



Copper-based metalwork in Roman to early Islamic Jerash (Jordan): Insights into production and recycling through alloy compositions and lead isotopes

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ABSTRACT

Metallographic, chemical and lead isotopic analyses of copper-based artefacts recovered from the Northwest Quarter in Jerash (ancient Gerasa) in Jordan provide new information on the civic life and material culture from a key urban site in the Roman Empire's eastern provinces. The samples span the city's occupation from its flourishing under Roman rule into the Byzantine and early Islamic periods. We examined 49 copper-based artefacts using reflected light microscopy and micro-X-ray fluorescence. A subset of these artefacts was analysed by electron microprobe spectroscopy for major and minor elements at higher spatial resolution, and by multi-collector inductively coupled plasma mass spectrometry for lead isotopes. Results imply that binary bronze dominated the Roman period, (leaded) brass characterised the Byzantine period, while tin-containing alloys were prevalent during the Islamic period. Lead isotopes suggest that during the Roman and Byzantine periods some of the metal in Jerash came from European and/or Mediterranean sources, while copper used during the Islamic period may have been sourced more locally from Timna. The changes in alloy types and lead isotopes suggest that recycling of metals took place in Jerash possibly as early as the Roman period and more frequent from the Byzantine period onwards.

1. Introduction

Jerash (ancient Gerasa) in northwest Jordan (Fig. 1) was founded in the Hellenistic period and flourished under Roman rule as one of the cities of the Syrian Decapolis, a group of nominally ten cities on the empire's eastern frontier with a shared cultural background. The city plan of Gerasa reflects strong influence of Roman urban planning (Kraeling, 1938; Lichtenberger et al., 2016; Lichtenberger and Raja, 2018b; Zayadine, 1986), and in late antiquity the urban layout and high monetarisation (Birch et al., 2019a; Schulze and Schulze, 2018) attest to economic prosperity. Activity in Jerash declined markedly following a devastating earthquake in January 749 CE. Small-scale occupation revived in the Ayyubid-Mamluk period (12th-15th century CE) (Lichtenberger et al., 2016; Lichtenberger and Raja, 2018b). Excavations by the Danish-German Jerash Northwest Quarter project during 2011–16 yielded a corpus of copper-based objects that provide new

insights into Jerash's material culture (Lichtenberger and Raja, 2018c, 2018b, 2017, 2016).

This study examines the copper-based assemblage from domestic contexts recovered in the Northwest Quarter in Jerash to determine their metalworking, chemical and isotope compositions, and place them within their cultural-historical context (Table 1). The type of metals and alloys recovered from Jerash and their associated technologies over more than six centuries (Roman to Islamic periods) are analysed and discussed. The samples stem from different contexts including secondary fill layers or dumps, and domestic contexts. Since the sample size is small, it is not possible to directly generalise from the spatial distribution to function and use of the objects and the relation to the metallurgical technology. Nonetheless the analysis and discussion undertaken here add valuable knowledge about metal artefacts across a long timespan. The elemental analyses of the copper-based artefacts aim to monitor the continuity and change of alloy types in relation to

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Fig. 1. Map showing the locations of Jerash and other sites mentioned in the text in the Eastern Mediterranean (adapted from Natural Earth).

shifting socio-cultural dynamics, and to elucidate technological aspects to better understand aspects of specialisation and standardisation of the metallurgical production. Of particular interest is a group of objects identified as a scrap metal hoard (the box group) kept together for re-use in a wooden box of which only the metallic hinges are preserved (Lichtenberger et al., 2016). The deposition of this small hoard dates to the last phase of the early Islamic occupation and was preserved due to the site's abrupt abandonment following the 749 CE earthquake. Examination of the fourteen copper-based samples from the metal hoard addresses aspects of recycling management in the early Islamic period. Metal hoarding for re-use raises basic, but crucial, questions about Jerash's early Islamic copper-based technology and, by extension, contemporary production and social organisation. Investigation of the metals' provenance and potential changes over time address questions about the supply of resources during a changing political framework at Jerash from Roman to Islamic rule. Jerash's proximity to the rich copper ore district in the Arabah Valley and the sites of Wadi Faynan and Timan (Fig. 1) with evidence of intense mining and smelting activity from prehistory onwards (Ben-Yosef, 2012; Hauptmann, 2007; Levy et al., 2002) raises additional questions regarding the nature of the copper used.

2. Materials and methods

Metallographic, elemental and lead isotopic analyses were conducted at the Aarhus Geochemistry and Isotope Research (AGiR) Platform, Department of Geoscience, Aarhus University. Additional chemical data were obtained at the Department of Earth and Planetary Sciences, UC Davis, by electron microprobe spectroscopy.

2.1. Sample description

A total of 49 objects were examined in this study, including utensils and jewellery, many in fragmentary state, as well as 14 objects from a scrap metal hoard (Table 1, Fig. 2). The samples include a range of object types and their typology is discussed in detail by Eger (in press). The hoard contained both ferrous and non-ferrous metals. Only data for the copper-based objects are reported here. Attribution based on typology and context show that 5 objects are Roman, 3 Roman or

Table 1
Description and chronological period of copper-based objects examined in this study.

Sample ID	Find No. ^a	Description ^b	Chronological period ^c
J1	J12-B-2-1253	Ring	Byzantine/Early Umayyad
J2	J12-Bd-34-1	Nail	Byzantine/Early Umayyad
J3	J12-Bb-62-36	Pin/utensil	Byzantine/Early Umayyad
J4	J13-Ed-18-8	Ring	Late Byzantine
J5	J13-Ed-18-9	Strip (sheet metal)	Late Byzantine
J6	J13-Ed-18-10	Sheet	Late Byzantine
J7	J13-Ed-23-9	Pin (see also J62)	Late Byzantine
J8	J13-Ed-18-80	Plate (scale?)	Late Byzantine
J9	J14-Kefgh-3-166	Knob or Button ^d	Hellenistic/Roman to Mamluk
J10	J13-D-25-6	Weight	Ayyubid/Mamluk
J11	J14-Kf-3-3x	Bracelet	Byzantine to Mamluk
J12	J14-Ke-3-208	Sheet	Byzantine to Mamluk
J13	J14-Ke-3-211	Former/model	Late Byzantine
J14	J14-Ke-3-214	Tube	Hellenistic/Roman or younger
J29	J14-Icd-35-3	Pin	Umayyad or Ayyubid/Mamluk
J36	J15-Qc-18-1	Earring	Late Byzantine
J37	J15-Ob-108-13	8-shaped ring	Umayyad (?)
J38	J15-Qc-23-4	Sheet	Early Byzantine
J39	J15-Qc-31-1	Needle	Roman
J40	J15-Qd-38-27	Needle	Roman
J41	J15-Oe-90-3	Needle	Umayyad
J42	J15-Qd-52-2	Nail	Roman
J44	J15-Qf-60-13	Object fragment	Roman
J45	J15-Pe-16-193	Sheet	Umayyad
J47	J16-Sc-13-7	Pin	Late Roman/Byzantine
J49	J16-Sb-23-6	Pin	Roman
J50	J16-Uc-19-8	Hook	Late Byzantine/ Umayyad
J51	J16-Sd-22-44	Utensil (folded pin)	Late Byzantine/ Umayyad
J52	J16-Vc-61-1x	Sheet	Umayyad
J54	J16-Vg-67-4x	Needle	Umayyad
J57	J16-Tb-35-12	Pin	Umayyad
J59A	J16-Vg-69-5x	Object fragment	Umayyad
J59B	J16-Vg-69-5x	Object fragment	Umayyad
J62	J16-Sg-22-145	Pin (see also J7)	Late Byzantine/ Umayyad
J63	J16-Td-52-2x	Hook with chain	Ayyubid/Mamluk
J64	J16-Uc-60-12x	Sheet	Late Byzantine/Early Umayyad
J105	J14-Ki-3-215	Hook	Undefined
<i>Samples from the box (Box group)</i>			
J15	J14-Kh-34-1	Weight	Late Byzantine
J16	J14-Kh-34-21	Spindle	Hellenistic/Roman and later
J17	J14-Kh-34-25	Utensil	Hellenistic/Roman and later
J18	J14-Kh-34-27	Belt part	Late Byzantine
J20A	J14-Kh-3-21x	Key with ring (ring)	Late Byzantine
J20B	J14-Kh-3-21x	Key with ring (key)	Late Byzantine
J21	J14-Kh-34-22	Hinge	Late Byzantine
J22	J14-Kh-34-23	Utensil	Hellenistic/Roman and later
J24	J14-Kg-3-14x	Lockplate with keyhole	Late Byzantine
J28	J14-Kh-34-26	Utensil	Late Roman/Byzantine
J33.1	J14-Kh-34-5	Utensil	Late Byzantine
J33.2	J14-Kh-34-10	Bracket	Late Byzantine
J34	J14-Kh-34-11	Lid	Hellenistic/Roman and later
J35	J14-Kh-34-12	Vessel foot	Late Roman/Byzantine

^a Find nos.: the first part (e.g. J13) refers to the year of excavation and the second part (e.g. Kh) refers to the trench and the respective sectors. The consecutive numbers refer to the evidence/locus and numbers ending in x refer to special find numbers.

^b Based on field observations and typology.

^c Chronological classification by C. Eger for the Danish-German Jerash Northwest Quarter Project based on typology and context.

^d Pers. com. D. Ignatiadou.



Fig. 2. Key with ring J20A-B (A), lockplate with keyhole J24 (B) and square plate weight J15 (C) recovered at Jerash, all date originally to Late Byzantine period. B and C and were recovered from the context of the Umayyad wooden box.

Byzantine, 14 Byzantine, 7 Byzantine or Umayyad, 7 Umayyad, 3 Umayyad or Ayyubid/Mamluk, and, finally, 10 are of undefined or too broad chronological periods. The significant gap between the Umayyad and Ayyubid/Mamluk periods in the assemblage reflects the hiatus in the Northwest Quarter's habitation. Objects from the hoard date to the Umayyad period and earlier (Lichtenberger et al., 2016). The typological analysis was done by Dr. Christoph Eger, Berlin, and will be published in the final publications of the Danish-German Jerash Northwest Quarter Project (Eger, *in press*).

2.2. Sample preparation

Objects were sampled by cutting small pieces from fragmented artefacts (31 samples) using a jeweller's saw or by drilling complete objects (18 samples) using a 0.5–1 mm diameter drill bit (twist drill consisting of tool steel by Fisher; fresh drill bits were used for each sample to avoid contamination); see Table 2 for sampling method of all objects. For metallographic examination and chemical mapping, samples were prepared as standard metallographic blocks, ground and flatly polished (Scott, 1991). Material for lead isotope analysis was transferred directly to Teflon vials for acid dissolution.

2.3. Reflected-light microscopy

Reflected light microscopy (RLM) using a Nikon Eclipse E600 POL microscope equipped with a Nikon digital shift camera and imaging software was employed on cut samples to characterise metallographic microstructures and relate these to manufacturing methods; see Table 2 for a list of examined samples). Metallographic observation considered metalworking techniques such as casting, hammering or annealing (repeated hot and cold working) as indicated by the presence of dendrites or recrystallised grains with or without annealing twins, respectively (Scott, 1991).

2.4. Micro-X-ray fluorescence analysis

Chemical maps and spot analysis of all 49 artefacts were performed using a Bruker M4 Tornado micro-X-ray fluorescence (μ XRF) system under vacuum with a Rh-anode X-ray tube operating at a 50 kV, anode current of 600 mA, and polycapillary X-ray optics focusing the beam to a spot size of ~ 20 μ m. Secondary X-ray fluorescence was quantified using two silicon drift detectors in different positions that permit discrimination between fluorescence and diffraction peaks. For area analysis we used a scan speed of 800 μ m/second for two cycles and over

1–2 mm². Area maps were especially useful for correlating microstructural information obtained by RLM with composition, and for identifying areas of corrosion (Nørgaard, 2017; Orfanou and Rehren, 2015). The instrument's operation was evaluated with the analyses of 6 certified reference materials, including 3 from the CHARM set specifically designed for ancient metals (Heginbotham et al., 2015). Further analytical details and certified reference materials analyses are reported in supplementary material A (Fig. S1, Table S1). Detection limits (DL) are for arsenic 0.05 wt%, for lead, iron, nickel and sulphur 0.1 wt%, for tin 0.2 wt%, for zinc 0.5 wt%, while for lighter elements related to corrosion products such as calcium, chlorine, potassium and silicon 0.5 wt%.

2.5. Electron probe microanalysis

Polished cross-sections of 35 objects', used for μ XRF analysis, were also analysed by electron-probe microanalyzer (EPMA) for their major and minor element compositions; see Table 2 for list of examined objects. Compared to the μ XRF, EPMA has the advantage of direct calibration using pure metal standards (from CM Taylor), better precision and a spot size down to 1–2 μ m. The selected samples were analysed by EPMA to monitor possible effects of corrosion that would not have been detected by the μ XRF's lower resolution and to examine minor elements at lower detection limits. We used a Cameca SX-100 microprobe at UC Davis equipped with wavelength and energy dispersive spectrometers and we routinely analysed for S, Fe, Ni, Cu, Sn, Zn, As, S, Sb and Pb (see also Table S2 in Supplementary Material B for further analytical details). Here, detection limits for copper, zinc and lead were 0.01 wt%, 0.03 wt% for iron, 0.02 wt% for sulphur, 0.05 wt% for tin, nickel and antimony, and 0.06 wt% for arsenic.

2.6. Lead isotope analyses

Lead isotope ratios were determined on copper-based alloys using standard analytical methods (Klein et al., 2009). A representative subset of 7 samples was selected covering the whole assemblage chronologically (Table 2). Drillings were first leached with cold 6 N HNO₃ for several minutes following the procedure of Ling et al. (2014). All the samples were dissolved in 6 N HNO₃ at 110 °C in 7 ml Teflon beakers. After dissolution, solutions were dried down and taken up in hydrobromic acid. These solutions were loaded on Teflon columns containing BioRad™ AG 1-X8 (200–400 mesh) resin for ion exchange separation of Pb using hydrobromic-hydrochloric acid dilutions (e.g. White et al., 2000). Following Pb elutions, samples were again dried

Table 2

Samples obtained and analyses conducted (reflected light microscopy/RLM, μ XRF, EPMA and lead isotope analyses) in the copper-based objects during this study; 'x' indicates samples investigated with the respective analytical techniques.

Sample ID	Sample (for RLM, micro XRF, EPMA)	RLM	μ XRF	EPMA	Pb isotope analysis
J1	drilling	–	x	x	–
J2	cut	x	x	x	–
J3	cut	x	x	x	–
J4	cut	x	x	x	–
J5	cut	x	x	x	–
J6	cut	x	x	x	x
J7	cut	x	x	x	–
J8	drilling	–	x	x	–
J9	drilling	–	x	–	–
J10	cut	x	x	x	–
J11	drilling	–	x	–	–
J12	cut	x	x	–	–
J13	drilling	–	x	x	–
J14	cut	x	x	–	–
J15	drilling	–	x	x	x
J16	drilling	–	x	–	–
J17	cut	x	x	–	–
J18	cut	x	x	x	–
J20A	drilling	–	x	x	x
J20B	drilling	–	x	x	–
J21	drilling	–	x	x	–
J22	drilling	–	x	–	–
J24	drilling	–	x	x	–
J28	drilling	–	x	–	–
J29	drilling	–	x	–	–
J33.1	drilling	–	x	x	–
J33.2	cut	x	x	x	–
J34	cut	x	x	–	–
J35	drilling	–	x	–	–
J36	cut	x	x	x	–
J37	drilling	–	x	x	–
J38	drilling	–	x	–	x
J39	cut	x	x	x	–
J40	cut	x	x	x	–
J41	cut	x	x	x	–
J42	cut	x	x	x	–
J44	cut	x	x	–	–
J45	cut	x	x	x	–
J47	cut	x	x	–	–
J49	drilling	–	x	x	x
J50	cut	x	x	x	–
J51	cut	x	x	x	–
J52	cut	x	x	x	–
J54	cut	x	x	x	x
J57	cut	x	x	x	x
J59A	cut	x	x	x	–
J59B	cut	x	x	x	–
J62	cut	x	x	x	–
J63	cut	x	x	x	–
J64	cut	x	x	x	–
J105	cut	x	x	–	–

down and taken up in 2% HNO₃ in preparation of analysis. Just prior to analysis, Tl was added at a Pb:Tl ratio of 1:3 for the mass fractionation correction by simultaneously measuring the ²⁰³Tl/²⁰⁵Tl ratio and Pb isotope ratios for samples and NBS 981 standard. Solutions were analysed on a Nu Plasma II multi-collector ICP-MS equipped with 12 Faraday cups and 3 ion counters in a fixed array. We monitored mass 202 (²⁰²Hg) for Hg interference on mass 204 (²⁰⁴Pb + ²⁰⁴Hg) – which was found to be insignificant on the ²⁰⁶Pb/²⁰⁴Pb, ²⁰⁶Pb/²⁰⁴Pb and ²⁰⁶Pb/²⁰⁴Pb ratio. The procedural blank contained 19 pg of Pb. ²⁰⁶Pb/²⁰⁴Pb, ²⁰⁶Pb/²⁰⁴Pb and ²⁰⁶Pb/²⁰⁴Pb ratios for USGS basalt standard BHVO-2 (n = 2) are 18.649 ± 0.019, 15.546 ± 0.006 and 38.235 ± 0.009, respectively, which are within uncertainties of expected values of 18.634 ± 0.034, 15.524 ± 0.025 and 38.146 ± 0.373 (GeoRem – preferred values). CRM bronze and brass

standards (n = 5) gave constant values of ²⁰⁶Pb/²⁰⁴Pb, ²⁰⁶Pb/²⁰⁴Pb and ²⁰⁶Pb/²⁰⁴Pb ratios of 17.580 ± 0.052, 15.553 ± 0.038 and 37.414 ± 0.146, respectively (see Supplementary Material C – Table S3).

3. Results

3.1. Elemental compositions

3.1.1. Major elements

The μ XRF and EPMA analyses confirm that all objects are copper or copper-based alloys. Alloy types were determined considering the approaches of similar studies and based on the results' distribution. Tin and lead amounts above 2 and 3 wt%, respectively, are considered intentional additions including the possibility of mixing scarp copper alloys during recycling, as lower values could reflect natural abundances in tin- or lead-rich copper ores (Hauptmann, 2007; Pernicka et al., 1990; Tylecote et al., 1977). Even though up to 5 wt% Pb has been suggested as naturally occurring from lead-rich copper ores (Pernicka et al., 1990), here we marked > 3 wt% Pb as 'leaded', based on a gap in the distribution of lead values in the analysed assemblage. Similarly, the limit for tin is based on a gap in the data between approx. 2 and 4 wt%. Alloys with < 5 wt% Zn are not considered brasses, also following Caley (1964, p. 69), as such amounts could be accidentally formed by mixing scrap brass or by smelting natural alloys rather than reflecting the production of copper-zinc alloys via cementation (e.g. Burnett et al., 1982; Craddock et al., 2004, 1980; Merkel, 2018). The levels of the rest of the alloying elements further suggest the accidental presence of < 5 wt% Zn in these samples. Of the 51 analyses including 2 composite objects, 19 are classified as bronze, 18 as brass and 14 as copper. Of these, 21 with lead > 3 wt% are grouped as leaded. The 2 composite objects are a brass key and ring (J20) both of brass and object J59 made of bronze and brass (Fig. 10).

Co-variations of tin, lead and zinc are presented in Figs. 3–5 (see Supplementary materials D & E for elemental analyses results and Table 5 for alloy types of all samples). Bronze objects are found with as much as 11.5 wt% Sn, while leaded bronze objects can contain as much as 23 wt% Pb. Likewise, brass objects have up to 23 wt% Zn while the leaded type typically has < 8 wt% Pb. Copper objects all have tin and zinc concentrations < 2 and 4 wt%, respectively, while those of the leaded type have as much as 17 wt% Pb.

3.1.2. Minor elements

The concentrations of minor elements (typically < 0.5 wt%) detected in the sample by μ XRF and EPMA methods are presented in Tables S4 and S5 in Supplementary material D and E, respectively. Iron

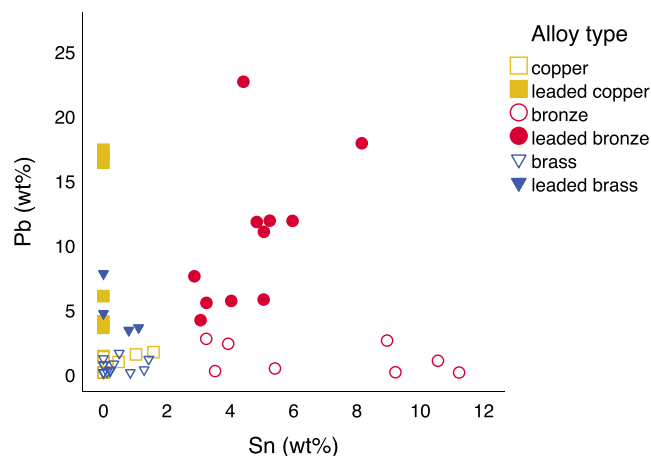


Fig. 3. Co-variation of lead and tin for Jerash objects classified by alloy type as measured with μ XRF.

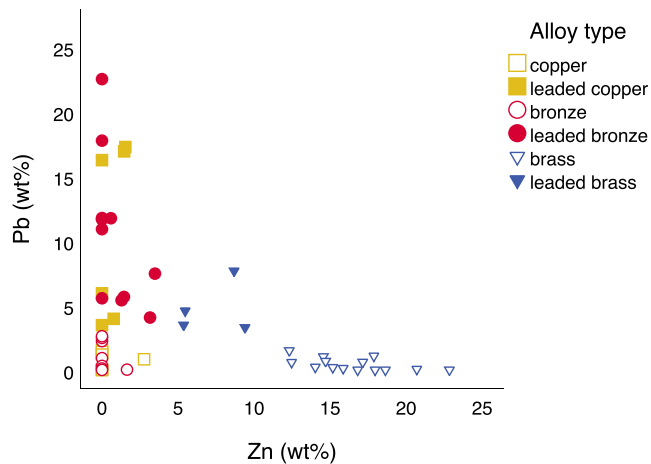


Fig. 4. Co-variation of lead and zinc for Jerash objects classified by alloy type as measured with μ XRF.

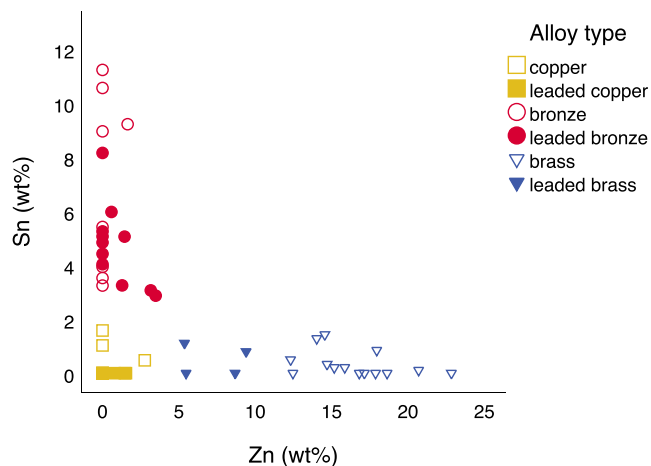


Fig. 5. Co-variation of tin and zinc for Jerash objects classified by alloy type as measured with μ XRF.

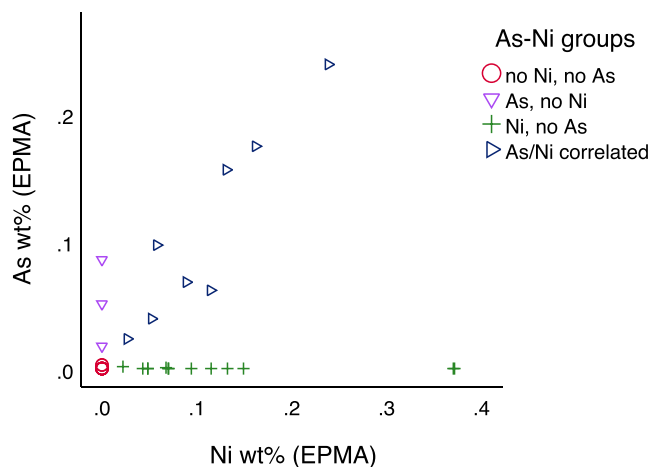


Fig. 6. Co-variation of nickel and arsenic in the 36 samples as analysed by EPMA. Four groups are visible: one with arsenic and nickel below the limit of detection (red circles), a second one with detected arsenic but no nickel (purple triangles), a third one with detected nickel but not arsenic, and a fourth one with detected arsenic and nickel positively correlated (blue triangles).

and arsenic concentrations below are based on the μ XRF data set as the values correlate well with the EPMA analyses, though nickel was better detected by the EPMA. Iron is present in most of the samples up to

0.5 wt%, but typically < 0.3 wt%. Iron concentrations are notably higher in (leaded) brasses (total of 18 samples) than other alloy types with means of 0.25 wt%. Arsenic is detected in about half of the analysed objects. Arsenic in leaded copper (6 samples) is 0.3 wt% whereas it is ≤ 0.1 wt% in all other alloy types. Nickel is detected in 21 of 36 objects analysed by EPMA with concentrations of ~ 0.07 wt%. Fig. 6 shows 4 sub-groups regarding arsenic and nickel concentrations as analysed by EPMA, namely one with none of the two elements detected (12 samples), a second one with arsenic detected but with no nickel (3 samples), a third one with nickel detected but no arsenic (13 samples), and a fourth one with both nickel and arsenic present and correlated with a factor of 0.9 (8 samples) (see Table 5 for information on individual samples). Arsenic-nickel groups do not correlate with alloy type. Antimony was detected in 2 samples in the EPMA data set with values ~ 0.1 wt%. Finally, in the box group (see above Sections 1 and 2.1), the minor elements are comparable to the rest of the assemblage.

3.2. Lead isotope analyses

Lead isotopes were measured in 7 objects spanning the alloy types and chronological periods. All results are reported in Table 3 and shown graphically in Fig. 7. Lead isotopes for the artefacts range from 17.603 to 18.773, 37.511 to 38.852 and 15.629 to 15.668 for $^{206}\text{Pb}/^{204}\text{Pb}$, $^{208}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$ respectively. $^{207}\text{Pb}/^{206}\text{Pb}$ ratios range from 0.835 to 0.888 and from 2.070 to 2.131 for $^{208}\text{Pb}/^{206}\text{Pb}$. The core and surface analyses of the Roman bronze spindle (J49) are distinctly different.

3.3. Metallography

Representative microstructures revealed in mounted polished cross-sections of metal samples are shown in Figs. 3–5, and observations of 32 samples are summarised in Table 4 (more details on individual samples are provided in Table 5 and Supplementary material F – Table S6). Dendrites in 11 samples suggest casting (Fig. 8) and 14 samples show polygonal grains suggesting cold working, i.e. hammering with or without annealing as suggested by slip planes and twins in 7 samples (Fig. 9, Table 5). Most of the samples contained minute sulphide inclusions (28 objects) of which 15 objects also contain lead globules. Large vesicles, such as shown in Fig. 8, were found in 3 samples (J5, J10 and J14). Weight J10 contains a bluish phase having high tin content compartmented to the surrounding yellowish dendrites rich in copper. Umayyad object J59 is a cast shallow hemisphere (diameter ~ 5 mm) attached to a hammered sheet (Fig. 10).

4. Discussion

4.1. Metalworking

A range of metalworking techniques from single step casting to repeated cold working were employed in the objects' production. Hammered objects span the full range of alloy types while 18 out of 21 samples are unleaded (Table 4). Samples with annealing twins comprised only bronze and brass, while cast objects featured ternary alloys with various ratios of copper, tin and lead, but with a marked preference for leaded alloys. The choice of mostly unleaded alloys for cold working that would have rendered them brittle in the process shows the craftspeople's sense of the metals' physical properties. Similar choices have often been noted in the archaeological record, placing the Jerash assemblage within a long tradition of technological choices. The relatively low lead content of 3 cast objects (leaded brass J6, and leaded bronze J47 and J50) between 3.5 and 5 wt% would have allowed their cold working.

The Umayyad period object J59 consists of cast bronze and hammered brass alloys, and is the only object in the assemblage for which two distinct alloys were used (Fig. 10). This object is fragmented and so

Table 3

Lead isotopes for Jerash metal artefacts from the Byzantine (Byz), Roman (Rom) and Umayyad (Uma) periods and standard materials.

Sample ID	Period	Alloy type	Tested for Cu/Pb	$^{208}\text{Pb}/^{206}\text{Pb}^3$	$^{207}\text{Pb}/^{206}\text{Pb}^3$	$^{206}\text{Pb}/^{204}\text{Pb}^3$	$^{207}\text{Pb}/^{204}\text{Pb}^3$	$^{208}\text{Pb}/^{204}\text{Pb}^3$	Model Age (Ma) ⁴	Broad ⁵	Euclidean neighbours ⁶
J6	Byz	leaded brass	Cu/Pb	2.0801	0.84204	18.589	15.653	38.667	137	W. Europe	Massif Central
J15	Byz	brass	Cu	2.0791	0.84096	18.617	15.657	38.707	124	W. Europe	Massif Central
J20A	Byz	brass	Cu	2.0696	0.83451	18.773	15.666	38.852	28	Mediterranean	Cyprus
J38	Byz	bronze	Cu	2.0833	0.84253	18.576	15.651	38.701	143	W. Europe	Massif Central
J49 Core ¹	Rom	bronze	Cu/Pb	2.0729	0.83651	18.730	15.668	38.825	64	Unknown	Cyprus
J49 Surface ¹			Cu/Pb	2.0810	0.84141	18.607	15.656	38.720	132	–	–
J54(1) ²	Uma	leaded bronze	Pb	2.0808	0.84176	18.606	15.662	38.715	144	Unknown	Romania/ Bulgaria
J54(replicate) ²			Pb	2.0803	0.84163	18.603	15.656	38.698	135	Unknown	Romania/ Bulgaria
J57	Uma	bronze	Cu	2.0935	0.85542	18.270	15.629	38.247	326	Unknown	Timna

¹ J49 analysed with sample from the object's core and surface.² J54 analysed twice for checking the comparability of produced results.³ Total analytical error estimated from long-term reproducibility of NBS981 are 0.1% or lower for ratios against ^{204}Pb and 0.02 or less for $^{208}\text{Pb}/^{206}\text{Pb}$ and $^{207}\text{Pb}/^{206}\text{Pb}$.⁴ Model Age refers to those as calculated by [Stacey and Kramers \(1975\)](#).⁵ Broad refers to suggested regional provenance interpreted from lead isotope bivariate plots of reference data.⁶ Euclidean neighbours refers to the nearest reference data points in the Euclidean space.

its original form and use are unknown.

4.2. Alloy types

Despite the low sample frequency for the chronological periods, our results indicate systematic changes in alloy types in Jerash ([Fig. 11](#); see [Table 5](#) for the alloy type of all samples analysed). The Roman period samples are dominated by bronze, while the use of brass and leaded brass appears to be most pronounced during the Byzantine period. Leaded bronze is present from the late Roman to the Islamic times. Meanwhile, the occurrence of copper and leaded copper appears to increase from the Byzantine period onwards. Below, the alloy types are discussed in connection to their chronological distribution.

4.2.1. Copper & leaded copper

Copper is present in the Byzantine and Umayyad periods in Jerash and absent from the Roman and Ayyubid/Mamluk periods. Tin, zinc, and lead are present at impurity levels as they could result from ore paragenesis, namely natural co-occurrence of minerals, making for a rather impure copper. The copper hook (J105) stands out by having somewhat higher zinc (3 wt%).

Leaded copper is absent from the Roman period, but it is potentially present from the late Roman period onwards as suggested by the vessel foot J35, and in the Byzantine period (J33.1), while the 2 remaining leaded copper objects are undated. Three high leaded objects with 16–17 wt% Pb suggest the production of leaded copper by mixing fresh or scrap copper with rather large and possibly controlled additions of lead. Object J33.1 with 6 wt% Pb contains no detectable zinc and tin, also suggesting that it was the result of mixing copper and lead, albeit in smaller proportions than for the high leaded objects noted above. Zinc concentrations of 1.5 wt% in J9 and J29 could reflect mixing of brass alloys or the primary ores used.

4.2.2. Bronze & leaded bronze

All 5 Roman objects, 1 Byzantine (J38) and 2 Umayyad objects (J57 and composite J59) consist of bronze. Meanwhile, 11 leaded bronze samples occur in all periods from the Byzantine (or late Roman) to the Islamic periods. The copper-tin ratios in both bronze and leaded bronze are statistically identical, namely 16.5 (± 6.3 2SE) for bronzes and 19.2 (± 4.2 2SE) for leaded bronzes. Given the individual copper-tin ratio distributions for the two alloy types the production of leaded bronze by mixing of bronze with lead or the primary mixing of the three distinct alloying components are both possible. The preferential

oxidation of tin over copper during remelting of bronze as observed in working of bronze alloys with Iron Age open crucibles due to the different oxidation degree between copper and tin ([Figueiredo et al., 2010](#); [Frohberg, 1994](#); [Klein and Hauptmann, 1999](#)), is not expected to have changed the copper-tin ratios significantly.

Zinc is present mostly at impurity levels (< 0.5 wt%), while four objects (J3, J50, J54, J63) from Byzantine and Islamic periods contain zinc between 1.5 and 3.5 wt%. These zinc concentrations are most probably the result of scrap brass re-use and are considered as rather unintentional as also observed elsewhere, while zinc uptake in copper during cementation can be lowered significantly by the presence of tin or lead ([Craddock, 1995](#); [Hook and Craddock, 1996](#), pp. 150–151).

4.2.3. Brass & leaded brass

Zinc levels in bronzes range between 12 wt% and 23 wt%, while most of the samples show zinc levels of 14–21 wt%. For producing brass with ~20 wt% Zn by cementation, operating temperatures of around or slightly below 1000 °C ([Craddock, 1979](#), p. 70; [Rehren, 1999](#), p. 1085; [Werner, 1970](#)) are needed, whereas lower temperatures will result in lower zinc uptake. A zinc range of 18–24 wt% is also prevalent in Roman bronzes ([Ponting, 2002a](#)). Variation in the zinc amounts and lower zinc values in bronzes can be the result of remelting of brass as ~10% of its original zinc content will be lost with every remelting and reworking, as further losses can happen during metalworking and heat treatment of brass objects as well ([Caley, 1964](#), p. 99; [Ponting, 2002a](#), p. 559). These estimates are based on industrial zinc losses ([Bassett, 1912](#)) and higher losses could have well taken place in pre-industrial metal workshops.

Brasses (including composite object J59) are characterised by overall low impurities. Lead and tin concentrations do not exceed 1.5 and 1 wt% respectively ([Fig. 4](#)), and are typical of copper ores such as found in the nearby deposits at Faynan in southern Jordan ([Hauptmann, 2007](#)) or zinc ores, such as calamine ([Bourgarit and Thomas, 2015](#); [Craddock, 1998](#); [Pollard and Heron, 2008](#)). It is also possible that lead was contributed along with zinc during sublimation of sphalerite ([Barnes, 1973](#); [Carradice and Cowell, 1987](#); [Craddock et al., 1980](#)). Two Byzantine brass objects (J4, J7) with ~1 wt% Sn suggest mixing with bronze scrap alloys. All arsenic-nickel groups ([Fig. 6](#)) are present in bronzes, including all of the 'As / no Ni' samples.

Leaded brass, as with the unleaded type, appears in the Byzantine (or late Roman) period. Some objects such as those in the box group indicate the use of leaded brass in the early Islamic period in Jerash. However, artefacts of leaded brass that were for sure produced during

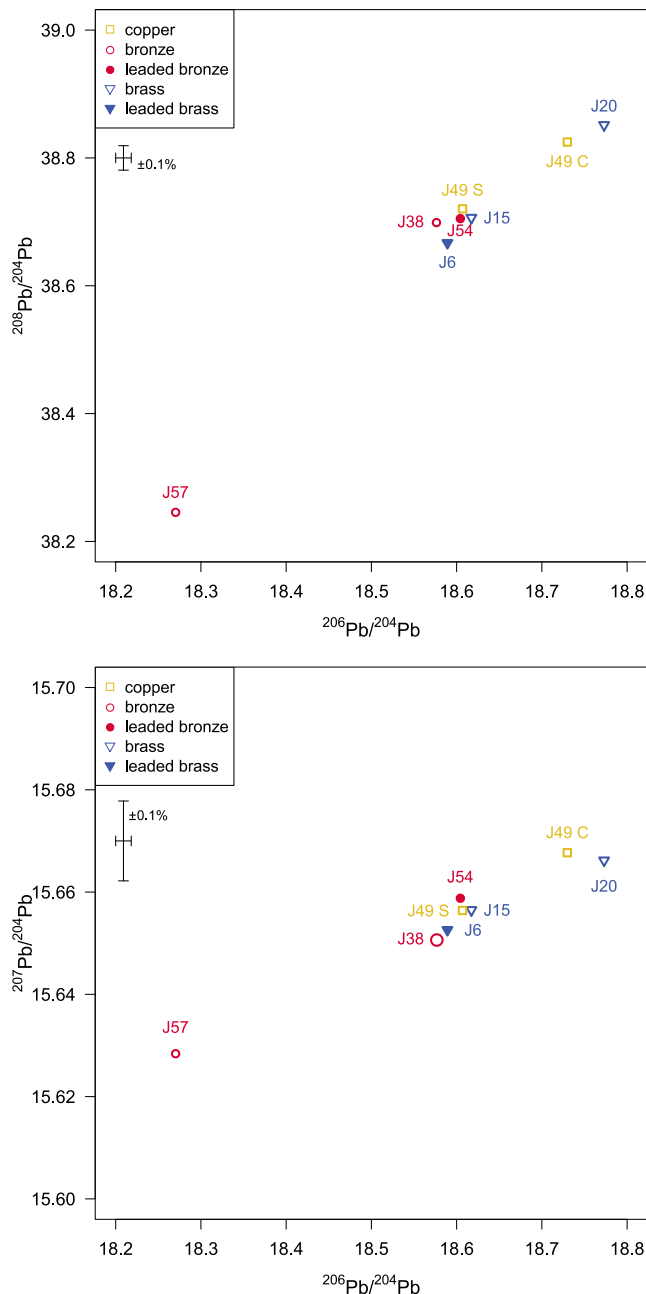


Fig. 7. Lead isotope biplots of $^{208}\text{Pb}/^{204}\text{Pb}$ - $^{206}\text{Pb}/^{204}\text{Pb}$ (top) and $^{207}\text{Pb}/^{204}\text{Pb}$ - $^{206}\text{Pb}/^{204}\text{Pb}$ (bottom) for copper-based objects from Jerash.

Table 4

Summary of metalworking techniques and alloy types (drilled samples are excluded).

	Copper	Bronze	Leaded bronze	Brass	Leaded brass	Total
Cast	1	3	7			11
Hammered	6	3	2	9	1	21
Total	7	6	9	9	1	32

the Umayyad period are missing from the contexts found until now in the Northwest Quarter of Jerash. Lead additions in brass are lower (3–8 wt%) compared to the leaded bronze or leaded copper types. A zinc range of 5–9 wt% is also consistently lower than in the brasses. Lower zinc values, compared to brass (12–23 wt%), can result from re-melting brass in (partly) oxidising conditions without infusing it with

fresh zinc vapour, as brass tends to lose some of its original volume with each re-melting (Hook and Craddock, 1996, p. 151). Additionally, lower zinc values in brass can also vary depending on the purity of copper used during cementation, as lead (and tin) impurities in copper can inhibit the uptake of zinc by copper during cementation (Craddock et al., 1980). Thus, in the case that leaded copper was used the resulting leaded brass would have a lower zinc content compared to a brass made with pure copper. Tin in leaded brass is again at impurity levels and compares to the tin concentrations in brass. This supports further the re-melting and mixing of brass with lead by intentionally avoiding the mixing of other metals or alloys, such as bronze, that would result in more varying quaternary (four-part) compositions.

Brasses and leaded brasses in Jerash show a higher iron content compared to the rest of the copper-based alloys analysed. On average 0.25 wt% iron is found in the (leaded) brasses, as opposed to 0.15 wt% for the rest of the samples as analysed with the μXRF , a pattern also observed in the EPMA data set too. Higher traces of iron have often been associated with the use of carbonate zinc ore, namely calamine or smithsonite, which unlike sphalerite can be used without sublimation and, thus, any iron impurities in the ore would be passed down to the metal product (Craddock et al., 1998; Craddock et al., 1980; Pollard and Heron, 2008; Ponting and Segal, 1998, p. 117). Finally, nickel was detected in the 2 leaded brass samples analysed by EPMA and not in other brasses, though no correlation between the lead and nickel contents was found.

4.3. Copper-based metalwork from the Roman to early Islamic periods in Jerash

The investigation of copper alloys recovered from the Northwest Quarter in Jerash reveal time-dependent patterns that offer new insights for the city's technological organisation. Below, we consider the alloys' technological characteristics in a chronological framework. Undated or chronologically weakly constrained objects (J16, J17, J22, J35) are excluded from this discussion.

4.3.1. Roman period

Samples dating to the Roman period are few (5 objects), however, it is worth noting that they comprise the majority of bronze objects in this study, while only 3 bronze samples date to later periods (Fig. 11). The finding of principally bronzes for the Roman period is noteworthy as brass was an established alloy by that time (Bayley, 1984). However, still in the Roman period, brass was reserved for specific artefact types such as coinage and military equipment (Bayley, 1998) or other decorative pieces (Ponting and Levene, 2015) not included in the sample here, which comprises needles, a nail and a simple pin. As seen in Fig. 12, Roman bronzes stand out from later ones due to their lower tin (4–6 wt% in 4 samples), and higher lead (2 wt% in 3 samples), as later bronzes contain ~10 wt% Sn and < 1 wt% Pb.

Even though we cannot determine the source of this lead, e.g. mixing of scrap leaded copper (exogenous) or lead-rich copper ores (endogenous), it is worth noting that lead-rich copper was produced at Faynan, often with lead values up to 2 wt% (Hauptmann, 2007, p. 201). Given the lead isotope results, if the provenance of Roman bronze J49 is indeed Cypriot (Table 3), then the case of added lead should be considered as Cypriot copper ores contain only low traces of traces (Constantinou, 1982). Our results for Jerash indicate a preference for the well-established copper-tin alloys of the Roman period. Even though it was the Romans who introduced brass technology, brass was more widely adopted in later periods, most notably from the 6th century CE onwards (Bayley, 1998; Brüggler et al., 2012; Craddock et al., 1998; Hanel and Bode, 2016). Thus, the predominance of bronzes at Roman Jerash is not surprising.

4.3.2. Byzantine period

Results for the Byzantine period suggest changes in alloying

Table 5
Summary table of microscopic, elemental (μ XRF, EPMA) and isotopic results.

Sample ID	Chronological period ¹	Alloy type ²	Metalworking ³	Provenance indications ⁴	Ni-As groups ⁵
J1	Byz/Uma	Leaded bronze	–	–	no Ni / As
J2	Byz/Uma	Leaded bronze	Cast	–	Ni, no As
J3	Byz/Uma	Leaded bronze	Cast	–	Ni, no As
J4	Byz	Brass	Hammered	–	As, no Ni
J5	Byz	Leaded bronze	Cast	–	As / Ni correlated
J6	Byz	Leaded brass	Hammered	Massif Central	Ni, no As
J7	Byz	Brass	Hammered	–	Ni, no As
J8	Byz	Brass	–	–	As, no Ni
J9	–	Leaded copper	–	–	–
J10	Ayy/Mam	Leaded bronze	Cast	–	no Ni / As
J11	–	Leaded bronze	–	–	–
J12	–	Brass	Hammered	–	–
J13	Byz	Copper	–	–	Ni, no As
J14	–	Leaded bronze	Cast	–	–
J15	Byz	Brass	–	Massif Central	no Ni / As
J16	–	Leaded brass	–	–	–
J17	–	Copper	Hammered	–	–
J18	Byz	Copper	Hammered	–	Ni, no As
J20A	Byz	Brass	–	Cyprus	Ni, no As
J20B	Byz	Brass	–	–	Ni, no As
J21	Byz	Brass	–	–	Ni, no As
J22	–	Leaded copper	–	–	–
J24	Byz	Leaded brass	–	–	As / Ni correlated
J28	Rom/Byz	Leaded brass	–	–	–
J29	Uma/Mam	Leaded copper	–	–	–
J33.1	Byz	Leaded copper	–	–	no Ni / As
J33.2	Byz	Brass	Hammered	–	no Ni / As
J34	–	Brass	Hammered	–	–
J35	Rom/Byz	Leaded copper	–	–	–
J36	Byz	Brass	Hammered	–	no Ni / As
J37	Uma	Leaded copper	–	–	no Ni / As
J38	Byz	Bronze	–	Massif Central	–
J39	Rom	Bronze	Hammered	–	no Ni / As
J40	Rom	Bronze	Hammered	–	no Ni / As
J41	Uma	Copper	Hammered	–	no Ni / As
J42	Rom	Bronze	Cast	–	no Ni / As
J44	Rom	Bronze	Cast	–	–
J45	Uma	Copper	Hammered	–	Ni, no As
J47	Rom/Byz	Leaded bronze	Hammered	–	–
J49	Rom	Bronze	–	Cyprus	no Ni / As
J50	Byz/Uma	Leaded bronze	Hammered	–	Ni, no As
J51	Byz/Uma	Brass	Hammered	–	As / Ni correlated
J52	Uma	Copper	Hammered	–	As / Ni correlated
J54	Uma	Leaded bronze	Cast	Romania/Bulgaria	As / Ni correlated
J57	Uma	Bronze	Hammered	Timna	Ni, no As
J59A	Uma	Brass	Hammered	–	Ni, no As
J59B	Uma	Bronze	Cast	–	As / Ni correlated
J62	Byz/Uma	Brass	Hammered	–	As, no Ni
J63	Uma/Mam	Leaded bronze	Cast	–	As / Ni correlated
J64	Byz/Uma	Copper	Hammered	–	As / Ni correlated
J105	–	Copper	Cast	–	–

¹ Chronological period grouping used in Fig. 11. Rom = Roman, Byz = Byzantine, Uma = Umayyad, Ayy = Ayyubid, Mam = Mamluk, dash (–) indicates undefined or too broad period.

² Alloy types attributed based on μ XRF and EPMA results shown in Figs. 3–5.

³ Dash (–) indicates drilled samples in which microscopic observation could not be performed.

⁴ Provenance after results shown in Table 3; dash (–) for samples not analysed.

⁵ Ni-As groups after results shown in Fig. 6, as analysed with EPMA; dash (–) for samples not analysed with EPMA.

practices, while all arsenic-nickel sub-groups are present in the Byzantine samples analysed with EPMA (Fig. 6). A gradual increase in arsenic accompanied often by a relative increase in nickel over time from the 1st century CE has been also observed by Hook and Craddock (1996, p. 152) possibly reflecting changes in smelting technology. Arsenic and nickel are both associated with copper and the above pattern suggests the introduction of copper with different trace element characteristics in the analysed assemblage and, thus, with a possible different origin or processing.

Several alloy types seem to be introduced in the sample no earlier than the late Roman period and certainly in the Byzantine period, including binary brass and the whole range of leaded alloy types, i.e.

leaded copper, leaded bronze and leaded brass (Fig. 11). Even though bronze was still present in the post-Roman periods, it seems to be used on a smaller scale, with only 1 Byzantine and 2 Umayyad bronze objects in the sample. As previously noted, the tin content of Byzantine and Umayyad bronzes is around double that of the Roman bronzes (Fig. 12). Unalloyed copper is present both in the Byzantine and Islamic periods, but Byzantine copper objects have fewer impurities.

Both brass and leaded brass emerge in the Byzantine period, while only 1 brass Umayyad object was found the sample. Meanwhile the use of old, namely Byzantine, metal in the Islamic period cannot be excluded as the hoarded metal finds (box group) in a house destroyed by the 749 CE earthquake indicate. Brass in the Byzantine and possibly the

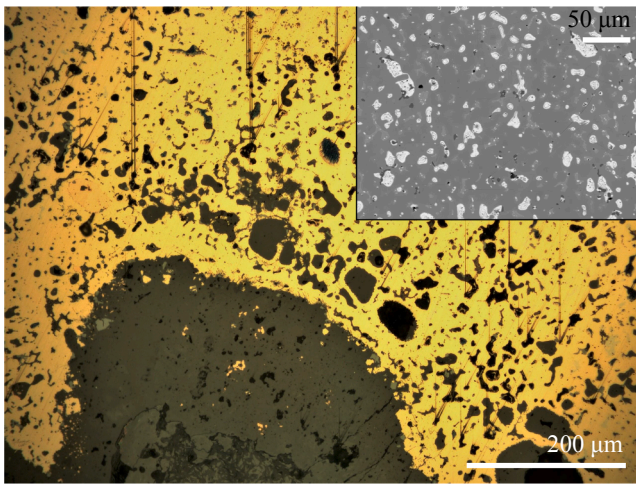


Fig. 8. Reflected light and back-scattered images of J10 showing a dense interconnected dendritic structure abundant in vesicles of varying size.

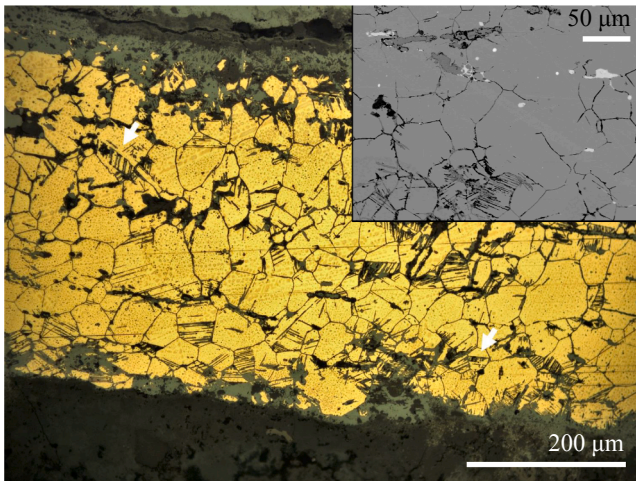


Fig. 9. Reflected light and back-scattered images of J4 showing a recrystallised metal structure forming polygonal and angular grains often with annealing twins (white arrows).

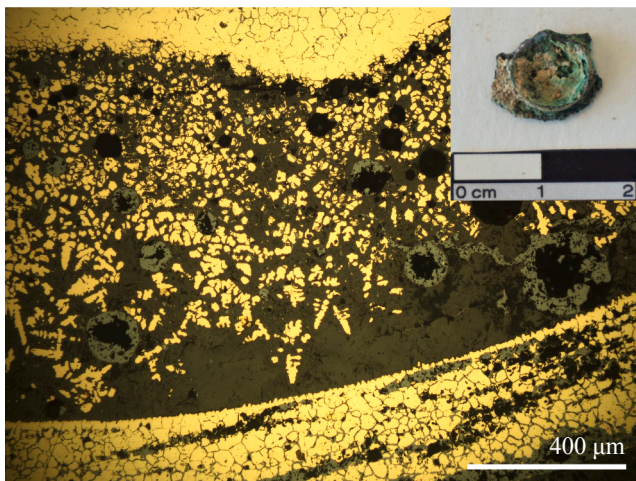


Fig. 10. Photograph (top right) and reflected light image of J59 showing a cast (dendritic) bronze structure surrounded by a hammered brass sheet with a characteristic recrystallised structure of polygonal grains.

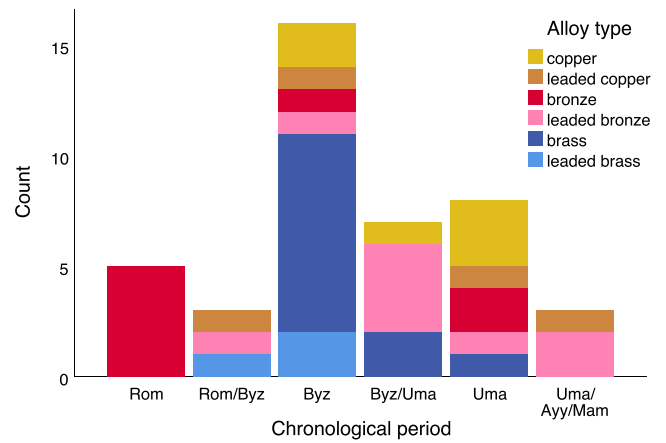


Fig. 11. Chronological distribution of alloy types; Rom = Roman, Byz = Byzantine, Uma = Umayyad, Ayy = Ayyubid, Mam = Mamluk.

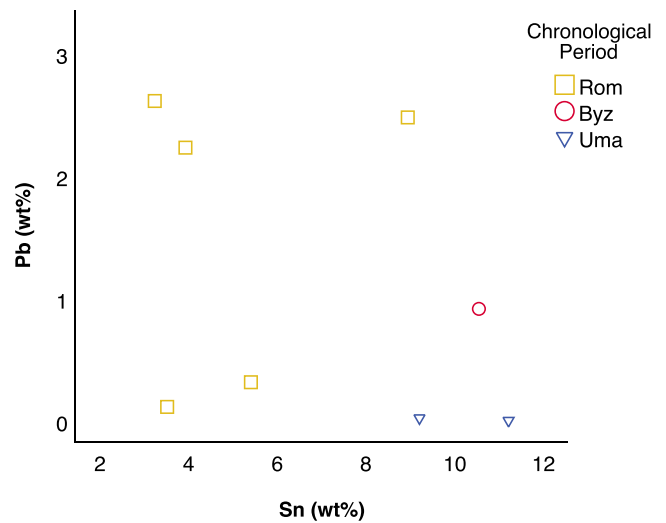


Fig. 12. Co-variation of lead and tin in bronzes for Roman, Byzantine and Umayyad periods as analysed with μXRF, where lower tin and higher lead contents are noted for the Roman period on average compared to the Byzantine and Umayyad periods.

early Umayyad periods is characterised by low impurities and a standardised zinc content. Similarly, leaded brasses mark the late Roman/Byzantine or Byzantine periods, while they are absent from later phases.

The presence of brass at Jerash suggests access to the products of a technology that requires a specific technological know-how regarding both brass-making (cementation) and reusing (recycling, remelting, mixing) that would not have been available to non-specialist craftspeople even if these workshops did not operate within the city. The presence of brass in a rather confined chronological period in the Byzantine (or late Roman) phase raises questions about the circumstances of its presence in Jerash. Being part of the eastern Roman Empire, meant that Jerash was also part of a network with potential access to zinc sources in Asia Minor (DeJesus, 1980) that could have sustained the zinc ore supply for the production of brass in the Near East. Proximity to such metal sources, for instance, could account for the majority of the Byzantine copper-based objects from Sardis being brasses (Rapp, 1983), especially as Anatolian tin sources would have been possibly quite limited and, thus, of little significance (Cierny et al., 2003). It is worth noting that both major tin sources in Europe and central Asia would have been outside the immediate sphere of influence of Eastern Roman Empire during the Byzantine period.

Currently, there is no evidence to support the local production or casting of brass in Jerash as no remains related to a metal workshop, such as lidded crucibles or moulds, have been recovered so far. Casting remains have been reported from the Roman period (Khalil et al., 2012), but not for later periods. Finished objects could have been produced elsewhere and then brought along with itinerant individuals or imported by (long-distance) trade to the city. The military might have contributed to the spread of brass objects in Byzantine Jerash. In Roman times, *militaria* (military weapons, equipment, etc.) were often made of brass, as well as various objects and vessels for civilian use (Bayley, 1998; Brüggler et al., 2012; Hamilton, 1996, p. 1; Ponting and Segal, 1998; Ponting, 2002a, 2002b). Unfortunately, detailed metallurgical analyses of Byzantine *militaria* are not available. However, analyses of various Byzantine objects shows the use of brass amongst other alloy types (Ponting, 1999). The Northwest Quarter yielded at least 2 pieces, namely belt tongue J18 and press model for belt fitting J13, which even though made of copper could have been used by Byzantine officials or military. The presence of Byzantine troops in Jerash during the second half of the 6th century is also attested by a mosaic inscription (Haensch et al., 2016).

The Byzantine period in Jerash represents a time of experimentation and enrichment of the copper alloy palette. On the one hand, as this study indicates the traditional Roman alloy, i.e. bronze, ceased to be used, while, on the other hand, the working of a much wider range of metals and alloys emerges including the specialised brass technology along with the frequent use of leaded alloys. Finally, analyses from Jerash stand out for the prolific use of low-impurity zinc-rich brass with a suggested European origin, as opposed to lead / tin-containing assemblages from surrounding regions or absence of tin from distant areas such as the Arabian Peninsula.

4.3.3. Umayyad and Ayyubid/Mamluk periods

Analyses of Islamic objects show a range of alloys. Copper and leaded copper are still in use. Bronze for the Islamic period shows a higher, more standardised tin content at 10 wt% compared to the smaller, more variable tin amounts in Roman bronze (Fig. 12). Thus, Islamic bronze emerges as the result of different technological choices, while conclusions on this are tentative due to the small number of Islamic bronzes available in this study. Leaded bronze which is present from the late Roman/Byzantine period seems to become much more widespread in the Islamic periods with 6 objects dated in the Umayyad (or late Byzantine/Umayyad) and Ayyubid/Mamluk periods. Finally, brass almost discontinues in the analysed sample except for the composite bronze-brass Umayyad object J59. As with the Byzantine samples, all arsenic-nickel sub-groups are present in the early Islamic phase and in the Umayyad period in particular.

The picture emerging from Jerash is comparable to Skythopolis (modern Beit She'an), also an ancient Decapolis city, where leaded bronze characterised the Umayyad period followed by brass during the Byzantine period (Ponting, 1999, p. 1319). Meanwhile, the vast replacement of brass by bronze during the Islamic period in Jordan has been noted before (Al-Saa'd, 2000). Tin is not locally available in Jordan and it would have to come from elsewhere such as Europe and / or central / eastern Asia (Moorey, 1994; Yener and Özalp, 1987). Thus, both Decapolis cities had access to a metal / alloy that would have only been available via a long distance network in the early Islamic period, possibly the silk road (Ponting, 1999, p. 1319). Mixing of old Roman bronzes could have contributed to the re-emergence of bronze as a dominant alloy in the Islamic period (Khamis, pers. comm. in: Ponting, 1999, p. 1320). Finally, Islamic period bronzes have been found elsewhere such as at Umm Qais to the north of Jerash (Arafat et al., 2013), thus raising additional questions about the technological choices emerging from the material at Islamic period Jerash.

Umayyad (or late Byzantine) utensil J51 and pin J62 were the only samples with detectable antimony up to 0.1 wt% (Supplementary Material E – Table S5). Even though their chronology is suggestive of a

technological change in the transition from the Byzantine to the Islamic phase of the site, further work is needed to demonstrate this.

4.4. The box group and the recycling issue at Jerash

Analyses of 14 copper-based objects from the scrap metal group found in a wooden box in a domestic context (the box group) dating from the Roman to the Byzantine periods, with the noticeable absence of Umayyad objects, showed the absence of tin-containing alloys as copper, brass, and their leaded counterparts dominate the group. The hoarding of these objects suggests that they were recognised by the city's inhabitants for their potential re-use showing a general appreciation for old metals, though other culturally led reasons for this small hoard may apply. Similar, though larger in scale, hoarding of copper-based objects in three ceramic vessels has been found in 11th century CE Tiberias (Fatimid period), where the majority of the object were of brass too (Ponting, 2008). Absence of leaded bronze objects from the box group would be justified if they were still in circulation in the Umayyad period as this alloy has been shown to be characteristic of late Byzantine and Umayyad periods onwards in Jerash, and elsewhere in Jordan (Al-Saa'd, 2000). The absence of unleaded bronze objects from the box group which, as discussed above, were the preferred alloy of the Roman period could be attributed either to the circumstantial exclusion of bronze from the small hoard or, less probably, the continued use of bronze objects in the Umayyad period.

Neither arsenic nor iron point to re-use as their levels are in accordance with the rest of the assemblage. As arsenic and iron tend to decrease with re-melting (recycling/mixing), they are often used as signifiers of distinct technological choices (Bray et al., 2015; Pernicka, 1999; Tylecote et al., 1977). Three of four arsenic-nickel sub-groups (Fig. 6) are present in the box group, the 'As, no Ni' group is absent.

However, this is not to say that re-use or mixing of copper and copper alloys did not take place as it has been also shown that efficient metal recycling is not always detectable and, thus, much more common than often thought (Ponting and Levene, 2015). Our analyses present evidence to suggest that certain objects were the result of copper and copper alloy mixing as noted in comparable assemblages from the region such as from Tiberias (Ponting and Levene, 2015; Ponting, 2008). For example, leaded brass in the analysed sample seems to be a version of the contemporary brass diluted with lead, though the possibility for the use of leaded copper during cementation cannot be excluded. The lead content is higher due to its intentional addition (or mixing with leaded copper) while the brass content drops from 12 to 23 wt% in binary bronzes to 5–9 wt% in the leaded alloy. This drop in the zinc content goes beyond issues of mass balance and suggests the loss of the element as vapour. Occasional higher values of tin, zinc and lead in copper group could result from re-use of scrap bronze / brass (leaded or unleaded), such as suggested by the erratic distribution of low values, thus also pointing to mixing of metals.

The arsenic-nickel sub-groups are possible indications of the use of different copper sources. The detection of arsenic and nickel from the Byzantine period onwards and into the early Islamic phase, though acknowledging the few Roman samples ($n = 4$), suggests that a range of copper sources were used for the metalwork present at Jerash's Northwest Quarter from the 6th century CE onwards. Except for the second group with arsenic detected but no nickel which is confined to bronzes, the rest of the arsenic-nickel sub-groups are found in all alloy types present in the EPMA sample. This possibly suggests that same source copper was used for a range of copper-based alloys. The fact that some samples contain nickel or arsenic and others both or none, shows that the copper-alloys analysed here were, if recycled, not recycled to the degree that a homogeneous copper impurity pool was formed.

4.5. Provenance indications of materials

Lead isotope ratios reflect the formation age of the ores from which

the metal derived, which can be used to distinguish between potential geological sources of the metal for provenancing purposes. Seven samples representative of all the studied periods were selected to preliminarily investigate the potential sources of copper or lead metal to test if these displayed any consistency chronologically: 4 Byzantine brasses (1 leaded), 2 Roman bronzes and 3 Umayyad bronzes (2 leaded). Caution is needed when interpreting lead isotope results from brasses, as the lead could derive from the zinc-based ore (calamine), the copper ore, or lead metal being added (cf. Merkel, 2018); the high lead level in J6 indicates that lead in this sample should be considered an addition.

The measurable difference in the lead isotope ratios between the two independent samples from the surface and the core of the Roman bronze pin (J49) shows the effect of external lead contamination on copper-based alloys due to post-depositional processes and highlights the importance of sampling pristine metal from the core of an artefact. Whilst this may be less of a problem for pure lead artefacts (Rehren and Prange, 1998), our findings show that there can be differences in lead isotope composition in copper alloys between the surface and the core.

The Pb-Pb model ages were calculated using Stacey and Kramers' (1975) two-stage model to identify potential geological sources of the ores used in Jerash. However, the significance of the model ages must be tempered by clear indications discussed above of anthropogenic mixing in the chemistry of the Jerash artefacts. Here, we compare our new lead isotope data to lead and copper ores from the Mediterranean, Central and Western Europe, North Africa, and more local sources in the Arabah (Jordan, Israel) and the Arabian shield (Saudi Arabia, Egypt, Yemen, Oman). The nearest reference data points (Euclidean neighbours) were calculated for each artefact as outlined elsewhere (Birch et al., 2019b; Stos-Gale and Gale, 2009). The combination of Pb-Pb model ages and nearest Euclidean neighbours provides an indication of the most relevant geological sources for hypothesising provenance.

The calculated Pb-Pb model ages correspond to geological ages between 28 and 326 Ma, with the exception of one sample with a model age of 821 Ma (Table 3). The results show high linearity in their $^{208}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ values (Fig. 7, $r^2 = 0.99$).

4.5.1. Massif Central/Alpine ores

Three of the objects

(J6, J15 and J38) have similar model ages of 137 Ma, 124 Ma and 143 Ma respectively, and from Euclidean neighbour analysis suggest that the ore could have come from the Massif Central in Western Europe (Table 3, Fig. 13). One copper ore sample from Oman features amongst the nearest Euclidean neighbours for J6, J15 and J38 and cannot be excluded as potential source. However, the lack of mining evidence in Oman between the Iron Age and early Islamic period (Begemann et al., 2010) should be taken into consideration. In contrast, the archaeological and historical evidence attest to long-term mining activities in the Massif Central.

For the late Byzantine brass sheet (J6) with 3.5 wt% Pb both lead and copper ores should be considered as lead could be exogenous. Even when both lead and copper ores are considered, the nearest Euclidean neighbours are almost exclusively copper ores from the Valais in Switzerland (Cattin, 2008; Cattin et al., 2011) or the Massif Central (Baron et al., 2006); Roman mining activity is well known from the Cévennes (Domergue, 2008). Similar results were obtained for J15 and J38. The late Byzantine brass weight (J15) contains around 0.5 wt% lead that could derive from either the copper or zinc (secondary lead phases in calamine). The lead isotope ratios are most consistent with copper ores from the Cretaceous sediments of the Massif Central in France (Baron et al., 2006; Brevart et al., 1982) or Alpine (Austria/Switzerland) copper deposits (Cattin, 2008; Höppner et al., 2005). The early Byzantine bronze sheet (J38) with 0.9 wt% lead has the same nearest Euclidean neighbours as J15.

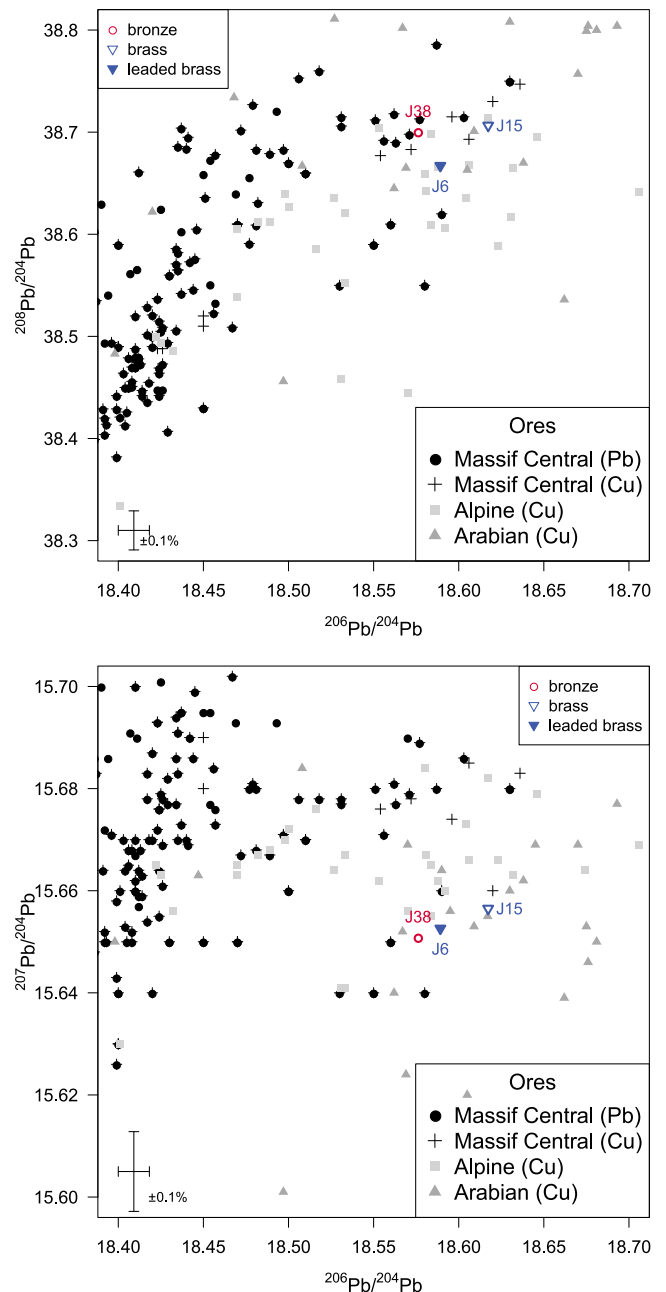


Fig. 13. Lead isotope biplots of $^{208}\text{Pb}/^{204}\text{Pb}$ - $^{206}\text{Pb}/^{204}\text{Pb}$ (top) and $^{207}\text{Pb}/^{204}\text{Pb}$ - $^{206}\text{Pb}/^{204}\text{Pb}$ (bottom) comparing J6, J15 and J38 with ores from the Massif Central (Baron et al., 2006; Brevart et al., 1982; Orgeval et al., 2000; Charef, 1986; Le Guen et al., 1991), Alps (Cattin, 2008), Arabah and Arabian shield (shown together as 'Arabian') (Hauptmann, 2007; Stacey et al., 1980; Begemann et al., 2010).

4.5.2. Cyprus ores

The late Byzantine brass ring with attached key (J20A) dates to the Alpine orogeny (28 Ma; Table 3). The nearest Euclidean neighbours are almost exclusively copper ores from Sardinia, specifically those from Castello di Bonvei (Stos-Gale et al., 1997) (Fig. 14). However the scale of copper production in Sardinia appears to be small, with much of the copper being imported from Cyprus (Kassianidou, 2001; Lo Schiavo, 2005; Lo Schiavo et al., 2005). If Sardinia is excluded, the next nearest copper ores come from Cyprus (Stos-Gale et al., 1997), for which there is ample evidence of copper production throughout antiquity.

When only the result from the Roman bronze spindle's (J49) core is considered, the same Sardinian copper ores as for J20A are the nearest

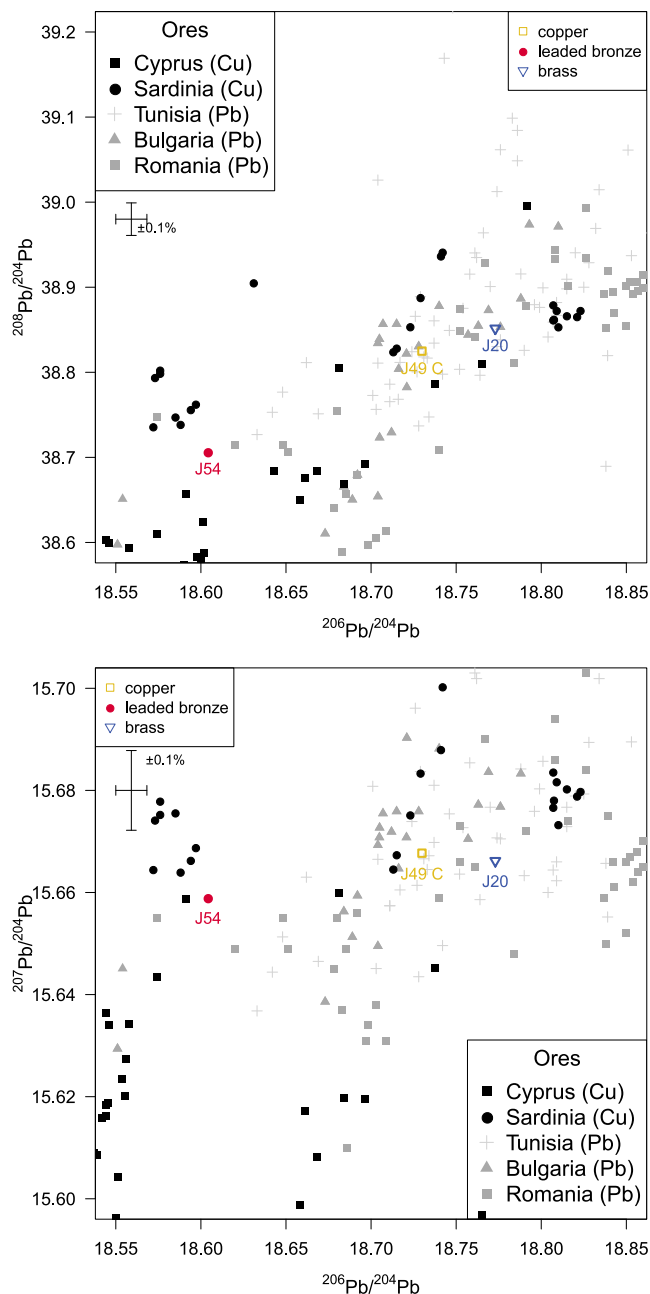


Fig. 14. Lead isotope biplots of $^{208}\text{Pb}/^{204}\text{Pb}$ - $^{206}\text{Pb}/^{204}\text{Pb}$ (top) and $^{207}\text{Pb}/^{204}\text{Pb}$ - $^{206}\text{Pb}/^{204}\text{Pb}$ (bottom) comparing J20A, J49 and J54 to ores from Cyprus (Gale et al. 1997; Stos-Gale et al. 1997), Sardinia (Stos-Gale et al. 1997), Tunisia (Skaggs et al. 2012), Bulgaria (Kouzmanov 2001; Stos-Gale et al. 1998) and Romania (Marcoux et al. 2002).

Euclidean neighbours which also shares the same Alpine geological age (Fig. 14). However, if the lead is exogenous, the ratios are consistent with lead mineralisations from both Tunisia (Skaggs et al., 2012) and Bulgaria (Stos-Gale et al., 1998).

4.5.3. Eastern Europe ores

The lead concentration of the Umayyad leaded bronze needle (J54) is 6 wt% and should be considered exogenous. Two separate fractions for this sample yielded near identical lead isotope compositions (relative difference of 1–4% of lead isotope ratios), which are on the threshold of the 2σ measurement error, yielding a lead model age range of 135–144 Ma (Table 3). When only lead related sources are considered, the lead isotope ratios of both J54 analyses are consistent with

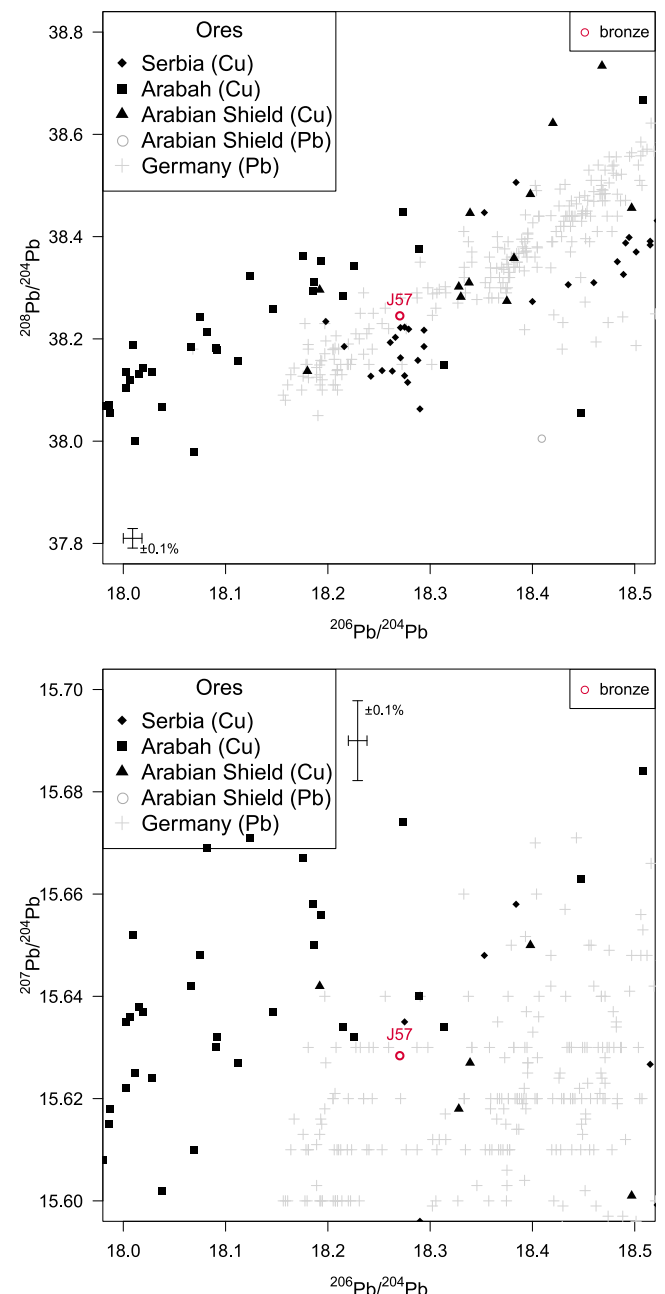


Fig. 15. Lead isotope biplots of $^{208}\text{Pb}/^{204}\text{Pb}$ - $^{206}\text{Pb}/^{204}\text{Pb}$ (top) and $^{207}\text{Pb}/^{204}\text{Pb}$ - $^{206}\text{Pb}/^{204}\text{Pb}$ (bottom) comparing J57 to ores from Serbia (Pernicka et al. 2009), Arabah (Timna) (Hauptmann, 2007), Arabian shield (Stacey et al., 1980; Begemann et al., 2010) and Germany (Bode, 2008; Bode et al., 2009; Gottschalk and Baumann, 2001; Harms et al., 2012; Schneider, 1999; Kimbauer et al., 2012).

ores from Tunisia (Skaggs et al., 2012), the Southern Apuseni mountains of the Romanian Carpathians (Marcoux et al., 2002), the Burgas district in eastern Bulgaria (Stos-Gale et al., 1998) and the Taunus in Germany (Kimbauer et al., 2012) (Fig. 14). The lack of evidence for pre-medieval mining means the Taunus should be disregarded. Romania and Bulgaria appear to be more prolific in the nearest Euclidean neighbours and are consistent with a Mesozoic age, indicating a potential Eastern European provenance for the exogenous lead in the bronze (Fig. 14).

Fig. 15 shows that the closest Euclidean neighbour to the bronze Umayyad pin (J57) is a copper ore from Aljin Do, Serbia (Pernicka et al., 1993). With the increase in evidence for Roman metallurgical

activity in this region up to the 6th century CE (Petković, 2009), this cannot be excluded as a potential ore source. The next two closest copper ores are from Timna (see Arabah in Fig. 15) (Hauptmann, 2007). Timna should be considered a candidate source of the copper used, as there is evidence for copper exploitation during the early Islamic period (7th and 8th centuries CE) such as the smelting sites of Be'er Ora and Khirbat al-Mana'iyya (Avner and Magness, 1998; Ben-Yosef, 2012; Jones et al., 2017; Rothenberg, 1988; Willies, 1991) (Fig. 1), as well as at other sites in the Wadi Arabah (Weisgerber, 2006) and Wadi Faynan (Hauptmann, 2007). Use of Arabah copper during the Islamic period in Jordan has been suggested previously by lead isotope analyses of a small group of everyday objects (Al-Saa'd, 2000).

4.5.4. Summary of provenance observations

The lead isotope results from Jerash exhibit a wide range of model ages corresponding with different geological ore deposits. The results indicate that copper may have been obtained from as near as Timna, or as far as Sardinia or the Massif Central/Alpine copper ore deposits. Similarly, added lead metal may have derived from the Carpathians. Although the lead isotope results from Jerash copper-alloys can directly be compared to published lead and copper ore data, hypotheses regarding the provenance are problematic due to potential re-use of metal and subsequent mixing of lead isotope ratios. Despite this limitation, the results are significant, as they confirm that a multitude of metal sources were utilised from the Roman through to the Umayyad periods.

Roman J49, and Byzantine J6, J20A and J38 showed a suggested western origin for the copper from Cyprus or the Massif Central, while Umayyad J57 has a suggested copper provenance from Timna (Table 3). A Near Eastern provenance for the Umayyad metals reinforces a hypothesis for an Eastern-looking supply network for the Islamic caliphates, as opposed to the more western / European outlook of the Roman and Byzantine Empires. Given the present data this is discussed as a suggestion as the analyses of additional Islamic period metals is needed, but it also ties with recently presented evidence for the exploitation of Timna copper ores in the early Islamic period (Ben-Yosef, 2012) and previous analyses of Islamic copper alloys from Jordan (Al-Saa'd, 2000, p. 395, Fig. 5).

Further still, the lead isotope results may provide useful information about long term re-use of the same metals. For example, the high linearity of the results from different copper alloys (J6, J15, J38) might indicate recycling of locally circulating copper-based metal / alloys, explaining the consistency in lead isotope ratios from the early through to the late Byzantine period. One of the most significant findings presented here is from J49, which shows the potential effects of lead contamination of surface samples, stressing the importance of unadulterated metal samples for lead isotope analyses.

5. Conclusion

Combined elemental, isotopic and metallographic examination of copper-based metalwork from the Northwest Quarter in Jerash (Table 5) revealed characteristics of the technology to which the city had access, as well as patterns of the diachronic development of the alloying practices from the Roman to the early Islamic periods. Jerash throughout its occupation had access to polymetallic copper-based objects. However, clear indications for a local production and with it a sophisticated knowledge of polymetallic copper-based technology are missing in the archaeological record of the Northwest Quarter. The relatively small number of samples that cover a wide chronological range renders any conclusions tentative. Elemental analyses of copper-based objects from Jerash showed the presence of copper, bronze, brass and their leaded counterparts, and revealed diachronic trends in the preference of certain alloy types. A low to medium tin bronze dominated the Roman period and (leaded) brass the Byzantine period. A return to a tin-containing alloy marks the Umayyad and Ayyubid/Mamluk periods during which leaded bronze emerges as the preferred

alloy, as also noted elsewhere in the region and in the Decapolis (Skythopolis) in particular. Unalloyed and leaded copper are present throughout the periods and point to the sustained need for a cheap functional metal with no enhanced mechanical or aesthetic properties. Binary alloy types in Jerash suggest well-controlled alloying choices as seen in the quite homogenous compositional groups of Roman bronze, Byzantine brass and leaded copper diachronically. The hoarding of old metal suggests that early Islamic occupants of the Northwest Quarter valued such pieces and were keen to make full use of the resources available to them, even if the remelting of scrap alloys did not take place on site. This ties in well to a general trend in Jerash as in the early Islamic period the production of glass saw an increase in glass re-use, compared to earlier periods (Barfod et al., 2018). The increase of recycling possibly relates to, on the one hand, shifting trade networks and, on the other hand, to an increase in local demand.

The above indicates technological experimentation, but also standardisation of alloying practices. Minor element compositions, namely arsenic, iron and nickel are remarkably uniform across chronological periods and most alloy types apart from arsenic-rich leaded copper or low-nickel Roman bronze. Provenance of copper suggests the western sourcing (Europe / Mediterranean) during the Roman and Byzantine periods, and a local Near Eastern provenance for Islamic metals. The western sourcing (Massif Central) during the Byzantine period is remarkable as Gaul was no longer under Imperial control from the second half of the 5th century CE onwards. This phenomenon still requires further attention and investigation of the long-distance trade of Byzantium with Visigoth and Frankish Gaul.

With respect to the local provenance of copper in Islamic times one must underline that Jordan had a long tradition of copper smelting and working due to the copper-rich Arabah Valley to the south, which was passed down throughout time. Though these results remain to be further tested by additional lead isotope analyses, they highlight Jerash's changing outlooks linked to subsequent administrations, namely the western-oriented Roman and Byzantine Empires and eastern-oriented Islamic caliphates possibly due to interruptions of established trade routes in the period during the Byzantine-Islamic conflicts.

Authors contributions

VO conducted the sampling and sample preparation, microscopic and μ XRF elemental analysis and preparations for EPMA analysis, drafted the text, including the main discussion and interpretation. GHB conducted the Pb isotope sample preparation, who together with CEL, guided the presentation of the geochemical data. TB contributed the interpretation and discussion of the Pb isotope results. AL and RR conceived the project, secured funding, and initiated and oversaw the selection of the objects and undertook – together with CE – the archaeological contextualisation of all items. CE assigned the artefacts to chronological periods. All authors contributed to drafts of the manuscript and approved the final version.

Declaration of Competing Interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jasrep.2020.102519>.

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