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Form and flow: the 'karmic cycle' of copper

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ABSTRACT

The analysis and interpretation of the chemical composition of copper-alloys is one of the longest ongoing research projects within archaeological science. Beginning in the late 18th century these data have been consistently used to try and link objects with distinct metal sources. This paper argues the traditional provenance model for copper alloys is fatally flawed. Through pursuing a 'pure' source signal, chemical and isotopic datasets have been removed from their context and history. Social engagement with metal through processes such as reuse, recycling, and curation were rarely considered important by analysts. We offer an alternative model that unites the available legacy scientific datasets with process-metallurgy, archaeological and geographical context, and new conceptual approaches. Rather than provenance, we offer an empirical model of metal flow. Here objects are seen as snapshots of a wider metal stream; their final scientific characterisation including echoes of their previous forms and contexts. Through a series of case studies we highlight how the reinterpretation of existing datasets can disentangle the complex life histories of units of copper.

A document dated to the 12th year in the reign of Edward I (1284), translated from the original Latin by Riley (1875: 77), records the casting of a new bell at Bridgewater (Bruggewauter) in Somerset, England. It provides a detailed account of donations received from the parish and the expenses incurred by Richard Maydous, Philip Crese Erl, Gilbert le Large and Richard de Dunsterre. The document also lists, on the reverse, the sources of the metal used in the production of the bell:

> '*Metal for the bell. They answer for 180 pounds of brass, received as gifts, as in pots, platters, basons, lavers, kettles (cacabis), brass mortars, and mill-pots (pottis molendini). Also, for 425 pounds received from one old bell. Also, for 40 pounds of brass, received by purchase. Also, for 896 pounds of copper (cupri), received by purchase. Also, for 320 pounds of tin, received by purchase. Sum 1861 pounds. Of which there has been melted in the making of a new bell, 1781 pounds; and there are 80 pounds remaining over.*'

This list highlights an important problem with many traditional approaches to archaeometallurgy, which have assumed a simple linear relationship between the composition of ore sources and archaeological objects, since at least 605 lbs of the 1861 lbs of metal collected for the bell (*c.* 33% by weight) are being recycled. This figure may possibly be much higher if the 'purchased' copper, brass and even tin were also obtained from scrap, rather than freshly smelted metal. Clearly, if we were to carry out a 'conventional' provenance study of metal from the bell – using trace element composition and/or lead isotope ratios – we would be in danger of misinterpreting the results. To make this single object, copper from at least four different sources (the scrap brass, the old bell, the purchased brass and the purchased copper) is combined. Each has a potentially distinctive trace element 'fingerprint.' The scrap brass could be very variable indeed, depending on the life histories of the individual objects used, and even the old bell could itself contain recycled material from a previous iteration. Likewise, each of these sources of copper might bring in lead with different isotopic ratios, resulting in a mixed signal, which would correspond to no real source.

Although the Bridgewater bell provides an unusually clear illustration of this problem, the example is far from unique. Even defenders of a traditional model of evaluating provenance have to concede that particular assemblages are undeniably the result of collecting and remelting old metal artefacts. Pernicka (2014) for example discusses Late Bronze Age bun-shaped ingots from Switzerland, within which are partially melted pieces of identifiable objects (Rychner and Kläntschi, 1995). Similar and overwhelming evidence from both historic and prehistoric contexts has

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led us to fundamentally re-evaluate our approach to the interpretation of chemical and isotopic data from archaeological copper alloy objects. We would argue that, in focussing exclusively on the search for static geological origins or 'provenance,' conventional object-based perspectives have ignored the complex effects of human action on the chemical and isotopic composition of metal. Accepting that human interactions with metal (reworking, recasting, mixing and re-alloying) may weaken and ultimately destroy the possibility of provenance in a traditional sense does not, however, diminish the value of archaeometallurgical chemical and isotopic research. We need to move away from hoping that recycling was often unimportant (Pernicka, 2014: 258), an argument that is often framed through inappropriate analogies with the modern metallurgy industry. If we approach such studies of ancient metal from a broader perspective, taking a life-history approach to both objects and the metal from which they are made, then the dynamic composition of copper can provide us with the key to understanding structure in the data. In short it is possible to *empirically* assess the level and nature of metal recycling in the past, along with other types of social and practical interaction. Here we offer a framework which allows us to explore how people used and related to metal as a material in the past. For the first time in more than 60 years, we can begin to re-write the history of human engagement with this remarkable material. Perhaps invoking a 'karmic cycle' for copper suggests a more spiritual model than we intend, but we do believe that copper was frequently recycled, and that in some cases the previous lives of an object may have an influence on the form that such reincarnation might take.

1. A new paradigm: 'form and flow'

In order to visualise this model, and to emphasise how it differs from previous ideas that consider objects in isolation, we liken the flow of metal through society to that of a river fed by a number of springs, which runs out into a desert where it disappears. The extraction of copper from its ores, like a spring, creates a pool of material, which flows out as artefacts through a shifting social landscape, like a series of streams which may ultimately merge to form a river. Near to a source, individual communities may rely exclusively on a single 'stream' of copper, although they may alter it in a variety of ways. A new stock of metal entering the flow – the metal in circulation – will change the composition of the flow, just as the tributaries of a river contribute water with different chemical characteristics and sediment load. The composition of the metal in circulation at any one time and place is dependent on the balance of inputs from these 'tributary' streams. The relationship between an individual copper alloy object (with a specific 'form') and the generalized metal in circulation at a given time and place (the 'flow') can, therefore, be compared to the relationship between a bucket of water and the river. Water can be temporarily taken out of the river and kept in a bucket. While it remains in that container it will retain the properties of the river water at the time it was removed from the flow of the river. If the water in the bucket is returned to the river, it will alter to some small degree the properties of the water currently flowing in the river. In this analogy, a quantity of metal is 'scooped out' from the flow of the river of metal, made into an object of fixed form with a composition systematically related to that in the river (analogous to the bucket of water) and is then either lost, buried (to be archaeologically recovered) or returned to the river of copper to become available for future use as a raw material. We are left only with what has been taken out of the river. The concept of metal flow in archaeology has been emphasized by many scholars (Bradley, 1988; Needham, 1998; Jin, 2008; Pollard, 2009), but it is only now that we have developed a quantitative methodology to disentangle this complex dynamic system. The 'flow' of copper at any particular time and place is in reality made up of all the available copper objects at that time and place, and its precise composition is, of course, generally unknown to the users and will change with time as metal is added to or removed from the flow. Though specialized ingot forms may exist, we would highlight the potential for all copper objects to be melted and returned to the broader flow of metal. For societies which do not exchange metal, the flows of metal will be independent, like two parallel river systems, but exchange of metal will create linkages between them. The only evidence we now have for the composition of the flow of copper is the chemistry of the surviving objects made from it. In order to reconstruct this flow, we therefore need to think about the life history of the copper from which these objects are made. Individual objects and their assemblages crystallize out snapshots of the ongoing, overarching course of copper.

From this perspective, individual copper objects have three intrinsic 'attributes,' which are interrelated, but not necessarily dependent on one another:

- **trace element composition**, derived primarily from the copper ore source(s), but altered by human manipulation of the metal,
- **alloy composition**, defined by intentional action, as craftspeople choose to add minerals and metals to modify the characteristics of their material (fluidity in casting, colour, hardness, etc.),
- **form** (described by typology), imposed by humans and reflecting the socio-technological context of production.

There is a fourth (extrinsic) property, 'context', which frames the life history of the object, allowing us to situate its intrinsic attributes within the wider physical and social world. We argue that none of these intrinsic attributes are fixed in time, and are contingent not only on the life history of the object, but also on the life history of the copper metal prior to its incorporation into the object. Because we see individual objects being made by extracting metal from the stock in circulation, and, if not lost or deposited, possibly being re-made into new objects by being combined with metal extracted from the 'river', the biography of the metal flow transcends any individual object biography. Lead isotope composition, which might be considered a separate intrinsic characteristic, is in fact dependent on mixing between the lead isotopic signal(s) in the copper itself and the lead (if any) carried by the alloying elements, and is therefore encapsulated within the first two attributes.

Copper metal is extracted from an ore, refined, and turned into a block of relatively pure raw metal which becomes an input into the flow of metal. In theoretical terms we consider this a 'unit' of copper. It might be visualised as an ingot of copper, but this implies that all ingots are made from a single primary source, and this is not necessarily true. It also implies that 'finished' objects cannot act as ingots of metal, when the fact that they often do so has been demonstrated (Bray and Pollard, 2012). This unit of copper will then go through a series of transformations in shape, alloy composition and trace element composition (and also isotopic composition) as it flows through different societal contexts until it is ultimately lost or deposited into the archaeological record. The period of time between the 'birth' and 'death' of this unit of copper may vary from almost instantaneous (e.g., being made into an object intended to be placed in a tomb) to several centuries or possibly even millennia. The trajectory a unit of copper might follow is highly variable, and will depend on time and place (recycling and reuse may be much more common at certain times and places), and also the social context (some objects may be more highly valued than others in some contexts and therefore not recycled), although this social context may also change through time, even if the object itself does not move. These ideas are best illustrated by some hypothetical examples.

1.1. A linear trajectory

An Emperor demands that a particular object be made using copper from a specific source. This primary unit of copper is likely to be alloyed with tin and perhaps lead. The object is made and performs a specific function for a period of time, and is then buried in the tomb of the Emperor, perhaps 20 years after making. The burial of the object in the tomb marks a significant change of context, and therefore function, for the object, and the life history of the object is, of course, much longer than the 20 years before burial, since it continues to have a function in the tomb, and also has an existence following excavation. For our purposes, we may therefore need to consider a more specific interpretation of the life history of copper artefacts, in which there are 'active' phases (when human-induced change can occur) and 'dormant' phases, when the only possibility of change is as a response to environmental parameters. Divisions of this type have been criticised in contemporary conceptual archaeology, which aims to highlight the blend of human and object agency (Gell, 1998; Gosden, 2005; Jones and Boivin, 2010). We agree with the worldview that people and things are inextricable linked; what we present here is a heuristic device to highlight periods where our specific metal attributes are most susceptible to change. If we accept this division, and ignore for present purposes the life in the tomb and afterwards, then this example illustrates a simple linear life history, lasting a mere 20 years, and the life histories of the unit of metal and the object are identical. The effect of this simple life history on the three intrinsic attributes can be easily described:

- The trace element chemistry is fixed at primary production, but may change due to volatilization and oxidation during alloying and casting, or metallurgical working more generally (Bray and Pollard, 2012). Following production, however, the trace element chemistry of the metal is fixed, apart from surface changes due to corrosion, cleaning, etc.
- The alloy composition is fixed by the choice of metals added to the copper to produce the desired mechanical and visual properties, and will not change after manufacture. The lead isotope signal will be on a linear mixing line between the value in the copper source and that in any alloying elements. For a leaded bronze, it will of course be overwhelmingly dominated by that in the source of the lead.
- The form is fixed on production and does not change. Its typology is therefore fixed, although we might imagine minor modifications such as the addition of new decorative features, or repair.

1.2. Long, complex, and branching trajectories

Although a simple, short, linear pathway between origin and deposition is possible, it is not the only trajectory we can consider. The mutability of copper and its relative resistance to corrosion lends itself to long and complex lifetimes. As a thought experiment we can easily imagine, for example, that the linear path of an object such as an axe could be significantly extended, if, instead of being buried at the death of its first owner, it was kept among the living as a blend of practical object, memento and heirloom. In this scenario, it might remain in use for several generations: inherited, curated, and passed along repeatedly. Over this long-lifetime it may accrue significant symbolic capital, it may still be retained but transformed, perhaps into an ornament or some other item. We could imagine that it may continue in this way for some time, until it is lost or deposited. At almost any point in this process the axe or ornament could be passed on to a new social context (traded, looted, exchanged, gifted, etc.). It may be the case that in this second context social norms have dictated that an axe can only be remade into another axe, because an axe is a special form. It is, however, the wrong shape, weight, or colour for an axe in this new context, so it is recast into an axe of a new shape, perhaps with new copper added (possibly from a different source from the original unit of copper) to increase the weight, and perhaps by the addition of a new quantity of tin because it wasn't quite shiny enough. This axe continues in this way for a further period of time, perhaps being sharpened or otherwise mechanically reworked. This cycle may go on for years or centuries, until it finally enters a cultural context which does not value or respect its form as an axe, at which point it becomes 'scrap metal', to be mixed with other unvalued object forms, and recast as part of a cauldron, or some other completely unrelated form. This object, may, in turn, be cycled through a different social system, until eventually entering the archaeological record in some form, possibly centuries after the original unit of copper was formed. By this time, this unit of copper may have been divided and diluted many times, so that it only forms a small proportion of the copper in the deposited object, and many otherwise completely unrelated objects may contain a proportion of the original unit of copper.

This complicated and branched life history may last many generations in active form and have gone through numerous changes of social context, and result in the original unit of copper (or parts of it) travelling many thousands of kilometres, through series of hops. The object which contains it is no longer directly recognisable chemically or isotopically as copper from the original mine, since the trace element and lead isotope signals will have been overprinted, blended, and/or diluted many times. Likewise, the form may have changed many times, sometimes only slightly (e.g., one shape of an axe to another), but sometimes very radically. Typological 'form' can therefore be seen as a manifestation of the 'flow' of copper, frozen in time and space. Similarly, the nature of the alloy may have changed, either as a result of one or more deliberate re-alloying steps, but also more subtly as mixing of metals results in a gradual dilution of the alloying components. The end result may be a 'leaded gunmetal' alloy, in which all the major alloying metals (tin, lead, and, in later periods, zinc) are present, but all at levels below those we would expect in a 'designed' alloy, and below the levels which would have a discernible effect on the physical properties of the object.

Previous interpretative models of chemical and isotopic data appear to have generally assumed (implicitly) that all archaeological copper alloy objects follow more or less the first (linear) path. For objects which do, then the traditional chemical and isotopic approaches to provenance are likely to be valid. If some or many do not, then they are flawed, and may lead to erroneous conclusions. Careful study of object biography and technology might allow linear as opposed to complex pathways to be detected for a particular time, place and social context, but in general they are not easily distinguished. The purpose of our proposed methodology is to help reveal such information. Our new approach requires no assumptions about whether a society employs high levels of recycling, consistent re-alloying, traditional use of heirlooms or any other practice which might upset simple linear provenance models. What we propose instead is a data-led approach, which in conjunction with archaeological context reveals structure derived from the specific, historically constituted flows of metal in the past. In this paradigm, the concept of 'source' or 'provenance' of the metal becomes multiple and fuzzy. For many archaeological questions the ultimate geological origins of the original metal object(s) may even become relatively unimportant as metal is mixed and traded from hand to hand many centuries after, and many kilometres away, from where it was mined. This is the basis of our recent critique of provenance as generally carried out on prehistoric material in general (Pollard et al., 2014) – the fact that the dimension of the time elapsed between primary extraction and archaeological deposition is rarely if ever taken into account.

At this point, it is better to change the question completely, and ask not where the copper or the alloying metals originally come from, but to focus attention on the changing nature of the flow of metal, through multiple objects and typological forms, since this gives information about how metal was actually used in the past, how it travelled, and even how it was thought of. That is not to say that provenance is no longer of importance to archaeologists – the balance between the different tributaries of the flow of copper is primarily a reflection of the balance between the different sources of metal in use at the time. We would argue, however, that the composition of the flow of metal contains information of far more value than simple notions of 'source' – but they are complex and socially-embedded. Many archaeologists are now beginning to appreciate the potential importance and impact of recycling in the past, and we suggest that the methodology outlined below is a way of directly addressing these issues.

2. New approaches

We have devised a new integrated methodology which combines a model for the chemical changes in copper-alloys caused by technological processes (well-established in metallurgy, but largely overlooked by archaeometallurgy) with a re-definition of the terminology for alloy composition which does not assume deliberate alloy design, and a new way of interpreting lead isotope data that is more sensitive to mixing. We then combine **ubiquity analysis** (the percentage of a particular assemblage made up of a particular type of copper or alloy) with spatial and temporal analysis to follow these subtle chemical shifts caused by human intervention. This allows chemical change – in a full geographical, archaeological and chronological context – to act as a proxy for aggregated socio-technological practice, tracing traditions of human engagement with copper and bronze over time. Because our approach requires a large dataset to be meaningful (it is a 'Big Data' approach to archaeometallurgy), we also consider the issues which arise when combining chemical data from many sources.

2.1. Trace elements and 'copper groups'

For interpreting trace element data in copper alloys, we use a two stage process. Firstly, we carry out a simple presence/absence classification system based on the four most commonly-reported trace elements – arsenic (As), antimony (Sb), silver (Ag) and nickel (Ni). This is a simple heuristic sorting step, which allows us to see the dominant signals running through the data (Bray, 2009; Bray and Pollard, 2012; Pollard and Bray, 2014, in press: see Fig. 1). These elements are related to ore-source since they tend to be either present or absent in the ores known to have been used in antiquity, but we make no assumptions about allocating a particular copper group to a specific source, known or unknown, since mixing and recycling can move an object from one group to another. At this stage, we are only interested in the geographical, typological or chronological relationships that are immediately apparent, with no prior assumptions required about mines or geology. Tracing these changes over time, through a landscape, and between social contexts is at the heart of interpreting metal flow.

We allocate a metal composition to one of 16 categories, on the basis of presence/absence (Y/N) of each trace element. Thus, a metal with arsenic but nothing else would be YNNN (assigned Group 2: see Fig. 1). For most datasets, we use a figure of 0.1% (after mathematical removal of any major alloying elements present and renormalization) as the division between presence and absence, but this can (and should) be tested for stability by systematic variation of the cut-off value in any particular dataset, and the exact value for each element may depend on the specifics of data consistency and quality. We emphasise that these metal types do not necessarily correspond to specific 'sources' or 'mine sites'. A single metal group may contain copper from one or more discrete geological sources, and conversely a particular mine may supply copper of more than one group. We then use mapping to determine the extent, movement and timing of the circulation of particular metal types. An example is shown in Fig. 2, where Group 2 metal (YNNN, or copper with only arsenic) originating in southern Iberia (as evidenced by ubiquities >50%) moves up the Atlantic coast of France and then into the UK. In the original work (Bray, 2009) another source entering Europe was indicated by the appearance of Group 2 metal in western Anatolia and the Balkans. The distribution of this throughout the Near East is confirmed by subsequent work extending the map eastwards (Cuénod et al., submitted for publication), showing possible source regions in Cyprus and Afghanistan. Clearly, we need more data to 'fill in' the gaps in these large-scale maps. A key issue in this work is obviously the choice of chronological resolution and region size in the mapping. Often these decisions are imposed pragmatically by the quality of the archaeological chronologies available for the metalwork and the number of objects analysed, combined with the need to keep the numbers of objects in each regional and chronological group above a significant minimum. The methodology becomes increasingly powerful, however, when combined with Geographical Information Systems (GIS: e.g., Perucchetti et al., submitted for publication), which allows more sophisticated mapping (specific locations rather than arbitrary regions) and allows the use of geospatial statistics to investigate the relationship between metal distributions and topographical features such as mountains and river systems. An important aspect of this methodology is that it is independent of, but not oblivious to, archaeological evidence for mining and processing sites, and the known ore geochemistry of such sites. We can

Copper Category	Copper with...	As	Sb	Ag	Ni
1	None	no	no	no	no
2	As	YES	no	no	no
3	Sb	no	YES	no	no
4	Ag	no	no	YES	no
5	Ni	no	no	no	YES
6	As+Sb	YES	YES	no	no
7	Sb+Ag	no	YES	YES	no
8	Ag+Ni	no	no	YES	YES
9	As+Ag	YES	no	YES	no
10	Sb+Ni	no	YES	no	YES
11	As+Ni	YES	no	no	YES
12	As+Sb+Ag	YES	YES	YES	no
13	Sb+Ag+Ni	no	YES	YES	YES
14	As+Sb+Ni	YES	YES	no	YES
15	As+Ag+Ni	YES	no	YES	YES
16	As+Sb+Ag+Ni	YES	YES	YES	YES

Fig. 1. The 16 'copper groups' as defined by the presence/absence of the four commonly measured trace elements. 'Presence' is usually taken as greater than 0.1% (see text).

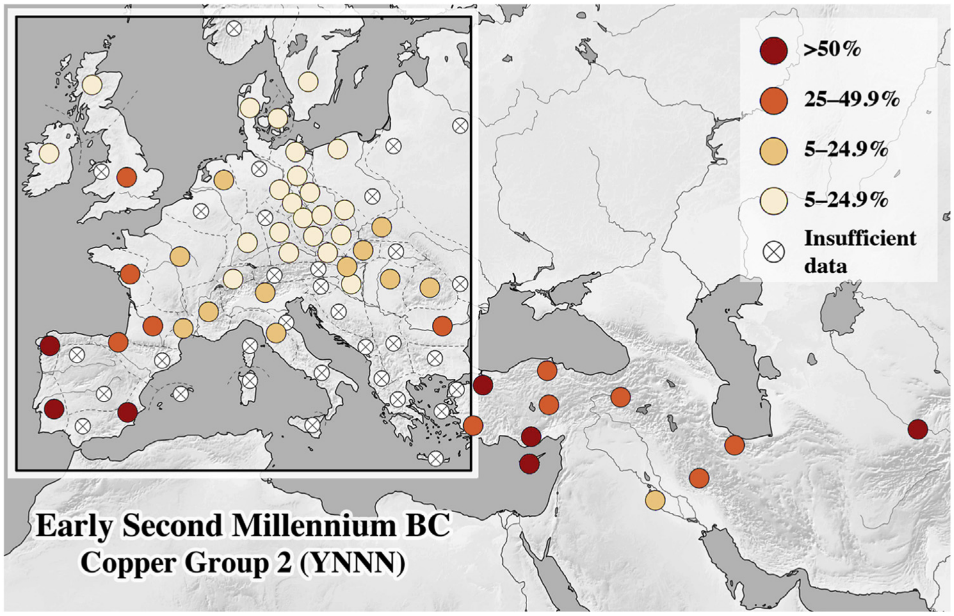

Fig. 2. Map of the ubiquity of Group 2 metal (YNNN, or copper with arsenic) in the European and Western Asian Early Bronze Age. The inset map of Europe is from Bray (2009), showing an Atlantic coast transport of such metal into France and Britain and possibly another source coming into SE Europe via Anatolia. The larger map (based on Cuénod, 2013) of Western Asia is less complete, but shows the distribution of this metal in the Near East with possible sources in Anatolia and Afghanistan.

infer the existence of a source for a particular type of copper from ubiquity mapping – 'hot spots' in the ubiquity map corresponding to likely source areas. This then allows us to cross-check against the locations of known mining or production sites and to compare ore geochemistry with copper group.

The second level of analysis aims to characterise the distribution and relationship between the elements present within these heuristic presence/absence copper groups. In short, arsenic and antimony are relatively vulnerable to oxidation and loss on recycling if the metal goes through a melting process, whilst silver and nickel are more resistant to removal. These observations are based on modern data from the metal recycling industry, where selective oxidation is used to extract precious metals from scrap, and have been confirmed by our re-melting experiments and fundamental thermodynamic calculations (Sabatini, in prep.). Because of their lower vulnerability to oxidative loss, a reduction in the average silver or nickel content is more likely to be caused by dilution with a unit of silver- or nickel-poor material. In Fig. 3 the average arsenic

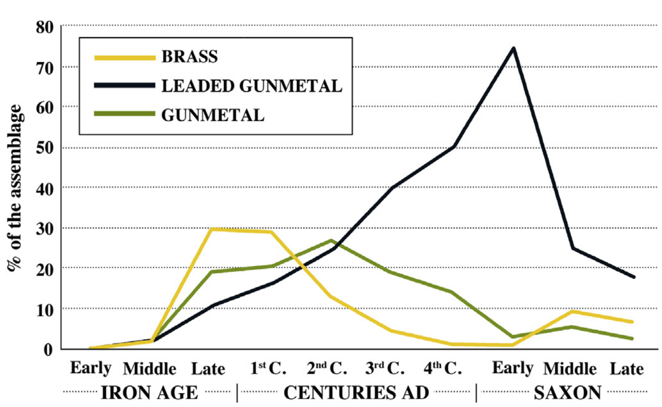

Fig. 3. Ubiquity of redefined alloy types in Roman Britain from the Iron Age to the Late Saxon period. This shows the injection of brass (BR; Cu plus >1% Zn) in the early 1st C AD, and the rise of leaded gunmetal (LG; Cu plus >1% Pb, Sn and Zn) through the end of Roman Britain into the Early Saxon Period, suggesting considerable recycling and the continuity of metal in circulation. From Pollard et al. (in press).

levels of Group 2 (YNNN) axes in Iberia are 2.2%, falling to 1.8% in Western France, and only 0.8% in Southern England. These changes in composition are associated with a change in the shapes of the axes, and thus the chemical shifts are consistent with the objects being recast into locally desired forms after each short range exchange of metal.

2.2. *Alloying elements*

A similar approach can also be used to classify the major alloying elements in a copper alloy object – here considered to be Sn, Pb and Zn, but in some cases As and Sb may have been deliberately added. Rather than using modern definitions applicable to designed alloys, we have adopted a presence/absence definition, in which we set the threshold of 'presence' at 1%. Thus we would classify an alloy containing 92% Cu, 2% Sn, 2% Zn and 4% Pb as a 'leaded gunmetal (LG)', despite the fact that this composition does not correspond to the definition of a modern 'leaded gunmetal' alloy. This system has the advantage of highlighting rather than hiding the presence of such mixed alloys in assemblages. If a high proportion of an assemblage is made up of such alloys, it suggests that those objects may be the result of mixing metals of more than one alloy type, rather than of deliberate alloy design. This interpretation is supported by the observation that such mixed alloys are unlikely to be designed, since alloying elements at low levels are unlikely to have affected the physical properties of the objects. Rather than denying the existence of 'designed alloys', however, it enables us to identify them more clearly when they do appear in the metal flow. For example, in a study of first millennium AD copper alloys in Britain, we have used this methodology to show continuity of metal circulation from the Late Roman period into the Early Anglo-Saxon, with a marked change occurring only in the Middle Saxon period, which we attribute to the arrival of fresh stocks of metal (Pollard et al., in press). Moreover, using the ubiquity of the quaternary alloy leaded gunmetal (defined as above) as a proxy for the amount of recycled metal in circulation, we have suggested that by the end of the Roman period, approximately 70% of objects analysed contained recycled metal (Fig. 3).

We can, however, take this idea further, by considering the distribution of the concentration of alloying elements within a particular assemblage, and also by mapping the ubiquities of different types of alloys. We can, for example, infer whether the bronzes in a certain region are primary or recycled by considering the shape of the distribution of tin concentrations in the assemblage. An approximately normal distribution of tin centred around 10% indicates that the assemblage consists of a primary alloy – i.e., that the objects are made from copper that has been alloyed with tin, and these bronzes have not been subsequently recycled. A distribution which peaks at a much lower level (or at <1%), and tends to only have a tail towards higher values, is indicative of a recycled alloy, where copper and bronzes of different tin levels have been mixed together. By plotting the ubiquity of tin bronzes, and the shapes of these tin distributions, on a series of time-resolved maps, it is possible to chart the development and spread of the use of tin bronze, and even to indicate where the tin itself might be coming from (see Fig. 4). 'Hot-spots' on these maps show the earliest locations of the widespread adoption of tin bronze, which might indicate the location of the tin sources. Even if, as may often be the case, these 'hot-spots' do not coincide directly with known tin sources, they do give us information about where tin bronzes were actually being used, in what social context this was occurring, and which objects were preferentially being made of bronze. This may be at least as important as knowing where the tin itself comes from.

A further advantage of our methodology with regard to trace elements and alloying is that it allows us to consider the circulation of copper independently of what it is alloyed with. There has been a general tendency in previous studies to focus solely on particular alloy types, once alloys appear, with the implicit assumption that alloys are immutable once formed (i.e., a tin bronze will always be a tin bronze). We make no such assumption. We can imagine copper being alloyed and re-alloyed any number of times, perhaps in 'consumer' regions rather than in primary metal-producing areas. In our methodology we can continue to discuss the underlying copper chemistry regardless of whether it is in the form of pure copper, or a leaded bronze, etc. This allows us to compare flows of different alloys in terms of their underlying copper chemistry (i.e., to think about whether different alloys are coming from the same copper source), and also to distinguish between the circulation of pre-alloyed metals and the practice of alloying (or re-alloying) in the consumer society. In effect, it is a powerful new tool, which allows us to ask fundamental questions about the concept of alloys, and the roles and identity of metal in society.

Fundamentally, therefore, we view trace element and alloy composition as properties which may vary through the life history of a particular object (through volatilization, oxidative loss, mixing, re-alloying, and general recycling). By combining details of the artefact typology, chronology, context and chemical characterisation at the assemblage level, a broad picture of the nature of the underlying metal flow may be inferred through these procedures. This perception of composition as being 'fluid' is in stark contrast to all previous methodologies (which we think of as 'static' models), and is why we believe that previous interpretations based on the cluster analysis of these 'static' compositions are potentially flawed. Of prime concern is the tendency of clustering to divide otherwise relatively homogeneous groups into subgroups on the basis of, for example, high and low arsenic. If we accept that arsenic (and other trace element) levels reflect a continuum which is contingent on the life histories of the objects, then it is inappropriate to use cluster analysis to define rigid groups based on data which are inherently variable and are not uniquely related to source.

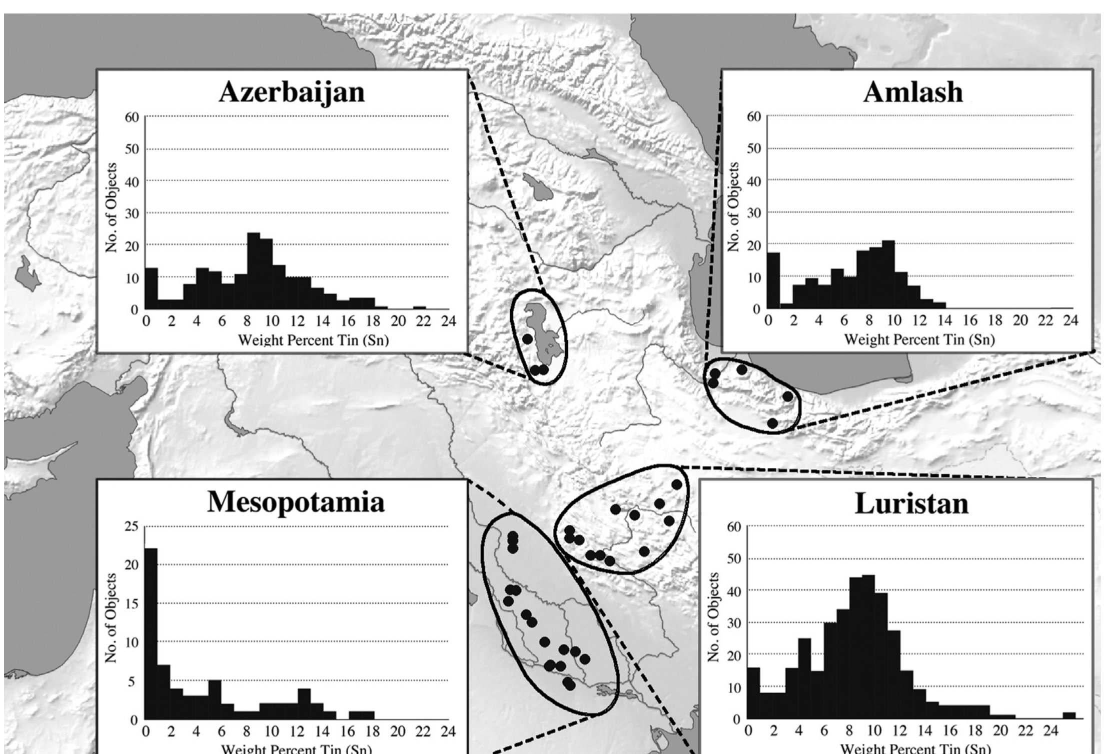

Fig. 4. Tin distributions in late 2nd to mid-1st millennium BC assemblages in Iran and neighbouring regions. Luristan has a distribution suggestive of the primary production of bronze (approximately normal Sn distribution centred on ~9%), whereas Mesopotamia has a typical 'recycled bronze' distribution (asymptotic to zero). Redrawn from Cuénod et al. (submitted for publication).

2.3. Chemical data quality

The use of chemical data compiled from published sources, many of them old, and using obsolete methods of analysis, is an obvious concern. It is well-known that some methods have systematic problems with certain elements. Moreover, virtually none of the literature contains information on primary or secondary standards, levels of detection, precision, or accuracy. How, then, can such datasets be combined? Firstly, we must ensure that the data we are using contain valid estimates for all of the elements we are interested in. For example, it is not always possible to interpret the meaning of '–' in some data. Does it mean 'absent' (i.e., below a probably unspecified level of detection), or 'not looked for'? – in which case we would not use the data. Similar considerations apply to the interpretation of entries such as 'tr', or semi-quantitative results such as '+', '++' etc. This process requires considerable historical and analytical knowledge. Secondly, our interpretations are based on group averages from a large number of analyses – the groups being defined on the basis of geography, chronology and typology. This is not an infallible defence against bias, since for some sites and periods the data are by a single analyst, but it is protection against rogue data. Most significantly, however, the use of 'presence/absence' for our preliminary classification renders our approach considerably less vulnerable to analytical variation than one which is based on absolute values (such as cluster analysis). The exception to this is our vulnerability to variations in limits of detection around our arbitrary cut-off values. Normally, when using data of mixed quality, we systematically vary the cut-off around the value of 0.1% to ensure that the results are not unstable. We believe that the difficulties involved in using old data are more than offset by the advantage of being able to use large datasets – often including objects on which it would now be impossible to obtain better quality data, given the tendency in most museums to restrict analysis to 'non-destructive' methods. This method therefore has the advantage that it can be applied without re-sampling, and allows us to use the archaeological equivalent of a 'Big Data' approach.

2.4. Lead isotope data

If we accept that, at least at certain times and places, metal recycling was a significant facet of human behaviour, then the uncritical use of lead isotope data on copper alloys is potentially misleading in terms of provenance (Pollard and Bray, in press). However, when viewed in a different way and when combined with chemical and other evidence, it provides a key to disentangling this complex picture. It is obvious that if metal from different sources is being mixed and recycled, then the measured lead isotope signature may not correspond to any one specific source. The conventional means of interpreting lead isotope ratios ($^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$, or similar) in archaeology has been to plot a pair of bivariate scatter diagrams, thus conveying all the available information in two diagrams. This is derived from geological practice, which plots a pair of isotope ratios to calculate the geological age of the lead deposit. When the method was extended to the lead impurities left in objects smelted from impure copper ores (Gale and Stos-Gale, 1982) it would appear that insufficient consideration was given to the most appropriate way to interpret the data. Hindsight suggests that the traditional isotope bi-plot is not optimal, particularly if mixing occurs. We have suggested a different set of three diagrams, which plot the Pb concentration against each isotope ratio. This is more akin to the interpretation of strontium isotope data, with the advantage that mixtures of two components, having different chemical abundances and isotopic ratios, show up as hyperbolic mixing lines, which become linear if plotted as isotope ratio against inverse concentration (Fig. 5). This method provides a highly appropriate model upon which to base a complete and radical re-interpretation of archaeological lead isotope data in copper alloys.

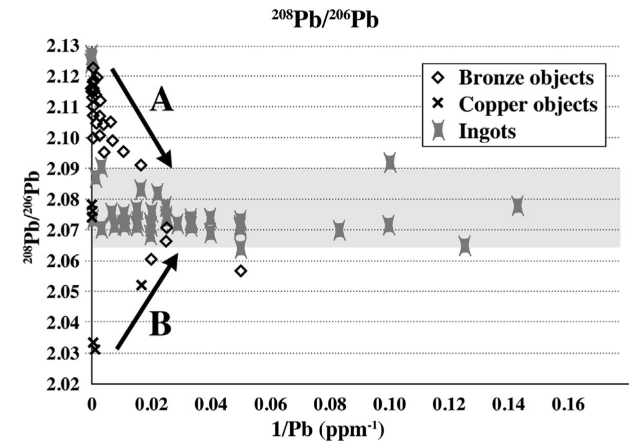

Fig. 5. Plot of 1/Pb against $^{208}Pb/^{206}Pb$ for Bronze Age copper artefacts and ingots from Sardinia. The ingots are shown to be consistent with a Cypriot source, but the lines marked A and B indicate mixing between local metal and the ingot metal, which was previously not recognised. Figure redrawn from Pollard and Bray (in press).

3. Conclusions

All things are transitory. Objects can be born and re-born. The form, chemical and isotopic composition and alloy formulation of a particular copper alloy object are but snapshots in time, with chemistry that is related to the composition of the underlying flow of copper when the object was temporarily removed from this flow. Therefore we argue that focussing on the provenance (i.e., the source of the copper, or perhaps the alloying metals) of individual objects is largely meaningless unless other information indicates that a specific object has had a very simple life history. The onus is on the analyst to demonstrate the validity of this assumption, rather than assume that mixing and recycling are unimportant. For the majority of objects, it is better to think of them as samples taken from an underlying flow of copper, the composition of which is dictated by a number of factors, including the geological source of the copper inputs, but also by recycling patterns, and modified by human practices such as re-melting and re-alloying. It is reconstructing the life history of this flow which gives us meaningful information about human interactions with metal. The strength of the method we propose is that it works in situations where 'linear' provenance studies are still appropriate, but also in the complex situations which we suggest better reflect the majority of archaeological reality. Determining the 'provenance' of the bell made in Bruggewauter in 1284 by Messrs Maydous, Erl, le Large and de Dunsterre is clearly not simply a matter of measuring the trace elements and/or lead isotopes in the object and matching them to a single source of copper. The results may therefore be highly misleading. We have outlined here a theoretical framework which leads to a more socially-embedded methodology for disentangling this story.

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