

Copper Metallurgy in the Jordan Valley from the Third to the First Millennia BC: Chemical, Metallographic and Lead Isotope Analyses of Artefacts from Pella

Graham Philip¹, Philip W. Clogg¹ and David Dungworth²
(with an appendix by Sophie Stos³)

¹ Department of Archaeology, University of Durham, The Dawson Building, South Road, Durham, DH1 3LE, UK

² Centre for Archaeology, English Heritage, Fort Cumberland, Portsmouth, PO4 9LD, UK

³ University of Oxford, Research Laboratory for Archaeology and History of Art, Keble Road, Oxford, OX1 3QJ, UK

This paper investigates Bronze and Iron Age metallurgy in the Levant while recognizing that metal artefacts were employed within a world in which social, economic and technological factors were closely intertwined. This theme is addressed through the investigation of sixty-eight copper-base artefacts from the site of Pella in the Jordan Valley, taken from contexts spanning the Early Bronze Age through to the early Iron Age. Data arising from a combination of chemical, metallographic and lead isotope analyses are considered in terms of artefact typology, manufacturing techniques and archaeological context, and their social and economic implications for ancient metallurgy discussed.

1. Research questions

As Knapp (2000, 33) has recently observed, the analytical data provided by science-based archaeology 'are likely to be open-ended, and subject to multiple interpretations'. From this standpoint, the growing body of archaeometallurgical data has not always been assessed in ways which are consistent with the investigation of those issues of greatest interest to archaeologists. For example, Budd and Taylor (1995) have argued that scholars working on ancient metallurgy should move away from assumptions of technological determinism and investigations of provenance, towards a greater stress upon understanding ancient metalworking within its socio-economic context. Obviously, technical studies are frequently of considerable interest in themselves; science-based archaeology will, quite reasonably, have its own research agenda. However, from the standpoint of the traditional archaeological concerns such as past social and economic organization, or the nature and scale of inter-regional contacts, the results of analytical programmes dealing with ancient metals are likely to be of greatest use when they are considered within a framework which addresses, not just

composition, but social processes such as production, distribution and consumption (Dobres and Hoffmann 1994), the evidence for which comes from careful consideration of archaeological contexts, and the wider patterning of material culture. The present project makes a few tentative steps in this direction, in that it seeks to identify patterning in the ways in which metal artefacts, as items of material culture, were produced and used within a world in which social and economic and technological factors were closely intertwined. This will be achieved by taking a diachronic perspective on: patterns of metal procurement, the range of manufacturing techniques, the relationship between alloy types and artefact categories, the nature and coherence of the metalwork occurring in different types of context, namely settlements, graves, and 'hoards'.

While there is now a reasonably large body of compositional and lead isotope data from pre-Classical artefacts from the east Mediterranean region, much of the data has limited value for the kind of research programme described above. This is because a good proportion of the data has been obtained from material purchased from antiquities dealers, or from artefacts in Western museums

which were acquired many years ago, and the provenance of which may be poorly documented (Branigan *et al.* 1976; Pigott 1996, 162; Rosenfeld *et al.* 1997). Secondly, data may have been obtained by one of several analytical techniques, creating significant problems of data comparability (Knapp and Cherry 1994, 19–24). Finally, the acquisition of the large numbers of analyses of artefacts from individual periods required in order to investigate data patterning is complicated by the existence of significant inter-site variability. This is documented both in terms of the simultaneous use of different alloys for the production of the same category of artefact at individual sites (Philip 1995a, 74), and the significant diversity, even at an intra-regional level, in the sources from which broadly contemporary communities obtained their raw materials (cf. Rehren *et al.* 1997; Hauptmann *et al.* 1999).

These difficulties can be allayed to some extent by a multi-disciplinary investigation focused upon a large body of material from a single site (e.g. Tadmor *et al.* 1995). However, given the vagaries of archaeological recovery, few sites can offer large groups of metalwork from reliable archaeological contexts covering the long time-depth necessary for a diachronic investigation of metallurgical development. Fortunately, an exception exists in the large tell site of Pella located on the east side of the Jordan Valley. In this case, the presence of substantial groups of material dating to several distinct periods, and recovered from a range of contexts (Table 1) permits the monitoring of diachronic change in metalworking practices. The sheer range and quantity of evidence available through the University of Sydney's Pella excavations highlights one of the major strengths of a long-term commitment to fieldwork at a single multiperiod site. Our programme involved the analysis of sixty-eight metal artefacts from the site, plus a distinctive flat axe from the nearby site of Tell al-Shuna.

In order to answer the questions outlined above, it was necessary to obtain data pertinent to all three areas of enquiry – metal composition, methods of manufacture and procurement of raw materials. These were investigated through a combination of elemental, metallographic and lead isotope analysis. The analytical programme was designed to find an effective compromise between sample size and laboratory costs on the one hand, and concern to minimize the impact of analysis upon the artefacts on the other, while working within the time-constraints imposed by a short-term study-loan.

2. Analytical procedures

Elemental analysis

Investigation of metal composition was undertaken through elemental analysis by energy dispersive X-ray fluorescence (EDXRF). The equipment consisted of a Link System XR200 EDXRF spectrometer with a Rhodium target X-ray tube running at 50kV. A copper filter was used to optimise the analytical conditions and to suppress the energy lines from the rhodium anode, whilst a 1 mm. diameter collimator was employed to limit the area of analysis. Analysis was undertaken for a set of metallic elements likely to provide information concerning alloy composition and/or manufacturing processes; Cu, Sn, Zn, Pb, As, Sb, Ag, Ni, Co and Fe. As many of the artefacts could be subject only to non-destructive surface analysis, non-metallic elements, which may have been significantly affected by differences in burial environments of individual artefacts, were not investigated. Calibration was by means of a full suite of single element standards supplemented by appropriate individual multi-element standards. The Fundamental Parameters Model (Sparks 1976) was used to correct for matrix effects. This combination of standards and correction model provided a relative error of approximately 1%.

Surface analysis

Surface analysis was undertaken on all the artefacts. Discrete areas were prepared for analysis by removing the corrosion products and exposing sound metal. Those artefacts that had little or no remaining sound metal were cleaned to their 'original surface' and the analysis undertaken on those areas. Where possible the analysis was repeated on four different areas for each artefact, with the results when consistent being averaged. In those cases where there were marked differences between the results of surface analysis undertaken on different parts of an artefact the relevant results are given separately (e.g. 80258, 170161, see Table 2). The most likely reasons for differences in surface composition include inhomogeneity within the artefact itself, resulting from variability within the casting and cooling process, or differential corrosion effects across the surface. These could result from local differences in composition as noted above, ancient repair work, or very localised variation in the burial environments to which different parts of an artefact were subject. A control on the results of surface analysis was provided by EDXRF analysis undertaken at points along the metallographic samples, when these were available (see below).

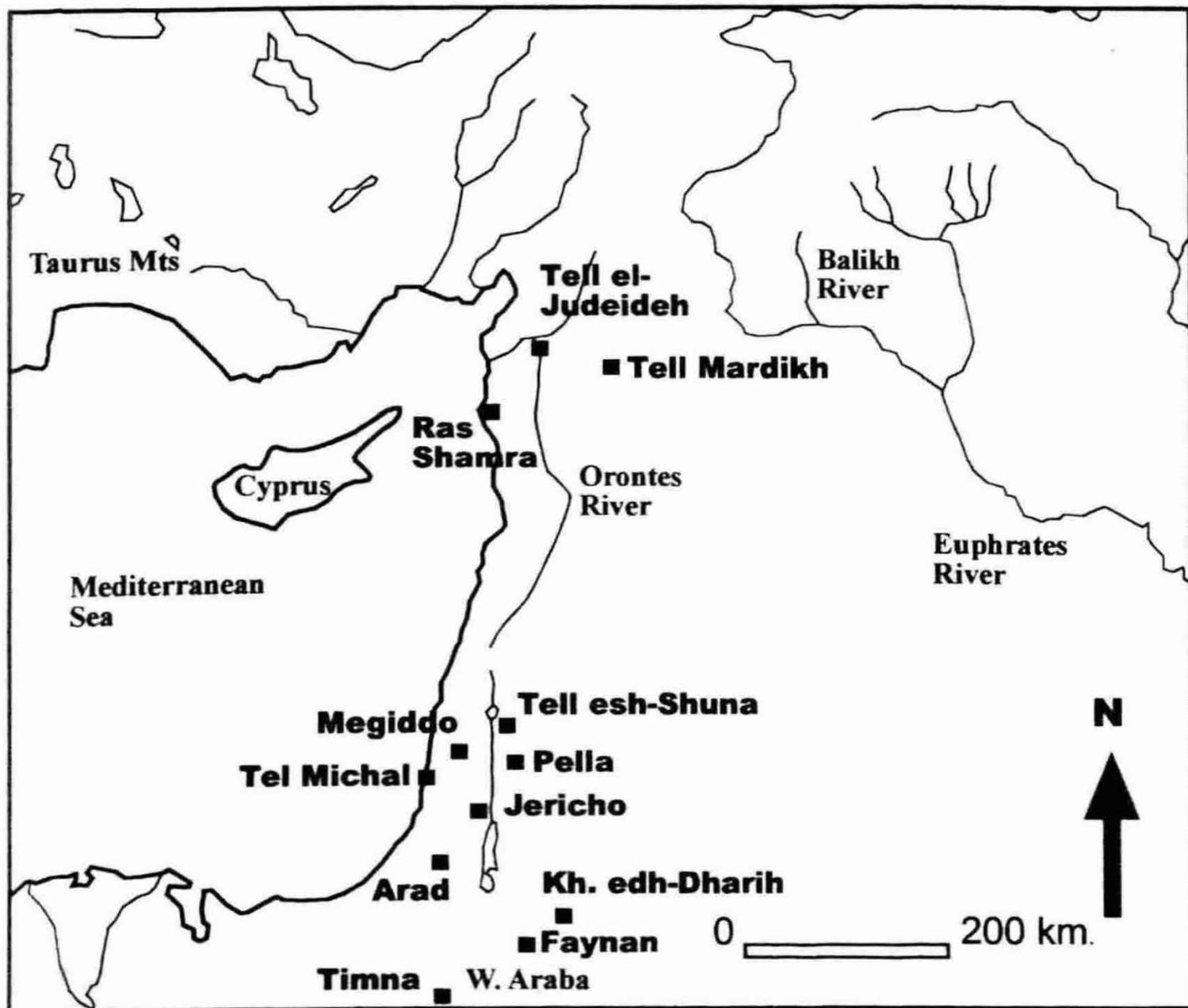


Figure 1. Map showing locations of Pella, sites and mining areas discussed in the text.

Analysis of metallographic samples

Twenty-seven artefacts were selected for metallographic examination, the sample being selected to reflect the typological and chronological range of artefacts. Samples were cut from the artefacts, mounted in epoxy resin, polished to a ¼ micron finish and etched in alcoholic ferric chloride. The samples were examined before and after etching using a Nikon Optiphot microscope at magnifications varying from $\times 25$ to $\times 1000$. Metallographic samples were cut close to areas of individual artefacts where damage was already apparent –

pre-existing breaks for example. EDXRF analysis of the sound metal in the prepared metallographic samples was undertaken in order to provide an assessment of the reliability of the surface analysis. Those artefacts that showed little or no remaining metal were analysed at points across the samples in order to establish the extent of variability within the corrosion layers (Table 2). A further assessment of the reliability of surface analysis was obtained from the EDXRF analyses carried out at the Oxford Isotracer Laboratory on the drilled samples collected for lead isotope analysis.

Reg. No	Object	Sub Type	Context	Type of Context	Date
TS 538	Axe	lugged-bladed	L 785	Settlement	EBA ?
32214	Pin	toggle-pin	XI T.21	Grave	MBIIC-LBI
32215	Pin	toggle-pin	XI T.19	Grave	
42114	Pin	toggle-pin	VI T.27	Grave	LB I
42116	Bracelet		VI T.27	Grave	LB I
60118	Edged tool	knife	IIIC 37.4	Settlement	MB IIB/C
70289	Pin	toggle-pin	XI T.62 1.2	Grave	MB IIC-LB I
70393	Pin	toggle-pin	XI T.62 1.2	Grave	MB IIC-LB I
70400	Pin	toggle-pin	XI T.62 1.E	Grave	MB IIC-LB I
70473	Ring	earring	XI T.62 1F	Grave	MB IIC-LB I
70517	Ring	earring	XI T.62 1G	Grave	MB IIC-LB I
70760	Pin	toggle-pin	XI T.62 3.D	Grave	MB IIC-LB I
70849	Pin	toggle-pin	XI T.62 4.D	Grave	MB IIC-LB I
70856	Ring	earring	XI T.62 3E	Grave	MB IIC-LB I
70889	Pin	toggle-pin	XI T.62 3.E	Grave	MB IIC-LB I
70901	Ring	earring	XI T.62 3E/F	Grave	MB IIC-LB I
70908	Pin	toggle-pin	XI T.62 4.D	Grave	MB IIC-LB I
70923	Pin	toggle-pin	XI T.62 4D	Grave	MB IIC-LB I
80258	Dagger	narrow bladed	XXXI T7.3.1.2	Grave	EB IV
90208	Tool	chisel ?	IIIN 49.33	Settlement	LB II
100142	Bracelet	small	II T.89 1.4	Grave	Iron Age
100201	Bracelet	anklet	II T.89 1.4	Grave	Iron Age
100214	Bracelet		II T.89 1.4	Grave	Iron Age
100222	Bracelet	anklet	II T.89 2.3	Grave	Iron Age
100252	Ring	finger	II T.89 1.4	Grave	Iron Age
100255	Bracelet	small	II T.89 1.4	Grave	Iron Age
110290	Edged tool	dagger ?	IIIP 104.50	Settlement	LB IIB
110603	Pin	toggle-pin	II T.94 1.2	Grave	LB I
110606	Pin	toggle-pin	II T.94 1.2	Grave	LB I
170058	Axe		IIIQ 121.12	Settlement	LB I
170065	Ring	finger	IVE 118.2	Settlement	MB IIB
170066	Pin	toggle-pin	IIIQ 109 F.118	Settlement	LB II
170084	Projectile	arrow	XXXIVF 17.3	Hoard	LB II
170085	Projectile	arrow	XXXIVF 17.3	Hoard	LB II
170086	Projectile	arrow	XXXIVF 17.3	Hoard	LB II
170087	Projectile	bolt	XXXIVF 17.3	Hoard	LB II
170088	Projectile	arrow	XXXIVF 17.4	Hoard	LB II
170089	Projectile	arrow	XXXIVF 17.4	Hoard	LB II
170090	Projectile	arrow	XXXIVF 17.4	Hoard	LB II
170091	Projectile	arrow	XXXIVF 17.4	Hoard	LB II
170092	Projectile	arrow	XXXIVF 17.4	Hoard	LB II
170093	Projectile	arrow	XXXIVF 17.4	Hoard	LB II
170094	Projectile	arrow	XXXIVF 17.4	Hoard	LB II
170095	Projectile	bolt	XXXIVF 17.4	Hoard	LB II
170096	Projectile	arrow	XXXIVF 17.4	Hoard	LB II
170097	Projectile	bolt	XXXIVF 17.4	Hoard	LB II
170160	Pin	toggle-pin	XXVIII A 32.12	Settlement	LB II
170161	Pin	toggle-pin	XXVIII A 31.22	Settlement	MB-LB
170167	Pin	toggle-pin	XXIIIB 14.26	Settlement	LB II
170202	Axe		XXXIID 42.14	Settlement	EB IB/II
180016	Spear		XXXIID 50.29	Settlement	MB IIB/C
180019	Projectile	arrow	XXXIID 50.35	Settlement	MB IIB
180043	Axe		XXXIVB 19.17	Hoard	EB II
180044	Axe		XXXIVB 19.17	Hoard	EB II
180045	Axe		XXXIVB 19.17	Hoard	EB II
180046	Axe		XXXIVB 19.17	Hoard	EB II
180047	Tool	chisel	XXXIVB 19.17	Settlement	EB II

Reg. No	Object	Sub Type	Context	Type of Context	Date
180048	Tool	chisel	XXXIVB 19.17	Settlement	EB II late
180049	Pin		XXXIVB	Settlement	EB II
180052	Tool?		XXXIID 53.5	Settlement	EB IB or MB
190073	Fitting	strap	XXXIIG 107.20	Settlement	LB IIB
190074	Harpoon		XXXIIG 107.24	Settlement	LB IIB
200013	Trinket	balance pan	XXXIIE 15.4	Settlement	LB IIB
200014	Trinket	balance pan	XXXIIE 15.4	Settlement	LB IIB
200015	Trinket	cymbal	XXXIIE 15.4	Settlement	LB IIB
200016	Trinket	cymbal	XXXIIE 15.4	Settlement	LB IIB
920630	Pin	toggle-pin	XXVIII A 26.3	Settlement	MB II
920631	Pin	toggle-pin	XXVIII A 26.3	Settlement	MB II
950477	Pin		XXXIVB 19.17	Settlement	EB II late

Table 1. *Artefacts included in analytical programme. (Note TS 538 is an axe from the site of Tell al-Shuna, located around 25 km. north of Pella).*

Variation in analytical results

Surface analysis of metal artefacts is often considered to be unreliable because of the differential corrosion and leaching of metallic elements during burial. However the availability of comparative results from three different sources allows for an assessment of the validity of the results presented in this work.

In addition to providing information on manufacturing techniques, the metallographic samples were intended to provide data on the degree to which different artefact categories were affected by corrosion. This issue is important because the bulk of the identifiable metal artefacts from almost any site in the Middle East, cemeteries in particular, comprises small objects such as rings, pins and thin blade fragments. Other things being equal these are the very kinds of artefact least likely to preserve significant quantities of sound metal. However, examination of such artefacts, which were both numerically important and perhaps to some extent symbolically charged, is vital if we seek to gain an overall picture of metallurgical activity. The larger objects within which substantial quantities of good metal are often preserved, in fact constitute a small proportion of the artefacts recovered at most sites. Moreover, being in better condition, such artefacts are not always available for laboratory analysis. In fact, an analytical project restricted to large objects alone would be compelled to work with a very small dataset and thus be unable to compare metalworking practices across the main artefact categories.

Those artefacts subject to metallographic analysis were divided into three groups: uncorroded, par-

tially corroded; substantially corroded (indicated as UC, PC and Corr in the 'Condition' column of Table 2). The analytical results for the first group are considered reliable, those in the second group may be less accurate because of internal corrosion but probably offer a reasonable general indication of alloy composition, while the values cited for those in the last group should be treated with caution. Although we have presented the data as they were collected, the numerical values quoted for artefacts classed as substantially corroded should be taken to indicate general alloy type only – tin bronze, arsenical bronze, leaded tin-bronze – rather than actual values. We feel that this approach permits the extraction of some useful information from the less well-preserved material, without assigning it an evidential value which it does not possess.

The variation in the tin content between the analysis of the surface and the drilled samples and metallographic samples is shown graphically (Figs 2–4). It is clear that for artefacts with sound metal core, generally larger artefacts such as axes, bracelets and chisels, differences between the concentration of tin in the surface analysis and those of the metallographic samples were relatively small (Fig. 2). In addition the drilled samples analysed by EDXRF at Oxford were compared with results of surface analyses in order to provide data on a range of objects from which it was not possible to prepare metallographic samples (Fig. 3). Once more, agreement was generally good, although certain artefacts revealed a reduced level of tin at the surface e.g. the bracelet 42116. This phenomenon was more clearly observed in those artefacts showing a greater degree of internal corrosion, with tin concentrations broadly consistent between analyses

I/D	Object	Date	Method	Condi- tion	Cu	Zn	Sb	As	Pb	Co	Ni	Au	Hg	Ag	Sn	Fe	TOTAL
538	Axe	EBA ?	Surface	UC	91.60	n.d.	det.	n.d.	1.70	n.d.	n.d.	n.d.	n.d.	det	6.50	n.a.	99.8
32214	Pin	MB IIC-LB I	Surface		88.1	det.	det.	n.d.	5.91	det.	det.	det.	det.	n.d.	5.37	det.	99.4
32215	Pin	MB IIB-C	Surface		95.7	det.	0.19	2.26	det.	n.d.	det.	det.	det.	det.	0.27	0.70	99.1
42114	Pin	LB I	Surface	PC	86.6	det.	det.	det.	det.	n.d.	det.	det.	0.32	n.d.	12.37	0.21	99.5
42114	Pin		Metalog	PC	85.6	det.	det.	det.	det.	n.d.	det.	det.	det.	det.	12.24	det.	97.8
42114	Pin		Metalog	PC	85.6	n.d.	n.d.	det.	det.	det.	det.	n.d.	n.d.	det.	12.33	1.01	98.9
42114	Pin		Metalog	PC	85.3	det.	n.d.	det.	det.	n.d.	det.	det.	det.	det.	12.55	0.63	98.5
42114	Pin		Metalog	PC	85.8	n.d.	n.d.	det.	det.	n.d.	det.	n.d.	n.d.	det.	13.54	det.	99.3
42114	Pin		Metalog	PC	84.8	det.	det.	det.	det.	det.	det.	n.d.	n.d.	det.	13.04	0.60	98.5
42116	Bracelet	LB I	Surface		89.8	det.	det.	0.28	0.72	det.	det.	n.d.	n.d.	det.	8.24	0.45	99.5
60118	Blade	MB IIB/C	Surface	Corr	93.9	det.	det.	n.d.	0.24	n.d.	n.d.	n.d.	n.d.	n.d.	5.58	det.	99.8
60118	Blade		Metalog	Corr	81.6	n.d.	det.	16.03	0.55	98.1							
60118	Blade		Metalog	Corr	85.94	det.	det.	det.	det.	n.d.	det.	n.d.	n.d.	det.	12.84	det.	98.8
60118	Blade		Metalog	Corr	80.4	n.d.	det.	det.	det.	n.d.	det.	det.	n.d.	n.d.	16.97	det.	97.3
70289	Pin	MB IIC-LB I	Surface		98.9	det.	det.	n.d.	0.44	det.	n.d.	n.d.	det.	det.	0.33	det.	99.6707
70393	Pin	MB IIC-LB I	Surface		97.4	n.d.	det.	det.	det.	n.d.	n.d.	det.	det.	n.d.	1.91	det.	99.2821
70400	Pin	MB IIC-LB I	Surface		97.6	n.d.	det.	det.	det.	n.d.	n.d.	det.	det.	n.d.	1.46	0.20	99.2424
70473	Ring	MB IIC-LB I	Surface	Corr	91.1	det.	det.	0.48	det.	n.d.	n.d.	n.d.	n.d.	det.	7.23	0.34	99.1383
70473	Ring		Metalog	Corr	81.9	n.d.	det.	0.75	1.82	n.d.	n.d.	det.	det.	det.	13.82	0.98	99.26
70473	Ring		Metalog	Corr	80.3	det.	det.	det.	2.38	n.d.	det.	n.d.	n.d.	det.	15.34	det.	98.03
70473	Ring		Metalog	Corr	80.6	det.	det.	det.	1.96	det.	n.d.	n.d.	n.d.	det.	14.84	1.33	98.71
70473	Ring		Metalog	Corr	80.0	n.d.	det.	det.	2.18	det.	det.	n.d.	n.d.	det.	14.42	1.72	98.2
70517	Ring	MB IIC-LB I	Surface		98.4	n.d.	det.	det.	det.	n.d.	det.	det.	det.	det.	0.98	0.25	99.6
70760	Pin	MB IIC-LB I	Surface		91.9	det.	det.	0.40	det.	n.d.	n.d.	det.	det.	det.	7.13	det.	99.5
70849	Pin	MB IIC-LB I	Surface		97.5	n.d.	det.	det.	det.	n.d.	n.d.	n.d.	det.	det.	1.91	0.28	99.8
70856	Ring	MB IIC-LB I	Surface		87.8	n.d.	det.	0.78	0.93	det.	det.	det.	det.	det.	8.13	0.55	98.2
70889	Pin	MB IIC-LB I	Surface		94.6	n.d.	det.	n.d.	det.	det.	det.	n.d.	det.	n.d.	4.88	0.21	99.7
70901	Ring	MB IIC-LB I	Surface		97.4	det.	det.	n.d.	0.40	det.	n.d.	n.d.	det.	det.	1.71	0.31	99.8
70908	Pin	MB IIC-LB I	Surface		98.2	det.	det.	n.d.	0.27	n.d.	n.d.	det.	det.	det.	1.11	0.18	99.8
70923	Pin	MB IIC-LB I	Surface	PC	94.4	n.d.	det.	det.	det.	n.d.	n.d.	det.	det.	det.	4.05	0.57	99.0
70923	Pin		Metalog	PC	83.3	det.	det.	det.	det.	n.d.	det.	n.d.	det.	n.d.	13.87	det.	97.2
70923	Pin		Metalog	PC	83.7	det.	13.55	det.	97.3								
80258	Dagger	EB IV	Surface	Corr	86.1	n.d.	0.88	7.03	0.59	det.	0.23	n.d.	det.	0.15	det.	5.02	99.9

ID	Object	Date	Method	Condi- tion	Zn	Sb	As	Pb	Co	Ni	Au	Hg	Ag	Sn	Fe	TOTAL	
80258	Dagger		Surface	Corr	88.0	det.	0.80	6.52	0.50	det.	0.22	n.d.	n.d.	0.15	det.	3.69	99.9
90208	Tool	LB II	Surface	PC	87.1	det.	det.	0.22	det.	det.	n.d.	n.d.	n.d.	det.	11.94	0.47	99.7
90208	Tool		Metalog	PC	81.3	det.	13.30	det.	94.4								
90208	Tool		Metalog	PC	81.6	n.d.	det.	det.	det.	n.d.	det.	det.	n.d.	det.	13.80	det.	95.46
100142	Bracelet	Iron Age	Surface		81.1	det.	det.	det.	0.87	det.	det.	det.	n.d.	det.	16.72	0.53	99.3
100201	Bracelet	Iron Age	Metalog		86.4	det.	det.	det.	1.25	det.	det.	det.	n.d.	det.	11.79	0.20	99.6
100214	Bracelet	Iron Age	Surface	PC	93.6	det.	det.	det.	0.39	det.	n.d.	n.d.	n.d.	det.	5.51	0.21	99.7
100214	Bracelet		Metalog	PC	89.9	det.	det.	det.	det.	n.d.	det.	det.	det.	det.	5.74	det.	95.6
100222	Bracelet	Iron Age	Surface		83.6	det.	det.	n.d.	1.11	n.d.	det.	n.d.	n.d.	n.d.	14.89	0.22	99.8
100252	Ring	Iron Age	Surface		90.0	det.	det.	n.d.	0.69	n.d.	det.	n.d.	det.	det.	9.76	0.41	99.8
100255	Bracelet	Iron Age	Surface	PC	87.1	det.	det.	n.d.	1.06	n.d.	n.d.	det.	n.d.	det.	10.82	0.55	99.5
100255	Bracelet		Metalog	PC	87.8	det.	det.	det.	0.97	det.	det.	det.	det.	det.	9.07	0.77	98.6
110290	Edged tool	LB IIB	Metalog	Corr	83.1	n.d.	n.d.	det.	det.	det.	det.	n.d.	n.d.	n.d.	14.70	det.	97.8
110290	Edged tool		Metalog	Corr	81.7	det.	det.	det.	det.	n.d.	n.d.	n.d.	n.d.	n.d.	16.64	det.	98.3
110290	Edged tool		Metalog	Corr	82.4	n.d.	det.	det.	0.62	n.d.	det.	n.d.	n.d.	det.	15.33	0.73	99.0
110603	Pin	LB I	Surface		96.4	det.	det.	det.	det.	n.d.	det.	det.	n.d.	det.	2.86	det.	99.3
110606	Pin	LB I	Surface		95.3	n.d.	n.d.	det.	det.	n.d.	n.d.	n.d.	n.d.	n.d.	4.28	0.16	99.7
170058	Axe	LB I	Surface		89.8	det.	det.	n.d.	det.	det.	n.d.	n.d.	det.	n.d.	9.34	0.48	99.6591
170065	Ring	MB IIB	Surface		98.3	det.	det.	det.	det.	det.	n.d.	det.	det.	n.d.	0.79	0.18	99.2
170065	Ring		Metalog	PC	83.9	det.	12.00	det.	95.9								
170065	Ring		Metalog	PC	87.3	det.	det.	det.	det.	n.d.	det.	n.d.	det.	det.	10.19	0.80	98.3
170066	Pin	LB II	Surface	Corr	77.6	n.d.	det.	0.39	det.	n.d.	n.d.	det.	det.	20.56	0.38	0.72	99.6
170066	Pin		Metalog	Corr	61.3	det.	det.	det.	0.49	n.d.	det.	det.	det.	34.41	det.	1.07	97.3
170066	Pin		Metalog	Corr	61.2	n.d.	det.	det.	det.	det.	n.d.	det.	n.d.	36.25		det.	97.4
170084	Projectile	LB II	Surface		98.3	det.	0.08	0.62	0.16	n.d.	det.	det.	n.d.	det.	det.	0.63	99.8
170085	Projectile	LB II	Surface		99.3	det.	det.	0.16	det.	n.d.	det.	det.	n.d.	det.	det.	0.15	99.6
170086	Projectile	LB II	Surface		99.0	det.	0.08	0.21	0.15	det.	n.d.	n.d.	det.	det.	det.	0.43	99.9
170087	Projectile	LB II	Surface		99.3	n.d.	det.	det.	det.	det.	n.d.	det.	n.d.	det.	0.22	0.19	99.7
170088	Projectile	LB II	Surface		99.0	det.	0.08	0.22	n.d.	det.	n.d.	det.	det.	det.	det.	0.29	99.6
170089	Projectile	LB II	Surface		98.8	det.	0.09	0.38	0.18	n.d.	det.	det.	det.	det.	det.	0.30	99.8
170090	Projectile	LB II	Surface		98.8	n.d.	det.	0.20	det.	det.	det.	det.	n.d.	det.	det.	0.60	99.6
170091	Projectile	LB II	Surface		99.1	n.d.	0.07	0.34	0.14	n.d.	n.d.	n.d.	n.d.	det.	det.	0.27	99.9
170092	Projectile	LB II	Surface		95.0	det.	0.09	2.23	0.21	det.	det.	det.	n.d.	det.	0.34	1.92	99.8

17I/D	Object	Date	Method	Condi- tion	Cu	Zn	Sb	As	Pb	Co	Ni	Au	Hg	Ag	Sn	Fe	TOTAL
0093	Projectile	LB II	Surface	UC	98.7	det.	0.10	0.49	det.	det.	n.d.	det.	det.	det.	det.	0.49	99.7
170093	Projectile		Metalog	UC	86.9	det.	det.	det.	det.	n.d.	det.	n.d.	n.d.	det.	det.	3.79	90.7
170094	Projectile	LB II	Surface		98.4	n.d.	0.10	0.70	det.	det.	det.	n.d.	det.	det.	det.	0.55	99.8
170095	Projectile	LB II	Surface		98.2	n.d.	det.	det.	det.	det.	det.	n.d.	n.d.	det.	1.37	det.	99.6
170096	Projectile	LB II	Surface		98.1	n.d.	det.	0.54	det.	n.d.	n.d.	n.d.	det.	det.	det.	1.10	99.8
170097	Projectile	LB II	Surface		97.9	det.	det.	det.	det.	n.d.	n.d.	det.	det.	det.	1.44	0.13	99.5
170160	Pin	LB II	Surface		94.3	det.	det.	det.	det.	n.d.	n.d.	n.d.	n.d.	det.	4.38	0.63	99.3
170161	Pin	MB-LB	Surface	PC	87.1	n.d.	det.	det.	5.80	det.	n.d.	n.d.	det.	det.	6.61	0.31	99.8
170161	Pin		Metalog	PC	86.0	det.	det.	det.	5.14	det.	det.	n.d.	det.	det.	7.62	det.	98.8
170161	Pin		Metalog	PC	88.5	n.d.	det.	det.	3.19	det.	det.	n.d.	det.	det.	6.86	det.	98.5
170161	Pin		Metalog	PC	87.9	det.	n.d.	det.	4.10	det.	n.d.	n.d.	n.d.	det.	7.00	det.	98.9
170161	Pin		Metalog	PC	88.2	det.	n.d.	n.d.	4.50	det.	det.	n.d.	det.	det.	6.30	det.	98.9
170161	Pin		Surface	PC	86.8	det.	n.d.	det.	4.59	det.	det.	n.d.	n.d.	det.	7.26	det.	98.7
170167	Pin	LB II	Surface	PC	97.6	det.	det.	det.	det.	det.	n.d.	det.	det.	det.	0.86	0.29	98.7
170167	Pin		Metalog	PC	89.8	det.	det.	det.	det.	n.d.	det.	det.	n.d.	n.d.	7.06	det.	96.8
170202	Axe	EB IB/II	Surface		97.9	n.d.	det.	0.71	det.	det.	det.	det.	n.d.	det.	det.	0.90	99.6
180016	Spear	MB IIB/C	Surface		98.7	det.	det.	0.33	det.	det.	n.d.	n.d.	n.d.	det.	det.	0.57	99.6
180019	Projectile	MB IIB	Surface	UC	94.6	n.d.	det.	det.	0.82	det.	det.	n.d.	n.d.	det.	4.23	0.17	99.8
180019	Projectile		Metalog	UC	84.6	n.d.	n.d.	n.d.	1.54	det.	det.	det.	det.	det.	12.66	det.	98.8
180019	Projectile		Metalog	UC	84.1	det.	det.	det.	det.	det.	n.d.	n.d.	n.d.	det.	10.55	det.	94.6
180019	Projectile		Metalog	UC	86.4	n.d.	det.	det.	1.63	det.	det.	det.	det.	det.	10.77	det.	98.8
180043	Axe	EB II	Surface	UC	96.8	n.d.	0.10	1.52	0.21	n.d.	det.	n.d.	n.d.	det.	det.	1.16	99.8
180043	Axe		Metalog	UC	95.7	n.d.	det.	det.	det.	det.	det.	n.d.	det.	n.d.	det.	det.	95.7
180044	Axe	EB II	Surface		99.1	n.d.	det.	det.	0.22	n.d.	det.	det.	det.	det.	det.	0.33	99.7
180045	Axe	EB II	Surface	UC	99.5	n.d.	det.	n.d.	0.17	det.	n.d.	det.	det.	det.	det.	0.16	99.8
180045	Axe		Metalog	UC	98.1	det.	det.	det.	n.d.	det.	det.	det.	n.d.	n.d.	det.	det.	98.1
180045	Axe		Metalog	UC	98.2	det.	n.d.	n.d.	0.77	n.d.	det.	n.d.	n.d.	det.	det.	det.	98.9
180046	Axe	EB II	Surface	UC	99.0	n.d.	det.	0.15	0.20	det.	n.d.	det.	n.d.	det.	det.	0.33	99.6
180046	Axe		Metalog	UC	94.1	det.	det.	det.	det.	n.d.	det.	det.	n.d.	n.d.	det.	det.	94.1
180047	Tool	EB II	Surface		97.2	det.	det.	0.60	det.	det.	n.d.	det.	det.	0.17	0.49	0.91	99.4
180048	Tool	EB II late	Surface	UC	97.5	det.	det.	1.32	det.	n.d.	n.d.	n.d.	det.	det.	det.	0.87	99.7
180048	Tool		Metalog	UC	95.3	det.	det.	1.11	det.	det.	1.30	n.d.	n.d.	det.	det.	1.05	98.8
180049	Pin	EB II	Surface	Corr	98.6	n.d.	det.	det.	1.08	det.	n.d.	n.d.	n.d.	n.d.	det.	0.22	99.9

I/D	Object	Date	Method	Condi- tion	Cu	Zn	Sb	As	Pb	Co	Ni	Au	Hg	Ag	Sn	Fe	TOTAL
180049	Pin		Metalog	Corr	95.9	det.	n.d.	n.d.	det.	n.d.	det.	n.d.	det.	det.	det.	det.	95.9
180052	Tool?	EB IB or MBA	Surface	Corr	95.2	n.d.	det.	0.29	det.	det.	n.d.	det.	det.	det.	3.36	0.50	99.3
180052	Tool?		Metalog	Corr	91.8	det.	det.	det.	det.	det.	n.d.	n.d.	det.	det.	det.	det.	97.3
190073	Fitting	LB IIB/	Surface		95.8	det.	det.	0.34	det.	n.d.	n.d.	n.d.	n.d.	det.	3.24	0.30	99.6
190074	Harpoon	LB IIB	Surface		97.6	det.	det.	det.	0.29	det.	det.	det.	n.d.	0.30	1.25	0.22	99.6
190074	Harpoon		Surface		97.2	det.	det.	0.34	0.34	n.d.	det.	det.	n.d.	0.21	0.70	0.89	99.7
200013	Balance Pan	LB IIB	Surface		98.7	n.d.	det.	det.	det.	det.	det.	n.d.	det.	det.	0.88	0.17	99.8
200014	Balance Pan	LB IIB	Surface		97.7	det.	det.	0.11	det.	det.	n.d.	n.d.	n.d.	det.	1.84	0.17	99.8
200014	Balance Pan		Surface		98.0	det.	det.	0.16	det.	0.04	n.d.	det.	det.	det.	1.36	0.25	99.8
200015	Cymbal	LB IIB	Surface		97.9	0.24	det.	det.	0.38	det.	det.	n.d.	n.d.	0.08	1.04	0.20	99.9
200015	Cymbal		Surface		98.1	0.23	det.	det.	0.62	det.	det.	n.d.	det.	0.03	0.61	0.24	99.9
200016	Cymbal	LB IIB	Surface		95.7	0.32	det.	0.09	0.10	det.	0.12	det.	n.d.	n.d.	3.38	0.20	99.9
200016	Cymbal		Surface		96.7	0.17	det.	0.05	0.08	det.	n.d.	n.d.	n.d.	det.	2.65	0.26	99.9
200016	Cymbal		Surface		96.0	0.33	det.	0.09	0.08	det.	det.	det.	det.	det.	3.146	0.20	99.9
200016	Cymbal		Surface		94.6	0.31	det.	4.42	0.27	99.6							
920630	Pin	MB II	Surface	Corr	97.6	det.	det.	det.	det.	det.	n.d.	det.	n.d.	det.	1.14	0.31	99.1
920630	Pin		Metalog	Corr	65.8	n.d.	n.d.	det.	det.	det.	det.	n.d.	n.d.	det.	28.98	2.52	97.0
920630	Pin		Metalog	Corr	62.6	n.d.	det.	det.	2.03	n.d.	n.d.	det.	n.d.	det.	33.77	det.	98.4
920630	Pin		Metalog	Corr	68.6	det.	det.	0.55	0.77	n.d.	det.	n.d.	n.d.	det.	28.55	det.	98.4
920630	Pin		Metalog	Corr	69.3	n.d.	det.	0.48	0.96	det.	n.d.	det.	n.d.	det.	27.07	1.17	98.9
920630	Pin		Metalog	Corr	90.1	det.	det.	det.	0.66	det.	det.	det.	n.d.	det.	7.25	det.	98.8
920631	Pin	MB II	Surface	Corr	96.0	n.d.	det.	det.	det.	det.	n.d.	det.	n.d.	det.	3.33	0.20	99.6
920631	Pin		Metalog	Corr	75.0	n.d.	det.	det.	det.	n.d.	det.	n.d.	n.d.	det.	21.8	det.	97.5
950477	Pin	EB II late	Surface		98.7	det.	det.	det.	0.58	det.	det.	n.d.	det.	det.	det.	0.24	99.5

Table 2. Results of EDXRF analysis of prepared artefact surface, analysis of metallographic samples. These are indicated as (Surface) and (Metalog) respectively in column 'Method'. In column 'Condition' (UC) = core preserved, (PC) = core partially corroded, (Corr) = little uncorroded material remaining. When multiple analyses were carried out, either on different areas of the surface of an artefact or at different points along a metallographic sample, these are listed separately. det = detected, n.d. = not detected, n.a. = not available; the limits of detection under the analytical conditions were: Cu 0.1%; Zn 0.1%; Sb 500ppm; As 500ppm; Pb 500ppm; Co 0.10%; Ni 0.1%; Au 0.1%; Hg 0.1%; Ag 500ppm; Sn 500ppm; Fe 0.10%.

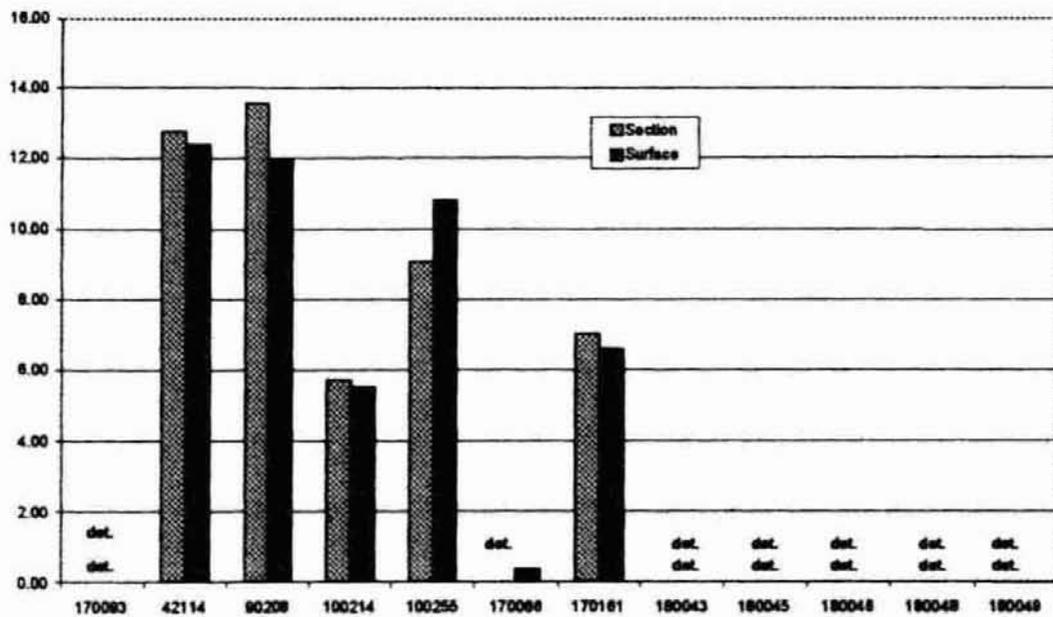


Figure 2. Comparison of tin concentrations obtained by EDXRF analysis of surface and of metallographic samples; artefacts with good preservation of metal core only.

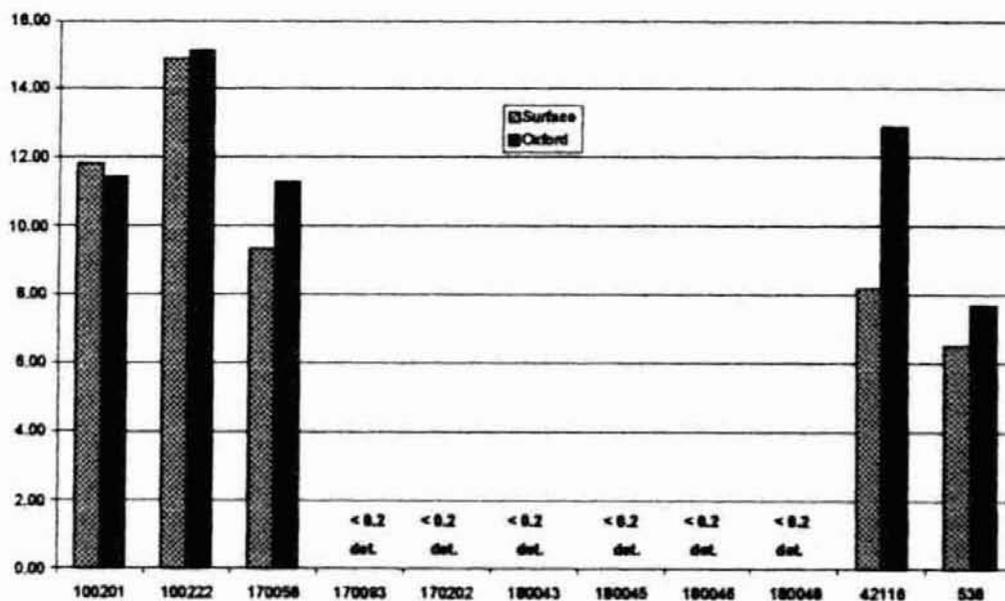


Figure 3. Comparison of tin concentrations obtained by EDXRF analysis of surface with 'semi-quantitative' EDXRF analysis of drilled samples undertaken in Oxford; artefacts with good preservation of metal core only.

taken on different parts of the metallographic samples, but markedly different from those obtained by surface analysis (Fig. 4). Values are generally higher in the interior than on the surface, presumably indicative of a degree of depletion of tin at the surviving surface, even though these samples had been cleaned to what appeared on visual

inspection to represent sound metal (see Section 3). We therefore believe that in the case of many of the smaller artefacts (not just those for which metallographic samples were examined), the surface analyses may considerably under-represent the quantity of tin present. Our conclusion is that artefacts classed as corroded, and all small ar

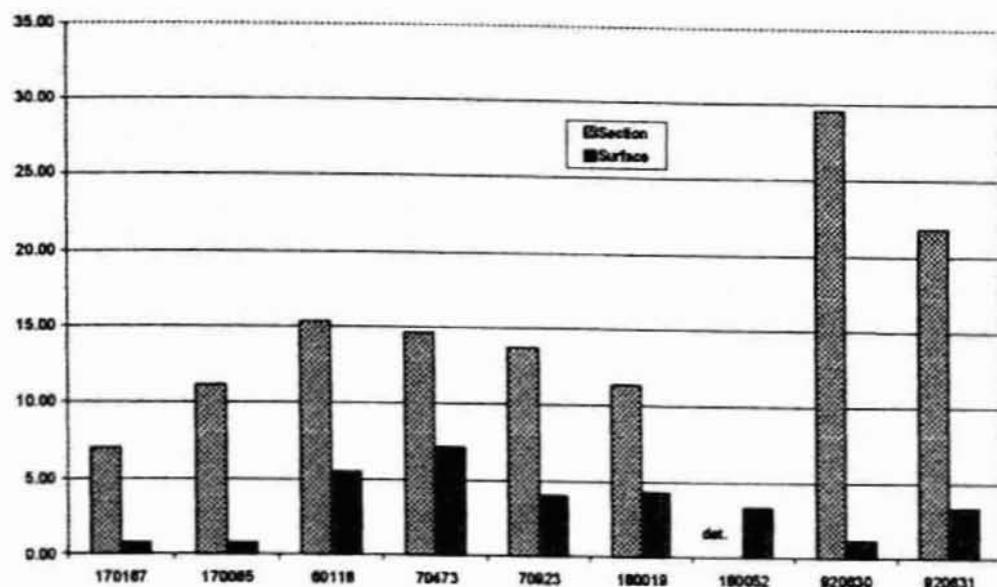


Figure 4. Comparison of tin concentrations obtained by EDXRF analysis of surface and of metallographic samples for artefacts with minimal preservation of metal core.

facts, mainly pins and rings, surface results indicating the presence of even quite low percentages of tin, should be taken to indicate that the artefact was made from tin-bronze.

Perhaps more difficult to explain are the differences in tin concentrations shown by Nos 920630 and 920631 which show consistent tin contents of 20% to 30% from the metallographic samples but surface concentrations of between 1% and 3% tin. Both toggle pins come from the same contexts so this might be attributable to circumstances relating to a particular burial environment, which have resulted in an amplification of the process documented in the other instances. Although in many instances there is generally good agreement between the results of the EDXRF analysis of drilled samples undertaken in Oxford and our analysis of metallographic samples, some differences can be observed.

Agreement between the results of EDXRF analyses undertaken at different points along a metallographic sample taken from a single artefact was frequently good e.g. 42114, 70923. In other cases, however, some variation was detected between the concentrations of individual elements at different points. For example 170161 revealed lead values ranging from 3.19% to 5.14%, while 920630 revealed a tin concentration that varied between 27.07% and 33.77%. In the case of lead, which tends to be concentrated in globules, this is most probably due to differences in its distribution throughout the artefact. In the case of other ele-

ments it is likely to relate to a degree of inhomogeneity resulting from the effects of specific casting procedures and different rates of cooling, and / or the variable effects of corrosion at different points in the artefact. Distinctions between the results of EDXRF analysis undertaken on metallographic samples at Durham (Table 2), and the 'semi-quantitative' results of EDXRF undertaken in Oxford (Table 5) may reflect the rather different nature of the metal collected in a drilled sample, as opposed to that from a single point on a metallographic sample, or perhaps differences in instrumentation, in addition to the possibilities outlined above.

3. Metallographic investigation

3.1 Introduction

The main microstructural features seen in the copper alloys are as follows: coring, recrystallised grains, annealing twins, cold-working, and inclusions – oxides, sulphides and secondary phase. The evidence is summarised in Table 3. Coring is the micro-segregation which occurs when copper alloys solidify. In all of the cases where coring could be seen in the samples the coring was not of the normal dendritic form (i.e. portions of the coring are perpendicular to each other). Instead the coring was present as parallel dark and light bands (Fig. 5). This is mostly likely to occur when a sample of cast metal is hammered flat. Heating copper alloys

Artefact	Object	Date	Cored	Recrystallised	Annealing twins	Cold worked	Copper-copper oxide eutectic	Sulphide inclusions	Second phase
180045 shoulder	axe	EBII		✓	✓	*	✓		
180045 blade	axe	EB II		✓	✓	*	✓		
180046	axe	EBII		✓	✓	✓	✓		
180049	pin	EBII	✓	✓	✓				
180043 blade	axe	EBII		✓	✓	✓		✓	
180048 back	axe	EBII late	✓	✓	✓	✓		✓	
180052	tool	EBII late		✓		✓		✓	
920630	pin	MBII		✓	?	?			
920631	pin	MBII	?	?				✓	
170065	ring	MBII B		✓	✓			✓	
180019 shaft	arrow	MBII B	?	✓	✓	✓			
180019 tip	arrow	MBII B		✓	✓				
60118	knife	MBII B/C	?	✓	?	✓			
70289	pin	MBII C-LB		✓	✓	?			
70473	earring	MBII C-LB		✓	✓			✓	
70923	pin	MBII C-LB		✓	✓	?		✓	
170161	pin	MB-LB		✓	✓				
42114	pin	LBI	?	✓	✓				
90208 head	chisel ?	LBII	?	✓	✓	?		✓	
90208 tip	chisel ?	LBII		✓	✓	✓		✓	
170066	pin	LBII		✓	✓				✓
170093	arrow	LBII	✓	✓	✓	*		✓	
170167	pin	LBII	✓	✓	✓			✓	
110290	dagger ?	LBII B		✓	?	✓		✓	
100214	bracelet	IA		✓	✓			✓	
100255	bracelet	IA	?	✓	✓				

Table 3. Summary of microstructure of copper alloys (Key to responses: ✓ = present, ? = possible, * = elongated grains [but no strain lines]).



Figure 5. 170167. LB II pin. Original magnification $\times 200$. Etched. Elongated sulphide inclusions indicate the metal has been worked. Recrystallised grains containing annealing twins can be seen imposed on distorted remnant coring (horizontal dark and light bands).

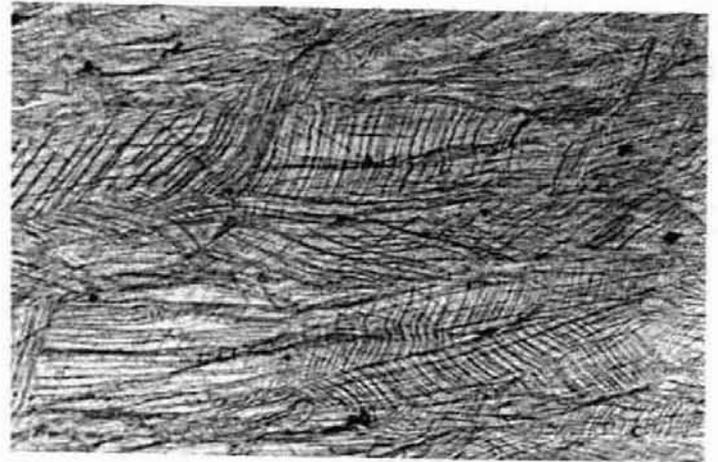


Figure 7. 180043. EB II axe (blade). Original magnification $\times 400$. Etched. Shows heavily distorted grains and abundant strain lines. Given the lack of remnant coring and the size of the grains this metal has been recrystallised. The severe distortion, however, makes it difficult to identify annealing twins with confidence. Some sulphide inclusions.

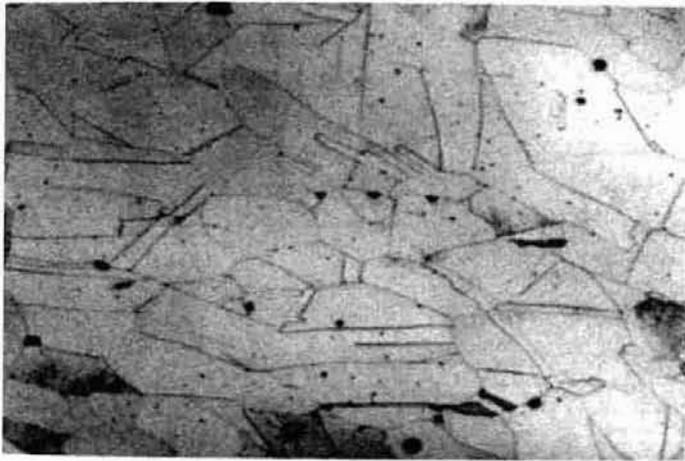


Figure 6. 170093. LB II arrowhead. Original magnification $\times 1000$. Etched. Shows recrystallised grains and annealing twins. The grains are not completely equi-axed but show no sign of strain lines. There is no sign of any coring. Some sulphide inclusions can be seen.

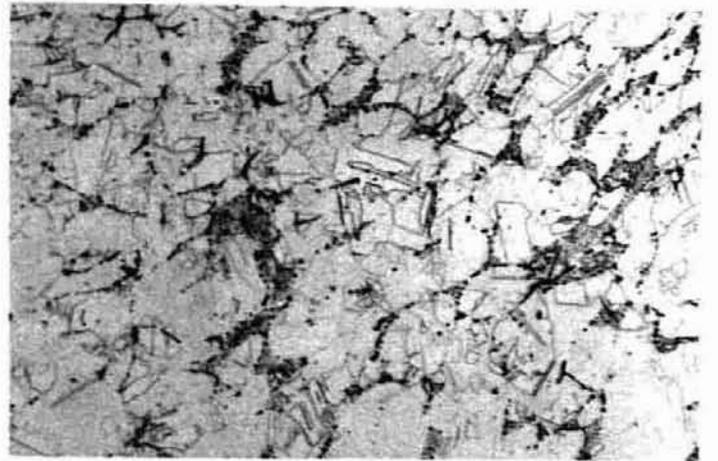


Figure 8. 180045. EB II axe (near to blade). Original magnification $\times 200$. Etched. Shows copper-copper oxide eutectic which is slightly distorted adjacent to the blade. Recrystallised grains and annealing twins are clearly visible.

allows recrystallisation (Fig. 6) although this is most likely to occur in samples which have been subjected to previous cold-working. Recrystallised grains are typically less than 50 microns in diameter (depending on the degree of cold-working and the temperature and duration of the annealing) while 'as cast' grains usually have a diameter of at least several hundred microns. The parallel-sided features seen within individual copper alloy grains are called annealing twins (Figs 5 and 6), and are produced when a cold-worked copper alloy is subjected to heat treatment. Evidence for cold-working takes two forms: distorted grains (Fig. 6)

and strain lines (Fig. 7). The grains are distorted (elongated) perpendicular to the direction of the hammer blows. In all of the cases where elongated grains could be seen in the samples these were orientated parallel to the long axis of the artefact. The distortion of grains occurs by a process of slip along regular crystallographic axes. This can be seen in polished and etched samples as a series of fine strain lines within grains (Fig. 7). Inclusions in copper alloys are those materials which are not



Figure 9. 180046. EB II axe (near to blade). Original magnification $\times 200$. Etched. Shows copper-copper oxide eutectic which is distorted into lines of 'droplets'. Recrystallised grains and annealing twins are visible. The grains are not equi-axed and the annealing twins also show some distortion. There is no sign, however, of any strain lines.

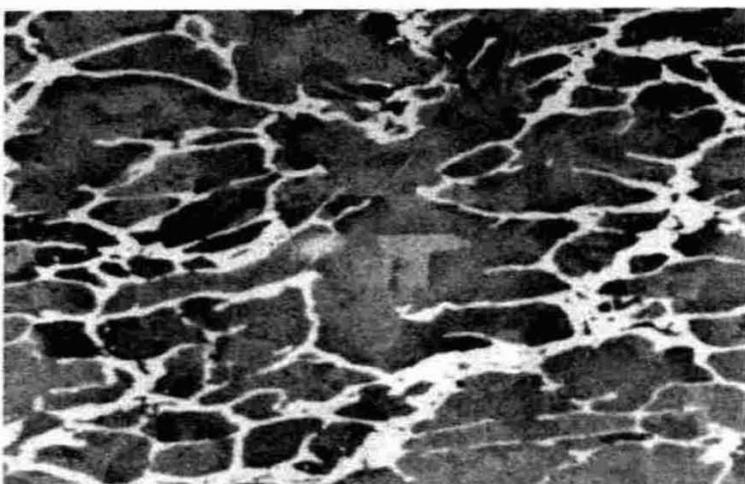


Figure 10. 170066. LB II pin. Original magnification $\times 200$. Etched. Shows silver-rich phase (white) and copper-rich phase (yellow). The copper-rich phase shows signs of recrystallisation.

soluble in the copper, especially sulphides (Figs 5–7) and oxides (Figs 8–9). The oxides are usually present as a copper-copper oxide eutectic. Copper sulphides on the other hand form as discrete nodules, usually in interdendritic spaces. When copper alloys are hammered the inclusions are deformed into plates or strings of droplets. Most of the samples examined are single-phased, that is any metallic elements are in solid solution in the copper. In one case (Fig. 10) a second phase was identified; in this case the β phase of the Cu-Ag binary system.

3.2. Oxide inclusions

Two of the Early Bronze Age II copper alloy artefacts contained copper-copper oxide eutectic (Figs 8 and 9). This inclusion is easily identifiable and distinguishable from copper sulphide (Figs 5–7): because they are light blue in normal brightfield illumination but oxides turn ruby red in plane polarised light (See Higgins 1991, 49–50). Oxide inclusions are generally not associated with more-or-less pure copper and indicate that the metal was not kept in a suitable reducing atmosphere when molten. If an alloy (such as bronze) was slightly oxidised then the dissolved oxygen would react with the alloying element (such as tin) rather than the copper. In modern foundry copper and its alloys are 'de-oxidised' by the addition of suitable elements. The relatively pure copper of the EB II metal would be easily oxidised during melting and casting. The lack of sulphides in the samples indicates the use of secondary ores.

Both EB II artefacts (Nos 18045 and 180046) containing copper-copper oxide eutectic have lead isotope signatures consistent with an origin in the Dolomite-Limestone-Shale (DLS) ores from Faynan (Tables 3, 4; Fig. 14), which would not normally be expected to contain significant amounts of sulphur (see above). That said, samples from other EBA artefacts (Table 3, 180043, 180048 and 180052) did contain sulphides. Two of these were subject to lead isotope analysis and are not compatible with an origin in the Faynan DLS ores. Thus the lead isotope data and the metallography appear to be in agreement.

3.3. Cored and recrystallised microstructures

A number of the Pella samples display a particularly interesting microstructure which has two apparently contradictory features: coring and recrystallised grains (Fig. 5). In the modern foundry copper alloys are hot-worked or homogenised before any cold working takes place. Both of these processes homogenise the alloy and remove coring (Higgins 1974, 328). Modern copper alloys display dendritic coring or recrystallised grains (Higgins 1991, 387–9). The examples from Pella show that ancient metalworking practice was able to produce a microstructure with coring and recrystallised grains (this microstructure can also be seen in Bronze Age samples from Europe and China). Recent experiments (Dungworth *et al.* forthcoming) have shown that a cored and recrystallised microstructure can be produced by annealing hammered copper alloys at relatively low temperatures. A 3% tin bronze was cold-worked (85% reduction in thickness) and then annealed under varying conditions (time and

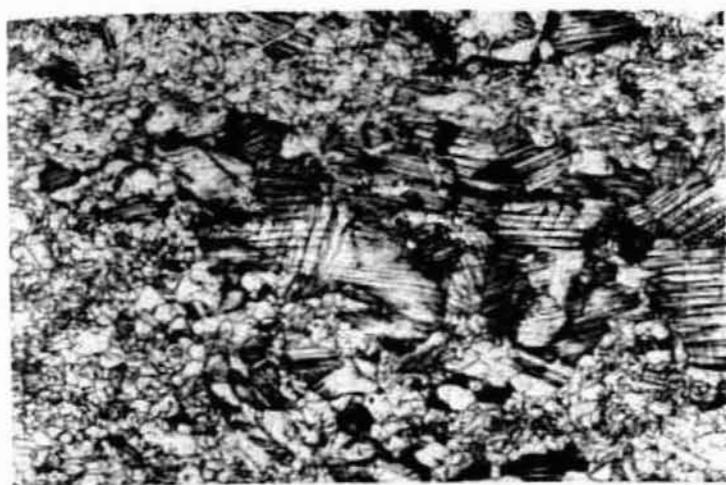


Figure 11. 70923. MB IIC-LB pin. Original magnification $\times 200$. Unetched. Grain boundaries are clearly shown by inter-granular corrosion. The small size of the grains indicates that the metal has been recrystallised. Intra-granular corrosion appears to indicate strain lines, however, compare this with Figure 12 (same sample etched).

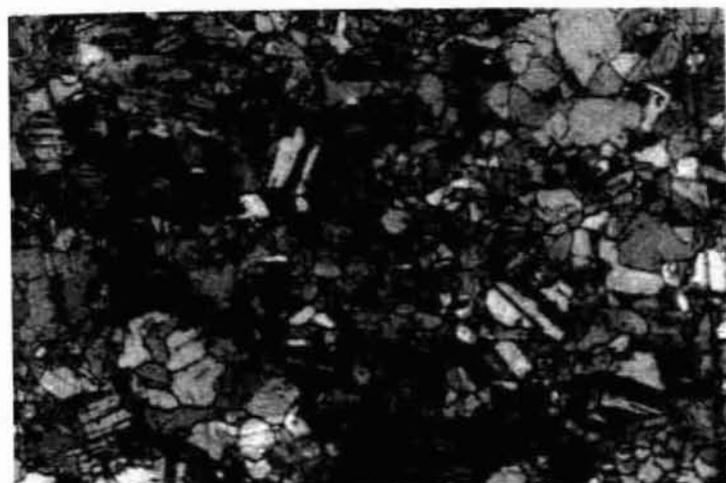


Figure 12. 70923. MB IIC-LB pin. Original magnification $\times 400$. Etched. Shows recrystallised grains and annealing twins. Note that many of the apparent strain lines in Figure 11 have now disappeared. Many of these are likely to have been an intra-granular corrosion phenomena.

temperature). This demonstrated that the cored and recrystallised microstructure can only be reproduced between 500° and 700°C . Below 500°C the metal is not recrystallised and above 700°C the metal is homogenised (further experiments are in progress to explore how these conditions change for different alloys and degrees of prior cold-working). A domestic hearth can produce temperatures in the range 500° to 700°C . The technology for this sort of copper smithing (as opposed to copper smelting or casting) is relatively simple and would require little in the way of specialised equipment, procedures or

knowledge. It is, therefore, possible that smithing was in many cases an everyday activity practised by 'non-specialists'.

3.4 Chemoepitaxy

In almost every case some corrosion products could be detected in the copper alloys from Pella. In some cases corrosion products had formed an outer crust and this often penetrated the remaining metal. The corrosion often followed grain boundaries helping to indicate the microstructure in the polished but unetched condition (Fig. 11). In several cases all of the original metal had been corroded. Despite this some elements of the microstructure could still be seen in the corrosion products (cf. Scott 1991, 43-7). A careful examination of the corrosion products in the Pella samples revealed that they fall into two types: amorphous corrosion products and coherent corrosion products (both types can on occasion be found in the same sample). In amorphous corrosion the copper oxides, carbonates and other compounds show little or no indication of the original microstructure. In coherent corrosion the metal is transformed into corrosion compounds but retains elements of the original microstructure of the metal - chemoepitaxy (Scott 1991, 43). Grain size and shape as well as internal features such as strain lines and annealing twins may be detected in coherent corrosion products (Fig. 13).

In some cases, however, microstructure preserved by chemoepitaxy needs to be interpreted with caution. When polished but before etching, sample 70923 revealed inter-granular and intra-granular

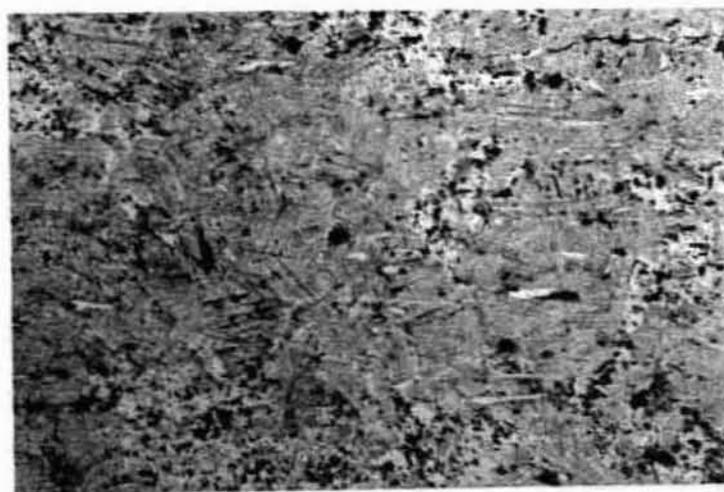


Figure 13. 180052. EB II (late) tool. Original magnification $\times 200$. Unetched. No metal survives in this sample. The original microstructure is preserved to a certain degree by chemoepitaxy. Some of the apparent strain lines may actually be intra-granular corrosion.

corrosion (Fig. 11). This showed that the grains were equiaxed and appeared to show strain lines. After etching, however, no extra strain lines were visible but annealing twins could be clearly seen (Fig. 12). It is suspected that the intra-granular corrosion is here following the same regular crystallographic axes along which slip occurs. For this reason caution was exercised in interpreting all evidence of 'strain lines' in corroded samples. The strain lines were interpreted as evidence of cold-working only where the grains were also distorted. Even in amorphous corrosion copper where all other aspects of microstructure fail to survive, the non-metallic sulphide inclusions may still be observed, as these are less susceptible to corrosion than the metal.

3.4 Summary

The metallographic examination of twenty-seven copper and copper alloy samples of Early Bronze Age to Iron Age date from the site of Pella in Jordan has shown how early copper smiths manipulated their metal to produce a range of artefacts (axes, pins, daggers, arrows, bracelets etc). The most striking feature of the microstructures as a whole is the preponderance of evidence for recrystallisation. The artefacts were all smithed (from blanks of some sort) rather than being cast directly into the final shape. In over half of the cases the samples displayed distorted grains and/or strain lines indicating that the final stage of working occurred below the recrystallisation temperature of the metal. The simultaneous presence of coring and recrystallised grains points to relatively low temperature hot-working or annealing which could have taken place in a domestic hearth. The indications are that artefact production of the kind documented in the microstructures of the Pella metalwork represented a relatively 'low-tech' style of production, perhaps indicative of an industry which was relatively small in scale but the technology for which may have been quite widely dispersed.

4. Metallurgy at Pella during the Bronze and Iron Ages

4.1. The Early Bronze Age

The EBA assemblage (Table 1) comes from late EB I and EB II contexts, and thus dates to the late fourth and early third millennia BC (Bourke 1997, 97, table 2). With one clear exception (80258, see below), the copper from Pella contains no more than

2 % arsenic. In fact, concentrations are often lower, typical of processed copper from the Fay area (see Levy *et al.* 2002, 534, fig. 6). Arsenic is certainly present at levels well below those at which would have had a significant impact on the work properties of the metal (Northover 1989, 113; Bland and Ottaway 1990, 139), and so provides little support for notions of deliberate alloying. In our view the levels of arsenic encountered indicates the presence of arsenic within at least some of the copper-ore bodies from which the metal employed at Pella derived.

The largest single group of EBA metalwork analysed here was a hoard from an EB II destruction deposit within a storage complex on Tell al-Husn, an eminence located south of the present tell-site (Bourke *et al.* 1999, 62–63). The hoard included four flat axes, each quite distinct in typological terms, and a distinctive hook-tanged spearhead with a pronounced v-shaped mic (Philip 1989, 79, tanged spearhead Type 7) and several smaller artefacts. The best stylistic parallels for this artefact come from Ras Shamra in coastal Syria (e.g. Schaeffer 1962, 335, fig. 4.10–11; Contenson 1972, 33, fig. 15). For a preliminary publication of the EBA hoard, see Bourke *et al.* (1999, 62–64, fig. 11).

Three of the axes from the hoard (1800045 and 1800046) were made from copper containing a very low level of impurities (Table 2). Such low impurity levels are consistent with analytical data on EBA artefacts from Arad in southern Palestine (Hauptmann *et al.* 1999, 9, table 3), which are characteristic of EBA copper smelted from ore bodies of the Wadi Araba region (Hauptmann 2000, 130–131). The lead isotope values for 1800045 and 1800046 appear consistent with an ore in the DLS ores of Faynan in southern Jordan (Hauptmann 2000, 137–8, fig. 115), a point supported by the absence of sulphides in the metallographic samples and the presence of a copper-copper oxide eutectic (Figs 8 and 9) (Hauptmann *et al.* 1992, 12), while 1800048 appears consistent with either the Timna or Faynan metal ores (see Appendix 1). As the Timna area has produced very little evidence for Early Bronze Age copper working, in contrast to the Faynan region some 100 km. to the north-east (Levy *et al.* 2002, 429), both possibilities should be considered. Similar lead isotope ratios to those reported from three artefacts have been reported from EBA copper bar ingots recovered from several locations in southern Palestine (Levy *et al.* 2002, 433, fig. 7), placing the Pella artefacts in a broad local tradition of Early Bronze Age metallurgy.

Reg. No.	Site	Description	Date	²⁰⁸ Pb/ ²⁰⁶ Pb	²⁰⁷ Pb/ ²⁰⁶ Pb	²⁰⁶ Pb/ ²⁰⁴ Pb	Consistent with:
PE100201	Pella	Bracelet/ anklet	Iron Age	2.12093	0.87042	17.970	Faynan
PE100222	Pella	Bracelet/ anklet	Iron Age	2.11850	0.86927	17.997	Faynan
PE170058	Pella	Axe	LB I	2.05914	0.83039	18.888	Taurus Mts.
PE170093	Pella	Arrow	LB II	2.05614	0.82865	18.911	Taurus Mts.
PE170202	Pella	Axe	EB IB/II	2.04997	0.82800	18.978	Taurus Mts.
PE180043	Pella	Axe	EB II	2.07630	0.83714	18.704	Cyprus
PE180045	Pella	Axe	EB II	2.12036	0.86961	18.017	Faynan
PE180046	Pella	Axe	EB II	2.11822	0.86941	17.980	Faynan
PE180048	Pella	Tool	EB II late	2.10000	0.85529	18.330	Timna/ Faynan
PE42116	Pella	Bracelet	LB I	2.08260	0.84337	18.596	Cyprus
TES538	Tell esh-Shuna	Lugged axe	EBA?	2.10022	0.85779	18.249	Timna/ Faynan

Table 4. Results of lead isotope analyses of artefacts from Pella (Isotracer Laboratory, Oxford, 2nd March 2000).

In contrast, axe 180043 contained higher levels of arsenic (1.5%) and iron (1.2%) on surface analysis although these were lower on the metallographic samples. It is also distinguished by the presence of sulphide inclusions visible in the metallographic sample (Table 3, Fig. 7), and on the basis of lead isotope data, which point to a derivation from the copper ores of Cyprus (Table 4, Fig. 16). This axe provides the first clear evidence for the import of Cypriot copper to the Levantine mainland. In addition, lead isotope data for chisel 170202 appears consistent with an origin in the region of the Taurus mountains of Anatolia (Table 4, Fig. 15).

The hoard was found in a destruction deposit, and is seen by the excavators as consisting of a group of material destined for recycling (Bourke *et al.* 1999, 64). If correct, it seems reasonable to assume that it included material drawn from that which was locally available, in which case the fact that it contained artefacts in metal originating at more than one ore-source is of particular interest. Firstly it implies that the circulation of metal was already a highly complex affair by the early third millennium BC, and that the emphasis upon metal from a single-source documented at Arad (Hauptmann *et al.* 1999) may not have been typical of all south Levantine EBA sites. Secondly, the fact that artefacts with quite distinct lead isotope signatures were recovered from a single hoard, where they awaited recycling, suggests that the mixing of copper from different ore sources may pose problems for the interpretation of lead isotope data from at least the beginning of the third millennium BC.

Broadly contemporary material from settlement contexts includes two chisels from the same area of the site, while another axe, an unspecified tool and

two pins come from settlement contexts on the main tell. No EBA I–III graves have yet been excavated at Pella. That said, the evidence from EBA cemeteries at Jericho and Bab al-Dhra (Kenyon 1960, 1965; Schaub and Rast 1989) suggests that EBA graves do not generally yield large quantities of metal artefacts, and that we will remain dependent upon settlement and hoard contexts for the bulk of our material.

The employment at EBA Pella of copper from sources outside the southern Levant may appear counter-intuitive, given the evidence for large-scale copper extraction and processing at Faynan during the EBA (Hauptmann 2000; Levy *et al.* 2002). While non-local copper was circulating in the region during the Chalcolithic period, this mainly concerned the use of a specific copper-arsenic-antimony ternary alloy for the production of certain forms of high-status material (Shalev 1994), rather than copper employed for more utilitarian artefacts. However, the use of copper originating in Cyprus is consistent with the presence at Pella of a hooked-tang spearhead of a type with its best parallels at Ras Shamra on the Syrian coast (itself located just over 100 km. from Cyprus), and with evidence from nearby Tell al-Shuna for the use of copper from Anatolian sources during EB I (Rehren *et al.* 1997). What is particularly striking about the Pella data is that with two of the five EBA artefacts made of non-local copper, it offers a striking contrast to the recently published evidence from contemporary Arad in the northern Negev, where the lead isotope analysis of copper artefacts indicated a near-exclusive use of metal from the Faynan ores (Hauptmann *et al.* 1999, 11–12, fig. 4).

Other EBA objects indicate broadly similar compositions, and are generally low in impurities. More

unusual is the presence of a pointed object of square section (180052) that surface analysis indicated as containing 3.36 % tin. However, examination of the metallographic sample revealed tin only at the limits of detection. This situation is in clear contrast to the marked loss of tin at the surface that was apparent in other small tin-bronze artefacts when analyses were undertaken both at the surface and across the metallographic samples, e.g. 42116, 60018, 170065 (see Table 2). One possibility is that these results indicate the presence of a tin-enriched surface layer, perhaps to achieve a desired visual effect. While perhaps unlikely in the case of a tool, it is possible that this small pointed object represented something rather more special, such as part of a miniature weapon which had become detached from an armed cult figurine of a kind well documented in the Levant during the earlier second millennium BC (Seeden 1980). As the excavator has observed that this tool came from a context that produced several Middle Bronze Age sherds alongside a large quantity of EBA pottery (Bourke pers. comm.), it cannot be taken as a reliable EBA instance of tin-bronze. The absence of evidence for the use of tin-copper alloys from the large quantity of metallurgical material recovered from the 'manufactory' site of Khirbat Hamra Ifdan in the Faynan area (Levy *et al.* 2002, 433), supports the view that tin played little part in the copper industry of the southern Levant during EB I-III.

The lugged, flat axe from Tell al-Shuna, belongs to Miron's (1992) class of 'lugged blades', and is quite distinct, typologically and in alloy composition, from the axes from Pella. It came from a mixed deposit, relatively high-up in a 3 × 3 m. sounding excavated in 1994 (Area L, Context 875), and the datable south Levantine parallels listed by Miron (1992, 42-3) run from the Middle Bronze Age through to the Iron Age. Although the lower levels of the sounding produced good *in-situ* EBA III material, including quantities of Khirbat Karak Ware, the axe did not derive from these deposits. However, the presence of a good parallel in a group of metal objects from Tell al-Judaiah Phase H (Braidwood and Braidwood 1960, 376, fig. 293.1), which is broadly equivalent to EB III in terms of the sequence in the southern Levant, does open the possibility that this axe represents a redeposited EB III artefact. As the metal includes both tin and lead, at levels notably higher than those observed in other EBA objects, the axe might represent an early example of a tin-lead bronze; however, this cannot be demonstrated. While the lead isotope composition appears consistent with an origin in the copper deposits of the Wadi Araba (see Table 4), the possi-

bility that lead was added intentionally to the copper must be considered, as this practice is now well-documented during the second millennium BC (Philip 1991; Rosenfeld *et al.* 1997, 859, tab. 1, 2; Shalev 2000, 279, tab. 13.2).

The single example (80258) of the distinctive narrow-bladed daggers characteristic of the EB IV period (Philip 1989, 102) provides a very different picture. This artefact, which dates to the late third millennium BC, comes not from the main tell, but from a cemetery excavated in the Wadi Hamme a few kilometres north of Pella (Potts *et al.* 1991, 123-128). It is badly corroded, and thus the analyses can provide no more than a general indication of its original composition (Table 2). However, high levels of arsenic recorded (6-7%) have not been observed in any of the other artefacts from Pella, and appear in keeping with practices documented through the investigation of EB IV narrow-bladed daggers from other sites in the southern Levant. A proportion of these have been shown to contain elevated levels of arsenic (Philip 1991; Shalev 1998). In fact, the investigation of a number of such weapons has provided good evidence for the presence of an arsenic-enriched surface layer (Shalev 1998, table 2), composed of the copper-copper arsenide (Cu_3As) eutectic and which would give the surface a distinct silvery appearance. Budd and Ottaway (1990, 138) note that this process, known as inverse segregation (Meeks [1993, 267-271] provides a detailed account of the metallurgy of this process), could be encouraged in alloys containing less than 8% arsenic by the use of casting techniques which would result in rapid cooling of something to which thin castings such as dagger blades were already predisposed, although there would be a corresponding depletion of arsenic in the core, which would therefore be less hard than the overall concentration of arsenic in the dagger might suggest. Budd and Ottaway also observe (1990, 136-139) that should the arsenic content rise above 6-7% the blade would become increasingly difficult to harden by cold working, and would be prone to cracking.

On present evidence the exploitation of this particular property to obtain a distinct surface coloration represents (as far as the EBA is concerned) a practice peculiar to the EB IV period, and perhaps to narrow-bladed daggers in particular. The evidence therefore points to the deliberate selection of high-arsenic copper for the production of some EB IV narrow-bladed daggers. This practice appears as a complete contrast to anything thus far observed in the earlier part of the third millennium BC, and suggests a desire to produce 'silvered' daggers, or perhaps even the g

colour which is taken by the tarnished surface (Northover 1989, 115). This development should probably be seen as a response to contacts with contemporary Syria where documents from Tell Aardikh /Ebla indicate that daggers made from, or decorated with, precious metals were highly desirable prestige objects (Archi 1985; Waetzoldt 1990).

It is probably worth restating that while there is now clear evidence for the working of the Faynan copper ores during the EB IV period (Adams 2000, 395–396), the low impurity levels which characterise this metal (see above) make it almost certain that the copper employed in the production of high-arsenic daggers originated outside the southern Levant. Thus despite the apparent insularity of the period, which was characterized by highly regionalized material culture, dispersed settlement, and a largely domestic mode of production (Palumbo 2001, 260), the presence of high-arsenic copper, points to participation in longer-range procurement networks when this was deemed worthwhile.

4.2. *The Middle Bronze Age*

The sample from the Middle Bronze Age period comprises 21 artefacts, 14 from graves and 7 from settlement contexts. The grave material includes 12 objects from Tomb 62 which spans the transition from Middle to the Late Bronze Age (Smith and Potts 1992, 69). It consists of eight toggle-pins and four rings, probably to be identified as ear-rings. A range of examples are illustrated in the publication of the tomb (Smith and Potts 1992, 76–77, pl. 61: 16–19). This group of personal ornaments should make a useful comparison for those from the Iron Age Tomb 89 (see below).

Both the rings and the pins show considerable variability in tin content, with surface analysis suggesting a range from very low to 7–8% tin. However, given that the concentrations of tin recorded in the metallographic samples were frequently higher, exceeding 10% in several cases, the indications are that by far the majority of these artefacts were produced in medium-high tin-bronze. There appears to be no obvious relationship between alloy selection and pin typology. Arsenic is generally present at low levels, often close to or below limit of detection. The alloy composition of the assemblage from Tomb 62 appears rather less homogenous than that of Tomb 89. However, the greater variability may be attributable, at least in part, to the fact that the former which contained between 100 and 150 interments and over 2000 artefacts (Smith and Potts 1992, 69–70), was almost certainly in use for a period spanning several decades. Three pins and one ring

from settlement contexts were examined, as well as several object types not available among the grave material. They show a similar range of tin concentrations to the grave material. When combined with the typological similarities between the artefacts from grave and settlement contexts, this suggests that the personal ornaments used in both contexts were, by and large, drawn from the same artefact population.

The curved-bladed knife (60118) from Tomb 62 (Smith and Potts 1992, 76, pl. 61.20) belongs to a type which is well documented in the southern Levant during the MBA (Philip 1989, 141). While the results of the surface analysis were clearly affected by corrosion, the data does appear at least to confirm that the object was made using tin-bronze, as might be expected in an object which would require a hardened cutting edge, a point confirmed by the metallographic evidence for substantial cold-working along the edge.

Two pins (32214 and 170161) revealed in the region of 4–5% lead, rather higher than one might expect to have resulted from the presence of lead in the original copper ore, and most likely indicative of its deliberate addition to the alloy. In the case of the latter the high lead levels were detected both through the analysis of the surface and metallographic samples. This view receives support from the generally low levels of lead in the other MBA artefacts. Occasional instances of similar copper-tin-lead alloys have been reported from other groups of MBA material (e.g. Philip 1991, 1995b; Rosenfeld *et al.* 1997; Shalev 2000). The significance of this practice is not clear, although one instance is a slender toggle-pin with a ribbed head, suggesting that lead may have been added to improve the fluidity of the molten metal and thus ensure even filling of what must have been a rather narrow matrix within a mould.

4.3. *The Late Bronze Age*

The corpus of artefacts from LBA contexts spans the entire LBA and comprises four artefacts from two different LB I graves, fourteen arrowheads from a hoard and eleven objects of various types from settlement contexts. Arsenic contents are generally below 0.5%, exceeding 1% in only one instance. Lead rarely exceeds 0.5%, and iron contents are generally low. On the basis of surface analysis, confirmed by the evidence of the one available metallographic sample (170093), the arrowheads from the LBA hoard were made from unalloyed copper. In contrast, tin is nearly always present in measurable quantities in the other LBA artefacts,

even when only surface analysis was possible. Apart from the arrowheads, the range and typology of the LBA artefacts is broadly comparable with that from the MBA, although the LBA group has produced no examples of leaded tin-bronzes. However, this observation has no statistical significance. Pins were generally made from bronze containing varying proportions of tin; such variability in alloy selection for pin production is seen as late as the Persian period (Muhly and Muhly 1989, 290). Oddly, one pin (170066) was made from copper containing a substantial admixture of silver – presumably to obtain a particular colour. Metallographic examination confirmed that this artefact had been considerably corroded and that the primary copper alloy phase had been removed preferentially, leaving a pale blue silver-rich, secondary phase (Fig. 10).

The projectiles come from a single hoard found in a deep, stone-lined silo, dated to the end of LB IIA (Bourke *et al.* 1999, 64, fig. 6). They were not broken or damaged, and there is no evidence to suggest that this group was intended for recycling. They fall into two main groups on typological grounds, the familiar, long, leaf-shaped arrowheads typical of the LBA (Bourke *et al.* 1999, 64, fig. 6. 1–11; Philip 1989, 146–147), and a group of three blunt-tipped ‘stunning’ bolts. The bolts consist of a straight square-section tang as in the leaf-shaped arrowheads, but the blade is made in a cylindrical or ‘teardrop’-shape, with a blunt tip (Bourke *et al.* 1999, 64, fig. 6. 12–14). These may have been intended for shooting targets, perhaps small game, where it was intended to produce a high-velocity, blunt impact rather than penetrate the body and cause bleeding. Preservation was generally good. While surface analysis indicated that all three bolts contained tin in measurable quantities, more than 1% in two cases, this was not the case with the leaf-shaped arrowheads. On the other hand, unlike the bolts, surface analysis of the arrowheads revealed arsenic at between 0.2 and 2.2%, while many also contained measurable quantities of antimony, something not documented in the other LBA artefacts. There is a suggestion therefore, that the bolts and the arrowheads were made from two different batches of metal. The single leaf-shaped arrow which was sampled for lead isotope analysis indicated that it was produced using copper from a source in the Taurus mountains of Turkey.

Ancient metalworking at sites such as Pella, which were located some distance from ore sources, would almost certainly have required a degree of opportunism as regards selection of material, and so it would be unreasonable to expect completely clear-cut distinctions to emerge. Given that qualification,

the data do appear to point to a distinction between the metallurgy of the leaf-shaped arrowheads, that of the bolts, and finally of the majority of the remaining LBA artefacts many of which contain higher concentrations of tin. Given that the majority of LBA artefacts were good tin-bronzes, the very low levels of tin in the leaf-shaped arrowheads might suggest that the metal from which these were produced contained relatively little local scrap. This could be taken to indicate that there existed some distinction between the metal used for the bulk production of projectiles, and that used for other tasks and it is tempting to attribute this to some degree of administrative involvement in the metallurgical processes at LBA Pella.

Axe 170058 is an example of an Egyptian axe form termed the Lugged Asymmetrical axe by Davies (1987) and Type G by Kühnert-Eggebrecht (1969). The form is characteristic of the Eighteenth Dynasty (Davies 1987, 53), and is one of a small number of such axes from the LBA southern Levant (Miron 1992, 92). Analysis of a drilled sample revealed a tin content of 11.3% (Table 5), reflecting the need for a hardened cutting edge, a pattern confirmed by a LB II chisel (90208), a metallographic sample from which contained over 13.0% tin. Interestingly, lead isotope data suggests that the axe was made from copper from sources in the Taurus mountains (Table 4, Fig. 15), and cannot therefore answer the question of whether or not it was made at Pella, or represents an Egyptian product. Most of the LBA toggle-pins contained tin although in variable quantities. However, none contained significant quantities of lead, and there is no obvious association between pin-style and choice of alloy.

Another good tin bronze, a bracelet also from LB I burial context, has been identified as made from Cypriot copper (Table 4, Fig. 16). That some have suggested (Budd *et al.* 1995; Knapp 2000, 42–43) that the large number of oxhide ingots which are often taken as made from copper from Cyprus, may actually represent the development of a common ‘pool’ of metal deriving from extensive mixing of ores as a result of the sheer scale and complexity of the metals trade in the east Mediterranean during the Late Bronze Age. In addition to regular trade in metals (Sherratt and Sherratt 1991), and the now extensive evidence for metal recycling documented in both Cyprus and Sardinia towards the end of the Late Bronze Age (Knapp 2000, 43–44, with further references), the documentary sources also indicate the large scale, long-distance movement of metal in the form of quantities of copper-alloy material taken as tribute, war booty

used as part of diplomatic gifts. Examples include bronze armour, vessels and tools taken by Tutmosis II following the battle of Megiddo (Pritchard 1958, 81) or quantities of bronze torques, armour, shields and various weapons being sent as royal gifts e.g. Amarna letter No. 22 (Moran 1992, 55–56).

A group of artefacts dating to the thirteenth century BC was excavated in Area XXXIII 15.4, interpreted as part of a cult structure. The group, which included two cymbals and two balance pans, is interpreted by the excavator as representing cultic paraphernalia, and came from the floor of a destruction that produced Mycenaean IIIB pottery (Bourke 1999, 152–155; Bourke *et al.* in press). Analysis, multiple analyses in some cases, revealed some variability between the various artefacts, but all were made from tin bronze. Although well preserved, the majority of these artefacts were made from relatively thin metal, and caution is required in the interpretation of the tin concentrations obtained from surface analysis. Context XXXIIG 107 produced additional material also dating to the Late Bronze Age / Iron Age transition. This included what appears to have been the rectangular tang of a dagger with a rivet still in place, and a harpoon with a distinctive barbed head. These were more robust and were likely to have a well preserved metal core, and appear to have been made from copper containing a low level of tin. The low tin contents may suggest some of these artefacts were produced from metal containing an admixture of bronze scrap.

4.4. The Iron Age

With a few exceptions (Curtis [ed.] 1988; Muhly and Muhly 1989; Moorey 1994), there has been relatively little interest in the technology of copper-alloy artefacts from Iron Age contexts in the Levant. This, in part, appears to reflect the gradual replacement of bronze by iron for the production of tools and weapons and its relegation to a more specialist repertory of artefacts, mainly those produced by casting or the working of metal sheet (Moorey 1994, 264). The latter includes an impressive range of frequently high-status objects such as bowls, cauldrons and drinking sets, to which the colour and lustre characteristic of bronze objects would have been significant. Of course, fine objects of this kind are not always readily accessible for laboratory analysis. However, when chemical data are available it appears to indicate, as might be expected in the manufacture of high-quality products, quite careful selection of alloys according to function, the technical requirements of sheet-metal working and what might be termed 'workshop

tradition' (Hughes *et al.* 1988). Thus groups of analyses drawn from such specialist artefacts may not be representative of Iron Age bronze-working practices generally, which as Craddock and Giunilia-Mair (1988, 321) have observed, appear to have involved considerable variability in alloy selection.

The Pella sample consists of six rings and bracelets, made by similar techniques and originating from a single tomb and so should offer an opportunity to assess the potential variability within a reasonably coherent group of artefacts. The tomb is dated to the later eleventh or tenth century BC, i.e. transitional Iron I–II (see Potts *et al.* 1988, 148; Bourke 1997, 113). Two large, heavy rings (Nos 100222, 100201), perhaps to be identified as anklets, are very similar in form, and composition. Both are composed of an alloy containing more than 10% tin, and a little over 1% lead. Neither of the pieces examined by metallography showed evidence of strain lines, although both indicated annealing. Presumably in the absence of a cutting-edge, there was no need for final cold-working. With the exception of one artefact (No. 100214) from which the results of surface analysis and that of a metallographic section were in good agreement at a little over 5%, the Iron Age bracelets revealed tin contents of around or above 10%, and concentrations of lead around 1%, suggesting a fairly standard set of alloying practices.

The main technical advantages in using bronze as opposed to copper for the production of artefacts which required neither great hardness nor complicated castings, lay in the lower melting point and longer casting interval which bronze offered. Thus the clear preference for using tin-bronze for personal items requires some consideration. Analysis of a large group of Persian period metalwork from Tell Michal in Palestine (Muhly and Muhly 1989, 269) revealed that bronze was preferred for the production of rings, leaded-bronze for bracelets, while unalloyed and arsenical copper remained in use for certain other categories of artefact. This suggests that rather than bronze having become the 'industry standard' for smiths, the choice of tin-copper alloys for rings, pins and so on reflected the desire to obtain artefacts with the distinctive yellowish colour characteristic of such alloys (Moorey 1994, 253), perhaps in emulation of gold: that is aesthetic, rather than technological considerations.

Two of the Iron Age artefacts gave lead isotope signatures consistent with copper from the Faynan DLS deposits (Hauptmann 2000, 137–8, fig. 115). This is consistent with the archaeological evidence that indicates the renewed exploitation of these ores

in the Iron Age following a hiatus during the Middle and Late Bronze Ages (Hauptmann 2000, 87–89, table 7). In a recent paper Klein and Hauptmann (1999) discussed metallurgical debris from Khirbat al-Dharih indicating the use of copper-tin-lead alloys at that site during the Iron Age. The site is located on the south Jordanian plateau some 40 km. north-east of the Faynan copper deposits, and the authors have tentatively suggested the Faynan ores as a possible source for this material (Klein and Hauptmann 1999, 1079). This suggestion receives support from the new evidence from Pella, which points to the existence of bronze-workshop(s) using copper sourced from south Levantine ores in the early Iron Age.

The exploitation once more of Transjordanian ores following on from what has been termed the 'commodification' of metals during the Late Bronze Age (Knapp 2000, 47) and which may have witnessed the creation of 'pooled' copper resulting from the mixing of metal derived from a range of ores, is interesting, and highlights the need for further investigation of the status and organization of copper metallurgy after the great internationalism which characterized the Late Bronze Age.

5. General conclusions

The Pella copper artefacts reveal a number of points that have important implications for developments in metallurgy generally.

First, while the EBA samples confirm the dominance of unalloyed copper as documented elsewhere (Shalev 1994; Hauptmann *et al.* 1999), the presence of copper likely to have originated from Anatolian and Cypriot sources was unexpected, and its use alongside metal from Faynan reveals that EBA resource acquisition systems were highly complex. The contrast between the near-exclusive concentration on Faynan ores at EBA II Arad (Hauptmann *et al.* 1999) and the much wider range of sources in use at contemporary Pella is striking. When viewed in the light of data for the use of metal from northern sources at Tell al-Shuna during the EB I (Rehren *et al.* 1997) this points to differences in the degree to which EBA communities in the northern and southern parts of the southern Levant participated in communications networks running along the Mediterranean littoral. The evidence from Pella therefore supports the claim (Philip 1999, 50; 2002, 219–220) that the well-documented second millennium BC east Mediterranean trading network (Sherratt and Sherratt 1991), had a thriving fourth/third millennium BC predecessor, the impor-

ance of which has frequently gone unnoticed by archaeologists, largely because the network was focused not upon pottery, but upon the transmission of consumables, raw materials and technological innovations, which are often less easy to identify in the archaeological record.

Second, in the light of the evidence for a substantial degree of metal recycling at sites in north and west Syria during the third millennium BC (Northover 2000, 113), the presence of artefacts made from copper from quite different ore sources within a single hoard at EB II Pella, points to the possibility that the mixing through recycling of metal from quite different geological settings – an issue which has been raised by various scholars (e.g. Budd *et al.* 1995) – may present a problem for lead isotope studies as early as the beginning of the third millennium BC. Although this does not appear to be the case in the current programme, this issue clearly requires further research.

There is no evidence from Pella for any significant use of tin during the EBA. Even were the possible bronze artefact (180052) of demonstrable stratigraphic integrity, this would not alter the overall picture of a reliance upon unalloyed copper. If the lugged flat axe from Tell al-Shuna was a redeposited EBA III find then it would represent a mid-third millennium BC instance of tin-bronze, but this remains uncertain. In contrast, the single EB IV dagger belongs to a group of high-arsenic alloys which appear to be characteristic of that particular period, and the production of which presumably reflects factors peculiar to that space-time locale.

The Middle and Late Bronze Ages witness the appearance of tin-bronze in quantity. While there is a general preference for producing edged tools and decorative items in good tin-bronze, presumably for reasons of hardness and colour respectively, its use was by no means universal. Alloys with low concentrations of tin may indicate the use of recycled metal. Many projectiles appear to be manufactured from less costly alloys, and there is evidence which points to an association between specific groups of material, projectiles in particular, and alloy composition, perhaps indicative of batch-production in a particular workshop.

While occasional instances of leaded-bronze, and a single instance of a copper-silver alloy are documented, there is little evidence for the use of high arsenic alloys after EB IV. Neither the evidence of typology nor that of artefact composition points to the production of a separate class of metalwork specifically produced for use in burial contexts. Rather, grave and everyday goods appear to have been drawn from a single artefact population (cf.

Dugay 1996, 181; Halotte 1995, 111–114; Philip 2001, 199–200). It is also worth underlining that either the EBA nor the LBA hoard was composed of artefacts which could be shown to come from a single ore source. Rather each hoard appeared to be composed of artefacts produced for other purposes, and which were subsequently interred as a group.

In comparison to that of the Early Bronze Age, the lead isotope data for the Late Bronze Age appears to indicate a shift away from the use of copper from local sources, and towards more extensive participation in wider Mediterranean trade networks. This is in agreement both with the situation in Faynan which shows little evidence for exploitation at this point (Hauptmann 2000), and the expectations of current models for the development of commercial networks in the east Mediterranean at this time (Sherratt and Sherratt 1991; Knapp and Cherry 1994).

The deliberate addition of lead to tin-copper alloys is documented in the southern Levant from the early second millennium BC (Philip 1991; 1995b; Rosenfeld *et al.* 1997, 859, tables 1, 2). Accordingly, one aim of the present research project was to assess the impact of the deliberate addition of metallic lead to copper alloys on the lead isotope ratios of the resulting artefacts by a comparison with those of contemporary objects from the same site containing the lower levels of lead consistent with an origin in the copper ores. In this way it was hoped to examine the potential impact of the recycling of tin-lead bronze artefacts upon lead isotope ratios in Bronze Age copper artefacts from the Levant. Unfortunately, it proved impossible to pursue this line of enquiry because the Pella assemblage produced relatively few objects containing significant concentrations of lead, and those that did were unable to provide the samples required for lead isotope analysis. However, the presence of tin-lead bronzes at Pella during the second millennium BC highlights the need for researchers to investigate this matter in more detail.

We believe that the present study has demonstrated quite clearly the value of an investigative programme that can deploy multiple analytical techniques in pursuit of a clear research design. While such approaches have been widely recommended, a variety of logistical and financial problems have militated against their achievement in practice. What is required now is not just more analyses, regardless of provenance or context, but carefully focused programmes, which will provide data complementary to the current project, and will address issues which we have raised, but have been unable to explore fully, because of the limitations of our material. Only in this way will researchers be able to develop an

understanding of patterns, and perhaps more revealing, discontinuities, in Levantine metallurgy, at general, site and context-specific levels, and thus begin to comprehend ancient metallurgy in terms of its social and economic setting.

Appendix: report on lead isotope analyses of 11 samples of copper-based artefacts from Pella and Tell al-Shuna.

Sophie Stos

Methodology

The eleven samples of copper-based artefacts submitted for lead isotope analyses by Dr Philip were analysed in the Isotrace Laboratory using Thermal Ionisation Mass Spectrometry. The analytical methodology of lead isotope measurements is described in detail in the first lead isotope database from the Isotrace Laboratory published in *Archaeometry* (Stos-Gale *et al.* 1995). The identification of the origin of copper ore used for making the artefacts is based on one-to-one comparisons of lead isotope characteristics of samples of ores from known locations and artefacts. The database of lead isotope characteristics of ore deposits used for comparisons included the following number of ores and copper slags from various deposits: 290 from Turkey, 650 from Cyprus, 260 from the Near East (Jordan, Egypt, Israel, Saudi Arabia, Oman) and 1500 from the Aegean. Most of the data is published in papers listed in the bibliography, although some comparisons are with unpublished data from the Isotrace Laboratory.

Discussion of the results

Dr Philip supplied, together with the samples of artefacts submitted for lead isotope analysis, the information about the elemental composition of these metals. However, all samples are routinely analysed before lead extraction using the semi-quantitative EDXRF method. The results of these analyses are summarised in Table 5. Only two samples submitted for lead isotope analysis contained lead in quantity of about 1% (538 and 100201). Lead in this amount is not unusual in copper metals smelted from polymetallic ores. This point will be discussed further in

I/D	Cu%	Pb%	Sn%	Ni%	As%	Fe%
100201	87.5	1.1	11.4	<0.2	<0.2	<0.1
100222	84.5	0.5	15.1	<0.2	<0.2	<0.1
170058	88.4	<0.2	11.3	<0.2	<0.2	0.3
170093	99.9	<0.2	<0.2	<0.2	<0.2	<0.1
170202	99.8	<0.2	<0.2	<0.2	<0.2	<0.1
180043	96.8	<0.2	<0.2	<0.2	2.5	0.6
180045	99.9	<0.2	<0.2	<0.2	<0.2	<0.1
180046	99.9	<0.2	<0.2	<0.2	<0.2	<0.1
180048	99.5	<0.2	<0.2	<0.2	<0.2	0.3
42116	86.9	<0.2	12.9	0.2	<0.2	<0.1
538	90.9	1.3	7.7	<0.2	<0.2	<0.1

Table 5. Results of 'semi-quantitative' EDXRF analysis undertaken at Oxford on drilled samples submitted for lead isotope analysis.

connection with the lead isotope composition of these samples. Differences between the Durham analytical data and our EDXRF analyses are most likely due to the inhomogeneity of lead distribution in the whole artefact and do not affect any of the conclusions drawn from their lead isotope composition.

The lead isotope composition of the artefacts from Pella and the axe from Tell al-Shuna (measured in the Isotraces laboratory) are listed in Table 4. In the same table the data is labelled with the ore deposit which is most consistent with the observed lead isotope ratios.

On Figure 14 the lead isotope (LI) data is presented in the form of two dimensional mirror-image plots for six of these artefacts and the LI data for ores from the mines of Faynan in Jordan and Timna in Israel. Four objects, two Iron Age bracelets (100201 and 100222) and two EB II axes (180045 and 180046) show nearly identical lead isotope compositions fully consistent with the copper ores and ancient copper from the smelting sites in the Wadi Araba (Faynan). The axes are made from 99% pure copper, the bracelets are high-tin bronzes with 0.5–1% of lead. The elemental and lead isotope compositions of these metals compare very well with the copper from smelting sites of Ras al-Naqab and Barqat al-Hatiya (EBA II/III), Faynan 5 and Khirbat al-Nahas (IA II) all located close to Wadi Faynan on the east side of the Wadi Araba (Hauptmann *et al.* 1992, 22, table 6 and 24, table 7). The amount of lead in the copper metal from the Iron Age smelting sites varies from 0.7% – 6.4%, the addition of 15% of tin would decrease the overall percentage of lead in the alloy by 15%.

The tool from Pella (180048) and the axe from Tell al-Shuna have lead isotope compositions con-

sistent with copper ores from Timna and the MBS ores from Faynan in the Wadi Araba (Gale *et al.* 1990; cf. Hauptmann 2000, 58–59, fig. 31). These lead isotope ratios appear also amongst the loaf-shaped ingots excavated by Galili in the sea off the coast of Israel, bronzes from Ras Shamra (Isotraces unpublished data) and amongst bronzes from al-Amarna (Stos-Gale *et al.* 1995b).

Two LBA artefacts, an axe (170058) and an arrow (170093) show lead isotope composition identical with artefacts from LBA Anatolian site (Fig. 15). The closest match for these lead isotope ratios is found amongst the ores from the Taurus Mountains, specifically the region of Bolkardag (Nigde). The history of ancient mining for copper in this area is not fully attested (Wagner *et al.* 1989; Yener *et al.* 1989 and 1991), but there are many Bronze Age artefacts from Egypt (Stos-Gale *et al.* 1995b), Greece, Cyprus and Ras Shamra falling in this lead isotope range and so far there are no other copper ores from the eastern Mediterranean matching them. The lead isotope data for these two artefacts from Pella are compared with ores from Bolkardag and artefacts from Anatolian sites on Figure 15. A third artefact from Pella, an EB axe (170202) has somewhat different lead isotope composition, but is also consistent with one sample from ore from the Bolkardag (Fig. 15).

On Figure 16 two artefacts, an EB II axe (180043) and an LB I bracelet from Pella (42116) are compared with the LI data for ore from Cyprus. They are both consistent with ores from the north eastern foothills of the Troodos Mountains where many mines have been exploited since the EB. (Larnaca axis) (Stos-Gale *et al.* 1997; Stos-Gale and Gale 1994).

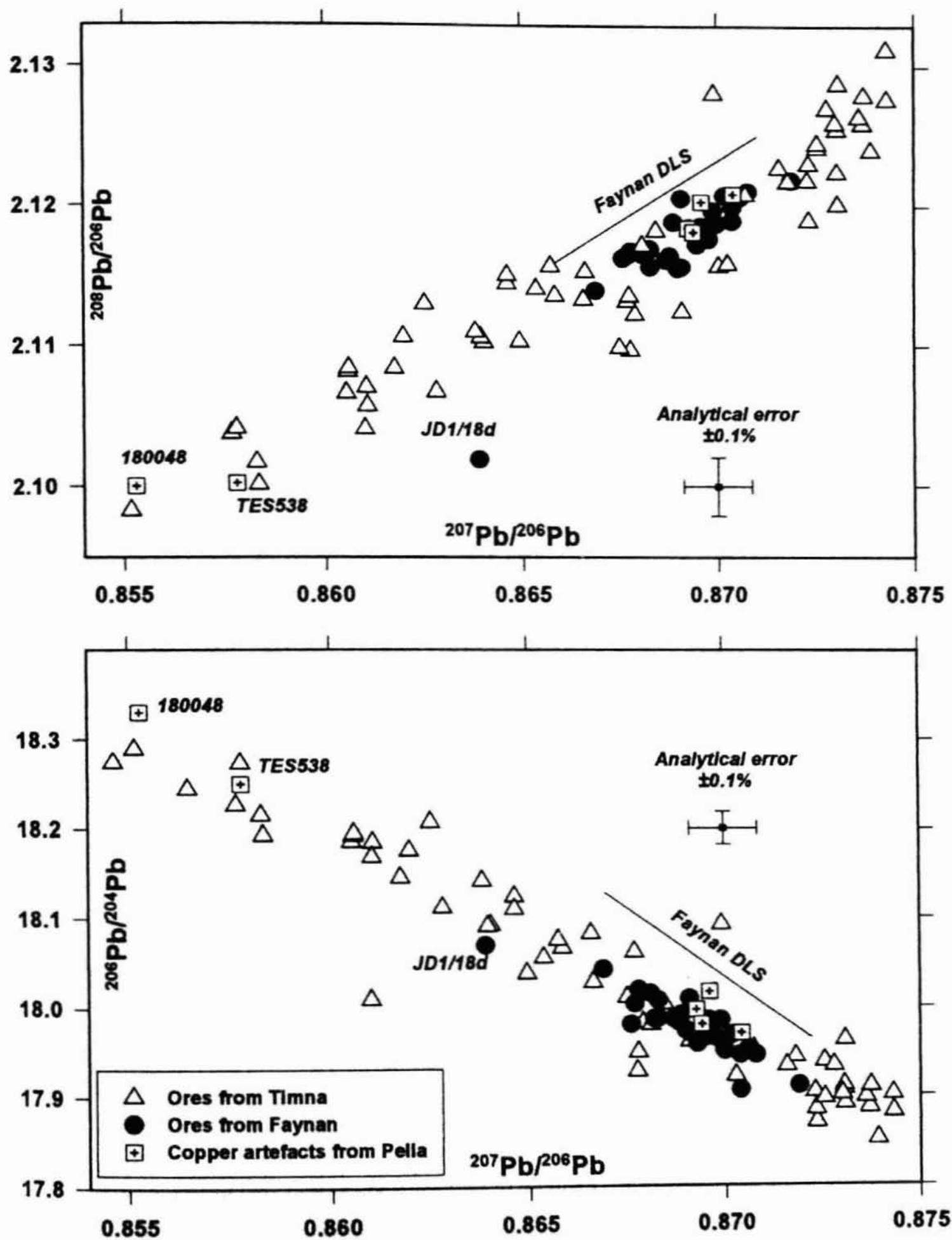


Figure 14. Lead isotope data for ores from Timna and Faynan and artefacts from Pella

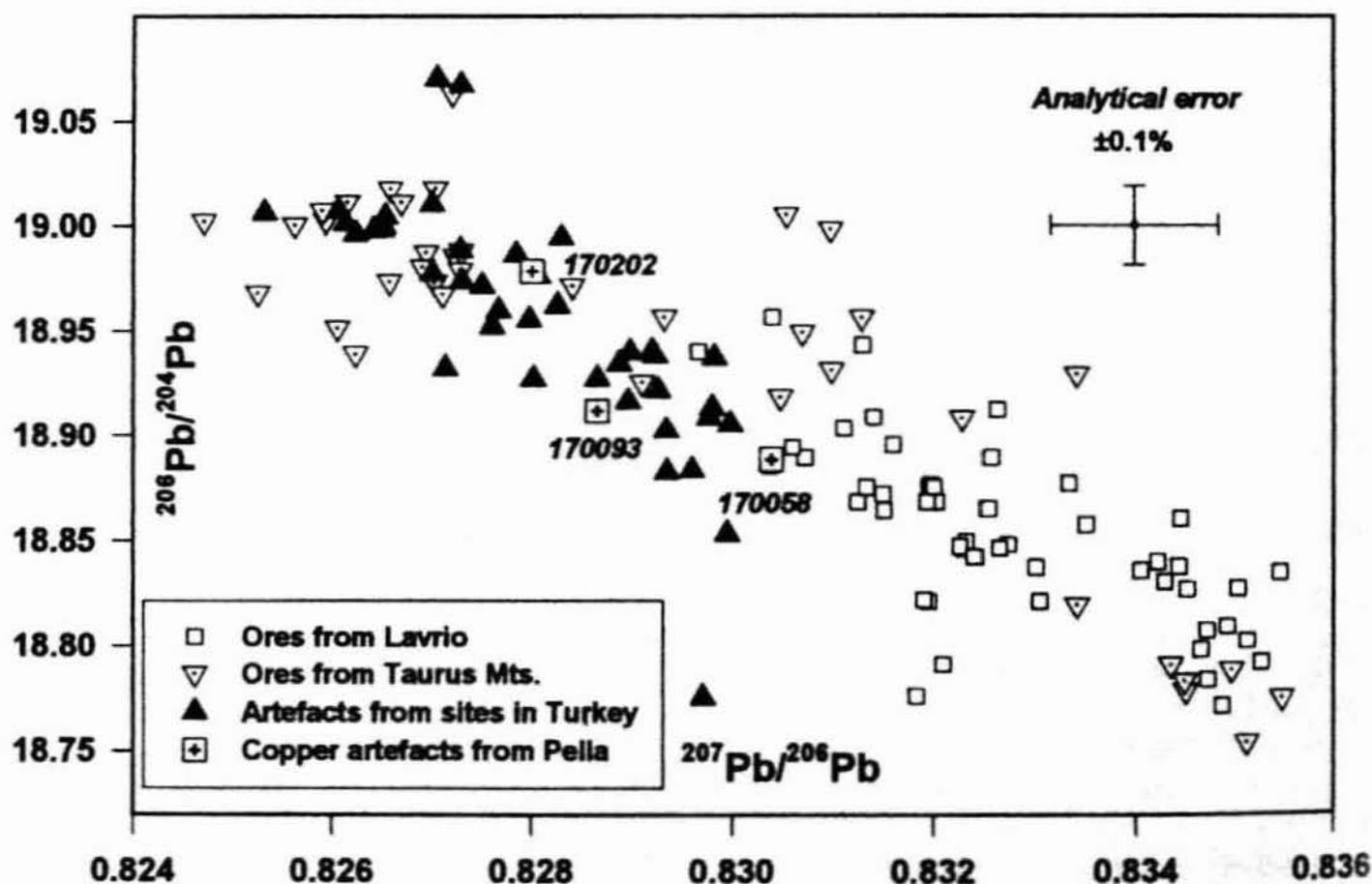
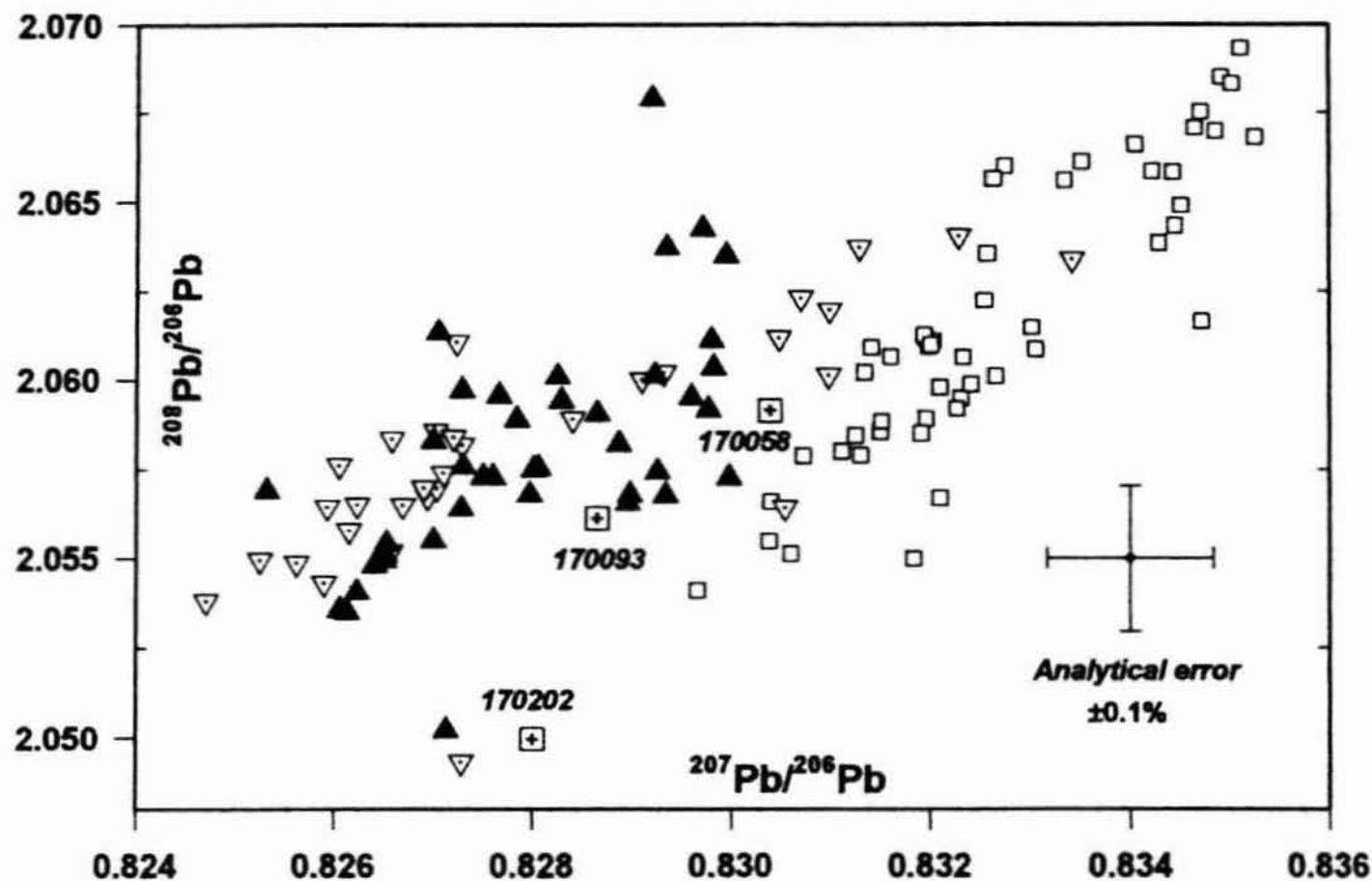


Figure 15. Ores from Taurus Mountains and Lavrio, Bronze Age metals from sites in Turkey and copper-based artefacts Pella.

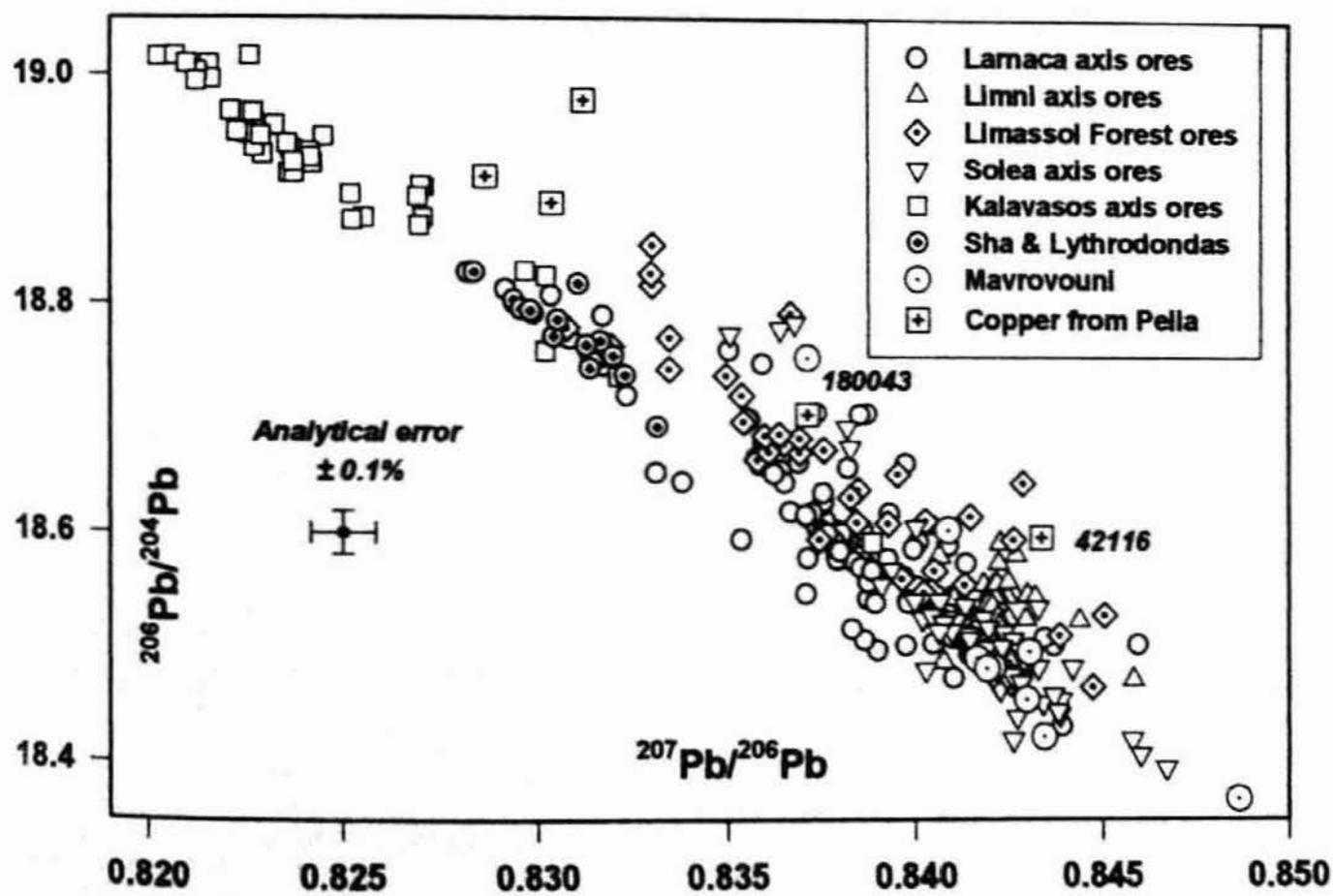
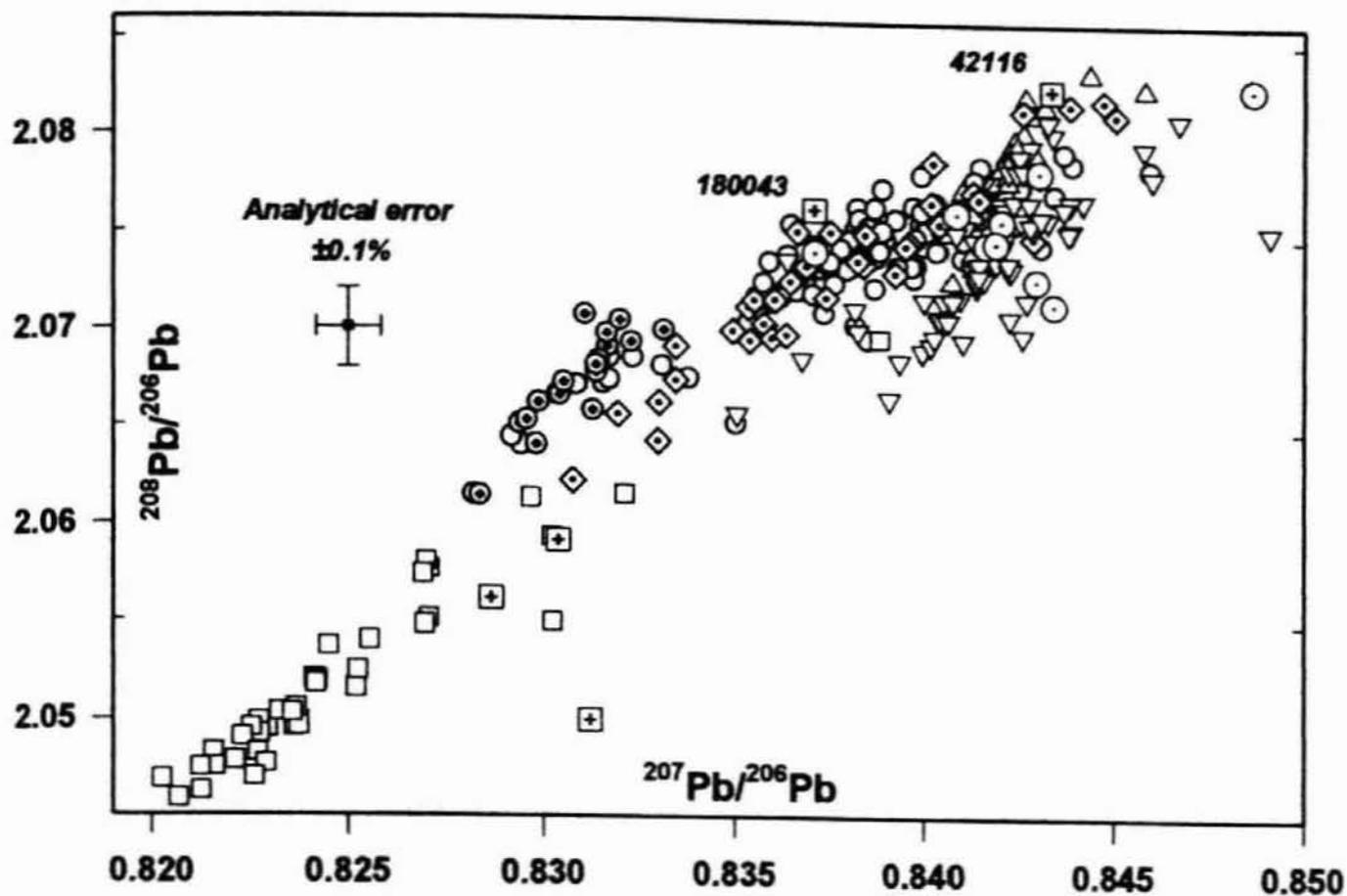


Figure 16. *Copper-based artefacts from Pella and lead isotope data for ores from Cyprus.*

Acknowledgements

The authors wish to express their gratitude to the British Academy Fund for Applied Science in Archaeology, which supported the work through Grant No. APN 7513. The material was made available for analysis by courtesy of Drs Ghazi Bisheh and Fawaz al-Khraishah, Directors General of the Department of Antiquities of Jordan and of Dr Stephen Bourke, director of the current excavations at Pella, to whom thanks are due. Drs Stephen Bourke and Peter Northover, and Prof. Thilo Rehren, kindly read and commented upon an earlier version of the text, but are in no way responsible for the final product.

Bibliography

- Adams R. (2000) The Early Bronze Age III-IV transition in southern Jordan: evidence from *Khirbet Hamra Ifdan*. Pp. 379-402 in G. Philip and D. Baird (eds) *Ceramics and Change in the Early Bronze Age of the Southern Levant*. Sheffield Academic Press: Sheffield.
- Allen, I.M., Britton, D. and Coghlan, H.H. (1970) Metallurgical Reports on British and Irish Bronze Age Implements and Weapons in the Pitt Rivers Museum. Pitt Rivers Museum: Oxford.
- Archi, A. (1985) Circulation d'objets en métal précieux de poids standardisé à Ebla. Pp. 25-34 in J.-M. Durand and J.R. Kupper (eds.) *Miscellanea Babylonica. Melanges offerts à Maurice Birot*. Editions Recherche sur les civilisations: Paris.
- Bourke, S.J. (1997) Pre-Classical Pella in Jordan: a conspectus of ten year's work (1985-1995). *PEQ* 129, 94-115.
- Bourke, S., Sparks, R.T and Mairs, L.D. (1999) Exploring prehistoric Pella in Jordan: the 1994-97 field seasons. *MA* 12, 147-156.
- Bourke, S., Sparks, R., McLaren, B., Mairs, L., Meadows, J., Hikade, T., Reade, W. (in press). Preliminary report on the University of Sydney's eighteenth and nineteenth seasons of excavation at Pella (Tabaqat Fahl) in 1996/97. *ADAJ* 46 (2002).
- Braidwood, R.J. and Braidwood, L.S. (1960) *Excavations in the Plain of Antioch. I. The Earlier Assemblages, Phases A-J*. OIP No. 61. Chicago University Press: Chicago.
- Branigan, K., McKerrell, H. and Tylecote, F.R. (1976) An examination of some Palestinian bronzes. *Journal of the Historical Metallurgy Society* 10.1, 15-23.
- Budd, P. and Ottaway, B.S. (1990) The properties of arsenical-copper alloys: implications for the development of Eneolithic metallurgy. Pp. 132-142 in P. Budd, B. Chapman, C. Jackson, R. Janaway and B.S. Ottaway (eds) *Archaeological Sciences 1989*. Oxbow Monographs 9. Oxford.
- Budd, P., Pollard, A.M., Scaife, B. and Thomas, R.G. (1995) Oxhide ingots, recycling and the Mediterranean metals trade. *JMA* 15, 1-32.
- Budd, P. and Taylor T., (1995) The faerie smith meets the bronze industry: magic versus science in the interpretation of pre-historic metal-making. *World Archaeology* 27, 133-143.
- Conn, G.K.T. and Bradshaw, F.J. (1952) *Polarized Light in Metallography*. Butterworths: London.
- Contenson, H. de. (1972) Rapport préliminaire sur XXXIIe campagne de fouilles à Ras Shamra. *AAAS* 2 25-43.
- Craddock, P. and Giunilia-Mair, A.R. (1988) Problems and possibilities for provenancing bronzes by chemical composition, with special reference to western Asia and the Mediterranean in the Early Iron Age. Pp. 317-331 in J.E. Curtis ed. (1988).
- Curtis, J.E. (ed.) (1988) *Bronze-working Centres of Western Asia c. 1000-539 BC*. Kegan Paul International: London.
- Davies, W.V. (1987) *Catalogue of Egyptian Antiquities, the British Museum. VII. Tools and Weapons. I. Axes*. British Museum Publications: London.
- Dobres, M.-A. and Hoffmann, C.R. (1994) Social agency and the dynamics of prehistoric technology. *Journal of Archaeological Method and Theory* 1, 211-258.
- Dugay, L. (1996) Specialized pottery production in the Bronze Age Cyprus and pottery use-wear analysis. *JMA* 9, 167-192.
- Dungworth, D., Catapotis, M., Darlinghaus, M., Howland, L. and Pederson, E. (forthcoming) An experimental study of remnant coring and recrystallization. In J.P. Northover and C. Salter (eds) *Foundations of Smiths and Platers*.
- Gale, N.H., Bachmann, H.G., Rothenberg, B. and Stos-Gale, Z.A. (1990) The adventitious production of iron in the smelting of copper in Timna, W. Araba. Pp. 182-191 in B. Rothenberg (ed.) *The Ancient Metallurgy of Copper*. Institute of Archaeometallurgical Studies, University College, London: London.
- Gale, N.H., Stos-Gale, Z.A., Maliotis G. and N. Anneti (1997) Lead isotope data from the Isotrache Laboratory Oxford: Archaeometry data base 4, ores from Cyprus. *Archaeometry* 39(1), 237-246.
- Halotte, R. (1995) Mortuary archaeology and the Middle Bronze Age southern Levant. *JMA* 8, 93-122.
- Hauptmann, A. (2000) *Zur frühen Metallurgie des Kupfers in Feinan/Jordanien*. Der Anschnitt Beiheft 1. Deutsches Bergbau Museum: Bochum.
- Hauptmann, A., Begemann, F., Heitkemper, R., Pernicka, E. and Schmitt-Strecker, S. (1992) Early copper produced in Feinan, Wadi Araba, Jordan: the composition of ores and copper. *Archeomaterials* 6(1), 1-33.
- Hauptmann, A., Begemann, F. and Schmidt-Strecker, S. (1999) Copper objects from Arad - their composition and provenance. *BASOR* 314: 1-17.
- Higgins, R.A. (1974) *Engineering Metallurgy. Metallurgical Process Technology*. 2nd edition. Edward Arnold: London.
- (1993) *Engineering Metallurgy. 1. Applied physical metallurgy*. 6th edition. London: Edward Arnold.
- Hughes, M.J., Lang, J.R.S., Leese, M.N. and J.E. Curtis (1988) The evidence of scientific analysis: a case study

- of the Nimrud bowls. Pp. 311–316 in J.E. Curtis (ed.) 1988.
- Benyon, K. M. (1960) *Excavations at Jericho*. I. British School of Archaeology in Jerusalem: London.
- (1965) *Excavations at Jericho*. II. British School of Archaeology in Jerusalem: London.
- Bein, S. and Hauptmann, A. (1999) Iron Age leaded tin bronzes from Khirbet Edh-Dharih. *Journal of Archaeological Science* 26, 1075–1082.
- Binapp, A.B. (2000) Archaeology, science-based archaeology and the Mediterranean Bronze Age metals trade. *European Journal of Archaeology* 3: 31–56.
- Binapp, A.B. and Cherry, J.F. (1994) *Provenience Studies and Bronze Age Cyprus*. Monographs in World Archaeology 21. Prehistory Press: Madison.
- Bühner-Eggebrecht, E. (1969) *Die Axt als Waffe und Werkzeug im alten Ägypten*. München Ägyptische Studien 15. Hessling: Berlin.
- Evans, T.E., Adams, R.S., Hauptmann, A., Prange, M., Schmitt-Strecker, S. and Najjar, M. (2002) Early Bronze Age metallurgy: a newly discovered copper manufactory in southern Jordan. *Antiquity* 76, 425–437.
- Feeks, N. (1993) Surface characterization of tinned bronze, high-tin bronze, tinned iron and arsenical bronze. Pp. 247–275 in S. la Niece and P. Craddock (eds) *Metal Plating and Patination. Cultural, Technical and Historical Developments*. Butterworth Heinemann: London.
- Feron, E. (1992) *Axes and Adzes from Canaan*. Prähistorische Bronzefunde Abt. IX Bd. 19. Steiner: Stuttgart.
- Goorey, P.R.S. (1994) *Ancient Mesopotamian Materials and Industries*. Clarendon: Oxford.
- Gorun, W. (1992) *The Amarna Letters*. Johns Hopkins University Press: Baltimore.
- Huhly, J.D. and Muhly, P. (1989) Metal artefacts. Pp. 267–295 in Z. Herzog, G. Rapp Jnr and O. Negbi (eds) *Excavations at Tel Michal, Israel*. Sonia and Marco Nadler Institute of Archaeology: Tel Aviv.
- Northover, P. (1989) Properties and use of arsenic-copper alloys. Pp. 111–118 in A. Hauptmann, E. Pernicka and G.A. Wagener (eds) *Archäometallurgie der alten Welt*. Der Anschnitt Beiheft 7. Deutschen Bergbau-Museum. Bochum.
- (2000) Analysis of Bronze Age metalwork from Tell Beydar. Pp. 117–124 in K. Van Lerberghe and G. Voet (eds) *Tell Beydar Environmental and Technical Studies*. Subartu VI. Brepolis: Turnhout.
- Palumbo, G. (2001) The Early Bronze Age IV. Pp. 233–269 in B. MacDonald, R. Adams, and P. Bienkowski (eds) *The Archaeology of Jordan*. Levantine Archaeology 1. Sheffield Academic Press: Sheffield.
- Pernicka, E., Begemann, F., Schmitt-Strecker, S. and Wagner, G.A. (1993) Eneolithic and Early Bronze Age copper artefacts from the Balkans and their relation to Serbian copper ores. *Prähistorisches Zeitschrift* 68(1), 1–54.
- Pernicka, E., Seeliger, T.C., Wagner, G.A., Begemann, F., Schmitt-Strecker, S., Eibner, C., Oztunali, O. and Baranyi, I. (1984). *Archäometallurgische Untersuchungen in Nordwestanatolien*. *Jahrbuch des Römisch-Germanischen Zentralmuseums* 31, 533–599.
- Philip, G. (1988). Hoards of the Early and Middle Bronze Ages in the Levant. *World Archaeology* 20, 190–208.
- (1991) Tin, Arsenic, Lead: Alloying Practices in Syria-Palestine around 2000 BC. *Levant* 23, 93–104.
- (1995a) Tell el-Dab'a metalwork: patterns and purpose. Pp. 66–83 in W.V. Davies and L. Schofield (eds) *Egypt, the Aegean and the Levant. Interconnections in the Second Millennium BC*. British Museum Publications: London.
- (1995b) The same but different: a comparison of Middle Bronze Age metalwork from Jericho and Tell el-Dab'a. *SHAJ* 5, 523–530. Department of Antiquities of Jordan: Amman.
- (1999) Complexity and diversity in the southern Levant during the third millennium BC: the evidence of Khirbet Kerak Ware. *JMA* 12, 26–57.
- (2001) The Early Bronze I–III Age. Pp. 163–232 in B. MacDonald, R. Adams, and P. Bienkowski (eds) *The Archaeology of Jordan*. Levantine Archaeology 1. Sheffield Academic Press: Sheffield.
- (2002) Contacts between the 'Uruk' world and the Levant during the fourth millennium BC: evidence and interpretation. Pp. 207–235 in J.N. Postgate (ed.) *Artefacts of Complexity. Tracking the Uruk in the Near East*. British School of Archaeology in Iraq, Archaeological Reports Volume 5. Aris and Phillips: Warminster.
- Piggott, V.C. (1996). Near Eastern archaeometallurgy: modern research and future directions. Pp. 139–176 in J.S. Cooper and G.M. Schwartz (eds) *The Study of the Ancient Near East in the Twenty-First Century*. Eisenbrauns: Winona Lake.
- Potts, T.F., Bourke, S.J.B., Edwards, P.C., Richards, F. and Wightman, G.J. (1988) Preliminary report on the eight and ninth seasons of excavation by the University of Sydney at Pella (Tabaqat Fahl) 1986 and 1987. *ADAJ* 32, 115–149.
- Pritchard, J.B. (1955) *Ancient Near Eastern Texts relating to the Old Testament*. 2nd ed. Princeton.
- Rehren, T., Hess, K. and Philip, G. (1997) Fourth millennium BC copper metallurgy in northern Jordan: the evidence from Tell esh-Shuna. Pp. 625–640 in H.-G. Gebel, Z. Kafafi, and G.O. Rollefson (eds) *The Prehistory of Jordan*. II. *Perspectives from 1997*. Studies in Early Near Eastern Production, Subsistence, and Environment 4. Ex oriente: Berlin.
- Rollason, E.C. (1949) *Metallurgy for Engineers*. Edward Arnold: London.
- Rosenfeld, A. Ilani, S and Dvoracheck, M. (1997) Bronze alloys from Canaan during the Middle Bronze Age. *Journal of Archaeological Science* 24, 857–864.
- Schaeffer, C.F.A. (1962) Contribution à l'étude des niveaux II et III de Ras Shamra. *Ugaritica* IV, 329–414. Geuthner: Paris.
- Schaub, R.T. and Rast, W.E. (1989) *Bab edh-Dhra: Excavations in the Cemetery Directed by Paul W. Lapp (1965–67)*. Reports of the Expedition to the Dead Sea Plain, Jordan. Vol. I. Eisenbrauns: Winona Lake.

- Scott, D.A. (1991) *Metallography and Microstructure of Ancient and Historic Metals*. Getty Conservation Trust: Los Angeles.
- Seeden, H. (1980) *The Standing Armed Figurines in the Levant*. Prähistorisches Bronzefunde, Abt.1 Bd. 1.C.H. Beck'sche Verlagsbuchhandlung: Munich.
- Shalev, S. (1988) Redating the Philistine Sword at the British Museum: a case study in typology and technology. *Oxford Journal of Archaeology* 7, 303–311.
- (1994) The change in metal production from the Chalcolithic period to the Early Bronze Age in Israel and Jordan. *Antiquity* 68, 630–637.
- (2000) Metallurgical analysis. Pp. 278–287 in M. Kochavi (ed.) *Aphek-Antipatris. I. Excavation of Areas A and B, the 1972–1976 seasons*. Emery and Claire Yass Publications in Archaeology. Tel Aviv University Institute of Archaeology: Tel Aviv.
- Sherratt, A. and Sherratt, E. (1991) From luxuries to commodities: the nature of Mediterranean Bronze Age trading systems. Pp. 351–386 in N.H. Gale (ed.) *Bronze Age Trade in the Mediterranean*. Studies in Mediterranean Archaeology 90. Paul Aström's Förlag: Jönsered.
- Smith, R.H. and Potts, T.F. (1992) The Middle and Late Bronze Ages. In A.W. McNicoll, P.C. Edwards, J. Hanbury-Tenison, J.B. Hennessy, T.F. Potts, R.H. Smith, A. Walmsley and P. Watson, *Pella in Jordan*. 2. Mediterranean Archaeology Supplement 2. Meditarch: Sydney.
- Sparks, C.J. (1976) Quantitative X-ray fluorescence analysis using Fundamental Parameters. *Advances in X-ray Analysis* 19, 19.
- Stos-Gale, Z.A. and Gale, N.H. (1994) The origin of metals excavated on Cyprus. Pp. 92–122 and 210–216 in A.B. Knapp and J. Cherry (eds.) *Provenience Studies and Bronze Age Cyprus*. Monographs in World Archaeology 21. Prehistory Press: Madison.
- Stos-Gale, Z.A., Gale, N.H. and Annetts, N. (1996). Lead isotope analyses of ores from the Aegean. *Archaeometry* 38(2), 381–390.
- Stos-Gale, Z.A., Gale, N.H., Annetts, N., Todorov, T., Lilov, P., Raduncheva, A. and Panayotov, I. (1998) Lead isotope data from the Isotrache Laboratory, Oxford: Archaeometry Database 5, Ores from Bulgaria. *Archaeometry* 40(1), 217–226.
- Stos-Gale, Z.A., Gale, N.H. and Houghton, J. (1997) The origin of copper metal excavated in El Amarna. Pp. 127–135 in W.V. Davies and L. Schofield (eds) *Egypt, the Aegean and the Levant: Interconnections in the 2nd millennium BC*. British Museum Press: London.
- Stos-Gale, Z.A., Gale, N.H., Houghton, J. and Speakman, R. (1995a) Lead isotope analyses of copper from the Western Mediterranean. *Archaeometry* 37(4), 407–415.
- Stos-Gale, S., Maliotis, G. and Gale, N.H. (1998) A preliminary survey of the Cypriot slag heaps and its contribution to the reconstruction of copper production on Cyprus. Pp. 235–262 in T. Rehren, A. Hauptmann and J. Muhly (eds.) *Metallurgica Antiqua, in Honour of Hans-Gert Bachmann and Robert Maddin*. Der Anschnitt 10. Deutsches Bergbau Museum: Bochum.
- Stos-Gale, Z.A., Maliotis, G., Gale, N.H. and Annetts, N. (1997) Lead isotope characteristics of the Cyprus copper ore deposits applied to provenance studies of copper oxide ingots. *Archaeometry* 39(1), 83–124.
- Tadmor, M., Kedem, D., Begemann, F., Hauptmann, P., Pernicka, E. and Schmitt-Strecker, S. (1995) The Neolithic Mishmar hoard from the Judean Desert: technology, composition, and provenance. *Atiqot* 27, 95–148.
- Waetzoldt, H. (1990) Zur Bewaffnung des heeres in Ebla. *Oriens Antiquus* 29, 1–38.
- Wagner, G.A., Begemann, F., Eibner, C., Lutz, O., Oztunali, O., Pernicka, E. and Schmitt-Strecker, S. (1989) Archäometallurgische Untersuchungen Rohstoffquellen des frühen Kupfers Ostanatolien. *Jahrbuch des Römisch-Germanischen Zentralmuseums Mainz* 36, 637–686.
- Yener, K.A., Özbal, H., Minzoni-Deroche A. and Alpar, B. (1989) Bolkardag: archaeometallurgy surveys in the Taurus Mountains, Turkey. *National Geographic Research* 5, 477–494.
- Yener, K.A., Sayre, E.V., Özbal, H., Joel, E.C., Barak, I.L. and Brill R.H. (1991) Stable lead isotope studies of Central Taurus ore sources and related artefacts in the Eastern Mediterranean Chalcolithic and Bronze Age sites. *Journal of Archaeological Science* 18, 541–572.

ID	Object	Date	Method	Condi- tion	Cu	Zn	Sb	As	Pb	Co	Ni	Au	Hg	Ag	Sn	Fe	TOTAL
80258	Dagger		Surface	Corr	88.0	det.	0.80	6.52	0.50	det.	0.22	n.d.	n.d.	0.15	det.	3.69	99.9
90208	Tool	LB II	Surface	PC	87.1	det.	det.	0.22	det.	det.	n.d.	n.d.	n.d.	det.	11.94	0.47	99.7
90208	Tool		Metalog	PC	81.3	det.	13.30	det.	94.4								
90208	Tool		Metalog	PC	81.6	n.d.	det.	det.	det.	n.d.	det.	det.	n.d.	det.	13.80	det.	95.46
100142	Bracelet	Iron Age	Surface		81.1	det.	det.	det.	0.87	det.	det.	det.	n.d.	det.	16.72	0.53	99.3
100201	Bracelet	Iron Age	Metalog		86.4	det.	det.	det.	1.25	det.	det.	det.	n.d.	det.	11.79	0.20	99.6
100214	Bracelet	Iron Age	Surface	PC	93.6	det.	det.	det.	0.39	det.	n.d.	n.d.	n.d.	det.	5.51	0.21	99.7
100214	Bracelet		Metalog	PC	89.9	det.	det.	det.	det.	n.d.	det.	det.	det.	det.	5.74	det.	95.6
100222	Bracelet	Iron Age	Surface		83.6	det.	det.	n.d.	1.11	n.d.	det.	n.d.	n.d.	n.d.	14.89	0.22	99.8
100252	Ring	Iron Age	Surface		90.0	det.	det.	n.d.	0.69	n.d.	det.	n.d.	det.	det.	9.76	0.41	99.8
100255	Bracelet	Iron Age	Surface	PC	87.1	det.	det.	n.d.	1.06	n.d.	n.d.	det.	n.d.	det.	10.82	0.55	99.5
100255	Bracelet		Metalog	PC	87.8	det.	det.	det.	0.97	det.	det.	det.	det.	det.	9.07	0.77	98.6
110290	Edged tool	LB IIB	Metalog	Corr	83.1	n.d.	n.d.	det.	det.	det.	det.	n.d.	n.d.	n.d.	14.70	det.	97.8
110290	Edged tool		Metalog	Corr	81.7	det.	det.	det.	det.	n.d.	n.d.	n.d.	n.d.	n.d.	16.64	det.	98.3
110290	Edged tool		Metalog	Corr	82.4	n.d.	det.	det.	0.62	n.d.	det.	n.d.	n.d.	det.	15.33	0.73	99.0
110603	Pin	LB I	Surface		96.4	det.	det.	det.	det.	n.d.	det.	det.	n.d.	det.	2.86	det.	99.3
110606	Pin	LB I	Surface		95.3	n.d.	n.d.	det.	det.	n.d.	n.d.	n.d.	n.d.	n.d.	4.28	0.16	99.7
170058	Axe	LB I	Surface		89.8	det.	det.	n.d.	det.	det.	n.d.	n.d.	det.	n.d.	9.34	0.48	99.6591
170065	Ring	MB IIB	Surface		98.3	det.	det.	det.	det.	det.	n.d.	det.	det.	n.d.	0.79	0.18	99.2
170065	Ring		Metalog	PC	83.9	det.	12.00	det.	95.9								
170065	Ring		Metalog	PC	87.3	det.	det.	det.	det.	n.d.	det.	n.d.	det.	det.	10.19	0.80	98.3
170066	Pin	LB II	Surface	Corr	77.6	n.d.	det.	0.39	det.	n.d.	n.d.	det.	det.	20.56	0.38	0.72	99.6
170066	Pin		Metalog	Corr	61.3	det.	det.	det.	0.49	n.d.	det.	det.	det.	34.41	det.	1.07	97.3
170066	Pin		Metalog	Corr	61.2	n.d.	det.	det.	det.	det.	n.d.	det.	n.d.	36.25		det.	97.4
170084	Projectile	LB II	Surface		98.3	det.	0.08	0.62	0.16	n.d.	det.	det.	n.d.	det.	det.	0.63	99.8
170085	Projectile	LB II	Surface		99.3	det.	det.	0.16	det.	n.d.	det.	det.	n.d.	det.	det.	0.15	99.6
170086	Projectile	LB II	Surface		99.0	det.	0.08	0.21	0.15	det.	n.d.	n.d.	det.	det.	det.	0.43	99.9
170087	Projectile	LB II	Surface		99.3	n.d.	det.	det.	det.	det.	n.d.	det.	n.d.	det.	0.22	0.19	99.7
170088	Projectile	LB II	Surface		99.0	det.	0.08	0.22	n.d.	det.	n.d.	det.	det.	det.	det.	0.29	99.6
170089	Projectile	LB II	Surface		98.8	det.	0.09	0.38	0.18	n.d.	det.	det.	det.	det.	det.	0.30	99.8
170090	Projectile	LB II	Surface		98.8	n.d.	det.	0.20	det.	det.	det.	det.	n.d.	det.	det.	0.60	99.6
170091	Projectile	LB II	Surface		99.1	n.d.	0.07	0.34	0.14	n.d.	n.d.	n.d.	n.d.	det.	det.	0.27	99.9
170092	Projectile	LB II	Surface		95.0	det.	0.09	2.23	0.21	det.	det.	det.	n.d.	det.	0.34	1.92	99.8

I/D	Object	Date	Method	Condi- tion	Cu	Zn	Sb	As	Pb	Co	Ni	Au	Hg	Ag	Sn	Fe	TOTAL
180049	Pin		Metalog	Corr	95.9	det.	n.d.	n.d.	det.	n.d.	det.	n.d.	det.	det.	det.	det.	95.9
180052	Tool?	EB IB or MBA	Surface	Corr	95.2	n.d.	det.	0.29	det.	det.	n.d.	det.	det.	det.	3.36	0.50	99.3
180052	Tool?		Metalog	Corr	91.8	det.	det.	det.	det.	det.	n.d.	n.d.	det.	det.	det.	det.	97.3
190073	Fitting	LB IIB/	Surface		95.8	det.	det.	0.34	det.	n.d.	n.d.	n.d.	n.d.	det.	3.24	0.30	99.6
190074	Harpoon	LB IIB	Surface		97.6	det.	det.	det.	0.29	det.	det.	det.	n.d.	0.30	1.25	0.22	99.6
190074	Harpoon		Surface		97.2	det.	det.	0.34	0.34	n.d.	det.	det.	n.d.	0.21	0.70	0.89	99.7
200013	Balance Pan	LB IIB	Surface		98.7	n.d.	det.	det.	det.	det.	det.	n.d.	det.	det.	0.88	0.17	99.8
200014	Balance Pan	LB IIB	Surface		97.7	det.	det.	0.11	det.	det.	n.d.	n.d.	n.d.	det.	1.84	0.17	99.8
200014	Balance Pan		Surface		98.0	det.	det.	0.16	det.	0.04	n.d.	det.	det.	det.	1.36	0.25	99.8
200015	Cymbal	LB IIB	Surface		97.9	0.24	det.	det.	0.38	det.	det.	n.d.	n.d.	0.08	1.04	0.20	99.9
200015	Cymbal		Surface		98.1	0.23	det.	det.	0.62	det.	det.	n.d.	det.	0.03	0.61	0.24	99.9
200016	Cymbal	LB IIB	Surface		95.7	0.32	det.	0.09	0.10	det.	0.12	det.	n.d.	n.d.	3.38	0.20	99.9
200016	Cymbal		Surface		96.7	0.17	det.	0.05	0.08	det.	n.d.	n.d.	n.d.	det.	2.65	0.26	99.9
200016	Cymbal		Surface		96.0	0.33	det.	0.09	0.08	det.	det.	det.	det.	det.	3.146	0.20	99.9
200016	Cymbal		Surface		94.6	0.31	det.	4.42	0.27	99.6							
920630	Pin	MB II	Surface	Corr	97.6	det.	det.	det.	det.	det.	n.d.	det.	n.d.	det.	1.14	0.31	99.1
920630	Pin		Metalog	Corr	65.8	n.d.	n.d.	det.	det.	det.	det.	n.d.	n.d.	det.	28.98	2.52	97.0
920630	Pin		Metalog	Corr	62.6	n.d.	det.	det.	2.03	n.d.	n.d.	det.	n.d.	det.	33.77	det.	98.4
920630	Pin		Metalog	Corr	68.6	det.	det.	0.55	0.77	n.d.	det.	n.d.	n.d.	det.	28.55	det.	98.4
920630	Pin		Metalog	Corr	69.3	n.d.	det.	0.48	0.96	det.	n.d.	det.	n.d.	det.	27.07	1.17	98.9
920630	Pin		Metalog	Corr	90.1	det.	det.	det.	0.66	det.	det.	det.	n.d.	det.	7.25	det.	98.8
920631	Pin	MB II	Surface	Corr	96.0	n.d.	det.	det.	det.	det.	n.d.	det.	n.d.	det.	3.33	0.20	99.6
920631	Pin		Metalog	Corr	75.0	n.d.	det.	det.	det.	n.d.	det.	n.d.	n.d.	det.	21.8	det.	97.5
950477	Pin	EB II late	Surface		98.7	det.	det.	det.	0.58	det.	det.	n.d.	det.	det.	det.	0.24	99.5

Table 2. Results of EDXRF analysis of prepared artefact surface, analysis of metallographic samples. These are indicated as (Surface) and (Metalog) respectively in column 'Method'. In column 'Condition' (UC) = core preserved, (PC) = core partially corroded, (Corr) = little uncorroded material remaining. When multiple analyses were carried out, either on different areas of the surface of an artefact or at different points along a metallographic sample, these are listed separately. det = detected, n.d. = not detected, n.a. = not available; the limits of detection under the analytical conditions were: Cu 0.1%; Zn 0.1%; Sb 500ppm; As 500ppm; Pb 500ppm; Co 0.10%; Ni 0.1%; Au 0.1%; Hg 0.1%; Ag 500ppm; Sn 500ppm; Fe 0.10%.

used as part of diplomatic gifts. Examples include bronze armour, vessels and tools taken by Tutmosis III following the battle of Megiddo (Pritchard 1958, 181) or quantities of bronze torques, armour, shields and various weapons being sent as royal gifts e.g. Amarna letter No. 22 (Moran 1992, 55–56).

A group of artefacts dating to the thirteenth century BC was excavated in Area XXXIII 15.4, interpreted as part of a cult structure. The group, which included two cymbals and two balance pans, is interpreted by the excavator as representing cultic paraphernalia, and came from the floor of a destruction that produced Mycenaean IIIB pottery (Bourke 1999, 152–155; Bourke *et al.* in press). Analysis, multiple analyses in some cases, revealed some variability between the various artefacts, but all were made from tin bronze. Although well preserved, the majority of these artefacts were made from relatively thin metal, and caution is required in the interpretation of the tin concentrations obtained from surface analysis. Context XXXIIG 107 produced additional material also dating to the Late Bronze Age / Iron Age transition. This included what appears to have been the rectangular tang of a dagger with a rivet still in place, and a harpoon with a distinctive barbed head. These were more robust and were likely to have a well preserved metal core, and appear to have been made from copper containing a low level of tin. The low tin contents may suggest some of these artefacts were produced from metal containing an admixture of bronze scrap.

4.4. The Iron Age

With a few exceptions (Curtis [ed.] 1988; Muhly and Muhly 1989; Moorey 1994), there has been relatively little interest in the technology of copper-alloy artefacts from Iron Age contexts in the Levant. This, in part, appears to reflect the gradual replacement of bronze by iron for the production of tools and weapons and its relegation to a more specialist repertory of artefacts, mainly those produced by casting or the working of metal sheet (Moorey 1994, 264). The latter includes an impressive range of frequently high-status objects such as bowls, cauldrons and drinking sets, to which the colour and lustre characteristic of bronze objects would have been significant. Of course, fine objects of this kind are not always readily accessible for laboratory analysis. However, when chemical data are available it appears to indicate, as might be expected in the manufacture of high-quality products, quite careful selection of alloys according to function, the technical requirements of sheet-metal working and what might be termed 'workshop

tradition' (Hughes *et al.* 1988). Thus groups of analyses drawn from such specialist artefacts may not be representative of Iron Age bronze-working practices generally, which as Craddock and Giunlia-Mair (1988, 321) have observed, appear to have involved considerable variability in alloy selection.

The Pella sample consists of six rings and bracelets, made by similar techniques and originating from a single tomb and so should offer an opportunity to assess the potential variability within a reasonably coherent group of artefacts. The tomb is dated to the later eleventh or tenth century BC, i.e. transitional Iron I–II (see Potts *et al.* 1988, 148; Bourke 1997, 113). Two large, heavy rings (Nos 100222, 100201), perhaps to be identified as anklets, are very similar in form, and composition. Both are composed of an alloy containing more than 10% tin, and a little over 1% lead. Neither of the pieces examined by metallography showed evidence of strain lines, although both indicated annealing. Presumably in the absence of a cutting-edge, there was no need for final cold-working. With the exception of one artefact (No. 100214) from which the results of surface analysis and that of a metallographic section were in good agreement at a little over 5%, the Iron Age bracelets revealed tin contents of around or above 10%, and concentrations of lead around 1%, suggesting a fairly standard set of alloying practices.

The main technical advantages in using bronze as opposed to copper for the production of artefacts which required neither great hardness nor complicated castings, lay in the lower melting point and longer casting interval which bronze offered. Thus the clear preference for using tin-bronze for personal items requires some consideration. Analysis of a large group of Persian period metalwork from Tell Michal in Palestine (Mulhy and Muhly 1989, 269) revealed that bronze was preferred for the production of rings, leaded-bronze for bracelets, while unalloyed and arsenical copper remained in use for certain other categories of artefact. This suggests that rather than bronze having become the 'industry standard' for smiths, the choice of tin-copper alloys for rings, pins and so on reflected the desire to obtain artefacts with the distinctive yellowish colour characteristic of such alloys (Moorey 1994, 253), perhaps in emulation of gold: that is aesthetic, rather than technological considerations.

Two of the Iron Age artefacts gave lead isotope signatures consistent with copper from the Faynan DLS deposits (Hauptmann 2000, 137–8, fig. 115). This is consistent with the archaeological evidence that indicates the renewed exploitation of these ores

Dugay 1996, 181; Halotte 1995, 111–114; Philip 2001, 199–200). It is also worth underlining that neither the EBA nor the LBA hoard was composed of artefacts which could be shown to come from a single ore source. Rather each hoard appeared to be composed of artefacts produced for other purposes, and which were subsequently interred as a group.

In comparison to that of the Early Bronze Age, the lead isotope data for the Late Bronze Age appears to indicate a shift away from the use of copper from local sources, and towards more extensive participation in wider Mediterranean trade networks. This is in agreement both with the situation in Faynan which shows little evidence for exploitation at this point (Hauptmann 2000), and the expectations of current models for the development of commercial networks in the east Mediterranean at this time (Sherratt and Sherratt 1991; Knapp and Cherry 1994).

The deliberate addition of lead to tin-copper alloys is documented in the southern Levant from the early second millennium BC (Philip 1991; 1995b; Rosenfeld *et al.* 1997, 859, tables 1, 2). Accordingly, one aim of the present research project was to assess the impact of the deliberate addition of metallic lead to copper alloys on the lead isotope ratios of the resulting artefacts by a comparison with those of contemporary objects from the same site containing the lower levels of lead consistent with an origin in the copper ores. In this way it was hoped to examine the potential impact of the recycling of tin-lead bronze artefacts upon lead isotope ratios in Bronze Age copper artefacts from the Levant. Unfortunately, it proved impossible to pursue this line of enquiry because the Pella assemblage produced relatively few objects containing significant concentrations of lead, and those that did were unable to provide the samples required for lead isotope analysis. However, the presence of tin-lead bronzes at Pella during the second millennium BC highlights the need for researchers to investigate this matter in more detail.

We believe that the present study has demonstrated quite clearly the value of an investigative programme that can deploy multiple analytical techniques in pursuit of a clear research design. While such approaches have been widely recommended, a variety of logistical and financial problems have militated against their achievement in practice. What is required now is not just more analyses, regardless of provenance or context, but carefully focused programmes, which will provide data complementary to the current project, and will address issues which we have raised, but have been unable to explore fully, because of the limitations of our material. Only in this way will researchers be able to develop an

understanding of patterns, and perhaps more revealing, discontinuities, in Levantine metallurgy, at general, site and context-specific levels, and thus begin to comprehend ancient metallurgy in terms of its social and economic setting.

Appendix: report on lead isotope analyses of 11 samples of copper-based artefacts from Pella and Tell al-Shuna.

Sophie Stos

Methodology

The eleven samples of copper-based artefacts submitted for lead isotope analyses by Dr Philip were analysed in the Isotrace Laboratory using Thermal Ionisation Mass Spectrometry. The analytical methodology of lead isotope measurements is described in detail in the first lead isotope database from the Isotrace Laboratory published in *Archaeometry* (Stos-Gale *et al.* 1995). The identification of the origin of copper ore used for making the artefacts is based on one-to-one comparisons of lead isotope characteristics of samples of ores from known locations and artefacts. The database of lead isotope characteristics of ore deposits used for comparisons included the following number of ores and copper slags from various deposits: 290 from Turkey, 650 from Cyprus, 260 from the Near East (Jordan, Egypt, Israel, Saudi Arabia, Oman) and 1500 from the Aegean. Most of the data is published in papers listed in the bibliography, although some comparisons are with unpublished data from the Isotrace Laboratory.

Discussion of the results

Dr Philip supplied, together with the samples of artefacts submitted for lead isotope analysis, the information about the elemental composition of these metals. However, all samples are routinely analysed before lead extraction using the semi-quantitative EDXRF method. The results of these analyses are summarised in Table 5. Only two samples submitted for lead isotope analysis contained lead in quantity of about 1% (538 and 100201). Lead in this amount is not unusual in copper metals smelted from poly-metallic ores. This point will be discussed further in