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A not so isolated fringe: Dutch later prehistoric (c. 2200 BCE-AD 0) bronze alloy networks from compositional analyses on metals and corrosion layers

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ABSTRACT

Using a corpus of over 370 compositional analyses of Dutch Bronze Age and Iron Age (*c.* 2000 BCE AD 0) copper alloy artefacts, long-term patterns in the types of alloys used for specific bronze objects are identified. As the Low Countries are devoid of copper ores and alloying elements, a combination of typo(chrono)logical and compositional analysis is used to identify through which European contact networks (such as Atlantic, Central European or Nordic exchange networks) these alloys were obtained. We employ a methodology that (following Bray et al., 2015) defines alloy groups by presence of As, Sb, Ag and Ni over 0.1 %wt, but expanded this classification to include Pb and to track high-impurity (>1%wt) alloys. Due to interfering soil-derived iron hydroxides, and preferent dissolution of copper from the objects' surface, the determination of tin is in most cases overestimated when using p-XRF, so Sn was not systematically reviewed. Objects were assigned a calendar age in years BCE to facilitate chronological sorting. Using this classification, we could show how different alloys (using different base ores) were used in different periods, and in different combinations. Moreover, particular alloys were used for different groups of functional types of objects. Also, we show diachronic differences in the influx of new (or less frequently mixed) alloys and chronological trends in the substitution of As by Sn as main alloying element in the Early Bronze Age as well as the rise of leaded alloys at the close of the Bronze Age. Combining information on the composition of the objects with their typological traits, allowed us to reconstruct the scales and geographic scopes of the European contact networks in which the copper alloys used throughout later prehistory were obtained.

1. Introduction

1.1. Problem definition

During the Bronze Age (*c.* 2000–800 BCE) the Netherlands were integrated into wider networks of contact and exchange. Every single bronze object from this period recovered forms a tangible proof thereof, as the area is devoid of ores (copper, tin) required for the production of bronze alloys (Arnoldussen, 2015, 17). The extent, duration and prime motivations for such – no doubt series of shifting – networks are more difficult to chart. Compositional analyses of the alloys, that show variations over time and correlate to specific functional types, can provide tantalizing clues on the presence, content and geographic scope of such contact networks. An example of such a temporal shift is the transition from arsenical copper to tin-bronzes at the start of the Bronze Age. As an example of the correlations between alloy groups and functional types, it can be shown that for the production of bracelets, other alloys are employed than those used for axes. However, for the Netherlands, compositional analyses of copper alloys dating to *c.* 2200 BCE-AD 0 have never been systematically compiled into a coherent corpus and analysed integrally. Therefore, this contribution aims to show how – over time – alloys of particular composition were obtained from different contact networks (*e.g.* Atlantic, central European, Nordic) and investigates whether particular alloys were favoured in certain periods or used preferably for specific object types. A specific challenge is using the corrosion layers on these objects to study trends in alloy composition.

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1.2. Bronze Age exchange networks in diachronic view

During the final neolithic and earlier Bronze Age (*c.* 2200–1700 BCE), the Rhine river has been postulated as a possible corridor for the 'amber routes' linking the Baltic and Mediterranean worlds (Harding, 2013, 376 fig. 202.2). Simultaneously, prestige goods such as faience beads circulated in a maritime zone (Needham et al., 2006, 75; 2009) encompassing the British Isles and a coastal fringe of continental Europe (Sheridan and Shortland, 2004, 269 fig. 21.5; 270; Haveman and Sheridan, 2006; Arnoldussen, 2014, 19). As early as in the start of the Early Bronze Age, axes of evident insular style traveled into the Low Countries (O'Connor and Cowie, 2001, 225; Jockenhövel, 2004, 156; Arnoldussen et al., 2020a, 45 fig. 3). Helpful as such 'smoking guns' may be, they have not yet led to any uniform mapping of contact networks for the Low Countries during the Early Bronze Age. Rather, the area is (partly) left blank, and/or mapped with varied labels/affinities or indicated as an impractically (and implausibly; *cf.* Needham et al., 2006, 75; Fokkens and Fontijn, 2013, 553) large geographical zone (*e.g.* Cunliffe, 1994, 255; Rassmann, 2002, 174 fig. 299a; Fokkens, 2005, 360 fig. 16.2; Mordant, 2013, 572 fig. 32.1a).

For the Middle Bronze Age (*c.* 1700–1100 BCE), the interaction networks in which the Low Countries are assumed to be integrated - as identified by funerary traditions, and stylistic traits of local (esp. pottery) and exchanged (esp. bronzes) items - are mapped with a similar broad brush approach: frequently placing the Netherlands in or at the boundary zone of Atlantic, Nordic and central European Tumulus Culture influences (*e.g.* Cunliffe, 1994, 248; 2008; 231; Rassmann, 2002, 174 fig. 299b; Fontijn, 2003; 77; 109; 143; Fokkens, 2005, 361 fig. 16.3; Mordant, 2013, 573 fig. 32.1b; Fokkens et al., 2016, 37). During the Late Bronze Age (*c.* 1100–800 BCE), the Netherlands are once more - depending on the (geographical) confines proposed by various authors - either well within the 'Atlantic zone' (*e.g.* Coffyn, 1998, 173; Harrison, 2004, 174; Quilliec, 2007b, 94), on the boundary (*e.g.* Brun, 1988, 602, 1991; figs. 3–4; Gouge and Peake, 2005, 336; Mordant, 2013, 574 fig. 32.1; Cunliffe, 2008, 257; *cf.* Deiters, 2008, 51; Verlinde and Hulst, 2010, 105–113), well within the 'Urnfield' and/or North-Alpine zone (Needham and Bowman, 2005, 125; Cunliffe, 2008, 231, *cf.* Cunliffe, 2009, 80) or left excluded from the various regions altogether (*e.g.* Brun, 2005, 140; Harrison, 2004, 167; Kristiansen, 2000, 87; Quilliec, 2007a, 121–123).

Evidently, there are grounds for a critical stance towards the extent, duration and contents of Dutch copper alloy contact networks. Paradoxically, the Bronze Age objects and their compositions themselves have played only a limited role in such discussions. They have served mostly to act as typological markers of the 'local' and the 'non-local' (*cf.* Butler and Fokkens, 2005, 382–383; Fontijn, 2008, 2009; Arnoldussen, 2015) and as chronological footholds. Whereas in a few studies the compositional analysis of alloys has been used to argue for specific non-local origins of alloys found in the Netherlands (*e.g.* Butler and Van der Waals, 1966, 81–2; Fontijn et al., 2012; Butler et al., 2014; Postma et al., 2017; Theunissen et al., 2017; Arnoldussen et al., 2020a, 2020b), these studies invariably pertain to single objects or single (multi-object) depositions. An overarching analysis of composition, age and origin for the full corpus of Dutch later prehistoric copper alloy artefacts has never been undertaken.

1.3. The potential of copper alloy datasets

We argue that using a sizeable corpus (*i.e.* over 370 analyses) of alloy compositions of copper alloy artefacts from the Netherlands, it is possible to better characterize the presence, content and geographic scope of contact networks in which such alloys were obtained. Also, such an overview of elemental compositions allows to trace diachronic changes in alloy preferences in general, and for functional object categories (*e.g.* swords, axes, ornaments) in particular. Such a diachronic and comparative perspective however requires a consistent way of representing the alloy composition of each object.

Historically, for the Low Countries there have been ample ways to represent and classify the alloys of prehistoric bronzes, and frequently tabulations and visualisations were chosen that allowed to highlight what elements (in addition to the main constituent copper, common alloying element (Sn, As, Pb) and minor or trace elements (*e.g.* Sb, Ni, Ag) characterised the alloy. Waterbolk and Butler (1965; Butler and van der Waals, 1966; Needham et al., 1989, 384; 2002, 106 fig. 4) proposed log-scale histograms to depict main alloy composition and trace elements in single plots, but in the *Studien zu den Anfängen der Metallurgie* (SAM) project (Junghans et al., 1960, 1968, 1974) a decision-tree approach was forwarded (Bray and Pollard, 2012, 854) that favoured compositional grouping. This was later refined for the British Isles by Northover (1977, 1980) who proposed the now well-known 'A' to 'H' alloy types. Whereas such classificatory approaches provide easy shorthands, they also tend to steer interpretations in terms of priority of certain elements (early tree decisions) and downplay within-group variation. Fortunately, most copper alloy repositories and collated datasets have retained primary compositional data (*e.g.* SAM (Junghans et al., 1960, 1968, 1974); OXSAM/OXALID (https://flame.arch.ox.ac.uk; Bray, 2009), Moving Metals (Ling et al., 2019 for references)). This allows new analyses to be undertaken that both take in isotopic approaches (*e.g.* Pollard, 2009; Pollard and Bray, 2015; Ling et al., 2019; Williams and Le Carlier de Veslud, 2019), but that are also more sensitive to discriminating between base (ore/alloy/ingot) composition and the effects of production strategies and recycling/mixing (Needham, 2002, 100; Bray and Pollard, 2012, 854–855; Bray et al., 2015; Pernicka, 2014; Nørgaard et al., 2019, 14–15; Radivojević et al., 2019, 156).

The corpus presented here reflects a long trajectory of compositional analyses, from initial chemical analyses from the 1930's, to spectral analyses undertaken in the 1960's and 1970's, but has been significantly boosted by the availability of handheld-XRF technology in the last two decades. It must be stressed that the dataset used as a base for this study is derived from a range of different analytical techniques and sample procedures that have been used and developed over a long period of time. Older analyses were done on pure metal samples, but a major portion of the dataset was collected using XRF surface measurements (*infra*; Fig. 2). The non-destructive character and ease of use, however, of this technique, makes it the only possibility for analyses of many archaeological and museum artefacts. However, these measurements can be seriously affected by corrosion processes, to the degree that it effectively measures the corrosion layer (*cf.* Vittiglio et al., 1999; Orfanou and Rehren, 2015; Nørgaard, 2017). Depending on the type and degree of corrosion, alloying elements like Sn, Zn, Pb Sb and As can concentrate or be depleted in this layer. For example, lead is present in copper alloys as a separate phase, often recognizable as globules. Lead is always present as an oxide present under current (soil) conditions that forms a passive layer around the globules that are at the outside of the object in contact with soil or atmosphere. As copper will dissolve preferentially to lead and tin under soil conditions in the presence of oxygen and water, the relative concentrations of the latter elements will increase. Therefore, on corroded copper alloy surfaces lead concentrations will be heightened with respect to the bulk lead concentration. As discussed at greater length in Roxburgh et al. (2019), such processes would hinder the use of p-XRF analyses in a pure 'provenancing' role. However, the variations in their alloy composition, despite deviations by corrosion effect, makes it still possible to address aspects relating to human interactions, use of specific alloys, recycling/mixing and other aspects that deal with socio-economic trends over larger time-frames (*cf.* Fernandes et al., 2013). This does, however, require large enough datasets to account for variability in type and degree of corrosion and - in the case of legacy datasets as discussed here - differences between analytical techniques. The present paper does specifically not aim to further this debate (but see Van Os et al., in prep.). That being said, some objects have been measured using various techniques or using both core and corrosion samples. Principally, however, we have to take the fact that over ¾ of our data is derived from p-XRF measurements at face value and have tailored our methodology accordingly.

2. Material and methods

2.1. Data origin

Here, we report on a total of 375 compositional analyses, representing 295 objects (Fig. 1/ES4). For the majority (n = 371) of objects,

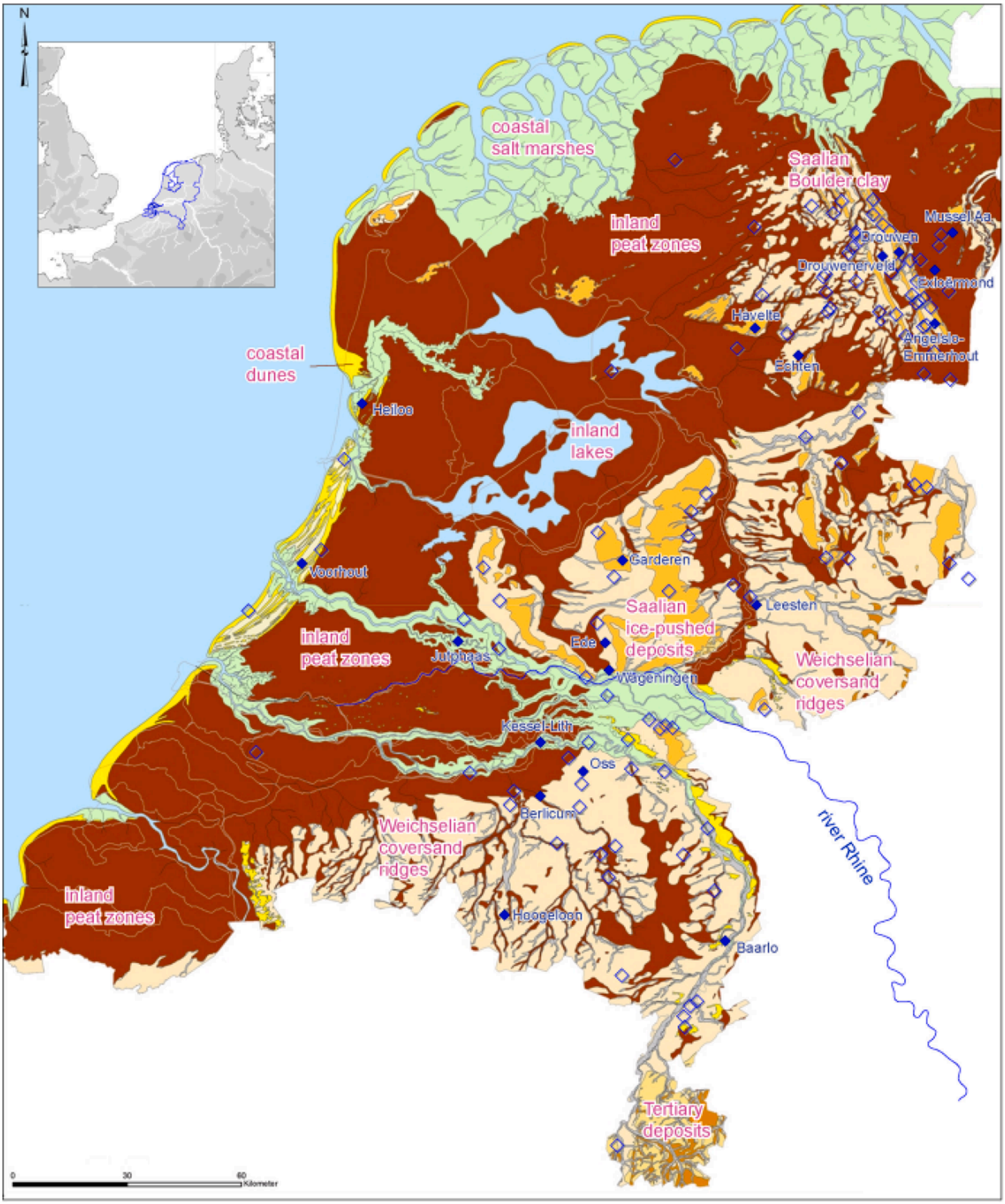


Fig. 1. Map showing the spatial distribution of the data in the corpus (solid blue diamonds mentioned in body text, open diamond other corpus findspots (see ES4 for a full map)), overlying a palaeogeographic map of the Netherlands around 1500 BCE (Deltares/RCE: after Arnoldussen et al., 2011). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

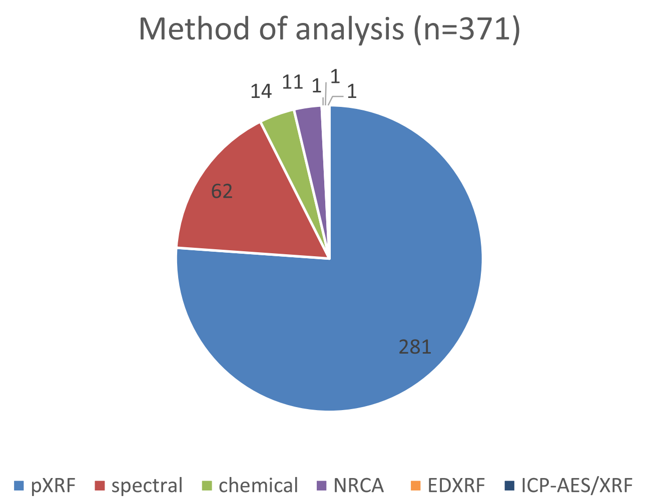


Fig. 2. Characterisation of method of analysis for the copper alloys in the corpus (pXRF = portable X-ray fluorescence, spectral = spectral analyses undertaken by the Studien zu den Anfängen der Metallurgie (SAM) project (Junghans et al., 1960, 1968, 1974), chemical = wet chemistry analyses done prior to 1960, NRCA = Neutron Resonance Capture Analysis, EDXRF = Energy dispersive X-ray fluorescence, ICP-AES = Inductively coupled plasma atomic emission spectroscopy, XRF- = X-ray fluorescence).

the method of compositional analysis was known (Fig. 2). Portable XRF measurements dominate, followed by optical emission spectral analyses (n = 62), and limited numbers of other techniques such as chemical (n = 11) or neutron resonance capture analysis (n = 11). In part (n = 100), the information is taken from published sources (some dating back to 1904; Jacobsen, 1904), but the other (n = 276) analyses reflect data that were either taken but never published (n = 72) or represent targeted p-XRF analyses undertaken by the authors (n = 204). Between 2015 and 2022, the authors have undertaken p-XRF studies of the prehistoric collections at the National Museum of Antiquities (Leiden, 2015), the Groningen Institute of Archaeology (Groningen, 2014–2020), Museum Twentse Welle (Enschede, 2018), and the Drents Museum (Assen, 2019). Moreover, an additional series of bronze artefacts recently found by metal-detectorists were analysed by the authors (*e.g.* Arnoldussen and Visser, 2014; Arnoldussen et al., 2020a; 2021; 2022; Theunissen et al., 2017) and which contributed further to the corpus of finds analysed.

For the 204 analyses undertaken by the present authors, more details of the methodology and setup is known. These measurements were taken with a Thermo Scientific NitonXL3t, that analyses up to 25 elements simultaneously (covering the elemental range from sulphur (atomic nr. 16) to uranium (atomic nr. 92) and which can moreover detect lighter elements (in the range of magnesium (atomic nr. 12) to chlorine (atomic nr. 17)). For calibration, factory standards and reference samples were used. Locations were measured using the 'small spot' beam in 'Electronic Mode' for a duration of 30–69 s. For 127 objects, multiple measurements were taken across different areas of the patina, allowing to better approximate original composition using averages (n = 37), selection of highest Cu content (n = 56) or compensation for iron content (n = 34; see Van Os et al., in prep. for method).

2.2. Data: Functional categories

In terms of functional categories, axes (n = 132, 35 %) dominate, followed by swords (n = 38, 10 %), bracelets (n = 36, 9 %) and spearheads (n = 33, 9 %). Daggers (n = 27, 7 %) make up the only other object category that is represented over 5 % (Fig. 3). This imbalanced distribution is not problematic, as particularly axes (Butler, 1995/1996; Butler and Steegstra, 1997/1998; 1999/2000; 2003/2004; 2005/2006), swords (Butler and Fontijn, 2007; Fontijn et al., 2012; Van der Sanden and Arnoldussen, 2017) and bracelets (Arnoldussen and Steegstra, 2021) are (1) published in more detail (and/or as overview studies) than other object categories, (2) show ample diachronic variation, and thus allow for (3) greater (typo)chronological control.

2.3. Data: Chronology

In terms of chronological control, individual date-ranges for the objects have been added, as well as midpoint ages to allow chronological sorting of the data-set. Ultimately, these are proxy dates of typo-technological traits, relying heavily on the proposed object date-ranges proposed by Fontijn (2003, esp. 56 fig. 5.2; 87 fig. 6.2; 117, fig. 7.2; 153 fig. 8.2), checked or refined by absolute dates where

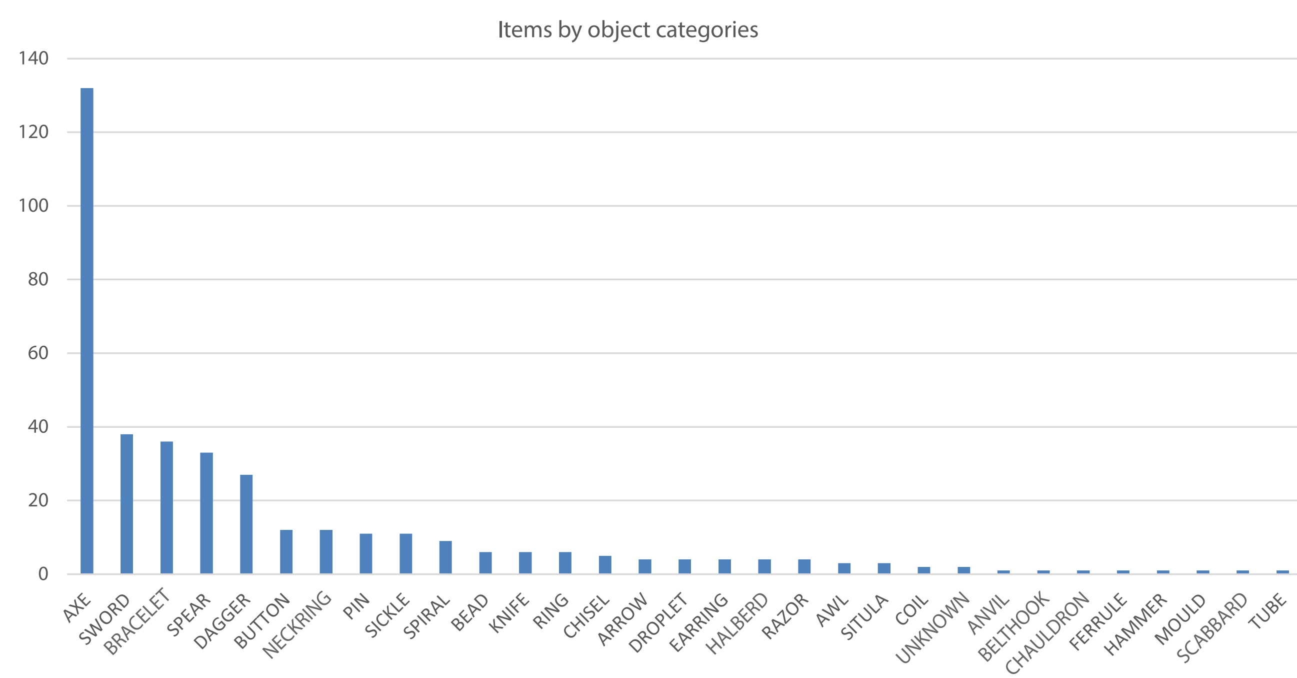
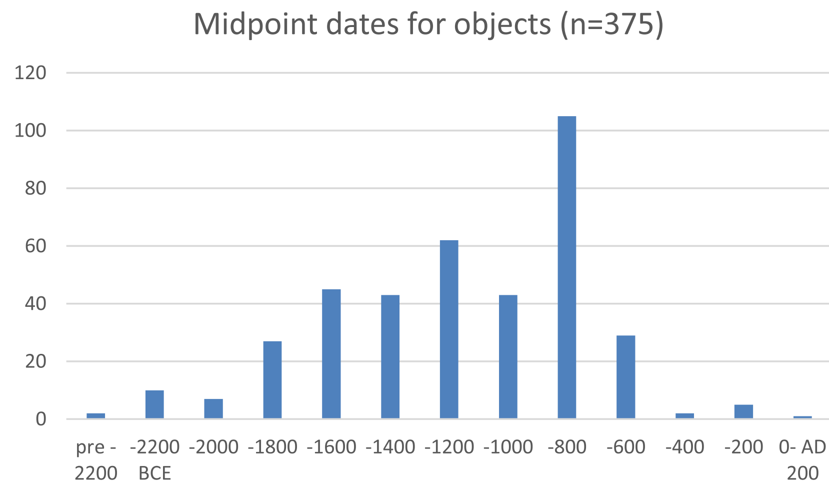


Fig. 3. Characterisation of typological variability of the compositional analyses, based on counts of main artefact types.



Period	Age (BCE)
1	pre 2200
2	2200 - 2000
3	2000 - 1800
4	1800 - 1600
5	1600 - 1400
6	1400 - 1200
7	1200 - 1000
8	1000 - 800
9	800 - 600
10	600 - 400
11	400 - 200
12	200 - 0 AD

Fig. 4. Characterisation of diachronic variability of compositional analyses, based on frequency of midpoint dates using 200 yr bins. To the right the table with periods and calendar dates is shown.

available (*cf.* Drenth and Brinkkemper, 2002, 20 table 1; Arnoldussen and Visser, 2014; Arnoldussen et al., 2020a, 2022). These typochronological characterisations allow for an – albeit crude – sequentially reliable chronological trajectory to place the data onto (Fig. 4).

It is clear that the dataset is skewed, with an overrepresentation of finds attributed to the Late Bronze Age-Early Iron Age period (c. 1100–800 BCE), but it should also be stressed that the span from Late Neolithic-B (c. 2400–2000 BCE), to Middle Iron Age (c. 600–400 BCE) is presented with no notable gaps. Using 200 year period bins, all but the Early Bronze Age (c. 2000–1800 BCE; 7 objects) are represented by >10 objects. Here, a more fine-grained chronological trajectory is favoured over larger bin sizes (and thus larger population per bin size), as even with 500 year bin duration statistically robust populations cannot be achieved.

2.4. Methods: Characterizing alloys

Here, we essentially follow the methodology proposed by Bray (et al., 2015, 205) for classification into alloy groups (Table 1), based on the difference between presence/absence of the main secondary (after copper) elements antimony, arsenic, silver and nickel (using 0.1 %wt as threshold values; Fig. 1). For alloys that display such elements in the 1 %–5%wt range, we add their elemental abbreviation to the numerical codes proposed by Bray et al. (nos. 1–16; 2015, 205 fig. 1). Moreover, we added an extra column in which the lead content can be registered (0 = < 0,1 %wt, 1 = 0,1–1 %wt, 2 = 1–5 %wt, 3 = 5–10 %wt and 4 = > 10 % wt).

For example, an axe showing 2 %wt As, 0.8 %wt Sb and 1.2 %wt Pb, would be classified as Alloy vGroup 9As-Pb2. The proposed revisions (compared to Bray et al., 2015, 205 fig. 1) serve different purposes. First, concentrations of volatile elements such as Sb and As diminish during oxidative heating of melted bronze (Tylecote et al., 1977, 329; Junk, 2003, 7; Merkl, 2010, 21; Bray et al., 2015, 206), which means that (the frequencies of occurrence of) alloys high (>1 %wt; *cf.* Pollard et al., 2015, 699) in Sb and As could signal the use/influx of fresh ores/ingots, and shifts from oxidic to sulphidic ores. Second, the addition of a column documenting the lead content, allows to trace the rise of leaded bronze after the Middle Bronze Age (*cf.* Van Impe, 1994; Wouters, 1994, 42; Johannsen, 2016, 153; 158 fig. 3; Radivojević et al., 2019, 166).

Despite being a common element of Bronze Age copper alloys, we have chosen not to include tin as part of the main alloy categorization. Principally, as most analyses pertain to surface p-XRF measurements and the fact that corrosion layers have a known tendency to overrepresent tin ('tin-sweating', or rather depletion of soluble elements in the burial environment; Meeks, 1986, 133; Wouters, 1994, 45; Orfanou and Rehren, 2015, 392; Nørgaard, 2017, 102; 105–106), it was excluded as a determinant element. Second, tin ores do not contribute to the trace elements studied here and are thus nondiagnostic in establishing (changes in) copper ore types. In our study, a high tin content (coupled with low copper content and high iron content) was interpreted as a caution that the measurements were presumably not representative for the bulk composition of the object. This almost complete dismissal of Sn, alas hampers the identification of the transition from arsenical copper alloys to tin-bronze proper (*cf.* Kienlin, 2013, 421 fig. 23.3; 425).

Table 1
Classification of alloys, adapted from Bray et al., 2015, 205 fig. 1.

Copper Category*	Copper with	As	Sb	Ag	Ni	Pb**
1	None	No	No	No	No	0–4
2	As	YES	No	No	No	0–4
3	Sb	No	YES	No	No	0–4
4	Ag	No	No	YES	No	0–4
5	Ni	No	No	No	YES	0–4
6	As + Sb	YES	YES	No	No	0–4
7	Sb + Ag	No	YES	YES	No	0–4
8	Ag + Ni	No	No	YES	YES	0–4
9	As + Ag	YES	No	YES	No	0–4
10	Sb + Ni	No	YES	No	YES	0–4
11	As + Ni	YES	No	No	YES	0–4
12	As + Sb + Ag	YES	YES	YES	No	0–4
13	Sb + Ag + Ni	No	YES	YES	YES	0–4
14	As + Sb + Ni	YES	YES	No	YES	0–4
15	As + Ag + Ni	YES	No	YES	YES	0–4
16	As + Sb + Ag + Ni	YES	YES	YES	YES	0–4

* Elements listed here occur with > 1 %wt.
** Pb 0 = < 0,1 %wt, 1 = 0,1–1 %wt, 2 = 1–5 %wt, 3 = 5–10 %wt and 4 = > 10 %wt.

2.5. Methods: cross-validating techniques

Two objects (both low-flanged axes datable to the Early Bronze Age) were analysed with three different techniques (chemical in 1934, spectral in 1966 and with p-XRF in 2019). These complementary analyses allow to reflect on the methodological differences (Table 2).

First, nearly all (n = 13) chemical analyses lacked information on arsenic and antimony (hindering comparison with other analyses). Second, for those objects for which both p-XRF, spectral and chemical alloy estimates were available, it is clear that that p-XRF performed better in establishing alloy composition (with notable better detection for Sb, As and Pb, albeit with moderate to severe overestimation of Sn; Table 2).

3. Results

3.1. Methodological considerations

The comparison between the different measurement techniques in Table 2 illustrates the challenge that is posed with legacy datasets that comprise analyses of metals and corrosion layers that were done with different methods (chemical, spectral, p-XRF). Some elements were not measured with all three techniques, while differences in concentrations between the different alloying elements can be attributed to differences in detection limits and precision of the techniques and heterogeneity of the alloy in addition to the effects of corrosion processes. Whilst corrosion processes are most probably the reason for observed elevated concentrations of Sn, As, Ag and in one case Sb, these differences would rarely affect the copper category (Table 1). Combined with a significantly large dataset, we are therefore confident that this classification is a robust approach and that occasional misclassifications due to instrumental differences or corrosion processes will not significantly disturb the overall temporal trends.

3.2. Diachronic shifts in As-Sn alloys

Despite the fact that we have chosen not to use tin as a criterion for alloy identification (*supra*), some diachronic information on the presence and amount of tin-alloying may still be glanced from a subset of the data filtered for copper content > 80 %wt (*i.e.* excluding samples most affected by 'tin-sweating'; Fig. 5). This shows that arsenical copper is rare beyond period 3, and that – on average – more tin is added to bronzes in periods 3–7 (after which the prevalence of leaded alloys (*infra*) reduces the average).

3.3. Diachronic shifts in alloy group preferences

Using the alloy categorization of Table 1 and after applying typological (mid)point chronological sorting, the presence and ubiquity of the various alloy groups can be outlined across the temporal trajectory (Fig. 6). It is clear that some alloy groups would remain in use over prolonged periods (such as alloy groups 2, 11, 12 and 16, that remained in use for over 600 years). Others, such as alloy groups 3, 4, 7, 10 and 15 have seen a much shorter and more punctuated use. Fig. 6 also shows that the numbers of objects fashioned in particular alloys is also variable: alloy groups 3, 10 and 15 are scarcely represented, whereas ample objects were crafted in alloys 12, 13 and 16. Clear diachronic trends in exclusivity (or rather: complementarity) of alloy groups can also be reconstructed. For example, in the decennia leading up to the Bronze Age proper (*i.e.* > 2000 BCE), the number of alloy groups current was limited (n = 5), whereas in the Late Bronze Age (period 8; 1000–800 BCE) no less than 14 alloy groups occur side-by-side. These observations merit a more detailed discussion of the diachronic patterns in copper alloy preference.

In the Early Bronze Age (period 3; 2000–1800 BCE), alloy groups 2 (n = 6, 21 %, *cf.* Needham, 2002, 109), 12 (n = 5, 18 %) and 16 (n = 8, 29 %) dominate. Dagger and halberd blades form the majority of the alloy group 2 finds: both the Wageningen hoard dagger and halberd are listed (Fig. 7, note that halberds may be even older: *infra*). Alloy group 16 contains three flat- and flanged axes of uncertain or doubtful provenance and also some of the Wageningen hoard items such as the awl, flat axe and halberd rivets (Butler, 1990, 68–71; 70 table. 2; Butler and Van der Waals, 1966, 76 Table X). The items in alloy group 12 comprise a flat axe, a flat axe/ingot and double axe of Westeregelen type (Postma et al., 2017, 52) as well as the blade of the Bargeroosterveld dagger (Glasbergen, 1956; 1960). While it is clear that the Wageningen hoard (Fig. 7) dominates the data-set for this period, the other items mostly concern stray flat- and low-flanged axes and two grooved ogival daggers.

In period 4 (*c.* 1800–1600 BCE, or the Dutch MBA-A), alloy groups 2 (n = 8, 18 %) and 16 (n = 15, 34 %) remain common like in period 3, but

Table 2
Comparison of alloy determination for objects measured thrice (chemical (c. 1930 CE), spectral (c. 1960–1970 CE) and p-XRF (c. 2005–2020 CE)).

Object	DB	Technique	Value	Cu-group	Pb-class	As-class	Sb-class	Ag-class	Ni-Class	Cu	Sn	Pb	Zn	As	Sb	Ag	Ni	Fe	Bi	Mn	References
Axe, flanged, low-flanged, emmen-type	79	Chemical	%wt	5 Ni	0				2	95.200	0.800	n.d.	n.d.	n.d.	n.d.	n.d.	2.800	0.140	<0,1	<0,1	Butler, 1995/1996, 186 cat. 48
Axe, flanged, low-flanged, emmen-type	79	Spectral	%wt	16 Sb Ni	0	1	2	1	2	n.a.	0.370	–	n.a.	0.200	2.300	0.700	2.100	–	–	n.a.	Butler and Van der Waals, 1966, table X
Axe, flanged, low-flanged, emmen-type	79	p-XRF	%wt	16 As Ag Sb	0	2	2	2	1	92.482	0.940	0.058	0.287	1.696	2.478	1.752	0.397	0.140	<0,1	<0,1	Butler, 1995/1996, 186 cat. 48
Axe, low-flanged; AXIE	66	Chemical	%wt	3	1	0	1	0	0	94.700	4.500	0.100	–	–	0.700	–	–	–	n.a.	n.a.	Unpublished note Drents Museum; letter to Van Giffen
Axe, low-flanged; AXIE	66	Spectral	%wt	16	0	1	1	1	1	n.a.	3.100	–	n.a.	0.200	0.200	0.310	0.150	–	–	n.a.	Butler and Van der Waals, 1966, table X
Axe, low-flanged; AXIE	66	p-XRF	%wt	16	1	1	1	1	1	80.011	17.352	0.197	0.116	0.731	0.533	0.655	0.237	0.379	<0,0	<0,1	Arnoldussen and Steffens, unpublished

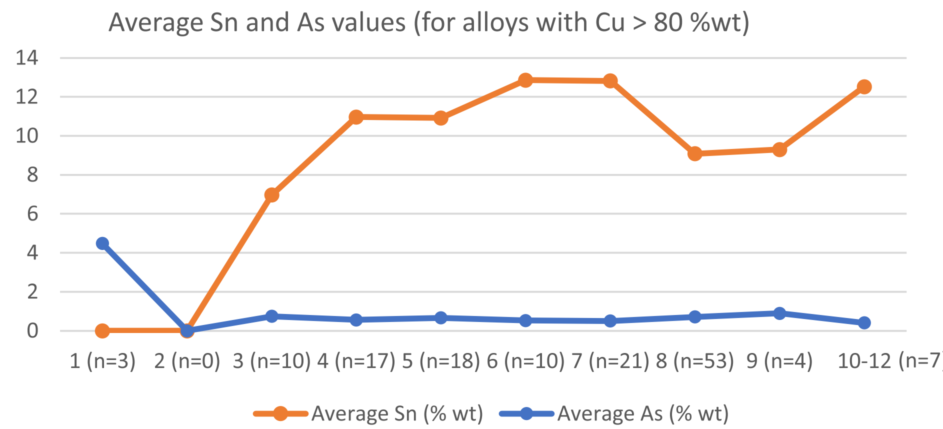


Fig. 5. Average values for tin and arsenic for those alloys with over 80 %wt in copper, plotted by period (for periods see Fig. 4).

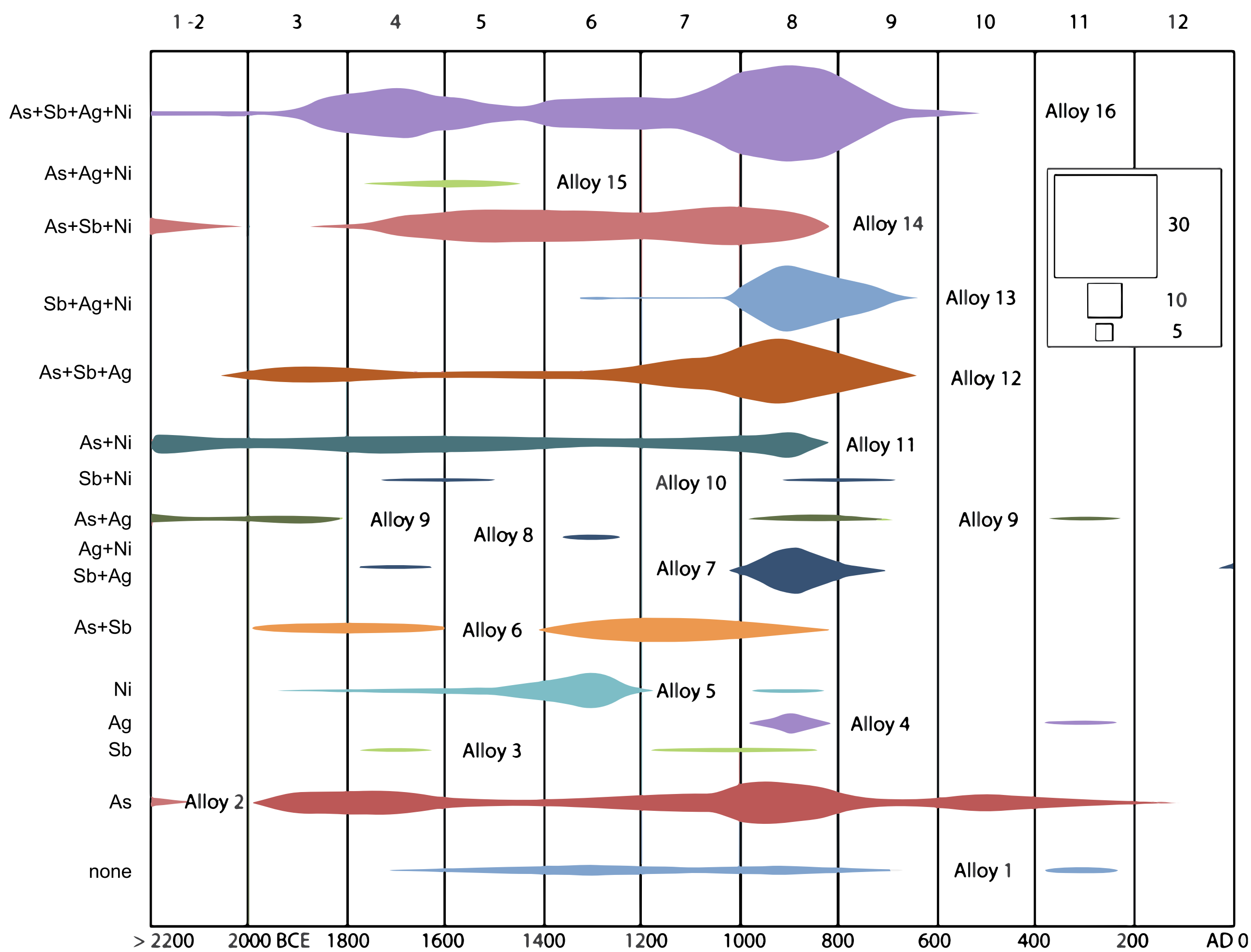


Fig. 6. Alloy ubiquity as counts (inset show numeric values for widths of violin plots) across time (top X-axis: periods, lower X-axis: years BCE), y-axis shows alloy group characterization following Table 1.

the importance of alloy group 12 is taken over by alloy groups 11 and 14 (each n = 6, 14 %). The items made in alloy 2 are mostly low-flanged axes (in both Atlantic/British Isles styles and local Emmen types of axes; Butler, 1995/1996, 171–178; 184–190 table. 1; *cf.* Arnoldussen et al., 2020a), but also comprise two rivets of two Sögel-Wohlde swords (Mussel Aa and Garderen- Bergsham; Butler and Van der Waals, 1966, 76 Table X). The blade from the Garderen-Bergsham grave (Bourgeois and Fontijn, 2015, 46) is made of alloy 11, as was the Wohlde blade of Echten (Van der Sanden and Arnoldussen, 2017). The Sögel blade of Drouwen was made in alloy 14 (Butler, 1990, 73 tab. 3), as were rivets of the Echten blade (Van der Sanden and Arnoldussen, 2017, 9). The items in alloy 16 are all flanged axes of local and Atlantic/Insular affinity (both Atlantic/British Isles styles and local Emmen types of axes; Butler, 1995/1996, 171–178; 184–190 table. 1; *cf.* Arnoldussen et al., 2020a).

Around the transition of the Middle Bronze Age-A to Middle Bronze Age-B (period 5; 1800–1600 BCE), alloy group 14 (As/Sb/Ni) forms the

Fig. 7. The Wageningen hoard (comprising amongst others a stone axe, flat axe, halberd with rivets, dagger blade, (neck)rings and bronze scrap ©Photo: RMO).

largest group (40 %), with alloys 16 (16.7 %) and 11 (13.3 %) second and third, all hinting at the importance of Ni ores/alloys in this period. In alloy 14, as in period 5, dagger and sword blades (Sögel-Wohlde and Grooved ogival daggers, two each) are represented, as are two flanged axes and two palstaves. It should be noted that in alloy 14 various evident Atlantic imports can be recognized. These comprise a Tréboul dagger from Berlicum (Ruijters, 2020, 136; Arnoldussen et al., 2021, 120 fig. 3, whose alloy is similar to Great Orme ores used for objects of the eponymous Tréboul hoard (*cf.* Williams and Le Carlier de Veslud, 2019, 1184 fig. 4; 1185)), a British basal-looped spearhead from Exloërmond (Butler, 1987, 18 fig 9.2; 32), as well as the famous aggrandized dirk of Jutphaas (Fig. 8; Butler and Sarfatij, 1970/71; Fontijn, 2001) that belongs to a group of masterfully crafted Plougrescant-Ommerschans dirks (Postma et al., 2017, 48–50) scattered across Atlantic Europe.

The Middle Bronze Age-B corpus (periods 6–7; 1400–1000 BCE) shows mostly alloys of groups 14 (As/Sb/Ni; 22.1 %) and 16 (As/Sb/Ag/Ni; 20 %), supplemented with alloys 12 and 5 (each 11.6 %). Strong differences between period 6 and 7 exist only for alloy 5 (Ni-only, which occurs exclusively in period 6 (comprising just the Voorhout hoard axes of Atlantic origins; Butler, 1990, 78–84; Fontijn, 2008; Williams, 2018, 44)) and alloy 12 (As/Sb/Ag, which increases from 2 to 9 objects between periods 6 and 7). Amongst the reliably dated finds in alloy group 14, palstaves, spearheads and *griffplatten*-swords are dominant. In alloy 16, comprising mostly palstaves and spearheads, new object types come to the fore such as bracelets (Arnoldussen and Steegstra, 2021, 47) and arrowheads (Van der Veen and Lanting, 1991, 216). Alloy 12 was used mainly for bracelets, spearheads and palstaves, but a single Atlantic Type Rosnöen sword in alloy group 12 should be noted (Essink and Hielkema, 2000, 309 no. 252).

The Late Bronze (1000–800 BCE; period 8) is one of the most diverse in alloy groups utilized, with no less than 14 used concurrently. Among these, alloy group 16 (23.1 %) is most numerous, followed by alloys 12 and 13 (each 14.9 %) and alloys 7 and 2 (each 10.7 %). Whereas alloy groups 12 and 16 were common in preceding periods, alloys 7 (Ag/Sb) and 13 (Ag/Sb/Ni) are only common in period 8, as is alloy 4 (Ag only), accounting for 4.9 % of the finds). Alloy 4 consists exclusively of a group of high-lead socketed axes in the Valkhof Museum, presumably of southern Dutch/Belgian affinity. Most of the alloy group 13 objects (often high in Sb/Ag) are ornaments from the Urnfield cemetery at Leesten (Van Straten and Fermin, 2012, 63–68). Alloy group 2 (As-only) comprises exclusively ornaments, such as torqued neckrings (*Wendelringe*; *cf.* Eimermann and Van den Broeke, 2016, 39), *Hohlwulstringe* and D-shaped bracelets (*cf.* Arnoldussen and Steegstra, 2021, 47–49 table 1).

For the Early Iron Age (period 9; 800–600 BCE), substantially fewer data are known than for the preceding period (19 records vs 129 respectively), but – as in period 8 – alloy groups 16 (36.8 %) and 13 (26.3 %) are dominant. In alloy 13, Urnfield ornaments such as disc-head pins and burnt droplets are found. For the later Iron Ages (Middle to Late: 600 BCE-AD 0; periods 10–12), again a smaller data-set (n =

Fig. 8. The aggrandized dirk of type Plougrescant-Ommerschans found at Jutphaas (length 42.5 cm. © Photo: RMO).

15) of items is available. Amongst these, alloy groups 2 (As only; 60 %) and 1 (13.3 %) are dominant. The items of alloy group 2 are all ornaments, the two items in alloy group 1 concern a Late Iron Age belt hook and bracelet from Kessel-Lith (*cf.* Roymans, 2004, 113–118).

3.4. Diachronic shifts in high-concentration alloys

Whereas the alloy groups proposed by Bray (et al., 2015, 205 fig. 1) proved insightful in charting alloy preferences over time and alloy-object correlations (*supra*), we argue that this classification does not capitalize on information on alloy constituents in the > 1 % ranges. For example, high values for volatile elements such as arsenic and antimony, could indicate the import of new (or low recycling/mixing frequency) bronze (scrap or ingots). These 'fresh' imports of course supplement the extant bronze corpus, so we can only look at this from a relative perspective: what is the percentage of high (>1 %wt, > 2 %wt) As and Sb alloys, and how does it change over time? As shown in Fig. 9, there is strong correlation for the patterns at > 1 %wt and > 2 %wt.

For the periods up to 2000 BCE (periods 1 and 2) high-arsenic copper (81.2 % > 1 %wt, 56,2% > 2 %wt) is the dominant type. In period 3, the percentage of high-arsenic alloys drops notably (35,7 % > 1 %wt, 17,8 % > 2 %wt), with just a few halberds, flat axes and (grooved) ogival daggers making up the high-arsenic group (Alloys 2 As, 11 As and 16 As). In period 4, high-arsenic copper plays just a marginal role. Only 13 % of the measurements are > 1 %wt and none are > 2 %wt. The objects with high (>1 %wt) antimony all have limited arsenic (0.1–0.3 %wt), moderate to high Ag and Ni (0.35–4.8 %wt) and are typologically classified as 'Emmen axes' (Fig. 10) crafted from Singen metal (Butler and Van der Waals, 1966, 86; Vandkilde, 2005, 25). In period 5 the percentage of high-arsenic alloys rises somewhat again (to 30 % for > 1 %wt, and 10 % > 2 %wt), but no correlations between object types and alloys/compositions are discernible. Period 6 and 7 form a continuation of the pattern observed for period 5: alloys with impurities > 1 %wt remain uncommon (<30 %) and those with impurities > 2 %wt even more so (<10 %; Fig. 9).

It is clear that in period 8, the frequency of high-antimony alloys rises

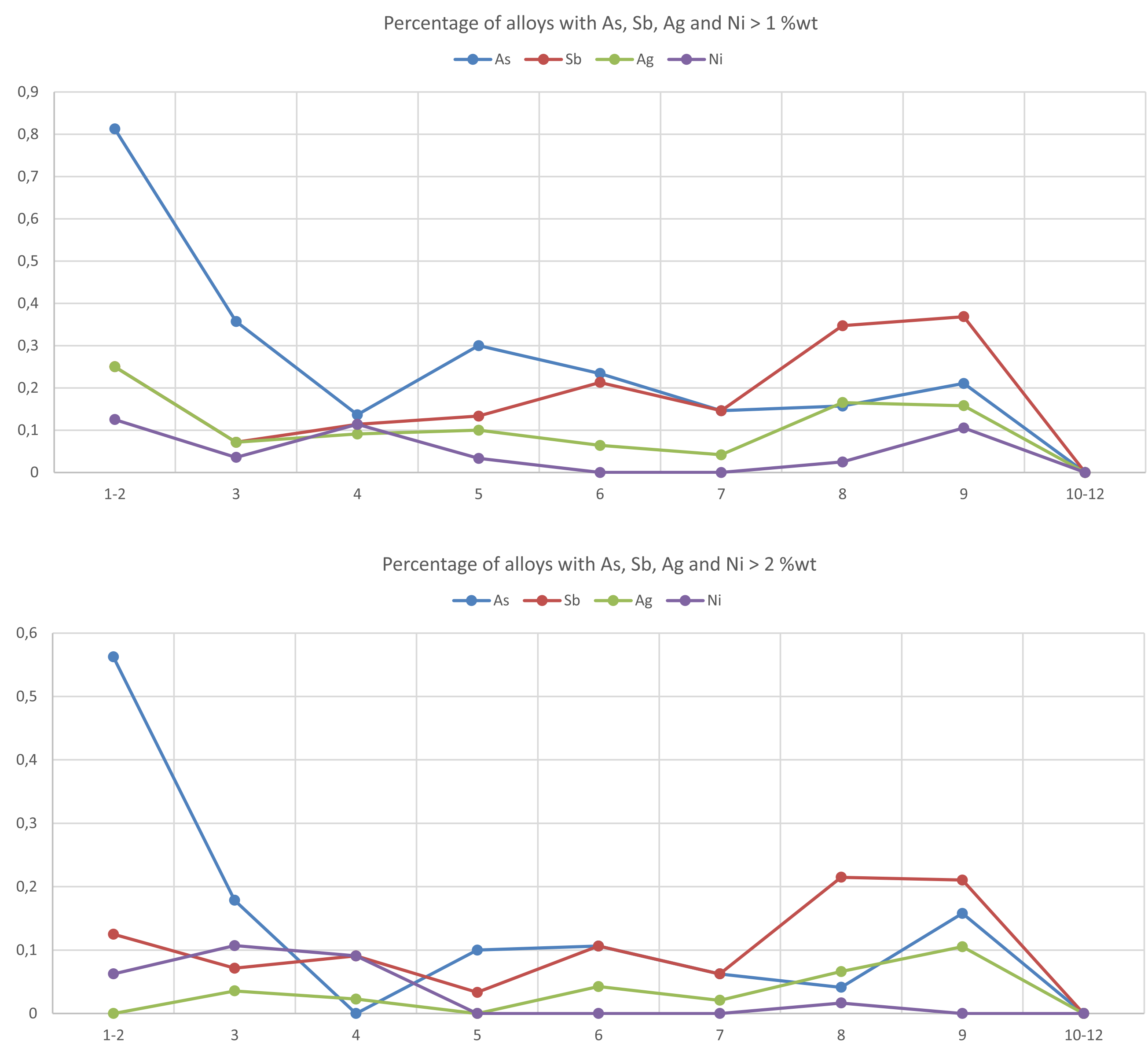


Fig. 9. Frequency of alloy compositions for elements over 1 %wt (top) and 2 %wt (bottom), for the chronological periods 1 to 12.

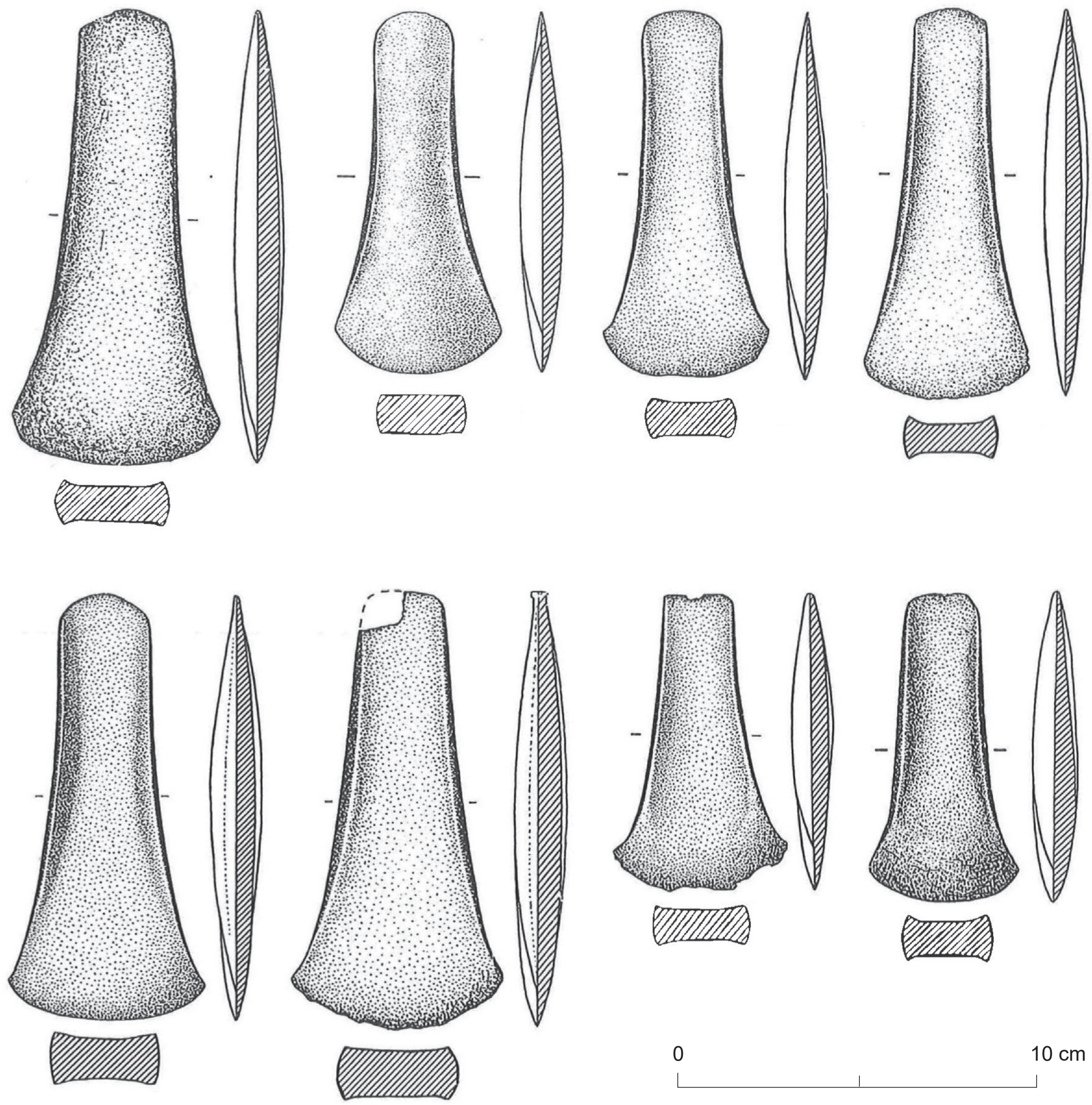


Fig. 10. Examples of Emmen-type low-flanged axes (from: Butler, 1995/1996, 184 fig. 14a).

steeply (>1 wt% at 34,7 % and > 2 %wt at 21.5 %). The fact that period 8 contains most data points (n = 121 observations) assures us that this is a real pattern and not due to overrepresentation of high-Sb alloys in a small sample. Not only is Sb present in more objects, it is also present more in the objects (26 observations with Sb between 3 and 9.6 %wt). Similar observations can be made for period 9, but here the representativity (n = 19 observations) is limited. All period 9 alloys with Sb > 1 %wt are Urnfield ornaments, sometimes (but not invariably) with Ag > 1 %wt and Ni 0.18–1.21 %wt – and always rich in lead (1.9–15.5 %wt). For periods 10 to 12, despite the few observations (n = 15), it could be telling that alloys with low (<1 %wt) concentrations of Ag, Sb, As and Ni were the norm – albeit that for ornaments and scabbards lead was added in high concentrations (2.1–20.6 %wt).

3.5. Diachronic shifts in leaded alloys

In the above section we have suggested that leaded alloys become more frequent towards the Iron Age. Here we discuss the general diachronic trends in leaded alloys for the periods under study. Fig. 11 shows the number of items assigned to each Pb class for all periods. It is evident that copper alloys with over 2 %wt in Pb were rare prior to *c.* 1400 BCE. After that, their prominence gradually rises: not only are more objects crafted from leaded alloys, the amount of lead added can also be much higher. This is a trend that culminates in the Late Bronze Age (per. 7; 1000–800 BCE). In what follows, we will look at these diachronic differences of leaded copper alloys in more detail.

For periods 1 up to 4 leaded alloys are rare in both relative and absolute terms. Minor (*i.e.* < 0.57 %wt) additions of lead to low-flanged axes, swords and daggers do occur, but this value is never higher. In periods 5 and 6 (1600–1200 BCE), items reliably dated that period and with lead at 1–5 %wt can be identified, such as several axes of the Voorhout hoard (Fig. 12; Butler, 1959, 131; Fontijn, 2008, 14–15) and some palstaves (*e.g.* Butler and Steegstra, 1997/1998, 229 cat 326). For the second half of the Middle Bronze Age (period 7; 1200–1000 BCE), an evident increase in the popularity of lead-alloying can be identified (average Pb class: 1.94; Fig. 11, bottom). Pb classes 2 and 3 consist only of seven spearheads, and in Pb class 4 bracelets dominate (*e.g.* Arnoldussen and Steegstra, 2021, 47; 54; 50; 65; 97).

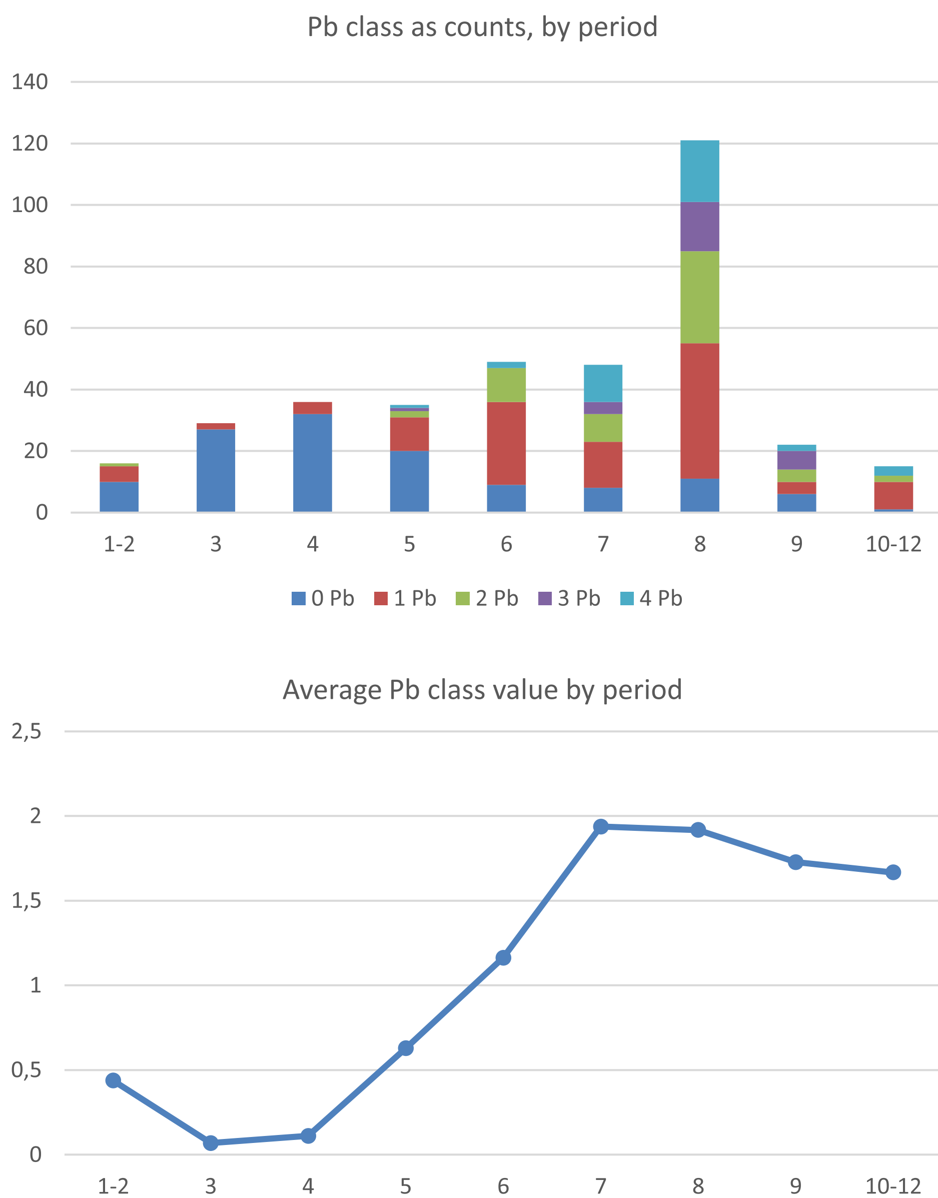


Fig. 11. Frequency of alloys with lead < 0.1 %wt (class 0), lead at 0.1–1 %wt (class 1), lead at 1–5 %wt (class 2), lead at 5–10 %wt (class 3) and lead > 10 %wt (class 4) for all periods. The top histogram shows the absolute numbers, and the lowermost trendline shows the average Pb class score for all periods.

Throughout the Late Bronze Age (period 8; 1000–800 BCE), the average Pb class value remains high (1.19; Fig. 11, bottom). The group of twenty Pb class 4 (*i.e.* Pb > 10 %wt) observations is dominated by socketed axes (75 %, often in alloy 4 or 7), of both local styles (Type Niedermaas; Butler and Steegstra, 2002/2003, 269–271) and evident imported ones (Type Seddin; *op.cit.*, 221–223, Type Plainseau; *op.cit.*, 279–281) alike. The Pb Class 3 objects (Pb 1–5 %wt) also comprise socketed axes (but now mainly of Type Geistingen (in alloys 14 As Sb Ni and 16 As Sb; Butler and Steegstra, 2002/2003, 303; Nienhuis et al., 2011, 47; 60), two socketed knives (in alloys 12 and 16, one of palafitte origin; Butler et al., 2012, 71), a pegged spearhead, a carp's tongue sword (alloy 16 and 16 Sb respectively) and a set of Urnfield ornament fragments (droplets, unknown fragments (alloys 7 and 11) and tinned ornaments (alloys 13 Sb/13 Sb Ag; Van Straten and Fermin, 2012, 63–68)). Pb class 2 contains 30 observations, with a few rarer object types (razor, ferrule, sickle), but no strong correlations of Pb class to alloys, nor of Pb class to object types that merit more detailed discussion.

For the Early Iron Age (period 9; 800–600 BCE), the average Pb class value appears to be still rather high (1.73; Fig. 11, bottom), but the small sample population (n = 19) may be a distorting factor. For Pb classes 4 and 3, seven of the eight observations pertain to Urnfield ornaments (pins, neckrings, droplets; of varied alloy types). The remaining high-lead (class 3) object concerns the Type Kurd situla in the chieftain's grave of Oss (van der Vaart-Verschoof, 2017, 26; 180–184; 194). Pb Class 2 items are again mostly Urnfield ornaments (but now exclusively in alloy 16) and a single socketed axe (also in alloy 16). The validity for inferences on leaded alloys in the later Iron Age (periods 10–12; 600 BCE-AD 0) is again limited due to the small sample size (n = 15), but two Pb class 4 and two Pb Class 2 items are listed. The former concern Middle Iron Age *Segelohrringe* with blue glass ornaments (in alloy 2; Kooi, 1983,

Fig. 12. Acton-park(like) palstaves and other axes from the Voorhout hoard (see Fontijn, 2008; 2015 for the origins of these axes). Thirteen axes contain over 1.5 % wt lead. © Photo: RMO, drawings: Groningen Institute of Archaeology/RUG (all to same scale).

Table 3
Distribution of artefact classes over Pb classes (<0.1 %wt (class 0), lead at 0.1–1 %wt (class 1), lead at 1–5 %wt (class 2), lead at 5–10 %wt (class 3) and lead > 10 %wt (class 4)).

Pb class	4	3	2	1	0
Axes	18 (33.9 %)	6 (18.7 %)	19 (29.2 %)	36 (26.3 %)	53 (43.4 %)
(of which socketed)	*15 (28.3 %)*	*5 (15.6 %)*	*6 (9.2 %)*	*6 (4.4 %)*	*4 (3.3 %)*
Ornaments	16 (30.1 %)	13 (40.6 %)	18 (27.7 %)	38 (27.7 %)	14 (11.5 %)
Swords/daggers	2 (3.77 %	1 (3.1 %)	6 (9.2 %)	18 (13.1 %)	42 (34.4 %)
Tools	3 (3.77 %	2 (6.2 %)	4 (6.1 %)	12 (8.8 %)	6 (4.9 %)
Spearheads		1 (3.1 %)	11 (16.9 %)	16 (11.7 %)	2 (1.6 %)
Other		1 (3.1 %)	1 (1.5 %)	11 (8.03 %)	1 (0.8 %)
Total	53	32	65	137	122

207), whereas the latter are an Early/Middle Iron Age *Hohlwulst* rivet (Arnoldussen and Steegstra, 2021, 84) and a Late Iron Age scabbard (*cf.* Roymans, 2004, 108–112).

If we for a moment ignore the diachronic trajectory and look at the correlations between object type and use of leaded alloys (or not) for specific artefact groups (Table 3), it is clear that heavily leaded alloys (Pb classes 3 and 4) were mostly used for casting ornaments. The conflation of axes of various types however masks an evident relationship: LBA and EIA socketed axes (reliant on *cire perdue* casting) dominate the Pb 4 and 3 classes (and comprise roughly a third of the Pb Class 2), whereas axes from other periods were mostly cast in alloys with < 1 % lead (Table 3).

4. Discussion

The above review of diachronic changes has highlighted correlations between alloys and periods as well as object groups. In the era leading up to the Bronze Age proper (*i.e.* pre 2000 BCE), arsenic alloys with (variable, but generally present) nickel (dubbed 'Dutch Bell Beaker Copper') are used for Bell Beaker metalwork (Butler and Van der Waals, 1966, 92 fig. 29; Vandkilde, 1996, 177–178; Wentink, 2020, 175). The ultimate origin of its ores could be Iberian (Butler and Van der Waals, 1966, 97) or Brittany (Vandkilde, 2005, 25). Halberd blades were crafted from arsenical copper (alloy 2 exclusively), albeit that in their rivets high impurity (high Sb, High Ag, high Ni) alloys ('Singen metal'; Butler and Van der Waals, 1966, 90 fig. 26; Vandkilde, 2005, 25) were used. In the Wageningen hoard (Fig. 7) both arsenic alloys and tin bronzes were combined (Butler and Van der Waals, 1966, 83–83; Fontijn, 2003, 73), marking the introduction of true tin-bronzes around 2000 BCE (for a discussion of this hoard's peculiar content and composition see: Visser, 2021; 85; 99–100). Based on recently proposed date-ranges for halberds in the final 3rd millennium BCE across Northwest Europe (*e.g.* Needham et al., 2015, 21 fig. 26; Horn and Schenck, 2016, 17), this transition is perhaps best placed one or two centuries prior to 2000 BCE). Given that in periods 1–2 high-arsenic copper remained the dominant type, we can postulate that most bronze artefacts were cast from (high-arsenic) ores that underwent limited melting cycles (average 2.68 %wt, st.dev. 1.99). Amongst the items with over 3.5 %wt As, bell beaker copper daggers (Butler and Van der Waals, 1966, 76 tab. X; Wentink, 2020, 172) are common. Sometimes objects of Singen composition are found associated with these (*e.g.* Exloo; Van Giffen, 1947, 123; Wentink, 2020, 163 fig. 6.21), indicating that by the end of the Bell Beaker period the metal pools exchanged integrated both Atlantic (Iberian?) and southern German regions (Wentink, 2020, 177, *cf.* Needham, 2002, 121–123; Kienlin and Stöllner, 2009; Merkl, 2010, 26).

For a group such as the low-flanged axes, it is clear that their compositions reflect a widening of sources/origins (and by proxy, supraregional contacts). Remarkably, low-flanged axes typologically interpreted as Atlantic/Insular imports are generally characterized by low silver and low nickel, whereas the Emmen axes (Fig. 10; Butler, 1995/1996, 184–189) deemed local are generally high in silver (>0.7 %wt) and high in nickel (0.39–4.8 %wt, av. 1.59 %wt, st.dev. 1.4, albeit that three 'Emmen' axes have alloys identical to the Atlantic/Insular ones; Butler and Van der Waals, 1966, 76 tab. X). Possibly, they represent re-melts/re-casts of Atlantic-British objects, scrap or ingots. Amongst this group of low-flanged axes cast into the local 'Emmen' shape, items in arsenical copper (alloy 2), Singen-copper (alloy 16 Sb Ni (Ag)) and arsenic-silver copper (alloy 9) are found. Contemporary typological imports (so-called low-flanged axes of Irish affinity) too are mostly made from alloy 2, once from 12 and once from 16 Ag, suggesting that both insular (British/Irish; Butler and Van der Waals, 1966, 82–84; Fontijn, 2009, 134; *cf.* Arnoldussen et al., 2020a, 45) as well as objects of Swiss/Southern German ores (Butler and Van der Waals, 1966, 84; Butler, 1995/1996, 190) were remelted into local types.

For the Sögel-Wohlde swords blades of Nordic affinity (Vandkilde, 1996, 156; Fontijn, 2003, 101; 345–347), alloy groups 11 and 14 were used exclusively (*i.e.* As-Ni alloys, with more (>0.1 %wt; alloy 14) or less (<0.1 %; alloy 11) antimony. This tallies well with the preference of As-Ni alloys for swords of the Sögel-Wohlde group as published by Ling (et al., 2019, tab. 4), who moreover showed that lead isotopes suggested eastern/southern Alpine ores (OEM863/965: Ni > As), Slovakian ores (MA-071222; As > Ni), Mitterberg ores (MA-071243: As > Ni) as well as Southern Iberian ores (FG 050575: As > Ni) for these swords. The group with antimony over 0,1% (alloy 14), has parallels in other MBA-A swords types such as Hajdusámson-Apa derivates and Valsømagle swords (*e.g.* ALM26/UM 40280_3006/B5469a: Ling et al., 2019 tab. 4), whose isotopic signals suggest Slovakian and Eastern Italian Alpine ores. Clearly, while the object styles (and their use in funerary assemblages) reflects an incorporation of the northern part of the Netherlands into a Nordic cultural realm (*cf.* Butler, 1986; Fontijn, 2003, 228; 345–347; Arnoldussen, 2015, 20–25; Arnoldussen and Steegstra, 2018, 37), to obtain the ores required to craft such blades central European and Italian Alpine contact networks were in place. Simultaneously, evident imports to the Netherlands from the Atlantic zone such as Tréboul and basal-looped spearheads were mostly crafted in alloy groups 11 and 14, and can perhaps be linked to Great Orme exploitation (*cf.* Williams and Le Carlier de Veslud, 2019, 1184 fig. 4; 1185).

During the MBA-A (*c.* 1800–1500 BCE), most objects for this period are low-impurity (<1 %wt) alloys, resulting from exchange networks that have insular, central European and Italian Alpine oxidic ore sources all at play simultaneously (*supra*). This suggests that in this period, 'fresh batches' of alloys with low recycle/mixing frequencies reached our areas less than in preceding periods, and that this is reflected by the homogenisation of alloys in period 4. For the MBA-B (*c.* 1500–1000 BCE) hoard assemblages such as the Voorhout hoard (Butler, 1959, 131; Fontijn, 2008, 14–15), the Drouwenerveld hoard (Butler, 1986, 135–137) and the Hoogeloon hoard (Fontijn and Roymans, 2019, 168–170) suggest that imported non-local bronze items (scrapped (Drouwenerveld) ornaments, or used/broken (Hoogeloon) or miscast/as-cast axes (Hoogeloon) rather than ingots may have been exchanged in order to facilitate local production. For the Voorhout hoard, at least three alloy groups are represented in that assemblage (Butler, 1995/1996, 194, maybe more if lead is taken into account) – hinting at significant heterogeneity in the imported alloys that were to be recycled

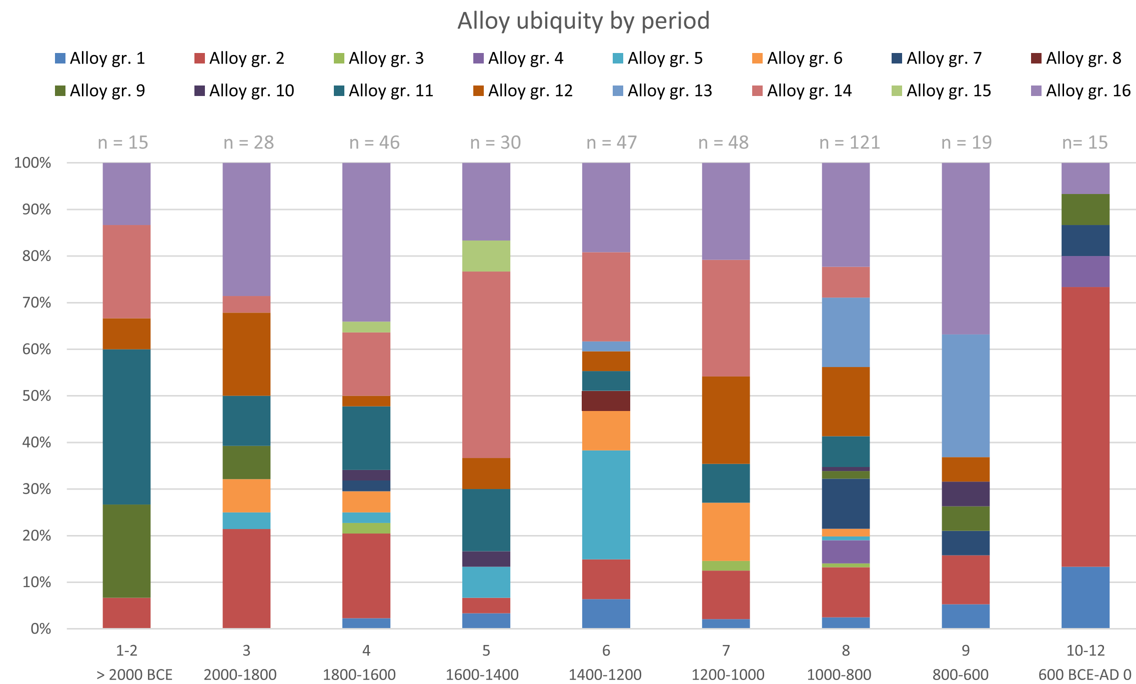


Fig. 13. Alloy ubiquity (for alloy types see Table 1) by chronological period (200 year bins, 1 = pre 2200 BCE, 12 = 200 BCE – AD 0).

into local forms. The two 'zero impurities', non-local, as-cast, and flawed axes of Angelslo-Emmerhout (Arnoldussen et al., 2020b, 51–52) could be examples of 'freshly imported, unrecycled' alloys (tentatively from Mitterberg ores; *op.cit.*, 52, *cf.* Pernicka and Lutz, 2016, 29; 30 fig. 8) that had yet to undergo their first melting. Moreover, the Voorhout hoard is a clear case in point that alloys with Pb > 1 %wt were available from the start on the MBA-B (*i.e.* 1500–1100 BCE) onward. This is in line with other parts of Europe. In Wales, lead was added to Acton Park assemblage alloys around 1500–1300 BCE (Johannsen, 2016, 153).

This diversification of alloys used increases in the Middle Bronze Age and Late Bronze Age (periods 6–8; Fig. 13). In these periods localized concentrations of object types such as Vlagtwedde palstaves and Hunze-Ems socketed axes (Butler, 1961, 199; Butler and Steegstra, 1997/1998, 270) are prominent. Moreover, production debris (Butler, 1961, 286, fig. 4.4; Butler and Steegstra, 2003/2004, 269 fig. 91a) and mould fragments (Butler, 1961, 286, fig. 4.4; Fontijn et al., 2002, 67–69; Kuijpers, 2008, 145–146) suggest more frequent and more localized production (but see Kuijpers, 2008, 46; Fontijn et al., 2002, 70 on the fact that for the Havelte and Oss moulds, the types castable do not appear to be local).

The enduring low percentages of high-impurity alloys, suggest that up to 1000 BCE, recycling/mixing was the norm and that only incidentally, objects of low recycling/mixing frequency alloys (*e.g.* high As, Sb) were added to the mix. For example, the period 8 group of items with Sb > 2 %wt comprises a restricted series of objects: Geistingen axes (Wielockx, 1986; Butler and Steegstra, 2002/2003, 303; Nienhuis et al., 2011, esp. 60), Urnfield ornaments (tinned studs/buttons and bracelets; Van Straten and Fermin, 2012, 63–68; Arnoldussen, and Steegstra, 2021, 61), two Late Bronze Age swords, a tanged knife from the Bargeroosterveld 1899 hoard (Butler et al., 2012, 81) and the Havelte socketed axe mould half (Butler and Steegstra, 2005/2006, 209–210). In contrast to their low numbers, the compositions and supra-regional affinities of the Voorhout, Hoogeloon, Drouwenerveld and Angelslo-Emmerhout hoards (*supra*) have been discussed as tangible illustrations of such freshly imported stocks (albeit that of these only the latter contained 'low recycle frequency' items). Evidently, by the Middle Bronze Age raw materials obtained by local communities were as a rule caches of (scrapped items and) axes that had seen prior smelting and mixing/recyling.

From the Late Bronze Age onward, leaded alloys gain in prominence. Even in Pb classes 2 and 1, ornaments figure prominently, albeit that axes are as frequently cast in these Pb classes. For bladed weapons, such as daggers and swords of all periods, alloys low in lead (<1%; Pb classes 1 and 0) were clearly favoured. Pb Classes 3 and 4 are dominated by ornaments (often in alloy group 13) and socketed axes. In that context, the addition of lead may have helped casting in finely worked moulds (*i. e.* cast-on details) and *cire perdue* types of casting required for socketed axes (*e.g.* Montero et al., 2003; Charalambous and Webb, 2020). The increase in popularity of leaded bronze alloys towards the Late Bronze Age is in line with developments elsewhere in Europe. For example, in Scandinavia (Johanssen 2016, 158 fig. 3) leaded alloys also increase onwards from period IV (*i.e.* 1125–925 BCE; Arnoldussen and Steegstra, 2018, 47 fig. 27). Certain axe types (Type Plainseau (*e.g.* Blanchet, 1984, 368–373; Van Impe, 1994, 16 fig. 1) and Type Seddin (Sprockhoff, 1956a, 92–39; 1956b, 22–23; Karte 9; Kibbert, 1984, 151–153; Johannsen, 2016, 158 fig. 3)) recovered from the Netherlands were cast in alloys of very high lead content. This underlines that (a) in both the Atlantic (Type Plainseau) and Nordic/North German (Type Seddin) networks high-leaded alloys were current, and (b) that Dutch communities were evidently integrated into these networks (note that based on socketed knives and bracelets, they were connected to palafitte networks too; Butler et al., 2012, 32 fig. 13C; Arnoldussen and Steegstra, 2021, 103–104). The preference for using leaded alloys persist into the Iron Age, and is more widely noted: In other parts of Northwest Europe, leaded bronze alloys were in use for Early Iron Age fibulae (*e.g.* Schwab, 2011, 271; 2014, 177), Early Iron Age ring ornaments (*e.g.* Tremblay-Cormier and Mille, 2018, 177; 179) and other Early Iron Age grave furnishings (*e.g.* Giumlia-Mair et al., 2003, 161).

The Late Bronze Age trend of alloy specificity (*e.g.* leaded alloys for ornaments) and alloy diversification continues into the Early and Middle Iron Age, albeit that again fewer correlations between alloys and objects can be identified. The most obvious remaining correlation is that between alloy group 13 and its use for ornaments (comprising beads, buttons/studs, earrings and pins), but one should keep in mind that this group consists mostly of high-lead, tinned beads whose copper values are low (tinning or tin-sweat, leading to an unfair (over)representation of antimony and silver (Van Straten and Fermin, 2012, 63–68). The usage of arsenic-only bronze (alloy group 2) for ornaments such as neck- and armrings is another notable pattern, albeit that some of the bronze situlae in Early Iron Age 'chieftain graves' such as those of Oss, Ede and Baarlo (*cf.* Van der Vaart-Verschoof, 2017) were also crafted from alloy 2 (in both low- and high-lead varieties).

5. Conclusions

The single-most important conclusion should be that the approach applied here indeed has yielded information on the relations between alloy groups, object types and typological periodization. Evidently, the limitations potentially posed by (a) crude typochronological dating, (b) mainly p-XRF compositional analyses, (c) predefined alloy groups (Table 1) and (d) modest data volumes (n = 375 observations) have not frustrated our approach but proved workable parameters to discuss diachronic patterns in alloy-object interrelations.

In the above, we have aimed to not only look at relationships between object types and particular alloy groups, although several correlations have been noted (*e.g.* alloy 14 for Sögel-Wohlde blades, alloy 5 for the Insular imports of the Voorhout hoard, alloy 4 for a group of socketed axes of southern affinity and alloy 13 for usage in LBA-EIA ornaments). In addition to these alloy-object correlations, we could use the prevalence of alloys high in volatile elements (*e.g.* As, Sb) as a proxy for 'new (*i.e.* low recycle/mixing) alloy influx'. Using this approach, we could show that particularly in periods 3, 5 and 8–9 (*i.e.* the Early Bronze Age, the MBA-A/B transition and the Late Bronze Age-Early Iron Age) objects and/or scrap that had not been mixed and recycled often were imported into the Netherlands. For each period, this aligns well with the cultural interpretations of (stylistic affinity of) several of the objects recovered.

For periods 1–3, halberds and several flat axes could be identified as imports from the British Isles. The alloy composition (Singen copper) of several rivets and low-flanged axes illustrates contacts with communities in possession of alloys melted from Alpine/Southern German ores. These connections may very well be embodied in the ('*mappa mundi*'; Fontijn, 2019, 37) hoards of this period, that show integration of local communities into networks spanning the Únětice region, the British Isles, and southern Germany (Visser, 2021, 104, *cf.* Arnoldussen, 2015; Fontijn, 2019, 36; Berger et al., 2021).

Period 5 (1600–1400 BCE) shows again that European connections of local communities comprised both Atlantic and Nordic exchange networks. From the former, Type Tréboul spearheads of presumably Great Orme metal and basal-looped spearheads were obtained that often ended-up as wetland depositions. Also, this is the period in which the enigmatic aggrandized Plougrescant-Ommerschans dirks (Fig. 8) circulated in the Atlantic network (West, 2015). As to the latter (Nordic) interaction sphere, in period 5 we find several examples of Sögel-Wohlde influences with a clear preference for alloys 11 and 14 (As-Ni alloys) for weapon blades (*supra*). Lead isotopes analysed for Scandinavian Sögel-Wohlde blades suggest that central European and Italian Alpine base ores were used to craft such blades (Ling et al., 2019, tab. 4), stressing the scale and intensity of contacts in this period (*cf.* Vandkilde, 1996, 243–246; Nørgaard et al., 2019, 24–25; Visser, 2021, 138; 164–167).

In periods 8 and 9 (1000–600 BCE), the frequency of high-Sb alloys was higher than ever (*cf.* Ling et al., 2013, 299 fig. 8), suggesting that imports of Fahlore type ores (of low-recycle/mixing frequency) was common. Ornaments and to a lesser extent socketed axes of Geistingen-type, but also swords and knife blades were cast in high-Sb alloys. Also, these are the periods in which leaded alloys become common. Simultaneously, these periods featured the widest variety of alloy compositions used and provided the most tangible evidence for local production (*e.g.* moulds, casting jets) suggesting that local production, surpa-local procurement of alloys/scrap and recycling/mixing were at their most intense (*cf.* Bradley, 1990, 79–98; Needham, 2002, 280; Fontijn, 2003, 214 fig. 10.1; 2019, 95; 98–102; 163; Timberlake, 2016, 724; Wiseman, 2017; Williams and Le Carlier de Veslud, 2019, 1193 fig. 10).

In conclusion, we have shown that geographical orientations of exchange networks proved not just identifiable with traditional typological means, but could be corroborated by both compositional analyses and object-alloy correspondances. This in turn, suggests that using an alloy characterisation approach is a successful tool to chart diachronic shifts in prehistoric alloy compositions (and in turn, provides insights into contact networks, frequency of imports and recycle frequencies). That fact that in this study mainly handheld p-XRF measurements of corrosion/patina were operationalised, testifies to the usability of such data (yet caution remains in terms of representativity, and critical evaluation of results/elements studied/included is key). This study thus highlights the potential of mixed heritage datasets of copper alloy compositions, even when different analytical methods were applied to corrosion layers as well as on core metal samples. Moreover, it underlines the value of the corrosion layers on the objects and its potential for non-destructive analyses that yield information on prehistoric socio-economic trends and changes. Using predominantly p-XRF analyses of museum objects, we could show that the data-variability observed pertained to four overlapping vectors: (1) typological provenance (*e.g.* Atlantic, Nordic, Central European) or stylistic origin, (2) Ore origins (*e.g.* Welsh versus Alpine ores), (3) fingerprinting alloys by phase (*e.g.* Neolithic Dutch Bell-beaker copper versus Later Bronze Age highly-leaded alloys), (4) fingerprinting alloys by object types (*e.g.* axes, ornaments, bladed weapons). That being said, the broad brush (lumping, rather than splitting of individual analyses of composition) approach applied here to object composition, cannot – and should not – replace approaches that target the (remaining) problems such as (a) the accuracy of alloy estimation due to corrosion effects, (b) core vs outer surface alloy estimations, (c) isotope provenancing and (d) individual object study.

CRediT authorship contribution statement

S. Arnoldussen: Conceptualization, Investigation, Formal analysis, Writing – original draft, Writing – review & editing, Visualization, Supervision. **D.J. Huisman:** Methodology, Validation, Formal analysis, Investigation, Writing – review & editing. **B. van Os:** Methodology, Validation, Formal analysis, Investigation, Data curation, Resources, Writing – review & editing. **B. Steffens:** Investigation, Resources, Writing – review & editing. **L. Theunissen:** Investigation, Resources, Writing – review & editing. **L. Amkreutz:** Investigation, Resources, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The full dataset is provided as Electronic Supplement 1

Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jasrep.2022.103684.

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