Approaches to interrogate the erased histories of recycled archaeological objects

Jonathan R. Wood¹,²

¹Institute of Culture and Heritage, Northwestern Polytechnical University, Xi’an, China
²University College London, London, UK

Abstract

Any archaeological artefact made from recyclable material may have been recycled before deposition. Three approaches are presented which have identified recycling in the archaeological record: (1) the application of log ratio analyses to investigate compositional data indicates that Roman glass was recycled and reapplied as a glaze on Parthian pottery, thereby suggesting that the paucity of Parthian and Sasanian glass in the archaeological record is due to recycling; (2) linear mixing lines on plots that combine compositional and isotopic data suggest that most silver found in the Iron Age hoards of the southern Levant was mixed, with vertical mixing lines indicating that some of it was melted down hastily in times of unrest; and (3) histograms of compositional data provide evidence of recycling accompanied by dilution of cobalt-blue glass in New Kingdom Egypt, potentially because the colourant was not available in later periods, thereby questioning the accepted provenance of the cobalt source. It is considered that application of these approaches can contribute to a better understanding of the motivations behind recycling in prehistory.

KEYWORDS
dilution, glass, glazed pottery, lead isotopes, log ratios, mixing, silver

INTRODUCTION

Until recently, recycling was regarded as being not particularly significant in prehistory. For example, the practice of burying silver objects with their owners, thereby removing silver from
circulation, was believed to have sustained a demand for new silver during the Aegean Bronze Age (Gale & Stos-Gale, 1981). Furthermore, isotopic matches between oxhide ingots recovered around the Mediterranean and copper ore sources in Cyprus were considered sufficient to negate the possibility that similar signatures could emerge by mixing metals from different sources (Gale, 2001). Recycling was also the first hypothesis to be rejected in favour of an assumed chemical compositional profile of an unprovenanced geological source to explain the presence of cobalt-copper glass in Egypt and Mycenae (Smirnou & Rehren, 2013). As Pollard et al. (2018: 38) highlight, many studies ‘have either ignored completely the potential complexities arising from the mixing or recycling of metal from different sources, or have acknowledged that such practices would invalidate the simple hypothesis of provenance, but have then largely dismissed the problem’.

To some extent, these issues are now being addressed by focussing not exclusively on the provenancing of objects (Pollard et al., 2018) and through the development of theoretical frameworks that advocate the inclusion of the mutability of artefacts in the chaîne opératoire (Sainsbury, et al., 2021)—that is, beyond a recursive arrow on a schematic diagram between the finished object and the workshop dealing with the raw materials (e.g. Martinón-Torres, 2002: Figure 1). Nonetheless, before any discussion can be held on the motivating factors for and the implications of recycling, it is necessary to identify whether the artefacts, which provide evidence for such discussions, had been recycled or not. In effect, any object that could be thrown back into a melting pot might have been recycled before it entered the archaeological record. Furthermore, an archaeological artefact made from recyclable material, such as glass or metal, may neither be the original object nor resemble it, with the consequence that a successful

![Logratio analysis (LRA) of the data in Table A1, where the additional samples in Table A2 are displayed as supplementary points (Roman samples 171, 163 and 156 in italic on the right) Notes: 95% confidence ellipses around the group means (large symbols: P, P-S and S) of the three groups, clearly demonstrate that S (Sasanian) is separate from P (Parthian), with P-S (Parthian-Sasanian) overlapping with both S and P. 74.0% of the variance is explained by the two principal dimensions](image-url)
ancient recycling economy can become invisible when recycling is synonymous with re-melting rather than reuse (Freestone, 2015; Ponting & Levene, 2015).

There are two main ways in which archaeology can approach recycling: the first is to identify material that is out of place in an archaeological context that can be deduced to have been recycled; the second is by investigating the recycling event, as episodes of recycling can result in the deliberate or accidental introduction, dilution, and loss of components compared with those in the original object. Both scenarios require that something is known about the original non-recycled material, such as its chemical composition (often referred to as an end member when discussing recycling in terms of mixing) before it can be suggested that recycling must have occurred.

This article examines how archaeological science can reveal evidence of recycling in different contexts, and shed light on the reasons (social, economic, political etc.) behind it, through examination of:

• Out of context materials: recycled Roman glass to glaze Parthian pottery in Mesopotamia
• Mixing lines: mixed silver found in hoards of the Iron Age southern Levant
• Distributions: dilution and contamination of cobalt-blue glass in New Kingdom Egypt

It should be noted that the approaches described in this article are not prescriptive but were chosen because they aligned with the data available. In effect, this article aims to demonstrate through the application of relatively well-known mathematical procedures to legacy data found in the published literature that episodes of recycling can be identified in the archaeological record.

OUT OF CONTEXT MATERIALS: RECYCLED ROMAN GLASS TO GLAZE PARTHIAN POTTERY IN MESOPOTAMIA

Alkaline glazes were first used on clay-based ceramics in Mesopotamia around 1,500 BCE, at the same time as the appearance of the first glass vessels, establishing a tradition of making glazes with plant ash, which continued through to the early Islamic period (Hedges & Moorey, 1975; McCarthy, Vandiver, & Gibson, 1995; Peltenburg, 1971; Vandiver, 1983). By contrast, the Roman Empire used lead-based glazes (Hatcher, Kaczmarczyk, Scherer, & Symonds, 1994; Walton & Tite, 2010), with alkaline natron (a form of hydrated sodium carbonate) glass being used only to produce objects of glass (Freestone, 1991).

On first inspection, these observations would appear to be unrelated to recycling, as pottery cannot be remelted. Glazed pottery, however, does have a recyclable component: the glass applied to glaze the pottery fabric. In effect, glazes may have been made from recycled glassware (Wood & Hsu, 2020), suggesting the existence of an organised recycling economy.

The approach applied to investigate this hypothesis was first to examine legacy data from chemical studies of glazed pottery from the Parthian and Sasanian periods, in order to identify compositional signatures (i.e., meaningful patterns in the data). Table A1 in the Supplementary Material presents legacy data from Parthian and Sasanian glazed pottery. Further details of the criteria for the selection of the legacy dataset and experimental protocols can be found in Wood and Hsu (2020). The main components of Mesopotamian glazes derive from silica and plant ash (Brill, 2005; Freestone, 2013; Hedges & Moorey, 1975; Simpson, 1997) and contain oxides of silicon, aluminium, iron, magnesium, calcium, potassium, and sodium (SiO₂, Al₂O₃, FeO, MgO, CaO, K₂O, and Na₂O, respectively). These components can also be found in pottery body fabrics that could influence the composition of the glazes during firing. However, McCarthy (1996) measured the compositional profile across Parthian glazes showing that there was very little change from the outer surface of the glaze to the glaze–body interface for all
measured oxides, suggesting that levels in the glaze do not change significantly. Similar experiments have found that any influence of the body fabric on the composition of the glaze is confined to a narrow layer (<150 μm from the fabric) immediately adjacent to the body (Wood & Hsu, 2020). It has therefore been assumed that compositions of the glazes predominantly reflect the recipes used to make the glass frit applied to the body.

The approach followed here treats the system as a series of subcompositional components, or parts, realising that the oxides being analysed are a subset of a potentially larger set, which affects their percentage values. The main idea is to analyse oxide ratios, which are unchanged whether observed in subcompositions or extended compositions, and to logarithmically transform the ratios to put them on an additive (interval) scale, resulting in log ratios. This method was first proposed by Aitchison (1986), who defined a relative geometry where only the pairwise log ratios of the different components were considered (see Aitchison 2005). The motivation for an archaeologist to use a method based on ratios of components is that this makes compositional data, and thus the results of the statistical analysis, invariant to the particular choice of components. In this study there were seven components in the form of major element oxides (Table A1). This choice depended on the availability of data. Extending or reducing the number of components (if greater or fewer oxides had been available) changes the compositional data, but the ratios between components remain unchanged.

Apart from the simple pairwise log ratios of parts, other log ratio transformations are possible, the most useful being the centred log ratios (CLR), the logarithm of each component (oxide) $x_j$ relative to the geometric mean of all the parts:

$$\log \left( \frac{x_j}{(\Pi_k x_k)^{1/D}} \right) = \log (x_j) - \left( \Sigma_k \log (x_k) \right)/D \quad j = 1, \ldots, D$$

It has been shown that the principal component analysis (PCA) of the $D$ CLRs is equivalent to the PCA of all $\frac{1}{2}D(D-1)$ pairwise log ratios (Aitchison & Greenacre, 2002). This method of dimension-reduction is called log ratio analysis (LRA) and its interpretation differs from that of regular PCA in that it is not the parts themselves that are interpreted in the solution but rather the directions linking pairs of parts, which represent the log ratios (for a practical introduction, see Greenacre, 2018). In essence, the log ratio approach considers from the outset that the interest lies in the relative magnitudes and variations of components, rather than their absolute values, and that the data are unconstrained (thereby retaining the proper covariance structure of any compositional data). The transformation to log ratios then allows the full range of standard statistical methods, both exploratory and inferential, to be available for identifying and confirming groups or patterns in the data.

Figure 1 shows the results of the LRA, explaining 74% of the log ratio variance using the first two principal components of the log ratios. This biplot is interpreted in terms of the directions linking the tips of the arrows rather than the arrows themselves. These connecting links represent the ratios between parts (on a logarithmic scale), so that large distances between pairs of oxides represent high variances of log ratios, for example those of FeO/Na₂O or FeO/Al₂O₃, and so on.

Figure 1 shows that Parthian and Sasanian glazes can be differentiated from the compositions of the major element oxides, with the more nebulous Parthian-Sasanian group falling between the two main chronological groups. The confidence ellipses were determined by bootstrapping, which allows the computation of a region enclosing a multivariate group falling with prescribed confidence, in this case 95%. In effect, these confidence plots show the variability in the mean rather than the variability in individual values, that is, there is a 95% chance that the ellipse includes the true mean (for more details and the R code on how Figure 1 and the confidence ellipses around the group means were constructed (see Wood & Greenacre, 2021).
Figure 1 indicates common links in the legacy data, which suggest that a projection of selected variables onto a particular direction is quasi-constant for the following sub-compositions: (a) Al₂O₃-MgO-CaO; (b) Na₂O-MgO-CaO, and (c) FeO-MgO-CaO. These sub-compositions were used to determine compositional signatures for Parthian and Sasanian glazes, and produced legacy data bands on log-ratio plots (Figure 2a-c). Figures 1 and 2a-c also plot compositional data (Table A2) from Parthian pottery recovered at the Roman military outpost of Ain Sinu in northern Mesopotamia (modern-day Iraq) excavated in the 1950s by Professor David Oates and Dr. Joan Oates. Ain Sinu was contested territory for both the Parthian and Sasanian empires, being occupied by the Romans between A.D. 197–364 (Oates & Oates, 1959).

Three of the Ain Sinu samples appear to lie in different regions of the biplot (Figure 1), that is, the samples in italics. The log ratio plots in Figure 2 further interrogate these samples with respect to the legacy data. The majority of the Ain Sinu glazes fall within the Parthian/Sasanian legacy data bands (Figure 2a-c), suggesting that these glazed wares are from the same tradition as the legacy data. However, it is apparent that three sherds from Ain Sinu do not fall with the legacy data bands. The MgO concentrations for these glazes are considerably lower than the remaining Ain Sinu samples and those of the legacy data (Table A2; Table 1). The low MgO concentrations for these three samples are more comparable with alkali glazes based on the evaporite natron, a form of hydrated sodium carbonate found as efflorescence around soda lakes (e.g., Wadi Natrun in Lower Egypt) rather than plant ash.

Table 1 shows that the typical composition of Roman glass, which was based on natron and is often classified as ‘LowMagnesia’ (Freestone, 1991), is similar to the glaze compositions for

![Figure 2](image-url)

**Figure 2**  (a-c) Logratio plots of the glaze compositions for the legacy data and the Ain Sinu glazed sherds. (d) Bivariate plot of K₂O (potash) vs. MgO (magnesia) Notes: Open circles are from the legacy data (Table A1). Triangles show the samples from Ain Sinu (Table A2). Open triangles are those glazes from Ain Sinu which appear to have been fluxed using natron (highlighted in Table A2), i.e., Roman glass.
the three Ain Sinu glazes with low magnesia levels. This would suggest that Roman glass was applied to Parthian fabrics to produce these glazes. Because Roman glaze production used lead-based glazes (Hatcher, Kaczmarczyk, Scherer, & Symonds, 1994; Walton & Tite, 2010), this would indicate that Roman natron glass was recycled to glaze Parthian pottery.

There are some differences between these three ‘natron’ glazes and Roman glass (Table 1). The K₂O concentration is several times higher in these glazes than for Roman glass. This could indicate that another source of potash entered the glaze systems, other than that associated with plant ash, that is, a source unrelated to the initial composition of the glaze. Paynter (2009) suggests that differences between Mesopotamian glasses and glazes could be a consequence of contamination from fuel ash or vapour, especially hard-wood fuel. Because glazes have a higher surface area to volume ratio compared to glass heated in a crucible, ash or vapourised potassium compounds are more likely to affect the composition of a glaze by adhering to the molten surface. This would suggest that levels of K₂O in the glazes investigated in this study (legacy and Ain Sinu glazes) could be a consequence of potassium compounds from the plant ash flux and the type of fuel used in Parthian and Sasanian kilns, thereby diminishing any co-dependence between K₂O and MgO from the plant ash. This is supported by the invariance of K₂O with increasing concentrations of MgO on a bivariate plot for the legacy data and the majority of the Ain Sinu glazes (Figure 2d). Moreover, the contamination argument outlined above could suggest that the three ‘natron’ samples (red open triangles in Figure 2d) were produced in the same tradition as the other glazes, but that the starting composition of the glass was different to the other glazes tested here, and to those of the legacy data. To some extent this feeds into the theoretical approaches on mutability (Sainsbury et al., 2021), in that changes in function can be viewed through variation in chemical markers that occur during recycling (in this case, increases in K₂O with the change from glass to glaze), in a similar fashion to how volatile compounds such as arsenic compounds found in copper alloys (Bray & Pollard, 2012; McKerrell & Tylecote, 1972) and cobalt-blue glass (Wood & Hsu, 2019) can be lost on remelting. It is therefore proposed that these three samples from Ain Sinu started out with lower concentrations of K₂O (because they are Roman glass, fluxed using natron) but were contaminated with K₂O because they were produced in the same tradition as the other glazes.

A further indication that these ‘natron’ glazes derive from Roman glass is evident from their antimony and manganese concentrations. Roman colourless glass from the first to third centuries A.D. was often decolourised using antimony (Jackson & Paynter, 2016) (mean = 0.54 wt% Sb₂O₅ - Table 1). The average results of the EPMA for Sb₂O₅ of the Ain Sinu samples are shown in Table 1 for the three ‘natron’ Ain Sinu glazes. Furthermore, two of the three ‘natron’ glazes have significant MnO levels (Table A2). All other sherds from Ain Sinu were below the detection limit for both antimony and manganese (Table A2). This could suggest that the Roman glass found on these two glaze samples had already been recycled, resulting in a mixture of decolourising agents, that is, Sb₂O₅ and MnO. In fact, at the extremes of the Roman world in the second and third centuries A.D., this recycled mixture appears to have become more readily available than the raw low-Mn blue–green glass (Jackson & Paynter, 2016). The
presence of mixed Roman glass, here reapplied as glaze on Parthian pottery, could reflect the potential difficulties experienced by this Roman outpost to acquire raw and ‘pure’ decolourised glass. 

Although only eight samples from Ain Sinu could be acquired for testing, the presence of Roman glass reapplied as a glaze on Parthian pottery is a good example of a material that is out of context and provides convincing evidence that glass was being recycled at Ain Sinu. In fact, if Parthian potters had not used Roman glass, it would have been much more difficult to identify that recycling had occurred. It also supports the possibility that the practice of glazing pottery in the Parthian Empire was to use a glass precursor in the form of a crushed glass frit applied in a slurry of local clay (Kingery & Vandiver, 1986: 58–62) rather than applying a fluxing mineral to react directly with the silica-rich clay surface during firing. For the three glazes that fail to lie within the legacy data bands in Figure 2a-c, this frit potentially derived from recycled Roman glass followed by typical Parthian methods of practice to fire a kiln and apply a glaze on a ceramic body (Figure 2d). This practice could indicate that glass and glaze were regarded as the same material in Mesopotamian production and that potters made their own frit specifically to glaze pottery and/or there are yet undiscovered glass production sites in Mesopotamia. Extending recycling to Indigenous glass production could provide an additional reason to weathering of glass as to the relatively low numbers of Parthian and Sasanian glass artefacts in the archaeological record, that is, each glass vessel could have been recycled to glaze two or three pots of equivalent size. Furthermore, the fact that none of the Sasanian legacy glazes appears to be made from Roman glass could suggest that the Parthian glass/glaze industry was less developed than its Sasanian successors, with Parthian potters, in some instances, having to recycle imported material.

MIXING LINES: MIXED SILVER FOUND IN HOARDS OF THE IRON AGE LEVANT

Recycling can involve mixing of components, either intentionally (perhaps to modify the colour or properties of the original material) or unintentionally (i.e. contamination) (Wood, Charlton, Murillo-Barroso, & Martinón-Torres, 2017a, 2017b). Mixing presents challenges for log ratios, as high values in the numerator or low values in the dominator can result in ratios with similar values. The problem for ‘standard’ methods, however, is that the whole data set is rarely (if ever) measured, as different instruments focus on different parts of the composition (e.g. pXRF cannot generally measure lighter elements). When amounts that are measured do not add up to the real total, their percentage values can change whether observed in sub-compositions or extended compositions.

Bivariate plots of elements and compounds are common in the archaeological literature. It is contended here that their application can lead to erroneous interpretations, particularly when datasets with different numbers of components are presented. This is a possible reason for the few studies in archaeological science that compare directly compositional data from different research groups. To demonstrate this point, the bivariate plot shown in Figure 3 plots iron oxide against silica for the legacy data on glazes described in Table A1. These samples are unlikely to have derived from the same melting pot. Nevertheless, a linear relationship can emerge, as shown in Figure 3. Here, the three graphs plot exactly the same data (FeOwt% vs SiO2wt%): The first plot closes the data to 100% for the following variables (SiO2, Al2O3, FeO, MgO, CaO, K2O, and Na2O); the second plot closes the data around the first 4 oxides (SiO2, Al2O3, FeO and MgO); the third plot closes the data around the two plotted variables (SiO2 and FeO). The plots clearly show that closing a subcomposition does not necessarily produce similar or compatible patterns, even if some of the components are irrelevant to the variables being studied. This occurs because of a geometrical artefact of compositional data: the
relationship between variables is not a real relation but one which is compelled, that is, as the proportion of one component increases, the proportions of other components must decrease, which in the extreme case of a two component system, would always result in a correlation coefficient of −1. Although this ‘spurious correlation’ (Chayes, 1949) may seem evident to many researchers, it is perhaps worth re-iterating that it means that any regression lines drawn to represent the interaction between the two components (in this case SiO₂ and FeO) cannot be trusted, that is, such an induced correlation may conceal true relationships among variables.

In addition to issues surrounding mathematical operations of closure or normalisation, linearity can emerge erroneously from plotting mathematically inconsistent combinations of compositional variables. For example, Freestone (Freestone, 2015: Figure 2) and Schibille et al. (Schibille, Sterrett-Krause, & Freestone, 2017: Figure 5) plot MnO/(Sb₂O₃ + MnO) versus Al₂O₃ to produce what is considered to be a mixing line between batches of low-alumina-antimony decolourised glass and high-alumina-manganese decolourised glass. However, there is no reason for the percentages of decolourisers in glass to add up to a unique value, which makes limiting values difficult to interpret: That is, if MnO approaches zero, MnO/(Sb₂O₃ + MnO) approaches zero, regardless of the amount of Sb₂O₃; similarly, when Sb₂O₃ approaches zero, MnO/(Sb₂O₃ + MnO) will always approach 1, regardless of the

FIGURE 3 Plots of legacy data from Table A1 normalised to 100%: Closed around 7 oxides (left) and 4 oxides (middle) and 2 oxides (right) Notice the change in scales and the emergence of linearity in the data as the number of elements used to close the data decreases
amount of MnO. Furthermore, when the denominator is zero, the limiting value is undefined. In effect, although mixing may have occurred, forced linearity can affect archaeological interpretations as it could imply, perhaps incorrectly, the mixing of two components (or batches) rather than simply non-structured variation.

To address issues such as these, compositional data can be combined with lead isotope analytical (LIA) data as both can reflect episodes of recycling but are not interdependent. For example, silver is often geologically associated with low concentrations of gold that, due to its chemical inertness, can survive silver smelting and refining operations. Moreover, silver is often geologically linked with lead, and lead is often used in silver extraction and cupellation. The isotopic signatures of the lead and the concentrations of gold associated with silver can therefore be used to provenance silver from ore to object (Wood et al., 2019).

This method of combining compositional data and LIA data has been used to investigate silver from Iron Age hoards in the southern Levant by applying the following strategy (Wood et al., 2017a; Wood et al., 2019): (1) LIA values are used to calculate the Pb crustal age (also known as the model age) of the ore, measured in millions of years (Ma), from which the silver derived (using a two-stage evolution model and the parameters of Desaulty, Telouk, Albalat, & Albarède, 2011); this age is then (2) plotted against the levels of gold found in silver, given that gold is a useful geological indicator for a silver source as it is likely to survive silver processing. This approach allows two types of data to be plotted together, providing two parameters which would encourage any variation due to the mixing of components to form mixing lines. In effect, because time (Pb crustal age) and concentration (Au/Ag) are linear variables, linear mixing lines should emerge when two components are mixed.

Figure 4 shows Au/Ag versus Pb crustal age for hacksilver (i.e. broken pieces of silver) recovered from in the later Iron Age hoards (c. late seventh century BC) of Ein Gedi and Miqne-Ekron in the southern Levant. The vertical axis shows the amount of gold in the silver, which could be either a geological remnant or introduced from recycling silver objects without consistently removing gold parts (i.e. gold rims, handles, gilt, etc.). The horizontal axis is the calculated crustal age of the ore from which the silver (or more precisely, the lead associated with the silver) originally derived.

A cluster is apparent at low crustal ages (around 0 Ma) and low gold concentrations (Au/Ag × 100 is approximately 0.2) from the hoards at Miqne-Ekron. It should be noted that the detection limit for these ED-XRF measurements was about 0.2% Au as indicated by the horizontal black dotted line. As such, the cluster appears more pronounced, as any value described as < 0.2 Au% was given the value 0.2 Au%. The lower plot in Figure 4 presents hand-drawn lines through the points. This process, albeit subjective, was considered preferable to fitting the data using linear trend lines, as it is unknown a priori which points correspond to which mixing line. In effect, these lines are interpretations of the patterns observed in the data, and it is appreciated that other interpretations could be made.

The green dashed line probably extends to even lower levels of Au/Ag than indicated on the plot, because of the detection limits of the ED-XRF, and is potentially a mixing line, formed by placing silver objects derived from one geological source (i.e., the Pb crustal age is relatively invariant, which suggests a single source) into a melting pot without removing all their gold parts, in order to make new silver objects. Such vertical mixing lines (with constant Pb crustal age but variable Au) may indicate the melting down and mixing of silver in times of unrest, both here and in other contexts. That is, silver of the same origin was hastily melted down without removing all gold embellishments into something easier to conceal, transport or trade. In fact, the six hoards of Miqne-Ekron, comprising 305 pieces of silver (Gitin, 1995), pre-date the final destruction of the city by the Neo-Babylonian King Nebuchadnezzar during one of his campaigns to Philistia (most probably in 604 BC) with researchers concluding that most of the silver was collected for its value as bullion, perhaps for safe keeping prior to the impending Babylonian attack (Porten, 1981).
The cluster of silver with low gold levels around 0 Ma suggest that one of the components was silver derived from the galena ores of Laurion on the Attic peninsula, that is, the cluster is consistent with the Pb crustal ages of Laurion ores (Wood, Montero-Ruiz, & Martinón-Torres, 2019: Figure 3). It is also consistent with the density plot of Athenian coins (Figure 4), which are believed to derive from the silver ores of Laurion (Gale et al., 1980). This further supports that the vertical mixing line in Figure 4 is a consequence of inadvertent mixing of gold, as gold concentrations are extremely low in silver derived from argentiferous galena (usually < 0.1% Au) (Wood et al., 2021).

More evidence of mixing can be observed for silver running along the Pb crustal age axis (the red dashed line in Figure 4). If silver with a higher crustal age was mixed in various proportions with silver from the cluster at 0 Ma, a series of silvers with relatively invariant gold levels but large differences in crustal ages would be produced.

The blue dashed line in Figure 4 shows, perhaps optimistically, a mixing line between silver from a high crustal age source with a low crustal age source that has a higher gold level than that from Laurion, that is, potentially silver from Laurion that had been mixed with gold to a level of Au/Ag × 100 of approximately 0.9. This mixing potentially produced silver with
intermediate levels of both gold concentration and Pb crustal age. The intersection of the blue and red dashed line would support that a high crustal age/low gold silver source was available at Phoenician-influenced Miqne-Ekron during the late seventh century BC.

Similar patterns can be observed in the earlier Iron Age hoards (Figure 5), where again the relatively linear lines provide the rationale behind these plots. A detailed interpretation of episodes of silver recycling in the Early Iron Age, as evidenced by the mixing of silver from trans-Mediterranean sources, is discussed elsewhere (Wood et al., 2020; Wood et al., 2019).

Although it has been speculated that the alloying of silver with copper could influence the isotopic signature (Eshel et al., 2021), thereby affecting the identification of the provenance of the silver, there are several reasons why the addition of copper would not generally affect the LIA of either original or recycled silver objects. First, copper concentrations are generally low in silver artefacts (often < 5%Cu) and copper can have a 'notoriously low lead content' (Stos-Gale & Gale, 1994:105). Second, although copper from Timna and Feynan (the proposed sources of copper for Levantine silver; Eshel et al., 2021) has relatively high concentrations of lead for copper, the levels of lead in copper ingots associated with these regions (Yahalom-Mack et al., 2014), as well as lead levels in copper ingots that isotopically match with these copper regions recovered at other locations (Montero-Ruíz, 2018), are not consistent with the absolute amounts of lead measured in the silver from these Levantine hoards. Furthermore, concentrations of lead and copper in the Levantine hacksilver are not correlated; that is, hacksilver with low copper levels (<5%Cu) has similar concentrations of lead to silver with higher levels of copper (>20%Cu) (see Eshel et al., 2021: Table B1). This suggests that it is
highly unlikely that the variation in lead isotopic signatures (and, thereby, the Pb crustal ages) of Levantine silver is due to variations in the concentrations of copper but rather because the hacksilver found in the Levantine hoards were mixtures of silver, of which one component was silver with a Western Mediterranean provenance (Wood & Montero-Ruiz, 2019; Wood et al., 2020; Wood et al., 2019).

In summary, the rationale behind these mixing plots is that compositional and isotopic data can be presented in a way that situates mixing events in time and space. In effect, silver objects deriving from mixtures of silver from different sources, which have been deposited at different locations and at different times, can be used to identify episodes of mixing. The patterns that emerge from analysing these mixing events, such as vertical mixing lines that potentially reflect social-political contexts when it became judicious to melt down silver objects rapidly without removing gold parts, provide a further approach to investigate the movement of artefacts that were recycled and reused prior to deposition in the archaeological record.

**DISTRIBUTIONS: DILUTION AND CONTAMINATION OF COBALT-BLUE GLASS IN NEW KINGDOM EGYPT**

Variation in compositional data of artefacts can be interrogated using histograms (e.g. Pollard et al., 2019; Wood & Hsu, 2019), with the caveat that samples must derive from many different objects. Furthermore, the underlying assumptions behind distributions could affect the identification of recycling, as mixing affects distributions by increasing variance (Perreault, 2019). This increase is potentially irrespective of whether mixing was a consequence an ancient melting pot or whether archaeologists combine artefacts from several contexts into one mixed assemblage after excavation. For example, if several archaeological contexts deposited at different points in time are analysed together, the variance within each context (i.e. the variance within a population at time $t$) will collapse into the variance that exists between contexts (i.e. the variance between a population at time $t$ and $t + 1$). Essentially, an archaeologist measuring and recording a continuous trait such as the diameter, weight, or chemical composition of artefacts and so forth, may combine different contexts, probably inadvertently, in the analysis. The greater the between-contexts variance, the more inflated the variance would be when assemblages are mixed together.

Figure 6a and b show that if assemblages have been mixed, the variance in the time-average assemblage would be much larger than it ever was at any single point in time, regardless of whether the mean of a trait changes linearly and gradually over time (Figure 6a) or whether the mean fluctuates over time (Figure 6b): That is to say, even for a constant variance, the distribution of the mixture would be wider, flatter, and converging to a uniform distribution. Similarly, a stable mean but changing variance would result in a time-averaged distribution in which the central values are overrepresented (Figure 6c). A similar increase in variance would happen if the same process occurred when different locations (perhaps on the same site) were combined (Figure 6d), that is, different areas with different means and/or variance from the same chronological setting.

A punctuated shift in the mean or a gap in the sequence of mixed contexts, however, would lead to a bimodal distribution. Figure 7a and b shows slow change punctuated with a period of rapid change. For compositional data, this means that if it can be assumed that punctuation is not a consequence of missing data (i.e. the absence of part of the sequence), a bimodal distribution for compositional data could suggest a sudden change in recipe.

The following histograms (Figure 8) plot cobalt-blue glass from New Kingdom Egypt (Wood & Hsu, 2019), showing a very similar pattern to those in Figure 7. The upper plot shows 30 compositional analyses from Abe et al. (2012): 26 different 18th Dynasty objects from museum collections in Japan, provenanced to Malkata and Amarna, and four 18th Dynasty
objects recovered at the site of Dashur. The lower figure plots 20 compositional analyses from different objects from 19th–20th Dynasty contexts at Dashur. Although differences in composition between the two periods have been attributed to the exploitation of a new cobalt source in the 19th Dynasty (Abe et al., 2012), it is proposed here that glass recycling is perhaps a more parsimonious interpretation.

Figure 8 (upper) shows that the distribution of cobalt oxide concentration in the 18th Dynasty glass appears to be bimodal, with the lower peak having about half the CoO level of the higher peak. The 19th–20th Dynasty glass (Figure 8 – lower) has a distribution which is similar to the lower peak of the 18th Dynasty cobalt-blue glass. Assuming that the bimodality is not due to missing data (which seems a reasonable assumption for sites that have been thoroughly excavated), this could suggest that the original glass was the higher peak in the upper
plot (i.e. the peak with a mean around 0.16wt%CoO), which was diluted in a 1:1 ratio with a base glass (i.e. glass without cobalt colourant) thereby diluting the amount of cobalt in the mixture to about 0.8wt%CoO. The 19th–20th Dynasty glass objects were potentially made from this mixture (or perhaps from a 1:1 dilution of remelted glass from both distributions found in 18th Dynasty contexts with a base glass). This dilution is also reflected in the concentrations of other oxides associated with the colourant, such as NiO and MnO (Wood & Hsu, 2019). In effect, this bimodal distribution would be consistent with a recycling process that also included dilution. Although such dilutions were probably derived empirically, it is interesting to note that the cobalt level in the glass from 19th–20th Dynasty Dashur (average CoO = 533 ppm) was around the lower limit to maintain a rich, deep blue (Henderson, 2000). This fits well with the narrative that glass was not produced in significant amounts after c. 1,250 BC (Shortland, 2016: 95), with glassmakers reducing the levels of cobalt through dilution with a base glass to the point where the colour was still effective.

Further support for mixing is found by examining possible contamination from lead and antimony (Figure 9). Recycling coloured glass requires that colours are separated prior to remelting. This is unlikely to be a completely efficient process, with elements required to make other colours potentially contaminating the cobalt-blue glass.

Although an indirect measure, the fact that the 18th Dynasty glass from the museum collections has an average of about 0.02%PbO, whereas 19th–20th Dynasty glass from Dahshur has an average of 0.04%PbO strongly suggests that yellow glass (coloured by lead antimonate) entered the system during recycling. The distribution (Figure 9) shows that most of the 18th

**FIGURE 8** Frequency histograms of the cobalt oxide (CoO %) in cobalt-blue glass from Egypt in the 18th and 19th–20th Dynasties Notes: Data from Abe et al. (2012). The upper plot exhibits a bimodal distribution with the higher peak having about twice the concentration of CoO as the lower peak from the 18th Dynasty objects. The 19th-20th Dynasty cobalt glass has similar levels of CoO to the lower peak from the 18th Dynasty cobalt glass.
Dynasty glass has low levels of PbO, which is potentially the trace levels in the original cobalt-blue glass prior to any recycling (the peak in Figure 9 is about 0.01% PbO). Some 18th Dynasty glass has elevated levels of PbO, providing further evidence that recycling (and inadvertent mixing of glasses of different colours) occurred during this period. The 19th–20th Dynasty glass has a much higher proportion of samples with higher levels of lead than the 18th Dynasty glass, suggesting that more of this glass has been contaminated with lead as a consequence of recycling.

Similarly, Sb$_2$O$_3$ was found to exhibit distributions with high concentrations over a broad range for both chronologies, which is potentially because contamination from this component could have derived from white opaque glass (calcium antimonate) as well as yellow glass (lead antimonate) during the recycling process.

The broad distributions of PbO and Sb$_2$O$_3$ (Figure 9) appear to indicate unintentional mixing (due to the failure to separate all the white and yellow glass from cobalt-blue glass during recycling), whereas the bimodal distribution for cobalt oxide (Figure 8) reflects the deliberate dilution of cobalt-blue glass with a base glass during the recycling process. This mechanism would also suggest that certain Egyptian and Mycenaean blue glasses, which have both copper and cobalt as colourants, are not deliberate mixtures to produce specific shades of blue or due to compositional variability in the geology of the colourant source (Smirnou & Rehren, 2013) but rather the accidental mixing of different types of blue glass during recycling.

Essentially, the presence of the broad distributions for levels of PbO and Sb$_2$O$_3$ in cobalt-blue glass supports that the later 19th–20th Dynasty glass has lower concentrations of cobalt oxide because of recycling and dilution (and its associated accidental introduction of
contaminants) rather than a deliberate reduction in the amounts of cobalt added to new glass in this later period. This suggests that recycling became necessary in New Kingdom Egypt and that the change in composition of cobalt-blue glass was not because a new source of cobalt was exploited in later periods (Abe et al., 2012) but because cobalt sources were no longer available to provide the colour cobalt blue. In effect, if a source was no longer available, but cobalt glass continued to be used, and a new source was not exploited, then recycling must have occurred. This would question conventional wisdom concerning the accepted sources of cobalt for the New Kingdom period, that is the Kharga and Dakhla Oases of Egypt’s Western desert (e.g. Kaczmarczyk, 1986; Rehren, 2001; Shortland, 2000) and raise the possibility that less local sources were used (e.g. Lucas, 1962; Dayton, 1981, 1993; Stern & Schlick-Nolte, 1994; Lee & Quirke, 2000:111; Wood & Hsu, 2019), which would have been more susceptible to interruptions due to changes in the political landscape affecting supply chains.

CONCLUDING REMARKS

There is no one-size-fits-all approach to investigate ancient recycling when re-melting is involved. There are methods, however, that align with the data available that allow hypotheses to be examined and inferences to be made. The three approaches presented here identify recycling from examining artefacts in the archaeological record. The presence of Roman glass applied as a glaze on Parthian pottery clearly indicates that glass, probably discarded after being broken, was recycled by Parthian potters for this purpose. It could also suggest that the paucity of Parthian and Sasanian glass in the archaeological record could reflect an organised recycling economy. Similarly, recycling accompanied with dilution of cobalt-blue glass with a base glass hints at the possibility that glass becomes less conspicuous in late New Kingdom Egypt because cobalt was no longer available. This raises questions regarding the provenance of this valued colourant. Finally, for the Iron Age silver hoards of the southern Levant, it appears that recycled silver was common. In fact, mixing lines emerging from combining isotopic and compositional data not only shed light on when certain sources of silver became available in the southern Levant but may also identify silver melted down hastily in times of unrest without first removing gold embellishments.

Understanding the materials from which artefacts have been manufactured can have a significant influence on how archaeological contexts are understood. For materials that can be recycled and reused through melting, such as metals and glass, the analysis of scientific data may provide insight well beyond elemental composition, including the evolution of raw material availability and supply chains over time, the process of manufacture, and how these might relate to external economic and political circumstances. It is therefore imperative that recycled material can be identified with some certainty in the archaeological record.

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DATA AVAILABILITY STATEMENT

The data that supports the findings of this study are available in the supplementary material of this article.

ORCID

Jonathan R. Wood https://orcid.org/0000-0001-6630-6916
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