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Lead isotopes in the context of the provenance of copper alloys and mutability processes in Lithuania from the second half of the 1st century to the 13th century AD

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Keywords

lead isotope ratios, mutability, copper groups, coper alloy provenance, the second half of the 1st century to the 13th century AD, exchange networks, Lithuania

Abstract

This paper presents the results of analysis of lead isotope ratios (208Pb/204Pb, 207Pb/204Pb, 206Pb/204Pb, 208Pb/206Pb, 207Pb/206Pb) in copper alloys combined with chemical composition studies of archaeological artefacts by inductively coupled plasma mass spectrometry (ICP-MS). The study covers a total of 208 samples collected from 55 sites spread all over Lithuania. The chronological range of the study encompasses the period from the second half of the 1st century AD to the 13th century AD. The repeated recycling of copper alloys, the addition of scrap materials with varying compositions, and the mixing of lead from different geological ore deposits naturally alter the original chemical composition of the alloy. The continuous recycling and alteration of materials pose challenges in identifying the original connections between the regions of origin of copper alloy ores in southern Eurasia and the users of these raw materials in the eastern Baltic Sea region. Together with lead isotope ratio analysis, investigation of copper alloy types, copper groups, and metal working technological development fundamentally changes the idea of a linear exchange of non-ferrous metals. The analysis carried out in this research has enabled the identification of the provenance and dissemination of non-ferrous metal raw materials (alloys and scrap metal) as part of the European exchange network at local (the present territory of Lithuania), regional (the eastern Baltic region), and trans-European levels.

Introduction

The communities of the eastern Baltic Sea region did not, in prehistory as today, have their own ore deposits to produce copper alloys and artefacts. Therefore, all these materials, like any other imports, could only be obtained by exchange. The availability of raw metals depended directly on the intensity of the flow of imports, as well as on their quantity and quality. Moreover, the flow of raw metals and

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other imports reached the different regions around the Baltic Sea differently.

Nearly seventy years ago, the first analyses of lead and silver isotopes were conducted, and this method was soon extended to the copper-based alloys (Brill and Wampler 1965; Pernicka 2014). However, a real breakthrough was made in 1982, when it was suggested that it is possible to extract lead from copper-based ancient metals to identify the provenance of the non-ferrous metal alloys (Gale and Stos-Gale 1982; Stos-Gale 2019, pp. 85–118; Pollard et al. 2018, pp. 146–149).[1] Stable isotope analysis has fundamentally transformed our understanding of the past. It is now possible to have a more realistic comprehension of how exchange processes operated and how the routes were interconnected during different periods of antiquity. Undoubtedly, the determination of lead isotope ratios in copper ores as the imprints of their geographical origin and the application of the method to the analysis of ancient metals and archaeological artefacts have brought about a major positive change in archaeometallurgical research. From the 1970s, lead isotope ratios were analysed by Thermal Ionisation Mass Spectrometry (TIMS) as a standard method. With TIMS, the lead isotopes ratios are measured using the exactly specified chemistry of lead separation and deposition, and the methodology of running the mass spectrometer and the raw data are corrected by comparisons with lead isotopic values using a standard reference material (NIST SRM 981). This methodology allows the measurement of lead isotope ratio compositions with very high accuracy (usually better than 0.1% for annual runs of the NIST SRM 981 standard) and very good reproducibility of measurements taken in different laboratories (Durali-Müller 2005, Figure 2.1; Stos-Gale 2019, pp. 90–91). Further advancements in analytical methods led to the development of laser ablation techniques combined with ionisation of lead in a plasma source, using multiple Faraday cups as ion collectors in multicollector inductively coupled plasma mass spectrometers (MC-ICP-MS). This approach allows analysis of much smaller samples with very high overall accuracy, commonly achieving better than 0.01% precision for all ratios within a single run. The main advantage of MC-ICP-MS relative to TIMS instruments is the efficient ionisation of most elements and the operation of the instrument at steady-state, which allows full control of mass fractionation (Durali-Müller 2005, pp. 15–18, Figs. 2.1–2.5). The method requires a sample of the metal core of the find (1–20 mg or 0.001–0.02 grams), but is debated in terms of precision and reliability (e.g. Scott and Gauthier 1996; White et al. 2000; Woodhead 2002; Baker at al. 2006; Pernicka 2014; Stos-Gale 2019, pp. 90–91). We have evaluated the relative advantages of the MC-ICP-MS and TIMS methods for analysing lead isotope ratios and tested the ICP-MS method. Using the same samples, we also examined the chemical composition of archaeological finds and the lead isotope ratios of copper alloys. The lead isotopic ratios we obtained are consistent with those produced by other methods.

There are four terrestrial stable isotopes of lead (^{204}Pb, ^{206}Pb, ^{207}Pb and ^{208}Pb), three of which are radiogenic products of radioactive decay: ^{208}Pb is produced by the decay of ^{232}Th (thorium), ^{207}Pb by that of ^{235}U (uranium), and ^{206}Pb by that of ^{238}U. ^{204}Pb is not radiogenic nor the product of any radioactive decay but is an Earth or primordial element belonging to the proto-solar forming system from the creation of the Earth 4.57 billion years ago to modern times (Todt et al. 1996; Durali-Müller 2005, p. 31; Pernicka 2014; Pollard et al. 2018, p.145; Stos-Gale 2019, p. 93). Because radioactive decay is irreversible, the Pb isotope ratios in any system containing uranium and thorium increase consistently over time. Therefore, the ratios of four radiogenic lead isotopes to the primordial ^{204}Pb serve as an immediate indicator of the mineral's age. Measurements of the lead isotope compositions are therefore always expressed as ratios (Krause 2003; Niederschlag et al. 2003; Pernicka 2014; Stos-Gale 2019, p. 90; Gale and Stos-Gale 2000; García de Madinabeitia et al. 2021).

Geologists use lead isotope data from ore deposits to estimate the Earth's age and geological age of the various metalliferous deposits. However, using lead isotopes to characterise metal deposits is challenging because very few lead deposits are truly conformable, and deposits with highly radiogenic lead isotope ratios are relatively rare and often isotopically heterogeneous (Pollard et al. 2018, p. 149). Despite these challenges, lead isotope ratios typically remain consistent within a geological ore body and unchanged through the smelting and refining, making them reliable for archaeometallurgy studies. The ratios of ^{208}Pb/^{204}Pb with the most variable values are most commonly between 38 and 40; for ^{207}Pb/^{204}Pb most commonly between 15 and16; for ^{206}Pb/^{204}Pb most commonly between 17 and 19; for ^{208}Pb/^{206}Pb most commonly between 2.05 and 2.12; and ^{207}Pb/^{206}Pb most commonly between 0.825 and 0.837 (Stos-Gale 2019, pp. 91–92). Besides this, the accepted average natural abundances are approximate, with a large variation for all four isotopes (^{204}Pb = 1.4%, 2^{06}Pb = 24.1%, ^{207}Pb = 22.1%, and ^{208}Pb = 52.4%) (Pollard et al. 2018, pp. 145–46).

However, the investigation already carried out and the collected data show the variation in lead isotope ratios (e.g. 206Pb/204Pb) across Europe (Pollard et al. 2018, p. 148, Fig. 1). There is a general east–west trend in isotopic values, with lower values in the west and higher values in the east. The lowest values are found in northwest Scotland and Fennoscandia, while a belt of much higher val-

[1] It has to be admitted, however, that the copper alloys of Bronze Age Eurasia are the subject of the most research, for well over a century and also currently.

ues occurs between Sweden and Norway. This results in significant overlap in the isotopic values of metalliferous ores from different sources, particularly across mainland Europe and Asia Minor (Pollard et al. 2018, p. 147). The slightly varying lead isotope ratios in the composition of different copper ores can also be identified. Even within the same deposit, there may be a variety of copper ores, ranging from bornite and chalcopyrite with very low impurities to tetrahedrites containing significant amounts of antimony (Sb), arsenic (As) and silver (Ag) (Pernicka 2014; Stos-Gale 2019, p. 88). Different artefacts may have grossly different lead contents, but the lead will have the same isotopic composition. It is extremely unlikely that the lead isotope ratios will be altered by any of the physical and chemical processes that occur during the metallurgical process from ore to finished artefact, save for the mixing of lead of different origins. In other words, a key prerequisite for using lead isotopes to study the provenance of ores is ensuring that the ancient copper alloy has not been recast with alloys of different origins or chronologically distinct metal scrap, as the isotopic composition of lead varies across different regions. Attention should be paid to the fact that the application of isotope analysis requires that the raw material of the artefact to have remained unchanged in its chemical or physical state. Any such alterations could induce additional isotopic fractionations, potentially erasing the original differences associated with the geological sources.

Despite the problems encountered in lead isotope analysis in archaeometallurgical research, it is possible to shed light on the chronological changes in the composition of copper alloys and to explain the possibility of anthropogenic mixing of lead from different sources during smelting and metal production. Lead isotope analysis contributes to understanding the chronological changes in raw non-ferrous metals and identifying long-distance, regional and local exchange routes for imports entering the region. These data enable assumptions about the mines from which copper alloy ingots and/or artefacts entered the region. The use of lead isotope ratios data in studies of geological ore deposits and metal provenance analysis in archaeology has proven to be a useful tool for investigating the relationship between the regions with ore mines and the copper alloys used by various societies from the Bronze Age onwards. In other words, lead isotope analysis is expected to support the hypothesis regarding the 'origin of copper alloys as raw metal'. However, like any method, lead isotope analysis has advantages, disadvantages and challenges.

The first disadvantage might be considered to be the uncritical use of lead isotope data on copper alloys, which can be potentially misleading in determining provenance (Bray and Pollard 2015; Pollard et al. 2018, p. 150). This issue arises from the inherent inhomogeneity of ores on all scales, and this fact, in many cases, which often challenges the prevailing assumption of a straightforward correlation between artifacts and their ore sources. The origin of the ore can, at least partially, be resolved by the ratios of basic impurities in copper alloys and the rare earth elements associated with specific ore deposits (Pernicka 2014, Fig. 11.1 and 11.5). Experimental studies currently provide no evidence that smelting alters the isotopic composition of the copper in the resulting metal, as previously proposed (Gale et al. 1999; Durali-Müller 2005, p. 77, Table 5.5). However, it must be acknowledged that establishing a linear relationship between ore deposits and artifact production sites in the north remains unproven.

Even if it has to be re-emphasised, the usefulness of lead isotope analysis in archaeological studies has been a subject of controversy. However, its suitability for metal provenance studies is now widely accepted, provided there is a proper application that would take into account complementary archaeological and/or geological data (Pernicka 1999; Durali-Müller 2005; Pollard 2009; Holmqvist et al. 2019; Radivojević et al. 2019; Artioli et al. 2020; Killick et al. 2020; García de Madinabeitia et al. 2021; Merkel 2018; 2021; 2022). Once again, one of the greatest dangers in lead isotope traceability of non-ferrous metals is the re-melting and re-casting of archaeological artefacts into the new copper alloys. This is known as mutability, which offers the possibility for artefacts produced from copper alloy to be recycled multiple times. This property is remarkably versatile, allowing copper alloy to be reused, reshaped, merged, split and finally recontextualised into new forms of artefacts almost indefinately. However, this exceptional quality has proven frustrating for both archaeometallurgical and archaeological research (Pernicka et al. 1993; Pernicka 2014; Pernicka et al. 2016; Fleming 2012; Pollard et al. 2018; Stos-Gale 2019, pp. 96–97; Bray 2020; 2022; Sainsbury et al. 2021). It has significant implications, namely the fact that identifying and quantifying recycling poses a notorious and considerable challenge, especially within the framework of the 'provenance hypothesis'. This hyphotesis seeks to establish a direct link between a chemical or isotopic signature of an artefact and its original ore source (Wilson and Pollard 2001; Pollard et al. 2018, p. 37–40; Bray 2020, pp. 237–239; 2022; Pernicka 2014, pp. 239–247; Holmqvist 2019). Despite the rapid increase in published results from lead isotope analysis, no unequivocal links have been established between the sources and artefacts.

However, the positive aspect of this complex process is that this mutability can reveal much about the social context and significance of archaeological material culture. Ultimately, it may offer information about the movement of commodities as a part of exchange networks. To summarise the importance of the provenance of raw metal in archaeometallurgical studies, it is clear that, in addition to lead isotope ratios analysis, extensive comparative geochemical investigations of the ore mines and wide range of archaeological materials are essential. These materials must span regional and trans-European cultural and communication networks.

It is also worth noting the lack of sufficient analyses of lead isotope ratios and comparable material from the Roman Iron Age to the end of the Viking Age. This is perhaps due to the severe scarcity of non-ferrous metals during this period, a problem that significantly affected large parts of Europe. s This issue was particularly acute in regions without local ore deposits, such as the eastern Baltic Sea region (for this, see Čivilytė 2014, pp. 50–58; Roxburgh 2023).

This article presents the first ever lead isotope ratio analysis carried out on archaeological finds from the second half of the 1st century AD to the 13th century in Lithuania. The samples, mostly drilled from artefacts produced locally in Lithuania, were analysed to determine lead isotope ratios, copper alloy chemical composition, alloy types and copper groups. Consequently, the scientific questions addressed in this article are based on four main, distinct yet interrelated analyses.

The primary aim was to determine lead isotope ratios, characterise copper alloy types based on their main alloying elements, and classify copper groups according to their primary impurities. Additionally, the study sought to identify the set-up technologies used in artifact production. The results provide insights into the flow of non-ferrous alloys exchange at local (present-day territory of Lithuania), regional (Eastern Baltic region and Finland) and trans-European levels. This analysis challenges the prevailing linear exchange model, which links ore mines directly to artifact production, by instead highlighting the complexity of far-reaching exchange networks. All this gives opportunity for better understanding of past people's behaviour trying to secure long lasting vital model for obtaining necessary raw materials.

1. Materials and methods

1.1. Archaeological background and sampling

The authors analysed 180 artefacts and 208 samples taken from the artefact metal core (metal shavings). These samples were taken from 57 archaeological sites in Lithuania dating from the 1st to the 13th century AD (Fig. 1). A wide range of finds typical of the region and made locally were examined (Michelbertas 1986; Tautavičius 1996; Bliujienė 2023), with the addition of 10 imported finds including Roman coins (sestertii and dupondius), a brooch (Type A69), pieces of a horse bridle chain (Type B) and a series of pieces of jewellery of types widely distributed in Barbaricum and in Baltic lands: a neck-ring with trumpet-shaped terminals, eye brooches and bracelets with knob-shaped ends (Michelbertas 1986, pp. 107–110, 135, 189–190; Riha 1979, Table 78.2.3, 2.4, 2.6; Wilbers-Rost 1994, Table 8.B). Other find samples are drilled of typical to Lithuania artefacts and are produced by local craftsmen (Michelbertas 1986, pp. 84–154; Tautavicius 1996, pp. 791–810; Bliujiene 2023, Figs. 139–141, 154–159).

Prior to sampling the archaeological finds, in order to assess their condition, i.e. the degree of corrosion, construction and design subtleties were checked firstly using a portable diagnostic X-ray device as well as detailed observation of artefacts under a microscope (method as described in Bliujienė et al. 2020). Based on the radiographs and microscope observation considering the level of corrosion and differences in the thickness of the metal core, points for artefact drilling were selected. Samples were drilled from artefacts, separating the patina, subpatina and the metal core. The artefacts were sampled by drilling with an electric drill and bits with a diameter of 1.5–2.0 mm. After the overall review of the radiographs and artefacts and selection of drilling points, it became clear that it was not feasible to take samples of 0.5 g from archaeological finds. Therefore, our drilling went as far as was possible without endangering the survival of the fragile artefact and with minimal damage to it. The weights of the samples are small, ranging from 0.010 g to 0.11 g (or 10.12–150.45 mg).2 The mass spectrometry require a sample of the metal core of the find (1–20 mg or 0.001–0.02 g) and the method accuracy commonly ran at better than 0.01% for all ratios (Baker at al. 2006; Pernicka 2014; Stos-Gale 2019, pp. 90–91). Thus, the weights of the metal core samples drilled from the archaeological finds met the requirement for such analysis.

1.2. Methods

The ICP-MS method for analysing the chemical composition of archaeological finds was first applied to a series of tests on the certified reference materials (CRM). The analysis of shavings drilled from the Cultural Heritage Alloy Reference Material (Brass alloy CHARM= 31XTB5.B) was carried out. The need for these standards is well known in the history of their development and can be deduced from the historiography of archaeometallurgical research and the comparisons that have already been made (Heginbotham et al. 2015; Heginbotham and Solé 2017; Pollard et al. 2018, pp. 69–70). The shavings of the standards were obtained by drilling the discs laterally at 5 mm from the top surface and then up to 10 mm thickness, prepared as samples weighing 0.007 to 0.073 g or 7–73 mg (Table 1).

[2] All ICP-MS analysis results are available from: http://lydiniai.lt/.

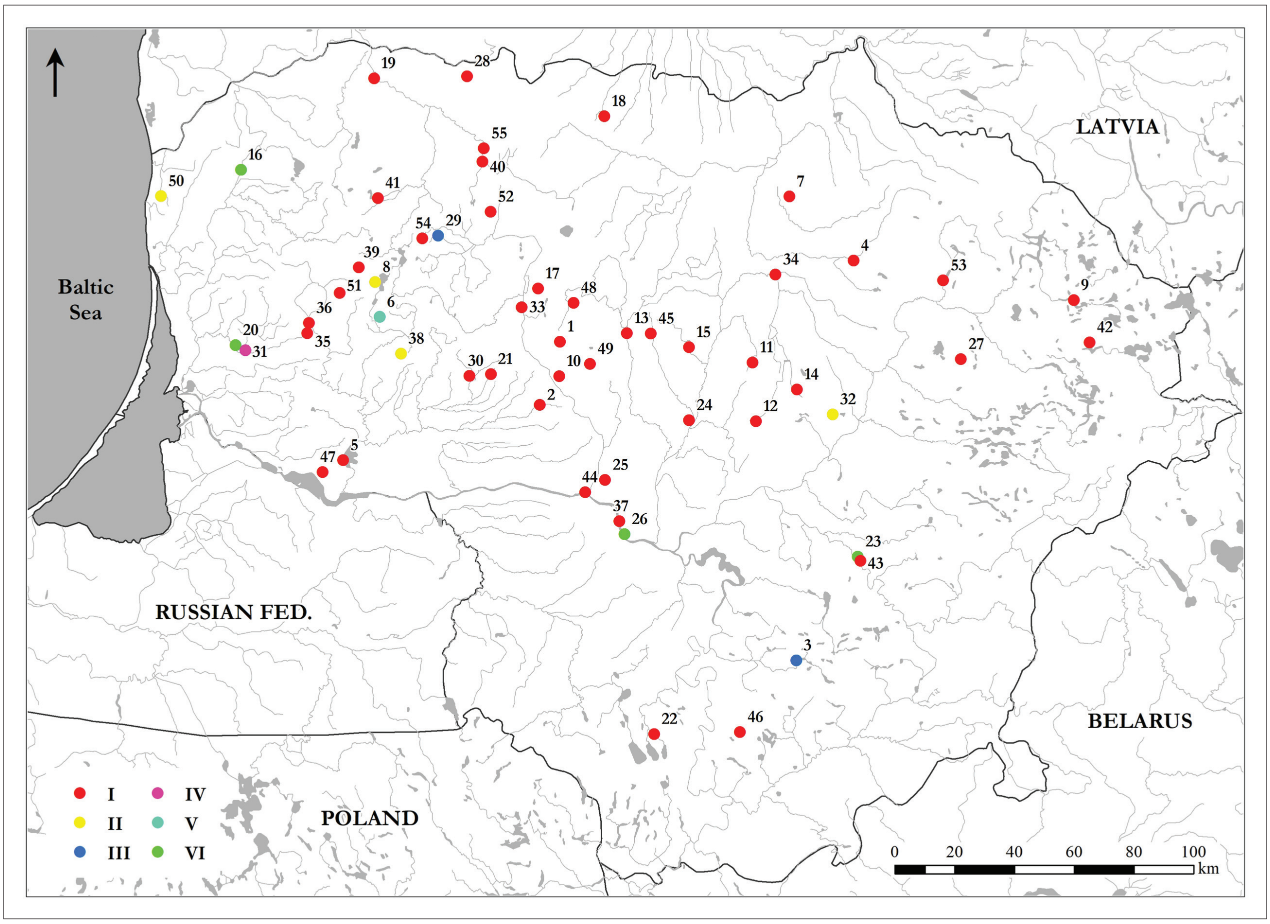

Figure 1. Locations of sites with analysed lead isotope ratios, dating to the Roman Iron Age (I), from the Roman Iron Age to the Migration Period (II), from the Roman Iron Age to the Viking Age (III), the Roman Iron Age and the Viking Age (IV), the Roman Iron Age, the Viking Age and the Middle Ages (V), and the Viking Age (VI): 1. Akmenė (Sandrausiškė), Raseiniai dist.; 2. Anulynas, Raseiniai dist.; 3. Aukštadvaris, Trakai dist.; 4. Bajoriškiai, Kupiškis dist.; 5. Barzūnai, Pagėgiai mun.; 6. Bilioniai, Šilalė dist.; 7. Daujėnai (Baluškiai), Pasvalys dist.; 8. Drobūkščiai, Telšiai dist.; 9. Eikotiškis, Zarasai dist.; 10. Fliorencija, Raseiniai dist.; 11. Gabulai, Panevėžys dist.; 12. Glaušiai, Kėdainiai dist.; 13. Grinkiškis, Radviliškis dist.; 14. Gružos (Užulėnis), Ukmergė dist.; 15. Ibutoniai, Panevėžys dist.; 16. Imbarė, Kretinga dist.; 17. Jagminiškė, Kelmė dist.; 18. Jautakiai, Mažeikiai dist.; 19. Jauneikiai, Joniškis dist.; 20. Jurgaičiai, Šilutė dist.; 21. Jūkainiai, Raseiniai dist.; 22. Kaukai, Alytus dist.; 23. Kernavė, Altar Hill hillfort, Širvintos dist.; 24. Kėdainiai; 25. Krūvandai, Kaunas dist.; 26. Kulautuva, Kaunas dist.; 27. Leliūnai, Utena dist.; 28. Mančeliai, Akmenė dist.; 29. Maudžiorai, Kelmė dist.; 30. Nemakščiai, Raseiniai dist.; 31. Nikėlai, Šilutė dist.; 32. Obeliai, Ukmergė dist.; 33. Pabariukai (Eimančiai), Kelmė dist.; 34. Pajuostis, Panevėžys dist.; 35. Pajūralis (Skerdynai), Šilalė dist.; 36. Paragaudis, Šilalė dist.; 37. Paštuva, Kaunas dist.; 38. Požerė (Paežeris), Šilalė dist.; 39. Pribitka, Rietavas mun.; 40. Raganiai, Šauliai dist.; 41. Rainiai, Telšiai dist.; 42. Rokėnai, Ignalina dist.; 43. Semeniškiai II, Širvintos dist.; 44. Seredžius II, Jurbarkas dist.; 45. Skėmiai (Skienai), Radviliškis dist.; 46. Slabadėlė, Alytus dist.; 47. Strazdai (Ječiškės), Pagėgiai mun.; 48. Šiaulaičiai, Radviliškis dist.; 49. Tautušiai, Raseiniai dist.; 50. Užpelkiai, Kretinga dist.; 51. Vienragiai, Rietavas mun.; 52 – Visdergiai (Papelkiai), Šiauliai dist., 53. Visėtiškės; Anykščiai dist.; 54. Žaduvėnai, Telšiai dist.; 55. Žiliai-Šemetaičiai, Šauliai dist.Findspots in central Lithuania and on the banks of the Venta River are not marked on the map, as the exact locations are unknown. Map prepared by Petrauskas, after Bliujienė et al. 2023, open access database, available from http://lydiniai.lt/.

ICP-MS analysis was carried out at the Klaipėda University Marine Research Institute. For chemical analysis, copper alloy samples were prepared by dissolving them in aqua regia solution using microwave digestion (Anton Paar Multiwave Go). For mineralisation, the Inorganic programme provided by the producer Anton Paar was used. First, in all the samples, alloying elements copper (Cu), zinc (Zn), tin (Sn) and lead (Pb) were measured and also the main impurities iron (Fe), nickel (Ni), silver (Ag), arsenic (As), antimony (Sb), manganese (Mn), cobalt (Co) and bismuth (Bi). ICP-MS is capable of quantitatively determining trace elements in liquids in the range of fractions of a part per billion (Durali-Müller 2005, p. 14).

After that, the same samples were diluted 100 times and used for lead (208Pb/204Pb versus 206Pb/204Pb, 207Pb/204Pb versus 206Pb/204Pb, 208Pb/206Pb versus 206Pb/204Pb and 207Pb/206versus 206Pb/204Pb) isotope ratio measurements. The control sample NIST SRM 981 was used to measure the samples to ensure the accuracy of the isotope analysis. The lead isotopic standard was prepared by dissolving 0.5 g of NIST SRM 981 in the same aqua regia solution and mineralization programme as the samples. The final NIST SRM 981 concentration for the measurements was chosen as 100 μgL-1. The Pb ratio of all samples was analysed by ICP-MS (Perkin Elmer Nexion 2000) standard mode. NIST SRM 981 was analysed identically as an artefact sample blank was prepared in the same way as the samples and measured at the beginning of the measurements. The standard was measured every 6 samples to monitor the precision and accuracy of analyses.

Copper groups can be used to detect the recycling of metal, based on the thermodynamic properties of the different trace or impurity elements. As, Sb, Ag and Ni were used. For interpreting trace element data in copper groups, a simple presence/absence classification system based on the above-mentioned four elements was carried out, which can be used to detect the recycling of metal, based on the thermodynamic properties of the different trace elements (Bray et al. 2015; Pollard et al. 2015; Pollard et al. 2018, pp. 85–90) for that values of these four elements might be of 0.1%. As, Sb, Ag and Ni are related to the ore source since they tend to be either present or absent in the ores known to have been used in antiquity, but we make no assumptions about allocating a particular copper group to a specific source, known or unknown, since mixing and recycling can move an object from one group to another from a geographical perspective and, very importantly, in terms of chronological changes in relationships in the flow of non-ferrous metals.

2. Results

2.1. Lead isotope ratios

The lead isotope ratios determined allowed us to assess the reliability of the selected methods for the analysis of extremely low-weight samples. Lead isotope ratios of 158 samples and 155 artefacts from the Roman Iron Age (1st century to the third quarter of the 4th century) show relatively homogeneous compositions of ^{208}Pb/^{204}Pb versus ^{206}Pb/^{204}Pb isotope values in the range respectively approximately between 38.5–39.0 and 18.4–18.6 (for the results, see Fig. 2.1-4; Table 2; Supplement 1). However, the values of these lead isotope ratios for all of the archaeological sites that have been studied are spread over a wider range ^{207}Pb/^{204}Pb versus ^{206}Pb/^{204}Pb isotopes ratio values making the concentration between 15.6–15.7 and 18.4–18.6. Similarly, lead isotope ratios for all of the archaeological finds that have been analysed from the Roman Iron Age are spread over a wider range. For ^{208}Pb/Pb^{206} versus ^{207}Pb/^{206}Pb isotope ratios it appears that from a small uniform centre,

Table 1. Results of ICP-MS analysis of brass alloy standard (CHARM =31XTB5.B) shavings weighting from 0.007 to 0.073 grams.

Elements		Cu	Zn	Sn	Pb	Fe	Ni	Ag	Sb	As	Bi	Co	Mn
CRM value		61.49	35.62	0.129	0.575	0.094	0.106	0.216	0.299	0.396	0.292	0.0202	0.283
Uncertainly		0.07	0.12	0.002	0.008	0.002	0.002	0.003	0.005	0.007	0.008	0.0006	0.005
CRM ID	Weight	wt%											
31XTB5.B	0.042	62.503	34.650	0.128	0.545	0.096	0.109	0.226	0.282	0.484	0.270	0.021	0.272
31XTB5.B	0.007	61.463	34.188	0.129	0.588	0.095	0.091	0.219	0.297	0.369	0.291	0.018	0.278
31XTB5.B	0.073	61.291	35.382	0.125	0.547	0.091	0.105	0.091	0.320	0.399	0.296	0.019	0.267
31XTB5.B	0.051	61.478	35.171	0.120	0.562	0.090	0.090	0.195	0.274	0.395	0.261	0.021	0.227
31XTB5.B	0.010	59.459	35.548	0.125	0.443	0.076	0.103	0.196	0.282	0.364	0.296	0.017	0.274

The CRM certified values are the present best estimates of the true content for each element. Each value is a panel consensus based on the averaged results of an interlaboratory testing programme. The uncertainty was generated from the 95% confidence interval derived from the wet analysis result in combination with a statistical assessment of the homogeneity data.

Table 2. Chronological comparison of aggregated lead isotope ratios from lowest to highest fixed values and amount of lead in wt% (after data in Supplement 1). Data for Estonia after Roxburgh 2023, Table 1, Fig. 4;
Data for Finland after Holmqvist et al. 2019, Table 3, Fig. 10.

^{208}Pb/^{204}Pb	^{207}Pb/^{204}Pb	^{206}Pb/^{204}Pb	^{208}Pb/^{206}Pb	^{207}Pb/^{206}Pb	Pb wt %
LITHUANIA: ROMAN IRON AGE (n=159)					
n=154, 38.109-39.596	n=157, 15.519-15.958	n=158, 18.185-18.876	n=159, 2.058-2.122	n=159, 0.837-0.865	n= 113, 0.4-1%
n=4, 37.852-37.976	n=2, 16.08365-16.387	n=1, 19.404			n= 46, 1-14.25%
n=1, 40.888					
ESTONIA: ROMAN IRON AGE (n=12)					
n=12, 38.344-38.843	n=12, 15.632-15.688	n=12, 18.34-18.74	n=12, 2.087-2.094	n=12, 0.850-8.53	
FINLAND: ROMAN IRON AGE AND MIGRATION PERIOD (n=8)					
n=8, 38.283-39.418	n=8, 15.639- 15.668	n=8, 18.495-18.673	n=8, 2.07-2.085	n=8, 0.831-0.846	
LITHUANIA: MIGRATION PERIOD (n=20)					
n=16, 38.029-38.994	n=19, 15.524-15.781	n=18, 18,227-18,560	n=19, 2.066-2.122	n=19, 0.842-0.883	n=7, 0.1-1.8%
n=3, 39.048-39.129		n=1, 17,645			n=13, 3.4-11.44%
n=3, 37.443					
LITHUANIA: VIKING AGE (n=13)					
n=10, 38.223-38,997	n=13, 15.378-15.864	n=13, 18.086-18.834	n=13, 2.076-2.125	n=13, 0.838-0.858	n=13, 0.15-18.61%
n=3, 39.008-39.429					
LITHUANIA: EARLY MEDIAVIAL AGE (n=2)					
n=2, 38.61344-3.076	n=2, 15.637-15.619	n=2, 18.416-18.331	n=2, 2.097-2.077	n=2, 0.849-0.85	n=2, 6,8-11.463%

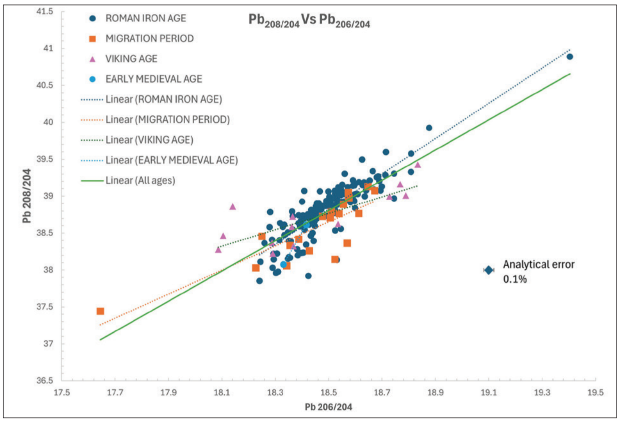

Figure 2.1. 208Pb/206Pb vs 206Pb/204Pb isotope ratios. The symbols indicating isotope ratios and chronology are common to all graphical representations in Figure 4. Plotted in dark blue circles – Roman Iron Age; plotted orange quadrangulars – Migration period; plotted pink triangulars – Viking Age; plotted light blue circles – Early Medieval Age. Diagrams prepared by Pudžaitis.

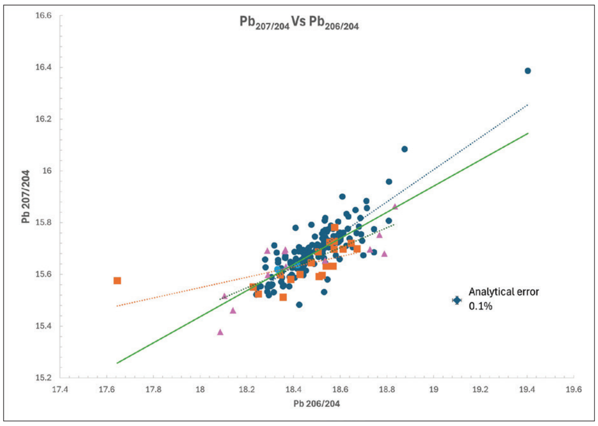

Figure 2.2. 207Pb/204Pb vs 206Pb/204Pb isotope ratios.

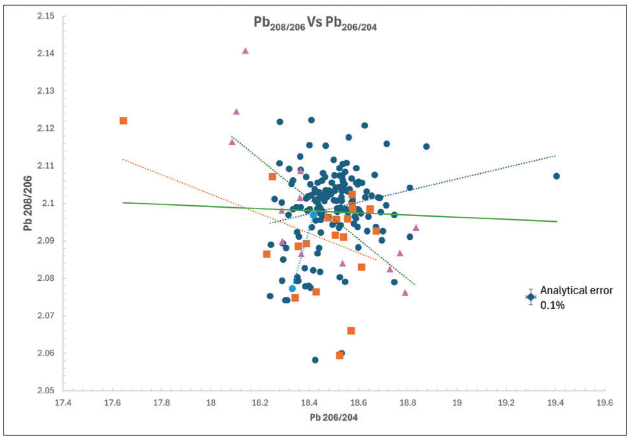

Figure 2.3. 208Pb/206Pb vs 207Pb/206Pb isotope ratios.

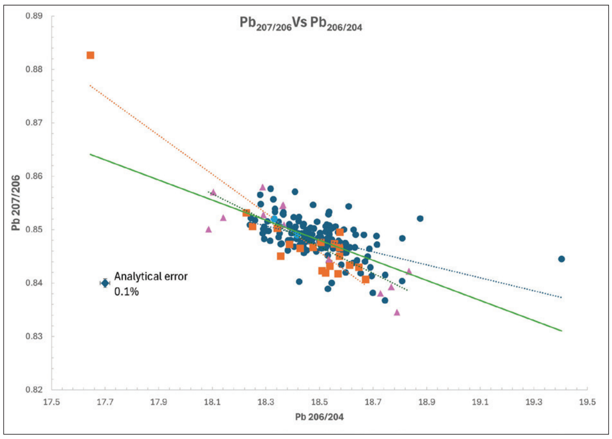

Figure 2.4. 206Pb/204vs 207Pb/206Pb isotope ratios.

the values range approximately between 2.09–2.10 and 0.845–0.85. However, values are spread over a relatively large area. ^{206}Pb/^{204}Pb versus ^{207}Pb/^{206}Pb isotope ratios are concentrated in the range approximately between 18.4–18.6 and 0.847–0.85. However, isotope ratios for all of the archaeological finds that have been analysed in the Roman Iron Age are spread over a wider range. In addition, it has been observed that there is a fairly consistent increase in the lead content of copper alloys over the Roman Iron Age (n=113, from 0.04 wt% up to 1 wt% and n=45, from 1 up to 14.25 wt%) (Table 2).

Fewer finds from the Migration Period (n=20) have been analysed, but the lead isotope ratio values are scattered in the same range as in the Roman Iron Age (Fig. 2.1–4; Table 2; Supplement 1). Lead levels also do not decrease in the Migration Period. Within the small sample studied, lead accounts for between 0.1 and 11.44 wt%.

Only 13 artefacts were investigated from the Viking Age.[3] The lead isotope ratios of this period fall into to two distinct groups according to the isotope ratios obtained (Fig. 2.1–4; Table 2; Supplement 1). There is also a trend towards higher lead content in copper alloys, from 0.15–18.61 wt%.

2.2. Artefact chemical composition analysis

The ICP-MS analysis made it possible to determine the chemical composition of the archaeological finds' metal core. Results of reference materials and shavings drilled from the Cultural Heritage Alloy Reference Material (Brass alloy CHARM= 31XTB5.B) are within the tolerance limits, as indicated in Table 1. Results are separated into the copper types accepted in modern archaeomaterial analysis (Pollard et al. 2018, p. 117, Table 1). The recalculated results obtained allowed the classification of the alloys into types (results are indicated in Supplement 1). The ICP-MS chemical composition analysis data of alloying elements and main impurities ranging from 95 wt% are to be considered fully reliable. Only a few results with lower values are included in the ICP-MS analysis. All the results included in Supplement 1and discussed in the text were converted to 100 wt% using the following formula: $Ei = E_{ki}. (\Sigma E_i / 100)$. On the basis of presence/absence (Yes/No) of main trace elements As, Sb, Ag and Ni copper groups were formed using the formula: $E_{ki} = E_i.(100 / \Sigma E_i - E_{Sn} - E_{Pb} - E_{Zn})$, i = all analysed elements from Ag(1) to Zn(12). Ei = initial content of the first element; Eki = percentage content of the first element after recalibration, where ki = Cu, As, Sb, Ag, Ni.

[3] For the Early Medieval Period only two artefacts were analysed for lead isotopes ratios, so these results are indicated in Figures 2.1-4, but are not discussed in the article text.

3. Discussion

3.1. Lead isotope ratios and possible non-ferrous metal ore deposits

Lead isotope ratios are particularly well suited to determining the provenance of the mine ores. Since the ratios of lead isotopes do not change during metallurgical processes or chemical reactions during recasting, the origin of the metal can be defined uniquely, with the one exception of where mixing of metals from different provenances occurs (Durali-Mueller et al. 2007).

In order to discuss and answer the main intertwined questions raised in this paper, it is necessary to ask where the non-ferrous metal alloys came from during the long chronological period covered by the study, in the eastern Baltic region and, of course, in Lithuania, where there are no metal ore deposits. If we compare ^{208}Pb/^{204}Pb versus ^{206}Pb/^{204}Pb, ^{207}Pb/^{204}Pb versus ^{206}Pb/^{204}Pb, ^{208}Pb/^{206}Pb versus ^{207}Pb/^{206}Pb and ^{206}Pb/^{204} versus ^{207}Pb/^{206}Pb analysis data from Lithuanian archaeological artefacts, it is possible to argue that from the Roman Iron Age to the Migration Period, lead isotope ratios show a certain apparent homogeneity, while at the same time there is a certain spread of values within the frame of characteristic values of lead isotopes in ore deposits. Therefore, it is possible to compare the lead isotope ratios of geological ore deposits and archaeological finds distributed in specific areas of Europe, and to prove or disprove a linear exchange model between the product of ore mines and archaeological finds (Gale and Stos-Gale 1982; Gale et al. 1999; Pernicka et al. 1993; Gale and Stos-Gale 2000; Stos-Gale 2019; Begemann et al. 2001; Höppner et al. 2005; Marcoux et al. 2002; Durali-Müller 2005; Durali-Mueller et al. 2007; Istenič and Šmit 2007; Bode et al. 2009; Klein et al. 2010; Holmqvist et al. 2019; Radivojević et al. 2019; Artioli et al. 2020; García de Madinabeitia et al. 2021; Mehofer et al. 2021; Roxburgh 2023; Kershaw and Merkel 2024).

Pliny the Elder, in his *Natural History*, was one of the first scholars who provided some information on the main precious and non-ferrous metal alloys used in the ancient world, naming some already exploited ore deposits in Cyprus, Campania (southern Italy), Cartagena (Spain), and mentioning active mines supplying similar 'copper' in Spain, Gallia and Germania (Healy 1999, pp. 271–326). Pliny the Elder also described technologies of artefact production. Previous and ongoing research indicates that many of the deposits exploited in the Bronze Age were not exhausted and continued to be exploited in the ancient world and later (Durali-Müller 2005, pp. 6–8, Fig. 3.1; Stos-Gale 2019, pp. 97–116, Fig. 3). The sources of lead and other metals in the Roman Empire were Spain, with over 560 mines and smelting places extracting silver, lead, gold, copper and mercury; France, where the most important districts of mining were in Gallia, Narbonensis and Aquitania; and Britain, which played an important role as a source of raw material, particularly for lead and tin, and became a major lead exporter in the Empire. Romans obtained gold, silver, lead and copper from several mining regions of Britain, Germany, the Eastern Alps, Italian Alps, Sardinia, Greece, Cyprus, the Aegean area, etc. (Durali-Mueller 2007). Many of the mines in the areas listed above have been worked since the Bronze Age and continue to be used. In fact, from a theoretical point of view, the means of access to non-ferrous metals for the eastern part of the Baltic Sea region has been extensive, encompassing southern Europe and Asia Minor (Fig. 3).

The main initial results suggest that from the Roman Iron Age and later on, societies of the present-day territory of

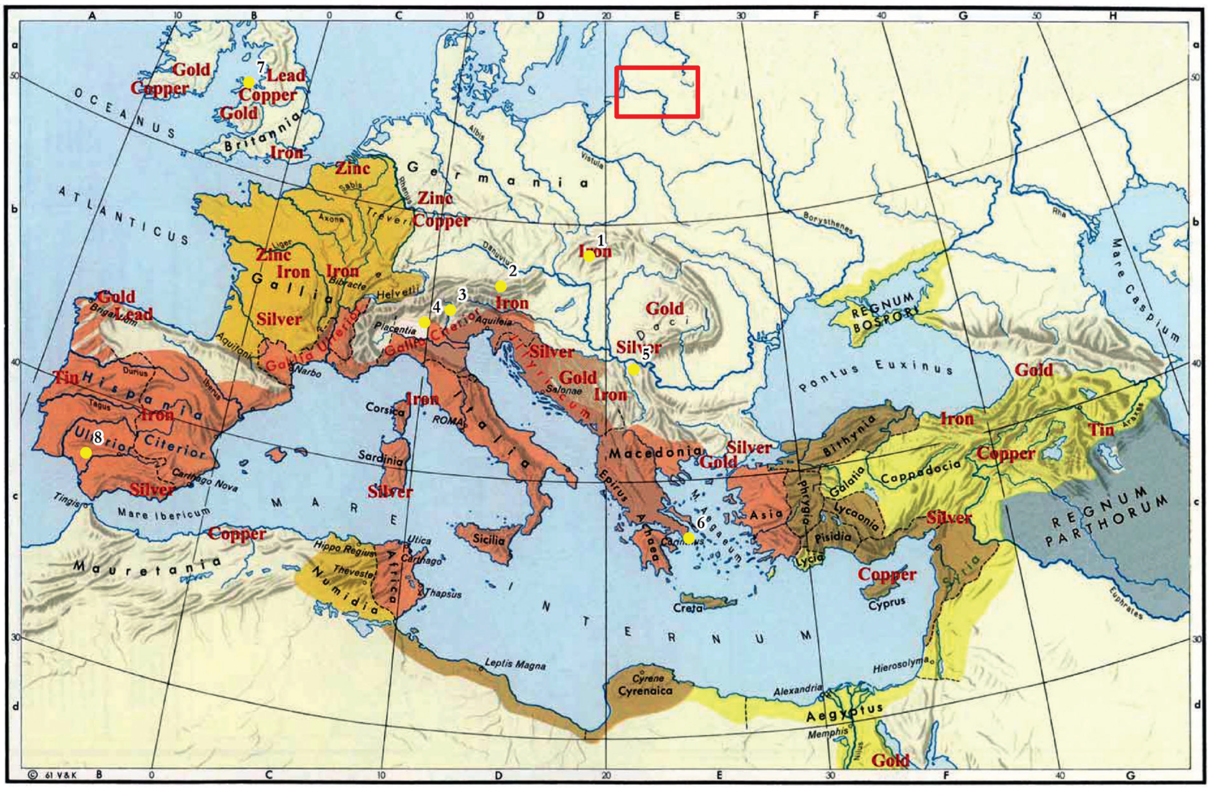

Figure 3. The map illustrates the principal metal ores of Eurasia and the regions from which non-ferrous metals were exported to the territory of present-day Lithuania between the second half of the 1st century and the beginning of the 13th century AD. 1. Slovak Ore Mountains (Hron Valley); 2. Mitterberg (Austrian Alps). 3. Italian Alps (Trentino-Bolzano), 4. Northern Italy (Dossena in Gorno district); 5. Serbia (Bor metallogenic zone); 6. Southern Greece (Lavrion); 7. Great Orme in Wales (British Isles); 8. Andalusia (southern Spain) (map after Durali-Müller 2005, p. 9, Fig. 1.3, the article authors have provided a regional account of metal flows to Lithuania).

Lithuania were receiving copper alloys from ores located in southern Europe in general. ^{208}Pb/^{206}Pb versus ^{207}Pb/^{206}Pb and ^{206}Pb/^{204} versus ^{207}Pb/^{206}Pb isotope ratios of the Roman Iron Age show a sufficiently wide dispersion of isotope ratio values, which, when compared with the published data from the ore deposits available in databases (OXALID https://oxalid.arch.ox.ac.uk/default.html, IBERLID https://www.ehu.eus/ibercron/iberlid etc.) would indicate that the copper alloys may have originated from ores in the region of Trentino-Bolzano (Italian Alps), Great Orme in Wales (British Isles), Slovak Ore Mountains (Hron Valley) or Mitterberg (Austrian Alps). It might be that the Lavrion mines in the southern Greece and Spain became suppliers of non-ferrous alloys in the Roman Iron Age (OXALID; Rohl and Needham 1998; Artioli et al. 2016; Modarressi-Tehrani et al.; 2016; Pernicka et al. 2016; Stos-Gale and Gale 2009; Stos-Gale 2019, pp. 93–116, Figs. 2–8) (Fig. 4.1-2). These ore mines were operating from the Bronze Age. The Slovak Ore Mountains, being the part of Western Carpathians ore mines region were one of the major ore deposits of Central Europe and are regarded as a major supplier of copper ores since the late 5th millennium BC to Poland and to Lithuania (Miarkiavichius 1980, p. 110; Čivilytė 2016, pp. 54–58, Figs. 30, 32; Stos-Gale 2019, p. 98). The lead isotope ratios from the Migration Period are largely consistent with the results of previous analyses. The isotopic studies carried out show that the main and dominant flow of non-ferrous metals during the Roman Iron Age was from the Slovak Ore Mountains, the Italian and Austrian Alps and the British Isles to the territory of present-day Lithuania (Fig. 3). Thus, the lead isotope values indicate that the copper alloys of the analysed Lithuanian Roman Iron Age artefacts originate from mines in the regions listed above. The graphs therefore show both the concentration of isotope ratios in a small range of values and the spread of ratios over a larger interval.

However, the isotopic composition of lead is altered by remelting when a new amount of copper alloy from another region and ore mine is added, i.e. the process of mutability kicks in. In trying to understand the movement of non-ferrous metal flows through long-distance exchange, it is necessary to point out that in the Barbaricum there were settlements where scrap metal, obtained by various routes and methods, was melted into new copper alloy ingots (for this see: Schmidt 2012; Voß 2016). Therefore, the 'fingerprints' of the original isotopic ratios of the lead could be lost during remelting and changes in the composition of the alloy led to the start of a long mutability process of alloy change (Durali-Mueller 2007). These remelted non-ferrous metal alloys were used to make new items, some of which may have been incorporated into the same long-distance exchange network, or the direction of movement may have changed. The perfect definition of radically changing the idea of the linear trajectory in the nonferrous metals exchange process was formulated in terms of a river flow, whereby a new stock of metal enters the flow and this addition in circulation will change the composition of the flow, just as the tributaries of a river contribute water with different chemical characteristics and sediment load (Bray et al. 2015, p. 203; Pollard et al. 2018, pp. 42–56, Figs. 1 and 3). The 'flow of the river of metal' is also a perfect reflection of the trans-European exchange process as

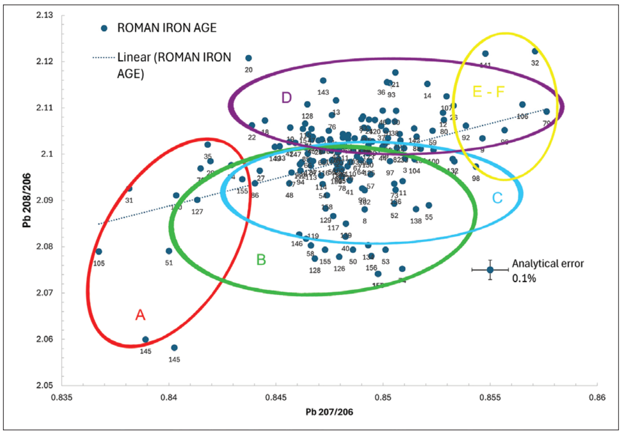

Figure 4.1. Roman Iron Age 208Pb/206vs 207Pb/206Pb isotope ratios of the archaeological artefact samples compared with the data for ores from the region of Trentino-Bolzano (Italian Alps), Great Orme in Wales (British Isles), Slovak Ore Mountains (Hron Valley) and Mitterberg (Austrian Alps). For lead isotopes data see supplement 1; after OXALID; Rohl and Needham 1998; Artioli et al. 2016; Modarressi-Tehrani et al. 2016; Pernicka et al. 2016; Stos-Gale 2019, pp. 93–116, Figs. 2–8; Merkel 2021). Diagrams prepared by Pudžaitis.

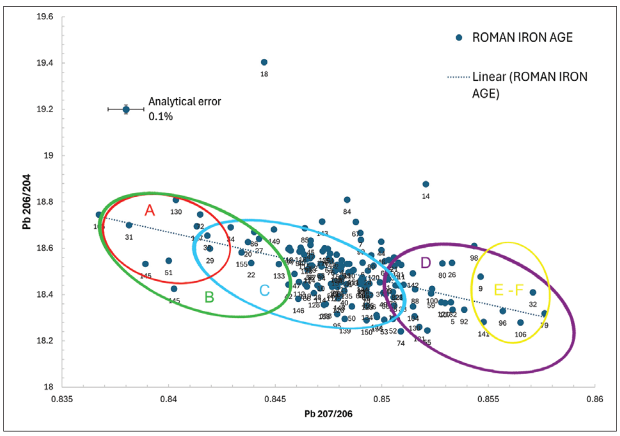

Figure 4.2. Roman Iron Age 206Pb/204Pb versus 207Pb/206Pb isotope ratios of the archaeological artefact samples compared with the data for ores from the region of Trentino-Bolzano (Italian Alps), Dossena in Gorno district (Northern Italy); Great Orme in Wales (British Isles), Slovak Ore Mountains (Hron Valley) and Mitterberg (Austrian Alps). For lead isotopes data, see supplement 1; after OXALID; Rohl and Needham 1998; Artioli et al. 2016; Modarressi-Tehrani et al. 2016; Pernicka et al. 2016; Stos-Gale 2019, pp. 93–116, Figs. 2–8; Merkel 2021).

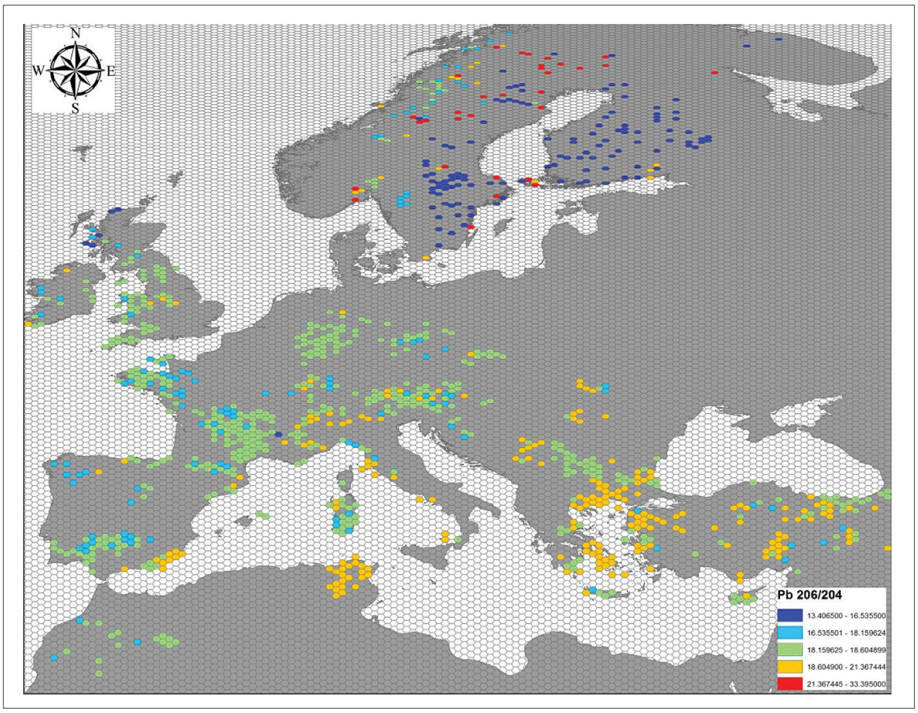

Figure 5. Variation in 206Pb/204Pb in ore deposits across Europe (after FLAME database; from Pollard et al. 2018, p. 148, Fig. 1).

a reality of the past and is an excellent metaphor. However, due to a lack of data, it is currently difficult to answer the question of where this river was replenished with new commodities and where commodities left the river. There is no doubt that the 'flow of the river of metal' has to be seen as a movement of commodities, even if it is not particularly easy to understand where this river was passing by.

Analysis of only 12 lead isotope samples from the Roman Iron Age in Estonia was undertaken. The results of these studies have not been elaborated. However, the main initial results clearly suggest that the territory of present-day Estonia was receiving brass produced from ore sources in southern Europe and from the Black Sea region and most likely new alloys were created by mixing the newly arriving copper alloy flow with older scrap (Roxburgh 2023, Figs. 4, 5). The Lithuanian Roman Iron Age part of the results overlaps with published Estonian lead isotope ratios. Estonian lead isotopic studies partially confirm that the eastern Baltic region may have been included in this common southern European non-ferrous metal flow. However, to confirm or disprove these hypotheses, further research is needed, which would include Latvia.4

Although there is not a lot of data on the Roman Iron Age and Migration Period available from the eastern Baltic Sea region for lead isotope comparison, recently eight copper alloy artefacts were recovered from the Iron Age water burial site of Levänluhta in western Finland dated to a large chronological period from 100 to 800 AD (Holmqvist et al. 2019, Fig. 1). The ores that present the closest comparison for the Levänluhta samples from the three lead isotope ratios of non-ferrous metal sources might be located in Spain, Bulgaria, Greece or Cyprus. In these regions, copper/lead mines were exploited from the Bronze Age (Holmqvist et al. 2019, Table 4, Fig. 12). In addition, the 207Pb/204Pb versus 206Pb/204 isotope ratios for Estonia and Levänluhta are in agreement, but there is no correlation between the Swedish and Finnish Uralic ores (Roxburgh 2023, Fig. 5a). Our lead isotope ratios analysis also shows no relationship between the Swedish and Finnish Uralic ores. However, Bronze Age and Early Iron Age archaeological material from Lithuania and Latvia shows the long-distance exchange relationships with the eastern European forest zone and some artefacts appear to result from the process of exchange (Podėnas and Čivillytė 2019, pp. 183–188, Figs. 7 and 10; Bliujienė et al. 2021b). There are also significant differences in 206Pb/204Pb isotope ratios between southern Europe and Asia Minor, southern Finland, and southeast and northern Sweden (Pollard et al. 2018, p. 148, Fig. 1). Most of the 206Pb/204Pb isotope ratio values in Lithuania, Estonia and Finland lie between 18.185 and 18.834 and are common to southern Europe and Asia Minor (Fig. 5). This suggest that the non-ferrous metal has been transported to the eastern Baltic Sea region by the trans-European exchange network from the southern regions during the whole period of time discussed in this paper.

[4] There are no published results of lead isotope studies from Latvia yet.

The Viking Age lead isotope ratios of this period fall within two distinct groups which are outside of or bordering on the lead isotopic ratios of the Roman Iron Age and the Migration Period. This is an indication that there has been a change in the direction of the inflow of copper alloy, because Migration Period shows that the composition of the lead isotopes has changed somewhat. The fingerprints of the Mitterberg (Austrian Alps) and the ore mines of southern Greece (Lavrion) and Spain are no longer present (Fig. 6.1–2; Table 2; Supplement 1). Perhaps the continuation of this line of enquiry another question, that of whether primary metal alloys, or even individual metals (e.g. lead), originated from ores already available during the Migration Period, and presumably may have functioned earlier in northwestern and central Sweden and northern Finland (Forshell 1992, pp. 39–45; Billstrom et al. 1997; Blichert-Toft et al. 2016; Bindler et al. 2017; Kershaw and Merkel 2024).

Our Viking Age isotopes ratios of the archaeological artefact samples might be compared with the data for ores from Bor and Majdanpek metallogenic zones in Serbia or even of Andalusia (southern Spain) (IBERLID; Pernicka et al. 1993; Merkel 2018; 2022). (Fig. 7.1–2). However, for analogues we might look to the isotope ratios obtained in Sweden have been sought in assessing the Viking Age contacts of the eastern Baltic Sea region with Scandinavia. Firstly, attention is drawn to studies at the 'central place' of Helgö in Lake Mälaren which uncovered a range of primary metal alloys, mainly in association with a Migration Period workshop (19 artefacts of lead, copper and brass, including ingots, rods and casting waste). Three lead melts come from Swedish lead ore. However, the published lead ratios do not match the ones we obtained; in contrast, the brass was imported over long distances as part of routinised trade with the Byzantine Empire, and potentially the eastern Mediterranean region (Kershaw and Merkel 2024).

A hoard of 25 copper-alloy bar ingots was discovered on the edge of the harbour of the Viking period settlement of Hedeby. The brass bar ingots found there are possibly products originating from the Balkans, specifically the metallogenic zones in Serbia, or Andalusia (southern Spain). The current lead isotope data points to the metallogenic zones in Serbia as the best fit for the source of the Hedeby bar ingots, and thus provides further evidence of the importance of the long-distance trade of raw materials in the Viking Age (Merkel 2018). The comprehensive study by Stephen W. Merkel (2018) on serial production of high-quality brass and the astonishing level of standardisation of the raw materials (down to trace element levels) indicates that the ingots were made in a well-organised workshop using firmly established metallurgical techniques and freshly extracted raw materials.

Our Viking Age artefacts collection comprises only 13 artefacts, but there are five brasses and two brass-based finds. At Kernavė 'Altar Hill' hillfort, a brass bar ingot was found which, according to the ICP-MS chemical alloy composition analysis (Cu 68.43 Cu, Zn 29.88, Sn 0,02 and Pb 0.77 wt%, also see Supplements 1 and 2), is close to the Hedeby bar-shaped ingots, but which differed in terms of lead isotope ratios (Merkel 2018, Table 3). However, at Aukštadvaris hillfort, two bar-shaped ingots and jewellery were found which, according to the lead isotope ratios, are close to the Hedeby ingots. This is reflected in both the chemical composition and the lead isotope ratios of the Lithuanian artefacts which have been analysed. It is pos-

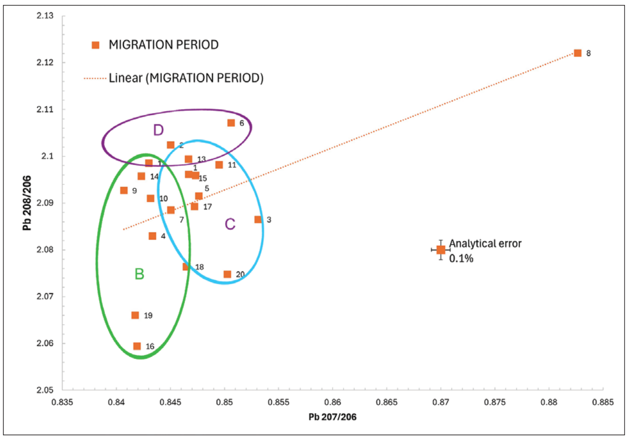

Figure 6.1. Migration period 208Pb/206versus 207Pb/206Pb isotope ratios of the archaeological artefact samples compared with the data for ores from the region of Trentino-Bolzano (Italian Alps), Great Orme in Wales (British Isles), Slovak Ore Mountains (Hron Valley) and Mitterberg (Austrian Alps). For lead isotopes data, see supplement 1; after OXALID). Diagrams prepared by Pudžaitis.

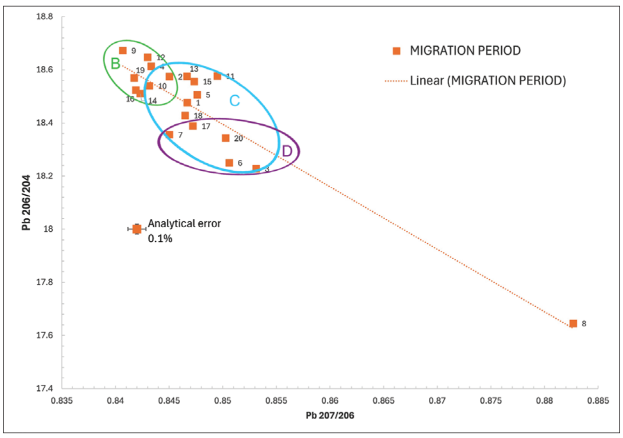

Figure 6.2. Migration period 206Pb/204Pb versus 207Pb/206Pb isotope ratios of the archaeological artefact samples compared with the data for ores from the region of Trentino-Bolzano (Italian Alps), Great Orme in Wales (British Isles), Slovak Ore Mountains (Hron Valley) and Mitterberg (Austrian Alps). For lead isotopes data, see supplement 1; after OXALID).

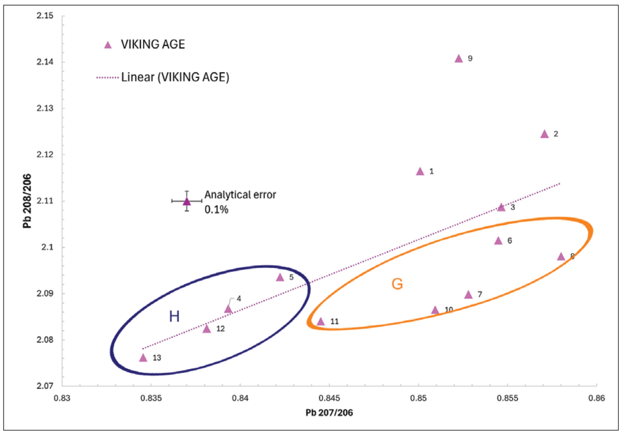

Figure 7.1. Viking Age 208Pb/206 versus 207Pb/206Pb isotope ratios of the archaeological artefact samples compared with the data for ores from Bor metallogenic zone in Serbia or Andalusia (southern Hispania). For lead isotopes data, see supplement 1; after OXALID; Merkel 2018; Kershaw and Merkel 2024.

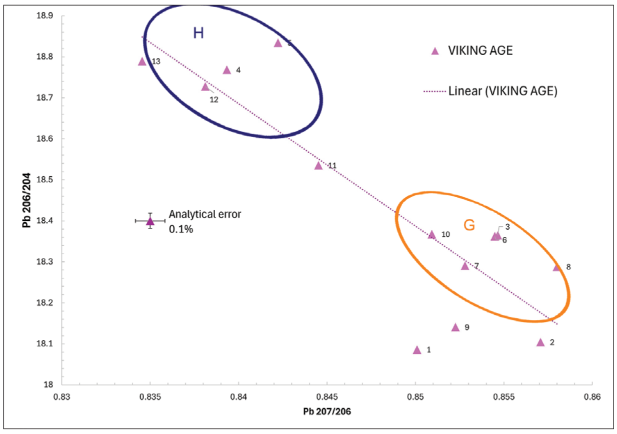

Figure 7.2. Viking Age 206Pb/204Pb versus 207Pb/206Pb isotope ratios of the archaeological artefact samples compared with the data for ores from Bor metallogenic zone in Serbia or Andalusia (southern Hispania). For lead isotopes data, see supplement 1; after OXALID; Merkel 2018; Kershaw, and Merkel 2024.

sible that the bar ingots were transported eastwards to the Black Sea region where they could have been converted to brass and traded via the eastern European forest zone river systems, mainly the Dnieper (Dnipro) reach, Nemunas (Memel) and eastern, central and western Lithuania, as well as the Baltic Sea. However, it is still possible that the copper alloys came from Scandinavia during the Viking Age, in this case from Hedeby, as there were close contacts between the areas. The process of remelting and reusing copper alloys can therefore explain certain isotopic discrepancies in the analyses.

In this context of the origin of non-ferrous raw materials, an important source of information is the Viking Age archaeological evidence and written sources, as well as the nature of the economic relations of the period which were based on long-distance exchange and trade, tribute, looting and warfare (Bliujienė 2008). As mentioned in Scandinavian sagas and other written sources, the Viking warfare raids, collection of tribute, and trade missions ending in looting enabled the Gotlanders to collect 20 kg of non-ferrous metal from jewellery in the eastern Baltic Region between the Estonian and Lithuanian coastal areas and the Lower Daugava (Western Dvina) and Lielupe Rivers in Latvia (Huttu 2004; Widerström 2004; Thunmark-Nylen 2006, pp. 701–706; Bliujienė 2008). It was not difficult for Scandinavian Vikings to obtain different artefacts made of copper alloys as tribute or ransom, or by outright looting of populations and graves. Anyway, the Gotlanders brought the 20 kg of non-ferrous metal to Gotland in Spillings (Othem parish) and hid it as valuable raw material until around the end of the 9th century to the beginning the 10th century AD. However, individual artefacts of this collection might be dated from 620 to 790 AD, because some of the Spillings artefacts are earlier and came from plundered graves (Huttu 2004; Bliujienė 2008).

All the Spillings artefacts are produced from copper alloys and are crudely broken ornaments, chopped up into little pieces and hurriedly partially melted in campfires so that metal scrap could be fitted into a fir chest. Other examples of the use of non-ferrous metals as scrap metal are related to grave plundering and deliberate, but temporary, hoarding of the valuables obtained in cemeteries during times of danger (for the multifaceted hoarding phenomenon in burial sites, see Heijne 2007). Hoarding in Lithuanian burial sites is known from Sargėnai (Kaunas city), Graužiai (Kėdainiai district) and Siraičiai (Telšiai district) cemeteries. All these non-ferrous metal scrap collections were deposited in the cemeteries during the Viking Age, and are undoubtedly raw materials destined for recycling and reuse. In addition to this, the looting phenomenon includes the perennial grave robbing and the storage of collected non-ferrous scrap metal in hillforts or settlements until the scrap metal was recycled and reused or exchanged for other goods.[5]

Although the main question is whether primordial non-mixed brasses or mixed ones, or alloys fused from various non-ferrous scrap metals, entered Lithuania, this remains somewhat open. However, as shown by ICP-MS surveys, supplemented by pXRF spectrometry analysis,[6]

[5] Scrap metal from the 11th-12th centuries was found in the Paverkniai (Birštonas municipality) hillfort (Zabiela 1994). Grave robbing was a particularly good source of non-ferrous metals. In fact, almost all the graves of the 4th–5th-century Eitulioniai (Trakai district) barrow cemetery in eastern Lithuania were plundered (Bliujienė and Bliujus 2021, pp. 346–47, Figs. 4 and 5). Most of the 9th–11th-century graves in Laiviai (Kretinga district), western Lithuania, have also been robbed (Gintautaitė-Butėnienė and Butėnas 2002).

[6] pXRF results from 2440 artefacts are available in a database at http://lydiniai.lt/, some of which are reproduced by ICP-MS. pXRF results are still under analysis and trends in copper alloy types and copper groups are emerging.

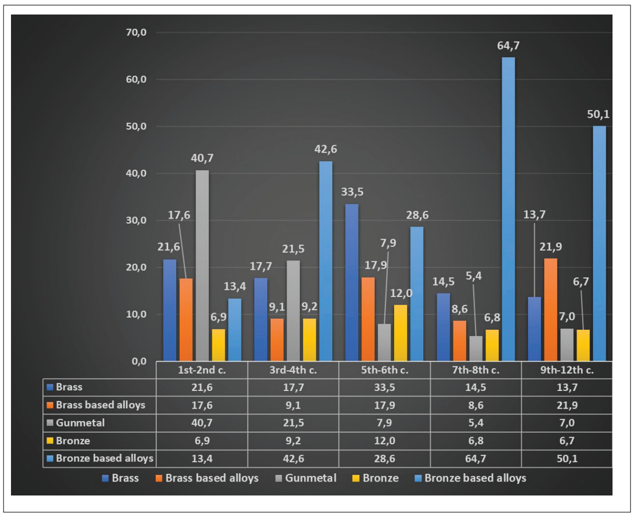

Figure 8. Percentage of brass, brass-based and bronze and bronze-based alloys produced in the 1st–12th centuries. Diagram based on dataset of pXRF analysis (prepared by Bliujienė).

new brasses were continuously arriving in Lithuanian archaeological material, but judging by the low zinc content of Roman Iron Age artefacts (usually only 10-20 wt%, 20-26 wt%, and up to 30 wt% zinc), these are not alloys from working ore mines. There are only a dozen finds of 20-26 wt% Zn from the Viking Age (for results see: http://lydiniai.lt/). The large-scale, multi-period legacy dataset study of Pollard et al. (2015) shows that pure copper was as inaccessible as pure brass in northwest Europe in the late 5th/early 6th century (Kershaw and Merkel 2024). The same process of zinc increase identified in the pXRF spectrometry analysis is evident in the analysis carried out on Lithuanian archaeological material (Bliujienė et al. 2023) (Fig. 8). This confirms once again that the chemical composition of copper alloys over time has followed the same rhythm, both in areas with non-ferrous ore mines and in those with easy access to them due to their geopolitical position. It should be noted, however, that for a variety of reasons some ore deposits were closed. Many of the ore deposits continued to operate and, of course, new ores were discovered. As a result, the routes along which non-ferrous metals moved changed.

3.2. Trace copper alloy elements and deliberate use of mutability

In addition to providing information on the origin of copper, isotope ratios may also offer insight into the incorporation of lead during manufacturing. However, it is important to consider the potential for alterations due to the mixing of alloys by craftspeople. Therefore, the copper groups can be used to detect the recycling of alloys and to understand the mutability process in archaeometallurgical analyses. The copper groups are based on the thermodynamic properties of the four trace elements or impurities and focus on information derived primarily from the copper ore source(s), which may potentially be altered by subsequent human manipulation of the metal (Bray et al. 2015; Pollard et al. 2018, p. 2; Radivojević et al. 2019). Whatever the effect of mutability on the chemical character of ancient copper alloys, the process began with the invention of the first non-ferrous metal alloys in the Bronze Age.

Specifically, arsenic (As) is volatile, while antimony (Sb) is slightly less volatile, which is the opposite of nickel (Ni) and silver (Ag) which are stable in thermodynamic conditions during the molten copper oxidising process. Therefore, these trace elements are the keys for establishing copper groups. All possible combinations of presence/absence for four elements give 16 possible copper groups (Bray et al. 2015; Bray 2020; 2022; Pollard et al. 2018, pp. 85–97, Table 1; Radivojević et al. 2019). Copper groups described using the same set of elements (and the same cut-off values) for all analyses came to be the universal approach to the comparison of copper alloys from different times and different areas in Eurasia (Pollard et al. 2018, p. 86).

The Lithuanian archaeological material analysed contained less than <0.1 wt% of the major impurities (As, Sb, Ag and Ni), which does not allow classification into all 16 copper groups (see Table 3). In the Roman and Migration Periods, and even in the Viking Age, three main trends emerged among the identified copper groups. Firstly, in all periods, regardless of the number of artefacts whose elemental composition has been analysed, the predominant copper alloys are those of the first group, with major impurities of As, Sb, Ag and Ni below >0.1 wt%. Therefore, the second trend is the domination of 3, 4 and 7 as more resistant during re-melting copper groups were dominant in the Lithuanian archaeological material analysed. These copper groups in their elemental composition have antimony (Sb) and silver (Ag) as more resistant to re-melting impurities. The third trend shows that the composition of copper groups is also an agreement with the attitude of existing multiple re-melting of non-ferrous metals.

Thus, optical emission spectrometry (OES) analysis carried out in Lithuania between 1970 and 1984 (Merkevičius 1984) and current large-scale analysis by pXRF, as well as 208 samples analysed by ICP-MS shows similar results and demonstrate in the overwhelming majority of archaeological finds and samples studied, wherein these elements account for less than 0.1wt%. In addition, bismuth (Bi) and cobalt (Co) can be extremely useful in distinguishing non-ferrous metals from different ore deposits, but analysis shows that they are below the detection limits.

The chemical copper alloy composition analysis presented by the authors was carried out by the ICP-MS method and

Table 3. The copper groups defined by the presence/absence of the trace elements: arsenic (As), antimony (Sb), nickel (Ni) silver (Ag). 'Presence' is usually taken as greater than 0.1%. CG – copper group; Σ – total number of finds studied in groups (table prepared after Pollard et al. 2018, p. 86, Table 1).

CG	**As**	**Sb**	**Ag**	**Ni**	**Σ**	**AsSbAgNi**	**Description**	**Shorthand**
ROMAN IRON AGE								
1	<0.1%	<0.1%	<0.1%	<0.1%	56	NNNN	Copper alloy	Copper alloy
2	>0.1%	<0.1%	<0.1%	<0.1%	3	YNNN	Cu+As	CuAs
3	<0.1%	>0.1%	<0.1%	<0.1%	57	NYNN	Cu+Sb	CuSb
4	<0.1%	<0.1%	>0.1%	<0.1%	19	NNYN	Cu+Ag	CuAg
5	-	-	-	-	-	-	-	-
6	0.1%	0.1%	<0.1%	<0.1%	16	YYNN	Cu+As,Sb	CuAsSb
7	<0.1%	>0.1%	>0.1%	<0.1%	23	NYYN	Cu+Sb,Ag	CuSbAg
8								
9	>0.1%	<0.1%	>0.1%	<0.1%	5	YNYN	Cu+As+Ag	CuAsAg
10	-	-	-	-	-	-	-	-
11	-	-	-	-	-	-	-	-
12	>0.1%	>0.1%	>0.1%	<0.1%	11	YYYN	Cu+As,Sb,Ag	CuAsSbAg
13								
14	>0.1%	>0.1%	<0.1%	>0.1%	3	YYNY	Cu+As,Sb,Ni	CuAsSbNi
15	-	-	-	-	-	-	-	-
16	-	-	-	-	-	-	-	-
CG	**As**	**Sb**	**Ag**	**Ni**	**Σ**	**AsSbAgNi**	**Description**	**Shorthand**
MIGRATION PERIOD								
1	<0.1%	<0.1%	<0.1%	<0.1%	5	NNNN	Copper alloy	Copper alloy
3	<0.1%	>0.1%	<0.1%	<0.1%	1	NYNN	Cu+Sb	CuSb
5	<0.1%	<0.1%	<0.1%	<0.1%	1	NNNY	Cu+Ni	CuNi
6	0.1%	0.1%	<0.1%	<0.1%	2	YYNN	Cu+As,Sb	CuAsSb
9	>0.1%	<0.1%	>0.1%	<0.1%	3	YNYN	Cu+As+Ag	CuAsAg
12	>0.1%	>0.1%	>0.1%	<0.1%	6	YYYN	Cu+As,Sb,Ag	CuAsSbAg
14	>0.1%	>0.1%	<0.1%	>0.1%	1	YYNY	Cu+As,Sb,Ni	CuAsSbNi
16	>0.1%	>0.1%	>0.1%	>0.1%	1	YYYY	Cu+As,Sb,Ag,Ni	CuAs,Sb,Ag,Ni
CG	**As**	**Sb**	**Ag**	**Ni**	**Σ**	**AsSbAgNi**	**Description**	**Shorthand**
VIKING AGE								
1	<0.1%	<0.1%	<0.1%	<0.1%	2	NNNN	Copper alloy	Copper alloy
2	>0.1%	<0.1%	<0.1%	<0.1%	2	YNNN	Cu+As	CuAs
3	<0.1%	>0.1%	<0.1%	<0.1%	1	NYNN	Cu+Sb	CuSb
6	0.1%	0.1%	<0.1%	<0.1%	1	YYNN	Cu+As,Sb	CuAsSb
7	<0.1%	>0.1%	>0.1%	<0.1%	2	NYYN	Cu+Sb,Ag	CuSbAg
11	>0.1%	<0.1%	0.1%	<0.1%	1	YNYN	Cu+As,Ag	CuAs,Ag
12	>0.1%	>0.1%	>0.1%	<0.1%	2	YYYN	Cu+As,Sb,Ag	CuAsSbAg
14	>0.1%	>0.1%	<0.1%	>0.1%	1	YYNY	Cu+As,Sb,Ni	CuAsSbNi
15	>0.1%	<0.1%	>0.1%	>0.1%	1	YNYY	Cu+As,Ag,Ni	CuAs,Ag,Ni

the obtained results allow the classification of the alloys into types, defined on intentional addition of metals by craftspeople to modify the characteristics of the material (e.g. for fluidity in casting, colour, hardness, etc., or perhaps to give additional symbolic significance). But subsequent mixing and recycling might move the assemblage away from the originally-designed alloy compositions. At the same time, it must be stressed that the theoretical approaches to grouping copper alloys into the types vary somewhat, as they are based on the slightly differently understanding of relationships between the main alloying elements Zn, Sn and Pb to copper (e.g. Riederer 1984; Bayley and Butcher 2004, pp. 12–13, Table 5; Pollard et al. 2018, p. 117, Table 1; Radivojević et al. 2019; Roxburgh 2023, Fig. 3; van der Meulen-van der Veen 2023).

It is widely accepted that recycling would cause some chemical alteration of the metal but, as with all archaeometallurgical processes, there are hazards which are difficult to avoid. It is clear that more than 0.1% of the alloys can be grouped into the copper groups using modern spectrometric techniques (for the results, see Table 3). It is true that the copper groups have an aspect of universality in space and time. The remelting process then presents the reasonable question of how to estimate the amount of metal lost during each occurrence of remelting, recycling and addition of scrap metal, if we take into account an estimated loss of 5% during metal remelting (Radivojević et al. 2019, Fig. 9). Furthermore, constant metal mixing copper alloys would have homogenised the metal stock, which is what we see in the Lithuanian archaeological evidence after lead isotope ratio analysis (Figs. 2; 4; 5 and 7). One more danger in interpreting the mutability process is highlighted by mixed trace element signatures of different ore provenance, for example fahlore and chalcopyrite copper (Pernicka et al. 2016; Radivojević et al. 2019, Fig. 10).

The Lithuanian artefacts under analysis are not part of all 16 possible copper groups. Although some of the artefacts analysed contained less than 0.1 wt% As, Sb, Ni and Ag. The low levels of arsenic and antimony found in the copper alloys suggests a long history of recycling. It is reasonable to assume that this has occurred because both metals are volatile. Other trace elements, Ni and Ag, which are thermodynamically stable in molten copper, are also of low values level (for the results, see Table 3). It is therefore possible for raw metals from different sources to be melted together. Due to the lack of research, it is difficult to say how many of the Lithuanian finds were made from recycled alloys. It is possible that the situation was similar to that in Britain from the Roman Iron Age to the end of the Anglo-Saxon period. About 250 years after the collapse of the Roman occupation in Britain to the end of the Early Saxon period, at least 75% of the copper alloy in circulation was still recycled Roman objects (Pollard et al. 2015). It should be noted, however, that the Lithuanian archaeological evidence was replenished with brass raw material, whether in the form of alloys, scrap or artefacts. The influx of brass is particularly noticeable at the beginning of the Roman Iron Age and during the Early Migration period, i.e. during the great epochal transformations (Fig. 8).

3.3. Craftsmanship tradition and copper alloy properties

Judging by the weight and the quantity of jewellery and other grave goods that were taken to the afterlife, copper alloys must have travelled to the eastern Baltic region and Lithuania in a fluctuating but continuous flow. In the context of the craftsmanship tradition, copper alloys were closely connected to the notion of metal value, their functional properties, and aesthetic and technological qualities. The chemical composition of copper alloys determines the main characteristics of the alloy, primarily the colour, the fluidity necessary for casting, and the malleability and hardness necessary for forging, etc., but also the shapes of the artefacts, which were imposed by societies and reflect the socio-technological context of production (Bliujienė 2023 pp. 305–321). For the period covered by the article, it is not only theoretical considerations that we have of craftsmen's understanding of the main alloys' functional properties (flexibility, fluidity, colour etc.) when recasting alloys from metal scrap waste. A good example is the Viking Age, when bronze and bronze-based alloys accounted for almost 67 wt% of the Lithuanian total based on X-ray Fluorescence analysis (Fig. 8). Although ICP-MS analyses from the Viking Age are limited, the tendency remains the same, with an increasing level of bronze and bronze-based alloys. Lead-based alloys also encouraged an increase in cast jewellery production (mostly dress-pins, some types of brooches). Casting requires more fluid alloys and this in turn influenced the simplification the technological processes (e.g. technologically complicated Viking Age brooches which towards the end of their development started to be cast in one casting mould). In addition, the Balts liked big, heavy ornaments, and the properties of lead were suitable for the production of such jewellery, so this means that lead was constantly added during casting. It is also possible that lead from Swedish and to an extent Finnish ores found its way to the eastern Baltic Sea region. The high-lead brasses were the dominant alloy used for the production of the Viking Age Gotlandic box brooches and the crossbow animal head brooches (Forshell 1992, pp. 61–63).

It seems, however, that the importance of the colour of copper alloys is overestimated. In fact, the colour of gold, due to its high zinc content of 30 wt%, was essentially equivalent to the *aurichalcum* used by the Romans. In the eastern Baltic, the zinc content of copper alloys rarely reaches 25 wt%, and even small amounts of tin and lead change the colour and quality of the alloys. As mentioned above, non-ferrous alloys in the eastern Baltic region have followed the same pattern as in the rest of Europe. In addition, as lead isotope analysis and the concept of mutability perfectly illustrate, colour as a property of alloys is altered by the craftsmen's manipulation. In terms of access to metal alloys in the eastern Baltic region, the technological capabilities of craftsmen may suggest that the choice of colour was a secondary consideration. The fact that the desired product was one shade or another of yellow was sufficient. Craftsmen, however, used different colours of copper alloy as far as they could, and such examples were recorded during the research. Some of the neck-ring bows and their conical ends may have a slightly different alloy composition, as ICP-MS shows some differences in the lead amount, which could have had an effect on the properties and colour of the alloy on pieces of this jewel-

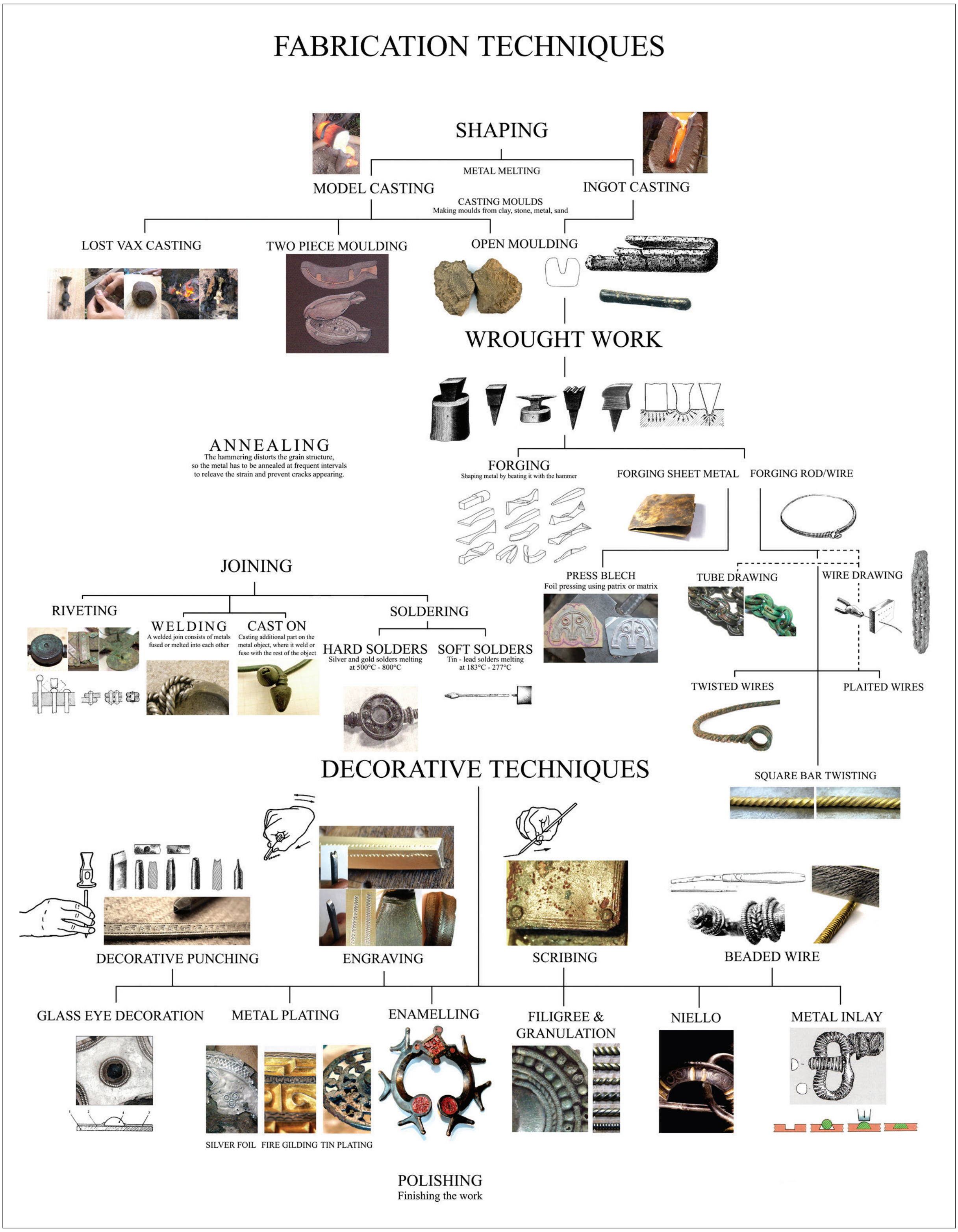

Figure 9. Technologies for the working of non-ferrous metals and techniques of surface treatment used in Lithuania from the second half of the 1st century to the 9th century AD (figure prepared by Babenskas).

lery.[7] However, it can be argued that non-ferrous metals were one of the elements of the region's polychrome style, both in terms of the technical skills of the craftsmen and in terms of aesthetics, including yellow alloys of whatever shade.

Other metals that, together with copper alloys, created the polychrome style of the region's wares were silver and tin (sometimes lead). These were metals with different shades

[7] Neck-rings from Ibutoniai (Ibutoniai ID: Ibu.536.1a-b) and Daujėnai (Baluškiai) burial grounds (ID: Dauj 526.16). The lead content reaches 3.27-3.30 in Ibutoniai neck-rings and 4.52-4.9 and 2.18 wt% in Daujėnai (Baluškiai) neck-rings (for ICP-MS values, are available from: http://lydiniai.lt/).

of silver. Of course, these are metal alloys of different values, but by the end of the 2nd century, silver jewellery of local manufacture was already appearing in Lithuania, and silver raw material must have reached the region. At the same time, the surface of the jewellery was coated with tin (pewter). Although this technology was widespread throughout Europe, what is important is that a clear distinction was made between the values of silver and pewter. Baltic craftsmen started making solders from different ratios of tin-lead, tin and lead in the late 2nd and early 3rd centuries AD (Volkaitė-Kulikauskienė and Jankauskas 1992, Fig. 2; Bliujienė et al. 2021a; Bliujienė et al. 2021b). A distinctive feature of the region's polychrome style is the blue glass inlays that decorate the silver plates soldered onto the copper alloy surfaces of artefacts. This feature is characteristic of the entire period covered by the article. In the Roman Iron Age, the polychrome style is complemented by enamel jewellery. This is a special technique called champlevé (for production technologies and treatment of artefacts, see Fig. 9). Finally, the technological, economic and social contexts in which jewellery is made matter greatly, as they ultimately give it both aesthetic value and the ability to demonstrate social status

Conclusions

208Pb/206Pb versus 207Pb/206Pb and 206Pb/204 versus 207Pb/206Pb isotope ratios shows a sufficiently wide dispersion of lead isotope ratios. In the Roman Iron Age, lead isotope ratios show that the copper alloys could have come from ore deposits in the region of Trentino-Bolzano (Italian Alps), the Great Orme in Wales (British Isles), the Slovak Ore Mountains (Hron Valley) or the Mitterberg area (Austrian Alps). It could be that the Lavrion mines in the southern Greece, and Spain were a source of non-ferrous alloys in the Roman Iron Age.

Due to the limited number of Migration Period and Viking Age finds that have been analysed, it is not possible to say with certainty which Eurasian metal ore deposits are the source of the metal flows in the eastern Baltic region. Isotope signals from the Mitterberg (Austrian Alps), southern Greece (Lavrion) and Spain were no longer recorded in surveys during this period.

In the Viking Age, it is possible that the flow of metal to the eastern Baltic originated from the Balkans, particularly the Bor and and Majdanpek metallogenic zones in Serbia, or even Andalusia (southern Spain). The changing geopolitical situation in Eurasia naturally altered the direction of long-distance trade networks and led to changes in ore sources. Changes in the ratios of the lead isotopes analysed capture this situation.

The repeated recycling of copper alloys (mutability process), the addition of scrap materials with varying compositions, and the mixing of lead from different geological ore deposits naturally alter the original chemical composition of the alloy. The continuous recycling and alteration of materials pose challenges in identifying the original connections between the regions of origin of copper alloy ores in southern Eurasia and the users of these raw materials in the eastern Baltic Sea region.

Together with lead isotope ratio analysis, investigation of copper alloy types, copper groups, and metal working technological development fundamentally changes the idea of a linear exchange of non-ferrous metals. The analysis carried out in this research has enabled the identification of the provenance and dissemination of non-ferrous metal raw materials (alloys and scrap metal) as part of the European exchange network at local (the present territory of Lithuania), regional (the eastern Baltic region), and trans-European levels. However, these networks were subject to constant change. This was due to the European geopolitical situation.

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Abbreviations

Archaeol. Anthropol. Sci. – Archaeological and Anthropological Sciences

Archaeol. Austriaca – Archaeologia Austriaca

Archaeol. Baltica – Archaeologia Baltica

ATL - Archeolgoginiai tyrinėjimai Lietuvoje/ Archaeological Investigations in Lithuania

Chem. Geol. – Chemical Geology

Eur. J. Archaeol. – European Journal of Archaeology

Geochem. Geophys. Geosyst. – Geochemistry, Geophysics, Geosystems

Int. J. Mass Spectrom. – International Journal of Mass Spectrometry

J. Anal. At. Spectrom. – Journal of Analytical Atomic Spectrometry

J. Archaeol. Res. – Journal of Archaeological Research

J. Archaeol. Sci – Journal of Archaeological Science

J. Archaeol. Sci.: Rep. – Journal of Archaeological Science: Reports

Lietuvos archeol. – Lietuvos archeologija

Ore Geol. Rev. – Ore Geology Reviews

Period. Mineral. – Periodico di Mineralogia

Praehist. Z. – Praehistorische Zeitschrift

Precambrian Res. – Precambrian Research

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Supplements

Supplement 1. Inductively Coupled Plasma Mass Spectrometry (ICP-MS) lead isotope ratios of artefacts on the metal core. All artefacts description, chronology and photographs are available from: http://lydiniai.lt/; Bliujienė et al. 2023). The types of copper alloys determined based on traditional typology (Pollard et al. 2018, p. 116, Fig. 1).

Sample ID	Site	Artefact	Alloy type	Sample weight (gr)	Pb wt%	Pb isotope ratios				
						208/204	207/204	206/204	208/206	207/206
ROMAN IRON AGE										
1.Bil.52	Bilioniai	Fibula	Gun-metal	0,010	4,995	38,508	15,718	18,184	2,118	0,864
2.Bil.56	Bilioniai	Roman coin	Brass	0,003	0,55	39,004	15,747	18,572	2,100	0,848
3.Bil.60b	Bilioniai	Pendant	Gun-metal	0,003	2,237	38,707	15,694	18,442	2,099	0,851
4.Bil.75	Bilioniai	Roman coin	Bronze	0,004	5,885	39,012	15,731	18,547	2,104	0,848
5.Bil.76b	Bilioniai	Pendant	Brass	0,008	0,976	38,471	15,645	18,334	2,098	0,853
6.Bil.78	Bilioniai	Roman coin	Brass	0,005	0,3	39,032	15,741	18,538	2,106	0,849
7.Bil.82	Bilioniai	Roman coin	Brass	0,003	0,559	39,338	15,848	18,666	2,108	0,849
8.Bil.85b	Bilioniai	Roman coin	Brass	0,005	5,727	38,501	15,659	18,440	2,088	0,849
9.Bil.89b	Bilioniai	Roman coin	Leaded bronze	0,003	12,182	38,861	15,791	18,477	2,103	0,855
10.Bil.93	Bilioniai	Roman coin	Bronze	0,002	5,833	38,858	15,654	18,477	2,103	0,847
11.Bil.94b	Bilioniai	Roman coin	Leaded bronze	0,006	13,771	38,797	15,766	18,528	2,094	0,851
12.Par.721-5	Paragaudis	Neck-ring (bow)	Brass	0,008	0,063	38,730	15,662	18,366	2,109	0,853
13.Par.721-19	Paragaudis	Neck-ring (trumpet)	Brass	0,011	0,041	39,059	15,685	18,500	2,112	0,848
14.Par.721-20	Paragaudis	Fibula	Brass	0,004	0,074	39,925	16,084	18,876	2,115	0,852
15.Par.721-34	Paragaudis	Neck-ring (bow)	Brass	0,004	0,274	39,138	15,733	18,591	2,105	0,846
16.Par.721-49	Paragaudis	Fibula	Brass	0,004	0,286	38,812	15,657	18,435	2,106	0,849
17.Par.721-68	Paragaudis	Fibula	Brass	0,009	0,309	38,976	15,711	18,523	2,104	0,848
18.Par.721-69	Paragaudis	Bracelet	Brass	0,004	0,113	40,888	16,387	19,404	2,107	0,844
19.Par.721-70-1	Paragaudis	Neck-ring (bow)	Brass	0,013	0,054	39,168	15,732	18,601	2,106	0,846
20.Par.721-70-2		Neck-ring (trumpet)	Brass	0,004	0,081	39,494	15,710	18,626	2,121	0,844
21.Par.721-70-3		Neck-ring (bow)	Brass	0,004	0,061	39,302	15,788	18,561	2,118	0,851
22.Par.721-72	Paragaudis	Bracelet	Brass	0,019	1,225	39,038	15,643	18,536	2,106	0,844
23.Par.721-73	Paragaudis	Bracelet	Brass	0,010	0,194	38,775	15,689	18,439	2,103	0,851
24.Par.721-81	Paragaudis	Fibula	Brass	0,014	0,21	38,991	15,716	18,503	2,107	0,849
25.Par.721-86	Paragaudis	Fibula	Brass/ Gun-metal	0,010	0,099	38,681	15,630	18,433	2,099	0,848
26.Par.721-93	Paragaudis	Fibula	Brass/ Gun-metal	0,006	0,114	39,118	15,817	18,536	2,110	0,853

Sample ID	Site	Artefact	Alloy type	Sample weight (gr)	Pb wt%	Pb isotope ratios				
						208/204	207/204	206/204	208/206	207/206
27.Par.721-94	Paragaudis	Neck-ring (bow)	Brass	0,011	0,163	39,076	15,738	18,641	2,096	0,844
28.Par.721-123a	Paragaudis	Bracelet	Brass	0,006	0,081	38,775	15,619	18,439	2,103	0,847
29.Par.721-128	Paragaudis	Temple ornament	Brass/ Gunmetal	0,005	0,183	39,024	15,659	18,598	2,098	0,842
30.Par.721-144	Paragaudis	Bracelet	Brass	0,015	0,419	39,070	15,757	18,523	2,109	0,851
31.Par.721-147	Paragaudis	Fibula	Brass	0,009	0,083	39,127	15,674	18,699	2,093	0,838
32.Par.721-149a	Paragaudis	Neck-ring (bow)	Brass	0,025	0,063	39,067	15,780	18,409	2,122	0,857
33.Par.721-149b	Paragaudis	Neck-ring (bow)	Brass	0,023	0,105	38,635	15,648	18,388	2,101	0,851
34.Par.721-150	Paragaudis	Fibula	Brass	0,012	0,139	39,203	15,755	18,690	2,098	0,843
35.Par.721-151	Paragaudis	Bracelet	Brass	0,013	0,145	39,210	15,704	18,654	2,102	0,842
36.Par.721-152	Paragaudis	Bracelet	Brass	0,009	0,037	38,925	15,645	18,401	2,116	0,850
37.Par.721-156	Paragaudis	Dress-pin	Brass	0,008	0,242	38,860	15,684	18,452	2,106	0,850
38.Par.721-157	Paragaudis	Neck-ring (bow)	Brass	0,021	0,156	38,742	15,641	18,388	2,107	0,851
39.Par.721-167	Paragaudis	Fibula	Brass	0,020	0,063	38,779	15,675	18,462	2,101	0,849
40.Par.721-182	Paragaudis	Neck-ring (bow)	Gunmetal	0,022	0,993	38,342	15,622	18,416	2,082	0,848
41.Paju.572-25a	Pajūralis (Skerdy-nai)	Neck-ring (bow)	Brass/ Gunmetal	0,042	3,326	38,732	15,693	18,495	2,094	0,848
42.Paju.572-4	Pajūralis (Skerdy-nai)	Neck-ring (cone)	Brass/ Gunmetal	0,096	4,163	38,775	15,597	18,443	2,103	0,846
43.Jag.16-14	Jagminiškė	Massive spiral	Brass/ Gunmetal	0,020	0,279	38,787	15,645	18,453	2,102	0,848
44.Jag.16-15	Jagminiškė	Dress-pin	Brass	0,031	0,113	39,145	15,834	18,628	2,102	0,850
45.Jag.16-16	Jagminiškė	Dress -pin	Brass	0,035	0,226	39,185	15,776	18,633	2,103	0,847
46.Jag.16-17	Jagminiškė	Dress -pin	Bronze/ Gunmetal	0,022	0,112	39,214	15,803	18,591	2,109	0,850
47.Jag.16-19	Jagminiškė	Dress -pin	Brass/ Gunmetal	0,025	0,201	39,188	15,761	18,614	2,105	0,847
48.Jag.16-20	Jagminiškė	Dress- pin	Bras	0,023	0,276	38,932	15,727	18,596	2,094	0,846
49.Jag.16-21	Jagminiškė	Dress- pin	Brass	0,017	0,201	38,694	15,649	18,408	2,102	0,850
50.Pab.462-4	Pabariukai (Eiman čiai)	Bracelet	Gunmetal	0,053	3,019	38,149	15,571	18,348	2,079	0,849
51.Mau.11596	Maudžio-rai	Fibula	Brass	0,084	0,814	38,556	15,580	18,546	2,079	0,840
52.Mau.11469a	Maudžio-rai	Bridle: horsehead pendant	Brass	0,049	2,619	38,219	15,560	18,294	2,089	0,851
53.Mau.11522	Maudžio-rai	Buckle	Brass	0,083	1,362	38,029	15,550	18,290	2,079	0,850
54.Mau.11512	Maudžio-rai	Bracelet	Brass	0,054	1,885	38,772	15,691	18,521	2,094	0,847
55.Mau. 13454	Maudžio-rai	Bracelet	Brass	0,057	1,441	38,109	15,547	18,244	2,089	0,852
56.Vie.620-24	Vienragiai	Bracelet	Brass	0,056	0,187	38,709	15,635	18,446	2,099	0,848

Sample ID	Site	Artefact	Alloy type	Sample weight (gr)	Pb wt%	Pb isotope ratios				
						208/204	207/204	206/204	208/206	207/206
57.Vie.620-18	Vienragiai	Bracelet	Brass	0,082	0,196	38,654	15,670	18,447	2,096	0,849
58.Vie.620-17	Vienragiai	Bracelet	Brass	0,098	0,512	38,534	15,685	18,525	2,080	0,847
59.Vie.620-19	Vienragiai	Bracelet	Brass	0,039	0,192	38,739	15,687	18,404	2,105	0,852
60.Prib.641-17	Pribitka	Neck-ring (bow)	Brass/ Gunmetal	0,046	0,287	38,885	15,698	18,512	2,101	0,848
61.Prib.641-2	Pribitka	Dress-pin	Brass	0,065	0,129	38,870	15,713	18,480	2,103	0,850
62.Prib.641-12	Pribitka	Fibula	Brass	0,047	0,094	39,033	15,765	18,543	2,105	0,850
63.Prib.641-6	Pribitka	Bracelet	Brass	0,070	0,129	39,025	15,728	18,557	2,103	0,848
64.Prib.641-23	Pribitka	Fibula	Gunmetal	0,088	0,104	38,709	15,660	18,446	2,099	0,849
65.Prib.641-11	Pribitka	Fibula	Brass	0,064	0,179	38,688	15,650	18,453	2,097	0,848
66.Bar.820-1a	Barzūnai	Fibula	Brass	0,016	0,175	38,712	15,605	18,434	2,100	0,846
67.Bar.2-1	Barzūnai	Bracelet	Gunmetal	0,007	0,254	39,285	15,884	18,713	2,099	0,849
68.Bar.3-2	Barzūnai	Fibula	Bronze	0,014	0,33	38,927	15,700	18,532	2,101	0,847
69.Bar.14-1b	Barzūnai	Bracelet	Brass/ Gunmetal	0,012	0,226	38,976	15,723	18,532	2,103	0,848
70.Bar.14-2a	Barzūnai	Bead	Brass/ Gunmetal	0,090	0,293	38,737	15,646	18,422	2,103	0,849
71.Bar.14-3a	Barzūnai	Bead	Brass/ Gunmetal	0,017	0,319	38,942	15,667	18,508	2,104	0,847
72.Bar.14-5b	Barzūnai	Bracelet	Brass/ Gunmetal	0,015	0,901	39,305	15,775	18,746	2,097	0,841
73.Bar.21-3	Barzūnai	Bracelet	Bronze	0,010	6,574	38,543	15,660	18,411	2,094	0,851
74.Bar.20-1	Barzūnai	Neck-ring (cone)	Bronze	0,043	6,681	37,852	15,522	18,241	2,075	0,851
75.Bar.21-1b	Barzūnai	Neck-ring (bow)	Brass/ Gunmetal	0,026	3,044	38,780	15,649	18,457	2,101	0,848
76.Bar.24-1	Barzūnai	Bracelet	Brass	0,138	0,253	39,099	15,720	18,546	2,108	0,848
77.Str.859-4	Strazdai (Ječiškės)	Fibula	Brass/ Gunmetal	0,018	1,128	38,962	15,723	18,590	2,096	0,846
78.Str.859-22	Strazdai (Ječiškės)	Fibula	Brass/ Gunmetal	0,013	1,033	38,774	15,697	18,508	2,095	0,848
79.Str.859-23	Strazdai (Ječiškės)	Fibula	Brass	0,013	0,696	38,634	15,711	18,318	2,109	0,858
80.Str.859-31	Strazdai (Ječiškės)	Fibula	Brass	0,011	0,119	39,056	15,807	18,534	2,107	0,853
81.Str.859-33b	Strazdai (Ječiškės)	Bracele	Brass	0,019	0,181	38,744	15,686	18,439	2,101	0,851
82.Ser.441-21	Seredžius II	Neck-ring (bow)	Brass	0,052	0,238	38,904	15,671	18,456	2,108	0,849
83.Nik.832-1	Nikėlai	Fibula	Brass	0,020	0,079	39,575	15,958	18,809	2,104	0,848
84.Nik.832-3	Nikėlai	Fibula	Brass	0,043	0,145	39,266	15,817	18,687	2,101	0,846
85.Nik.832-6	Nikėlai	Briles: eight shaped spacer	Brass	0,037	0,161	39,088	15,759	18,670	2,094	0,844
86.Fli-1-1	Fliorencija	Fibula	Brass	0,006	1,279	38,871	15,686	18,525	2,099	0,847
87.Tau.1-1b	Tautušiai	Fibula	Bronze	0,011	0,207	38,754	15,689	18,422	2,104	0,852
88.Tau.2-1b	Tautušiai	Fibula	Brass/ Gunmetal	0,003	0,157	39,044	15,776	18,570	2,103	0,850
89.Juk.1-1	Jūkainiai	Pendant	Brass/ Gunmetal	0,015	0,226	38,903	15,700	18,464	2,107	0,850

Sample ID	Site	Artefact	Alloy type	Sample weight (gr)	Pb wt%	Pb isotope ratios				
						208/204	207/204	206/204	208/206	207/206
90.Anu.1-1	Anulynas	Fibula	Brass	0,013	0,091	38,612	15,656	18,334	2,106	0,854
91.Nem.1-1	Nemakš-čiai	Fibula	Gunmetal	0,008	0,173	39,062	15,703	18,467	2,115	0,850
92.San.1588.2	Sandrau-siškė (Akmenė)	Neck-ring (bow)	Brass	0,049	0,174	38,962	15,727	18,586	2,096	0,846
93.San.1588.5	Sandrau-siškė (Akmenė	Neck-ring (terminal)	Brass	0,031	0,113	38,404	15,531	18,316	2,097	0,848
94.San.1588.10	Sandrau-siškė (Akmenė)	Fibula	Brass	0,096	0,132	38,582	15,684	18,328	2,105	0,856
95.San.1588.18	Sandrau-siškė (Akmenė)	Fibula	Brass	0,039	0,129	38,923	15,774	18,549	2,098	0,850
96.San.1588.19	Sandrau-siškė (Akmenė)	Fibula	Brass	0,024	0,1	39,026	15,900	18,609	2,097	0,854
97.San.1588.23	Sandrau-siškė (Akmenė)	Dress-pin	Brass	0,052	0,301	38,994	15,823	18,636	2,093	0,849
98.San.1588.25	Sandrau-siškė (Akmenė)	Bracelet	Brass	0,043	0,16	38,705	15,705	18,424	2,101	0,852
99.San.1588.26	Sandraus-iškė (Akmenė)	Bracelet	Brass	0,069	0,092	39,005	15,772	18,537	2,104	0,851
100.Kru.86.9	Krūvandai	Bracelet	Brass	0,050	0,539	38,594	15,666	18,448	2,092	0,849
101.Kru.86.10	Krūvandai	Bracelet	Brass	0,049	0,535	38,863	15,700	18,500	2,101	0,849
102.Pas.657.7	Paštuva	Fibula	Brass	0,072	0,153	38,509	15,623	18,348	2,099	0,851
103.Vidu.383-6	**C Lithuania	Neck-ring (bow)	Brass	0,019	1,697	38,968	15,685	18,745	2,079	0,837
104.Vidu.383-11	**C Lithuania	Neck-ring (bow)	Brass	0,013	0,142	38,580	15,657	18,279	2,111	0,857
105.Vidu.383-13	**C Lithuania	Neck-ring (bow)	Brass	0,013	0,123	38,791	15,664	18,364	2,112	0,853
106.Vidu.383-29	*C Lithuania	Fibula	Brass	0,013	0,245	38,882	15,725	18,532	2,098	0,848
107.Vidu.383-43	*C Lithuania	Bracelet	Brass	0,018	0,164	38,822	15,668	18,473	2,102	0,848
108.Vidu.383-59a	*C Lithuania	Bracelet	Gunmetal	0,005	0,296	38,709	15,614	18,453	2,098	0,846
109.Vidu.383-67b	*C Lithuania	Bracelet	Brass	0,008	0,199	38,921	15,712	18,557	2,098	0,847
110.Vidu.383-69b	*C Lithuania	Bracelet	Brass	0,009	0,912	38,610	15,610	18,427	2,095	0,847
111.Ked.34-2	Kėdainiai	Neck-ring (bow)	Brass	0,013	0,478	39,121	15,719	18,572	2,107	0,846
112.Ked.34-3	Kėdainiai	Neck-ring (bow)	Brass	0,021	0,315	39,003	15,749	18,588	2,098	0,847
113.Ked.34-4	Kėdainiai	Bracelet	Brass	0,020	3,404	38,462	15,627	18,434	2,087	0,848
114.Ked.34-7	Kėdainiai	Bracelet	Brass/ Gunmetal	0,011	0,149	39,028	15,741	18,602	2,098	0,846

Sample ID	Site	Artefact	Alloy type	Sample weight (gr)	Pb wt%	Pb isotope ratios				
						208/204	207/204	206/204	208/206	207/206
115.Gla.830-1	Glaušiai	Neck-ring (terminal)	Brass	0,042	1,07	38,393	15,612	18,444	2,082	0,846
116.Paj.554-8	Pajuostis	Bracelet	Brass	0,032	0,133	39,034	15,739	18,524	2,107	0,850
117.Paj.554-23	Pajuostis	Neck-ring	Brass	0,031	0,156	38,664	15,625	18,396	2,102	0,849
118.Paj.554-24	Pajuostis	Dress-pin	Brass	0,055	0,103	38,850	15,674	18,513	2,099	0,847
119.Paj.554-132	Pajuostis	Bracelet	Brass/ Gunmetal	0,044	2,07	38,615	15,631	18,403	2,098	0,849
120.Gab.479.4	Gabulai	Neck-ring (bow)	Brass	0,109	0,54	38,196	15,589	18,384	2,078	0,848
121.Gab.479.4	Gabulai	Bracelet	Brass	0,082	1,162	39,072	15,729	18,695	2,090	0,841
122.Ibu.536.1a	Ibutoniai	Neck-ring (cone)	Brass/ Gunmetal	0,076	3,052	39,155	15,704	18,551	2,111	0,846
123.Ibu.536.1b	Ibutoniai	Neck-ring (bow)	Brass/ Gunmetal	0,053	3,091	38,234	15,587	18,406	2,077	0,847
124.Zil.1-1	Žiliai-Šemetaičiai	Ring	Brass	0,004	2,238	38,838	15,760	18,599	2,088	0,847
125.Rag.1-1	Raganiai	Temple ornament	Brass/ Gunmetal	0,005	1,098	39,328	15,807	18,809	2,091	0,840
126. Visd.670.13	Visdergiai (Papelkiai)	Fibula	Brass	0,057	0,116	38,364	15,553	18,260	2,101	0,852
127. Visd.670.17	Visdergiai (Papelkiai)	Fibula	Brass	0,041	0,36	38,546	15,671	18,366	2,099	0,853
128.Rai.23.4	Rainiai	Bracelet	Gunmetal	0,095	3,483	38,944	15,663	18,531	2,102	0,845
129.Zad.4861a	Žaduvėnai	Dress-pin	Gunmetal	0,046	2,94	38,175	15,587	18,352	2,080	0,849
130.Ven.137-1	**Venta	Neck-ring (bow)	Gunmetal	0,037	2,739	38,681	15,647	18,444	2,097	0,848
131.Ven.137-3	**Venta	Bracelet	Leaded bronze	0,036	6,629	38,394	15,610	18,351	2,092	0,851
132.Jau.675-10	Jautakiai	Fibula	Brass	0,018	0,174	38,835	15,698	18,514	2,098	0,848
133.Jaun.537-4	Jauneikiai	Bracelet	Gunmetal	0,147	0,645	38,222	15,589	18,306	2,088	0,852
134.Man.1-1b	Mančeliai	Bracelet	Gunmetal	0,011	3,861	38,141	15,519	18,294	2,085	0,848
135.Gru.1-1	Gružos (Užulėnis)	Bracelet	Brass	0,013	0,754	38,842	15,681	18,467	2,103	0,849
136.Baj.93-3	Bajoriškiai	Neck-ring (trumpet)	Brass	0,012	0,082	38,785	15,627	18,281	2,122	0,855
137.Baj.93-6b	Bajoriškiai	Bracelet	Brass	0,018	0,11	38,932	15,745	18,492	2,106	0,851
138.Baj.93-7	Bajoriškiai	Bracelet	Brass	0,021	0,069	39,596	15,856	18,715	2,116	0,847
139.Baj.93-10	Bajoriškiai	Bracelet	Brass	0,020	0,14	38,844	15,686	18,473	2,103	0,849
140. Dauj.505.16-1	Daujėnai (Baluškiai)	Neck-ring (cone)	Brass/ Gunmetal	0,106	4,482	37,918	15,482	18,425	2,058	0,840
141. Dauj.505.16-2	Daujėnai (Baluškiai)	Neck-ring (bow)	Brass/ Gunmetal	0,081	4,576	38,134	15,532	18,532	2,060	0,839
142.Gri.33.1a	Grinkiškis	Neck-ring (bow)	Bronze	0,092	6,536	38,281	15,553	18,382	2,083	0,846
143.Gri.33.1b	Grinkiškis	Neck-ring (cone)	Bronze	0,101	5,881	38,588	15,598	18,393	2,098	0,848
144.Gri.33.2a	Grinkiškis	Neck-ring (bow)	Leaded gunmetal	0,101	5,881	38,588	15,598	18,393	2,098	0,848
145.Gri.33.2b	Grinkiškis	Neck-ring (cone)	Leaded gunmetal	0,076	4,645	38,868	15,640	18,488	2,102	0,846

Sample ID	Site	Artefact	Alloy type	Sample weight (gr)	Pb wt%	Pb isotope ratios				
						208/204	207/204	206/204	208/206	207/206
146.Ski721-1	Skėmiai (Skienai)	Bracelet	Brass	0,049	0,195	39,025	15,746	18,577	2,101	0,848
147.Sia.711-2	Šiaulaičiai	Bracelet	Brass	0,059	0,158	39,256	15,785	18,680	2,102	0,845
148.Kauk.500-495	Kaukai	Dress-pin	Leaded brass	0,043	12,467	38,407	15,533	18,289	2,100	0,849
149.Sla.103-9	Slaba-dėlė	Pendant	Brass	0,028	0,987	38,921	15,705	18,505	2,103	0,849
150.Sla.103-10	Slaba-dėlė	Pendant	Brass	0,018	0,516	39,052	15,719	18,564	2,104	0,847
151. SemG2.7976a	Semen-iškiai II	Raw metal	Brass	0,007	0,597	38,885	15,710	18,528	2,099	0,848
152. SemG2.7976b	Semeniš-kiai II	Raw metal	Brass	0,037	0,504	38,689	15,646	18,454	2,097	0,848
153.Vis.729-286	Visėt-iškės	Bracelet	Brass	0,018	0,242	39,144	15,761	18,597	2,105	0,847
154.Eik.76-8a	Eiko-tiškis	Neck-ring (cone)	Brass/ Gunmetal	0,068	6,403	38,166	15,553	18,356	2,079	0,847
155.Eik.76-8b	Eiko-tiškis	Neck-ring (bow)	Brass/ Gunmetal	0,105	3,464	38,918	15,673	18,582	2,095	0,843
156.Lel.81-1a	Leliūnai	Neck-ring (bow)	Brass/ Gunmetal	0,082	3,019	38,229	15,629	18,398	2,078	0,849
157.Lel.81-1b	Leliūnai	Neck-ring (cone)	Brass/ Gunmetal	0,083	2,997	37,960	15,554	18,303	2,074	0,850
158.Dro.179-14b	Drobūkš-čiai	Bracelet	Leaded brass	0,038	14,253	37,976	15,560	18,310	2,074	0,850
159.Rok.667-12	Rokėnai	Neck-ring (bow)	Gunmetal	0,042	3,834	38,387	15,557	18,358	2,091	0,847
MIGRATON PERIOD										
1. Uzp.55118	Užpelkiai	Clasp	Unreliable result	0,0723	1,04	38,723	15,644	18,476	2,096	0,847
2.Uzp.55000	Užpelkiai	Fibula	Bronze/ Gunmetal	0,086	3,540	39,048	15,696	18,574	2,102	0,845
3.Imb.523-14	Imbarė	Bracelet	Leaded brass	0,042	11,439	38,029	15,551	18,227	2,087	0,853
4.Poz.426.2	Požerė (Paežeris)	Fibula	Bronze	0,064	3,085	38,768	15,697	18,613	2,083	0,843
5.Poz.469.15	Požerė (Paežeris)	Bracelet	Bronze	0,008	3,454	38,703	15,687	18,506	2,092	0,848
6.Poz.469.19	Požerė (Paežeris)	Fibula	Bronze	0,070	3,787	38,455	15,524	18,250	2,107	0,851
7.Poz.469.85	Požerė (Paežeris)	Fibula	Bronze	0,064	4,818	38,333	15,512	18,355	2,089	0,845
8.Poz.469.172	Požerė (Paežeris)	Neck-ring (bow)	Brass	0,094	1,514	37,443	15,576	17,645	2,122	0,883
9.Mau.13216	Mau-džiorai	Fibula	Brass	0,102	1,194	39,073	15,698	18,672	2,093	0,841
10.Mau.1153	Mau-džiorai	Bracelet	Brass	0,119	3,341	38,764	15,632	18,539	2,091	0,843
11.Mau.11541	Mau-džiorai	Neck-ring (bow)	Bronze	0,150	3,072	38,975	15,781	18,576	2,098	0,850
12.Mau.13241	Mau-džiorai	Bracelet	Gunmetal	0,085	1,309	39,129	15,720	18,647	2,099	0,843
13.Mau.11543	Mau-džiorai	Bracelet	Bronze/ Gunmetal	0,085	1,791	38,994	15,727	18,575	2,099	0,847

Sample ID	Site	Artefact	Alloy type	Sample weight (gr)	Pb wt%	Pb isotope ratios				
						208/204	207/204	206/204	208/206	207/206
14.Dro.179-12	Drobūkš-čiai	Dress-pin	Gunmetal	0,024	4,888	38,790	15,592	18,510	2,096	0,842
15.Obel.621-120	Obeliai	Fibula	Brass	0,078	0,100	38,890	15,724	18,556	2,096	0,847
16.Obel.621-209	Obeliai	Fibula	Brass	0,091	0,576	38,146	15,596	18,523	2,059	0,842
17.Dauj.505.18	Daujėnai (Baluškiai)	Bracelet	Brass/ Gunmetal	0,040	2,662	38,416	15,580	18,388	2,089	0,847
18.Auks.276.64	Aukšta-dvaris	Bracelet	Brass	0,079	0,657	38,261	15,600	18,428	2,076	0,846
19.Auks.276.367	Aukšta-dvaris	Fibula	Brass	0,058	4,219	38,363	15,632	18,570	2,066	0,842
20.Auks.276.513	Aukšta-dvaris	Fibula foot (raw metal)	Leaded bronze/ Gunmetal	0,058	11,101	38,056	15,597	18,343	2,075	0,850
VIKING AGE										
1.JurAp.724	Jurgaičiai	Chain	Lead brass	0,075	5,838	38,279	15,378	18,086	2,116	0,850
2Nik.833.5	Nikėlai	Neck-ring (bow)	Leaded bronze	0,088	5,981	38,464	15,517	18,104	2,125	0,857
3.Nik.833.8	Nikėlai	Bracelet	Brass	0,055	0,149	38,724	15,696	18,365	2,109	0,855
4.Nik.833.20	Nikėlai	Fibula	Leaded brass	0,099	5,534	39,162	15,753	18,768	2,087	0,839
5.Nik.833.51	Nikėlai	Bracelet	Bronze/ Gunmetal	0,073	6,042	39,429	15,863	18,834	2,094	0,842
6.Mau.10946	Maudžio-rai	Bracelet	Leaded gunmetal	0,091	3,524	38,586	15,691	18,362	2,102	0,854
7.Kul.1-1	Kulautuva	Neck-ring (bow)	Brass	0,019	2,292	38,223	15,599	18,291	2,090	0,853
8.Auks. 276.11	Aukšta-dvaris	Fibula	Gunmetal	0,079	1,182	38,368	15,692	18,288	2,098	0,858
9.Auks.276.517	Aukšta-dvaris	Ingot	Brass	0,082	5,472	38,860	15,461	18,141	2,141	0,852
10.Auks.276.441	Aukšta-dvaris	Fibula	Bronze/ Gunmetal	0,085	1,251	38,323	15,630	18,368	2,087	0,851
11Auks.235.77	Aukšta-dvaris	Fibula	Brass	0,046	2,021	38,626	15,654	18,535	2,084	0,845
12.Auks.235.148	Aukšta-dvaris	Ingot	Leaded brass/ Gunmetal	0,073	18,614	38,997	15,697	18,728	2,082	0,838
13.Auk.561	Kernavė 'Altar hill' hillfort	Ingot	Brass	0,045	0,704	39,008	15,681	18,789	2,076	0,835
EARLY MEDIEVAL PERIOD										
1.Bil.65b	Bilioniai	Fibula	Brass	0,021	6,800	38,614	15,636	18,416	2,097	0,849
2. Bil.99	Bilioniai	Fibula	Leaded brass	0,026	11,463	38,076	15,619	18,331	2,077	0,852

*Central Lithuania, exact location unknown; ** Shores of the River Venta, exact location unknown.

Švino izotopai vario lydinių kilmės ir perlydymo procesų kontekste Lietuvoje nuo I a. antrosios pusės iki XIII a. po Kr.

Audronė Bliujienė, Irma Vybernaitė-Lubienė, Veronika Biveinytė, Vaidas Pudžaitis, Evaldas Babenskas, Gediminas Petrauskas

Santrauka

Šiame straipsnyje pateikiami švino izotopų santykių ($^{208}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, $^{206}Pb/^{204}Pb$, $^{208}Pb/^{206}Pb$, $^{207}Pb/^{206}Pb$) vario lydiniuose rezultatai, derinant juos su archeologinių artefaktų elementinės sudėties tyrimais, atliktais induktyviai susietos plazmos masių spektrometrijos (ICP-MS) metodu. Iš viso šis tyrimas apima 208 mėginius, paimtus iš 180 radinių, pasklidusių visoje Lietuvoje (1 pav., 1–3 lentelės, 1 priedas). Chronologinis tyrimų diapazonas apima laikotarpį nuo I a. antrosios pusės iki XIII a. po Kr. pradžios. Daugkartinis dirbinių, pagamintų iš vario lydinių, perlydymas ir įvairios sudėties metalo laužo pridėjimas gaminant naują spalvojo metalo žaliavą, suprantama, keičia buvusią cheminę lydinio sudėtį ir izotopų santykius. Todėl perlydymo procesas tampa sunkiu iššūkiu, nustatant buvusių ryšių tarp pirminių vario lydinio kilmės regionų (rūdynų) pietinėje Europoje ir Mažojoje Azijoje bei žaliavos naudotojų Baltijos jūros regione pobūdį (2–8 pav.). Suprastas perlydymo procesas iš esmės keičia spalvotojo metalo žaliavos mainų linijinės trajektorijos idėją.

Šio tyrimo autoriai, be švino izotopų santykių, kas yra vienas iš pagrindinių veiksnių vario lydinių kilmės tyrimuose, į tyrimų lauką siūlo įtraukti vario grupes, kurios leidžia įvertinti spalvotųjų lydinių perlydymo mastą ir šį procesą susieti su tendencijomis, vyravusiomis Europoje (3 lentelė, 3 priedas). Vario grupės nustatytos remiantis arseno (As), stibio (Sb), sidabro (Ag) ir nikelio (Ni), kaip pagrindinių vario lydinių priemaišų, termodinaminėmis savybėmis ir nuostata, kad lydinyje kiekvieno iš šių keturių elementų turi būti ne mažiau kaip 0,1 %. Nustatyti švino izotopų santykiai, apibūdintos vario grupės, kartu su juvelyrų naudotomis dirbinių gamybos technologijomis (9 pav.) leido atskleisti prekių judėjimą kaip europinio metalo srauto dalį, vietiniu (dabartinės Lietuvos teritorijos), regioniniu (Rytų Baltijos regionas) ir transeuropiniu lygmenimis.