LEAD ISOTOPES REVEAL THE ORIGIN OF MIDDLE BRONZE AGE ARTEFACTS FOUND IN SIDON (BURIAL 42)

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ALAIN VÉRON
ANDRÉ POIRIER
GAËL LE ROUX

Introduction

Since 1998 extensive excavations in Sidon by the British Museum in collaboration with the Department of Antiquities of Lebanon have revealed the extent of the city's growth from as early as the third millennium BC (Moran, 1987) right up to the Persian period (Doumet-Serhal, 2003, 2007). Furthermore these investigations have highlighted the economic and cultural role of Sidon and its importance within the Eastern Mediterranean during the Middle Bronze Age (MBIIA/B-C, 2000-1550BC). Sidon, along with the cities of Tyre and Byblos (Dunand, 1963; Jidejian, 1969, 1971; Elayi, 1989), is considered to be one of the oldest settlements of ancient Phoenicia (Herodotus, *History* I: 1). To date, ninety-two graves have been found in Sidon some of which contained numerous bronze artefacts (weapons and jewels) (Doumet-Serhal, 2007; Doumet-Serhal and Griffiths, 2007).

Bronze Age archaeological excavations invariably produced more pottery artefacts than bronze. Nonetheless, these bronzes constitute a sample of widespread material available at the time thus allowing the investigation of the early exploitation in the Mediterranean basin of metals such as copper (Cu), tin (Sn) deposits used for bronze alloys as well as silver (Ag) ores (Gale and stos-Gale, 1982). Geological investigation on the origin of these metal deposits has brought significant insights on metal trades during the Bronze Age and helped reconcile textual and archaeological findings (Muhly, 1973, 1993; Stech and Pigott, 1986). Meanwhile, concentration patterns of minor and major elements can be misleading when searching for the geographical origin of these deposits owing to ore heterogeneities as well as changes induced by smelting processes (Griffitts *et al.*, 1972; Craddock, 1976; Catling and Jones, 1977; Muhly, 1977). For the sake of accuracy, geochemical tracers ought to clearly and distinctly imprint the geographic origin of ore bodies and remain unchanged through smelting processes and metal corrosion. Lead (Pb) isotopes display such characteristics and have proven their usefulness to evidence the geographical origin of the raw material used to manufacture artefacts during the Bronze Age (see ref in Brill and Wrampler, 1965; Patterson, 1971; Gale and Stos-Gale, 1982; Gale, 2001; Véron *et al.*, 2004). Indeed, Pb is commonly found in Cu, Sn and Ag deposits that can easily be identified owing to their relative proportion of stable Pb isotopes (^{204}Pb, ^{206}Pb, ^{207}Pb, ^{208}Pb) of which the three latest are end-members of the uranium (U)-thorium (Th) decay chains. Depending on their initial U, Th content and their age, ore bodies will display specific Pb isotope imprints (Doe, 1970) that are not affected by subsequent chemical and biological fractionation processes.

Here we propose to geochemically evidence the origin of metal alloys used for the manufacturing of bronze artefacts found in Sidon's grave 42. This burial was discovered in 2005 and constitutes the first constructed grave in which more than one individual was found (Doumet-Serhal and Griffiths, 2007). Preliminary investigations date this burial to the MBII A/B period (around 2100-1600 BC). Bronze artefacts recovered from burial 42 comprised daggers, decorative discs which were part of a belt and a javelin head of which further description can be found in Doumet-Serhal and Griffiths (2007). In order to improve their preservation these objects were treated by Isabelle Skaf with mechanical and chemical procedures involving scalpel scrapping, brushing (soft corrosion) and the use of a corrosion inhibitor and the stabilizer Benzotriazole (BTA). A layer of Incralac lacquer 20% in toluene was also applied over the entire surface of the object. In order to remove every possible contamination due to such chemical treatments, samples were partly dissolved in a concentrated mixture of HCl and HNO_3 acids, before rinsing with MilliQ water. Cleaned samples were then oxidized with concentrated HCl-HNO_3 before purification on an AG1X8 anionic exchange column (Mahnès *et al.*, 1978). The ratios of Pb isotopes were measured by MultiCollector-Inductively Coupled Plasma Mass Spectrometry (MC-ICPMS) at GEOTOP (ISOPROBE, UQAM, Montréal). Calibration and mass fractionation were determined from concurrent analyses of thallium and the SRM981 NIST standard.

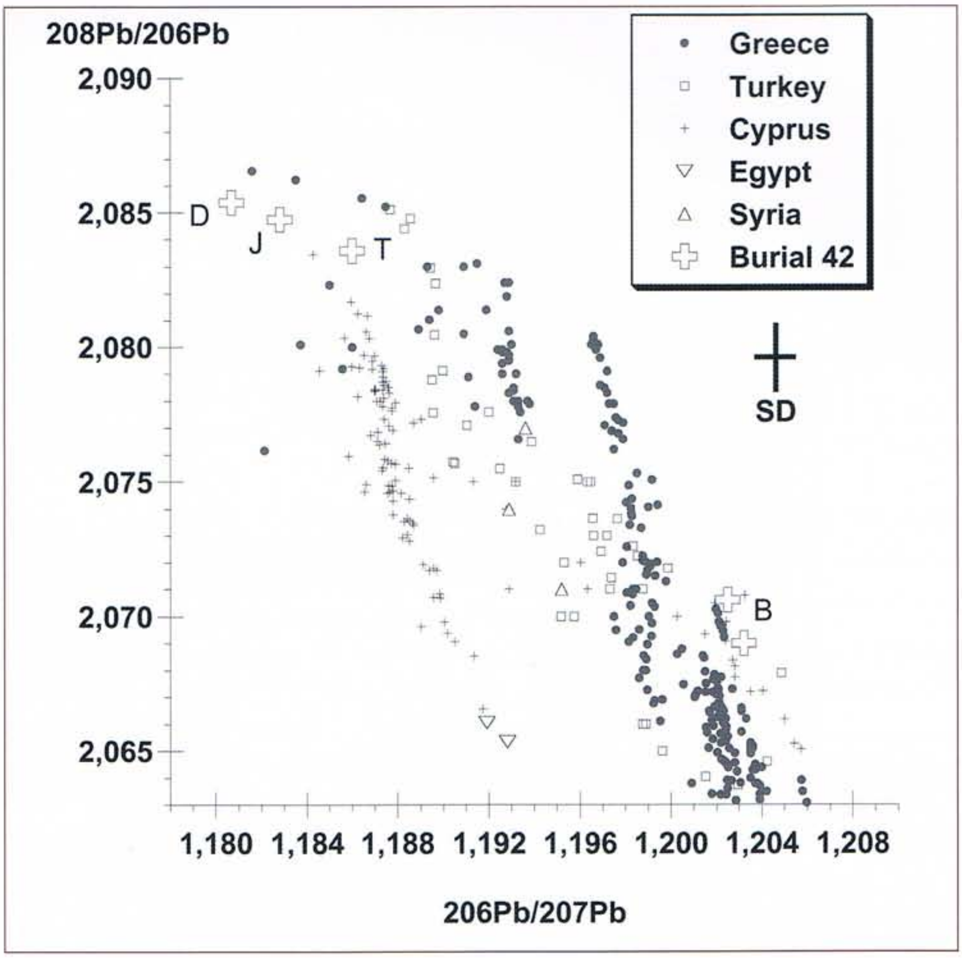
Lead isotope systematics

Bronze artefact	Ref	$^{206}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$	$^{208}Pb/^{206}Pb$	$^{206}Pb/^{207}Pb$
Torque	3580	18,580	15,666	38,714	2,0836	1,1860
Dagger	3541	18,520	15,686	38,622	2,0854	1,1807
Javelin head	3544	18,559	15,691	38,691	2,0847	1,1828
Belt disk	3617	18,890	15,700	39,083	2,0690	1,2032
Belt disk	3619	18,874	15,696	39,081	2,0706	1,2025

Table 1: Lead isotope imprints of bronze artefacts from burial 42. Standard deviation for lead ratios are 0,01% and 0,05% for $^{208}Pb/^{206}Pb$, $^{206}Pb/^{207}Pb$ and $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, $^{208}Pb/^{204}Pb$.

Lead isotope ratios determined for the burial 42 artefacts are reported in table 1. The ratio of ^{206}Pb to ^{207}Pb are used in the discussion as they are the most accurate ratios and have been used extensively in previous studies for ore characterization (see ref. in Gale and Stos-Gale, 1982; Yener *et al.*, 1991; Sayre *et al.*, 1992; Stos-Gale *et al.*, 1997; Weeks, 1999). Differences as low as 0,05% in $^{206}Pb/^{207}Pb$ ratios generally allow the distinguishing of ores from distinct origins. From table 1, it clearly appears that the two belt discs have a significantly different imprint (mean $^{206}Pb/^{207}Pb$= 1,203) than the three other bronze artefacts ($^{206}Pb/^{207}Pb$= 1,180 to 1,186). In order to restrict the geographical origin of the raw material used to fabricate these objects, we compared their isotopic ratios to those of well-known body ores from eastern Mediterranean geological deposits from Greece, Turkey, Cyprus, Egypt and Syria (fig. 1). Ore body isotope imprints that significantly fell off the range of our artefact signatures are not reported (Jordan, Iran, Iraq, Sardinia, Italy, Spain). As expected from table 1, belt discs and other artefacts do not show the same provenance in figure 1 as

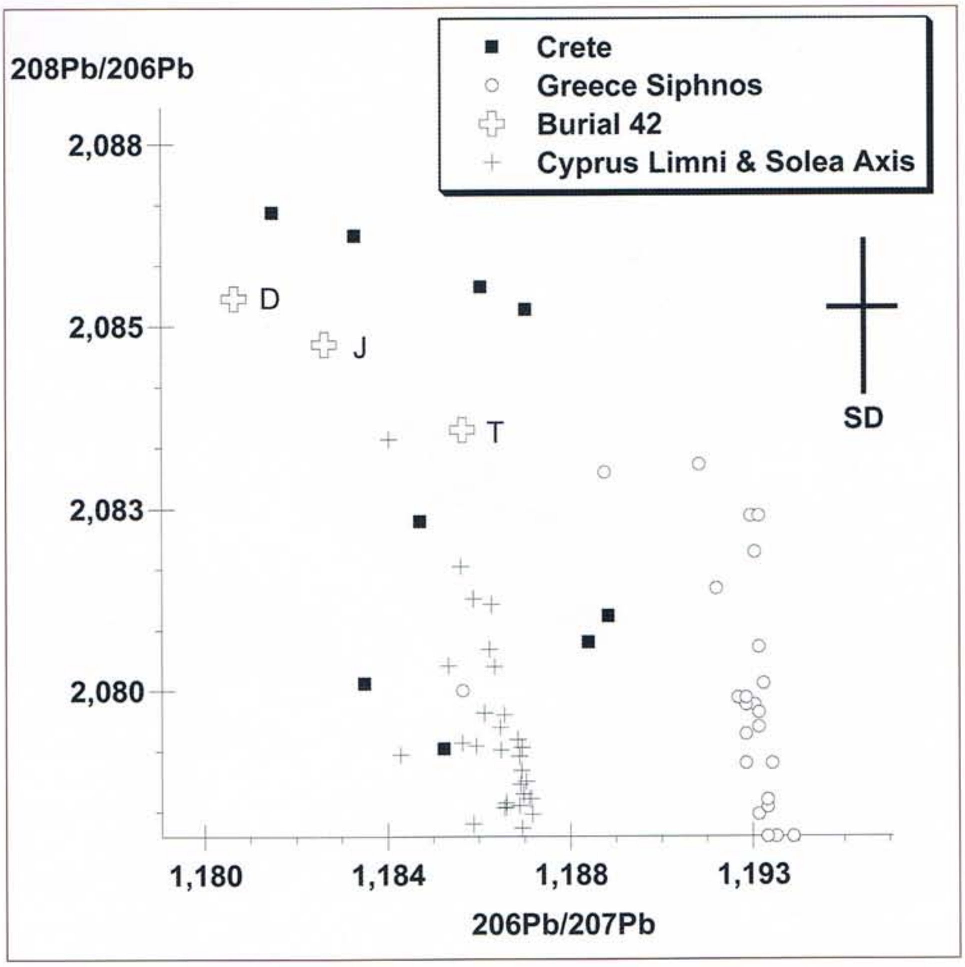
1 Comparison of isotopic imprint (208Pb/206Pb vs. 206Pb/207Pb ratios) of (B42) burial bronze artefacts to those of Eastern Mediterranean ore deposits (data from Gale *et al.*, 1981, 1988; Sayre *et al.*, 1992; Seeliger *et al.*, 1985; Stos-Gale and Gale, 1981, 1992; Stos-Gale *et al.*, 1981, 1986, 1996, 1997; Vavelidis *et al.*, 1985; Wagner *et al.*, 1985; Yener *et al.*, 1993). SD denotes the maximum Standard Deviation from reported published data. Specific annotations identify burial 42 artefacts: T (Torque), D (Dagger), J (Javelin head), B (Belt discs).



they display signatures that are consistent with Greek and/or Cypriot metal sources. While the occurrence of Turkish ores has often been evidenced as a source for bronze, Ag and Pb artefacts during the Bronze Age (Stos-Gale *et al.*, 1984; Gale *et al.*, 1985; Wagner *et al.*, 1985; Gale and Stos-Gale, 1992; Sayre *et al.*, 1992; Yener *et al.*, 1993), none of the burial 42 objects appear to display such geographical origin (fig. 1). This is in contradiction with previous findings that showed an Anatolian origin (Central Taurus Mountain) for Ag jewels found at Sidon in burial 27 (Véron *et al.*, 2004). A few outliers from the Aladag region at the edge of the Taurus isotope field could reconcile the origin of the metal used for the burial 42 torque in the absence of any other possible source (fig. 1). Meanwhile, as seen in figure 1, these outliers do not overlap at all with any of the other bronze artefacts carrying similar isotope imprints (i.e. dagger and javelin head) while other regional ore bodies from Cyprus and/or Greece do.

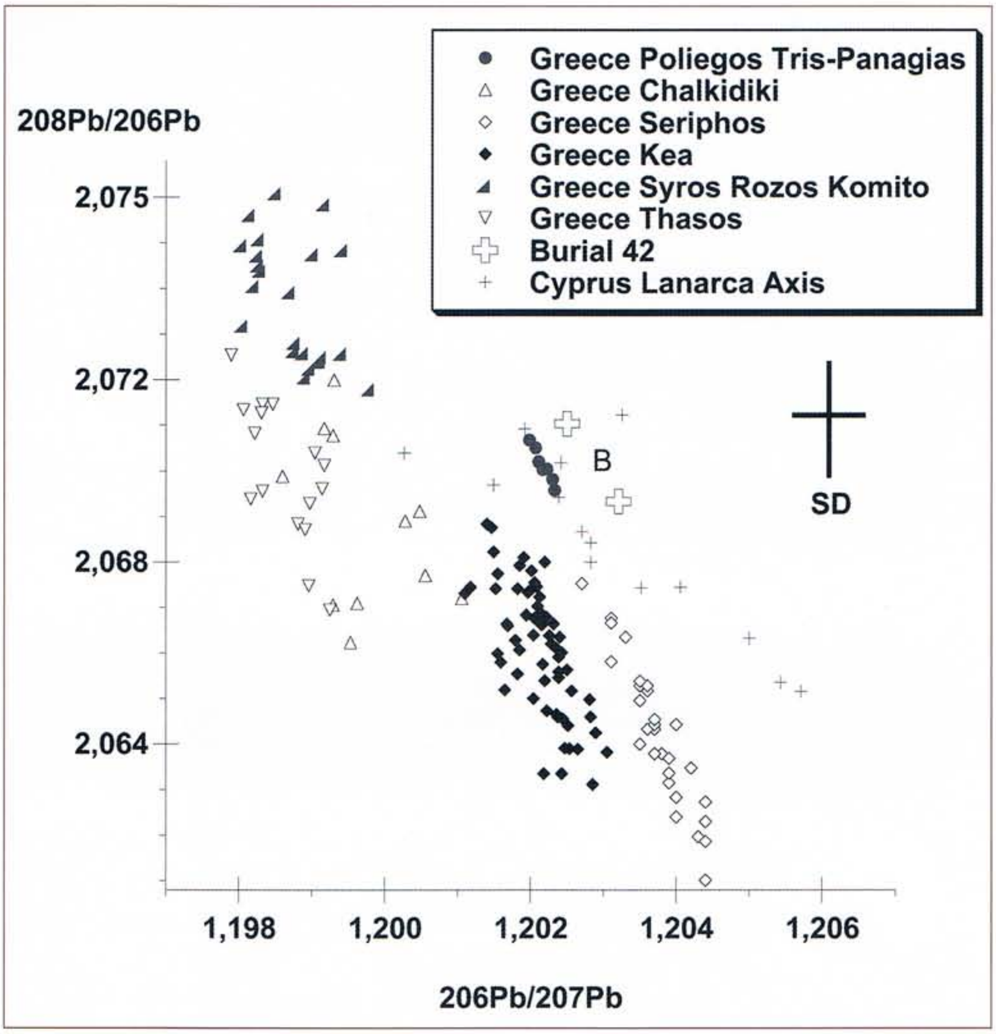
Based on these findings, more details on the provenance of metals could be assessed with a thorough investigation of regional Greek and Cypriote ore isotopic imprints (fig. 2, 3). The origin of the metal used to manufacture burial 42 weapons (javelin head and dagger) can be determined by invoking Cu ores from Crete (Gale and Stos-Gale, 1986) (fig. 2). Matching isotope imprints between this isotope field and burial 42 weapons suggest trading activities between Sidon and the Minoan civilization that flourished between 2700 and 1450 BC. This does not come as a surprise since it is well known that during the second millennium the Minoans were actively involved in the development of Sulphide metallurgy along

2 Blow up of fig. 1 with detailed isotopic imprints of regional ore deposits from Greece and Cyprus that might conform to Torque, Dagger, and Javelin head artefact signatures (see references in fig. 1).



with the Anatolians, the Cypriotes, and later, the Mycenaeans (Muhly, 1972; Muhly, 1973; Wertime, 1973). As previously mentioned, the sources of the metal's used for the torque (noted "T" in fig. 1 and 2) are not as clear. While a Turkish origin could reasonably be disregarded, the Cypriote isotope field slightly overlaps that of Cretan ores with an outlier from Solea Axis (Chalcopyrite, Stos-Gale *et al.*, 1997) (fig. 2). This would be consistent with the intermittent mining activity that has been revealed in Cyprus since the Early Bronze Age (Wertime, 1973; Koucky, 1982). Meanwhile the exact role of Cyprus in the Bronze Age metal trade is still in dispute (Stos-Gale *et al.*, 1986, 1997; Muhly *et al.*, 1988) and such findings would need to be confirmed in other artefacts. It is at this juncture that the question of metal recycling and possible multiple sources for an artefact owing to re-melting and mixing of metals could be raised (Budd *et al.*, 1995). Indeed the torque found in burial 42 could originate from ingots produced in Cyprus and re-melted in Crete for example. Low content of Sn and Pb in artefacts often precludes the addition of bronze scraps to ingots (Gale and Stos-Gale, 1982; Stos-Gale *et al.*,1997). The trace metal content of Sidon's bronze artefacts are generally below 1% (Le Roux *et al*, 2004, 2009) suggesting that not only re-melting is unlikely, but also that Pb from these artefacts derives from the original ore as a minor impurity, and therefore has not been added to improve the fluidity of the molten bronze. In the torque's case, the question regarding metal mixing from various sources cannot be completely ruled out and might only be resolved by trace metal analyses of the artefact.

3 Blow up of fig. 1 with detailed isotopic imprints of regional ore deposits from Greece and Cyprus that might conform to Belt discs artefact signatures (see references in fig. 1).



When compared to regional mining districts (fig. 3), the belt disc isotope ratios fit ore signatures from the Greek Cyclades (Poliegos) and Cyprus (Lanarca Axis). Once again we face two possible geographical sources for ores used to manufacture these discs. The Poliegos imprint corresponds to galena (Stos-Gale *et al.*, 1996) while the Lanarca Axis deposits correspond to enriched Cu sulfide ores (Stos-Gale *et al.*, 1997) that are more likely to be used in bronze alloys. Furthermore there is no evidence of Cu deposits in Poliegos (Gale 1978; Gale and Stos-Gale, 1981; Stos-Gale *et al.*, 1996). Therefore it is reasonable to infer that the metals from the bronze belt discs found in burial 42 would originate from Cyprus rather than the Cyclades.

Isotope imprints of bronze artefacts found in Sidon (burial 42) suggest Greek and Cypriote origins of the alloys. Both weapons (dagger and javelin head) display very similar Pb isotope ratios that can only be explained by a Cretan source while the torque might also originate from Cyprus (trace metal content of the artefact could help resolve this uncertainty). Both bronze belt discs are much more likely to originate from Cypriote ore deposits. None of the burial 42 bronze artefacts display Anatolian signatures as found for the silver jewels of burial 27.

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