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Copper quality and provenance in Middle Bronze Age I Byblos and Tell Arqa (Lebanon)

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ABSTRACT

Forty-four Middle Bronze Age I weapons discovered at the sites of Byblos and Tell Arqa in Lebanon were investigated in order to study their copper quality and provenance. The evaluation of copper qualities is based on quantifying permanent inclusions such as copper sulfide and lead globules. The provenance of copper was studied using lead isotope analyses. For further discrimination between copper groups and sources elemental analyses by PIXE were performed on some of the weapons investigated. The results revealed two copper groups that could be qualified as "dirty" copper and "clean" copper. The former was used in most of the weapon types whereas the latter was reserved for items made of high-tin bronzes (>11 wt%) which underwent heavy hammering during the manufacturing process. Even though several potential copper sources were identified, the data point to Iran and Oman as the most probable areas of origin for the metal used in these weapons. These results contribute to the study of inter-regional exchange networks in the ancient Near East.

1. Introduction

Copper, tin and silver are one of the major trade and exchange goods documented in Near Eastern Middle Bronze Age (*c.* 2000–1600 BC) textual sources. Their importance probably stemmed from their scarcity and their properties. As a consequence, these metals were used in a wide range of objects such as tools, weaponry, ornaments and other symbols of wealth and prestige. They were also used as offerings to the deities and as "gifts" exchanged especially among members of the elite to forge or consolidate alliances and allegiances (Durand, 1997; 398–408; Ilan, 1992, 1995a; Philip, 1988, 1989; 160–1). Therefore, acquiring these metals and controlling their circulation and distribution was of major strategic, financial and political importance for the governing entities. As a consequence, identifying metal sources and trade routes have constituted an important research topic for historians, archaeologists and archaeometallurgists working on the Ancient Near East (Moorey, 1999; 245–9; Pollard and Heron, 2008; 302–3).

In contrast to Mesopotamia, few textual sources, belonging mainly to the Royal Archives of Mari (Arkhipov, 2012; Bonechi, 1992; Malamat, 1960, 1970), mention metal circulation in the Levant area. These 18th century BC texts report only the exchange of metal items or ingots between the city of Mari – situated on the present day Syrian/Iraqi borders – and some cities from western Syria and Canaan but almost never mention the source of the ore. In the southern part of the Levant, this lack of information has been compensated by several archaeometallurgical and metal provenance studies (for example Gale et al., 1990; Hauptmann, 2007; Hauptmann et al., 1999; Segal et al., 1996–7, 1999, 2000, 2004; Philip et al., 2003). In northern Levant however, apart from the Amuq valley and Cilicia (Gale et al., 1985a,b; Yener, 2007; Yener et al., 1991), few attempts have been made to determine ore sources exploited during the Middle Bronze Age: in the area of modern day Lebanon for example, only Sidon located to the South of Beirut has provided such data (Le Roux et al., 2004, 2009; Véron et al., 2012). As a consequence, it is quite difficult to propose an inter-regional model for metal exchange networks in this area. In this paper we hope to contribute to this field by presenting copper provenance data obtained by using elemental and lead isotope analyses on Middle Bronze Age I (MB I, *c.* 2000–1800 BC) copper

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alloy weapons discovered at the sites of Byblos and Tell Arqa in Lebanon.

In addition, the copper qualities of these weapons will be investigated. Even though they are mentioned in several Early Bronze Age IV (*c.* 2400–2000 BC) and Middle Bronze Age textual sources such as the Royal Archives of Mari (Arkhipov, 2012; Joannès, 1997; Limet, 1986) and Ebla (Waetzoldt and Bachmann, 1984; 5) in western Syria, the use of different copper qualities has never been confirmed by material evidences. Their definition varies according to the textual sources. These copper qualities could imply products from different stages of the smelting and refining processes, or copper with different levels of what Michel (2008) referred to as "impurities". Having only finished objects at our disposal, only the latter aspect could be investigated. The aim of this work is to find out whether or not the MB I copper alloy craftsmen of Byblos and Tell Arqa were able to distinguish between various copper qualities. In fact, we suspect that these qualities were related to the metal's mechanical properties. In that case the craftsmen would probably have been able to test copper properties such as hardness or malleability by hammering and annealing. Permanent inclusions, such as copper sulphide inclusions and lead globules, have been shown to have a harmful effect on the workability (i.e. the extent of the plastic domain) of a copper alloy during plastic deformation (Andrieu et al., 2000) and on the recrystallization rate during annealing (Pernot, 2000; Pernot and Montheillet, 1994). These inclusions are qualified as permanent as they are not eliminated by plastic deformation and annealing processes. In this paper the quantity of these inclusions is investigated as it is very likely to have an important impact on the quality of copper. This aspect will be referred to here as the "cleanness". Compared to a "dirty" copper, a "clean" copper contains fewer permanent inclusions and consequently it is less prone to cracking during cold working and easier to recrystallize during annealing.

The sites of Byblos and Tell Arqa are situated respectively 45 and 110 km north of Beirut. The weapons analysed in this work were found mainly in the so-called "champs des offrandes" deposits at Byblos (Dunand, 1954) and in burial 14.14 (Thalmann, 2006; 34) at Tell Arqa. Due to the excavation method used at Byblos (Dunand, 1939, 1954), contextual and chronological problems occurred preventing a thorough study of the site. The subsequent work of Saghieh (1983; 38, 132) and Lauffray (2008) concerning the stratigraphy of the main architectural complexes allowed the "champs des offrandes" deposits to be ascribed to the beginning of the MB I. This was also confirmed by the work of Philip (1989) and Gernez (2007) on the typology of the copper base alloy weapons. The stratigraphy of Tell Arqa, the ceramic vessels and copper alloy weapon types allow us to safely assume that burial 14.14 is contemporary with the deposits of Byblos (Gernez, 2010; Thalmann, 2006; 45).

2. Corpus presentation

For this work, 45 items (Tables 1 and 2) were selected from a corpus of 72 MB I weapons studied previously. This selection was made according to the morpho-typology of the weapons, the availability of samples, the technological features and the alloy recipes. The last two parameters were previously determined using metallographic observation and major and minor element analysis by energy dispersive spectrometry attached to a scanning electron microscope (SEM–EDS). These data are accessible in El Morr and Pernot (2010, 2011) as well as in the unpublished PhD thesis of El

Table 1
Site, context, Dimension, and typology of the socketed spearheads. The methods used for the study of each weapon is also mentioned.

Site	Weapons	Inv. number[a]	Pub. number[b]	Context	Current state[c]	Weight (g)	Preserved length (mm)	Max thickness (mm)	Type[d]	Method
Byblos	Large spearheads	9028	9669	Deposit λ	Incomplete[f]	187	246	15	2	Mt[i], EDS[h], Img[j]
		10268	8311	Deposit β	Fragment[g]	95	207	12	2	Mt, EDS, LIA
		10336	8441	Deposit γ	Fragment	85	199	11	2	Mt, EDS, Img
		11856	9523	Deposit ι	Incomplete	230	404	12	2	Mt, EDS, PIXE, LIA, Img
		11913	8352	Deposit β	Incomplete	399	453	18	2	Mt, EDS, LIA
		11914	8346	Deposit β	Incomplete	344	484	15	2	Mt, EDS, LIA
		11921	8348	Deposit β	Incomplete	465	555	21	2	Mt, EDS, LIA
		27855	9626	Deposit λ	Fragment	187	260	11	2	Mt, EDS, PIXE, LIA, Img
		27856	9667	Deposit λ	Fragment	137	296	9	2	Mt, EDS, PIXE, LIA, Img
		27884	9632	Deposit λ	Fragment	138	217	10	2	Mt, EDS, PIXE, LIA, Img
	Multi-piece spearheads	10977	10836	Deposit χ	Incomplete	65	188	10	8	Mt, EDS, PIXE, LIA, Img
		10978	10834	Deposit χ	Fragment	69	276	8	4	Mt, EDS, PIXE, LIA, Img
		10982	10846	Deposit χ	Incomplete	49	184	9	8	Mt, EDS, LIA
		10997	10845	Deposit χ	Incomplete	91	173	9	8	Mt, EDS, PIXE, LIA, Img
	Small spearheads	10274	8288	Deposit β	Complete[e]	37	114	6	10	Mt, EDS, PIXE, LIA
		10277	8298	Deposit β	Complete	64	185	6	6	Mt, EDS, LIA
		10279	8282	Deposit β	Complete	50	133	5	10	Mt, EDS, PIXE, LIA, Img
		10280	8291	Deposit β	Incomplete	60	158	6	9	Mt, EDS, PIXE, LIA, Img
		10301	8289	Deposit β	Incomplete	49	195	4	10	Mt, EDS, LIA
		10303	8279	Deposit β	Complete	24	124	2	10	Mt, EDS
		10810	4059	Mur LvXI/XX	Fragment	34	132	6	?	Mt, EDS, Img
		10870	13854	Carré 8/12	Incomplete	200	216	11	?	Mt, EDS, Img
		11200	10105	Deposit ξ	Complete	54	197	8	9	Mt, EDS, Img

[a] Inventory number for archaeological objects situated in the National Museum of Beirut.
[b] Inventory number given and published by the site archaeologists (Dunand, 1954).
[c] Current state of the object. Does not refer to its corrosion.
[d] The weapon types have been defined by Philip (1989).
[e] Complete: The object is not missing any part.
[f] Incomplete: The object is missing some parts but most of its dimensions are still available.
[g] Fragment: Only a part of the object remains.
[h] SEM–EDS.
[i] Metallography.
[j] Image analyses.

Table 2
Site, context, Dimension, and typology of riveted spearheads, daggers and duckbill axes as well. The methods used for the study of each item are also mentioned.

Site	Weapons	Inv. number[a]	Pub. number[b]	Context	Current state[c]	Weight (g)	Preserved length (mm)	Max thickness (mm)	Type[d]	Method
Byblos	Riveted spearheads	9031	8374	Deposit γ	Incomplete[f]	40	123	5		Mt[i], EDS[h], Img[j]
		9046	9616	Deposit κ	Incomplete	217	271	8		Mt, EDS, Img
		9050	8361	Deposit γ	Incomplete	34	103	5		Mt, EDS, Img
		9051	8382	Deposit γ	Incomplete	56	129	6		Mt, EDS, Img
		10334	8418	Deposit γ	Incomplete	119	225	6		Mt, EDS, Img
		11102	10703	Deposit υ	Complete[e]	88	171	7		Mt, EDS, Img
		11109	10704	Deposit υ	Complete	198	230	9		Mt, EDS, Img
		11110	10698	Deposit υ	Complete	182	241	9		Mt, EDS, Img
		12329	8837	Deposit ε	Incomplete	120	230	7		Mt, EDS, Img
		12332	8839	Deposit ε	Incomplete	85	227	5		Mt, EDS, Img
Byblos	Daggers	9021	9665	Deposit λ	Incomplete	126	225	5	12	Mt, EDS, Img
		10811	3806	Bat. II sale C	Incomplete	44	172	3	13	Mt, EDS, Img
		11671	10122	Deposit ο	Incomplete	49	149	4	30	Mt, EDS, Img
		11686	10134	Deposit ο	Complete	65	203	5	32	Mt, EDS, Img
		11697	10132	Deposit ο	Incomplete	47	145	4	16	Mt, EDS, Img
		11854	9535	Deposit ι	Incomplete	48	202	4	30	Mt, EDS, Img
		11930	8344	Deposit β	Fragment[g]	9	80	2	?	Mt, EDS, Img
		27092	9172	Deposit ζ	Fragment	21	138	3	16	Mt, EDS, LIA
		27857	8365	Deposit γ	Incomplete	32	101	3	30	Mt, EDS, Img
Tell Arqa		26757	98.334–001	Burial 14.14	Complete	294	266	N/A[k]	13	Mt, EDS, LIA
Byblos	Duckbill axes	6273	10646	Deposit σ	Incomplete	127	103	14	1	Mt, EDS, PIXE, LIA
Tell Arqa		26759	98.334–004	Burial 14.14	Complete	425	123	N/A	1	Mt, EDS, LIA

[a] Inventory number for archaeological objects situated in the National Museum of Beirut.
[b] Inventory number given and published by the site archaeologists (Dunand, 1954).
[c] Current state of the object. Does not refer to its corrosion.
[d] The weapon types have been defined by Philip (1989).
[e] Complete: The object is not missing any part.
[f] Incomplete: The object is missing some parts but most of its dimensions are still available.
[g] Fragment: Only a part of the object remains.
[h] SEM–EDS.
[i] Metallography.
[j] Image analyses.
[k] N/A: not available.

Morr (2011). To facilitate the reader's access to the SEM–EDS elemental analyses results, an illustration showing the composition in arsenic and tin expressed in weight percentage is provided in Inline Supplementary Fig. S1.

Inline Supplementary Fig. S1 can be found online at http://dx.doi.org/10.1016/j.jas.2013.05.025.

The sub-set includes 22 socketed spearheads, 10 riveted spearheads, 10 daggers and 2 duckbill axes. These weapons will be designated by the inventory numbers assigned by the National Museum of Beirut. Previous morpho-typological works (Gernez, 2007; Miron, 1992; Philip, 1989) have shown that most of these objects are typical of MB I Levant. The typological affinities of each weapon will be given according to Philip (1989) in Tables 1 and 2 along with its dimensions, archaeological context and the type of analyses it underwent.

The socketed spearheads are separated into three groups (Fig. 1a–c) according to their size, technological features (El Morr, 2011; El Morr and Pernot, 2010, 2011), types (Gernez, 2007; Philip, 1989) and, in some cases, their archaeological context (Dunand, 1954). All three groups display a homogenised grain structure which resulted from heavy deformation and annealing operations. The first group is composed of socketed spearheads 9028, 10268, 10336, 11856, 11913, 11914, 11921, 27855, 27856, 27884 discovered at Byblos in the deposits β, ι, γ and λ (Dunand, 1954). They represent the largest type, dimension-wise, of MB I socketed spearheads in the Levant (Gernez, 2007; 351–2; Philip, 1989; 89). They are made of bronze containing between 11 and 15 wt% tin. Even though their grain structure is homogenised, occasional ($\alpha + \delta$) eutectoid phases which were formed after annealing are observed (Inline Supplementary Fig. S2). This group is designated here as "large spearheads".

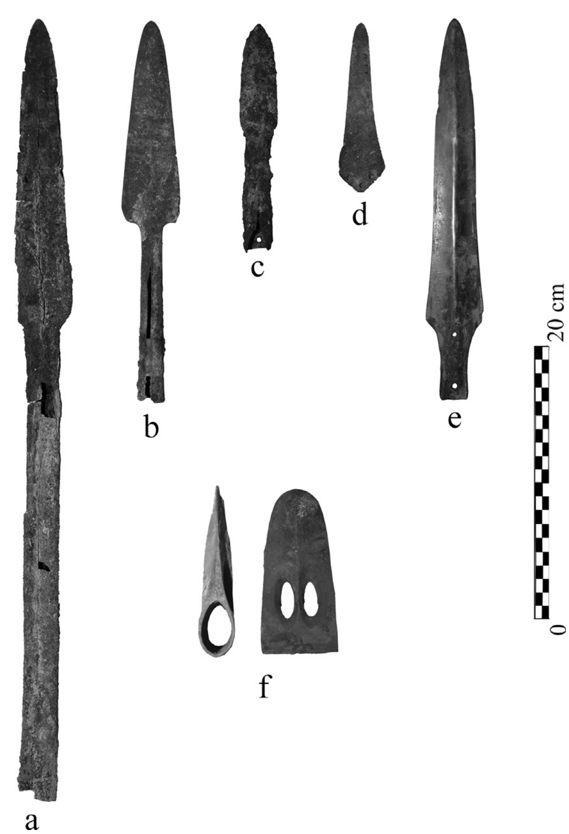

Fig. 1. Main weapons discussed in this paper. a – large spearhead; b – multi-piece spearhead; c – small spearhead; d – dagger; e – riveted spearhead; f – duckbill axe.

Inline Supplementary Fig. S2 can be found online at http://dx.doi.org/10.1016/j.jas.2013.05.025.

The second group is composed of socketed spearheads 10977, 10978, 10982 and 10997 discovered in deposit χ at Byblos. They are distinguished from the other socketed spearheads studied here by the presence of one or several collars surrounding the socket part (Fig. 2). These different elements are attached together by silver or silver–copper alloy braze situated exclusively over the joint separating the socket in length (El Morr and Pernot, 2011). These spearheads are made of bronze containing between 6 and 8 wt% tin. They will be designated as “multi-piece spearheads”.

The last group of spearheads, designated here as “small spearheads”, is morphologically more diverse. It includes weapons 10274, 10277, 10279, 10280 and 10301 discovered in the deposit β and 11200 from deposit ζ. We also included spearheads 10810 and 10870 which were found in poorly defined contexts at Byblos. The small spearheads are made of arsenical copper. The arsenic is generally present in amounts varying between 2 and 5 wt% except for spearhead 10277 where it is at 10 wt%. In spearheads 10274, 10277, and 10301 arsenic is mostly concentrated in arsenic rich inclusions (El Morr, 2011; 272–5). In the case of 10274 these inclusions (Inline Supplementary Fig. S3) also contain high amounts of antimony.

Inline Supplementary Fig. S3 can be found online at http://dx.doi.org/10.1016/j.jas.2013.05.025.

The riveted spearheads (Fig. 1e) were all discovered at Byblos. They include 9031, 9046, 9050, 9051, 10334, 11102, 11109, 11110, 12329, 12332 from deposits γ, κ, υ and ε. The group of daggers (Fig. 1d) contains 9021, 10811, 11671, 11686, 11697, 11854, 11930, 27857, and 27092 from deposits β, γ, σ, ζ and λ as well as dagger 26757 from Tomb 14.14 of Tell Arqa. SEM–EDS analyses revealed that the daggers and the riveted spearheads are made either of arsenical copper or bronze with varying amounts of arsenic and tin. The alloy recipes of 27092 and 26757 could not be verified due to heavy corrosion.

In addition to these groups two duckbill axes were selected. Duckbill axe 6273 was found in deposit σ at Byblos. It is mostly left as-cast and is made of bronze with around 8 wt% tin. Duckbill axe 26759 was discovered at Tomb 14.14 of Tell Arqa. It is heavily corroded so its alloy recipes could not be determined even though SEM–EDS analyses revealed the presence of copper and tin (El Morr, 2011; 259).

3. Method

As exemplified by previous work (Cattin et al., 2009, 2011), lead isotope analyses (LIA) were used to identify groups of data and to help constrain the potential provenance(s) of copper. The evaluation of copper “cleanness” was done using image analyses. In addition, elemental analyses using Particle Induced X-ray Emission (PIXE) were performed on several samples. This method is more sensitive than the previously used SEM–EDS and provides data that allow a better discrimination among copper coming from different minerals as well as among copper qualities. PIXE analyses were applied to samples prepared for metallographic examinations. The work method, the operating conditions and the resulting data for SEM–EDS (JEOL JSM 6460-Lv and Oxford Instruments X-max Silicon Drift Detector) and metallographic examinations are detailed in El Morr and Pernot (2010, 2011) and therefore will not be exposed here.

Metal chips were sampled from each object using a jeweller’s saw. In some cases the samples used for LIA were not cut in close proximity to the ones that were used for metallographic examinations and elemental analysis by SEM–EDS and PIXE. This was crucial when sampling the multi-piece spearheads. As has already been mentioned, these weapons contain a copper–silver braze on their socket joints that could contaminate the spearhead’s copper alloy lead isotopic composition. Therefore, in the case of multi-piece spearheads, LIA samples were collected from the blade or from the socket on the opposite side of the joint.

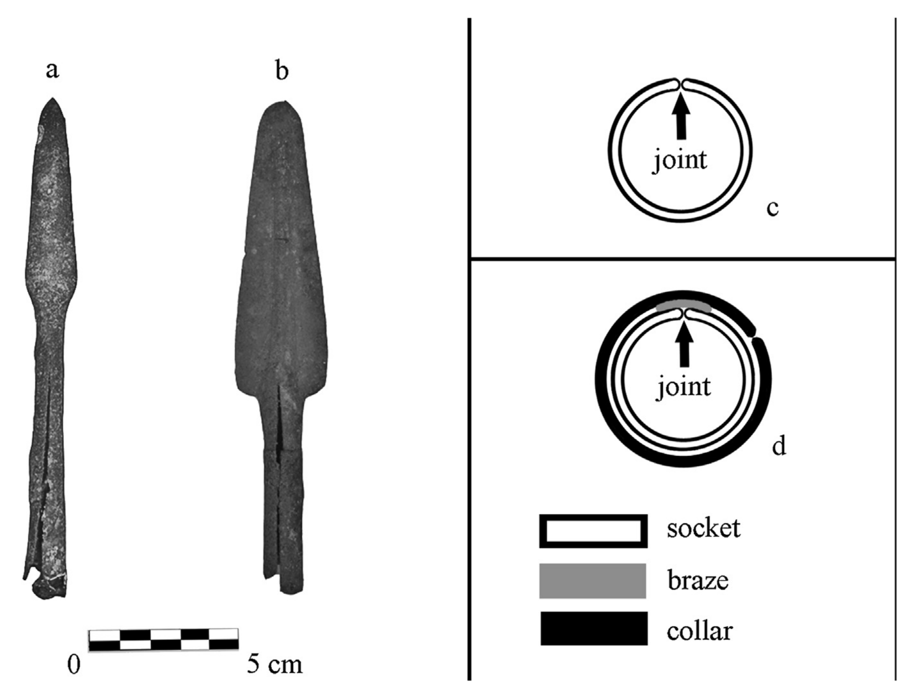

Fig. 2. Socket form of common MB I socketed spearheads (a) at Byblos and Tell Arqa and the form of the multi-piece spearheads with a collar around the socket part (b). Also included in this illustration is a sketch of common MB I spearheads socket section (c) and the socket section of a multi-piece spearhead showing the position of the braze and the collar in respect to the socket’s joint (d).

The lead isotope analyses were carried out on a Thermo-Scientific Neptune multi-collector ICP – mass spectrometer at the University of Ghent (Belgium), Department of analytical chemistry. They were applied to large spearheads 10268, 11856, 11913, 11914, 11921, 27855, 27856, 27884, to multi-piece spearheads 10977, 10978, 10982, 10997, to small spearheads 10274, 10277, 10279, 10280, 10301, to daggers 27092, 26757 and to duckbill axes 6273 and 26759. Previous metallographic and SEM–EDS work didn't reveal any indication of added lead in any of these weapons.

In order to avoid contamination, it was necessary to clean the solid samples (from a few mg to about 200 mg) in two steps, (1) in diluted (0.5 M) HCl coupled with ultra-sonic bath and (2) in Micropure water. Although the standard procedure implies the repetition of this process twice, the corroded, powdery, and soluble samples 10268, 10978, 10982, 10997, 11913, 11914, 11921, 26757, 26759, 27884 and 27092 received only a rapid washing procedure. There are no indications that the single or repeated washing procedure or the resin material – used as a means of consolidation and conservation of the weapons of Tell Arqa – affected the analytical results.

The cleaned samples were dissolved in aqua regia. Lead was extracted from ion-exchange resin (Pb spec™) using Bio-Spin Disposable chromatography columns from BioRad. The samples were loaded in 1 M HNO_3, rinsed with 0.1 M HNO_3 and the lead was collected using 0.05 M $(NH_4)_2C_2O_4$ according to the procedure developed by D. De Muynck and F. Vanhaecke (De Muynck et al., 2007; see also Huelga-Suarez et al., 2012).

Subtraction of analytical blank representing less than 0.05% of total Pb was performed and a correction for ^{204}Hg was made based on monitoring the ^{202}Hg peak. The influence of the procedural blank was evaluated on each sample and was generally found to be negligible (<0.45% of the total lead content) except for analyses of 10997, 26759, but they appear to fit well within the other data, and 27884, 10301, 10277, due to the very low lead content in the copper. The lead isotope data of the later three weapons have to be considered with caution.

Analytical results were corrected for mass bias using Tl NIST SRM 997 and Pb NIST SRM 981 reference materials – exponential factor, $^{205}Tl/^{203}Tl = 2,3871$, lead reference values from Galer and Abouchami (1998). Repeated measurements of the NIST SRM 981 reference material provided the following values, based on 30 analyses (uncertainties at two sigma): $^{208}Pb/^{204}Pb = 36.725 \pm 0.004$, $^{207}Pb/^{204}Pb = 15.500 \pm 0.001$, $^{206}Pb/^{204}Pb = 16.942 \pm 0.002$, $^{208}Pb/^{206}Pb = 2.16770 \pm 0.00002$, $^{207}Pb/^{206}Pb = 0.91490 \pm 0.00001$.

Elemental analyses were carried out by μ-beam Particle Induced X-ray Emission (PIXE) on the AGLAE accelerator facility at the Centre de Recherche et de Restauration des Musées de France (Dran et al., 2000). A specific protocol for elemental analysis of copper-based alloys, which includes Co filtering and scanning for homogenisation, was used. A proton beam extracted in helium was used. The spot size was around 50 μm but homogenisation scanning of circa 1 mm^2 was performed. The PIXE's analytical performances were checked using bulk metallic certified reference materials (CRM). This procedure is detailed in Bourgarit and Thomas (2012). The analyses were applied to uncorroded chips of metal samples from weapons that were also investigated by LIA: large spearheads 11856, 27855, 27856, 27884, multi-piece spearheads 10977, 10978, 10997, small spearheads 10274, 10279, 10280 and duckbill axe 6273. All the results of PIXE analyses are expressed in weight percentage.

Image analyses were applied to optical microscope micrographs of polished samples in order to evaluate the quantities of permanent inclusions observed in the copper alloys such as copper sulphides and lead globules. The evaluation consists in measuring the surface fraction occupied by these inclusions in comparison to the surface of the copper alloy matrix. The quantification of permanent inclusions is expressed in area percentage (A %). Only the inclusions larger than 6 μm were measured. This parameter was chosen to avoid counting in polishing products left from the sample preparation phase. The ($\alpha + \delta$) eutectoid phases observed in the large spearheads were not measured. These were considerably bigger than the permanent inclusions (Inline Supplementary Fig. S2) and could be easily omitted from the measuring process. The measurements were done by using scripts from Pandore (version 6.4), which is a standardized library of image processing operators, developed by GREYC laboratory (http://www.greyc.ensicaen.fr/~regis/Pandore/index.html). The samples investigated here represent 32 pre-selected objects (Table 4). The pre-selection process involved investigating the sections (transversal and longitudinal in regards to the long axis) and the polished surface of uncorroded samples collected from 66 MB I weapons from Byblos and Tell Arqa. This multi-section metallographic examination allowed omitting samples with inter-granular corrosion, flattened copper sulphide or arsenic and antimony rich inclusions. All objects containing more than 2 wt% of lead were not included as they could be deliberate leaded copper alloys. Consequently, it was possible to study samples presenting annealed homogenised structure with round, cylindrical or "cigar" shaped copper sulphide inclusions and lead globules. In this work, only micrographs representing the polished surface of an object were analysed. These are the only ones guaranteed to show the full length of the elongated copper sulphide inclusions. Consequently, measurements on micrographs of transversal or longitudinal sections were avoided as the elongated inclusions are not necessarily oriented parallel or perpendicular to the object's long axis. The results obtained from image analyses remain an approximate value destined to show general tendencies only. This is partly due to the heterogeneity in the distribution of inclusions and the "irregular" form of some of these elements. Furthermore, the maximum dimensions of the inclusions on the polished surfaces are not always totally uncovered. Due to this lack of precision, attempts to separate between copper sulphide inclusions and lead globules were avoided as this could complicate the data without improving the results. The comparison of the proportion of inclusions (Table 5) from several micrographs of different magnifications but from the same object generally shows a difference in measurement varying between 0.01 and 0.1 A %. Occasionally, in cases where the amount of inclusions is high, this difference could reach 0.4 A %. The selected 32 objects are all from Byblos, they include large spearheads 9028, 10336, 11856, 27855, 27856, 27884, multi-piece spearheads 10977, 10978, 10997, small spearheads 10279, 10280, 10810, 10870, 11200, riveted spearheads 9031, 9046, 9050, 9051, 10334, 11102, 11109, 11110, 12329, 12332 and daggers 9021, 10811, 11671, 11686, 11697, 11854, 11930 and 27857.

4. Results

4.1. Elemental analyses by PIXE

As major elements have already been published elsewhere (El Morr and Pernot, 2010, 2011), the focus of this work will be on minor and trace elements data obtained by PIXE (Table 3). The PIXE results partially confirm the separation of the three spearhead groups mentioned earlier.

The composition (Fig. 3) of multi-piece spearheads 10977, 10978 and 10997 appears to be homogeneous as they contain almost similar Fe, As, S and Pb contents. This said, 0.1 wt% silver is found in spearhead 10997 while in the other two spearheads there is less than 0.04 wt% of the same metal (Table 3). The high amount of silver in 10997 is probably the result of a contamination by the silver–copper braze due to the proximity of the sample taken for PIXE to the socket's joint.

Table 3
Elemental analyses by PIXE.

Inv. nb.[a]	S	Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Se	Ag	Cd	In	Sn	Sb	Te	Au	Hg	Pb	Bi
11856	<0.02	<0.011	<0.004	0.13	<0.012	<0.02	87	<0.14	<0.08	<0.023	<0.02	<0.024	<0.029	12.9	<0.03	<0.043	<0.02	<0.02	0.27	<0.03
27855	<0.03	<0.011	<0.002	0.06	<0.009	<0.01	88	<0.07	<0.09	<0.013	<0.02	<0.022	<0.047	12.0	<0.04	<0.051	<0.04	<0.03	0.35	<0.03
27856	<0.04	<0.011	<0.001	0.11	<0.005	<0.01	88	<0.16	0.19	<0.005	<0.01	<0.023	<0.024	11.8	<0.03	<0.050	<0.04	<0.04	0.22	<0.03
27884	<0.02	<0.007	<0.002	0.05	<0.011	<0.02	87	<0.09	<0.03	0.023	<0.01	<0.019	<0.031	13.2	<0.04	<0.044	<0.04	<0.03	<0.04	<0.02
10977	0.15	<0.003	<0.004	0.23	<0.009	<0.02	94	<0.26	0.12	<0.013	<0.02	<0.018	<0.019	5.7	<0.04	<0.037	<0.03	<0.03	0.09	<0.03
10978	0.14	<0.010	<0.004	0.29	<0.006	<0.02	92	<0.16	0.28	<0.013	<0.04	<0.023	<0.026	7.2	<0.04	<0.043	<0.03	<0.05	0.07	<0.03
10997	0.16	<0.009	<0.004	0.40	<0.006	<0.02	92	<0.16	0.59	<0.007	0.11	<0.015	<0.026	6.9	<0.03	<0.038	<0.04	<0.06	0.12	<0.03
10274	<0.03	<0.009	<0.003	0.03	<0.010	0.14	96	<0.18	3.50	<0.020	<0.01	<0.005	<0.016	<0.02	0.40	<0.034	<0.02	<0.19	0.08	<0.03
10279	<0.06	<0.006	<0.004	0.90	<0.024	<0.01	97	<0.18	2.49	<0.005	<0.05	<0.005	<0.008	<0.08	<0.05	<0.015	<0.02	<0.23	0.19	<0.04
10280	0.09	<0.008	<0.002	0.54	<0.006	<0.02	97	<0.15	2.59	<0.027	<0.02	<0.010	<0.007	<0.01	0.55	<0.025	<0.12	<0.15	<0.03	<0.03
6273	0.23	<0.007	<0.003	0.06	0.040	0.82	89	<0.10	1.52	<0.018	<0.02	<0.018	<0.016	8.6	<0.03	<0.049	<0.06	<0.11	0.91	<0.04

All elements are expressed in weight percentage.
The sign “<” refers to the detection limit.
[a] Inventory number of the National Museum of Beirut.

Table 4
Area percentage (A%) of permanent inclusions.

Weapons	Inv. nb.[a]	Pub. nb.[b]	A %[c]
Riveted spearheads	9031	8374	1.4
	9046	9616	1.0
	9050	8361	1.1
	9051	8382	1.5
	10334	8418	1.4
	11102	10703	1.7
	11109	10704	1.6
	11110	10698	0.4
	12329	8837	1.8
	12332	8839	1.1
Large spearheads	9028	9669	0.1
	10336	8441	1.5
	11856	9523	0.1
	27855	9626	0.4
	27856	9667	0.3
	27884	9632	0.2
Multi-piece spearheads	10977	10836	1.5
	10978	10834	1.3
	10997	10845	1.2
Small spearheads	10279	8282	1.1
	10280	8291	1.5
	10810	4059	1.0
	10870	13854	0.4
	11200	10105	0.5
Daggers	9021	9665	0.2
	10811	3806	0.9
	11671	10122	1.7
	11686	10134	0.2
	11697	10132	0.9
	11854	9535	2.4
	11930	8344	0.9
	27857	8365	1.4

[a] Inventory number of the National Museum of Beirut.
[b] Published inventory number (Dunand, 1939, 1954).
[c] Area percentage representing the surface fraction occupied by the permanent inclusions in a copper alloy matrix.

Large spearheads 11856, 27855, 27856 and 27884 also have similar compositions even though some exceptions are observed: spearhead 27856 contains 0.2 wt% As while in the remaining three large spearheads the amount of arsenic is less than 0.09 wt%. Lead in spearhead 27884 is below the detection limit (DL Pb = 0.04 wt%) whereas in the other large spearheads it is around 0.3 wt%. Apart from lead, the large spearheads generally contain amounts of Fe, As and S that are lower than the other groups of weapons.

The small spearheads display more heterogeneous compositions especially when it comes to antimony and nickel: in spearheads 10274 and 10280, Sb is respectively at 0.4 and 0.5 wt%. In

Table 5
Comparison of the area percentage (A%) of inclusions measured from micrographs of different optical microscope magnifications.

Inv. nb.[a]	Pub. nb.[b]	Avrg. A %[c]	Diff. A %[d]	Inclusion % according to microscope magnification			
				×50[e]	×100[e]	×200[e]	×500[e]
27855	9626	0.44	0.03			0.46	0.43
27856	9667	0.34	0.12			0.4	0.28
27884	9632	0.24	0.02	0.25			0.23
10978	10834	1.31	0.08			1.27	1.35
10811	3806	0.85	0.08			0.92	0.8
11200	10105	0.53	0.03	0.51	0.54		
10279	8282	1.06	0.38		1.38		1

[a] Inventory number of the National Museum of Beirut.
[b] Published inventory number (Dunand, 1939, 1954).
[c] The average of the measurements of area percentage of permanent inclusions from micrographs of different magnifications.
[d] The difference between the measurements of area percentage of permanent inclusions from micrographs taken at different magnifications.
[e] Optical microscope magnification.

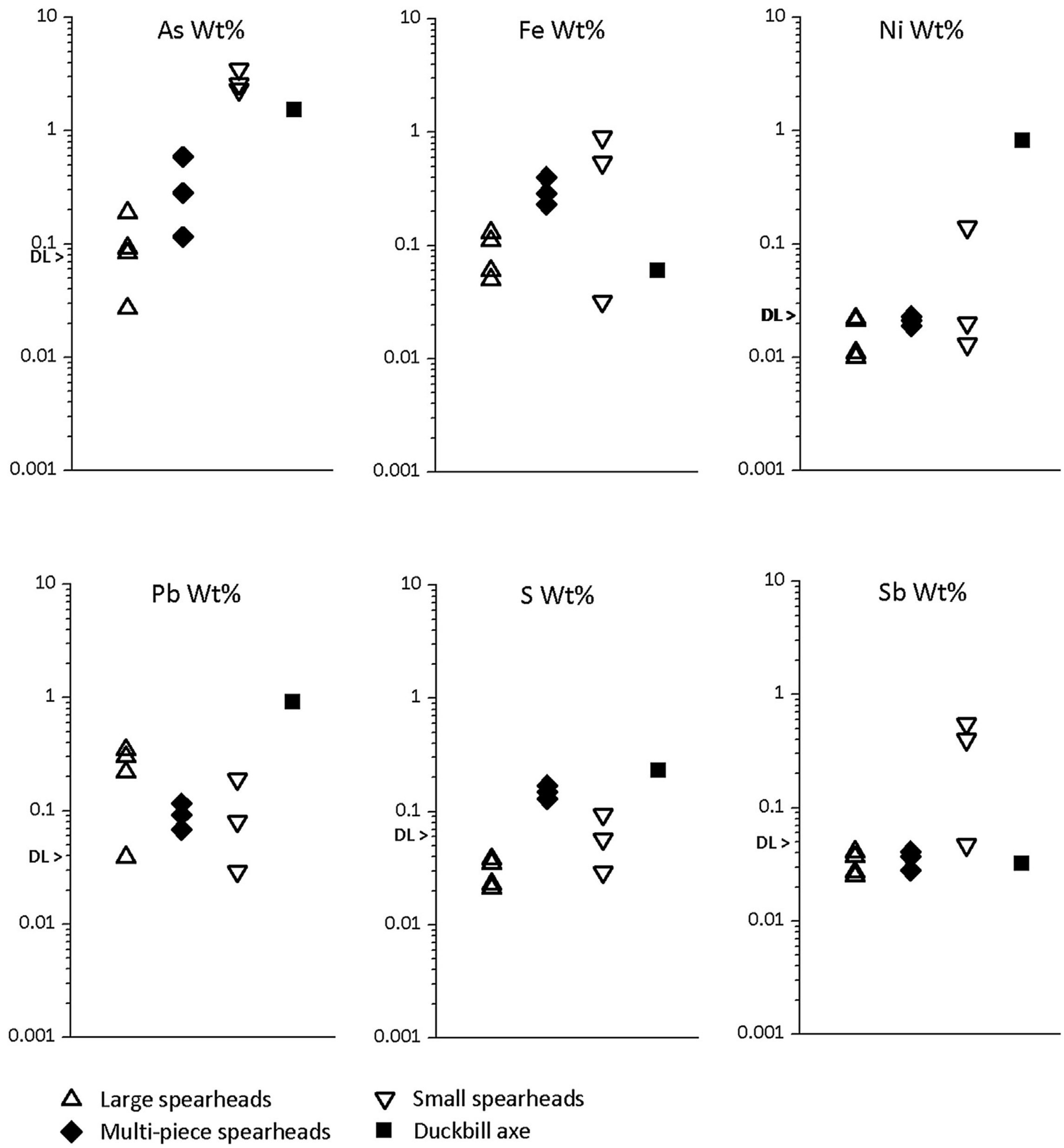

Fig. 3. Minor and trace elements composition (wt%) of the weapons analysed by PIXE. DL stands for Detection limit.

10279, Sb is below the detection limit (DL Sb = 0.05 wt%). Small spearhead 10274 revealed relatively high amounts of Ni (0.14 wt%) and low amounts of Fe (0.03 wt%). This is not the case in the other two spearheads where Ni is below the detection limit (DL Ni = 0.02 wt%) and Fe is higher than 0.5 wt%. The differences in S and Pb contents are less striking.

Duckbill axe 6273 is characterized by high amounts of impurities, such as 0.8 wt% Ni and 0.04 wt% Co, which are below limits of detection in the other weapons and particularly in the ones made of bronze. In comparison with bronze spearheads, 6273 exhibits also higher amounts of lead (0.9 wt%) and arsenic (1.5 wt%). This diversity in composition is very common in duckbill axes (Philip, 2006; Shalev, 2009; Shalev et al., 2013) which might be the result of recycling and thus does not necessarily imply an intentional alloying or a particular copper source derivative, therefore it is useful to keep in mind that the copper alloy used in this particular weapon might contain recycled components.

4.2. *Permanent inclusion quantification*

The work on copper “cleanness” of the 32 weapons of Byblos (Fig. 4) shows that most of the large spearheads (9028, 11856, 27855, 27856, 27884) present very low amounts of permanent inclusions (between 0.1 and 0.5 A %). The three multi-piece spearheads revealed higher, but coherent, amounts of inclusions (between 1.2 and 1.5 A %). The difference in the amount of permanent inclusions between the former and latter group is visually evident from the metallographic micrographs (Fig. 5). In the case of the riveted spearheads, small spearheads and daggers there is a wide distribution in the quantity of inclusions where variations from 0.2 to 2 A % are observed. The riveted spearheads and the daggers present generally diverse recipes of both arsenical copper and bronze. The diagram in Fig. 4 shows that there is no clear correlation between the inclusions’ A % and the tin content. Both low- and high-tin bronzes exhibit low and high rates of inclusions.

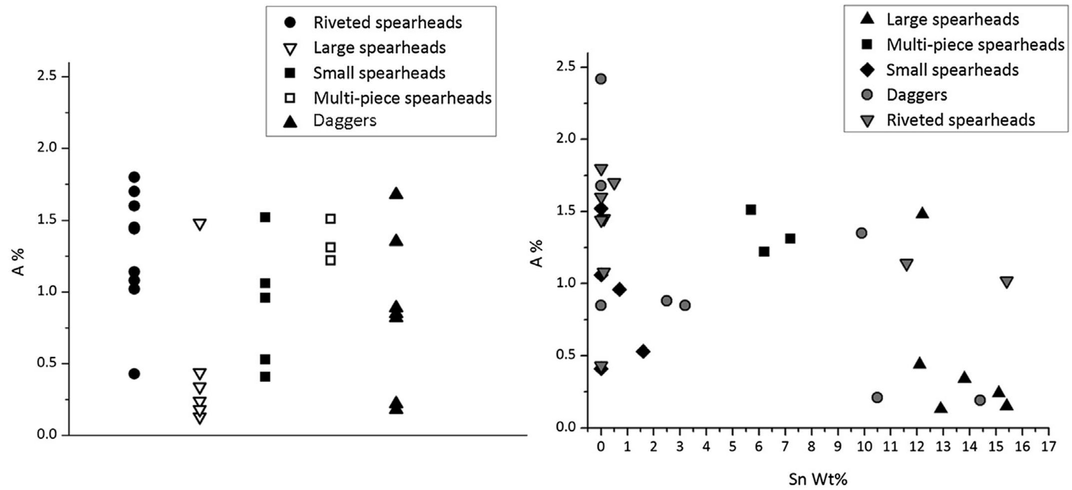

Fig. 4. On the left, diagram showing the area percentage (A %) of permanent inclusions of each of the 32 investigated weapons sorted by their main groups. On the right, diagram plotting the Sn composition (wt%) of these weapons versus their area percentage (A %) of permanent inclusions.

4.3. *Lead isotope analyses*

The lead isotope ratios of the Byblos and Tell Arqa weapons are represented on scatter diagrams $^{208}Pb/^{206}Pb$ vs. $^{207}Pb/^{206}Pb$, $^{207}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ (Fig. 6) in order to facilitate their description and interpretation.

Compared with the overall set of LIA data (Table 6 and Fig. 6), the lead isotope ratios of large spearheads 10268, 11856, 11913, 27855, 27856 and 27884, multi-piece spearheads 10977, 10978, 10982 and 10997, dagger 27092 and duckbill axe 26759 could be seen as grouping with $^{207}Pb/^{206}Pb$ ratios ranging from 0.84513 to 0.85362, and $^{208}Pb/^{206}Pb$ ratios from 2.08698 to 2.09984 (Fig. 6). Slightly lower on the $^{208}Pb/^{206}Pb$ vs. $^{207}Pb/^{206}Pb$ diagram, duckbill axe 6273 and small spearhead 10279 have very close $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ isotope ratios, but diverge considering the ratios including the ^{204}Pb. On the $^{207}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ diagram, all these weapons display lead isotope ratios which are between the young and the old upper crust lines as defined by Kramers and Tolstikhin (1997). The lead evolution model developed by these authors offers additional help for provenance analyses. It permits the identification of the origin of the mineralization fluid (mantel, young/old, upper/lower crust), the potential contribution of the host-rock, the ore-forming events of mineralization and – with all due caution – their age (Guénette-Beck et al., 2009; Kramers and Tolstikhin, 1997). The display of the Kramers and Tolstikhin model along the artifacts data allows their interpretation in a larger geological context.

On the $^{208}Pb/^{206}Pb$ vs. $^{207}Pb/^{206}Pb$ diagram small spearheads 10301, 10280, 10274, large spearheads 11914 and 11921 and dagger 26757 display higher and more scattered lead isotope ratios than the former weapons. They seem to evolve on the same trend line except for spearhead 10301 that is made from a copper with low lead content and therefore potentially subject to lead contamination. On the $^{207}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ (Fig. 6) the lead isotope ratios of 10280, 11914 and 11921 are close to or on the young upper crust line whereas 10274, 10301 and 26757 are below it.

Small spearhead 10277 has lead isotope ratios distinct from the other weapons (Fig. 6). They are anomalous with respect to the Kramers and Tolstikhin (1997) lead isotope evolution model. This can be explained by highly radiogenic lead isotope ratios, probably reflecting input from uranium enriched minerals in the ore source.

5. Discussion

5.1. *Copper groups and copper qualities*

The multi-piece spearheads 10977, 10978 and 10997 are all made of similar "dirty" copper as testified by the high proportion of permanent inclusions and the relatively high contents in As, Fe, and S. The similarity of impurity patterns and the lead isotope ratios is not incompatible with a common copper source(s). This said, the discrepancy shown by the lead isotope ratios does not allow the confirmation of their belonging to the same batch of ore or metal.

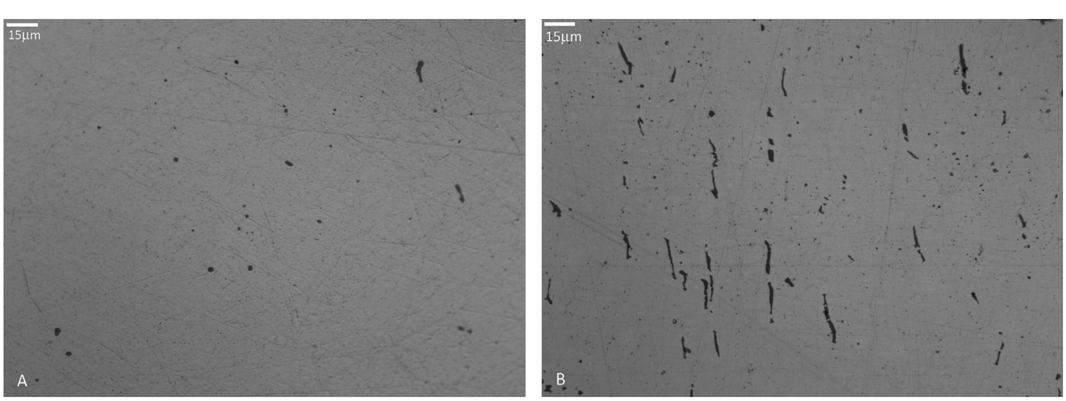

Fig. 5. Two micrographs (reflected light microscopy) showing the difference in the quantity of permanent inclusions (in dark colours) between large spearhead 27884 (A) and multi-piece spearhead 10978 (B).

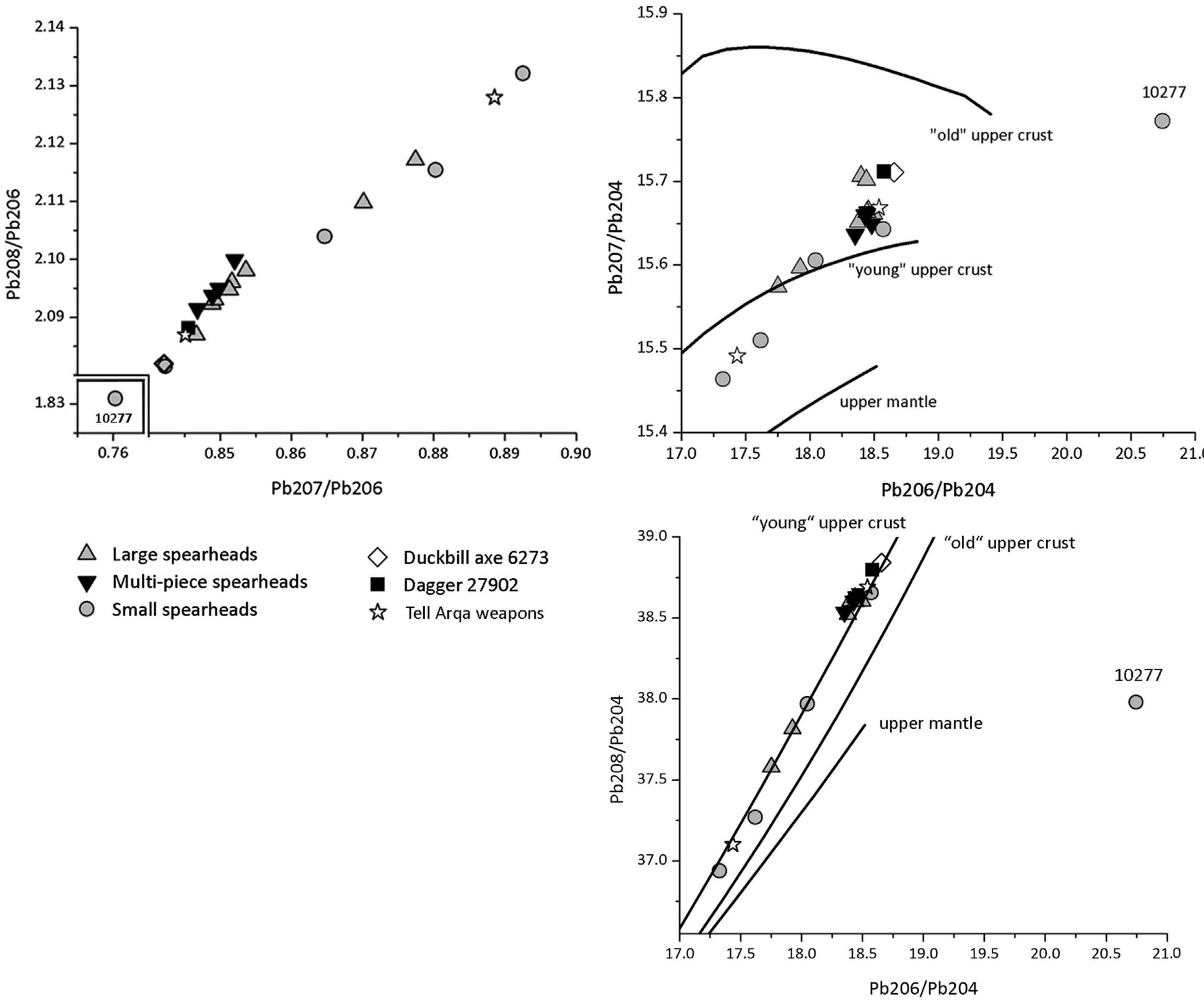

Fig. 6. Lead isotope ratios of the weapons of Byblos and Tell Arqa represented on the $^{208}Pb/^{206}Pb$ vs. $^{207}Pb/^{206}Pb$, $^{207}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ scatter diagrams.

Table 6
Lead isotope ratios of the weapons investigated.

Site	Weapons	Inv. nb.[a]	Pres.[b]	$^{206}Pb/^{204}Pb$	Error 2σ	$^{207}Pb/^{204}Pb$	Error 2σ	$^{208}Pb/^{204}Pb$	Error 2σ	$^{208}Pb/^{206}Pb$	Error 2σ	$^{207}Pb/^{206}Pb$	Error 2σ
Byblos	Large spearheads	10268	C.C[c]	18.453	0.007	15.665	0.007	38.610	0.016	2.09234	0.00023	0.84890	0.00009
		11856	S.M[d]	18.378	0.006	15.651	0.005	38.521	0.014	2.09608	0.00020	0.85166	0.00007
		11913	C.C	18.498	0.005	15.661	0.005	38.604	0.013	2.08696	0.00023	0.84665	0.00007
		11914	P.C[e]	17.924	0.004	15.597	0.003	37.816	0.009	2.10981	0.00020	0.87014	0.00006
		11921	C.C	17.750	0.005	15.574	0.004	37.580	0.012	2.11721	0.00026	0.87742	0.00006
		27855	S.M	18.400	0.011	15.706	0.011	38.604	0.025	2.09808	0.00039	0.85362	0.00013
		27856	S.M	18.443	0.006	15.663	0.006	38.602	0.014	2.09305	0.00023	0.84926	0.00007
		27884	S.M	18.439	0.115	15.701	0.105	38.626	0.279	2.09475	0.00171	0.85132	0.00107
Byblos	Multi-piece spearheads	10977	S.M	18.480	0.006	15.649	0.005	38.650	0.014	2.09141	0.00020	0.84679	0.00006
		10978	S.M	18.427	0.005	15.660	0.004	38.603	0.011	2.09493	0.00020	0.84982	0.00007
		10982	S.M	18.451	0.005	15.663	0.004	38.631	0.011	2.09375	0.00024	0.84892	0.00007
		10997	S.M	18.352	0.014	15.636	0.013	38.535	0.032	2.09984	0.00045	0.85205	0.00014
Byblos	Small spearheads	10274	S.M	17.325	0.005	15.464	0.005	36.940	0.012	2.13211	0.00020	0.89254	0.00008
		10277	S.M	20.744	0.046	15.772	0.036	37.981	0.090	1.83091	0.00084	0.76031	0.00026
		10279	S.M	18.571	0.005	15.643	0.004	38.656	0.012	2.08149	0.00019	0.84231	0.00006
		10280	S.M	18.047	0.004	15.606	0.004	37.970	0.010	2.10394	0.00021	0.86471	0.00007
		10301	S.M	17.620	0.035	15.510	0.031	37.270	0.077	2.11542	0.00106	0.88029	0.00039
Byblos	Daggers	27092	C.C	18.581	0.004	15.711	0.004	38.798	0.011	2.08806	0.00019	0.84557	0.00006
Tell Arqa		26757	C.C	17.435	0.005	15.491	0.004	37.102	0.012	2.12801	0.00022	0.88854	0.00007
Byblos	Duckbill	6273	S.M	18.657	0.005	15.711	0.004	38.843	0.012	2.08194	0.00021	0.84209	0.00007
Tell Arqa	axes	26759	C.C	18.539	0.018	15.669	0.016	38.691	0.041	2.08698	0.00045	0.84513	0.00018

[a] Inventory number of the National Museum of Beirut.
[b] Preservation refers to the presence or absence of corrosion. The description is based on metallographic observation of polished sample sections.
[c] Completely corroded.
[d] Sound metal.
[e] Partially corroded.

Based on its lead isotope ratios, multi-piece spearhead 10982 could belong to this group as well, but the lack of minor and trace element data does not allow any confirmation.

Compared to all other investigated weapons, large spearheads 11856, 27855, 27856 and 27884 are made of the "cleanest" copper, bearing the lowest amount of permanent inclusions and impurities (As, Fe and S). They also display similar lead isotope ratios and elemental composition despite occasional disparities. These results point to a similar copper source, and possibly to a similar degree of copper refining. Based on their lead isotope ratios, large spearheads 10268 and 11913 could belong to this group as well, but the lack of minor and trace element data prevents any firm attribution. The large spearheads 11914 and 11921 show much higher $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ abundance ratios than the rest of the other large spearheads, which may pertain to completely different copper sources. This in turn would mean that contextual, stylistic and technologic similarities do not directly imply the use of the same copper. Yet, one must bear in mind that these two spearheads are heavily corroded, and it is quite possible, as previously shown by Hauptmann et al. (1999), that their lead isotope composition was affected by soil contamination. Large spearhead 9028 is also made of a "clean" copper, however the lack of LIA and trace element data prevents us from assimilating it to any group of weapons.

The clear choice of high quality copper for most large spearheads demonstrate that the MB I craftsmen of Byblos were able to distinguish between "clean" and "dirty" copper. Moreover, they probably knew of the influence of copper quality on mechanical properties and on the efficiency of metallurgical treatments. High quality copper was indeed almost exclusive to the large spearheads made of bronze containing high amounts of tin (between 11 and 15 wt%) – which are rather fragile alloys (Lechtman, 1996; Piccardo et al., 2004) – that were subject to heavy plastic deformation that could reach 85% of thickness reduction (see El Morr, 2011; 149; El Morr and Pernot, 2011). The relationship between alloy recipe, heavy hammering and weapon dimensions as well as the sample condition (see selection criteria in Section 3) restricted the number of weapons where "clean" copper is observable. Indeed, the large spearheads, which are the most likely candidates to display this relationship, are rare, especially outside Byblos. Even though the number of cases investigated is not sufficient, we chose to generalize this pattern of selective use of copper qualities to the craftsmen of MB I Levant as we hope this will encourage other scholars working on the Levant metallurgy to investigate this aspect.

Lead isotope ratios of both multi-piece spearheads (10977, 10978, 10982 and 10997) and large spearheads (11856, 27855, 27856 and 27884) belong to the same area in the diagrams and in some cases almost overlap. It is possible that the metal of all these weapons refers to the same copper source area even though the impurity patterns are not completely identical. Indeed, if linked to various degrees of refinement, the different impurity patterns are not incompatible with this suggestion. We may thus assume that at least two coppers with two different qualities but coming from the same area were used at Byblos during the MB I for the manufacture of weapons. The copper qualities could have been evaluated through the observation of their thermo-mechanical behaviour (softening, cracking…) during hammering and annealing tests. Moreover, as suggested by Moorey (1999; 243) for Mesopotamia, copper might have been further refined by the Byblos's craftsmen themselves in order to obtain the required quality.

5.2. Copper sources

The results were compared to the available data of lead isotope field signatures of possible Bronze Age copper sources form the eastern Mediterranean, the Near East and Eastern Europe. This comparison shows that the lead isotope ratios of the Byblos and Tell Arqa weapons are completely incompatible with the ore signatures of Bulgaria (Gale et al., 1991, 2000), Cyprus (Attanasio et al., 2001; Gale et al., 1997), Greece and the Aegean (Chalkias et al., 1988; Gale et al., 1985a,b; Stos-Gale, 1989; Stos-Gale and Gale, 2003; www.oxalid.arch.ox.ac.uk), Saudi Arabia (Bokhari and Kramers, 1982; Stacey et al., 1980), Serbia (Gale et al., 1991, 2000; Pernicka et al., 1993) and Timna (Gale et al., 1990; Hauptmann, 2007).

With regard to the Anatolian mining districts (Hirao et al., 1995; Sayre et al., 2001), weapons 11913, 26759 and 27092 overlap, on the three scatter diagrams, with the ore field signature of Trabzon on the Black sea (Fig. 7). The lead isotope ratios of spearhead 10279 are compatible with the ore field signature of Artvin situated east of Trabzon. The lead isotope ratios of these four weapons and of fenestrated axe 6273 are compatible with the copper ore signatures of the Massive Brown Sandstone (MBS) outcrop (Fig. 7) at Faynan (Hauptmann, 2007; 79–80) in Transjordan. They show lesser affinities on the $^{207}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ diagram. It is possible, however, that the copper used at Byblos and Tell Arqa derives from a mix of ores from the two main outcrops at Faynan, i.e. the Massive Brown Sandstone (MBS) and the Dolomite–Limestone–Shale (DLS). If this is true, then these five weapons along with 10268, 10978, 10977, 10982, 10997, 11856, 26759, 27855, 27856 and 27884 could be included in the common signature of these two outcrops. Copper rich in arsenic (>1 wt%) is unlikely to have been produced from Faynan ores (Hauptmann, 2007; 76) and thus the copper used in spearhead 10279 containing 2.5 wt% As could not come from this source unless arsenic was added separately (Rehren et al., 2012; Thornton et al., 2009). Overall, the composition of minerals in MBS and DLS is of no help in rejecting this region as a possible source as it is not incompatible with the elemental data of the copper-based artefacts. According to Hauptmann (2007; 152) however, there are no or very limited mining activities at Faynan during the Middle Bronze Age I compared to the Early Bronze Age and the Iron Age. The few traces of activity are mostly found at the DLS outcrops and thus it is unlikely that these 15 weapons were made with Faynan copper.

Comparing the lead isotope ratios of the weapons of Byblos and Tell Arqa to the available data from Iran (Fig. 8) shows that duckbill axe 6273 and small spearhead 10279 have lead isotope ratios similar to copper ores from the Karkas mountain (Pernicka et al., 2011). Radiocarbon dating has revealed that copper was extracted from the Vešnāve mine situated in the Qom-Kāšān region during the first half of the 2nd millennium BC (Stöllner et al., 2011). Dagger 27902, large spearhead 11913 and duckbill axe 26759 show signature affinities with ores from the Anārak region on the western central Iranian plateau (Pernicka et al., 2011). As no elemental composition could be obtained from those highly corroded samples, it is not possible to discuss their compatibility with the minerals. Spearheads 10268, 10977, 10978, 10982, 27855 and 27856 are compatible with the Pb–Zn ore deposit fields of the Sanandaj–Sirjan Zone (Mirnejad et al., 2011) and the Deh-Hossein mine lead isotope field (Nezafati et al., 2009). The copper ores of the Deh-Hossein mine – mainly chalcopyrite, arsenopyrite, cuprite – are not incompatible with the trace element patterns of the above-mentioned artefacts. This mine contains both copper and tin ores and was worked at least during the first quarter of the 2nd millennium BC. According to recent lead isotope data (Begemann and Schmitt-Strecker, 2009), north-west Iran is a very plausible copper source as it appears that it provided at least Mesopotamia with this metal.

It has been demonstrated before that the multi-piece spearheads 10977, 10978, 10982, 10997 and large spearheads 11856, 27855, 27856 and 27884 have elemental and lead isotopic

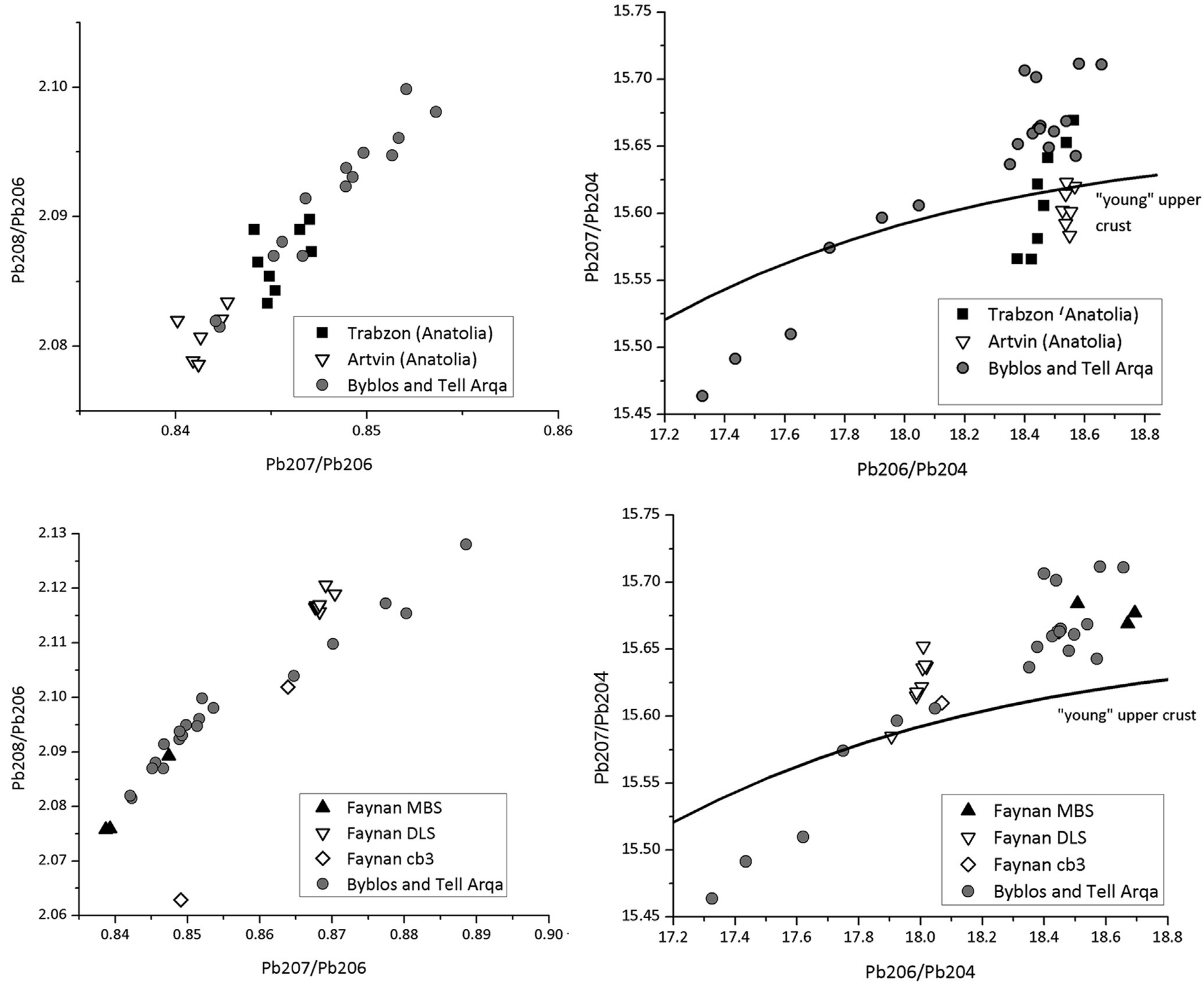

Fig. 7. Lead isotope ratios of the weapons of Byblos and Tell Arqa plotted on the $^{208}Pb/^{206}Pb$ vs. $^{207}Pb/^{206}Pb$ and $^{207}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ with the copper ore field signatures of Trabzon and Artvin in Anatolia and Faynan MBS (Massive Brown Sandstone), DLS (Dolomite–Limestone–Shale) and cb3 (Variegated Sand- and Clay-stones).

affinities. It would be expected then that the copper ore field to which these two groups belong should include all of their constituent members. The fact that 10997, 11856 and 27884 are not included in the Iranian ore fields casts doubt on the compatibility of these sources. This, of course, might be related to an insufficient number of samples defining ancient Iranian sources as Pernicka et al. (2011) remarked. Until further data are made available, the copper ores of Iran could not be considered as the sole source for the copper used in the Byblos weapons even though they remain a strong contender given the compatibility of their isotopic signature and the date of exploitation.

The copper ore field of Masirah Island (Begemann et al., 2010), situated off the east coast of Oman, overlaps with the highest number of analysed weapons from Byblos and Tell Arqa (Fig. 8). It includes weapons 6273, 10268, 10279, 10977, 10978, 10982, 10997, 11856, 11913, 26759, 27855, 27856, 27884 and 27902. The overall signature of other Oman ore deposits from Bidbid, Nizwa, Samad and Suhar (Begemann et al., 2010) correlates also with weapons 6273, 10268, 10279, 11913, 26759, 27855, 27856 and 27902 on the $^{208}Pb/^{206}Pb$ vs. $^{207}Pb/^{206}Pb$ diagram. According to archaeological evidence, Oman actively supplied Mesopotamia with this copper until 1750 BC (Weisgerber, 2007).

The arsenic and nickel compositions in the Oman ores are correlated and generally display an As/Ni ratio ranging between 0.1 and 10. Based on this correlation Begemann et al. (2010) considered that artefacts from Mesopotamia, Bahrain and Oman presenting less than 0.1 wt% nickel and an As/Ni ratio higher than 10 are not made of Omani copper. Following this reasoning, duckbill axe 6273 could be considered as compatible with these ores (Fig. 9). Spearheads 10279, 10978, 10997 contain respectively 2.5, 0.3 and 0.6 wt% As while the amount of nickel is at or below 0.02 wt% (detection limit for Ni). If we suppose that the amount of nickel in these 3 weapons is at 0.02 wt% then the As/Ni ratio are 125, 15 and 30 (Fig. 9) which casts doubt on the chemical compatibility of these objects with the Omani ores. The same could also be said concerning large spearhead 27856 which displays an As/Ni ratio of 20 (As 0.2 wt%, Detection limit for Ni is at 0.01 wt%).

Hauptmann remarks (2007; 204–7), in his work on the partitioning of trace elements between metal and slag in the case of the copper ore exploitation at Faynan, that indeed As and Ni are likely to enrich the metal more than the slag regardless of the firing condition during smelting. It seems, however, that the partitioning coefficient $D_{Cu/S}$ of arsenic is higher than that of nickel (Hauptmann, 2007; Fig. 6.37). According to Hauptmann:

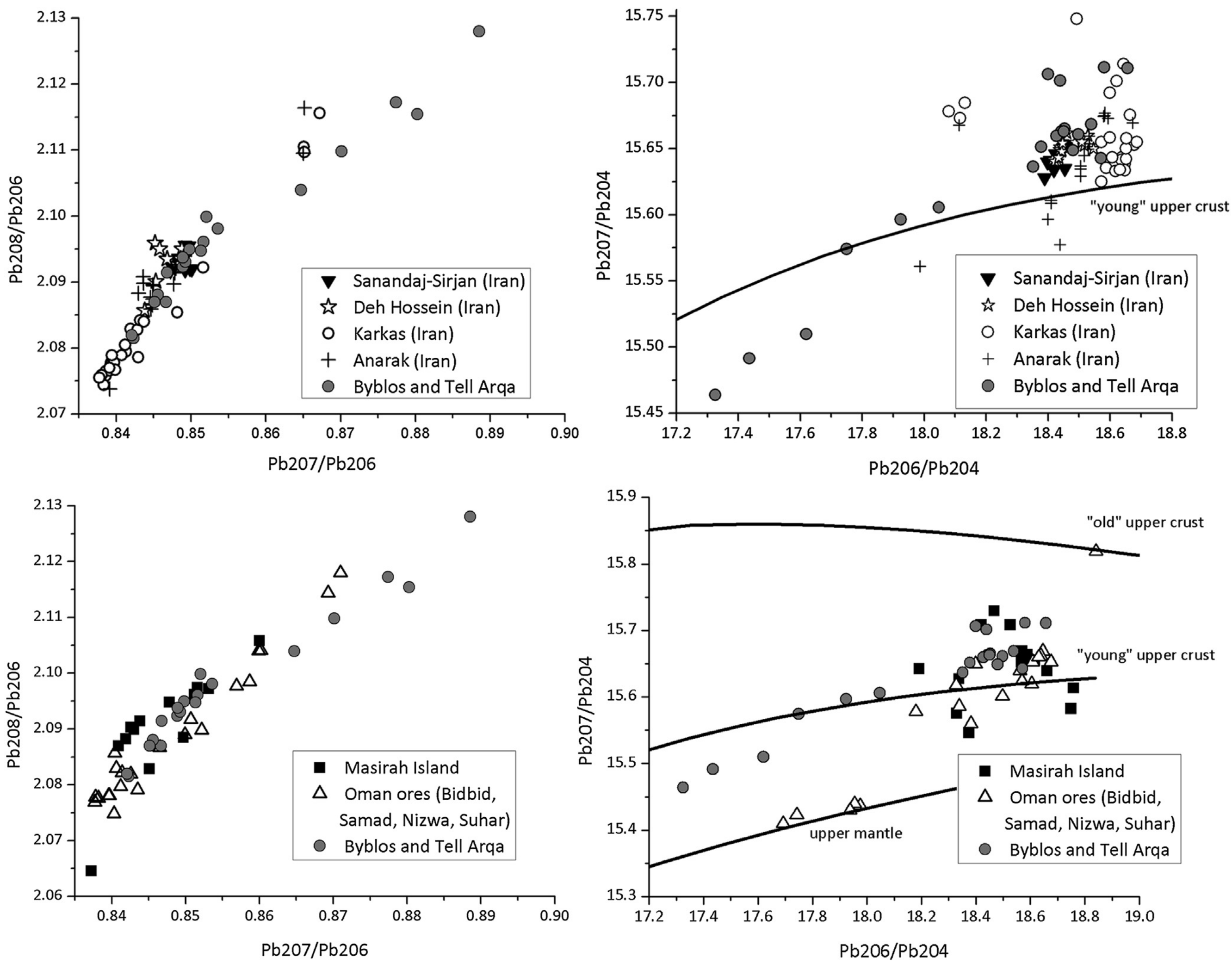

Fig. 8. Lead isotope ratios of the weapons of Byblos and Tell Arqa plotted on the $^{208}Pb/^{206}Pb$ vs. $^{207}Pb/^{206}Pb$ and $^{207}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ with the Pb–Zn ore field signature of Sanandaj–Sirjan zone and the copper ore field signatures of Deh-Hossein, Karkas and Anārak in Iran as well as the copper ore field signature of Masirah Island, Bidbid, Nizwa, Samad and Suhar in Oman.

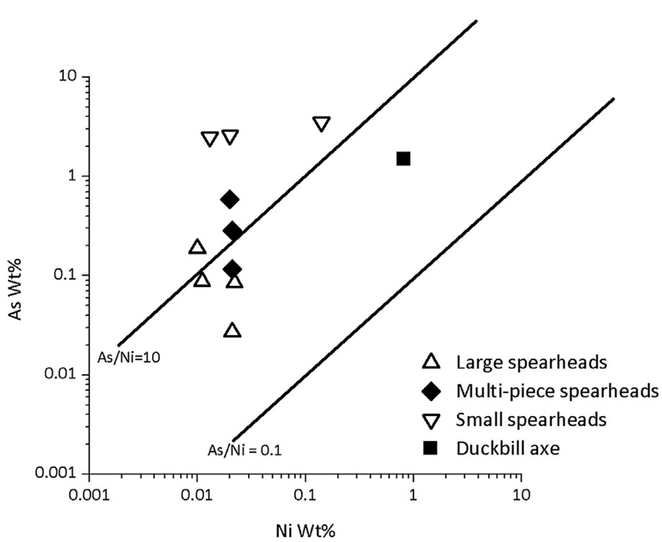

Fig. 9. Plot showing the As/Ni weight percentage ratios of some of the Byblos weapons analysed by PIXE.

$$D_{Cu/S} = (\%\ M\ \text{in copper})/(\%\ M\ \text{in slag})$$

M stands for the impurities such as Zn, Fe, Pb, As, Ni, Co.

This shows that arsenic will generally concentrate in the metal more than nickel and thus it should be expected that the As/Ni ratio of the Omani ores is slightly lower than their copper product.

The majority of the analysed metal objects dated to the Umm an-Nar (*c.* 2600–2000 BC) and Wadi Suq (*c.* 2000–1250 BC) period from Oman and Bahrain (Begemann et al., 2010; Prange, 2001) display Ni composition varying between 0.01 and 1%. Taking the limit of detection of the PIXE analyses into account, it is clear that the Ni composition of most of the Byblos and Tell Arqa weapons lies, at most, at the low end of this range, if not lower. Once again, these evidences, even though not conclusive, argue towards discarding the Oman ores as a compatible copper source for several of the Byblos and Arqa objects (10977, 10978, 10997, 11856, 27855, 27856 and 27884).

Weapons 10274, 10280, 10301, 11914, 11921, displaying $^{207}Pb/^{206}Pb$ ratios higher than 0.87, do not overlap with any known source. However, metal items presenting such high ratios, for example objects, ingots, prills and slags from Crete (Gale and Stos-Gale, 1986; Stos and Gale, 2006; www.oxalid.arch.ox.ac.uk), Egypt (Abdel-Motelib et al., 2012), Hebron in Palestine (Segal and Halicz,

2005) and Tell Abraq in United Arab Emirates (Weeks, 1999) are not uncommon in the Ancient Near East. These, however, cover a wide chronological (*c.* 3200–1500 BC) and geographical range and thus it is hazardous to use them as a definite indicator of inter-regional exchange with Byblos and Tell Arqa. The lead isotope ratios of ingots of the Barbar period from Al-Nasaria, Bahrein (Begemann et al., 2010; Prange, 2001), are interesting in this respect as they are consistent with the weapons from Byblos and Tell Arqa with $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ ratios respectively below 2.11 and 0.86. These weapons, however, are chemically different as they display Ni composition (<0.02 wt%) lower than the Bahrain ingots and hence a trade connection between the Levant and Bahrain cannot be established based on this particular set of evidence.

Spearhead 10277 display distinctively low isotope ratios on the $^{208}Pb/^{206}Pb$ vs. $^{207}Pb/^{206}Pb$ diagram which might be compatible with ores form southern Sinai (Abdel-Motelib et al., 2012; Hauptmann et al., 1999, 2012; Pfeiffer, 2013 cited from Abdel-Motelib et al., 2012). According to Abdel-Motelib et al. (2012), these ores display generally low amounts of arsenic (<1 wt%) which is not the case with 10277 which contains around 10 wt% As. This might indicate a chemical incompatibility or might reflect an intentional alloying of copper with arsenic by using "imported" products such as speiss (Rehren et al., 2012; Thornton et al., 2009).

These results highlight two possible copper exchange networks: the first one is a short distance network where cities of the Levant were supplied with Faynan copper. The lack of archaeological evidence of a massive MB I exploitation of its outcrops leads us to discard this option for the time being. The second is a long distance network where Levant cities received, via Mesopotamia, copper coming from Iran and possibly Oman. On one hand, exchange activities between Mesopotamia and these regions are established for late third and early second millennium BC by archaeological materials and textual sources (Begemann et al., 2010; Begemann and Schmitt-Strecker, 2009; Potts, 1993a, 1993b; Weisgerber, 1981, 2007). On the other hand Syrian and Mesopotamian cultural influence in the Levant is evident from the adoption of some pottery and weapon types, architectural elements and burial practices (Gernez, 2007; Ilan, 1995a, 1995b; Maeir, 2000; Philip, 1989; Philip et al., 2003). Occasional evidence of long distance contact between Byblos and lower Mesopotamia is also attested from an Ur III (*c.* 2112–2004 BC) cuneiform tablet from Derhem, near Nippur, mentioning an "Ensi" of Byblos (Genz, 2010; Saghieh, 1983; 131; Sollerberg, 1959–60).

As for the modalities of copper exchange and a circulation network several possibilities exist. Copper could have travelled through one or several Mesopotamian cities such as Mari and Ešnuna (Joannès, 1991; Michel, 1996, 2008), on the Middle Euphrates, before being sent to the Levant. Textual sources from Mari have already revealed that tin, silver and gold in the form of ingots and finished items were sent to cities such as Hazor (Bonechi, 1992; Malamat, 1960, 1970) in northern Canaan, Qatna (Arkhipov, 2012; 321) and Ugarit (Arkhipov, 2012; 365, 366, 369) in western Syria. These texts however only mention what has been sent by the king of Mari and do not necessarily represent the whole exchange and trade activity. The possibility of copper being exchanged by merchants, as in the case of the Anatolian Karum (Michel, 2008), should not be discarded. Nomadic or semi-nomadic populations moving along the desert margins of the Fertile Crescent could also have contributed to copper circulation. Their interaction with the sedentary populations is considered to have facilitated the spread of several elements of the same material culture in the Levant, inland Syria and Mesopotamia (Dever, 1980; Gernez, 2007; 651–652; Ilan, 1995a). This might also explain why the socketed spearheads first appear (*c.* 2100–2000 BC) in two distinct regions which are the Levant and in Oman (see Gernez, 2007; 652).

6. Conclusion

The various methods applied to the copper alloy weapons of Byblos and Tell Arqa revealed in at least one case a clear relationship between weapon type, alloy recipe, manufacturing techniques and "clean" copper. The craftsmen of Byblos thus differentiated at least between two copper qualities: the "dirty" copper used in most of the weapons and the "clean" used in a limited type of objects presenting technical difficulties during their manufacturing process. These results highlight the sensitivity of the craftsmen to the behaviour of their materials and their ability to find solutions to technical difficulties in order to meet the "customer's" expectations. These results might explain the information reported from the textual sources and provide new explanations to the nature of copper qualities. Demonstrating the selective use of several copper qualities adds important data to the growing repertoire of know-how of MB I Levant craftsmen. It also shows that Middle Bronze Age metallurgy was technologically rich and complex and that many exciting aspects of this craft are yet to be unravelled.

The copper provenance data place this work in an inter-regional context. It provides, for the first time, hard evidence showing the association of Byblos, historically known for its political, commercial and cultural ties to Egypt, and Tell Arqa to a wide eastern exchange network extending to Iran and possibly to Oman. Most of the possibilities explaining the modalities of these exchanges point to the role of Mesopotamia and Inland Syria as areas of "transit" or redistribution. Archaeology has already demonstrated a Syro-Mesopotamian cultural influence on the Levant during the MB I, our results however reveal direct or indirect trade relations between Byblos and these two regions. The object of this trade being of strategic importance, it is now crucial to start reconsidering the political ties of Byblos and Tell Arqa with their eastern counterparts and to reassess the influence of Egypt on Byblos under a new light at least for the beginning of the second millennium BC. For the time being the limited number of samples and sites studied could not entirely lead to tracing the ancient exchange networks of MB I. Evidently, recycling practices and the overlapping isotopic fields of many Near Eastern ancient ore deposits imposes caution when interpreting the data. This work, however, has demonstrated the potentiality of using multi-disciplinary approaches to answer highly complicated and important Levantine and Near Eastern archaeological issues. From this perspective, it is highly recommended to continue this work on a larger geographical and chronological scale.

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