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Metal provenance of Iron Age *Hacksilber* hoards in the southern Levant

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ABSTRACT

Hacksilber facilitated trade and transactions from the beginning of the second millennium BCE until the late fourth century BCE in the southern Levant. Here we demonstrate the use of new, data-driven statistical approaches to interpret high-precision Pb isotope analysis of silver found in archaeological contexts for provenance determination. We sampled 45 pieces of *Hacksilber* from five hoards (Megiddo Area H, Eshtemoa, Tel Dor, ʿEn Gedi, and Tel Miqne-Ekron) and combined our data with recent literature data for the same hoards plus five more (Beth Shean, Ashkelon, Tell Keisan, Tel ʿAkko, and ʿEn Ḥofez) thus covering silver from the Late Bronze Age III (c.1200 BCE) to the end of the Iron Age IIC (586 BCE).

Samples were taken by applying a new minimally destructive sampling technique. Lead was extracted using anion-exchange chromatography, and Pb isotopic compositions were measured by MC-ICP-MS. Data were treated using a new clustering method to identify statistically distinct groups of data, and a convex hull was applied to identify and constrain ore sources consistent with the isotopic signature of each group. Samples were grouped by minimizing variance *within* isotopic clusters and maximizing variance *between* isotopic clusters.

We found that exchanges between the Levant and the Aegean world continued at least intermittently from the Late Bronze Age through to the Iron Age III, demonstrated by the prevalence of Lavrion (Attica), Macedonia, Thrace (northern Greece), southern Gaul (southern France), and Sardinia as long-lived major silver sources. Occasional exchanges with other west Mediterranean localities found in the isotopic record demonstrate that even though the Aegean world dominated silver supply during the Iron Age, exchanges between the eastern and the western Mediterranean did not altogether cease. The mixture of silver sources within hoards and relatively low purity of silver intentionally mixed with copper and arsenic suggest long-term hoarding and irregular, limited supply during the Iron Age I.

1. Introduction

Hacksilber is irregularly cut silver bullion made from broken pieces of silver ingots, jewelry, and other pieces of scrap silver. It served as a store of wealth and means of payment in the Ancient Near East for thousands of years. Used in local and international transactions, the value of *Hacksilber* was determined by weighing it on scales against standardized weights presumably allowing for the purity of silver. Evidence exists of cupellation being used for silver extraction since the fourth millennium BCE at sites in Greater Mesopotamia (Helwing, 2014), and *Hacksilber* itself has been discovered in archaeological excavations in the region (for Middle Bronze Age II *Hacksilber* hoards, see Kletter, 2003:148; Thompson, 2009 for Early Iron Age I hoards). In the southern Levant, *Hacksilber* facilitated trade and transactions from the beginning of the second millennium BCE (Kletter, 2003:148) until the late fourth century BCE (Farhi, 2014), with mixed hoards composed mainly of uncut coins supplemented by *Hacksilber* in equal or greater quantities (Gitler, 2006). A hoard allegedly found in the vicinity of Samaria illustrates that the practice of using cut coins alongside *Hacksilber* continued into the second half of the fourth century BCE (Gitler, 2006).

Hacksilber hoards have been found in excavations throughout the southern Levant in:

- Ceramic containers – e.g. ʿEn Gedi (Kletter and Degroot, 2007), Eshtemoa (Kletter and Brand, 1998), ʿEn Ḥofez (Thompson and Skaggs, 2013), Tel ʿAkko (Thompson and Skaggs, 2013), and Tel Miqne-Ekron (Golani and Sass, 1998).

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- Bundles wrapped in linen bags and kept in ceramic containers – e.g. Megiddo Area H (Arie et al., 2019), Tell Keisan (Thompson and Skaggs, 2013), and Tel Dor (Shalev et al., 2014).
- Bundles wrapped in linen bags – e.g. Tel Beth-Shean (Thompson, 2009), Ashkelon (Thompson, 2020), and Tel Miqne-Ekron (Golani and Sass, 1998).
- Assemblages where no remains of the container or the textile bag have been found – e.g. Tel Miqne-Ekron (Golani and Sass, 1998) and Beth Shean (Thompson, 2009).

Recent research into *Hacksilber* has mostly concentrated on two questions. First, the extent to which *Hacksilber* filled the functions of money prior to the invention of coinage (Stos-Gale, 2001; Thompson, 2003; Eshel et al., 2018). Second, identifying *Hacksilber*'s silver sources with a focus on determining when and to what extent the Phoenicians were engaged in long-distance silver trade prior to their colonization of the western Mediterranean (Thompson, 2003; Thompson and Skaggs, 2013; Murillo-Barroso et al., 2016; Eshel et al., 2019, 2021; Wood et al., 2019; Gitler and Tal, 2020).

Arguably, the Hebrew phrase *bẓʿ ksp*, meaning “intentionally cut pieces of silver”, was the biblical term for money itself (cf. Judges 5:19). The bundling of *Hacksilber* evoking the biblical expression “pouch of silver” (*ẓrwr ksp*, Genesis 42:35) has led to the contention that it functioned as money (Thompson (2003) or at least as pre-coinage (Gitin and Golani, 2001; Silver, 2006; Kroll, 2008; Gestoso-Singer, 2013, 2015; Heymans, 2018a). Eshel et al. (2018; 2019) addressed this by investigating the context, chronology, weight, and typology of *Hacksilber*, and comparing this information with a subset of major and minor elemental compositions. They found that neither the weight nor the fineness, that is the quantity and quality of *Hacksilber*, are sufficiently consistent to support the theory that *Hacksilber* was a precursor to coinage. Rather, it would appear that the Phoenician perspective on silver before the 5th century BCE was to treat it as a commodity, not a currency (Gitler and Tal, 2006; Elayi and Elayi, 2009; Vanalfen, 2002; Albarède et al., 2020). This is an important finding which matches Davis' (2012) conclusions about contemporary Archaic Athenian Greek use of *Hacksilber* and goes to the question of whether there was an adequate and reliable supply of silver to make true monetary use viable. If there was, then the expectation would be mostly homogeneous hoards, especially within the bundles, a question tested in this research. Albarède et al. (2021) further investigated the apparent resistance to the adoption of money in the western Mediterranean, tracing silver refining techniques in the region using lead and silver isotope analysis.

Eshel et al. (2021) raise the point of whether systematic debasement by copper and arsenic during the period 1200-950 BCE in the southern Levant was intentional or just common economic practice in a region that could only acquire silver from abroad. They argue that the use of arsenic (up to 4.6 wt% in Megiddo Area H) to maintain a silvery shine indicated deliberate debasement. However, there is no evidence to support Eshel et al.'s statement that “the local administrations initiated sophisticated devaluation methods to compensate for the lack of silver”, and full-scale tampering with all silver circulating in the southern Levant in its various forms is unlikely. An equally reasonable assumption is that relatively low-quality silver alloys were broadly accepted during the Late Bronze to Iron Age transition period. Understanding silver sources and thus trade routes in use at the time will help in better understanding this phenomenon.

The focus of the present research is on Iron Age *Hacksilber* hoards in the southern Levant when the geopolitical circumstances were different from the preceding Late Bronze Age. Material found in controlled archaeologically excavated hoards and single finds provides the best evidence because it has secure contexts. Research questions for this study to be addressed by Pb isotopic analysis are:

1. How many silver sources were there, and which ones can be identified?
2. Do the silver sources change with time?
3. To what extent is the silver in each hoard internally homogenous?

The hoards described in this work have been dated based on their archaeological context, and a generally agreed ceramic pottery typology (Mazar, 2011). For a description of the methodology behind the dating of each hoard, see Eshel et al. (2018; 2021). For details of the archaeological context and relative dating for individual sites, see Rowe (1940) (Beth Shean), Arie et al. (2019) (Megiddo Area H), Balmuth (2001 p.15) (Ashkelon), Nodet (1980 pp. 323–326) (Tell Keisan), Stern (1998) and Shalev et al. (2014) (Tel Dor), Thompson and Skaggs (2013) (Tel ʿAkko), Alexandre (1997) p. 53) (ʿEn Ḥofez), Kletter and Brand (1998) (Eshtemoa), Golani and Sass (1998) and Gitin and Golani (2001) (Tel Miqne-Ekron), and Kletter and Degroot (2007) (ʿEn Gedi). For a broader understanding of the hoards in relation to each other, see Thompson (2003) and Eshel et al. (2018). The Bronze Age and Iron Age are convenient constructs understood to start and end at different times in different places. In the southern Levant, the Bronze Age is considered to end around 1200 BCE with the cultural collapse of the dominant states, and the Iron Age ends in 586 BCE marked by the Babylonian conquest and destruction of the temple in Jerusalem. Within these two periods are separations into shorter time phases (e.g. Iron Age I, Iron Age II), which can be further divided (e.g. Iron Age IIA, Iron Age IIB, Iron Age IIC). The dates associated with each division remain intensely debated (e.g. Gilboa et al., 2008; Finkelstein and Piasetzky, 2011; Mazar, 2011), with ranges for the transition between each period being accepted within ‘low’ and ‘high’ chronologies. The time periods are being tightened as further data are gathered in archaeological excavations, especially by radiometric dating (Webster, 2015), but the exact dating does not affect the arguments presented in this paper.

Lead isotope analysis of *Hacksilber* hoards has been conducted before, most notably through the ‘*Hacksilber* Project’ undertaken by Balmuth and Thompson with Stos-Gale (Stos-Gale, 2001; Stos-Gale and Gale, 2012; whose data are often referred to as ’legacy’ data). These data were produced by thermal-ionization mass spectrometry (TIMS), which does not control analytical mass bias as efficiently as multiple-collector inductively-coupled plasma mass spectrometry (MC-ICP-MS). The ‘*Hacksilber* Project’ publication included a chapter on lead isotope analysis of excavated *Hacksilber* at Tel Miqne-Ekron, Shechem, and Selinus (Stos-Gale, 2001). When interpreting results from lead isotope analysis in these *Hacksilber* hoards, Stos-Gale observed that they are consistent with lead isotope ratios of ores from the Aegean, Spain, and Iran. A further observation made by Stos-Gale was that the data did not support a hypothesis of widespread mixing, positing that melting and re-casting was done on individual objects on a small scale. Stos-Gale pointed out that a silver object of unknown provenance can only be assigned an ore source if data from that ore source exist and called for further lead isotope analyses of *Hacksilber*. We add in the same vein that further Pb isotope analyses of ores likewise are warranted.

Stos-Gale's conclusions were re-examined by Thompson and Skaggs (2013) for hoards from Tel ʿAkko, Tel Dor, ʿEn Ḥofez, and Tell Keisan against geological data from the Forum of the European Geological Surveys (FOREGS) Geochemical Baseline Mapping program. Thompson and Skaggs concluded that at least one sample from each of the *Hacksilber* hoards under consideration could have lead isotope ratios consistent with those of silver-bearing ores from Sardinia and southern France and inconsistent with Aegean and Anatolian sources. Apparent in this paper and elaborated on in Thompson's unpublished “A Brief History of *Hacksilber* Project Research” (2017), is the criticism of previous assumptions that *Hacksilber* was unlikely to originate in the western Mediterranean. Further, with regards to the problem of mixing, Thompson (2017) asserted that even before 800 BCE silver sources were multiple and bullion travelled long distances.

Eshel et al. (2019) analysed numerous pieces of *Hacksilber* from the same hoards analysed by Stos-Gale (2001) for lead isotope composition, comparing their results to lead isotope compositions of silver-bearing ores available in the Oxford Archaeological Lead Isotope Database (OXALID). The study focused on the hoards from Tel Dor, Tel ʿAkko, ʿEn Ḥofez, and Eshtemoa. The authors were critical of Thompson and Skaggs (2013) for assuming that *Hacksilber* hoards were not mixed. They suggested that *Hacksilber* from Tel Dor and Tel ʿAkko originated from both Anatolia and Sardinia, while *Hacksilber* from ʿEn Ḥofez and Eshtemoa originated from Iberia (Spain). Based on the chronology of these hoards, they proposed that the Phoenicians brought knowledge of silver refining acquired in Anatolia to tribes in Iberia in the mid-10th century BCE, 150–200 years prior to Phoenician settlement in Iberia. However, while the data may be consistent with the conclusions, they do not necessarily go so far as to support them.

Wood et al. (2019) used lead isotope analysis to further investigate whether *Hacksilber* silver sources were mixed. They re-examined the lead isotope ratios in *Hacksilber* produced by Stos-Gale (2001) against compositional data. Wood et al. (2019) proposed a comparison of the crustal age of the metal calculated from the lead isotope ratios against the compositional ratio of gold to silver. Where the gold to silver ratio did not match the crustal age, Wood et al. (2019) proposed that mixing of silver from different sources is evident. In this way, mixing lines were identified, and the authors proposed considerable mixing between several potential ore sources. The authors used the chronology of the hoards, identified via their archaeological contexts, to suggest where and when mixing of silver may have taken place. The study focused on the use of mixing lines to identify mixing events, with only some of the end-member clusters identified in terms of specific geological sources. Crucially, the authors noted that further work is needed to contextualize their findings, acknowledging that apart from the archaeological chronology, there is little evidence to support where or when potential mixing took place. The authors applied their proposed methodology in Wood et al. (2020) by reanalysing legacy lead isotope data from Stos-Gale (2001) in order to determine the source(s) of the Tel Dor *Hacksilber* hoard. The authors concluded that the Phoenicians learned silver refining in Cyprus, rather than Anatolia as Eshel et al. (2019) had suggested, and introduced the methodology to Iberia. The authors further suggested that silver objects which were previously identified as having originated from Thera, Kythnos, or Cyprus (Wood et al., 2019) are in fact silver mined at Kalavasos in Cyprus, or lead used for cupellation of silver from a different, so-far unidentified source. These conclusions are unsupported. The geochemistry of ores on the islands of Kythnos and Thera is inconsistent with significant silver production. As to the silver extraction from the ores in Kalavasos, one of the copper mines of Cyprus, this speculation is also unfounded. Even though intrusions of porphyries are ubiquitous from the Aegean to modern Afghanistan and contain substantial traces of silver (Zürcher et al., 2019), the historical context shows that silver from these ores could never compete with peri-Aegean mines. A simple interpretation is technological: Cu and Ag form a eutectic barrier at 28% Cu (Baker, 1992) making silver purification by cupellation of Cu-rich alloys a major hurdle. This problem does not exist with Ag–Pb alloys. In a more geological context, finding Ag and Pb ores in Cyprus, a textbook example of an ophiolite, would be highly unexpected.

Eshel et al. (2019; 2021) conducted further research on *Hacksilber* using lead isotope analysis, with the premise that interpretations based on lead isotopes can be further refined using elemental compositions. The elements they considered diagnostic are Cu (<5.5%), Pb, Au, and Bi, which may be indicative of silver that has been cupelled, and Co, Ni, Zn, and Sn, the *absence* of which likewise is indicative of silver that has been cupelled but the *presence* of which is consistent with the addition of smelted copper. A caveat, however, is that some elements (Zn, Bi, As) (Honig and Kramer, 1969) are more volatile than silver and, according to Henry's Law, should be at least partially lost during the metallurgical process. Arsenic and antimony were also considered carefully, as they may have been added intentionally to the alloy, and not introduced via the addition of copper. All compositional data were collected using quadrupole ICP-MS. Rather than using compositional information for provenance determination, this information was directed towards identifying mixing and alloying. When various mixes or alloys were identified, the results were placed within their specific archaeological context (particularly hoard location and approximate date of deposition) to draw conclusions about the broader political situation at the time. Their key claim was that high copper contents in *Hacksilber* date from around the Bronze Age collapse (c.1200 BCE) correlated with a purported end to ready access to Aegean silver in the Levant due to breakdown of trade connections. They proposed that a perceived influx of silver from Anatolia and the west Mediterranean (based on lead isotope results in Eshel et al., 2019) is reflected in considerably lower copper contents in *Hacksilber* deposited after approximately 950 BCE.

As discussed in the literature, a geological perspective is crucial to understanding lead isotope analysis of archaeological objects (Albarède et al., 2012; Blichert-Toft et al., 2016; Albarède et al., 2020). The advantage of using the geological perspective approach, which involves calculating Pb model ages and time-integrated parent-daughter (U/Pb) and parent-parent (Th/U) ratios from measured Pb isotope data, is that it is data-driven and is performed independently of any archaeological or numismatic conclusions drawn about the samples. The former set of evidence thereby complements the latter two in an objective manner. As a caveat, mixing trends are linear only if the x and y axis denominators are identical, as in Pb–Pb isotopic plots. Lead model ages are, to a first approximation, proportional to $^{207}\text{Pb}/^{206}\text{Pb}$ (Albarède et al., 2021) and mixing lines using Pb model ages vs elemental ratios such as Au/Ag (e.g. Figs. 4 and 6 in Wood et al., 2019) are strongly curved as a function of the Pb contents of the coins. Mixing patterns, therefore, should be interpreted with the utmost caution.

Prior attemptsat provenance -assignments have relied on one-to-one comparisons between individual ores and *Hacksilber* fragments (Thompson and Skaggs, 2013; Delile, 2014; Westner et al., 2020). Our approach is different. We provide new high-precision Pb isotope compositions of 45 *Hacksilber* fragments from seven southern Levantine hoards and combine them with the data from Eshel et al. (2019). Eshel et al.'s data were all obtained by MC-ICP-MS, as were the new data of this study, hence justifying the merging of the two data sets. We further apply a new, more encompassing approach to statistically interpret the data, based on 'convex hulls' of the 3-dimensional Pb isotope data of each hoard (i.e., the smallest convex volume that contains the data). By using a data-driven approach to the statistical interpretation of the lead isotope compositions of the present *Hacksilber* samples, combined with lead isotope data for likely ore sources, we identify which ore sources could have contributed to the *Hacksilber* hoards and determine the homogeneity of silver sources for each hoard. By then combining these data with the archaeological chronology of the hoards in question, we identify changes in silver sources over time.

2. Materials

Thirty-four *Hacksilber* hoards dating to the Iron Age (c.1200-600 BCE) from 15 different sites in Israel and under the control of the Palestinian Authority were identified by Thompson as part of the '*Hacksilber* Project' (2017). The 13 sites listed in Table 1 were selected for the present research project to provide representative samples from the end of the Late Bronze Age through the Iron Age, from different find-spots in the southern Levant which can best answer the research questions identified in this paper. There is a strong focus on the rich material from the seventh century sites of Tel Miqne-Ekron, with its multiple hoards, and ʿEn Gedi, as this was the century when coined money was invented and monetary use of *Hacksilber* might be expected.

Pre-MC-ICP-MS legacy data are dominated by more 'noise', and therefore less precision, than data from the MC-ICP-MS era (post-2000). Precision and accuracy of early (pre-2000) TIMS (which generally did not include double- or triple-spike data) and modern MC-ICP-MS Pb isotopic data may differ by up to two orders of magnitude. due to the poorer control of instrumental mass fractionation by TIMS than by MC-

Table 1
Hoards selected for analysis (for the description and dating of these hoards see Heymans, 2018b, pp. 256–269).

Hoard	Period	Approximate date (BCE)	Contents	Sampled for new lead isotope analysis	Previous lead isotope analysis available in the literature
Beth Shean 10704 (L.88866)	Late Bronze Age III	1200–1150	Cut ingots, wires, broken jewellery stored in a bundle	0	13 (Eshel et al., 2021)
Megiddo Area H	Iron Age I	1070	3 linen-wrapped bundles of *Hacksilber* - cut ingots, jewellery	5	13 (Eshel et al., 2021)
Beth Shean 1095	Iron Age I	1150–950	Ingots, wires and jewellery stored in a bundle	0	5 (Eshel et al., 2021)
Megiddo 2012	Iron Age I	1050–950	Cut ingots, sheet fragments, and jewelry stored in three bundles	0	11 (Eshel et al., 2021)
Ashkelon	Iron Age I	1050–950	2 linen-wrapped bundles of *Hacksilber*	0	12 (Eshel et al., 2021)
Tell Keisan	Iron Age I	1050–950	Silver fragments including sheet silver + jewellery stored in a single jar	0	20 (Eshel et al., 2021)
Tel Dor	Iron Age IIA	2nd half of 10th century	17 linen-wrapped bundles of *Hacksilber* stored in a single jar + ingots	14	34 (Eshel et al., 2019)
Tel ʻAkko	Iron Age IIA	10th-9th centuries	Cut ingots, tokens, wires, sheets stored in a juglet	0	12 (Eshel et al., 2019)
Beth Shean 1029a and b	Iron Age IIA	950–800	Jewellery and ingots store in a ceramic vessel	0	15 (Eshel et al., 2021)
ʿEn Ḥofez	Iron Age IIA	9th century	*Hacksilber*, ingots + some jewellery stored in 3 jugs	0	29 (Eshel et al., 2019)
Eshtemoa	Iron Age IIB	8th century	*Hacksilber*, ingots + jewellery stored in five jugs	5	0
Tel Miqne-Ekron	Iron Age IIC	Late 7th-early 6th century	*Hacksilber*, ingots, jewellery + a foundation deposit. Hoards 1, 3 and 5 were stored in a jug, Hoard 2 shows traces of textile impressions, Hoard 4 was found as an assemblage and Hoard 6 was covered by an overturned bowl.	16	0
ʿEn Gedi	Iron Age IIC	Late 7th-early 6th century	62 pieces of silver, mostly ingots stored in a cooking pot	5	0
Total				**45**	**164**[a]

[a] Not including OXALID data.

ICP-MS. Furthermore, the archaeological literature tends to reference Pb isotope data relative to ^{204}Pb (e.g. Eshel et al., 2018; 2019; 2021), which is the least abundant isotope rather than, for example, ^{206}Pb or ^{208}Pb (Albarède et al., 2012, 2020). The measurement precision of ^{204}Pb is always several times lower than the measurement precision of the more abundant and Hg-interference-free isotopes ^{206}Pb, ^{207}Pb, and ^{208}Pb, and as a result the ^{206}Pb/^{204}Pb, ^{207}Pb/^{204}Pb, and ^{208}Pb/^{204}Pb ratios are strongly correlated. When lead isotope data are examined using ^{204}Pb as the denominator on both the x and y axes, 'noise' is thus greater and, hence, precision is poorer. Interpreting geological provenance from data represented in this way, therefore, is not ideal.

In order to avoid merging data of heterogeneous quality in the same database, ^{204}Pb-referenced legacy data as well as Pb isotope data measured by TIMS, mostly from the OXALID database (which includes *Hacksilber* data published in Stos-Gale, 2001), have not been included in this study. This choice does not imply that the inclusion of these data would have modified the conclusions reached here but was made to limit the present assessment to high-precision data only. The merging of our data with those of Eshel et al. (2019; 2021) is justified by their similar level of precision acquired via solution purification chemistry and MC-ICP-MS measurement.

3. Methods

3.1. Artefact sampling

All samples were etched on their edges using a novel technique (Milot et al., 2021b) which removes only a few micrograms of material, less than one-millionth the total weight of the object (or less depending on the size of the object in question). To briefly summarize the technique, the artefact is rolled for about 90 s onto a strip of chromatographic paper impregnated with a solution of H_2O_2, NH_4OH, and H_2O in the proportions of 1:1:1 using a custom-made set of felt-covered pliers (Milot et al., 2021b). In cases where the shape of the object did not permit rolling, such as is often the case with unevenly shaped *Hacksilber*, a cotton bud was used instead of the pliers and chromatographic paper. The strips and cotton buds were air-dried under an IR lamp and placed into clean 10 mL centrifuge tubes, which were closed tightly with screw lids and wrapped in multiple layers of film for transportation back to the Lyon clean laboratory for Pb separation and isotopic analysis (as artefacts were usually sampled on-site in a distant museum). There, 10 mL double-distilled 1 M HBr, for which Pb has strong affinity, were added to the tubes, thereby submerging the strips (or cotton buds) which were subsequently left to leach at ambient temperature for 24 h. The HBr containing the Pb leached from the strips (or cotton buds) was then transferred to a clean Savillex (PFA) beaker and evaporated to dryness on a hotplate at approximately 130 °C. Meanwhile, the HBr-leached strips (or cotton buds) were submerged in 10 mL distilled 0.5 M HNO_3 and left to leach further at ambient temperature for another 24 h. The HNO_3 was transferred to the same beakers containing the now dried-down HBr solutions and evaporated to dryness under the same conditions.

3.2. Lead purification

Lead was eluted from the samples following a one-step anion-exchange (AG1-X8, 100–200 mesh) column chromatography protocol. The dry residues from the leaching procedure were dissolved in 1 mL double-distilled 1 M HBr, and alternately heated at 110 °C on a hotplate and placed in an ultrasonic bath to ensure total dissolution. The cooled-down samples were loaded onto the anion-exchange columns, the sample matrix eluted with 1 M double-distilled HBr, and Pb subsequently collected with 6 M distilled HCl. The Pb fractions were evaporated to dryness at approximately 110 °C, redissolved with a few drops of concentrated distilled HNO_3 to remove any traces of HBr and organic material, dried down again and finally dissolved in 1 mL distilled 0.05 M HNO_3 with 5 ppb Tl prior to Pb isotope analysis.

3.3. Lead isotope analysis

Lead isotope analysis was done using a Nu Plasma 500 HR MC-ICP-MS at the Ecole Normale Supérieure de Lyon. Blichert-Toft et al.

(2003) and White et al. (2000) describe in detail the analytical method of Pb isotope analysis by MC-ICP-MS. Instrumental mass bias was corrected with added Tl using the reference values for Tl and Pb of Eisele et al. (2003). Sample-standard bracketing relative to the NIST 981 Pb standard, which was run systematically every two samples throughout each analytical session, further ensured the accuracy of the measured Pb isotope compositions of the unknowns. The repeated measurements of NIST 981 yielded an external reproducibility of 30–100 ppm (0.003–0.01%) for ^{204}Pb-normalized ratios and of 6 ppm (0.0006%) for ^{207}Pb/^{206}Pb and ^{208}Pb/^{206}Pb. Estimation of internal analytical uncertainties was based on the measurement of 60 isotope ratios with 10 s integration time for each sample and standard expressed as twice the standard deviation (2SD). The Pb isotope data of the 45 *Hacksilber* samples analysed in this study are listed in Supplementary Table S1.

3.4. Parameter calculation and statistical analysis

Traditionally in archaeometric research, lead isotopes are used for provenance determination based on the interpretation of plots of the raw isotopic data. In this study, we additionally take the geologically-informed approach proposed by Albarède et al. (2020) and further developed in Albarède et al. (2020) to explore Pb model ages T_{mod} and the ^{238}U/^{204}Pb (μ) and ^{232}Th/^{238}U (κ) parameters of each sample.

3.5. Provenance assessment

In previous studies (e.g. Delile, 2014; Westner et al., 2020), the ‘consistencies’ (Thompson and Skaggs, 2013) are established by testing the error-weighted or unweighted distance between a particular ore and a particular archaeological sample in one of the 3-dimensional spaces of Pb isotopes (with isotopic ratios preferentially normalized to ^{206}Pb to avoid the strong correlations arising from ^{204}Pb-normalized ratios; see Albarède et al., 2020). The pair-wise approach ignores the fact that ore data tend to form regionally coherent populations, which the geologically-informed parameters (model age T_{mod}, μ, and κ) clearly confirm (Blichert-Toft et al., 2016; Milot et al., 2021a). These groups must, in one way or another, manifest themselves in the *Hacksilber* Pb isotope record. Here we adopted the alternative strategy of detecting the ores that are consistent with the range of Pb isotopic ratios defined by the samples. This led us to introduce the concept of the ‘sample convex hull’, which is the smallest volume in the 3-dimensional space of Pb isotopic ratios that contains all the sample values. As a preliminary step, we used the standard clustering technique applied to Iberian galenas by Albarède et al. (2020) with the expectation that distinct isotopic groups would be found. The next step consisted in separating the data within each hoard into distinct groups, typically one to three as a function of the number of samples. At that point, a convex volume circumscribing the points of the group, known as a convex hull and meshed with tetrahedra, is calculated by Delaunay triangulation (e.g. Aurenhammer et al., 2013) (Fig. 1). Convexity is required to ensure that points do not lie in gulfs or embayments. Finally, an algorithm was run for each datum of the ore database to determine if the corresponding point lay inside or outside the volume. Minor improvements were added: the hoard data were scaled to unit variance and zero mean, and some slack around the volume was allowed for. The successful data finally were plotted on a map of the circum-Mediterranean regions. The necessary algorithms were all implemented with the MATLAB software.

The convexity of the hull guarantees that if extreme points lie in the hull, so will all the points that are lying on the mixing lines between the extremes. The problem is that if a single sample with a completely different isotopic composition (i.e., an outlier) is included in the sample set, the hull will include the empty volume between the outlier and the rest of the data. We found that a preliminary identification of distinct groups, as explained above, avoids including data-intermediate free volumes in the search. As will be shown below, all the hoards include one or two groups. It must be remembered that the extreme points are still included and should show up in the search.

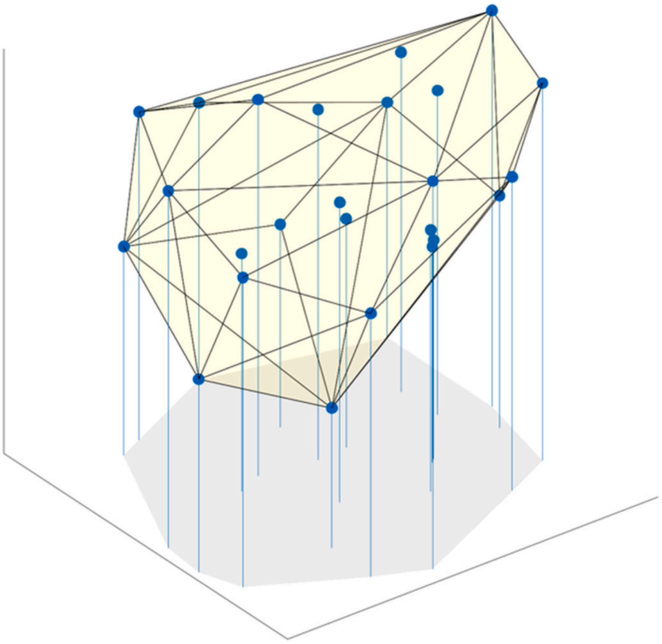

Fig. 1. Sketch of a convex hull (in yellow) built by Delaunay triangulation. The original data (blue circles) were generated as a set of 25 random points in 3-dimensional space. In practice, the convex hull is the 3D convex volume that contains all the isotopic coordinates of the samples in each hoard. The Delaunay triangles have been made transparent to show the points inside the hull. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

4. Results

We first explored the space of the 209 Pb isotope data (including those produced by Eshel et al. (2019; 2021) and the 45 Pb isotope data produced in the present work), from the 13 different *Hacksilber* hoards listed in Table 1. We proceeded hoard by hoard by:

(1) identifying significantly different isotopic groups in each hoard,
(2) constructing the corresponding convex hulls for each of them by Delaunay triangulation,
(3) searching the ore database for the points lying within each hull, and
(4) identifying the location of potential sources.

4.1. Constructing the convex hulls

Convex hulls were obtained for each hoard by Delaunay triangulation. By maximizing the smallest angle of all the triangles, Delaunay triangulation makes fewer elongated triangles. In order to avoid enclosing large unpopulated (‘empty’) volumes, a rough cluster analysis was run for each hoard defining a maximum of three groups (in practice one or two) in the conventional space of ^{204}Pb-normalized ratios. Choosing a different normalization isotope does not affect the definition of the clusters. Fig. 2 shows how different the hulls are from hoard to hoard.

A remarkable property of this technique is that, since the hull is convex, all the data corresponding to intermediate mixtures of different isotopic components plot within the hull defined by the data corresponding to the extreme mixtures. Let us assume a series of samples resulting from mixing components A and B, and label x_1, x_2, x_3 three of these mixtures with proportions of component B increasing in that order.

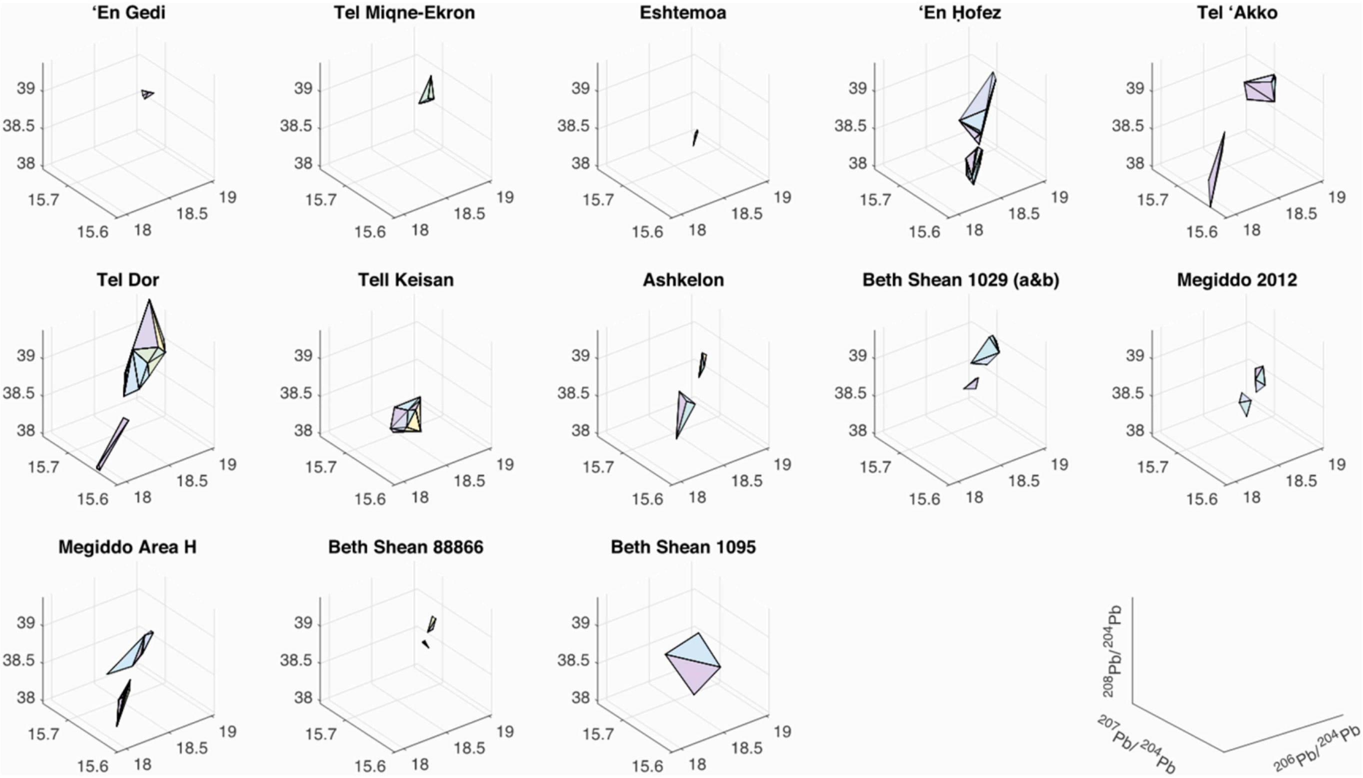

Fig. 2. Scaled convex hulls obtained by Delaunay triangulation in the space of ^{204}Pb-normalized ratios. The data were first standardized to zero mean and unit variance and cluster analysis allowing for up to three separate groups to help define tighter hulls.

If samples x_1 and x_3 plot in the convex hull, so will sample x_2. A potential risk is that a mixture of components A and B may end up fortuitously producing a mix similar to an unrelated component C, thereby leading to erroneous provenance assignment. Our experience with the available data, however, is that whereas the risk of such fortuitous coincidence in two-dimensional plots does exist, it essentially disappears in three-dimensional space, or at the very least, concerns only isolated hits. We found that 'hot spots' with multiple hits are robust against inadvertent coincidence and that the robustness may be conveniently illustrated by frequency peaks of model ages.

4.2. Permissible silver sources

Once the hulls were defined, each sample from our ore database, which includes about 7000 samples of galena (Milot et al., 2021a), was tested for being inside or outside the hulls defined for each hoard. Although this database includes many references from the OXALID database, only Pb sulphides were retained and samples such as slags were carefully and systematically eliminated. A tolerance of 2% was allowed for on the standardized data, which is equivalent to considering the hull boundary as fuzzy. In-hull ores with Pb isotope compositions falling within the hull of each hoard are considered 'hits.' The results are displayed for each hoard in Fig. 3. Each hit should not be considered as decisive, but only as a permissible source. Given the large number of ore samples in the database, a handful of geographically consistent hits may not be significant (e.g. Eshtemoa). Hits in unexpected localities, such as modern United Kingdom or modern Tunisia can be disregarded on two grounds: a very small number of hits or lack of archaeological evidence for mining. In contrast, ores that are no-hit can be excluded with a high degree of confidence.

The most common hot spots are from Lavrion, Thrace, Macedonia, Sardinia, and the Cévennes (southern Gaul) (Table 2). Fewer hits are found for some hoards in northern Sardinia, Illyria, the Betics, the Ossa-Morena Zone (southern Iberia), and Taurus. Only one hit was found for Eshtemoa, which we considered insufficient for source assignment.

5. Discussion

Eshel et al. (2021) argue that the Pb isotope data of the Levant can be accounted for by mixing different end-members and we have tested this model here. We normalized the data to ^{206}Pb (^{204}Pb/^{206}Pb, ^{207}Pb/^{206}Pb, and ^{208}Pb/^{206}Pb) to minimize error correlations and then ran a principal component analysis both globally and hoard by hoard. Table 3 shows that, with the exception of Tel Miqne-Ekron, the third principal component accounts for only a negligible fraction of the variance in each hoard. Two principal components (i.e., three end-members, but often only two) therefore adequately account for the observed isotopic variability, which in general is consistent with the number of modes in the histograms of Fig. 3, and agrees with the conclusions of Eshel et al. (2021).

A concern raised by Eshel et al. (2021) is that Pb isotopes in *Hacksilber* may have been drastically altered by copper-based debasement. A puzzle in this context is the contrast between the well-defined mixing arrays observed by these authors for some hoards (Tell Keisan, Megiddo Area H) in Pb isotope diagrams and the lack of mixing hyperbolae observed in the ^{206}Pb/^{204}Pb vs Cu diagram (Eshel et al., 2021, Fig. 8). The key to solving this puzzle is the limited solubility of Pb in Cu (a few hundred ppm, Vaajamo et al., 2013). In contrast, Pb solubility in Ag is substantial (up to a few percent, Karakaya and Thompson, 1987). It therefore takes up to several tens of percent of Cu addition to silver to alter the original Pb isotope composition. This is the case for the Tell Keisan and Megiddo Area H hoards. The mixing lines observed by Eshel et al. (2021) are therefore better interpreted as a mixture of silver components than as an effect of debasement by copper addition. The range of ^{206}Pb/^{204}Pb observed at nearly constant Cu concentration by Eshel et al. (2021, Fig. 8) for Beth Shean 1029a and b and 1095 and for Ashkelon confirms the lack of correlation between the two variables and cautions against overemphasizing the effect of debasement.

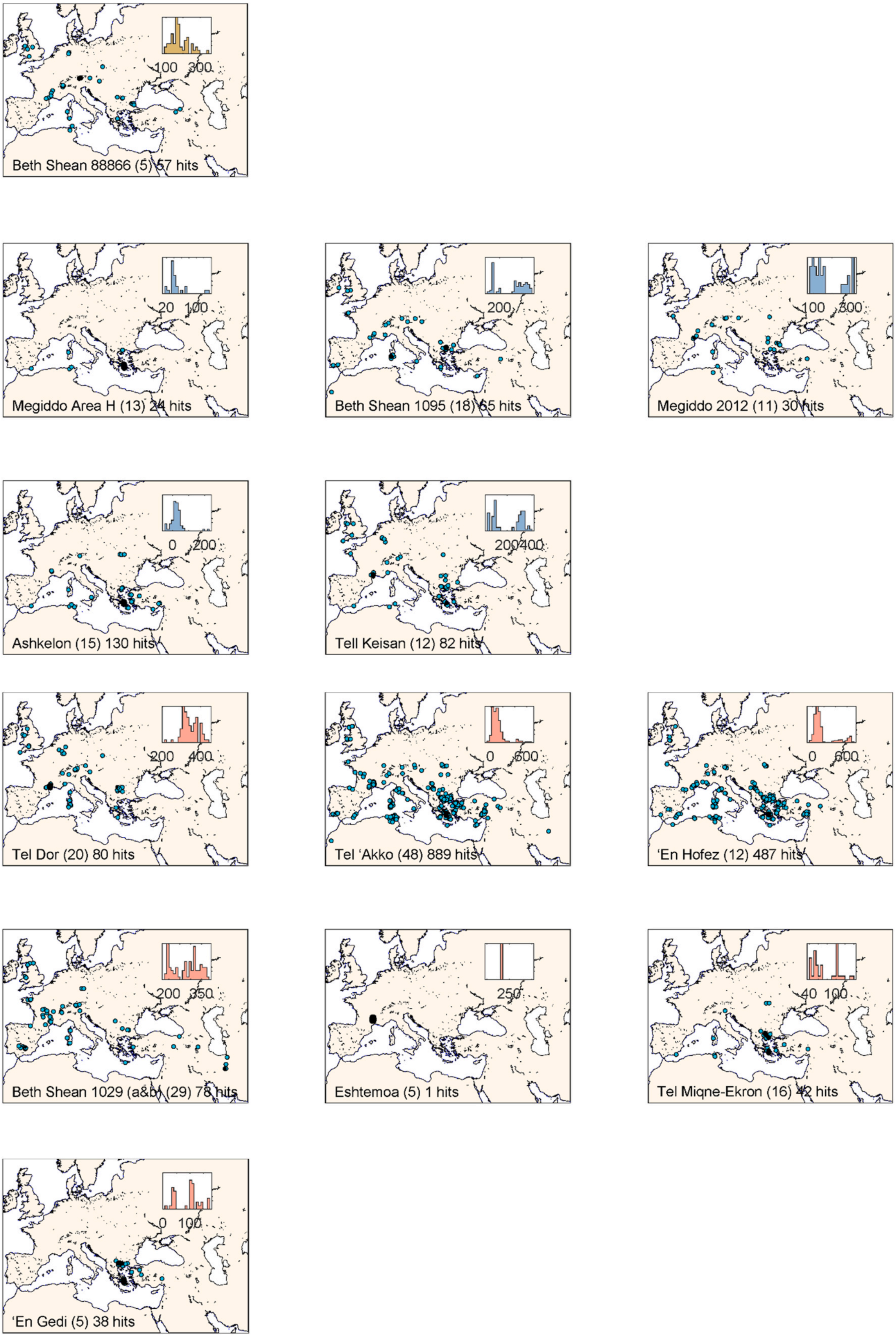

Fig. 3. Map hits for the different hoards (in cyan). Each hit represents an ore falling in the sample convex hull of each hoard. The Pb isotope compositions of ~7000 ores were tested for inclusion in the convex hull of each hoard. Top panel: Bronze Age (yellow). Middle five panels: Iron Age I (blue). Bottom seven panels: Iron Age II (red). The black fields (ʻhot spotsʼ) include areas making up more than 10% of the hits (an equivalent of bi-dimensional histogram peaks). Upper right-hand corner insets are Pb model age histograms (Ma). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 2
Approximate dates of deposition of the *Hacksilber* hoards analysed for Pb isotopes, number of hits, and probable origin of the metal.

Hoard	Period	Years (BCE)	Ref.	Hits	Probable silver source(s)
Beth Shean 88866	Late Bronze Age III	1200–1150	1	24	Unknown
Megiddo Area H	Iron Age I	1070	2	56	Lavrion
Beth Shean 1095	Iron Age I	1150–950	3	57	Thrace, Sardinia
	Iron Age I	1050–950	4	31	Cévennes
	Iron Age I	1050–950	6	78	Lavrion
Tell Keisan	Iron Age I	1050–950	7	86	Lavrion, Cévennes
Tel Dor	Iron Age IIA	2nd half 10th century	8	786	Cévennes
Tel ʿAkko	Iron Age IIA	10th to 9th centuries	9	422	Lavrion
Beth Shean 1029a and b	Iron Age IIA	950–800	3	134	Sierra Morena, eastern Persia
ʿEn Ḥofez	Iron Age IIA	9th century	9	78	Lavrion
Eshtemoa	Iron Age IIB	8th century	10	1	Unknown
Tel Miqne-Ekron	Iron Age IIC	Late 7th early 6th centuries	11	35	Lavrion, Thrace
ʿEn Gedi	Iron Age IIC	Late 7th early 6th centuries	12	33	Lavrion, Macedonia

References: 1 – Thompson (2009); 2 – Arie (2013); 3 – Rowe (1940); 4 – Paice (2004); Hall (2016); 5 – Balmuth (2001); 6 – Nodet (1980); 7 – Stern (1998, 2001); 8 - Thompson and Skaggs (2013); 9 – Alexandre (1997); 2013; 10 – Yeivin (1990); Kletter and Brand (1998); 11 - Golani and Sass (1998); Gitin and Golani (2001); 12 - Kletter and Degroot, 2007.

Table 3
Results of principal component analysis in ^{204}Pb/^{206}Pb–^{207}Pb/^{206}Pb–^{208}Pb/^{206}Pb space for each hoard. PC1, PC2, and PC3 represent the percentage of variance accounted for by the corresponding component. One, two, and three significant percentages correspond to the presence of two, three, and four end-members, respectively.

Hoard	PC1	PC2	PC3	N° of end-members
Beth Shean 88866	98.1	1.9	0.0	2–3
Megiddo Area H	99.8	0.1	0.1	2
Beth Shean 1095	77.8	21.4	0.8	3
Megiddo 2012	95.5	4.5	0.0	2–3
Ashkelon	99.3	0.7	0.0	2
Tell Keisan	96.0	4.0	0.1	2–3
Tel Dor	99.2	0.8	0.0	2
Tel ʿAkko	98.8	1.2	0.0	2
ʿEn Ḥofez	99.7	0.2	0.1	2
Beth Shean 1029a and b	99.4	0.5	0.1	2
Eshtemoa	97.9	2.1	0.0	2–3
Tel Miqne-Ekron	85.4	10.8	3.8	4
ʿEn Gedi	98.5	1.5	0.0	2–3

We took advantage of the mixing model of Eshel et al. (2021) to calculate the hits for each of the convex hulls displayed in Fig. 2, with the expectation that potential end-members would be more frequent in the ore database if they could be safely considered as end-members. Evidence obtained from these convex hull analyses is extremely rich and found to support or falsify previous theories as follows:

- The prevalence of Lavrion, Macedonia, Thrace, Sardinia, and southern Gaul (the Cévennes) as silver sources is particularly strong. Communication between these areas and the Levant, whether peacefully by trade or by way of war, never really stopped during the Late Bronze Age through the Iron Age IIC, and/or *Hacksilber* was recycled from earlier times.
- The prevalence of hits from southern Gaul is particularly interesting, as these mines have been known to be active since Roman times (Baron et al., 2005; Elbaz-Poulichet et al., 2017). The present study, however, indicates mining activity in the area centuries prior. We posit that this activity would conform to a broader pattern of silver exploitation and sale by dominant local tribes across Thrace and Iberia.
- The case for Sardinia is only strong for Beth Shean 1095. Iron Age Nuragic settlements are known from Sardinia and Corsica (Balmuth, 1992), but evidence of contemporaneous mining is missing. Taurus provided a few hits (Tel ʿAkko, ʿEn Ḥofez), but the small number of ore samples does not make this area an incontrovertible source of silver. Central Asia, Turkey and Cyprus, with few hits and inadequate geology for hosting Ag-bearing ores, are unlikely to have been significant silver providers.
- Evidence for exploitation of silver ore sources in Iberia is surprisingly scant, with one exception being the Sierra Morena (Beth Shean 1029a and b). Hits in the Betics exist but are in general second to more productive areas in the Aegean domain. Possible hits near Huelva and in southern Portugal are uncommon, limited to Meggido Area H and Tel Dor. This area may be underrepresented because silver needed to be complemented with Pb for cupellation. Silver was mined from jarosite in the Lower Guadalquivir (Tartessos), while Pb was extracted from galena mined in the distant Betics and Ossa Morena Zone (Anguilano et al., 2010). This may be why the latter district is found only in the Tel ʿAkko and ʿEn Ḥofez hoards.
- The assignment of silver to other localities is more speculative, either because the number of hits is too small or because little evidence is known from archaeological and textual contexts. This is the case of the Eifel, Germany, the Palaeozoic basement of the southern Massif Central of France (the Cévennes), the Alps, and the Carpathians. Hits in Crete, Peloponnese, Tunisia, and even the British Isles may at this point be considered coincidental because they are unsupported by archaeological evidence.

One possible conclusion is that exchanges between the southern Levant and the Aegean world never entirely came to a full stop during the Late Bronze and Iron Ages. What supported these exchanges is not always clear; war, as attested to by the violent demise of large towns, and trade, the only possible ways to attract silver to the Levant, a territory where silver mines are absent. In the context of the overall fineness of *Hacksilber* (Eshel et al., 2018, 2021), occasional silver famines may be the reason for unusually high Cu contents in Beth Shean 88866 and Megiddo Area H (Eshel et al., 2021) rather than deliberate debasement but other explanations are possible.

Alternatively, the prevalence of mixing in some hoards, the low percentage of silver, and the relatively few hoards from over half a millennium suggest that bullion was hoarded over long periods of time. The scatter of isotopic values within some hoards can be taken as evidence of such long-term hoarding, while the scatter in the plot of ^{206}Pb/^{204}Pb vs Cu in wt.% (Eshel et al., 2021, Fig. 8) shows the lack of coherent mixing hyperbolae for hoards such as Tell Keisan, Megiddo Area H, and Beth Shean 1095, and requires that the analysed samples were produced from silver with widely different Pb contents and not from a single source. This pattern may be usefully contrasted with early minting of silver coinage in the Late Archaic period which overwhelmingly came from single ore sources refined to a very high percentage of silver (Stos-Gale and Davis, 2020; Birch et al., 2020). If mixing due to recycling is the case, we should in theory expect to see a linear array trending parallel to the Pb evolution curve (lines of constant μ and κ) (Albarède et al., 2020). Good examples of mixing are those of Megiddo Area H and the Tell Keisan hoards (Eshel et al., 2021). However, mixing is by definition between different end-members (A and B) which should manifest in ore ’hits’ on the end-members A and B, but not on the mixtures between A and B which have no natural match. If the mixture is between different ‘pure’ issues (without remelting), it would show up, not as alignments, but as separate groups of points corresponding to each issue. Although the hoards seem to show both types of behavior, there are not enough samples analysed at present to conclusively separate the groups. Hence, until enough data points have become available to conclusively demonstrate via statistical means long-term hoarding and mixing, trade continuity is not contradicted by the present data and overall remains a valid working hypothesis.

Exchanges between the eastern and western Mediterranean are more problematic. They may have been less active during the turmoil at the end of the Late Bronze Age but clearly were revived in the Early Iron Age I.

6. Conclusions

This study has demonstrated the applicability of a data-driven approach to lead isotope analysis. Principal component analysis of the most precise Pb isotope data (this work combined with Eshel et al., 2019; 2021) confirms that the Pb isotope compositions of each *Hacksilber* sample in each hoard can be accounted for by a small number of end-members (2–3). The new ‘convex hull’ approach applied to the ore database of Pb isotopes identifies more previously unrecorded potential source end-members. The concepts of ‘hits’ and ‘hot spots’ introduced here are critical in assigning provenance to individual isotopic data and data groups.

The prevalence of Aegean sources including Lavrion, Macedonia, Thrace, Sardinia, and southern Gaul as long-lived major sources of silver shows that exchanges between the southern Levant and the Aegean continued at least intermittently from the end of the Late Bronze Age through the Iron Age III. A caveat is that long-term hoarding suggested by isotopically heterogeneous hoards and use of low-purity silver may have blurred evidence of trade disruption. Southern Gaul is proposed as an Iron Age silver source. Occasional exchanges with Sardinia and southern Iberia in the Iron Age are confirmed. The Aegean world, including Thraco-Macedonian sources, dominated silver supply presumably because of its proximity, but exchanges between the eastern and western Mediterranean did not disappear from the isotopic record.

Declaration of competing interest

None.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jas.2021.105472.

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