Journal of Archaeological Science 118 (2020) 105147

Contents lists available at ScienceDirect

Journal of Archaeological Science

journal homepage: http://www.elsevier.com/locate/jas

Combining chemical and lead isotope analyses with 3-D geometric–morphometric shape analysis: A methodological case study of socketed bronze arrowheads from the southern Levant

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A R T I C L E I N F O

Keywords:
Socketed bronze arrowheads
Typology
Chemistry
LIA
Geometric morphometric shape analysis
Statistics

A B S T R A C T

In this study, the shape of socketed bronze arrowheads is analysed and expressed as a series of mathematical trends which are then compared to chemical and lead isotope composition, as well as to the categorization of traditional non-computerized typology. It is shown that while traditional typology has statistical validation, additional important information can be gleaned from 3-D geometric morphometric shape analysis (3DGM), particularly when combined with material analyses. For example, arrowheads that are traditionally categorized as a single type demonstrate minute shape differences that correlate with the sites where they were found. This micro-variability, detected only through 3DGM, has potential cultural, chronological and regional implications. Most importantly, this pilot study shows that chemical and isotopic composition is correlated to a specific shape trend, revealed through computerized analysis, rather than to the traditional typological classification. This opens up new vistas for a more advanced analysis of archaeological finds.

1. Introduction

The analysis of copper-based arrowheads most often involves the creation of a morphological-based typology. Sometimes, chemical composition is reported and more rarely, lead isotope analysis, but the three, as far as we know, are never combined. The formulation of an objective set of criteria by which to classify arrowheads is an exceedingly complex task, particularly in light of the multitude of variations in form that may exist within any particular corpus (e.g., Cross and Milik, 1956). This process inherently entails some degree of subjectivity, as scholars must judge which features should carry the greatest diagnostic weight and what constitutes a reasonable division between any two proposed types among finds which may vary along a continuum. Likewise, scholars may be consciously or subconsciously influenced by attempts to infer intent on the part of the arrowheads' original manufacturers without means by which to validate such assumptions, since very little evidence exists regarding how ancient people themselves distinguished between types. Consequently, such arrowheads tend to be categorized quite differently by various scholars, each of whom emphasizes the characteristic that dominates to his or her eye (e.g., Stern, 1982: 154–156 versus Dugaw, 2017; Dugaw, Lipschits and Stiebel, forthcoming). Theoretically, computerized typology, which takes into account multiple variables, should be particularly well-suited to address these shortcomings. However, since arrowheads from the Bronze Age and onwards are made of copper-alloys or iron, the poor state of preservation of many finds presents a significant obstacle. Iron is particularly problematic, as it tends to swell. While copper-alloy arrowheads are sometimes found chipped and corroded, they often remain preserved well enough for computerised analysis.

In the present pilot study, in addition to the objective 3-D geometric morphometric shape analysis (henceforth 3DGM), we incorporate statistical analysis of the multiple shape trend variables, chemical analysis and lead isotopic measurements. This creates a synergetic effect, in which valuable information may be retrieved, taking into consideration the raw material used. Lead Isotope Analysis (henceforth LIA) is a method used for provenancing, that is based on the comparison of the

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https://doi.org/10.1016/j.jas.2020.105147
Received 1 September 2019; Received in revised form 1 April 2020; Accepted 9 April 2020
Available online 29 April 2020
0305-4403/© 2020 Published by Elsevier Ltd.

isotopic ratios of lead or lead-containing end products to lead isotopic ratios of potential parent ores (see below). In the case of socketed bronze arrowheads, lead may have been used to facilitate casting and improve penetration potential, the lead isotope ratios in some of the arrowheads thus point to the origin of the added lead rather than that of the copper.

1.1. Definition of socketed bronze arrowheads

Numerous socketed bronze arrowheads, often referred to as "Scythian", have been found throughout the Southern Levant. These arrowheads are characterized by having sockets with which to affix the arrowhead to the shaft of the arrow and exhibit either two blades, three blades, or a solid pyramidal point. They first appear in the southern Levant, in significant numbers, during the beginning of the sixth century BCE in destruction layers associated with Neo-Babylonian military activity (Dugaw, 2017; Dugaw, Lipschits and Stiebel, forthcoming). These finds are markedly distinct, in form and manufacture, from the local leaf-shaped iron arrowheads common at this time (Stern, 1982: 154–156). The use of copper-alloys made them amenable to mass-production via casting, which was not possible with the wrought iron arrowheads that had to be individually forged (Moorey, 1980: 65). Chemical composition has been reported for several socketed copper-alloy arrowheads, such as those from Tel Michal and Tell el Hesi (Bennett and Blakely, 1989:291; Lupu, 1989; Muhly and Muhly, 1989:271). All appear to be composed of tin-bronze with varying amounts of added lead.

Since the late nineteenth century, scholars have used the term "Scythian" to describe socketed bronze arrowheads due to their association with the nomadic people who inhabited the Eurasian steppe during the first millennium BCE, referred to as Skythai by the ancient Greeks (Schmidt, 1908: 183; Snodgrass, 1964: 149; Sulimirski and Taylor, 1991: 551–552). Such arrowheads have been found in substantial numbers among the grave goods of the pre-Scythian and Classical Scythian cultures (Diakonoff, 1985: 91; Hellmuth, 2014: 4, 8). Most scholars attribute the initial introduction of socketed copper-alloy arrowheads into the Near East to the Scythians or the Scythians and the Cimmerians who settled south of the Caucasus in the late eighth and early seventh centuries BCE (Derin and Muscarella, 2001: 197–203). Although the earliest forms of socketed bronze arrowheads to appear in the Southern Levant have parallels in the Eurasian Steppe and may have been initially associated with contingents of soldiers of steppe origin in the service of Mesopotamian armies, such arrowheads appear to have been widely adopted. Many subsequent forms found in the Southern Levant are almost certainly of Near Eastern origin with no parallels on the Eurasian Steppe (Dugaw, 2017; Dugaw, Lipschits and Stiebel, forthcoming). Consequently, the common term "Scythian" is somewhat of a misnomer where it appears in the academic literature as a generic descriptor for socketed copper-alloy arrowheads, since it is easily misunderstood as an ethnic attribution. As such, it will be avoided hereafter.

1.2. The find contexts of the arrowheads

The present study focused on combination analyses of 29 socketed

Table 1
Arrowheads included in this study.

	Site	Sample	Reg. No.	Locus	Phase	Remarks	Type	Length (mm)	Weight (g.)	Figure
1	Azekah	Azekah 1	31170	13/E1/284			I	50.6	6.9	2.1
2	Azekah	Azekah 2	40020	12/T1/102		Topsoil	I	45.2	6.9	2.2
3	Azekah	Azekah 3	70262	14/W3/253		Topsoil	I	36.4	4.3	2.3
4	Qeiyafa	Qe-20	3604	–		Unstratified, Area D, dump	III	25.4		3.11
5	Qeiyafa	Qe-21	C8397	C6096	III	Accumulation on floor Area C, Sq. FF8/9	II	30.6	4.4	3.5
6	Qeiyafa	Qe-22	C10683	C6651	III	Accumulation on floor Area C, Sq. MM18 (Building C100)	II	32.6	3.2	3.6
7	Qeiyafa	Qe-23	C10163	C6527	III	Accumulation on floor Area C, Sq. DD9	II	33.2	2.8	3.3
8	Qeiyafa	Qe-24	C8706	C6148	III	Accumulation on floor Area C, Sq. FF8/9	II	33.8	3.6	3.1
9	Qeiyafa	Qe-25	C9071	C6083	III	Accumulation on floor Area C, Sq. FF10	III	18.8	2.4	3.12
10	Qeiyafa	Qe-26	C8418	C6095	III	Accumulation on floor Area C, Sq. EE9	III	27.7	4.5	3.10
11	Qeiyafa	Qe-27	C8410	C6095	III	Accumulation on floor Area C, Sq. EE9	II	32.8	4.4	3.4
12	Qeiyafa	Qe-28	C8876	6188		Topsoil Area C, Sq. EE10	II	33.1	5.3	3.2
13	Qeiyafa	Qe-29	C10258	6571		Topsoil Area C, Sq. KK18 (Building C100)	II	31.0		3.9
14	Qeiyafa	Qe-30	C8409	C6095	III	Accumulation on floor Area C, Sq. EE9	II	33.8	3.3	3.7
15	Qeiyafa	Not sampled	8855	6184 6174	III	Accumulation on floor Area C, Sq. FF11	II	29.3	3.0	3.8
16	Ramat Raḥel	Not sampled	2573/60	10102			I	42.9	6.7	4.2
17	Ramat Raḥel	Not sampled	341	17064			II	30.5	3.8	4.11
18	Ramat Raḥel	RH-1	2619/60	10111	B2-3b	Fill IA-to Hellenistic	I		1.8	–
19	Ramat Raḥel	RH-2	3468/60	820	C1-5	Accumulation on floor	I	30 .6	2.6	4.13
20	Ramat Raḥel	RH-3	3116/60	322	C1-4	Fill IA-to Hellenistic	II	31.9	3.8	4.9
21	Ramat Raḥel	RH-4	3060/60	316	C1-3	Fill IA-to Hellenistic	II	30.3	3.7	4.12
22	Ramat Raḥel	RH-5	3117/60	322	C1-4	Fill IA-to Hellenistic	I	31.3	3.0	4.7
23	Ramat Raḥel	RH-6	8843/60	11014		Topsoil	I	35.8	3.5	4.5
24	Ramat Raḥel	RH-7	8782/60	43	C1-6	Garden soil	I	29.5	2.5	4.8
25	Ramat Raḥel	RH-8	5752/60	15074	D4-3	Fill below church	II	32.3	4.6	4.10
26	Ramat Raḥel	RH-9	8592/60	16		Fill IA-to Hellenistic	I	32.6	3.9	4.6
27	Ramat Raḥel	RH-11	8637/60	20	C1-3	Fill IA-to Hellenistic	I	43.5	6.2	4.1
28	Ramat Raḥel	RH-12	2209/60	10006	B2-1	Fill	I	40.6	5.8	4.4
29	Ramat Raḥel	RH-13	1456/60	12059		Topsoil	I	43.3	4.8	4.3

copper-alloy arrowheads from three sites in the southern Levant, specifically in the south of Israel: Ramat Rahel (n = 14), Khirbet Qeiyafa (n = 12) and Azekah (n = 3). For contextual details of the arrowheads, see Table 1. The location of the sites can be found in Fig. 1, and scanned images of the arrowheads are in Fig. 2–4.

1.2.1. *Khirbet Qeiyafa*

The site is located on top of a hill overlooking the Elah Valley. The major occupation phase here is Stratum IV, a fortified city from the Iron Age IIA, dated to the early 10th century BCE (Garfinkel et al., 2016). After some 700 years, in the Late Persian – Early Hellenistic era, Khirbet Qeiyafa was reoccupied (Stratum III, Freikman et al., 2014: 101–128; Garfinkel, in press). Based on numismatic data and the pottery, two phases were securely related within Stratum III (Farhi, 2016; Sandhaus and Kreimerman, 2015). The earlier phase is dated to the late fourth century BCE and the later phase is dated to the early third century BCE. Most of the socketed bronze arrowheads from the site originated in a basement of an abandoned building of Area C, which belong to the early phase, with no evidence of any military attack. This area was never re-occupied and none of the coins found in Area C date later than the mid-fourth century BCE.

1.2.2. *Tel Azekah*

Fragmented remains dating to the Iron IIB were found in most of the excavation areas, but this level was heavily damaged by building activities dating to the Persian and Early Hellenistic periods. A clear destruction layer from late eighth century BCE was found in two excavated areas (S1 and T1). After a settlement gap in the first half of the seventh century BCE, the settlement at the site was renewed, with a probable fortress that was built in the northern side of the site (Area N1). In the other excavated areas Iron IIC finds are very rare and are usually unearthed in later mixed fills, but the amount of rosette stamped handles discovered at the site are a clear indication for the importance of the site during the last decades of the First Temple period (Lipschits et al., 2017). Two of the arrowheads discussed in this paper were discovered in topsoil, and one was found in Area E1, in locus 204 (B. 77749) defined as a fill from the Hellenistic period.

1.2.3. *Ramat Raḥel*

The first building phase at the site should be dated to the late eighth–early seventh centuries BCE. A citadel with unique architectural features, such as volute capitals and window balustrade, was built on the top of the mound. Many jar handles with LMLK and "private" stamp impressions were assigned to this phase, representing all the known varieties dated from the late eighth to the mid seventh centuries BCE. In the second building phase, dated to the last quarter of the seventh century BCE, a new citadel was built, surrounded by a garden to its north, west and south. Many rosette-stamped handles were assigned to this phase. The citadel was not destroyed in the late Iron Age. Many lion, early YHWD and sixth-century "private" stamped handles, were assigned to the transitional stage between the second and third building phases, dated to the sixth century BCE. From the third building phase 365 YHWD stamp impressions of the Persian and Hellenistic periods were discovered, a clear indication that during the Persian and early Hellenistic periods the edifice at Ramat Raḥel continued to function as an administrative/governmental center, until it was destroyed in the second century BCE (Lipschits et al., 2011). Most of the arrowheads were discovered in fills and in the garden soil, dated to the second and third building phases.

2. Research approach and methods

The particular question that guided us throughout this study is methodological and focuses on whether a correlation between typology, chemical and isotopic compositions of the arrowheads in the sample can be discerned. The socketed bronze arrowheads included in this study were divided into three main types on the basis of traditional observational morphological typology (see details below). In order to validate this typological division and to facilitate a statistical analysis of the results, the arrowheads were scanned using a 3D scanner (see below). This was done in order to minimize as much as possible the subjectivity of the morpho-typological classification. The chemical and isotopic analyses were directed towards clarifying the possible correlation between shape and compositional variables, i.e., whether each type would be made of a different copper alloy or from copper or lead from different sources.

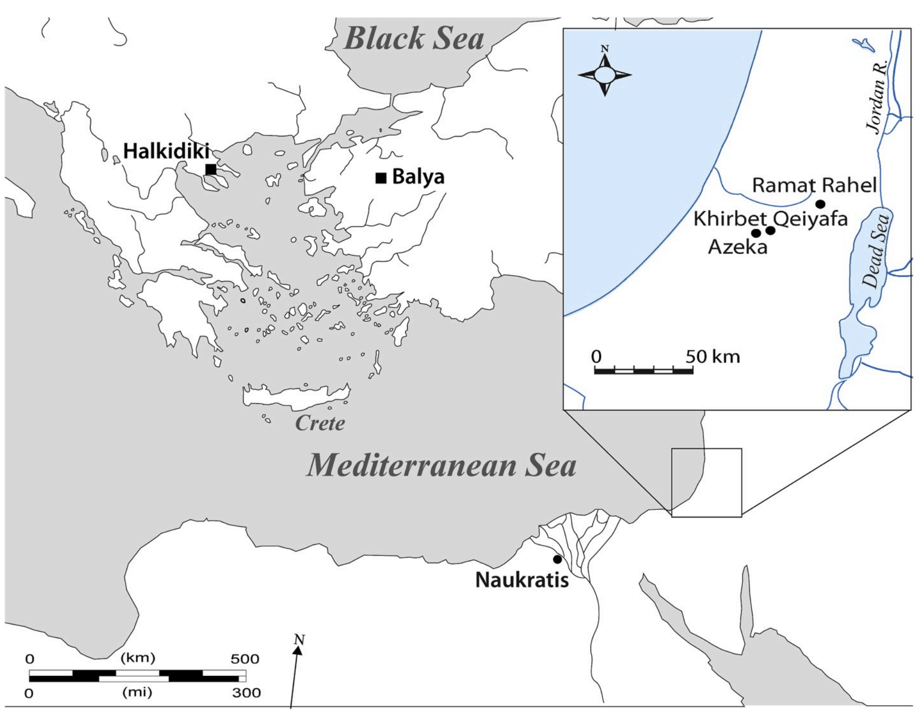

Fig. 1. Map showing the location of sites mentioned in the text.

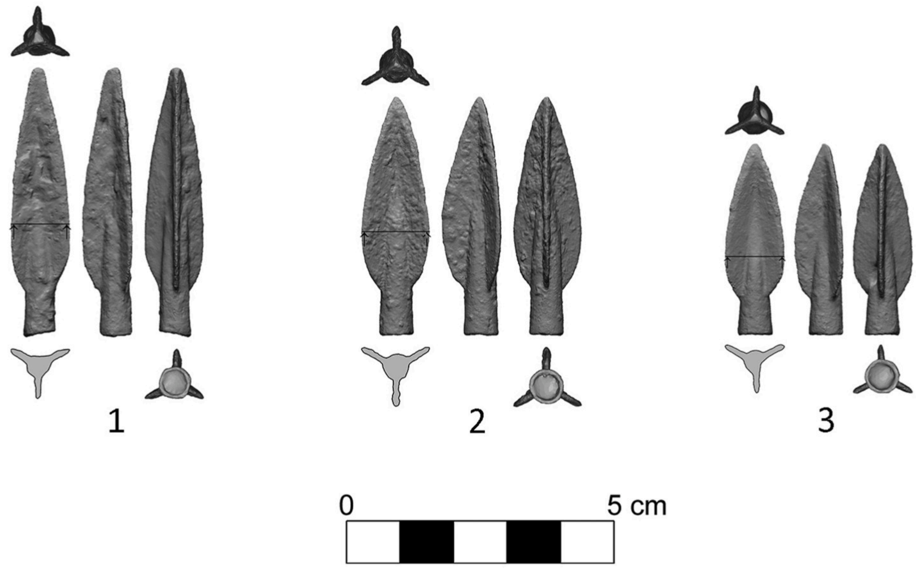

Fig. 2. Scanned images of arrowheads from Azekah (For identification of the arrowheads see data in Table 1).

2.1. Morphological classification

2.1.1. Traditional typology

Traditional arrowhead classification relies primarily upon visual examination of variations in morphology, such as the number of blades, blade shape, proportions, and the means by which the arrowhead was affixed to the arrow shaft. Specific features, such as the particular blade shape, are used as the criteria by which arrowheads are sorted into types, so that all arrowheads that share a common set of specific features are defined as a single type. Based on typological criteria defined by Dugaw (2017: 28–29; Dugaw, Lipschits and Stiebel, forthcoming) in his recent catalogue of socketed bronze arrowheads from the Southern Levant, the arrowheads of the present study may be separated into three types:

Type I: This arrowhead type has a leaf-shaped trilobate blade and long prominent socket. When viewed from the side (perpendicular to the axis), the blades form a shape reminiscent of a typical leaf – a vaguely ovoid shape coming to a point at the tip. When viewed from the tip, the three blades extend from the axis of the arrowhead approximately 120° apart. The socket is "prominent" in that it extends well behind the anterior edge of the blades. The type is analogous to Dugaw's Type T1 (2017: 38–41; Dugaw, Lipschits and Stiebel, forthcoming).

Multiple lines of evidence associate this type with Neo-Babylonian military activity from the end of the seventh century to the early sixth century BCE. One arrowhead of this type was found in Jerusaelm in a destruction layer dated to the 586 BCE Babylonian conquest (Shiloh, 1984: 19, Pl. 33:2) An additional three were found there in stratified fills amongst pottery dated to the late seventh to early sixth centuries BCE (Vejil and Mazar, 2015: 469–470, Fig. 11.1:1,13,17). Two were found at Tel Goren in a stratum dated from roughly 630 BCE until the site's destruction in circa 582 BCE, likely at the hands of Nebuchadnezzar II's army (Mazar, 1993: 404; Stern, 2007: 179, 181). At least one arrowhead of this type was found imbedded in a wall at Tel Goren, which Stern (2007: 179, 181) suggested can be assumed to have been set by the Babylonian archers during their conquest of Judean En-Gedi. Further evidence of Neo-Babylonian use of this arrowhead in the grater Near East is derived from late seventh century BCE Babylonian assaults on Nineveh (Pickworth, 2005: 310, Fig. 35), Nimrud (British Museum ND 4149), and Carchemish (British Museum 116207, 116208, 116209).

Type II: This type has a rhomboid-shaped trilobate blade and a flat or very slight socket. Being trilobate, its blade configuration appears similar to Type I when viewed directly from the tip, with three blades situated roughly equidistant around the axis of the arrowhead. The type is analogous to Cleuziou's Type F3 (1977: Fig. 1) and Dugaw's Type T2 (2017: 41–44; Dugaw, Lipschits and Stiebel, forthcoming).

Including those from the present study, Dugaw (2017: 41–44; Dugaw, Lipschits and Stiebel, forthcoming) was able to identify eighty-three clear examples of this type in the southern Levant. Well-provenanced arrowheads of this type date almost exclusively to the Persian and Hellenistic Periods, with a few outliers dated to the Roman Period. The type has clear Achaemenid associations. Great numbers of this type have been found at Pasargadae (Stronach, 1978: 218–219 Pl. 165: a,b), in the royal treasury at Persepolis (Schmidt, 1957: 97–99, Pl. 76:8), and at the sites of several major battles and sieges of the Greco-Persian wars (Dodwell, 1819: 160, Fig. 2; 44; Schumacher, 1890: 144 f, No. 748, Pl. XIV:23–28; Walters, 1899: No. 2806; Broneer, 1933: Fig. 13 a-c; 1935: 114–117, Fig. 4; Robinson, 1941: 378 ff, Pl. CXX; Snodgrass, 1964: 151–153; Cleuziou, 1977: 194–196, Fig. 1). No finds of this type predate the Persian Period.

Type III: This type is rhomboid-shaped with a solid pyramidal point. Its form is similar to the Type II trilobate, except that a solid plane exists between each of the three points situated 120° from each other in relation to the axis, resulting in the entire arrowhead forward of the widest point forming a solid pyramid shape. Behind the midpoint, the three corners of the base of the pyramid turn into three ridges that become progressively narrower as they approach the socket. The socket is flat or slightly prominent. This type is analogous to Dugaw's Type P1 (2017: 48–49; Dugaw, Lipschits and Stiebel, forthcoming). It has been suggested that the pyramidal blade shape evolved from rhomboid trilobate form as means to increase the arrowhead's armour penetrating potential (Petrie, 1928: 16; Snodgrass, 1964: 153). This is supported by the fact that the two types are often found together, implying that the difference between the two was functional, since the trilobate form would likely cause a more severe wound when employed against an unarmoured or lightly armoured target. This type first appears in the archaeological record somewhat after Type II (Dugaw, 2017: 49; Dugaw, Lipschits and Stiebel, forthcoming). It was not found in Persepolis and Pasargadae and appears to be a Near Eastern type. Among the current assemblage, this type was only found at Khirbet Qeiyafa.

2.1.2. Geometric morphometric shape analysis

Homologous landmarks-based 3DGM is one of a number of geometric morphometric methods designed to quantitatively and objectively describe and analyse the shapes and shape variability of physical

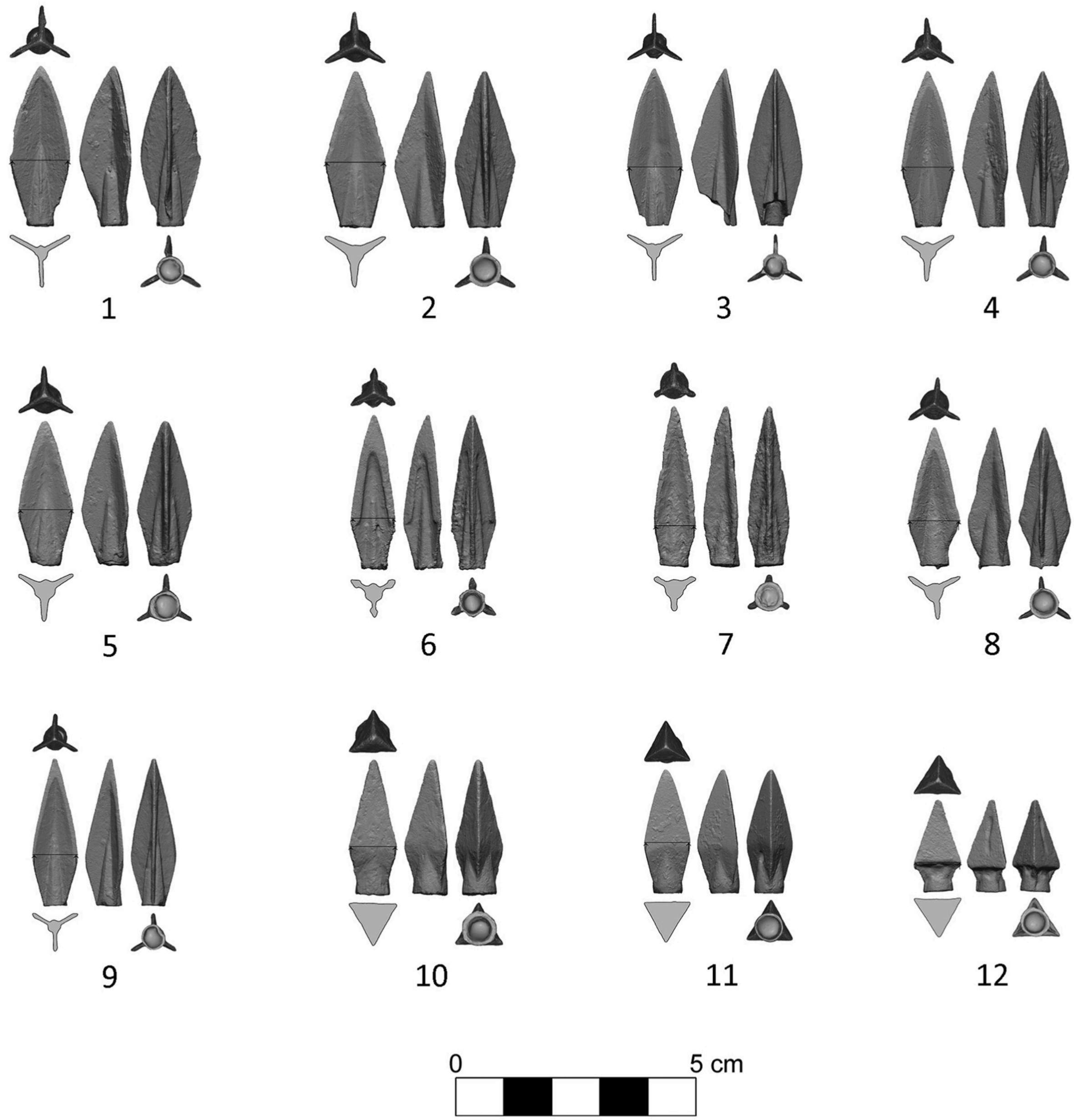

Fig. 3. Scanned images of arrowheads from Khirbet Qeiyafa (For identification of the arrowheads see data in Table 1).

objects (Dryden and Mardia, 1998). It is based on the positioning of a finite number of landmarks, each having three coordinates in Euclidean space, onto the surfaces of a set of physical objects or their 3D digital models. These landmarks must be homologous across all objects in the analysed sample, that is, each landmark should have a corresponding landmark on each object. In the case of manmade material culture, the landmarks homology stems from an explicit, objective and consistent positioning and orientation of all objects in space, as well as from consistent positioning of the landmarks onto the surfaces of the items (Lycett and Chauhan, 2010). The 3D landmarks' coordinates are subjected to a series of multivariate statistical procedures and analyses to quantitatively describe the shapes of the objects, the degree of shape variability and the trends which underlie it. These can then be used in statistical clustering procedures to facilitate a morphologically-based classification of objects into groups.

Of the 29 items included in the present study (Table 1), 27 copper-alloy arrowheads were chosen based on the preservation state for high resolution 3DGM shape analysis. Firstly, all artefacts were scanned using a high resolution structured-light scanner (produced by Polymetric™) in the Computational Archaeology Laboratory at the Institute of Archaeology, The Hebrew University of Jerusalem, in order to provide high resolution 3D closed mesh models. These models are publically available for download from an online repository (Yahalom-Mack et al., 2020). Subsequently, the models were automatically positioned to planform view by rotation along the X and Y axes following the polygons' normal distribution (Grosman et al., 2008) using the Artifact3-D software (Grosman, 2016). Next, the models have been automatically positioned by rotation along the Z axis so that the axis which maximizes the bilateral symmetry would be parallel to the Y axis (Herzlinger and Grosman, 2018). Lastly, 2500 three-dimensional homologous semi-landmarks have been positioned on the upper and lower faces of each model in a 50X50 deformed grid configuration, with the top and bottom latitudes sampling the proximal and distal edges. Thus, the morphology of each artefact was expressed in the analysis by 5000 three-dimensional landmarks. Landmarks positioning was performed using Artifact GeoMorph Toolbox 3D (AGMT3-D) 3.0 (Herzlinger and

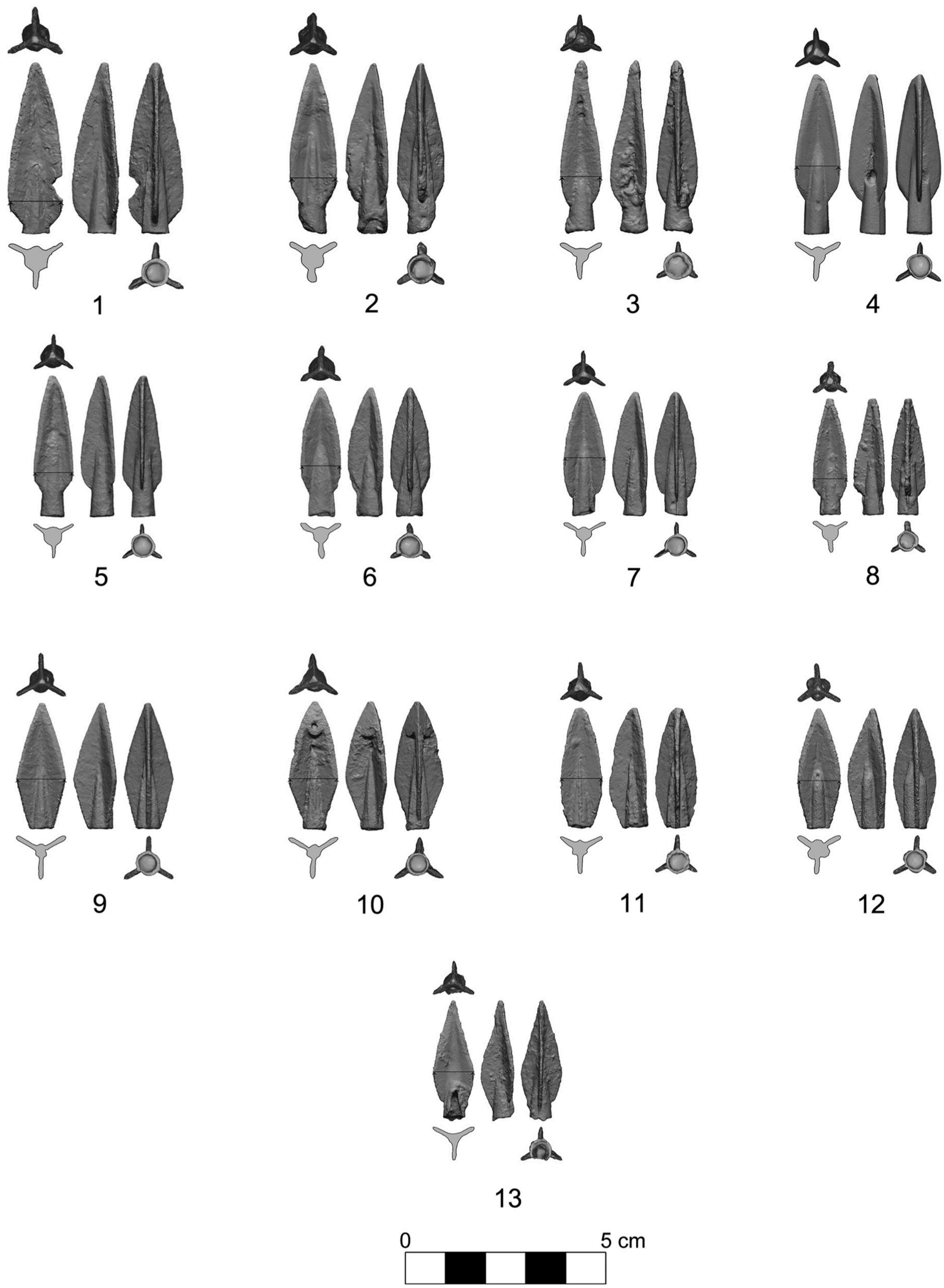

Fig. 4. Scanned images of arrowheads from Ramat Raḥel (For identification of the arrowheads see data in Table 1).

Grosman, 2018). The files containing the landmarks coordinates, and allowing the following shape analysis are publically available for download from an online repository (Yahalom-Mack et al., 2020).

Following the data acquisition stage, the combined dataset has been subjected to a geometric morphometric analytical procedure following common protocol (Dryden and Mardia, 1998), using AGMT3-D 3.0 (Herzlinger and Grosman, 2018) and SAS JMP 14.0. The procedure consisted of Generalized Procrustes Analysis (GPA), serving as a superimposition procedure, which removed non-shape related variability stemming from differences in location, rotation and scale. Next, the GPA modified coordinates were subjected to a principal component analysis (PCA). This analysis was used to reduce dimensionality and express shape differences in terms of principal component or shape trends, which are a series mutually perpendicular axes in multidimensional shape space, corresponding to co-variable groups of landmarks, sorted in descending order according to the proportion of shape variability they explain. Principal components scores were tested against various chemical, isotopic and archaeological variables to detect significant correlations, using linear discriminant analysis and standard least squares models.

2.2. Chemical composition and lead isotope analyses

Of the 29 arrowheads, 26 arrowheads were selected for chemical compositional analysis and twenty-five were subjected to lead isotope analysis (Table 1; Figs. 5–8). The latter is based on numerous observations that no significant isotope fractionation occurs during smelting and re-melting processes. The lead isotope ratios thus serve as a 'fingerprint' of the mineral ore deposits, which can be compared with the end-product (for discussion and bibliography, see Gale and Stos-Gale, 1982; Hauptmann, 2007: 31–38; Stos-Gale and Gale, 2009; Pernicka, 2014).

The morphology of the arrowheads, being hollow and relatively small, presented difficulty in obtaining a clean sample from the core of the object. The thickest area, in the junction between the shaft and the tip was drilled, usually from more than one direction. After surface drillings were discarded 10–20 mg of sample was collected. The sample obtained from a few of the arrowheads (RH1, RH 5, RH 9 and RH 11) was less than 10 mg (Table 1), and some of the corrosion must have been included, resulting in a relatively low sum of elements; between 60% and 80% in six samples (RH1, RH2, RH5, RH6, RH8 and RH9) and lower than 60% in two samples (RH4 and RH7) (see Table 1).

The drillings were dissolved using *Aqua Regia.* Metal concentrations

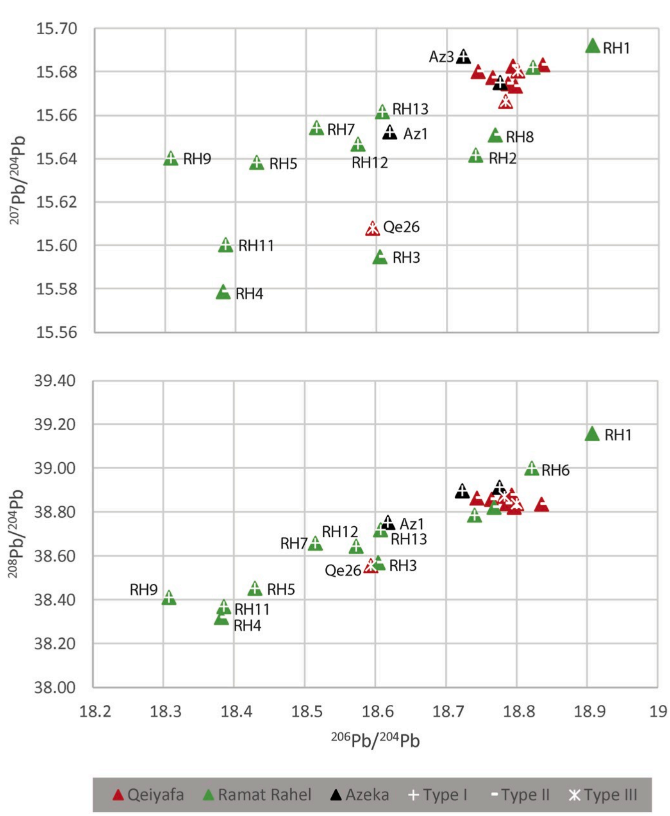

Fig. 5. Lead isotope ratios of arrowheads samples for this study, identified by type (see data in Table 2).

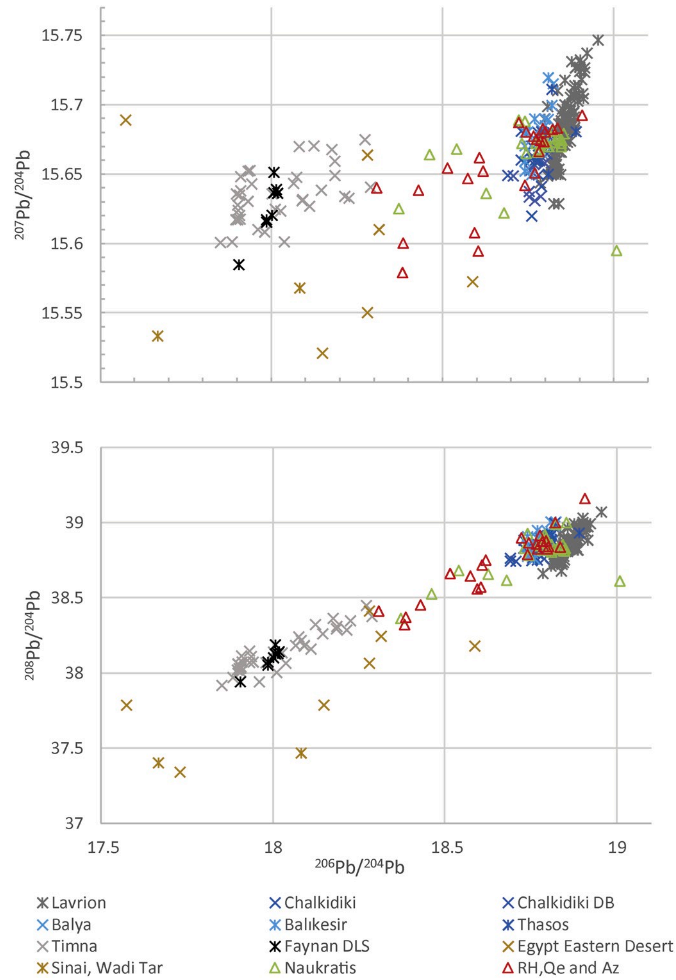

Fig. 6. Lead isotope ratios of arrowheads sampled for this study along artefacts from Late Period Naukratis (Masson-Berghoff et al., 2018), plotted against relevant lead ores from the Aegean and western Anatolia (Chalkias et al., 1988; Stos-Gale et al., 1996; Vavelidis et al., 1985; Wagner et al., 1985; Wagner et al., 1986; http://oxalid.arch.ox.ac.uk), as well as ores from the Arabah, Sinai and Egypt (Gale et al., 1990; Hauptmann et al., 1992; Hauptmann, 2007;Abdel--Mutelib et al., 2012).

were determined using a quadrupole Inductively Coupled Plasma – Mass Spectrometer (ICP-MS, Agilent 7500cx) in the Institute of Earth Sciences, The Hebrew University of Jerusalem. The ICP-MS was calibrated with a series of multi-element standard solutions (Merck; ME VI), standards of major elements and a blank. Drift was corrected with the help of internal standards (750 μg/L Sc, 100 μg/L Re and 50 μg/L Rh). Standard reference samples (US Geological Survey standard reference samples T-201 and T-209) were examined after calibration for accuracy assessment. Estimated precision of the major and trace elements are 3% and 5%, respectively. Major and trace elemental accuracy was <5% for all elements. Following the separation of lead in columns (Erel et al., 2006), lead isotopic ratios were measured using Neptune plus multi-collector ICP-MS in the Institute of Earth Sciences, The Hebrew University of Jerusalem. Thallium was used for mass-bias correction. SRM-981 standard was run with the samples yielding the following values ($n = 13$): $^{206}Pb/^{204}Pb = 16.931 \pm 0.002$, $^{207}Pb/^{204}Pb = 15.484 \pm 0.003$, $^{208}Pb/^{204}Pb = 36.675 \pm 0.009$.

3. Results

The significance of our results lies both in the data obtained from the analyses and, more so, in the correlations detected between the chemical

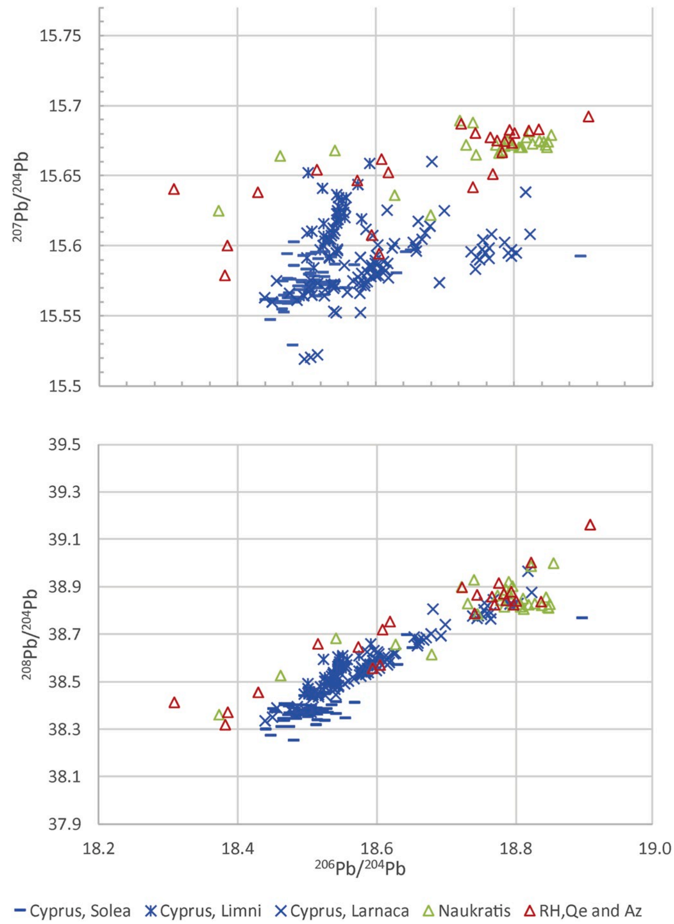

Fig. 7. Lead isotope ratios of arrowheads sampled for this study along artefacts from Late Period Naukratis (Masson-Berghoff et al. 2018), plotted against copper ores from Cyprus (http://oxalid.arch.ox.ac.uk).

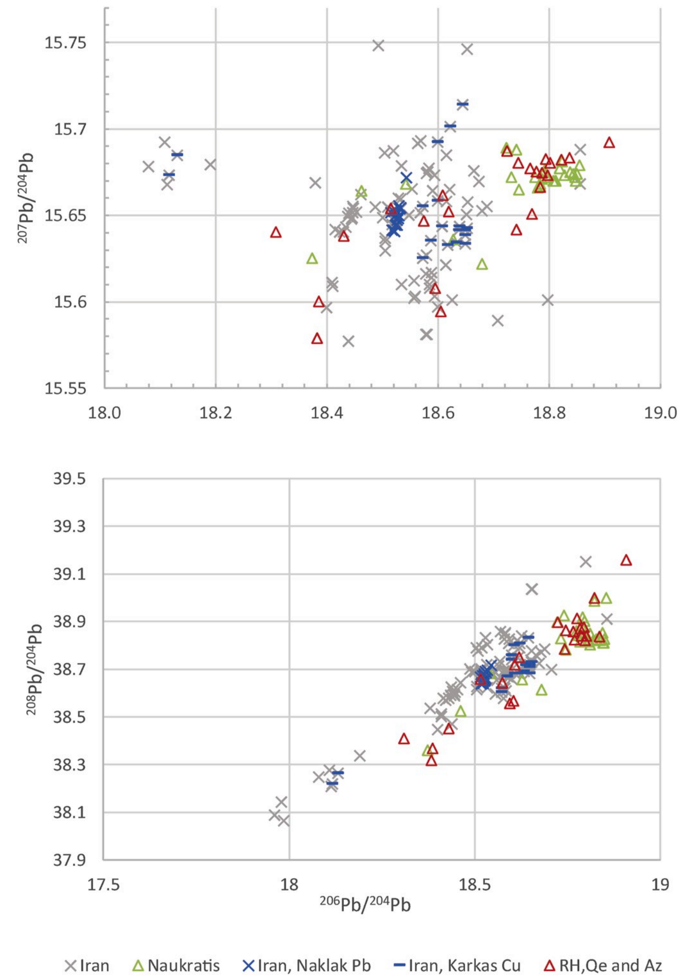

Fig. 8. Lead isotope ratios of arrowheads sampled for this study along artefacts from Late Period Naukratis (Masson-Berghoff et al., 2018), plotted against lead and copper ores from Iran (Shafiei, 2010; Nezafati et al., 2009; Pernicka et al., 2011).

and isotopic composition and the various shape trends of the analysed objects. These correlations enable to discern micro-variability, particularly on the level of the site (find place) vis-à-vis the type and composition. First, we present the results of the chemical and isotopic analyses and then proceed to examine these results in correlation with those obtained by the shape analysis.

3.1. Chemical composition

The results of the chemical analysis presented in Table 2 show that the majority of the arrowheads are indeed copper-based, mostly bronze with varying tin content. One arrowhead from Khirbet Qeiyafa (Qe-23) is made of arsenical copper (3.6% As) and four arrowheads from Ramat Raḥel with relatively low tin contents (RH-4, RH-6, RH-8, RH-9) contain antimony in a percentage level (1.6–9.6% Sb). Eight arrowheads, two from Khirbet Qeiyafa and six from Ramat Raḥel are relatively poor in lead (<0.7% Pb). Thirteen arrowheads (four from Khirbet Qeiyafa, three from Azekah, and six from Ramat Raḥel) contain c. 1–4% Pb and six (all from Khirbet Qeiyafa) contain c. 9–23% Pb. While the arrowheads in the latter group (9–23% Pb) likely contain artificially added lead, some of the arrowheads in the former group contain lead which may or may not be naturally associated with the copper. This has bearings on our ability to determine the source of the metal, as an artificial addition of lead would significantly alter the lead isotope ratios so that they indicate the source of the added lead rather than the source of the copper.

3.2. Lead isotopic composition

The majority of the arrowheads form a rather tight cluster (Table 2, Fig. 5). Since, as noted above, many of the arrowheads in this cluster contain added lead, the source of the lead rather than that of the copper is indicated. The results appear to be consistent with lead ores from western Turkey and Chalkidiki in northern Greece (Fig. 6). Lavrion partially overlaps these sources and cannot be ruled out. This observation is reinforced by recent results of LIA performed on objects from the roughly contemporaneous Egyptian trading port of Naukratis (Masson-Berghoff et al., 2018). The remaining arrowheads, have a wide range of isotopic ratios; while the cluster is comprised mainly of Khirbet Qeiyafa arrowheads, the scatter includes arrowheads mainly from Ramat Raḥel. Several arrowheads in the scatter are relatively poor in lead (Qe-26, RH-4, RH-11, RH-12 and RH-13), others contain up to 3% Pb, except for RH-3 containing 13% Pb.

Based on the $^{207/204}$Pb vs $^{206/204}$Pb two mixing lines are apparent. One group (the upper mixing line) is comprised mainly of Type I arrowheads and the second includes arrowheads of all three types. While, the relatively high isotopic raios of Sample RH-1 suggests that copper ores from the Taurus Range in Anatolia(not shown) may represent one end member, the two additional end members are unknown. Generally, for the first group (Type I), with higher $^{207/204}$Pb values, sources of copper in the Araba and Sinai, which may contain Pb in percentage level, are possible, while for the second group, Egypt, Sinai and Iran (?)

Table 2
Chemical and Lead Isotope Composition*. All concentrations are in % weight.

	Fe	Co	Ni	Cu	Zn	As	Ag	Sn	Sb	Pb	Bi	Total	$^{206}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$
Azekah-1	0.05	0.026	0.06	70		0.09	0.010	18	0.69	2.1	0.003	91	18.62	38.75	15.65
Azekah-2	0.48	0.016	0.12	72		0.52	0.028	6.1	0.15	2.1	0.013	82	18.78	38.91	15.68
Azekah-3		0.009	0.05	87		bdl	0.010	7.0	0.15	1.1	bdl	95	18.72	38.90	15.69
Qe-20	0.2	0.002	0.02	73	0.073	0.21	0.012	0.8	0.03	2.6	0.004	77	18.78	38.87	15.67
Qe-21	0.1	0.020	0.05	79	0.002	0.15	0.014	8.1	0.10	3.9	0.035	91	18.79	38.88	15.68
Qe-22	bdl	0.003	0.02	74	0.003	0.08	0.011	2.9	0.02	9.2	0.005	86	18.79	38.84	15.67
Qe-23	3.2	0.001	0.03	61	0.000	3.6	0.010	0.1	0.51	12	0.125	81			
Qe-24	bdl	0.004	0.05	84	0.001	0.06	0.026	3.4	0.03	0.20	0.004	88	18.77	38.86	15.68
Qe-25	bdl	0.014	0.06	69	0.002	0.1	0.003	3.7	0.25	23	0.022	97	18.80	38.84	15.68
Qe-26	0.1	0.080	0.02	100	0.032	0.24	0.004	0.0	0.01	0.04	0.002	101	18.59	38.56	15.61
Qe-27	0.20	0.036	0.05	85	0.004	0.15	0.022	7.7	0.10	4.3	0.303	98	18.74	38.86	15.68
Qe-28	0.10	0.020	0.06	82	0.002	0.05	0.012	10	0.13	2.80	0.013	96	18.80	38.82	15.67
Qe-29	bdl	0.005	0.03	67	0.000	0.01	0.002	4.6	0.03	16	0.119	88	18.84	38.84	15.68
Qe-30	0.10	0.012	0.04	68	0.001	0.03	0.004	5.0	0.04	13	0.016	86			
RH-1		0.019	0.07	57	0.016		0.051			1.40		58	18.91	39.16	15.69
RH-2		0.003	0.01	62	0.006		0.041	0.8	4.0	1.00		68	18.74	38.79	15.64
RH-3	0.50	0.013	0.02	73	0.008	0.04	0.002	0.4	0	13.0	0.012	87	18.60	38.57	15.59
RH-4	bdl	0.000	0.10	97	0.001	1.7	0.008	2.9	2.9	0.03	0.204	105	18.38	38.32	15.58
RH-5	bdl	0.022	0.06	77	0.002	0.01	0.004	4.1	0.73	2.8	bdl	85	18.43	38.45	15.64
RH-6		0.018	0.09	62	0.010		0.144			3.3		66	18.82	39.00	15.68
RH-7		0.003	0.03	32	0.001		0.030			0.70		33	18.52	38.66	15.65
RH-8		0.006	0.01	69	0.007		0.018	1.3	1.6	2.8		74	18.77	38.82	15.65
RH-9		0.010	0.04	53	0.004		0.821	1.4	9.6	2.0		67	18.31	38.41	15.64
RH-11		0.005	0.04	99	0.033		0.170			0.60		100	18.39	38.37	15.60
RH-12		0.036	0.06	88	0.016		0.133			0.10		88	18.57	38.65	15.65
RH-13	0.10	0.061	0.05	91	0.008	0.19	0.011	4.3	0.01	0.30	0.022	96	18.61	38.72	15.66

are potential sources. However, there are problems with some of latter, as for the Sinai ores the quality of the data is low, there is a lack of data from Egypt and no archaeological data for their exploitation during the Persian period. Despite continuous investigations in Faynan and Timna, in the Araba, no evidence was found for mining or smelting operations between the Iron Age and the Roman period.

The results may be also viewed individually, rather than as a result of mixing. In this case, in addition to the north Aegean/Western Anatolian cluster, there are several consistencies and inconsistencies which may be pointed out, some in agreement with the data presented by Masson--Berghoff et al. (2018):

1) None of the Pb-poor samples are consistent with copper ores from the exploited units in the Araba (i.e. DLS in Faynan and Amir-Avrona formations in Timna, contra Masson-Berghoff et al., 2018 and see Fig. 6).

2) Cypriot copper sources are consistent with several of these samples (Fig. 7); Qe-26 which does not contain added lead is consistent isotopically with ores from Larnaca. The latter is also consistent with ores from Limni and from the Mavrovouni mining dump in the Solea region. RH-12, also without Pb addition, is consistent with ores from Limni. Additional data is required in order to substantiate these observations. It should be noted, however, that evidence for the exploitation of copper from the Solea region during the Cypro-Archaic and Cypro-classical periods has been presented (Kassianidou, 2013).

3) Iran: RH-12 along with RH-13 is also consistent isotopically with copper ores from Karkas, Iran (Fig. 8). The possibility that some of the arrowheads were brought from the Persian homeland is further suggested by RH-7 which is consistent isotopically with lead ores from Naklak. Again, much more data is required in order to corroborate this hypothesis, however, it should be noted that all three are Type I arrowheads, originating from Ramat Raḥel. A situla from Naukratis (Cat. No. 12) produced similar results (Masson-Berghoff et al., 2018:331). Notably, Dugaw (2017; Dugaw, Lipschits and Stiebel, forthcoming) relates all Type I arrowheads in the southern Levant, to the Babylonian destruction levels (see above).

3.3. Correlation between chemical composition and LIA

The $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ ratios were tested as dependent variables against the concentrations of Fe, Co, Ni, Cu, Zn, As, Ag, Sn, Sb, Pb and Bi using a stepwise regression control for a standard least square model with the stopping rule being maximal R^2 value for the validation set. Positive validation R^2 values have been reached only for $^{208}Pb/^{204}Pb$, with 12 observations as the training set and four observations for validation consisting of all items which had observations for the relative variables. The results indicate that the proportions of Sb, Co, As and Sn are significantly correlated with the $^{208}Pb/^{204}Pb$ ratio at the 0.05 level ($R^2 = 0.96$; RMSE = 0.04; P-value < 0.0001; R^2 (validation) = 0.19 see S1 for further details). Based on the small number of observations, only very general conclusions can be drawn: It appears that artefacts which differ isotopically from the main cluster (north Aegean/western Anatolian, see Fig. 6), also differ in their chemical composition. Notably, two of the four Sb-rich samples (RH-9 and RH-4) plot at the end of each of the two mixing lines suggested, strengthening the possibility that Iranian sources were used (Fig. 8).

As will be detailed below, both chemical and isotopic composition correspond to both the traditional and to the 3DGM-based shape classifications.

3.4. Comparing traditional and 3DGM-based shape classifications

Fig. 9 presents a bivariate scatter-plot of principal component 1 and 2 scores for all items grouped by site and traditional typological classification (Types I-III). These two shape trends explain together approximately 51% of the shape variability in the entire sample. When considering only these two shape trends, the majority of artefacts seem to be sharing a similar shape space. However, a more in-depth inspection clearly shows that both provenance and typology can be distinguished using the geometric morphometric results.

Two stepwise forward linear discriminant analyses have been conducted for the principal component scores against the sites and typological classification of the artefacts, with 21 observations as the training set and six for validation. For site classification, the results show that using the scores of principal components 1, 2 and 5 allows a correct classification of some 86% and 84% in the training and validation sets, respectively. For type classification, the scores of same principal components provide a correct shape classification of some 95% and 100% for

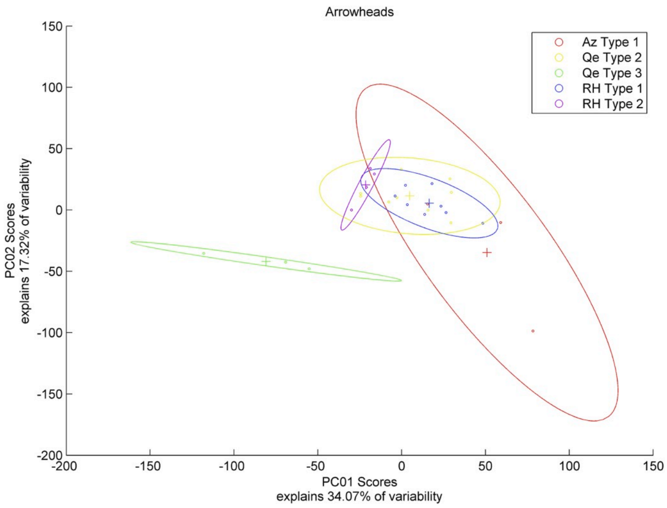

Fig. 9. Scatter plot of the principal component scores of the artefacts on the first two principal components. Artefacts are grouped by traditional typological classification and site. + signs represent mean group shape. Ellipses represent 95% confidence interval.

the training and validation sets, respectively. While the latter corroborate the validity of the traditional subjective typological classification, they also allow the classification of the artefacts by site, a possibility which cannot be achieved using traditional typological classes.

The principal component scores were also subjected to the same analysis in an attempt to classify the objects to both site and type. This analysis also incorporated the scores of PC19 in addition to those used in the former analyses. The results allowed a correct classification of 90% and 84% in the training and validation sets, respectively. Thus, it appears that specific shape trends were significantly different both between sites and between types to such an extent that allows their use as classificatory factors. This observation is further corroborated by a Wilcoxon rank-sum non-parametric test conducted on multidimensional inter-point distances between group's centroids and observations. The results indicate that a significant difference exists between the mean shapes of Type 1 arrowheads from Azekah and those from Ramat Raḥel (rank-sum = 87, p-value < 0.05) as well as between Type 2 arrowheads from Qeiyafa and those from Ramat Raḥel (rank-sum = 125, p-value < 0.05). These results show that small-scale, yet archaeologically significant shape differences exist between the artefacts in the different groups, differences which were not discerned by traditional typological classifications.

3.5. Correlation between chemical composition, lead isotopic ratios and morphology

The 3D geometric morphometric analyses, which produces specific high resolution quantitative shape trends, allows the application of powerful in-depth models for testing the relation between morphology on the one hand and chemical composition and isotopic signature on the other. The principal components scores were tested as dependent variables against both chemical and isotopic results. For the chemical analysis the Co, Ni, Pb and Ag concentrations were taken as factors in a standard least square model. These elements were selected in contrast to other tested elements, since for these the dataset is more complete, allowing to maximize the analysed sample. These were tested against each of the first 12 principal components (explaining together more than 91% of the shape variability in the sample), with 17 observations as the training set and 6 observations for validation. The results indicate that Ni, Pb and Ag are significantly correlated to artefacts' scores only on PC5 at the 0.05 level ($R^2 = 0.46$; RMSE = 13.54; P-value = 0.04; R^2 (validation) = 0.26, see S2).

For the isotopic analysis, the $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ ratios were taken as factors in a standard least square model. These were tested against each of the first 12 principal components (explaining together more than 91% of the shape variability in the sample), with 16 observations as the training set and 6 observations for validation. The results indicate that $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ ratios (similarly to Ni, Pb and Ag concentrations) are significantly correlated only to artefacts' scores on PC5 at the 0.05 level ($R^2 = 0.61$; RMSE = 10.5; P-value < 0.01; R^2 (validation) = 0.2 see S3).

While PC1 and PC2 separate the Type III arrowheads from Types I and II (Fig. 9), these differences, which explain the vast majority of variability in the sample, do not seem to be related to the chemical and isotopic signatures. In contrast, PC5 separates Type I arrowheads from Types II and III (Fig. 10), and is strongly correlated to chemical composition and isotopic signature. Thus, while in terms of general shape, Types I and II are more similar to each other, in terms of chemical composition (of several elements), isotopic signatures, and shape trend expressed by PC5, Types II and III are more similar to one another than Type I, which is distinctly different. If we associate the isotopic signature and chemical composition of the artefacts with their production origin, then we can conclude that the shape differences expressed by PC5 reflect a correlation with the place of production which might also be related to chronology.

4. Discussion and conclusions

Twenty-nine socketed bronze arrowheads from three sites (Khirbet Qeiyafa, Ramat Raḥel and Azekah) were subjected to chemical, isotopic and geometric morphometric shape analysis, with the purpose of examining the potential of such a combined methodology when studying complex artefacts.

The three types (denoted Types I-III) that were defined by visual examination in the present study were found to correlate with typology of the socketed bronze arrowheads from the southern Levant recently proposed by Dugaw (2017; Dugaw, Lipschits and Stiebel, forthcoming). Type I was found at Ramat Raḥel and Azekah and according to Dugaw, appears to have been used in the southern Levant mainly during the late seventh and early sixth cent. BCE. Type II was found at Ramat Raḥel but is most frequent at Khirbet Qeiyafa and is a common Achaemenid type, widely used during the fifth-fourth centuries BCE. Type III was found only at Khirbet Qeiyafa and developed in the Near East, likely later in the Persian period.

The 3D geometric morphometric analysis validated this tripartite classification. As expected, it showed that the difference between the mean shape of Type III and that of Types I-II was much larger than the

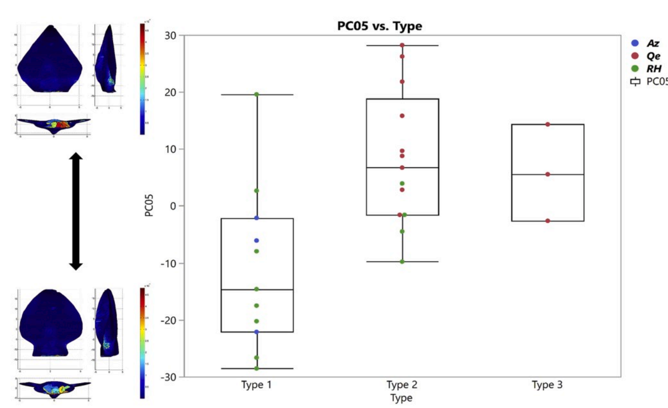

Fig. 10. Artefacts' scores on PC05. Colour coding reflect the site from which the artefact is derived while signs reflect traditional typological classifications. The two models reflect hypothetical artefacts on the extremities of PC05. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

difference between the mean shape of Types I and II. However, by allowing a quantitative comparison between mean groups' shapes it demonstrated that Type I at Azekah was significantly different than Type I at Ramat Raḥel, and that Type II at Ramat Raḥel was significantly different than Type II at Khirbet Qeiyafa. These differences, highlighting the significance of the find place – the site – went unnoticed in the traditional subjective typology, indicating possibly the different production venues of such artefacts (and see more below).

In addition, application of 3D geometric morphometric analysis enabled the identification of a specific and relatively minute shape trend, described by PC5, that is significantly correlated with chemical and isotopic compositions of the arrowheads: PC5 separates Type I arrowheads from Types II and III, a separation that is reinforced by the chemical composition and isotopic signature. Thus, while in terms of general shape, Types I and II are more similar to each other, in terms of chemical composition (of several elements), lead isotopic signatures, and shape trend expressed by PC5, Types II and III are more similar to one another than to Type I. This result correlates with the chronology of these types wherein Types II and III are roughly contemporary and later than Type I, and possibly with their place of production; The minute difference between the Type I arrowheads from Ramat Rahel and the Type I arrowheads from Azeka (described above) suggests that the arrowheads used (possibly by the conquerors) in each site, were produced separately. The difference between arrowheads of Type II in different sites, suggests, on the other hand, local dispersed production.

The majority of the Khirbet Qeiyafa arrowheads (both Types II and III), which were relatively rich in lead, together with a large group of objects from Naukratis (see above) formed in terms of their isotopic composition a rather tight cluster that is consistent with the isotopic ratios of lead ores from western Anatolia and northern Greece. While this prevents us from identifying the source of copper, it suggests that if Types II and III were indeed locally produced, their production involved the addition of foreign lead. From both the southern Levant and from Naukratis there is ample evidence for contacts with the above mentioned regions (for Naukratis see Villing, 2015 and summary in Masson-Berghoff et al., 2018:334 with references therein). While the trade in metals such as iron, silver and lead from the Aegean to the southern Levant along with other products was indicated previously from texts (Van Alfen, 2002), it is evidenced here for the first time. Certain types of amphorae found in small numbers in the southern Levant during the Persian period were produced in the north and south-eastern Aegean (based on petrographic and NAA analyses), and may have been the by-product of this metal trade (Shalev, 2014:387).

The isotopic results show that Type II and III arrowheads were unlikely produced in the Persian homeland, which is rich in lead ores (e.g., Momenzadeh, 2004). While beyond the scope of this paper, these results have the potential to shed light on the *modus operandi* of the Persian Empire in their rule of distant lands. While the extent to which the Persian forces were actually present in the southern Levant is debated (e.g., Edelman, 2007; Tal, 2005; Shalev, 2014: 19–20, 86 and bibliography therein), it is apparent from our results that the arrowheads of Iranian origin were uncommon here and instead, socketed bronze arrowheads were probably locally made. Were these produced here for the use of Persian forces? Or, was this type adopted by local inhabitants or local elites for their own use? The origin of the socketed bronze arrowheads in what appears to be the basement of an abandoned building in Khirbet Qeiyafa certainly suggests that the arrowheads belonged to the inhabitants of the building which yielded no other 'Persian' material culture (see above).

A different picture emerges from the lead isotope analysis of the Type I arrowheads. These were generally inconsistent with lead ore sources in the Aegean. If we accept Dugow's suggestion that the arrowheads of Type I should be associated with the Babylonian forces, then the isotopic data and the different potential sources suggested above, do not preclude the production of these arrowheads in the Babylonian heartland which unlike the Persian homeland is devoid of metal sources and was dependent on sources in the Caucasus, Eastern Anatolia and based on our results, perhaps Iran. The relatively high antimony in some of the arrowheads from this groups would fit well with several ore formations in all of these regions.

In summary, although in the present study, 3D geometric morphometric analysis combined with chemical and isotopic analyses was performed on a relatively small group of objects, it shows great potential for future typological studies, mainly of large assemblages and particularly in the much-needed integration between morphological and material analyses. Beyond the methodological advantages, implications of these results can be significant also to our understanding of the organization of production and the mechanisms of distribution and consumption of the objects under study, in this case, the socketed bronze arrowheads from the Achaemenid period in the southern Levant and beyond.

Acknowledgments

We would like to thank Leore Grosman and Ortal Harush from the Computational Archaeology Laboratory at the Institute of Archaeology, The Hebrew University of Jerusalem, for producing the 3D scans. We would also like to thank Igor Kreimerman, Debi Sandhaus and Alla Rabinovich who provided data regarding the material from Khirbet Qeiyafa, and Leora Freund for providing contextual data for the arrowheads from Azekah and Ramat Raḥel. Aurelia Masson-Berghoff from the British Museum kindly discussed the material from Naukratis. Svetlana Matskevich prepared the illustrations. The work was made possible with the kind assistance of the Lady Davis Fellowship Trust.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jas.2020.105147.

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