF revealed 45 wt% of Ag, a typical result for a silver-coated surface, whereas only traces of silver were detected by ICP-MS for the drilled sample (Fig. 8).

There are undeniable benefits to the non-invasive pXRF analysis; nevertheless, we must conclude that the utility of the pXRF data was compromised by serious surface effects, sample surface morphology,

Table 2
Selected relevant geochemical concentrations measured via ICP-MS and non-invasive pXRF after normalization of all data. Concentrations at or below the limit of detection are excluded.

ID	Catalogue no	Fe		Ni		Co		Cu		Zn		As		Ag		Sn		Sb		Pb	
		ICP-MS	PXRF	ICP-MS	PXRF	ICP-MS	PXRF	ICP-MS	PXRF	ICP-MS	PXRF	ICP-MS	PXRF	ICP-MS	PXRF	ICP-MS	PXRF	ICP-MS	PXRF	ICP-MS	PXRF
		%		%		%		%		%		%		%		%		%		%	
A	NM2440:2	0.050	3.963	0.013	0.014	0.001	bdl	93.147	82.856	0.015	0.034	0.018	0.062	0.004	0.055	6.558	12.658	0.047	0.183	0.045	0.174
B	NM2440:3	0.011	0.048	0.084	0.047	0.010	0.016	99.419	99.069	0.140	0.235	0.079	0.105	0.004	0.019	0.232	0.376	0.000	bdl	0.021	0.057
C	NM2440:5	0.010	0.013	0.072	0.032	0.009	0.014	97.131	99.148	0.202	0.223	0.068	0.083	bdl	0.020	2.469	0.399	0.000	bdl	0.019	0.044
D	NM2441:1	0.075	0.212	0.021	0.014	0.001	0.010	91.179	88.897	0.141	bdl	0.026	0.023	0.014	0.062	7.895	10.216	0.050	0.171	0.597	0.391
E	NM2441:2	0.699	27.637	0.043	0.014	0.002	bdl	82.124	47.458	3.188	2.378	0.039	0.451	0.058	0.129	13.466	16.583	0.058	0.216	0.299	5.133
F	NM6373:2	1.206	15.973	0.175	0.014	0.006	bdl	70.815	61.442	24.810	18.456	0.223	0.400	0.004	0.112	0.075	0.075	0.076	0.033	0.135	3.519
G	NM6373:5	0.036	0.179	0.007	0.014	0.000	0.008	86.751	84.417	10.096	13.614	0.045	0.084	0.001	0.039	0.026	0.189	0.012	bdl	2.333	1.453
H	NM6373:6	0.139	7.936	0.062	0.014	0.001	bdl	91.857	43.748	1.687	bdl	0.070	0.033	0.004	44.880	4.559	2.666	0.049	bdl	0.614	0.690

and especially patina-related enrichment, not always visible to the naked eye. The pXRF analysis did not always provide secure alloy type determination, and the data quality was insufficient to evaluate the minor and trace elemental patterns of the artefacts. Compared to the ICP-MS data, the pXRF performed most accurately with flat and corrosion-free samples with a high Cu content (e.g., concave-convex arm-rings NM NM2440:3, 5; chain NM6373:5). The surface effects are particularly evident in the pXRF results of samples E and F (neck-ring NM2441:2; arm-ring NM6373:2), which give unusually high Fe (15–27 wt%) and Pb (3.5–5 wt%) concentrations due to the surface corrosion enrichment of these elements (see Orfanou and Rehren, 2014).

The lead contents of seven of the eight objects measured by ICP-MS are below 1% at 0.019–0.6 wt%, which indicates that there are no concerns of added Pb in the alloy endangering the lead isotope tracing of the copper sources. However, sample G, the chain fragment, presents a higher Pb value at 2.3 wt% (Table 2), suggesting intentional alloying with lead, or mixing with Pb-rich tin in this case (see Liversage, 2000; Ling et al., 2014: 117; Melheim et al., 2018: 103).

Geochemical data alone is not adequate for metal source identification, but the concentrations of As, Sb, Ag and Ni (Figs. 8 and 9) can be indicative of copper ore deposit characteristics. However, it can be difficult to acquire comparative geochemical ore data and the artefact concentrations may altered by e.g. remelting of the metals (i.a., Pernicka, 1999: 169; Bray and Pollard, 2012: 854; Pernicka et al., 2016: 39). In the case of the Levänluhta samples, their limited number and geochemical heterogeneity prevents any meaningful comparison with the original ore composition for provenancing purposes. Three of the analysed objects (samples B, C, and G; NM2440:3–5 and NM6373:5) show very low As and Sb levels (Fig. 9), whereas samples A, D, E, and H (NM2440:2, 2441:1–2 and 6373:6) are fairly similar with slightly higher Sb concentrations. There are no obvious typo-chronological explanations for these differences, as all of the objects dated to ca. 300–800 CE.

Interestingly, the arsenic and antimony levels of the Levänluhta artefacts are generally lower (0.02–0.22%, and 0.01–0.07%, respectively, Fig. 9) compared to values reported by Ling et al. (2013) for Swedish Bronze Age bronze objects. The reduced As and Sb levels in these Iron Age objects might be indicative of the fact that the metals were reused several times (even since the Bronze Age?), resulting in a loss of As through oxidization in re-melting (see, e.g. Bray et al., 2015; Pollard and Bray, 2015: 997). Furthermore, the typo-chronologically oldest artefact among the analysed eight artefacts, sample F, a small arm-ring dated to ca. 100 CE and onwards – hence centuries older than the other analysed objects dated to 400–800 CE – presents the highest Sb and As concentrations in the assemblage (Fig. 9). Although the absolute differences between the measured Sb and As values are not substantial, the higher concentrations might be indicative that the metals used in the manufacture of sample F were re-melted fewer times compared to the materials used in the later object forms. Based on the geochemical patterns, it appears that metal recycling played a role in the material acquisition for most of the analysed Levänluhta objects.

3.2. Tracing the metal source using Pb isotopes

Lead isotopic ratios of the eight Levänluhta metal objects (samples A–H) were determined via multi-collector-ICP-MS, in order to trace the copper ore extracted to produce the metal for the manufacture of the artefacts. The majority of the samples (B, C, D, E, F, and H, Table 3, Fig. 10) display relatively homogeneous lead isotopic compositions with $^{208}Pb/^{206}Pb$ ratios approx. at 2.077–2.085, $^{207}Pb/^{206}Pb$ at 0.831–0.846 and $^{206}Pb/^{204}Pb$ at 18.495–18.673. Samples A (arm-ring, NM2440:2) and G (chain, NM6373:5) are characterized by higher $^{208}Pb/^{206}Pb$ ratios (at ca. 2.099) and $^{208}Pb/^{204}Pb$ ratios (at 38.283 and 39.418, respectively), and $^{206}Pb/^{204}Pb$ ratios (18.245; 18.783) that fall outside the range presented by the rest of the samples. Sample G is

Fig. 5. Non-invasive pXRF analysis of a bronze cauldron recovered in Levänluhta at the National Museum of Finland exhibition hall in Helsinki (Photo E. Holmqvist).

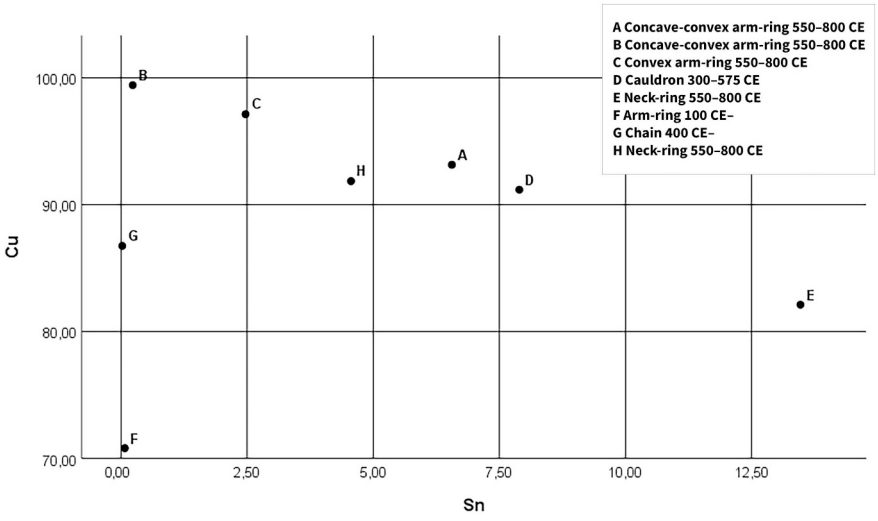

Fig. 6. Copper (Cu) vs tin (Sn) concentrations measured by ICP-MS for the eight Levänluhta objects.

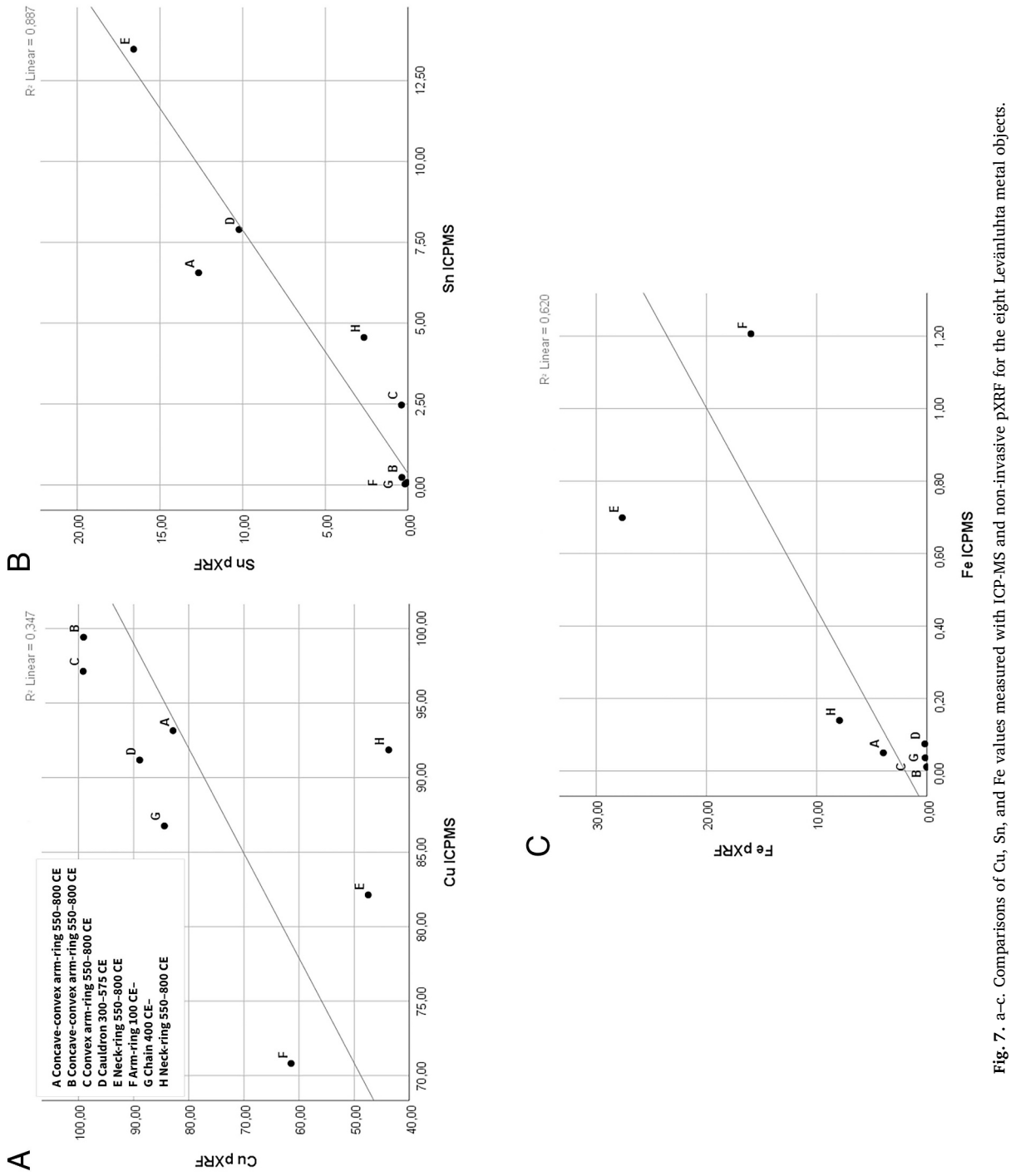

Fig. 7. a–c. Comparisons of Cu, Sn, and Fe values measured with ICP-MS and non-invasive pXRF for the eight Levänluhta metal objects.

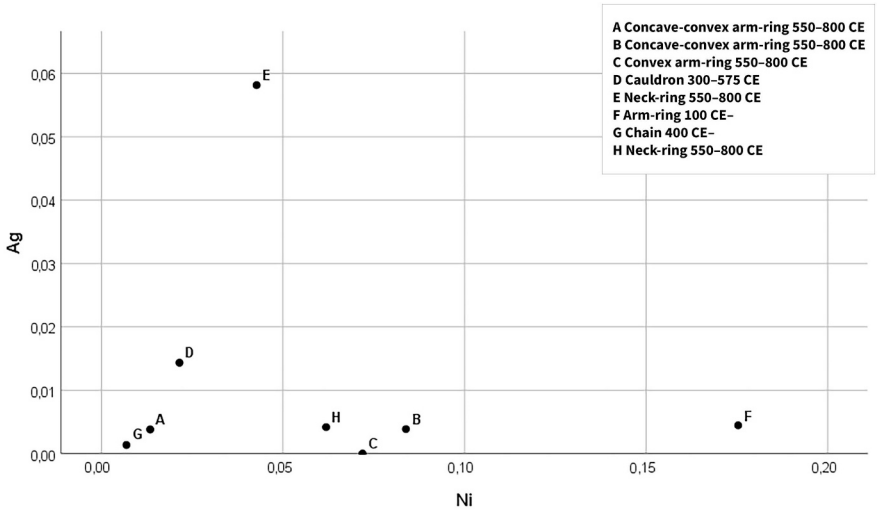

Fig. 8. Silver (Ag) vs nickel (Ni) concentrations measured by ICP-MS for the eight Levänluhta metal objects.

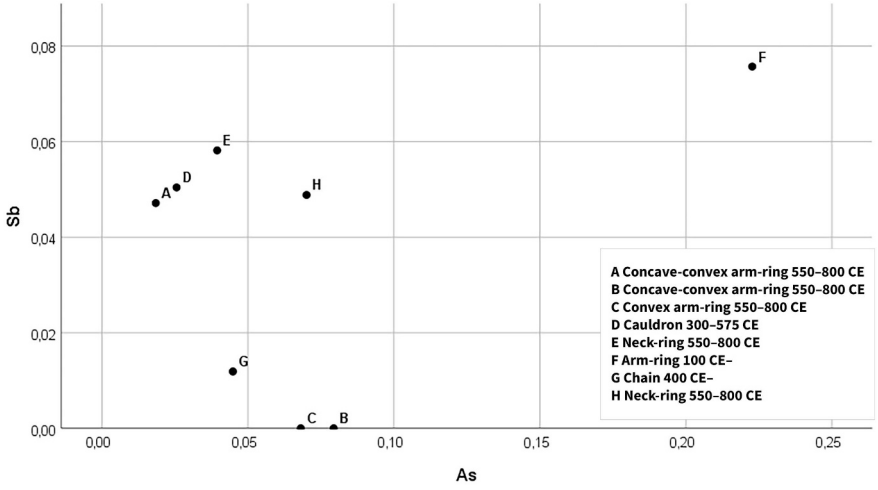

Fig. 9. Antimony (Sb) vs arsenic (As) concentrations measured by ICP-MS for the eight Levänluhta objects.

Table 3
Pb isotope compositions for the eight Levänluhta metal artefacts.

Levänluhta sample	Catalogue no	Object form	Date	$^{207}Pb/^{206}Pb$	$^{208}Pb/^{206}Pb$	$^{208}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{206}Pb/^{204}Pb$
A	NM2440:2	Concave-convex arm-ring	550–800 CE	0.857	2.098	38.283	15.639	18.245
B	NM2440:3	Concave-convex arm-ring	550–800 CE	0.840	2.080	38.782	15.671	18.647
C	NM2440:5	Convex arm-ring	550–800 CE	0.840	2.080	38.781	15.668	18.649
D	NM2441:1	Cauldron	300–575 CE	0.846	2.085	38.560	15.649	18.495
E	NM2441:2	Neck-ring	550–800 CE	0.844	2.082	38.634	15.652	18.553
F	NM 6373:2	Arm-ring	100 CE–	0.841	2.079	38.701	15.665	18.619
G	NM6373:5	Chain	400 CE–	0.834	2.099	39.419	15.665	18.783
H	NM6373:6	Neck-ring	550–800 CE	0.839	2.077	38.784	15.668	18.673

typologically a rare object, and also geochemically different compared to the other samples, with an ICP-MS measured Pb concentration of ca. 2.3 wt%. A lead content this high could indicate that lead was added to the original copper alloy in the manufacturing process, also creating a mixed Pb isotopic fingerprint for the artefact at the same time. For this reason, sample G was excluded from the statistical LI data comparisons.

In order to trace the copper sources, we first compared the LI data of the artefacts to Finnish and Swedish ores, i.e., the local geology nearby the Levänluhta site. As a result, it is clear that the Levänluhta samples present us with lead isotopic ratios (Table 3, Fig. 11a–b) inconsistent with the values reported by Vaasjoki (1981) and Billström et al. (1997) for the Finnish and Swedish deposits (e.g. the Archean basement, Svecofennian rocks, sulfride ore belt, the granite areas, Greenstone or Porphyry groups), thus the possibility of the metals originating from ores known from Finland or Sweden can be excluded. Due to the lack of comparative LI data from the region of modern Russia, it is currently impossible to exclude the possibility of eastern ore extraction being the source of the metal for these artefacts. However, the bedrock in the north-western Russia is similar to that of the Finnish region, i.e., much older than implied by the Pb isotope data of the analysed artefacts, and thus could not generate LI compositions similar to the Levänluhta objects, speaking against at least the north-western Russia as the origin of the metals (see Vaasjoki et al., 2005: 4–5 for the geological map of the Fennoscandian Shield and its explanation).

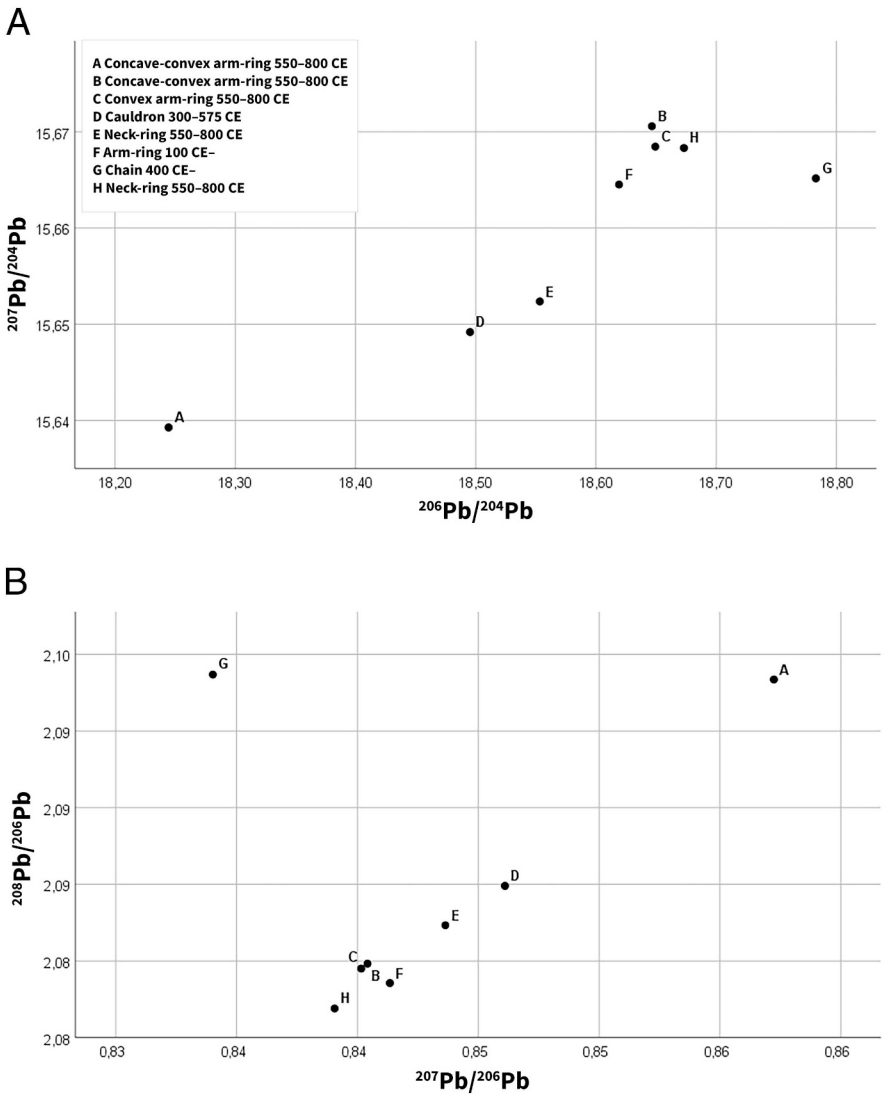

Fig. 10. a–b. Lead isotope ratios of the different artefact types from the Levänluhta site.

In light of the comparative data available, and in order to examine possible European sources for the Levänluhta metals, we compared the $^{208}Pb/^{206}Pb$, $^{207}Pb/^{206}Pb$ and $^{206}Pb/^{204}Pb$ isotopic composition of the Levänluhta artefacts with published LI data of different ores of known localities elsewhere in Europe. We followed the procedure suggested by Stos-Gale and Gale (2009) in order to identify possible sources. First, we searched published databases for potential source areas (i.a. OXALID; Stos-Gale and Gale, 1994, 2009; Stos-Gale et al., 1997; Begemann et al., 2001; Bielicki and Tischendorf, 1991; Bode et al., 2009; Chernyshev et al., 2007; Durali-Mueller et al., 2007; Höppner et al., 2005; Klein et al., 2010; Marcoux et al., 2002; Neubauer et al., 2005; Santos Zaldeuegui et al., 2004; Tornos and Chiaradia, 2004; Yener et al., 1991). Next, we tested the normalised Euclidian distance (SPSS software) of ca. 2000 potential ore samples against the Levänluhta LI data, searching for ore samples (Cu and Pb ore data only) with all three lead isotope ratios very similar (within the ± 0.1% of the error) to each individual Levänluhta sample. Finally, these ore data were plotted together with the artefact data in $^{208}Pb/^{206}Pb$ vs $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{206}Pb$ vs $^{206}Pb/^{204}Pb$ diagrams (sample G was excluded due to its suspected mixed isotopic signature).

Based on our data analysis, the ores that appear to show the most similarity with the Levänluhta metals (samples A–F, H) are located in southern Europe. It is apparent that the Levänluhta objects are not consistent with any single important copper/lead ore deposit. Considering the vast quantities of LI data published in the recent years, our data comparison may not be entirely exhaustive, and our interpretations are based on the currently existing evidence available to us. The ores that present the closest comparanda for the Levänluhta sample set, i.e., fall within a ± 0.1% analytical error from the three LI ratios presented by the sampled objects, are located in Spain (particularly for sample A), Bulgaria, Greece (Lavrion) and Cyprus (Table 4; Fig. 12). The Iberian peninsula, Bulgaria, the Lavrion region in Greece and Cyprus are known from their copper/lead mines exploited in the Bronze Age, the Roman period and later (see, e.g., Dušanić, 1977; Edmondson, 1989; Gale et al., 2003; Hirt, 2010; Huelga-Suarez et al., 2012; Wilson, 2007). Thus, they can be considered as potential source areas for the Levänluhta metals. However, the results should be treated cautiously, because a) the question of possible eastern sources cannot be excluded due to the lack of comparative data, and b) the relatively low As and Sb levels of the Levänluhta sample set pose the question of metal recycling, and mixing, in the manufacture process of these artefacts.

With regard to the most commonly reported ore-related elements As, Sb, Ag and Ni, the Levänluhta objects show concentrations below 0.1 wt% in the ICP-MS analysis (Table 2; see Bray et al., 2015), most likely deriving from the continuous reuse of the metals, largely preventing geochemical comparisons between the analysed objects and potential sources. An exception is made by the typologically oldest sample F with As and Ni values at 0.22 wt% and 0.18 wt%, respectively.

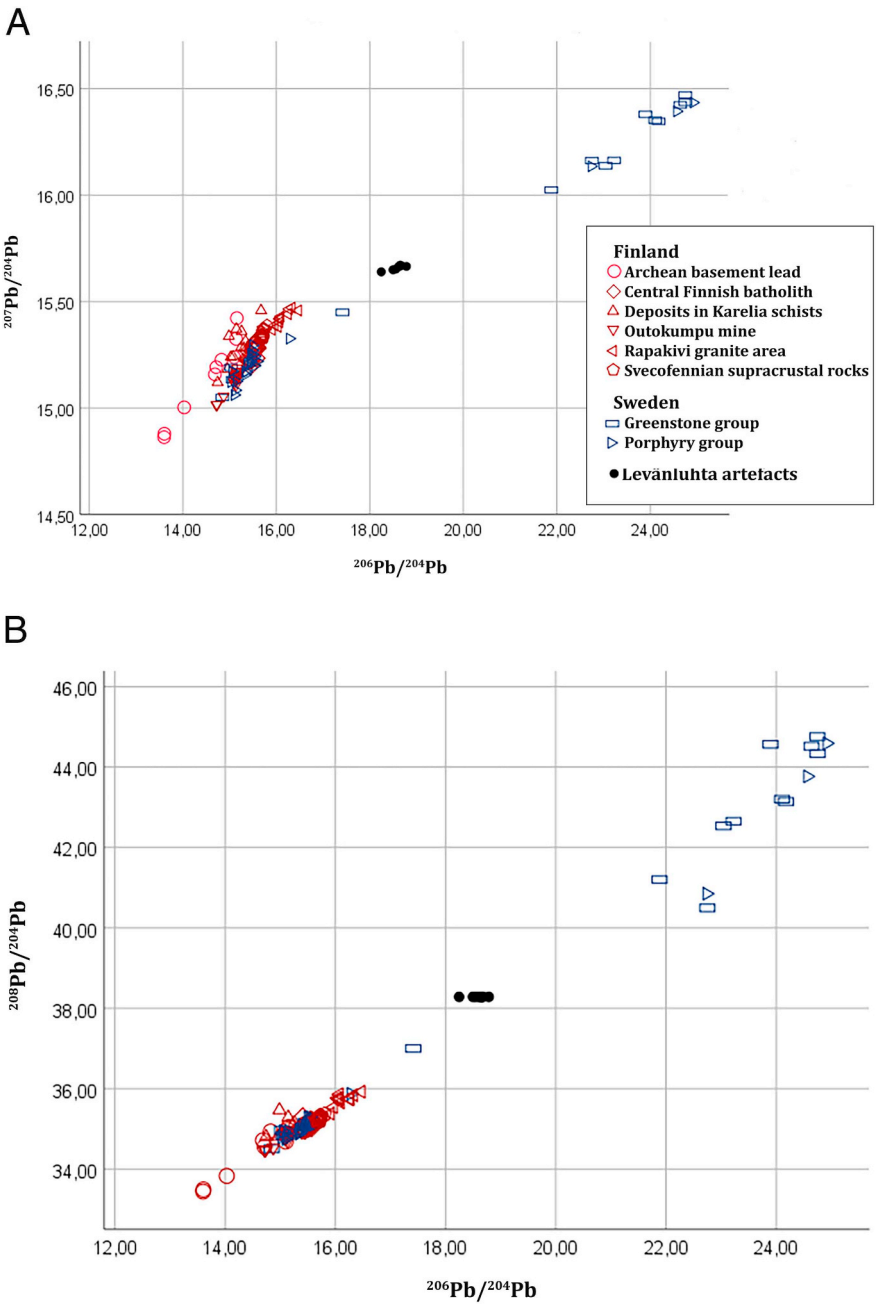

Fig. 11. a–b. Pb isotope ratios of the Levänluhta artefacts plotted against data from Finland and Sweden (Vaasjoki, 1981; Billström et al., 1997).

Considering the possible scenario of mixing metals extracted from different sources, the LI signature of the Levänluhta objects might, in fact, represent not the LI signature of an individual ore or region, but instead that of "mixing-lines" between different major copper (or lead) mining regions exploited perhaps in the Roman period or pre-Viking period Europe, or earlier. The cauldron, sample D, did not match German or Austrian ore samples in our data analyses, with Bulgarian ore data dominating the comparative data set (OXALID; Bielicki and Tischendorf, 1991; Bode et al., 2009; Durali-Mueller et al., 2007, Gale et al., 2003).

In light of this evidence, yet bearing in mind the possible eastern sources and metal mixing, it appears reasonable to suggest that there is a possibility that the metals used to manufacture the Levänluhta objects (apart from sample G with its inconclusive LI profile) were transported all the way through Europe and across the Baltic Sea to Finland, linking the region of modern western Finland to the broad pan-European network of metal transport. Compared to the LI signatures of the Swedish Bronze Age bronzes reported by Ling et al. (2014, Table 3), primarily associated with Central and Southern European sources, the Levänluhta artefacts show a more homogeneous LI profile, dissimilar to those of the majority of the Swedish bronzes. The Swedish bronzes that show the most similarity with the Levänluhta finds date to the BA periods I and II (EBA, 1700–1500 cal. CE, and MBA 1500–1300 cal. CE, respectively, Ling et al., 2014: 108). It is perhaps noteworthy that some of them were found in Uppland, Öland and Södermanland in eastern Sweden near the Baltic coast. Thus, although highly speculative, these could have been possible transit points for the metals *en route* across the Baltic and to Levänluhta on the Finnish coast, prior to their recycling (and mixing) in the Iron Age, if we accept that at least for some of the Levänluhta objects the metals link to the BA copper exchange.

Table 4
Mean values of the ore samples from the different regions with the closest Euclidian distance from the LI data of the Levänluhta objects (excluding sample G). All ore data given here are published in OXALID. Details of individual ore samples are given in the SI Table 2.

Levänluhta	Normalised Euclidian distance	Country	Ore sample	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{206}Pb/^{204}Pb$
Sample A				2.098	0.857	18.245
	0.016–0.147	Spain	μ (n = 11)	2.102	0.858	18.254
			σ	0.007	0.001	0.054
	0.054–0.060	Greece (Lavrion)	μ (n = 3)	2.101	0.857	18.258
			σ	0.006	0.001	0.044
Sample B				2.080	0.840	18.647
	0.008–0.138	Bulgaria	μ (n = 6)	2.073	0.840	18.603
			σ	0.003	0.001	0.045
	0.022–0.149	Cyprus	μ (n = 12)	2.076	0.840	18.551
			σ	0.001	0.001	0.042
	0.001–0.149	Greece (Lavrion)	μ (n = 40)	2.080	0.840	18.661
			σ	0.004	0.001	0.073
	0.024–0.122	Spain	μ (n = 3)	2.084	0.840	18.720
			σ	0.007	0.001	0.079
Sample C				2.080	0.840	18.649
	0.003–0.042	Bulgaria	μ (n = 5)	2.072	0.840	18.621
			σ	0.001	0.000	0.006
	0.014–0.133	Cyprus	μ (n = 7)	2.076	0.840	18.569
			σ	0.002	0.001	0.036
	0.013–0.148	Greece (Lavrion)	μ (n = 51)	2.079	0.839	18.681
			σ	0.003	0.001	0.068
	0.043–0.133	Spain	μ (n = 4)	2.081	0.839	18.733
			σ	0.008	0.001	0.070
Sample D				2.085	0.846	18.495
	0.014–0.139	Bulgaria	μ (n = 13)	2.083	0.845	18.487
			σ	0.003	0.001	0.025
	0.016–0.119	Greece (Lavrion)	μ (n = 6)	2.084	0.846	18.490
			σ	0.003	0.001	0.023
	0.020–0.119	Spain	μ (n = 4)	2.085	0.847	18.460
			σ	0.002	0.001	0.092
Sample E				2.082	0.844	18.553
	0.003–0.122	Bulgaria	μ (n = 20)	2.082	0.844	18.503
			σ	0.002	0.001	0.026
	0.012–0.111	Greece (Lavrion)	μ (n = 14)	2.083	0.843	18.557
			σ	0.005	0.001	0.065
	0.055	Spain	μ (n = 1)	2.079	0.843	18.576
Sample F				2.079	0.841	18.619
	0.070–0.138	Bulgaria	μ (n = 9)	2.076	0.841	18.586
			σ	0.005	0.001	0.044
	0.018–0.148	Greece (Lavrion)	μ (n = 32)	2.079	0.841	18.606
			σ	0.005	0.001	0.075
	0.044–0.136	Spain	μ (n = 3)	2.081	0.841	18.660
			σ	0.006	0.002	0.103
Sample H				2.077	0.839	18.673
	0.038–0.149	Bulgaria	μ (n = 29)	2.071	0.838	18.688
			σ	0.003	0.001	0.038
	0.002–0.147	Greece (Lavrion)	μ (n = 80)	2.078	0.839	18.696
			σ	0.003	0.001	0.061
	0.001–0.124	Spain	μ (n = 7)	2.081	0.839	18.734
			σ	0.006	0.001	0.067

4. Conclusions

This article presents the geochemical and Pb isotopic (LI) results of eight Iron Age copper-based alloy artefacts recovered at the Levänluhta water burial in western Finland. Non-invasive pXRF spectrometry and MC-ICP-MS analysis on prepared samples were carried out to allow alloy type identification and tracing of the metal source. The results show that the objects were manufactured of copper, brass, and bronze, and that different types of alloys were used to manufacture typologically related objects, e.g. arm-rings. The utilisation of different alloys in the artefacts' manufacture may simply be related to the availability of materials at the given time of production, but the compositional variation can also be indicative of separate lines of production, e.g. different manufacturers or workshops, different times of manufacture or material acquisition, or different patterns of mixing and remelting of the metals.

There were significant inconsistences between the pXRF and ICP-MS datasets due to surface effects (uneven surfaces, corrosion) affecting the pXRF data accuracy. Hence, the non-invasive approach was not always successful in alloy type determination. For distinguishing coating composition from the artefact material, on the other hand, the two methods were complementary, allowing both the alloy of the object and the coating metal to be determined with minimal damage to the object.

Despite the typological and geochemical diversity among the sampled artefacts, the majority of the Levänluhta objects form a relatively homogeneous group based on their LI ratios. One artefact, the chain fragment, was a clear outlier in terms of both LI and geochemical composition, interpreted to derive from added lead, and this prevented further examination of its provenance based on LI data. The stylistic attributes of the objects suggest that the Levänluhta metals were cast in regional workshops in the region of modern Finland, although potential workshop sites remain to be identified. However, it is apparent from the LI data that the copper utilised to manufacture these objects does not derive from Finnish or Scandinavian copper ores.

Compared to available published LI data on European copper ores from numerous locations, it appears that our LI and geochemical results

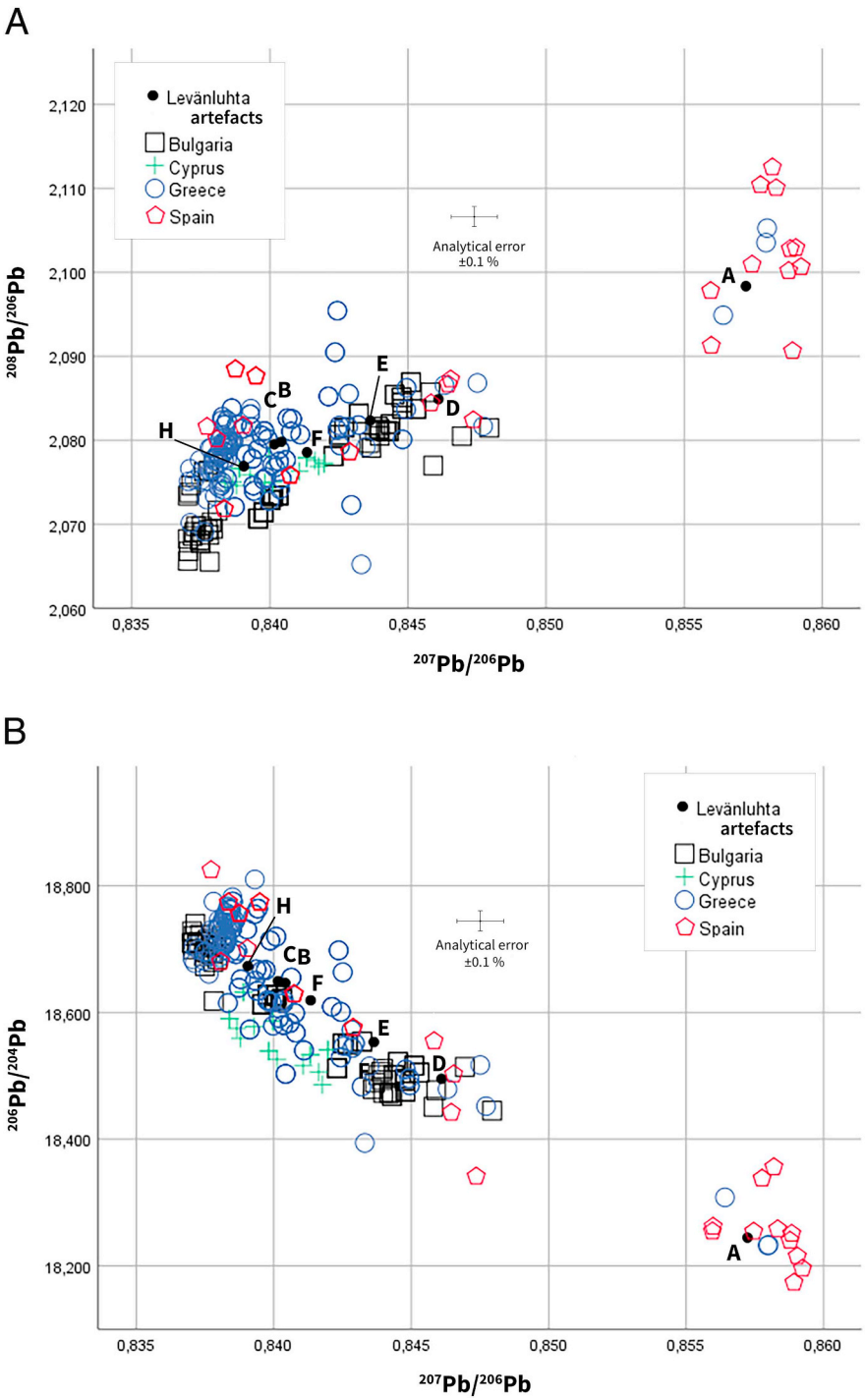

Fig. 12. a–b. Lead isotope ratios of the ore samples with the closest Euclidian distance from each of the Levänluhta artefacts (excluding sample G). All ore data published in OXALID, see Table 4 and Supplementary Information Table 2 for details.

for the Levänluhta objects show the most similarity with southern European copper ores, and the regions of modern Spain, Bulgaria, Cyprus and Greece in particular (see, e.g., Müller and Pernicka, 2009; Huelga-Suarez et al., 2012, 2014; Farci et al., 2017; Gale and Stos-Gale, 2000). Hence, these areas may represent potential source areas for the Levänluhta metals.

When interpreting the LI data from this geographical context, it is important to remember that the Bronze and Iron Age communities of Finland also had links with the east (i.e. modern Russia), from where copper ore databases are not openly available, preventing data comparison with the possible eastern source areas. The available comparative data is thus currently biased towards Europe, whereas culturally equally relevant links towards the east, and other regions lacking published LI data (see Pernicka et al., 2016: 38–42), cannot even be considered. However, based on the geological data available, at least north-western Russia can be excluded as a potential source area for the Levänluhta metals.

The chain fragment (sample G) which had LI signature unparalleled in the comparative datasets, could hypothetically be an eastern import to the Finnish region; or alternatively, its composite patterns could be a result of material recycling and metal mixing, creating a combination of metals from different sources over time. Furthermore, the Levänluhta

objects in general display relatively low As and Sb concentrations, a characteristic that may derive from material recycling over the centuries. During the Iron Age, local iron supply replaced imported bronze as the main material for weapons and tools in Scandinavia (see Ling et al., 2018: 492), thus the import of copper alloys likely also diminished in Finland, and any that were used were probably primarely from previously acquired metals that were recycled. Bray et al. (2015: 203) write that the life histories of unwanted artefacts can easily continue for several centuries, through different cultural phases and over long distances, as 'scrap metal', and the recycling and blending of materials from different sources may result in artefact materials "no longer directly recognisable chemically or isotopically as copper from the original mine" in provenance analysis. The practice of recycling Bronze Age metals to produce artefacts in later periods is probably one explanation for the limited amount of Bronze Age artefacts recovered in the region of Finland. The low As levels of the objects compared to bronzes found in earlier contexts in Scandinavia also supports the interpretation that the objects are products of metal recycling.

Therefore, although it is clear from our data that the Levänluhta objects were, indeed, manufactured of foreign metals, it is impossible to assess in detail the exact arrival routes, times, or formats of the metals found in Iron Age contexts in Finland. It is probable that at least some of these findings are the material reminiscences of objects or raw materials brought to Finland already during the Bronze Age. Accordingly, the suspected recycling means that we cannot ascertain the cultural phase during which the metals arrived in the Finnish region, or link the source metal with any social or technological context (see Pollard et al., 2014 for discussion). Taking into account that Levänluhta is located on the Ostrobothnian coast, one feasible option is that the metals arrived in the Finnish region via Sweden, which is known for its BA links with the southern European copper exchange (e.g., Ling et al., 2013, 2014), but the metals may have equally well arrived via, for instance, the Baltic region, or from the east (modern Russia), either during the BA or in the IA period. There is a possibility that, at least in the domestic arm-rings and neck-rings, we are in fact seeing recycled Bronze Age materials (see, e.g. Brandherm, 2018; Bray and Pollard, 2012; Bray et al., 2015), recast to make Iron Age jewellery types.

The cauldron (sample D), on the other hand, probably had a completely different arrival route to Levänluhta, in another chronological context, unrelated to the BA or IA bronze supplies in this region in general. It arrived in northern Europe as a ready-made (elite) object, and was buried as such in Levänluhta, relatively soon after its manufacture somewhere in the frontier provinces of the Roman Empire (of Bulgarian copper as indicated by its LI data?). Furthermore, it may have arrived to Finland via Norway, thus not by the "fastest route".

Accordingly, and again bearing mind the need for caution in the data interpretation discussed above, if we accept that copper mined in southern Europe was transported to western Finland sometime in the Bronze/Iron Ages, these results would link Finland to the well-documented pan-European network of copper circulation, extending into Scandinavia (e.g. Kristiansen, 2017; Earle et al., 2015; Ling et al., 2014; Pernicka et al., 2016; Shennan, 1999; Ling et al., 2018; Melheim et al., 2018), and which apparently also crossed another sea, the Baltic, to reach the coast of western Finland. Hence, the results confirmed our hypothesis that instead of Finnish or Scandinavian copper and tin ores, the metals used to manufacture the Levänluhta artefacts were transported via long-distance trade routes, from southern Europe to Finland, in accordance with findings by Ling et al. (2013, 2014) for Swedish bronzes.

As a final note, although the copper has an international profile, the individuals buried in Levänluhta during the Iron Age, who most likely used these objects and were buried with them, probably acquired them from local workshops (where they were manufactured from recycled metals), without any international links either on a personal or communal level. It is clear, nevertheless, that the metals travelled long-distances to the Finnish coast, through several hands, and probably over a long period of time, again providing material evidence (see, e.g. Holmqvist et al., 2018) that the Prehistoric Finnish region had active and far-reaching networks across the Baltic Sea and beyond.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jasrep.2019.05.019.

Declaration of Competing Interest

None.

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