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& SASJA **VAN DER VAART-VERSCHOOF** (EDS)

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Comparative science-based analyses of bronze grave goods

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10.1 Introduction

This chapter focuses on the results of technical analyses of metal objects found in Early La Tène elite graves. These are mainly thin-walled bronze objects that are susceptible to corrosion and post-depositional damage. Various techniques and measurement tools were employed. All bronze items were analysed by handheld X-ray fluorescence (XRF) and the spherical balls were scrutinised in several ways. In total, 161 objects were investigated. All the techniques used are non-invasive; no damage was caused to the objects or object surfaces (Sect. 10.2). In this contribution, we focus on three main categories of bronze objects – the situlae, phalerae and spherical balls (Tab. 10.1) – as these were found in relatively large numbers, thus allowing for comparative analyses. Small groups or single items such as wheel parts, body ornaments and a bronze bowl are reviewed within the context of the totality of studied objects (Fig. 10.2).

In the past two decades the availability of handheld-XRF has changed the study of copper alloy objects in the Netherlands. Its non-destructive nature and ease of use has boosted the application and number of XRF surface measurements, especially for objects from the Late Neolithic to the Bronze Age (Arnoldussen *et al.* 2022). The technique provides valuable data, in particular when the results are cross-checked with those from other types of chemical analyses. For copper alloy objects from the Iron Age, however, the number of metallurgical studies in Europe is still limited, which makes interregional comparisons within specific bronze categories not yet possible.

In the present study, we attempt to answer several questions. First, we focus on the type of material, asking basic questions like: *What is the composition of the bronze alloy? How homogeneous is the bronze alloy of sets of objects from different graves? What are the differences and similarities in the bronze used for the different objects?* Sets of identical objects are given special attention in this approach: *Do these show a homogeneous composition, pointing to standardised production?* When interpreting the results we also engaged in discussions about methods of production.[1] *How were objects manufactured? Did this involve assembling bronze sheet plates, the lost-wax technique, or other methods?*

In general, lost-wax casting – also referred as *cire perdue* (French), *Wachsauschmelzverfahren* (German) – is a method used to cast single objects, based on the almost complete formation of the artefacts in a wax model (Nørgaard 2018, 97-8). In Iron Age Britain and Ireland clear changes in bronze-working technology are ob-

1 We would like to thank Sophie Adams (British Museum) and Jeroen Zuiderwijk for exploring ideas with us and for answering our craft-related questions. We also wish to thank Stijn Arnoldussen (Groningen Institute for Archaeology) for reviewing an earlier draft of this chapter and Judith van der Leije (Archol) for providing the photo in Figure 10.18.

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site	grave	analysed objects
Heumen-Hessenbergseweg	chariot grave; Ch. 2	12 phalerae, 1 situla, 10 hollow spherical balls, 4 nave bands
Overasselt	situla grave; Ch. 3	1 situla, 1 bronze bowl, 3 phalerae, 5 hollow spherical balls
Nijmegen-Traianusplein	chariot grave 9; Ch. 4	4 phalerae, 1 domed phalera head
Andelst-Hoge Hof	disturbed grave finds; Sect. 7.2	1 phalera, 1 hollow spherical ball
Nijmegen-Hunerberg	disturbed grave finds; Sect. 7.12	2 hollow spherical balls
Nijmegen-unknown findspot	disturbed grave finds; Sect. 7.13	2 phalerae, 10 hollow spherical balls
Wijshagen-De Rieten	graves C, D, E, H; Ch. 5	2 situlae, 1 ribbed bucket, 7 phalerae, 8 hollow spherical balls, ferrules and belt clamps, 1 bracelet, 1 torc, 1 glass bead

Table 10.1. Overview of the investigated sites and objects.

served (Webley *et al.* 2020, 128). After c. 600 BC, alloy recipes tended to be very different from those of the Late Bronze Age, with a much lower lead content. Crucibles were generally smaller, in keeping with a focus on casting smaller objects. In southern Britain, the bivalve mould technology of the Bronze Age and earliest Iron Age seems to have been abandoned altogether in favour of lost-wax casting.

It was hoped that the analyses could help us to identify workshop traditions, or to reconstruct the production process for the bronze items and the copper alloys used. Subsequent questions aimed to discern patterns of coherence: *To what extent are there indications that specific objects were made in specialised workshops?* The variation in bronze composition, or lack thereof, could also help answer the question of whether goods were specifically made for elite burial events or whether they were acquired over a longer period before their final deposition.

10.2 Methodology

10.2.1 Portable XRF

XRF analysis was used to determine the composition of the bronze objects. Earlier studies have shown that the results achieved by handheld XRF are of great value (Arnoldussen *et al.* 2022). Although it is a surface technique, with a very limited penetration depth, the outcomes are useful if judiciously applied. This involves considering information on the burial environment, patination and corrosion. In practice, measurement locations are chosen in areas where the original patina is damaged or has been removed.

A Niton XL3t GOLDD+ XRF analyser was used in our study. It was factory calibrated with standards for metals and alloys; it also had a silicon drift detector with optimised geometry. The electronic metals mode was selected and used throughout the data-gathering phase. The advantage of this mode is that the same metals contained in late prehistoric alloys (copper, tin, silver, zinc, gold) are found in modern electronic equipment, including potentially hazardous metals (lead, mercury, selenium) that need to be identified in the recycling of scrap. A reading time of 30 seconds was used, which is sufficient to detect most trace elements at parts per million (ppm) level. Corroded metal is one of the most problematic materials to investigate with handheld XRF because the outer corrosion has an altered composition relative to its original (non-corroded) state. This bias introduced by corrosion was evaluated by analysing each object at several different spots, distributed across both sides. The thinking behind this method is that the thickness of a corrosion or patina layer varies across the object. By measuring at different locations and taking into account the behaviour of different elements during corrosion and the influence of the burial environment, an estimation of the composition of the non-corroded surface may be arrived at. The concentrations of the alloying elements are summed to 100%, without the concentrations of iron and light elements such as silicium and aluminium. However, the accurate determination of copper and hence tin, as the sum of both is almost 100%, is still hampered by

Fig. 10.1. The use of the Niton XL3t GOLDD+ XRF analyser on the remains from the chariot grave of Nijmegen-Traianusplein (RCE).

surface effects such as copper dissolution and the absorption of secondary copper X-rays by iron.

The six find complexes were measured with the handheld XRF at different times. The bronze objects of Heumen-Hessenbergseweg were analysed soon after discovery and before restoration. This allowed us to measure fresh fractures, where measurements were less hampered by corrosion or restoration substances. Due to the fragmentation, the situla of Heumen was exhaustively studied (46 measurements on one situla, compared with 45 measurements on two situlae and a single ribbed bucket from Wijshagen). The objects from the other locations (Wijshagen, Nijmegen-Traianusplein, Overasselt, the objects from Nijmegen in the collection of the National Museum of Antiquities and the finds from Andelst) were all restored prior to XRF analysis. The five phalerae and two rings hidden in the Nijmegen-Traianusplein metal jumble were the most challenging to measure (Fig. 10.1).

In total, 440 measurements were taken from five situlae, 29 phalerae, 36 spherical balls, the four nave bands of Heumen, the bronze bowl from Overasselt, ornaments from Wijshagen (bracelet and torque) and some smaller objects such as the ornamental bronze wire in the linchpin from Heumen. All detailed measurement data can be found in DANS (https://archaeology.datastations.nl/dataset.xhtml?persistentId=doi:10.17026/AR/QSFQHD).

10.2.2 X-ray and scanning electron microscopy (SEM-EDX)

The badly burned spherical ball of Heumen-17 was studied using two different techniques, both carried out at the Cultural Heritage Laboratory in Amsterdam. First, the ball was X-rayed. The instrument used was a General Electric Eresco 280 MF, with a rotating table, 2-millimetres-thick copper filter and a flat panel (FP) detector. Images were made at 120kV and 4 mA with a spot size of 1 millimetres.

The surface of a clay fragment found inside the Heumen ball was then studied by means of a JEOL JSM-IT700HR scanning electron microscope with a JED-2300-Fully integrated JEOL EDS system (100 mm^2 SDD). The SEM was operated in low-vacuum mode at a chamber pressure of 30 Pa, with an operating voltage of 20 kilovolt and a working distance of 10 millimetres. The SEM uses a focused beam of electrons that is scanned along the surface of a sample to create a magnified image. Interaction between the electron beam and the sample produces a wealth of information. The backscattered electrons (BSE) used in this study create an image whose intensity mainly reflects the chemical composition of the sample. Heavier components (e.g. iron) show up brighter than lighter ones (e.g. silica). Due to the bombardment with electrons, the atoms in the sample will also emit X-rays, each with a characteristic energy spectrum. Energydispersive X-ray (EDX) spectroscopy can be used to identify the elements present within the sample.

10.2.3 THM-Py-GC-MS

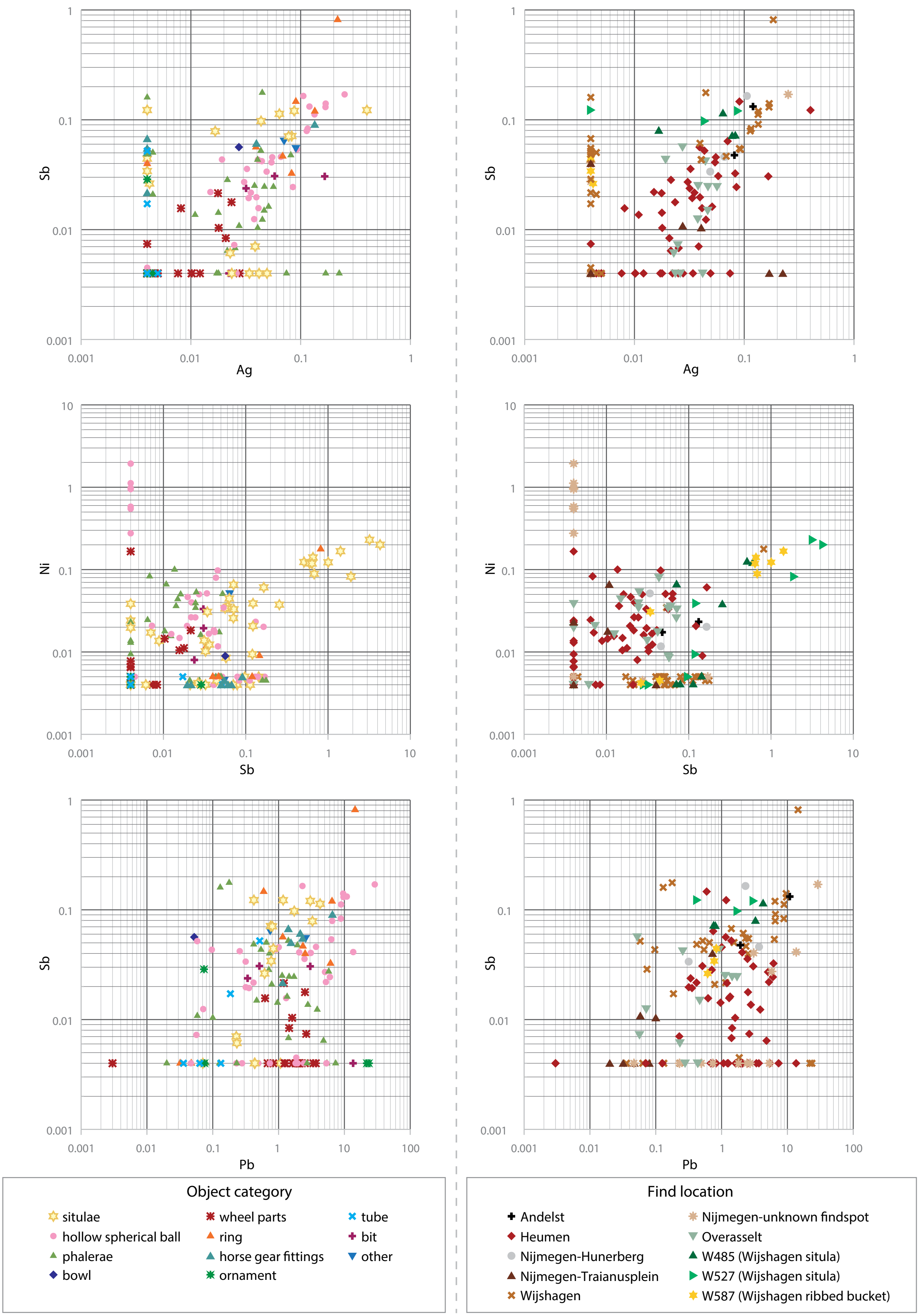
Thermally-assisted hydrolysis and methylation pyrolysis gas chromatography mass spectrometry (THM-Py-GC-MS) was used at the Cultural Heritage Laboratory in Amsterdam in order to identify a small amount of organic material from a spherical bronze ball from Heumen. THM-Py-GC-MS is a destructive method although the amount of sample needed is very small: about 0.1-10 milligrams depending on the amount of organic matter present in the sample. The method involves heating a small sample in the absence of oxygen to 700°C in a few seconds (pyrolysis). Small organic molecules will evaporate and large molecules decompose. To prepare polar molecules for the subsequent chromatographic analysis, the reagent TMAH is added to the sample before pyrolysis, which reacts with the polar sides of the molecules. The resulting small molecules and fragments of the large molecules enter a capillary glass tube coated on the inside with a non-polar layer (the chromatography column). This column is heated slowly from a temperature of 35°C to 320°C, causing the different substances to leave the column at different times, specific to each substance. In this way each substance passes through a connected mass spectrometer, which provides information about the molecular mass and molecular structure. The resulting mass spectra can be matched with a database to identify the substances.

Fig. 10.2 (opposite page). Summary graphs of the studied objects: element plots of silver antimony, antimony nickel and lead antimony (excluding the rivets of the situlae), expressed in wt % (RCE).

A Frontier Lab 3030D pyrolyser was used in combination with a Thermo Scientific Trace 1310 gas chromatograph and a Thermo Scientific ISQ7000 mass spectrometer. The pyrolysis technique involved a rapidly increasing temperature, ranging from 350°C to 700°C; the temperature of the pyrolysis interface was 290°C. The pyrolysis unit is directly linked to a SLB5 ms Supelco column (with a length of 20 m, an internal diameter of 0.18 mm and a film thickness of 0.18 microns) by a split connector. Helium with a programmed flow (0.5 to 1.2 ml/min) is used as carrier gas in combination with a temperature program of 35°C (1) – 60°C/min – 110°C – 14°C/min – 240°C – 5°C/min – 315°C (2). The column is directly coupled to the ion source of the mass spectrometer. The temperature of the interface was 250°C, the temperature of the ion source 250°C. Mass spectra were recorded from 29 to 600 AMU at a speed of 7 scans per second. Xcalibur software 4.6 was used to record and process the spectral data.

10.3 Results for metal composition

Most prehistoric bronzes consist of copper alloyed with tin (Sn), and sometimes lead (Pb). In addition, trace elements such as arsenic (As), antimony (Sb), nickel (Ni), bismuth (Bi) and silver (Ag) may be present. Early copper ores were most likely the oxidised parts of sulphidic ore deposits that could be easily distinguished by prehistoric miners. The amount of sulphidic bound elements, such as As, Sb, Ni, Bi and Ag, are generally low in these ores as they are not retained by the oxidised copper ore. Most copper in the Iron Age is derived from sulphidic ores in which these elements are concentrated. Depending on how the ore is processed, some of these elements may be preserved in the bronze.


1
0.1
0.01
0.001
Sb
0.001
0.01
0.1
1
Ag
10
1
0.1
0.01
0.001
Ni
Sb
10
100
Pb
Object category
situlae
hollow spherical ball
phalerae
bowl
wheel parts
ring
horse gear fittings
ornament
tube
bit
other
Find location
Andelst
Heumen
Nijmegen-Hunerberg
Nijmegen-Traianusplein
Wijshagen
Nijmegen-unknown findspot
Overasselt
W485 (Wijshagen situla)
W527 (Wijshagen situla)
W587 (Wijshagen ribbed bucket)

However, during recycling, volatile elements such as Bi, As and Sb will evaporate.[2]

In the corpus of over 370 compositional analyses of Dutch Bronze Age and Iron Age copper alloy artefacts, 15 are dated to the last six centuries of the first millennium BC (Arnoldussen *et al.* 2022, 10). This dataset suggests that alloys with low (<1%wt) concentrations of Ag, Sb, As and Ni were the norm, although lead was added in high concentrations (2.1-20.6%wt) to ornaments and scabbards.[3]

In the Late Bronze Age and Iron Age the amount of inherited elements[4] increases, most likely due to the decline in copper mines and the use of fahlores, which occur more abundantly across Central and Alpine Europe.[5] Fahlores (German *Fahlerz*) are ores with a pale colour (*fahl* in German), caused by the copper arsenic and copper antimony minerals (tennantite and tetrahedrite respectively) present in the ore, which can also include silver, zinc and bismuth.

In the Early Iron Age, bronze objects were no longer needed for agriculture or warfare, but became luxury goods used for display. They were elaborate in design and ornamentation, resembling much rarer gold and silver objects. As a result, the composition of the bronze became less critical, although more important for metalworking processing such as forging, casting, drawing, sinking, raising, chasing, coining, bending and repoussé. For fine casting, lead is added to increase the fluidity and reduce the melting point of the molten metal, allowing for the manufacture of thin-walled objects. The disadvantage of leaded bronze is that cold working, such as hammering and annealing, is difficult as lead does not dissolve in bronze but remains at the grain boundaries or as a separate chemical phase. Too much lead may lead to unwanted material characteristics as the material becomes more brittle, is more difficult to cold work and is weaker.[6]

In our analyses, we paid special attention to differences in the values of specific elements such as lead, silver, antimony, bismuth and zinc, as these values are more distinctive than the copper and tin ratios.

The element plots show that the sulphidic ore-associated elements have a log-normal distribution. This is because the occurrence of the amount of minerals or elements in nature is governed by processes of exponential growth or loss resulting in such distributions (Limpert *et al.* 2001). In the lead antimony plot, two groups can be immediately distinguished: one with lead-containing objects with no antimony and one containing both elements. As lead is most likely added if its value is higher than 2.5%, several types of copper ores were clearly used for the production of these objects, although groups (location and/or object types) with a similar lead/antimony ratio can be distinguished. Three groups can be distinguished in the plots for silver, antimony and arsenic: two in which one of these elements is very low compared to the other, and one showing a positive correlation between the elements (Fig. 10.2). The positive relationship between those elements indicates the use of fahlore, most likely mined at different locations, characterised by a higher arsenic concentration (tennantite-type fahlore) or a higher antimony concentration (tetrahydrite fahlore), both containing silver. In the silver/antimony plot, it is remarkable that most objects related to situlae contain almost no nickel but varying amounts of antimony. By contrast, the hollow spherical balls and most of the phalerae do have similar amounts of nickel and antimony. In addition, the finds of Nijmegen-Hunerberg, Overasselt and Heumen show a large overlap in their values for nickel, silver, antimony, bismuth and arsenic.

In order to determine and explain the differences and similarities *within* the object groups, we present the results by category in the following sections.

10.3.1 Situlae of the Rhineland-Tessin type and the ribbed bucket

We investigated four different situlae of the Rhineland-Tessin type, from the graves of Heumen, Overasselt and Wijshagen mounds C

2 L'Héritier *et al.* 2015; Merkl 2010; Pollard 2018; Tylecote *et al.* 1977.
3 The abbreviation 'wt' refers to 'percentage by weight'.
4 The inherited elements are the trace elements that were first embedded in the ores and were passed on after mining and processing to the bronze alloy of the objects.
5 Arnoldussen *et al.* 2022; Merkl 2010; Mödlinger/Trebsche 2020; Pernicka *et al.* 2016.
6 Gupta *et al.* 2016; Montero *et al.* 2003; Prasad *et al.* 1997.

situla	type	lead % of rivets (mean)	lead % of vessel plate
Heumen-Hessenbergseweg	situla Rhineland-Tessin type	0.6	0.1
Overasselt	situla Rhineland-Tessin type	7.5	0.3
Wijshagen mound C (WH-485)	situla Rhineland-Tessin type	12	3
Wijshagen mound E (WH-527)	situla Rhineland-Tessin type	2.5	1.4
Wijshagen mound H (WH-587)	ribbed bucket (cista)	16.3	0.5

Table 10.2. Differences in bronze alloys used for the bronze vessels.

(WH-485) and E (WH-527). The current thinking is that these vessels were produced in several workshops in the southern Alpine region of northwest Italy. Situlae of the Rhineland-Tessin type belong to a larger family of bronze vessels composed of prefabricated bronze plate material. The making process can be regarded as highly standardised (Nortmann 1998, 452; Nortmann/Grosskopf 2018). The bronze was cast as a slab: it may have been formed by casting directly into sand or a more formal stone mould may have been used (Joy 2014, 338). The plate was then hammered out over a wooden former with blows on the surface, to a thickness of 0.5-0.6 millimetres. The vessel design was constructed from quarter-circular plates cut from a circular disc 1 metre in diameter (Nortmann 1998, 453). The plates were put together with a row of rivets to form the body. The truncated cone-shaped form was created by plastically re-shaping the plates in the shoulder area.

The situlae of Overasselt and Heumen differ from the other situlae of this type in terms of the construction of the bottom. Instead of the usual *Falzboden*, where the bottom plate is folded over the bottom edge of the body plate, the bottom plate was attached to the lower end of the body by being clamped between the walls, probably with the aid of solder or an organic substance. This bottom construction seems to be unique, suggesting that the situlae of Overasselt and Heumen are the product of the same workshop (Sect. 2.3.2.2, especially footnote 12).

The bronze ribbed bucket of Wijshagen mound H (WH-587) was also investigated. The Wijshagen cista was also made of plate material, which – before being assembled – was carefully hammered into a ribbed surface. A region of origin cannot be precisely determined, but a production workshop in northern Italy or the east Alpine region seems most plausible (Sect. 5.2.3). Different parts of the vessels were measured in the XRF study: the body (made from one or two single sheets of bronze), the bottom plate, selected rivets and the handle. The bronze repair wire that was used to re-attach the bottom plate of Wijshagen situla 485 was also analysed.

10.3.1.1 Metallurgical differences

The first, more general observation, is that the rivets were made from a different bronze alloy than the bronze plate of the body and bottom, which suggests that different bronze-casting processes were used. Within the five vessels, the average lead content of the rivets in particular varied widely (Tab. 10.2).

The use of a highly leaded alloy for the production of small rivets makes sense from a practical point of view. The addition of more than 3% lead makes the bronze easier to cast (Heeb/Ottaway 2014). It is generally accepted that adding lead was a new step in the development of metal technology: it lowers the melting point and improves flowability and castability (Scott 1991, 2014). But a high lead content also leads to brittle material, which easily cracks and breaks. This is unfavourable for sheet bronze, but not for small fixtures such as rivets. The rivets from Heumen are an exception: apart from the ones used to connect the handle attachments, they all contain only a small amount of lead (0.3% on average). This suggests that the rivets of the Heumen situla were produced as a series or batch. Also, the rivets of the Wijshagen bucket have far higher antimony, silver and bismuth concentrations than all the other rivets and are grouped close together for these elements (Fig. 10.3). In the antimony-silver plot, all vessel rivets can be

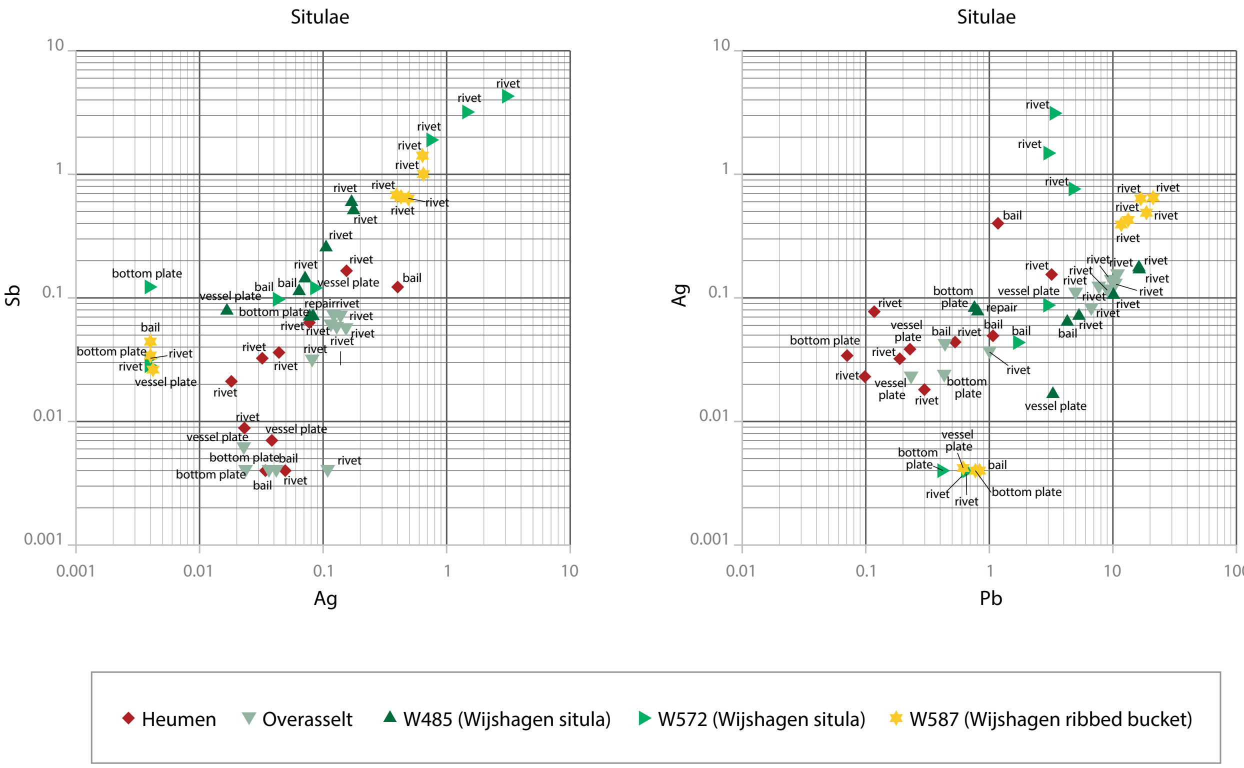


Fig. 10.3. Graphs of the minor elements antimony, silver and lead encountered in the situla components (RCE).

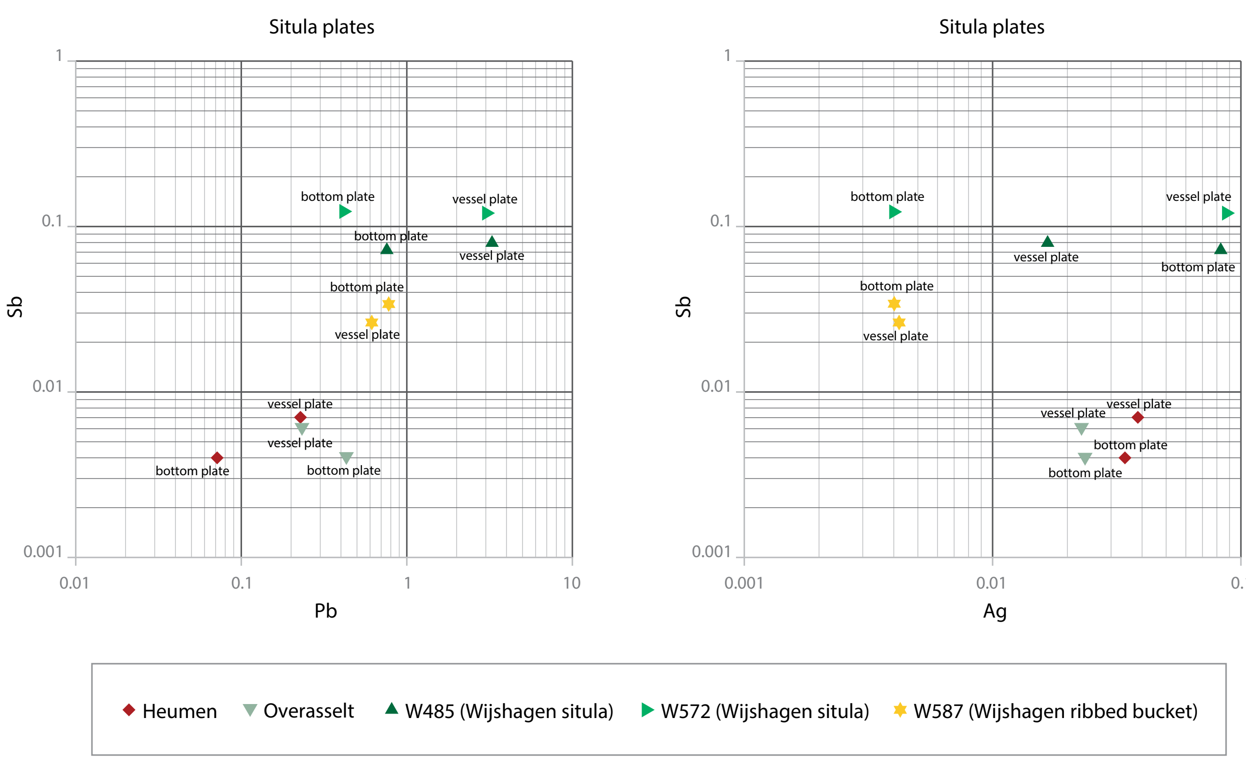


Fig. 10.4. Graphs of the antimony-lead and antimony-silver ratios of the situla plates (RCE).

distinguished on the basis of the concentrations of these elements. Only rivets 4 and 5 from the Wijshagen 527 differ from the main Wijshagen rivet group, having much lower silver concentrations than the other rivets. These, too, were not used to assemble bronze sheets but to connect the handle attachments to the shoulder. This may be because different rivet types were used in the production of the situlae, suggesting a different type of alloy for a different type of fastening object. Another possibility is that the rivets of the handles were repaired as they are subject to the most wear and tear. One of the handle rivets of the Overasselt situla is another outlier, possibly pointing to a repair.

The antimony-lead/silver graphs show that the bronze plates of the body and bases of both situlae of the Rhineland-Tessin type of Heumen and Overasselt are remarkably similar in terms of the alloy used (Fig. 10.4). Although the lead content of the sheet material for the body and bottom of W485 and W527 is different, the other trace elements (Sb, As, Ni) are comparable. Lead might have been added to enhance the casting of sheet material. It is plausible that the bronze sheet metal of the Overasselt and Heumen vessels was made from one alloy type with copper from the same ore, and processed in the same workshop. This fits in well with the unique attachment of the bottom plates of both situlae. However, rivets containing far more lead were used to fasten the vessel plates of Overasselt. Lead was added to the bronze alloy used for the Overasselt rivets, whereas an alloy similar to that of the bronze plate was used for the Heumen rivets.

A similar consistency can also be observed for ribbed bucket W587 [W-36] from mound H of Wijshagen, for which it is plausible that body and base were made from the same type of bronze alloy. The bronze composition of the rivets is again clearly different (i.e. highly leaded) from the rivets used to attach the bronze sheets of the situlae (Tab. 10.2). The bronze plates of the body and base of situlae W527 from mound E and W485 from mound C differ more from each other. The lead-antimony ratios suggest that the bottom and body plates were made from two different alloy types. This ties in with the macroscopic observation that the bottom plates of these two situlae have been renewed (Ch. 5). It is interesting to note that these two conical situlae have a slightly different construction: W485 is large and made of two separate sheets, while the smaller W527 is made from a single sheet. These situlae could have been assembled at different workshops. Also, compared with the Overasselt and Heumen situlae, all three situlae of Wijshagen have a higher silver and lead content, pointing to a different bronze source. For situla W485 from mound C, it is moreover notable that the bronze composition of the repair wire at the bottom is similar in composition to the handle and bottom plate. This observation supports the idea that the handle of this situla may also have been replaced in the past.

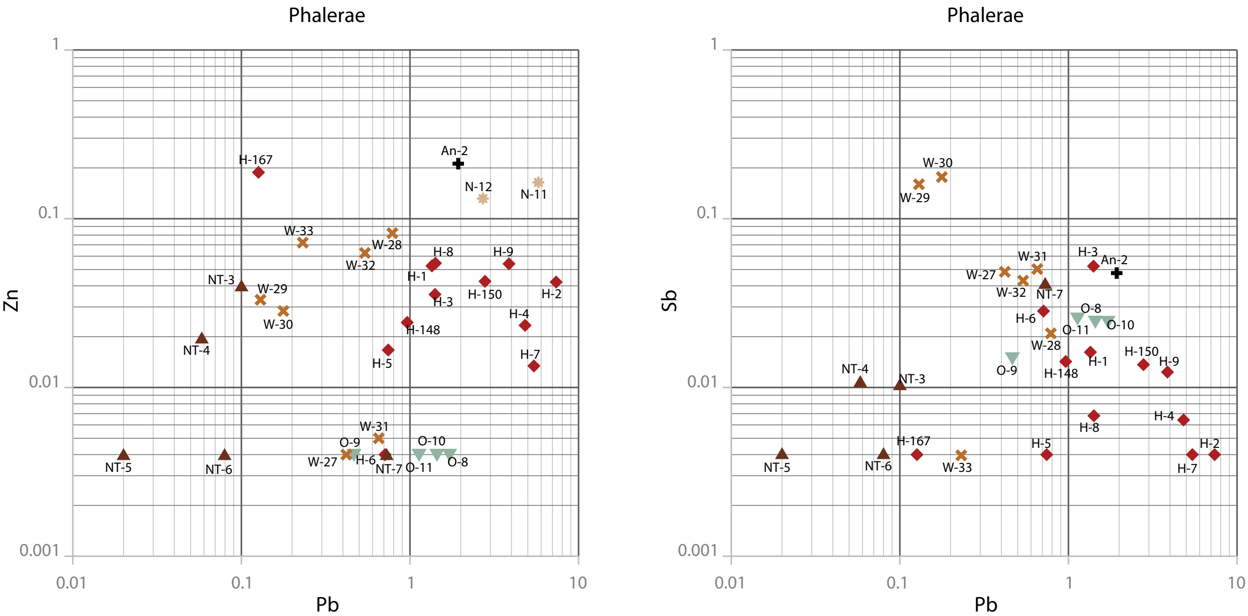
10.3.1.2 Conclusions about the bronze vessels

We can state that the Overasselt and Heumen situlae have not only typological similarities but also a comparable composition, except for the rivets used. Clearly different batches of rivets were used for both situlae. The composition of the rivets of all the vessels differs from the composition of the plate material. The rivets generally contain less tin and more lead. The difference in bronze composition of the different situla parts may indicate batch production. It is most likely that several parts were produced separately and assembled afterwards. There are indications of ancient repairs on all situlae.

The replacement of the bottom plates of situla W485 of Wijshagen mound C and W527 of mound E is confirmed by metallurgical analysis: the alloys of body and base plates have a different composition. The handle of W485 has also been renewed. The repairs indicate that both situlae had been used over a long period of time before being deposited in a grave. One of the handle attachments of the Overasselt situla may also have been repaired.

10.3.2 Phalerae

Phalerae, bronze decorative discs, are common elements in elite graves, often occurring in sets and interpreted as horse tack fittings. Different types can be identified, based on stylistic and technological features. Most common are phalerae of the Heumen type, characterised by discs of



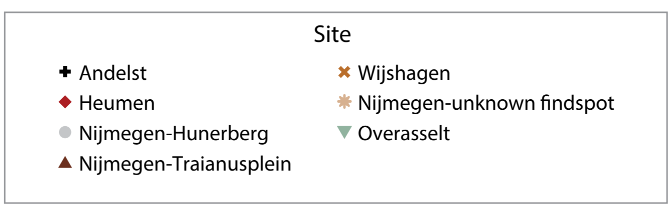


Fig. 10.5. Graphs of the lead-zinc and the lead-antimony ratios of the phalerae (RCE).

cast bronze with one or several circular grooves around a central 'navel'. Within this Heumen group, two subtypes can be distinguished: one with a looped attachment at the back, and one with a T-shaped central knob at the back; the latter are in fact composite artefacts.

The phalerae found at Nijmegen-Traianusplein are somewhat larger and have a pointed, domed head and are composed of a bronze disc with a separate tanged boss attached to the centre by an iron T-shaped sleeve (Bloemers 2016, 24; see Sect. 4.2.1.4).

10.3.2.1 Metallurgical differences

A total of 30 phalerae were analysed by portable XRF. The results as shown in various graphs in Figure 10.5 indicate that distinct groups can be identified.

Heumen

The XRF data on the twelve phalerae of Heumen shed some light on the bronze alloys used. The first observation is that although two distinct types occur in this group – eight with a T-shaped hook and two with looped attachments – their bronze composition is very similar. Phalerae 5 and 148, both equipped with a looped attachment, do not stand out. For the phalerae with a T-shaped hook, the results indicate that the alloy of the central button ('navel') at the front and the T-shaped knob at the back differs from that of the discs; the discs and the knob-hook attachments were cast with different alloys, made separately, meaning that these phalerae are composite objects. It is highly likely that the cast-on technique was used to assemble the two parts. The cast-on technique is a developed version of the lost-wax technique and was used primarily to cast complicated bronzes, such as belt plates (Nørgaard 2018, 113-26). 'Cast-on' means the mechanical connection of two cast pieces by enclosing or clasping these two parts through a third part that is subsequently cast on. The cast-on technique may have been applied as a variant of the lost-wax method or using two-part moulds. The similarities in appearance, thickness, regularity and size of the discs and hooks suggest that they could have been cast in the same impression moulds (see also Sect. 10.3.2). An impression mould could be created by pressing the bronze object into two separate pieces of soft clay, taking on the negative impression of its shape on both sides. When the clay hardened, the two pieces were removed to form a mould or at least a negative impression of the original bronze object, as a mnemonic device.

The different element plots for As, Pb, Ag, Bi and Sb show that phalerae 4, 5, 7, 8 and 167 appear as one cluster with a similar trace metal composition, as do phalerae 6, 8, 9 and 150. Phalera 3 differs in that it contains less lead but more trace metals such as Sb, Ni and As.

If we compare the Heumen items with the phalerae from other findspots, we see that they clearly represent a fairly distinct group (Fig. 10.5). Within this Heumen collection some of the phalerae have a fairly similar bronze composition: H-9 and H-150 show similar values of lead, zinc and antimony. They were probably made from the same bronze batch.

Wijshagen

Six phalerae from Wijshagen were studied, all with a looped attachment at the back. They were found in cista grave H and cast in one piece, as the bronze composition of the front plate is the same as the looped attachment at the back.

An examination of Wijshagen's six phalerae revealed the presence of sets: two show secondary holes (3 [W-33] and 6 [W-28]), two have clear, identical mould marks at the back (5 [W-29] and 1 [W-30]), indicating reusable moulds (of bronze or stone) or impression moulds, and two are slightly larger and better finished (4 [W-31] and 2 [W-32]). The question was whether their pairwise external properties would be mirrored by a paired bronze composition. The graphs of the trace elements indicate that this is indeed the case for the phalerae with the mould marks (Fig. 10.5). Phalera 1 and 5 were probably made from the same bronze batch. Both items have almost identical As, Pb and Sb concentrations. The same holds for pair 3 and 6, which are also strikingly similar, but different from pair 4 and 2, in their Sb, Pb, Zn and As concentrations. The set with the secondary holes are quite different from each other, suggesting that these were made in different workshops. In general, the Wijshagen phalerae contain far less lead than those from Heumen (Pb=0.4 versus 3.0%) but have much higher As contents (As = 0.76% versus 0.2%, except for pair W-29 and W-30 (As< 0.01%)).

Nijmegen-Traianusplein and the Nijmegen-unknown findspot

Four complete phalerae and a domed head (of an unrecovered or lost fifth specimen) from Nijmegen-Traianusplein were examined (Sect. 4.2.1.4) and two from the Nijmegen-unknown findspot (Sect. 7.13). Of the five phalerae, four occur in pairs (NT-5, NT-6 and NT-4 and NT-3). These have a low antimony, lead and arsenic content, but the pairs can be identified by their silver content. Phalera NT-7 does not match any of the other phalerae, showing higher lead and antimony concentrations and lower silver. Nijmegen is also the provenance of the other two phalerae, but unfortunately the exact findspot is not known. These phalerae, N-11 and N-12, have a far higher lead content and lower silver content than the Nijmegen-Traianusplein specimens. The small decorative discs N-11 and N-12 are typologically different.

The phalerae from Nijmegen-Traianusplein are larger (an average diameter of 10 centimetres), have a somewhat pointed, domed head, and are composed of a bronze disc with a separate tanged boss attached to the centre by an iron T-shaped sleeve (Bloemers 2016, 24; see Sect. 4.2.1.4). Like those from Heumen, these phalerae are composite objects: the discs are cast bronze work, and the tanged boss is also solid bronze, cast in a single piece with a domed head. Due to the difficulty of accessing the phalerae in the tangled iron work, we were only able to measure the disc plates (front or back) and the domed heads (at the front). As the results show no clear differences in bronze composition, it is likely that the same bronze alloy was used to cast both elements.

Overasselt and Andelst

The phalerae of Overasselt are also clustered very closely together in the Pb-Zn, As-Sb and Ag-Sb plots, having intermediate lead contents of about 2% and intermediate silver and antimony contents (0.4% and 0.2% respectively). The somewhat larger phalera from Andelst is only comparable to the Wijshagen phalerae in terms of its As-Sb ratio.

10.3.2.2 Conclusions about the phalerae

The phalerae composition differs per location and each find location has its own distinctive composition. Besides these groups, pairs can be distinguished at each location. Their compositional signature suggests that the phalerae were cast in pairs, and were probably used in horse tack in sets of two, four, etc. The sets remained together – they were not replaced – and were deposited as part of the burial ritual.

The T-shaped hook phalerae from Heumen and Traianusplein are composite objects, made of precisely produced and tight-fitting elements. The similarities in appearance, thickness, regularity of shape and decoration of the discs are remarkable, which also raises questions about the construction process. The cast-on technique was most likely used to cast these complex bronzes. The Heumen items were probably created from a small disc with a hole onto which a transverse pin was cast.

More work on the technological aspects of metalwork is needed in the future. Key questions to be answered include whether impression moulds were used to make the phalerae discs and pins and whether and how the cast-on technique was applied. Were all these phalerae pre-moulded in wax, or only the first specimen? And was this first original pressed into clay to create a mould that was used to make the other bronze copies? Or is it more likely that a reusable stone or bronze mould was used? The phalerae from the chariot grave of Amel-sur-l'Etang in France (Verger 1994, 583-92) are mentioned in Section 2.3.5.2, as these display the same T-shaped hook type as the Heumen ones. This resemblance could point to a common origin, to a same workshop. Verger (1994, pl. 209) schematically describes the different steps in the *chaîne opératoire* for the T-shaped hook type: a sequence of casting, hammering, assembling and finishing. The initial phase is yet unspecified, however. A bivalve mould is suggested by Verger, but whether this was a re-usable one or an impression mould, creating a negative impression of the original bronze object, is unclear. The mould marks of Wijshagen suggest that a re-usable solid bronze (or fine-grained stone) bivalve mould was used. Perhaps a lathe was used to create the decorative circle lines in the moulds of Heumen and Wijshagen. Verger (1994, pl. 209) suggests that the decoration was applied with a lathe after assembly, in the bronze metal rather than in the clay or wax. Further experimental work may reveal how these knob-hook attachments were produced.

10.3.3 Hollow spherical balls

The hollow spherical balls have been regarded for years as enigmatic objects.[7] These bronze balls are hollow-cast and have short, hollow 'legs' or shaft-cases. The most plausible possibility is that they are decorative horse tack fittings (see Sect. 13.6.3 and 14.8). At first glance, the balls are uniform in terms of appearance, size and signs of use. Their distribution (Fig. 14.7) shows a concentration in the Nijmegen region, which may point to a production in the Lower Rhine-Meuse region. A further three balls are known from settlement contexts in the region east of the Middle Rhine (Marquart 2010).

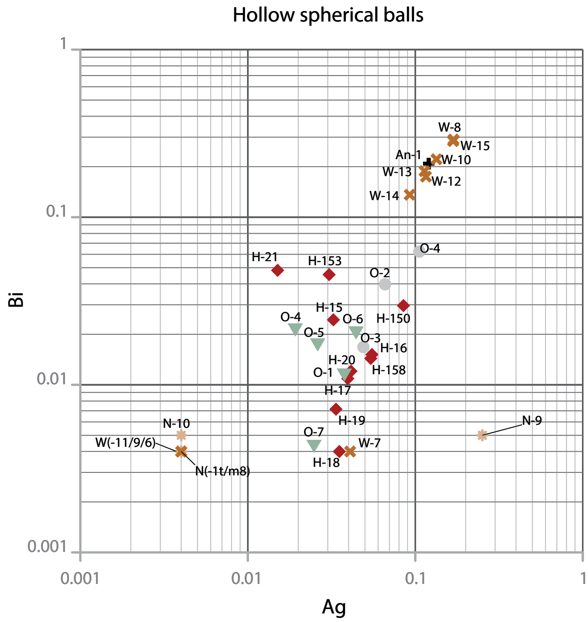
10.3.3.1 Approach

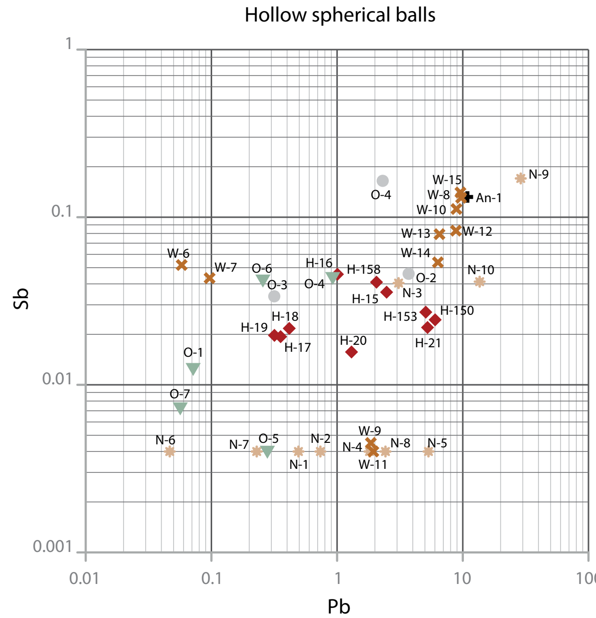
Several techniques were applied for a detailed examination of the spherical balls, focusing on their metal and clay composition. A total of 36 items were analysed by portable XRF. The heat-deformed ball with a clay core from Heumen-17 was analysed by X-ray, SEM-EDX and neutron tomography. Another ball from Heumen (16) and the one from Andelst were also examined by neutron tomography as both have clay remains inside. Organic remains were discovered during the restoration of ball Heumen-16. This residue was analysed by pyrolysis GCMS spectrometry.

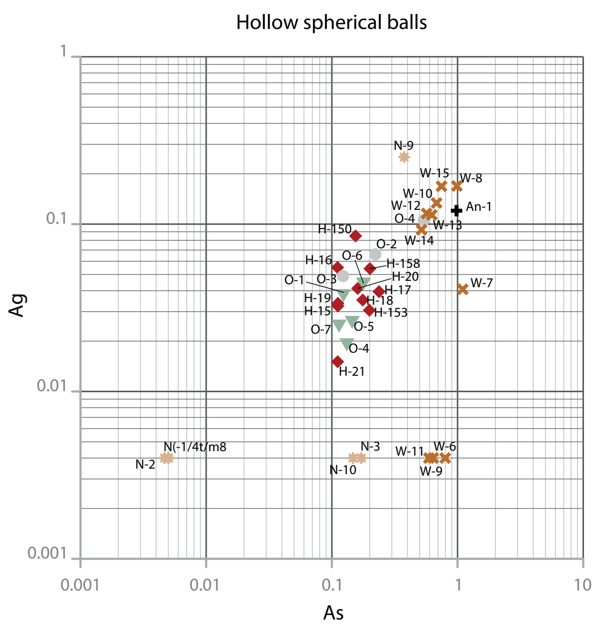
10.3.3.2 Results of the portable XRF

A total of 36 hollow spherical balls were examined with the portable XRF (Fig. 10.6). The distribution pattern shows very distinct groups for each find location (Fig. 10.6). The spherical balls from the Nijmegen-unknown findspot contain almost no silver, lead, antimony or bismuth. In contrast, these elemental concentrations in the balls from Wijshagen are a factor of 20-50 times higher than in the one ball from Andelst. The balls from Heumen, Overasselt and Nijmegen-Huner-

7 Marquart 2010; Swinkels 2010; 2011; Van Impe/Creemers 1991.







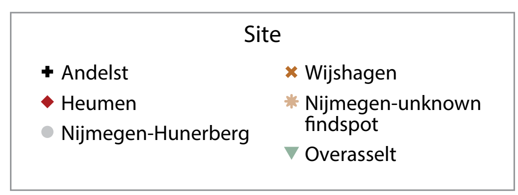


Fig. 10.6. The graphs of the trace elements of the hollow spherical balls (silver-bismuth, lead-antimony, arsenic-silver) (RCE).

berg have a similar trace metal composition. Like some of the phalerae, some balls appear to occur in pairs or small groups. Most prominent are the pairs Wijshagen balls W-11/W-9, Heumen H-150/ H-153 and the triple H-17/H-18/H-19.

These distinct bronze groups point to the production of balls with different bronze alloys and of different batches, except for the ones from Heumen and Overasselt, which have a very similar bronze composition. It is not only the trace metal compositions that are different: each set has very similar dimensions, which differ slightly from balls from the other sites (cf. Tab. 14.8). The balls from Wijshagen can be divided into two to three groups: two sets of two (W-9/W-11 and W-6/W-7) and a larger group (W-8, W-10, W-12 and W-15). The pair W-6/W-7 originate from mound E, while the other set and larger group of four are part of the group of eight from mound H. The specimens from the Nijmegen-unknown findspot and Nijmegen-Hunerberg are also two distinct groups.

Some balls from Wijshagen and the Nijmegen-unknown findspot group have similar trace metal compositions, as shown in Figure 10.6.

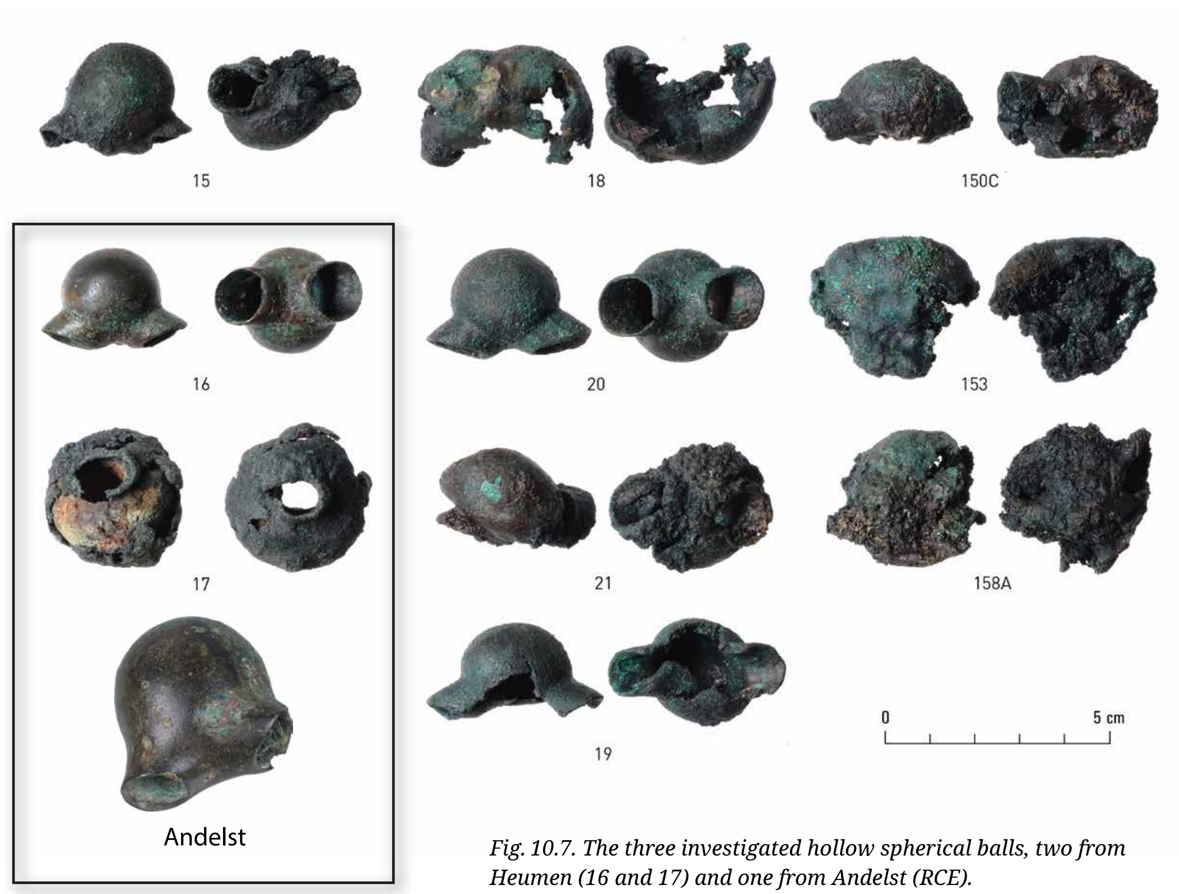


Fig. 10.7. The three investigated hollow spherical balls, two from Heumen (16 and 17) and one from Andelst (RCE).

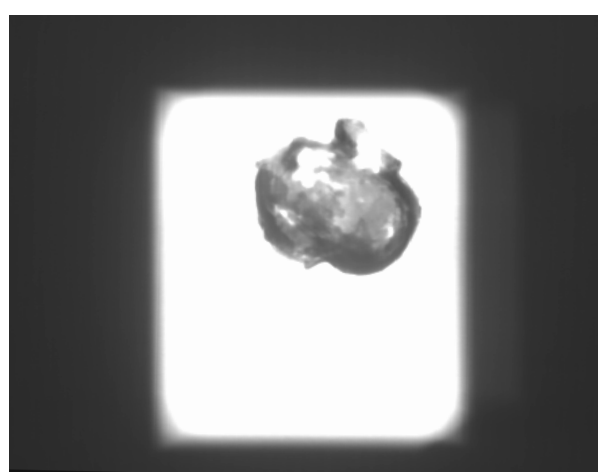


Fig. 10.8. Hollow spherical ball Heumen-17 in the X-ray setup at the Cultural Heritage Laboratory in Amsterdam (left), and the 120kV, 4 mA-image (right) (RCE).

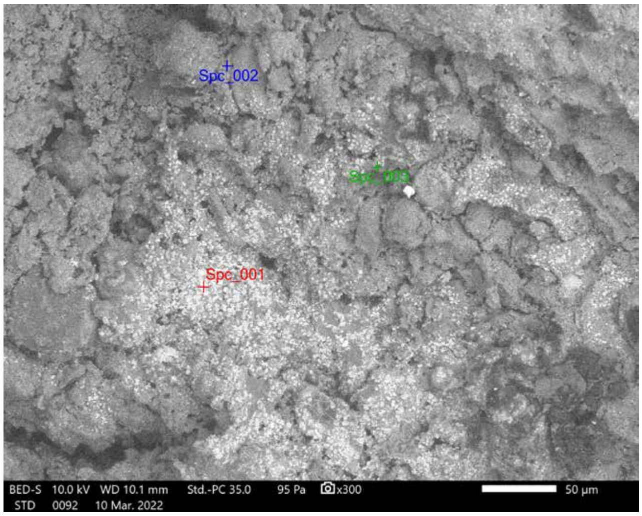
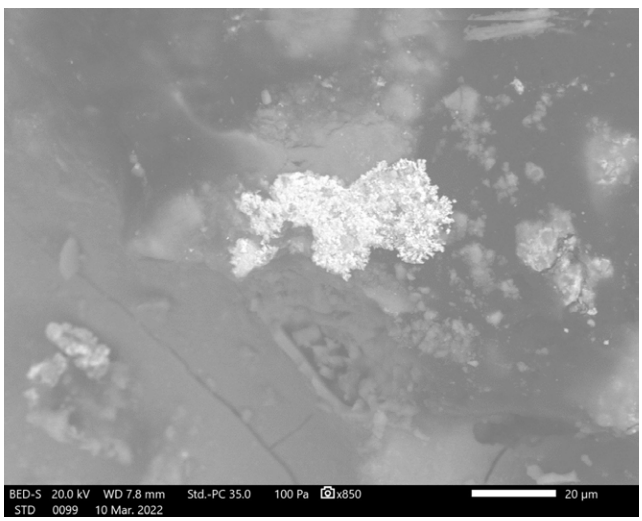
	BEI	Spectrum	Elements	Remarks
Clay		001 003	C, **O**, Al, **Si,** (K), (Ca), (Cu) C, **O**, Al, **Si,** (K), (Ca), Ti, Fe, (Cu)	clay matrix with a trace of copper
Metal		009	**Ag, S**	silver sulphide

Table 10.3. SEM-EDX results for the Heumen-17 ball summarised.

Ball N-9 from Nijmegen is plotted in the main Wijshagen compositional group and the two balls from Wijshagen mound H, W-9/W-11, are plotted in the middle of the main Nijmegen-unknown findspot group.

The identified sets were probably made from the same alloy batch. XRF analysis indicates that the bronze of the balls contains 0.3% silver. Silver occurs naturally in copper fahlores as a trace element. The similarity in Heumen, Overasselt and Nijmegen-Hunerberg is remarkable, hinting at a production in the same workshop.

Besides distinguishing different groups within the 36 balls, the XRF results also show differences within the objects. In a large number of balls, the spherical part contains more lead than the shaft-cases. This is striking, assuming that the objects were cast, which means that all the components in the alloy are evenly distributed within the object.

10.3.3.3 Results of the study of the clay remains (X-ray and SEM-EDX)

Three of the hollow balls contain traces of clay residues in their interior (Fig. 10.7). We used different techniques to try to explain their presence: *Is this a residue from a lost-wax casting process or was it added afterwards to fill up the inside of the balls?*

The badly burned ball Heumen-17 was the easiest to examine because the clay surface is clearly visible and accessible. The portable XRF revealed that the clay consists of silicon and is calcareous.

First, the Heumen-17 ball was X-rayed, 4 mA, providing an unclear picture of the bronze container and the internal clay volume (Fig. 10.8). No other features were visible.

The chemical composition of the clay and bronze surface was analysed in the scanning electron microscope (SEM) with energy dispersive X-ray spectroscopy (EDX). The imprints of plant remains at the surface of the clay core were clearly visible. These burnt plant remains were probably originally imbedded in the clay or added together with the clay.

The clay surface was measured at 20 locations, revealing aluminium silicates with potassium, iron and calcium, typically of clay minerals and lime. Copper is also present, which most likely derives from corrosion products. A small amount of silver sulphide was found in two places on the bronze surface. The most plausible explanation is that this silver also originates from the bronze (see above). A small amount of silver may have dissolved in copper (Chang *et al.* 1977), and also some copper sulphide may have survived the initial ore roasting, indicating that fresh (non-recycled) material was used to make the starting material for producing the balls.

Finally, the clay surface was examined for the presence of diatoms (or other phenomena) that could reveal something about the clay provenance, but no such indicators were found.

10.3.3.4 Results of the neutron tomography

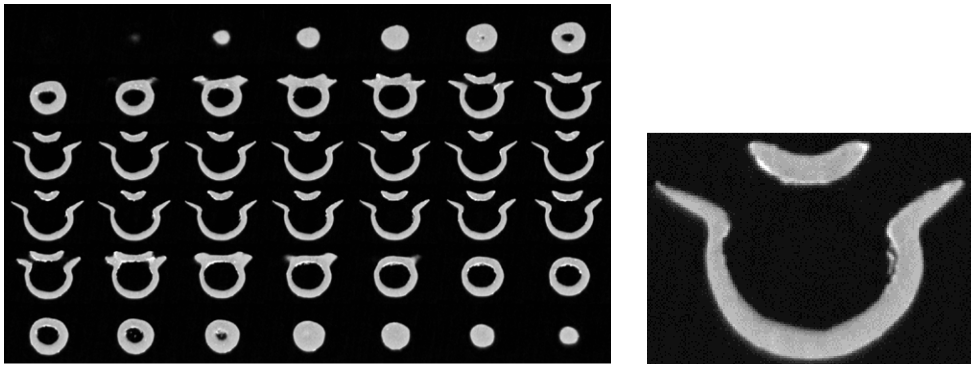
As the X-ray session at the Cultural Heritage Laboratory could not reveal a detailed picture of the inner structure, the three spherical balls were then examined at the TU Delft Reactor Institute by neutron-computed tomography (nCT). Like hospital CT scans, this technique allows us to visualise the interior of 3D objects in a non-invasive manner. By using neutrons instead of X-rays, thick metal objects can be inspected, as can be seen in Figure 10.9. These 2D-slices of the three 3D-reconstructed data show the shape, thickness and (to some extent) microstructural porosity of the bronze balls, the inner clay with pores inside, as well as some traces of other organic remains. These remains appear as brighter grey values than the clay and bronze because neutrons are particularly sensitive to light elements such as hydrogen and chlorine. For the same reason, the corrosion of the bronze on the outer and inner surfaces also appears 'bright'. The 3D data allows us to measure dimensional properties such as wall thickness and corrosion thickness with a typical resolution of between 0.2 and 0.5 millimetres.

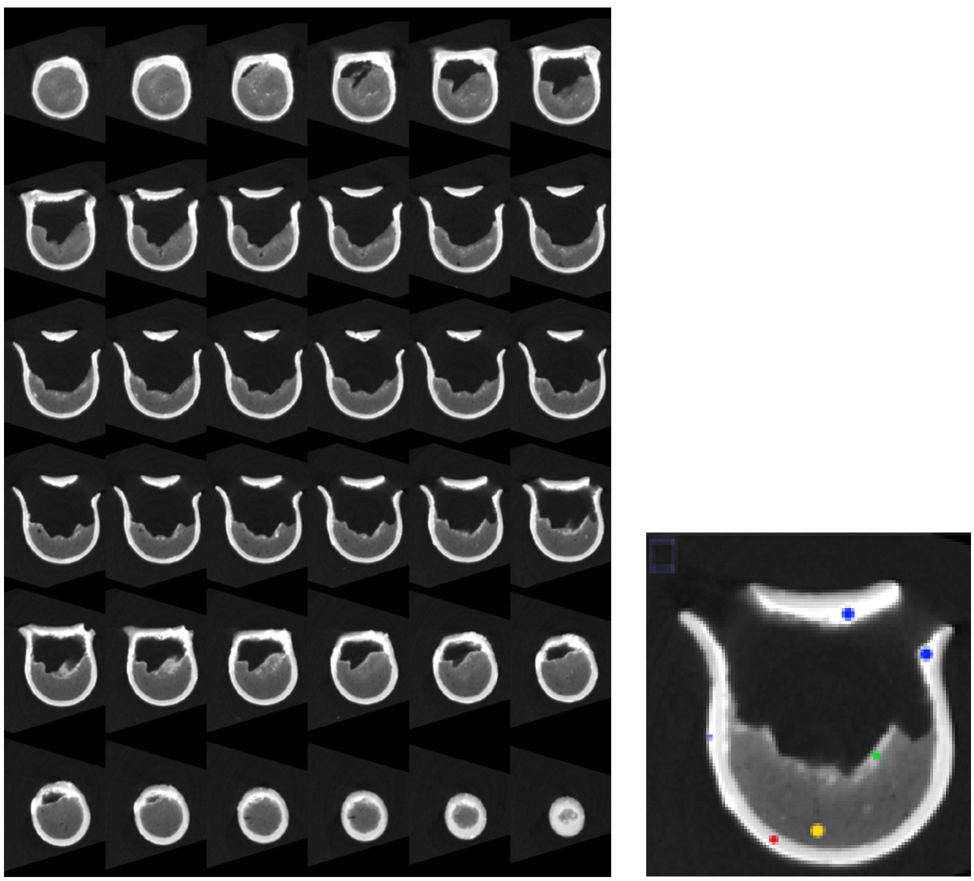
The images and videos of the three hollow spherical balls show a clear difference between the bronze layer and clay substance. There are no differences visible in the bronze layer of the balls, indicating that the objects were cast in one piece. Also, no holes or other anomalies could be noted: the bronze layer of each object is quite even in thickness.

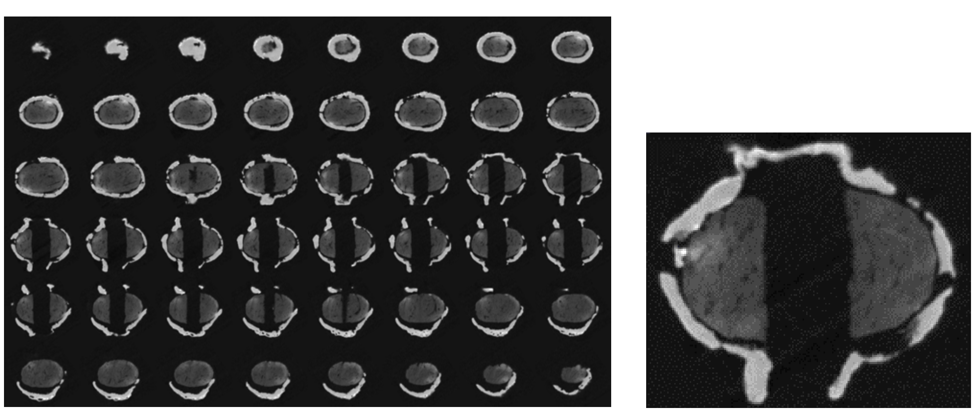
The images of the burnt clay core of Heumen-17 reveal a mass with small inclusions (organic temper?) and sometimes a layered structure (Fig. 10.10). This mass is clearly cut by something that had been inserted or 'stabbed' into both ends. This 'shaft' through the clay core has a larger diameter in the middle than at the ends. Perhaps a flexible element (rope?) was clamped in.

From the observation that the inclusions/voids inside the clay are mainly oriented along the shaft axis, we could infer that the clay was rolled (as a sphere) along the shaft. Another hypothesis would be that the orientation was caused by pulling the clay along the same axis, which seems less likely given its spherical shape.

Fig. 10.9 (Opposite page). A slice of each of the three objects' 3D neutron tomographic data (a Heumen-16; b Andelst; c Heumen-17). In Heumen-16 some corrosion can be seen on the surface and some flakes on the right-hand side. In the Andelst slice, the blue dots indicate the regions with either bronze corrosion or deposition of organic matter. The clay (yellow) is clearly visible, with a small especially bright region (green). The 'pristine' bronze has a grey value (red) that is distinct from the corroded part (blue) and is homogeneous (TU Delft).


a.


b.


c.

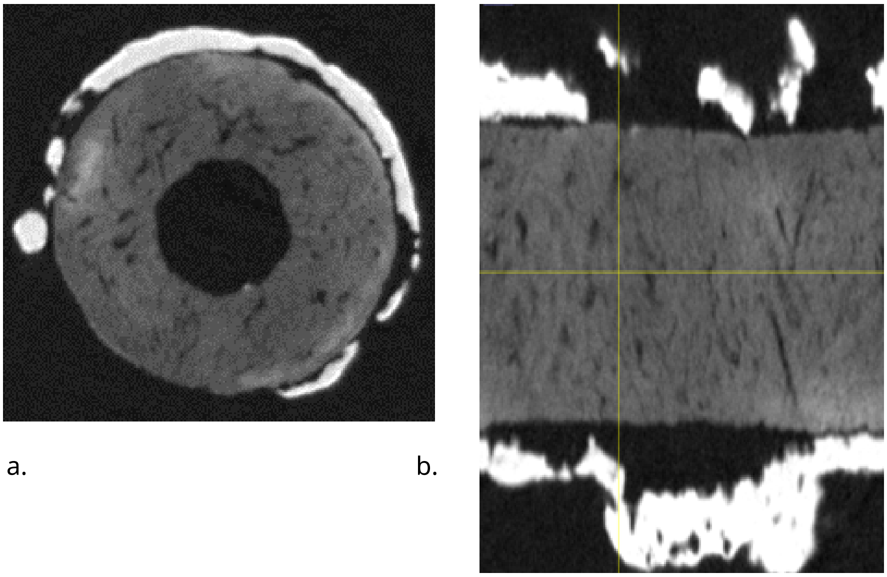


Fig. 10.10. A slice through the ball of Heumen-17 perpendicular (a) to the shaft shows that the porosity/inclusions do not have a marked radial or tangential orientation. The right-hand figure (b) shows a cylindrical slice, giving a '360-degree view' of the interior of the clay. Here, we can see that the inclusions are mostly oriented along the main rotation axis of the shaft (TU Delft).

10.3.3.5 Results of the THM-Py-GC-MS analysis

A small amount of deep brown to black material was recovered by Restaura from hollow spherical ball Heumen-16. It was secured in a snap cap and labelled 'charcoal'. First, this material was examined by archaeobotanical expert Otto Brinkkemper of the RCE. He concluded that the substance was not charcoal as no solid cellular tissue could be identified. Although the small lumps were difficult to identify, they could be vegetal, stem-like, hollow material (perhaps reed). His evaluation led to an assessment by archaeobotanical specialist Lucy Kubiak-Marten (BIAX). The question was whether a SEM analysis would be useful for determination. In her opinion, this was unhelpful as there was no evidence of anatomical structure. The matrices of the small lumps are very solid, only containing some air bubble hollows. It looks as though it was once liquid or semi-liquid, then solidified, perhaps indicative of a resinous or tarry matter. She proposed trying a chemical analysis by a pyrolysis-GCMS, which was performed at the RCE Laboratory in Amsterdam.

The pyrolysis chromatogram of the resinous material from ball Heumen-16 is dominated by short fatty acids (Fig. 10.11ab and 10.12), 2-ethyl-hexanol (RT 3:58) and a number of compounds whose mass spectra have no match in the databases. The short fatty acids present may indicate highly (microbially?) degraded fats. Dicarboxylic acids, of which hexanedioic acid (RT 5:24) in particular is present, may also indicate this but, like 2-ethyl-hexanol and dimethylphtalate (Fig. 10.12), hexanedioic acid could also reflect a contamination of the sample, as these compounds are widely used in the plastics industry, especially for plasticisers. Short fatty acids, especially when oxidised, can be easily transported by groundwater or mineralised completely in the soil (Evershed *et al.* 2002). However, the condition inside the balls might have been favourable for the survival of these compounds. Short fatty acids are more indicative of plant residue than animal residue (Evershed 2008).

In conclusion, pyrolysis-GCMS did not reveal any further clues about the nature of the material from hollow spherical ball Heumen-16.

10.3.3.6 Conclusions and discussions about the hollow spherical balls

The combined analyses of the hollow spherical balls yielded results on several topics discussed in this section.

Place of manufacture and exchange

The very distinct groups in terms of bronze composition of the 36 balls point to a more restricted compositional signature per site, including the sometimes pairwise occurrence of balls with identical composition. This similarity applies to all trace elements. A similar pattern was observed for the phalerae. Their compositional signature suggests that the balls were produced

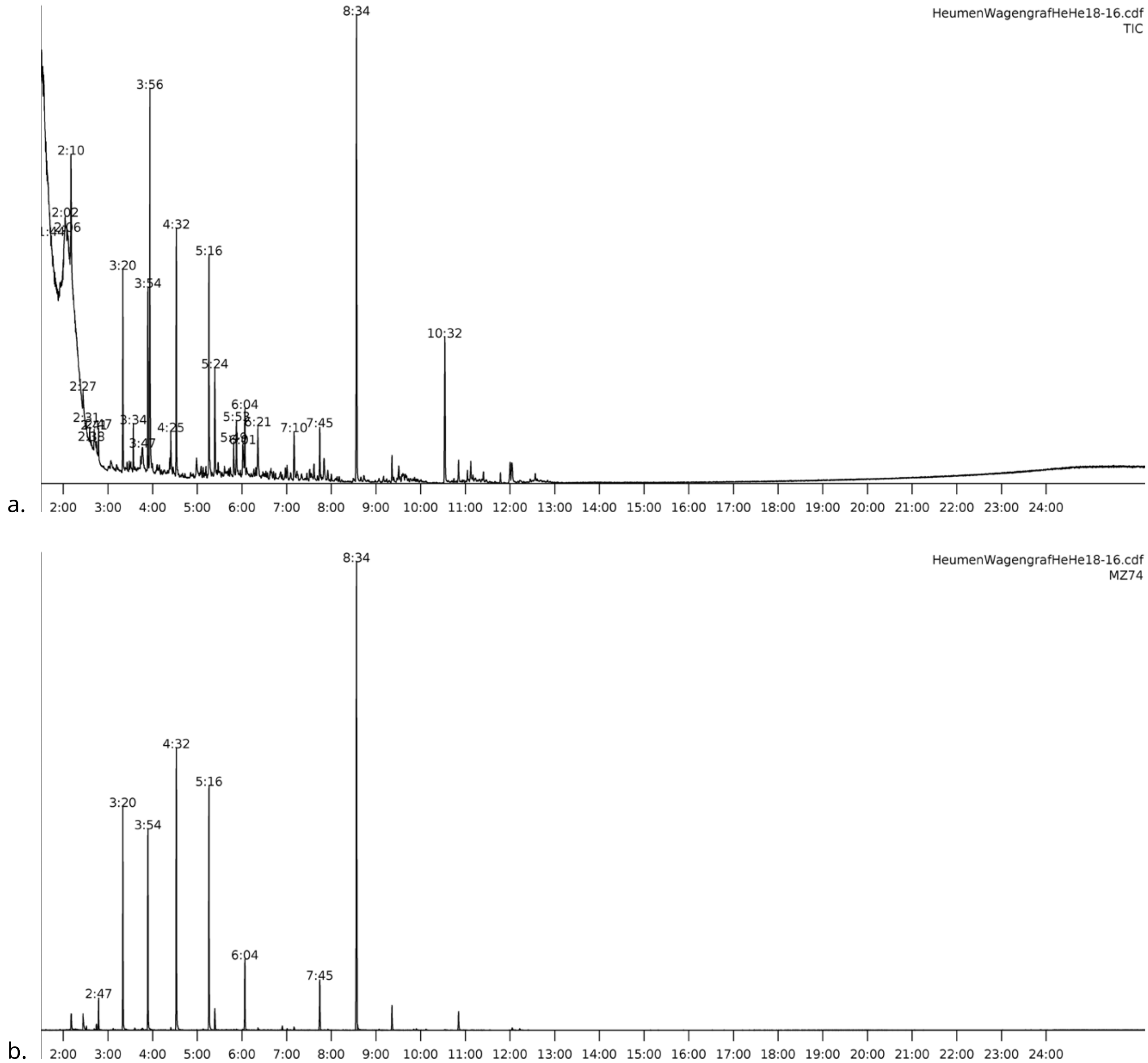


Fig. 10.11. (a) Total ion chromatogram of the pyrolysate of the resinous substance of the Heumen-16 ball. (b) Mass chromatogram m/z 74 showing the peaks of methylated fatty acids. The high peak at RT 8:34 is the internal standard (tridecanoic acid, methyl ester) (RCE).

in pairs – or even triples – in a single casting activity, as a batch-wise production from the same alloy. The sets remained together during use and were ultimately deposited in a grave. The alloys of the balls from Heumen, Overasselt and Nijmegen-Hunerberg all have similar trace elements, which points to the same, probably local, workshop.

The compositional signature of the Wijshagen balls and the Nijmegen-unknown findspot group can be divided into two clusters. This may point to a manufacture using different alloys. Within the Wijshagen group a clear distinction is visible between the sets from mound E and mound H. In this latter group of eight balls, a set of four and two can be identified. We can assume that the craftspeople at a workshop may have worked with different bronze alloys, depending on what was available as raw base and recyclable materials. All bronze in the Netherlands was imported and its origins will have varied. A predictable consequence would be differences in alloys

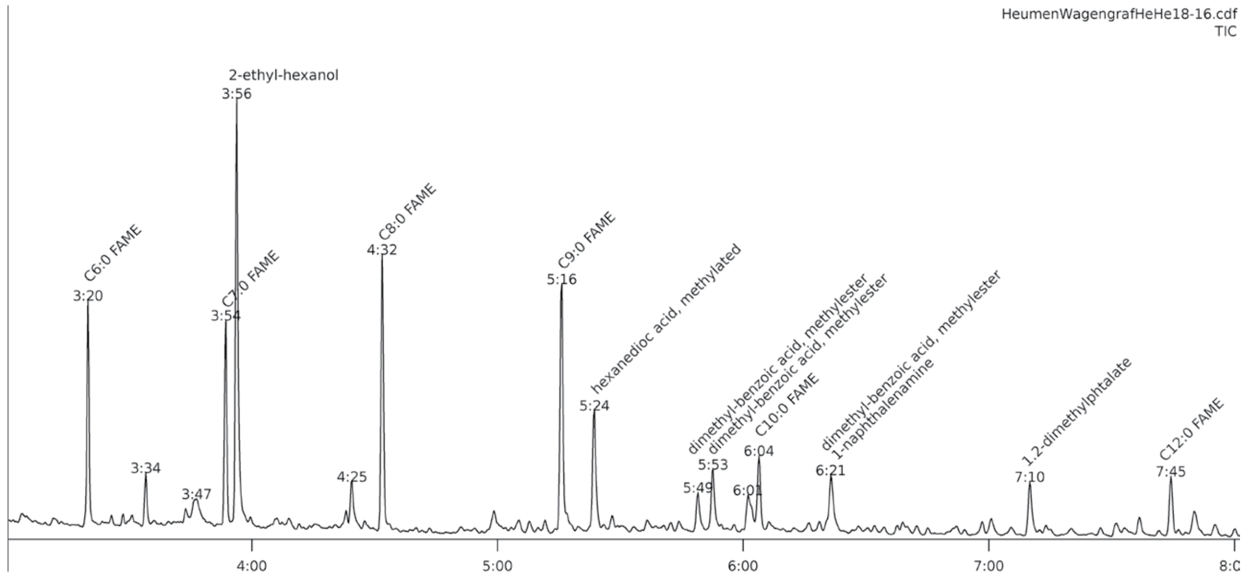


Fig. 10.12 Part of the total ion current (TIC) of the Heumen-16 ball. Cn:m FAME indicates fatty acids, where FAME stands for fatty acid methyl ester (=methylated fatty acid), n indicates the number of carbon atoms in the fatty acid chain and m indicates the number of double bonds in the fatty acid chain (RCE).

used within the same workshop. It is therefore all the more remarkable that some balls from Wijshagen, the pair found in mound H and the Nijmegen-unknown findspot group have similar trace metal compositions (Fig. 10.6). This correlation suggests that a ball from Nijmegen may have been produced in the same workshop as some items from Wijshagen, and that one pair from Wijshagen mound H (W-9/ W-11) may have been made in the same workshop as most of the Nijmegen balls. Although an exchange of single spherical balls seems implausible, we should bear in mind that some horse tack components may have ended up in other places through exchange, either as sets of spherical balls, separately, or fixed as part of horse tack.

Method of production and the use of clay

Most researchers do not address the making process, with the exception of Marquart (2010, 124; 2017, 127; 448), who states that the balls were cast. Considering their complex shape with curves and straight shafts, the manufacture of the balls using the lost-wax method is seen as the most plausible scenario. The clay remains in the inside of the balls from Andelst and Heumen-16 could then be interpreted as remnants of the primary clay core that was not completely removed. However, the clay core of Heumen-17 is not related to the casting process, but was probably used to fix a straight organic object (wood?, fibre or leather cord?). Clay with an organic temper seems to have been used as 'putty' – a rolled clay layer applied as a fixer – leaving a hollow 'shaft' after the ball was burned on the pyre. Unfortunately, it is not possible to interpret the organic matter from Heumen-16 – with short fatty acids more indicative of plant residue – in terms of the 'putty' hypothesis.

The remarkable discovery of Aschaffenburg-Bachsaal (Marquart 2017, 126-7; 447-8; 674) offers us more clues about the making process and method of attachment (Fig. 10.13). During construction work at a depth of 2 metres in the heart of the medieval centre of Aschaffenburg, a bronze hollow spherical ball was found together with a bronze tube remnant. Both objects were found in a secondarily relocated cultural layer that contained finds from the Early and Late Middle Ages. On the basis of the similar decoration on the tube and shafts of the ball, the similarity in diameter of the hollow openings and the comparable patina, Marquart proposes that they belong together as a bronze ensemble from the Early La Tène period. We also consider this plausible.

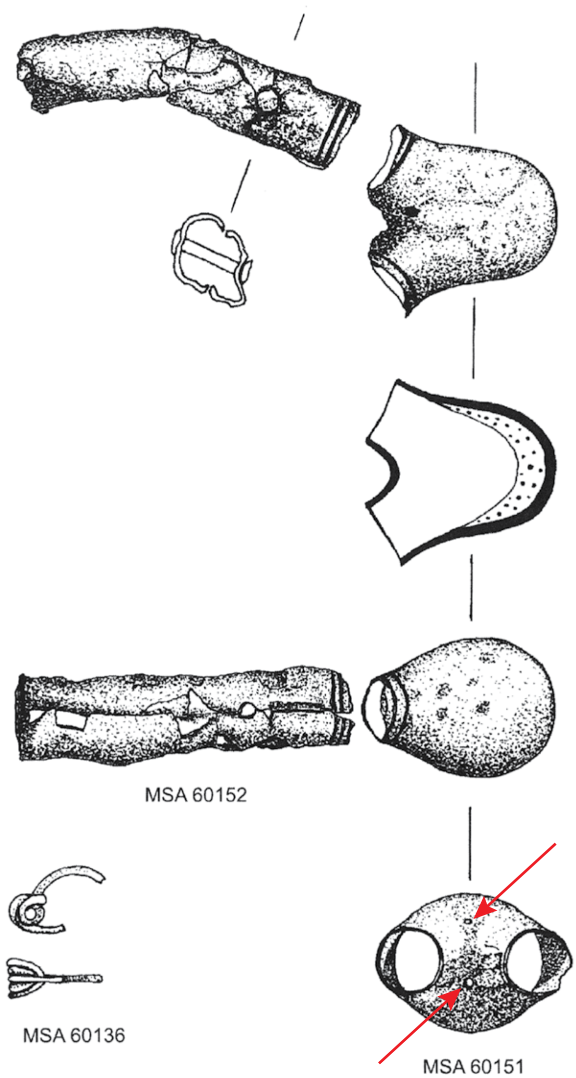


Fig. 10.13. The hollow spherical ball of Aschaffenburg was found together with a bronze tube. Two very small holes can be seen in the bronze body, near the shafts (red arrows; after Marquart 2017, 674, Taf. 118).

The hollow ball is very similar to the 36 examined in this study, but differs in two ways. The ends of the shafts have a ribbed decoration – like one side of the tube – and the body has two small holes near the shafts. It is thought that both small holes are remnants of the pins securing the mould to the clay core around which the wax model was formed (Marquart 2017, 448) as part of the lost-wax working process (Fig. 10.14). As holes of this kind are not visible in the spherical balls of Heumen and Andelst, analysed at the Technical University in Delft by neutron tomography, we believe that the Dutch balls were made without pins. The small holes in the Aschaffenburg ball may have been caused by air bubbles in the bronze alloy.

The tube is made from two bronze sheets, held together by a bronze peg that clamped the sheets into an oblong object, probably made of organic material (wood?, antler?), which was inserted into one of the shafts of the ball. It is remarkable that these two supposedly related objects had such different production methods, the ball being cast by the lost-wax process and the tube as two single bronze sheets, hammered out over or in a wooden former and connected by a bronze peg.

If we assume that the tube and hollow spherical ball from Aschaffenburg-Bachsaal belong together, this shows that the bronze smith had mastered a range of production techniques and was experienced in making these small, thin-walled bronze objects.

The reconstruction of the bronze-working technology used to make these enigmatic balls still leaves many questions unanswered, and the proposed operational sequence is a tentative suggestion only.[8] The key question is whether the wax-casting model of each ball was individually created by hand or whether a bivalve impression mould was used.

There is no macroscopic evidence of small holes, as in the Aschaffenburg ball, or of casting seams on the 36 balls. Nor did the neutron tomographic images reveal any casting seams on the three balls examined. Irregularities of this kind can be removed while the object is finished. As the dimensions of the balls in each set are quite similar, the use of a negative impression in clay, as a mnemonic, is plausible.

The making process by direct casting created by hand

The reconstruction of the making of the hollow spherical balls by direct casting (Fig. 10.14) would imply the following *chaîne opératoire*:

1. Making a small ball from soft clay;
2. Adding two small clay rolls, by pushing into the soft clay ball, as two protrusions;
3. Applying a thin layer of beeswax to the ball and protrusions, mixed with various

8 We wish to thank experimental bronzesmith Jeroen Zuiderwijk for sharing his thoughts on this topic.

additives (such as oil, talc and/or rosin) (Nørgaard 2018, 97);

4. Creating an assemblage of two beeswax balls and adding a larger wax sprue that can serve as a pouring cup and several wax rods that will allow displaced air to escape;
5. Wrapping the wax assemblage in clay layers (innermost layer the finest, followed by coarser layers) and drying or firing the block;
6. Firing the clay and melting the wax out;
7. Melting the alloy and pouring it into the fired clay block mould;
8. Releasing the bronze balls from the block mould and removing the bronze sprue and rods;
9. Removing the interior clay cores from the bronze balls;
10. Finishing the surface to enhance its golden colour;
11. The final ball, ready to be used.

In this process, the ball is modelled directly in beeswax, creating a unique casting, with the loss of the clay block mould.

The making process using an impression mould (indirect lost-wax casting)

The fairly fixed sizes of the balls could be an argument that a bivalve impression may have been used to produce hollow wax casting models of equal form and size. This would imply a negative impression of the sides of the bronze ball-to-be (coated with a layer of wax or uncoated or a dried clay model of a ball), impressed in two pieces of soft clay and, if hardened, used as a bivalve mould. In this scenario, hot wax was poured in such a mould and then poured out again, leaving a thin layer of wax on the inside of the mould. The desired thickness of the wax, and thus the future bronze wall of the ball, could be achieved in one go. That is a matter of waiting just long enough before emptying, based on experience. Or the pouring in and out was repeated until the desired thickness was reached. After that, the two halves of the mould were taken apart to release a hollow wax model of the ball. Next step is to fill the cavity in the hollow wax model with liquid clay silt and leaving this to dry. Then the casting process would continue as from step 4 in Figure 10.14.

Following this manufacturing process involving direct or indirect lost-wax casting, a more or less constant surface composition can be expected. However, the differences in lead and tin content between the body and shafts of most balls tell a different story. This discrepancy is perhaps due to local differences in the casting alloy itself or during the cooling of the bronze, as lead does not mix very well with copper. Or the bronze smith may have deliberately enriched the lead/tin on the surface of the ball to create a difference in colour or sparkle.

Further material research on the metallurgical structure of the bronze hollow balls, combined with a more detailed study of the inside and outside surface, could shed further light on the production method of the hollow balls. For instance, a study of the structure of the dendrites, the tree-like crystals, in the bronze would reveal the direction in which the metal was poured into the mould.

10.3.4 Other horse tack fittings

This category of horse tack fittings consists of various types of objects: rings in various sizes from the Heumen grave and two small rings from Nijmegen-Traianusplein and bronze connecting elements and tubes from Wijshagen mound H, belonging to the bridle of a horse. The larger rings of Heumen are probably related to the horse bits. How and where the smaller rings were attached is as yet unknown. W-20, a ring from the northeast quadrant of mound C, appears as an outlier in Figure 10.15. The bronze alloy of this ring contains far more lead and antimony then the other items.

As with the other objects (phalerae, hollow spherical balls), the silver content has proven to be an excellent discriminator between the sites. Most of the Wijshagen horse tack fittings have a very low silver content but varying contents of lead and antimony. The Heumen horse tack all contains silver and are clustered together in the arsenic-antimony and silver-arsenic plots. The two Nijmegen-Traianusplein horse tack fittings (rings) have very low silver, antimony, arsenic, nickel and bismuth contents and seem to be made of similar bronze.

Again, like the other metal find categories, the sites have their own trace metal signature. Although there is an overlap between some of the

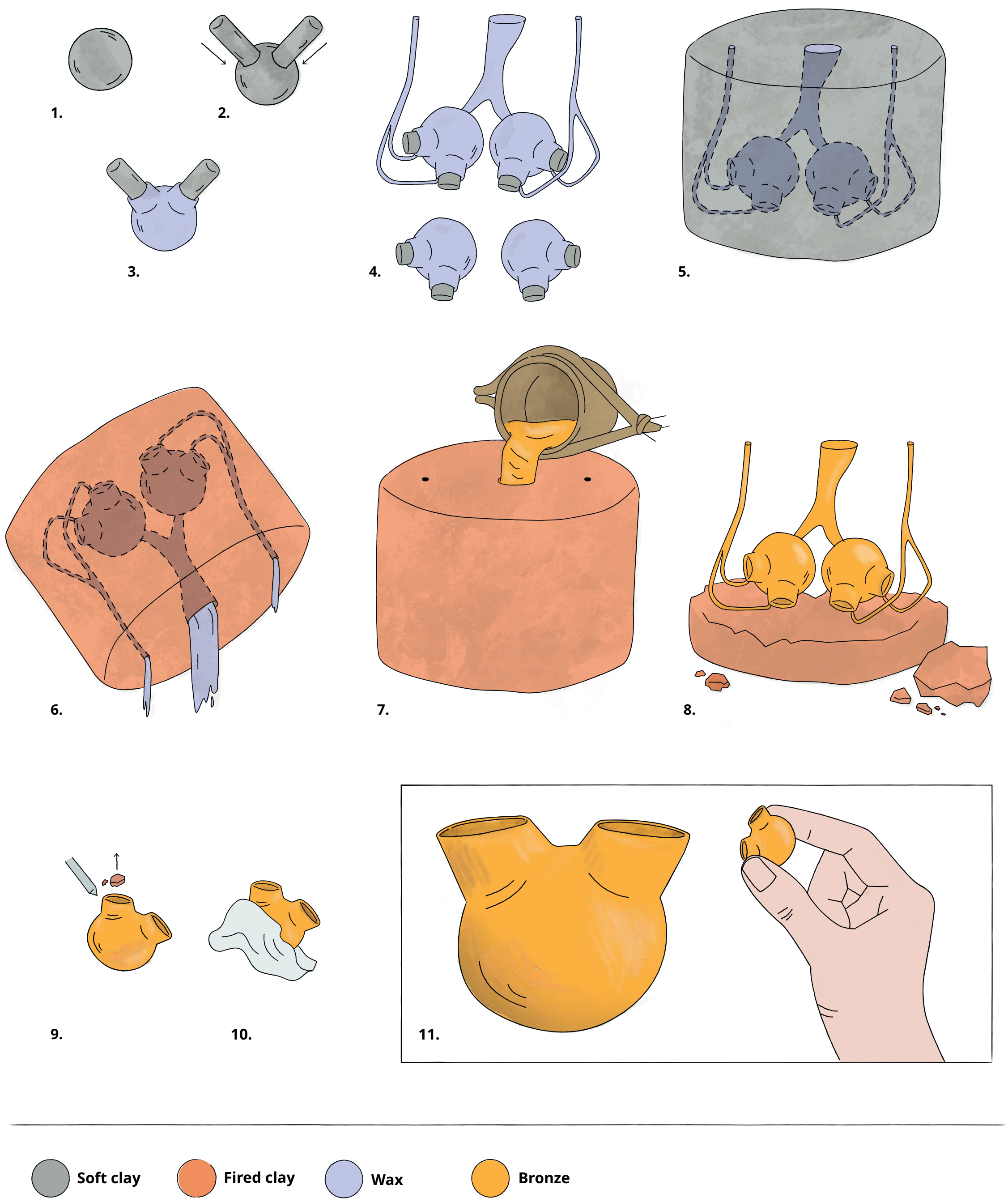
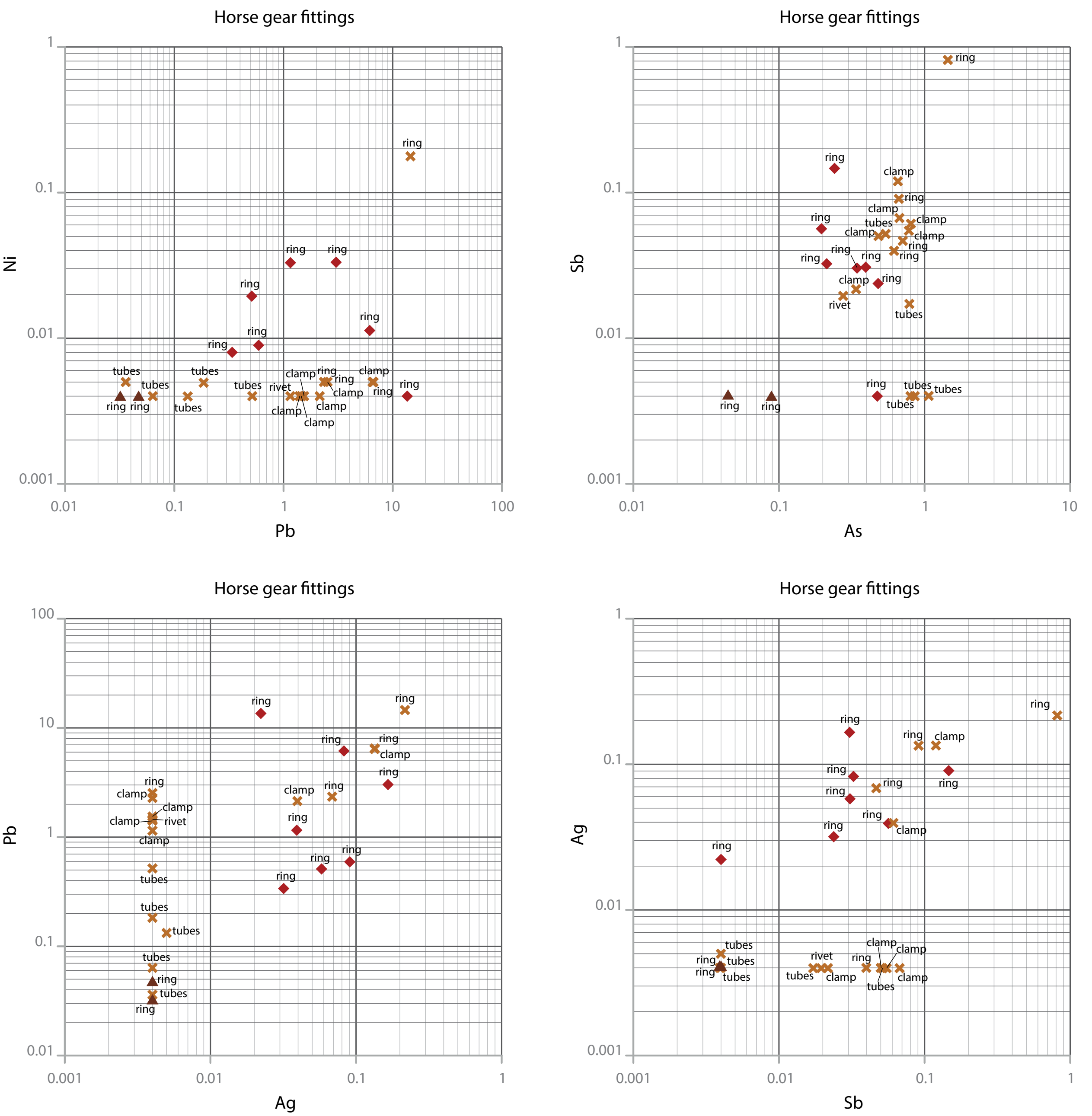


Fig. 10.14. Different steps in the making of a set of hollow spherical balls by direct casting using the lost-wax method (inspired by Huan/Jingnan 2022, fig. 8).

horse tack of Wijshagen and Heumen, which may point to a similar workshop or a bronze metal pool with similar characteristics depending on bronze circulation and new material input, three different types of copper for bronze production can be identified. Lead has been added to some of the bronze, most likely to facilitate better castings for some of the rings. However, the amount of lead is generally lower than 2%.

Comparing the different horse tack parts from one elite grave, such as Wijshagen mound H and Heumen (Fig. 10.16), the different clusters of the phalerae, hollow spherical balls and tubes stand out in the Wijshagen graph, whereas the cluster-



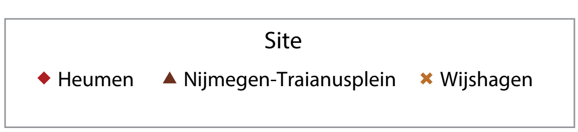


Fig. 10.15. Graphs of the trace elements (nickel-antimony-silver-lead) of the horse tack fittings in the elite graves of Heumen, Nijmegen-Traianusplein and Wijshagen (RCE).

ing of the phalerae and hollow spherical balls is clear in the Heumen graph. The latter indicates that the horse tack was made in the same workshop, using fairly similar alloys (only the ratio of lead differs slightly). This is an interesting outcome, as we assume that the Heumen horse tack belonged to three different horses. For the Wijshagen mound H horse tack, it is assumed that the objects belonged to a single riding horse. The trace elements of the different parts point to an assemblage of products from different workshops, or at least different alloys.

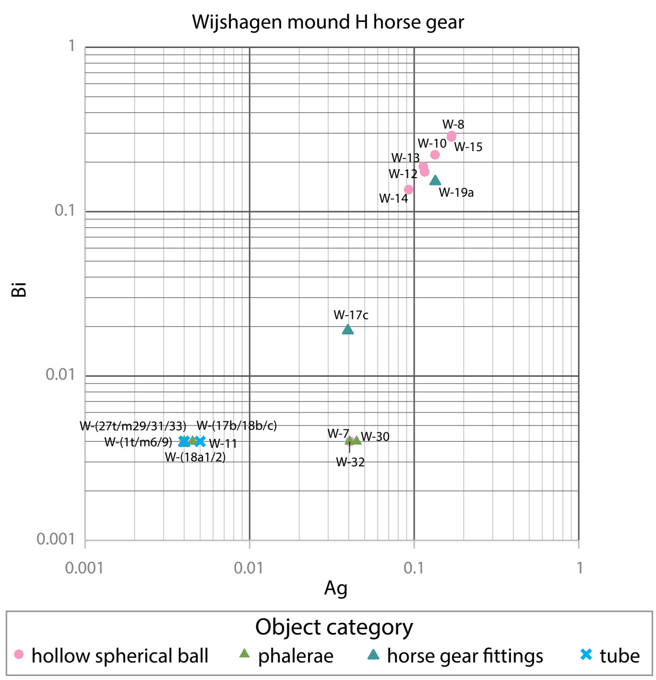


Fig. 10.16. The graphs of the trace elements of the horse tack of Wijshagen mound H and Heumen (silver-bismuth) (RCE).

10.3.5 Bronze wheel parts of the Heumen burial

The chariot of the Heumen elite burial contained the remains of four bronze nave bands, severely affected by the heat of the pyre (Sect. 2.3.4). Analyses of the nave bands revealed that they have an almost identical bronze composition (high tin bronze with about 2% lead). In particular, their trace elemental composition of Sb, Bi, Ag and As is almost identical. It is highly likely that the bronze bands were all cast from the same material (Fig. 10.17).

The bronze composition of a nail in the axle cap and the decorative bronze wire embedded in the iron linchpin is also very similar. It seems that both objects were made from the same bronze alloy. In comparison to the nave bands, only their nickel content differs slightly. No lead was found in the nail in the axle cap or in the wire, indicating that they were forged rather than cast.

The identical composition of the four nave bands indicates that they were made and used as a set. There are no indications that any of these bands were replaced.

10.3.6 Body ornaments from Wijshagen

Two pieces of bronze ornaments were discovered in mound C of Wijshagen: a fragment of a knobbed bracelet in the southwest quadrant and a fragment of a torc in the northwest quadrant (Sect. 5.3.4.1-2). The XRF results indicate that the bracelet was made of a leaded bronze alloy (22.5%wt). Leaded bronze was often used to create ornamental details.

The Wijshagen bracelet, typologically identifiable as an arm-ring with studded or knobbed plastic ornamentation (Arnoldussen/Steegstra 2019/2020, 87-8), ties in well with what we know from other parts of Northwest Europe where leaded bronze alloys were used for Early Iron Age fibulae (Schwab 2014, 177) and Early Iron Age grave furnishings (Giumlia-Mair *et al.* 2003, 161). A recent study on Dutch bracelets shows that most of the bracelets, dating from Period V (c. 925-750 BC), were made of leaded bronze (9-21%wt) (Arnoldussen/Steegstra 2019/2020, 50). Besides this leaded bronze, unleaded bracelets also occur with no silver, nickel or antimony and arsenic in the range of 0.24-0.77%wt. A remarkable outcome of this study is that arm-rings of similar appearance were constructed from dif-

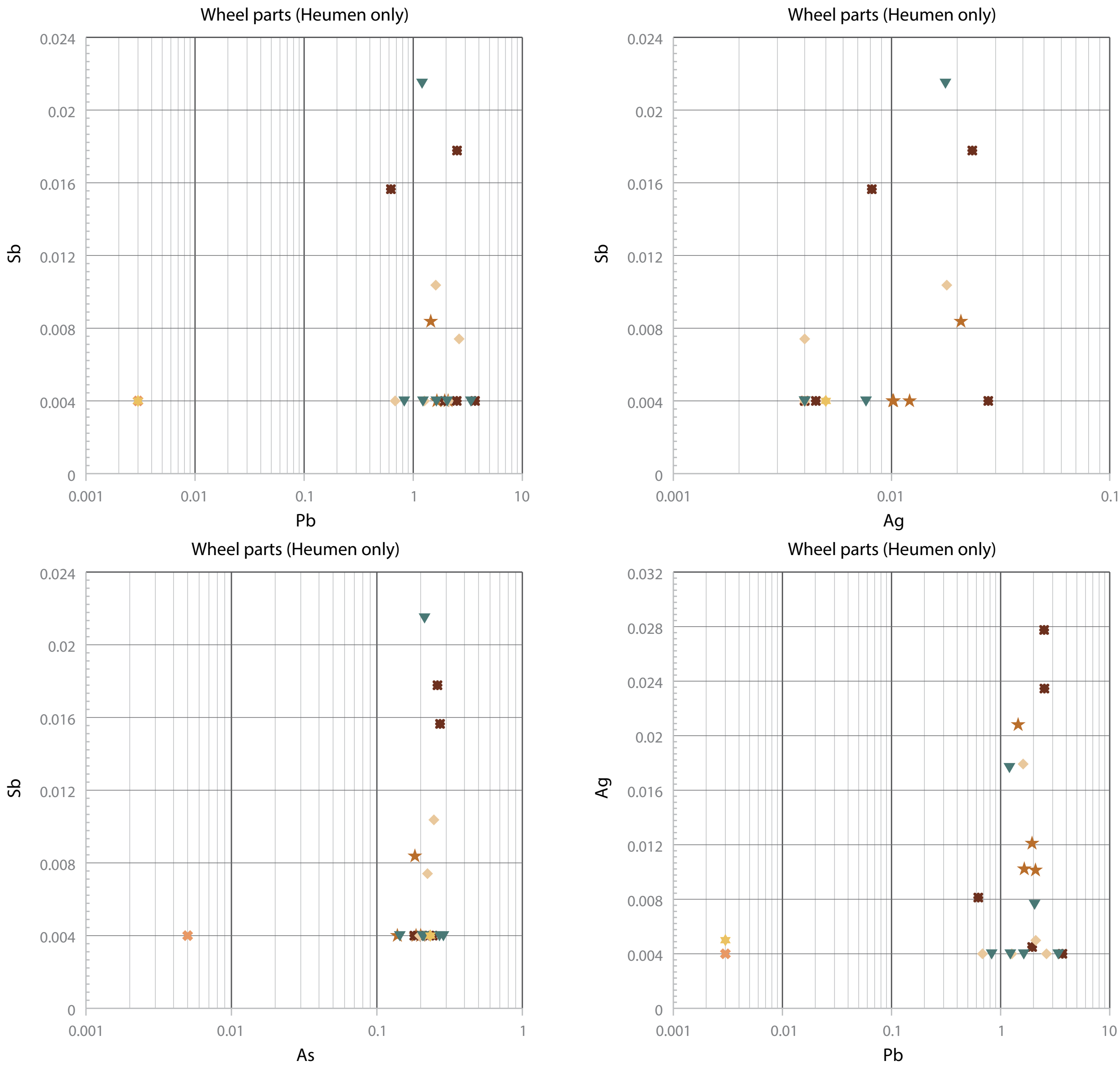


Fig. 10.17. The graphs of the trace elements of the wheel parts of Heumen: the four nave bands, a nail of the axle cap and bronze wire in the linchpin (lead-antimony-silver-arsenic) (RCE).

ferent base alloys, hinting at local fabrication or the copying of artefacts using different smelts.

The decorated torc, in contrast, is very low in lead (0.07%wt) and slightly higher in arsenic (0.83 versus 0.65%wt) in comparison to the knobbed bracelet. The fine decoration was probably added later, after the torc had been cast and cooled down. As stated above, the absence of lead in bronze allowed for fine cold-working techniques such as engraving, embossing and repoussage (Ashkenazi *et al.* 2012).

10.3.7 The bronze bowl from Overasselt

The bronze bowl from Overasselt is a remarkable object for Early La Tène grave goods, and without parallels for this period and cultural area (Sect. 3.2.2.2). However, many drinking bowls have been found in Hallstatt C and D elite burials north of the Alps, in combination with cauldrons and situlae (Kimmig 1991; Krausse 1996, 2003). The Overasselt bowl, for instance, resembles the silver *phiale* from Vix (Krausse 2003; Rolley 2003,

Fig. 10.18. The fragments of the open bowl of Didam-Kollenburg Noord, made of sycamore wood. Two small holes, which appear to have been made intentionally, are present in the base of the bowl (Van der Leije/Van Hees, fig. 2, 130 © Archol/ADC).

157-60). In addition, bronze bowls with an omphalos bottom are known from Moravia (Golec *et al.* 2022) in Hallstatt D2 contexts.

The XRF data shows that the bowl of Overasselt is made of tin bronze with a very small amount of lead (0.05%), antimony (0.06%) and silver (0.03%). The lead content is lower than in the bronze sheets of the situlae of the Rhineland-Tessin type (see Fig. 10.2). Considering that this piece is thought to be cast in bronze, based on the thickness of the material (2-3 millimetres) and the protruding ornamental decoration, a much higher lead content in the bronze was expected. We can conclude that this cup, as a cast product, differs from all other bowls from this period, which were all forged and hammered from bronze sheet metal. The absence of any clear parallels makes it difficult to link it to a region or workshop.

Also striking is the smooth execution of the ribbed decoration on the 2-3-millimetre thick wall of the bowl. This suggests that the wax model could have been created using a mechanical instrument. Woodturning is known in the south of Germany from the Early Iron Age onwards (Kossack 1959, 105-7). Two wooden bowls, recently found at Didam-Kollenburg Noord and dating to the Early to Middle Iron Age, are the only examples from the Netherlands made on a lathe (Van der Leije/Van Hees 2022). Both were found in fragments in the fill of a water pit. The open bowl was at least 34 centimetres in size and 11 centimetres high. The 1-centimetre-thick outside wall has three cut-out line decorations (Fig. 10.18). It was made from sycamore wood (*Acer cf. pseudoplatanus*), which did not occur locally at that time, suggesting that it was not locally made. In this light, it is tempting to see the Overasselt bowl as a bronze imitation of a bowl made of wood and regard it as an import. Another possibility is that the bowl was made locally as a copy of an imported wooden specimen.

Future metallurgical investigation may reveal more technological details on how the bowl was cast and its surface smoothed on a lathe. In this future research, it is important to keep an open mind for testing less obvious options as cold working (hammered). If the bowl was made from cold-worked plate material, it is very likely, given its shape and composition, that it was made in a workshop in northern Italy or the east Alpine region. In that case the bowl was an imported item that arrived with the situla as a set. Hopefully, future discoveries of similar bronze bowls will shed more light on this issue.

10.4 Conclusions

In conclusion, we have shown that handheld XRF results allow us to identify distinct alloy groups and clues as to production methods. More generally, the fact that we can identify distinct groups that are specific to a site suggests that metal objects in these Early La Tène elite graves were made and used as a set and were mostly kept together during their lifetime. For the horse tack items, this points to a relatively short lifespan, as replacing items would have led to greater variation in shape and composition and the separation of pairs or triples. Replacing, exchanging or inheriting these bronzes would undoubtedly have led to more variation in shape and composition within a find category and site. Instead, the objects were kept together as a package before being deposited as part of the burial ritual. The situlae, on the other hand, show clear signs of repair, suggesting a longer period of use, perhaps as heirlooms.

10.4.1 Provenance

The XRF data on the finds from Nijmegen-Hunerberg, Overasselt and Heumen show a large overlap in their values for nickel, silver, antimony, bismuth and arsenic. This points to a common pool of bronze or copper from the same fahlore mines. High silver, arsenic and antimony but low nickel bronzes are diagnostic for Ösenring copper that was used in Early Copper Age material, but is also seen in Late Bronze Age bronzes.[9] This material is thought to have been mined in Central Europe (Moravia) or the eastern Alps.[10] Future lead isotopic studies could reveal the provenance of the copper ore. The copper for the situlae is most likely to have come from Mediterranean sources such as Tuscany, Cyprus or Greece (Laurion) (Chiarantini *et al.* 2018), whereas the copper from more local objects such as the phalerae and spherical balls came from deposits that were nearer at hand, such as in Central Europe or the eastern Alps (as indicated by their higher antimony and arsenic contents).

10.4.2 Metallurgical groups, technological aspects and workshops

The composition of the four situlae of the Rhineland-Tessin type points to batch production as all rivets have more or less the same composition and contain more lead than the bronze sheet material. The sheet metal of the situlae from Heumen, Overasselt and Wijshagen shows a similar composition, although their rivets contain a varying amount of lead and trace metals. The ribbed bucket from Wijshagen has a different trace metal composition, pointing to another source or workshop of origin. The rivets of the situlae are made from a leaded bronze alloy and were most likely cast, whereas the bronze sheet of the body and bottom was forged and cold hammered. As the situlae were most likely made in the southern Alpine region of northwest Italy, the copper very likely came from Tuscany, Cyprus or Laurion. The rivets, however, have a similar trace element signature to the other metal categories and could have been made locally, suggesting that the situlae were repaired in local workshops.

The phalerae are also cast, the ones from Heumen and Traianusplein as composite, cast-on objects, made of precisely produced and tight-fitting elements. The mould marks on some of the Wijshagen phalerae suggest that a re-usable, two-part,

9 Mödlinger/Trebsche 2020; Pernicka *et al.* 2016; Van der Sanden/Van Os 2021.

10 Chiarantini *et al.* 2018; Radivojević *et al.* 2018, 2019; Tropper *et al.* 2019.

fine-grained stone or bronze mould was used. It is also possible that impression moulds were created.

For the hollow spherical balls, with their complex shape involving curves and straight shafts, the lost-wax process is the most likely method of manufacture. This idea is supported by the high lead content of the balls in general, but the variation in lead and tin between the body and shafts of most balls is difficult to explain. This could be a result of the casting process itself or a deliberate act such as metallurgical enrichment for colour/sparkling. The compositional signature also implies that the balls were produced as a pair – or even as triples – in one casting activity, as a batch-wise production from the same alloy. This same pattern – the occurrence in pairs or triples – is also visible in the compositional signature of the phalerae. Paired or serial production in one workshop seems to have been rather common.

The elite burials of Heumen and Overasselt bear strong similarities in several respects. The composition of the sheet metal work of both situlae, as well as the compositional signature of the phalerae and the hollow spherical balls, all show similar trace elements, which points to a specialised workshop. The phalerae are assumed to be imports from southern (possibly French) regions (see Ch. 14), while the balls may have been produced in the Lower Rhine-Meuse region. The fahlore signature of the phalerae and hollow balls points to a Central European or eastern Alpine copper source. The similarities of Heumen and Overasselt are inconsistent with the horse tack of Wijshagen mound H, suggesting an assemblage of products from different workshops (or alloys).

To summarise, XRF results allow us to draw conclusions about important questions regarding Early La Tène bronze grave goods, their provenance, and metallurgical and technological aspects. However, it is the multidisciplinary approach, such as the use of neutron-computed tomography, that brings more insights and raises new questions. Future research on this topic should be designed as a multidisciplinary framework, encouraging cooperation between experts on different techniques and experimental metal work, such as craftspeople. This will shed new light on the bronze metal work, exchange networks and connectivity of Early La Tène societies.

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