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RESEARCH ARTICLE

Early evidence of royal purple dyed textile from Timna Valley (Israel)

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Abstract

In the context of a broad study aimed at examining dyeing technologies in the Timna textiles collection, three samples of prestigious fibers dyed with murex sea snail were identified. Our identification is based on the presence of 6-monobromoindigotin and 6,6-dibromoindigotin components (detected using HPLC analysis), which is considered unequivocal evidence for the use of murex-derived purple dyestuff. Furthermore, by comparing the analytical results with those obtained in a series of controlled dyeing experiments we were able to shed more light on the specific species used in the dyeing process and glean insights into the ancient dyeing technology. The samples originated from excavations at the extensive Iron Age copper smelting site of "Slaves' Hill" (Site 34), which is tightly dated by radiocarbon to the late 11th-early 10th centuries BCE. While evidence for the important role of purple dyes in the ancient Mediterranean goes back to the Middle Bronze Age (early 2nd millennium BCE), finds of dyed textiles are extremely rare, and those from Timna are the oldest currently known in the Southern Levant. In conjunction with other observations of the very high quality of the Timna textiles, this provides an exceptional opportunity to address questions related to social stratification and organization of the nomadic society operating the mines (early Edom), the "fashion" of elite in the region during the early Iron Age, trade connections, technological capabilities, and more.

Introduction

Early iron age textiles from Timna Valley

Textile dyeing has been practiced since prehistoric times, using dyes that are extracted from both plant and animal sources. The color of textiles provides a window into various aspects of ancient societies, including the role of textile dyeing and technological achievements, fashion, social stratification, agriculture and trade connections [1,2]. However, textiles are rare in the archaeological record. Like any perishable organic material, they are usually subject to rapid decomposition and their preservation requires special conditions to prevent destruction by microorganisms [1,3]. Such conditions exist in the ancient copper-ore district of the Timna Valley (southern Israel). Starting in 2013, excavations in several copper production sites in the region by the Central Timna Valley Project [4,5] uncovered dozens of fragments of dyed textiles, which currently constitute the largest Iron Age assemblage in the Southern Levant [6]. This rare assemblage from Timna provides a window into aspects of past societies that are usually hardly visible in the archaeological research. Focusing on the dyeing technologies, we applied High Pressure Liquid Chromatography (HPLC) analyses to identify organic dyestuff; we found three items that were dyed with true purple, which is based on extracts from the murex sea snail. True purple—also known as 'royal purple'-was considered the most prestigious dye for textiles in many ancient societies. The finds from Timna currently constitute the earliest physical evidence on dyed fiber of true purple in the Southern Levant. Their context—in a marginal region inhabited by seminomadic tribes, and within debris of copper smelting activities-sheds new light on the society that operated the mines at the turn of the 1st millennium BCE, with implications for our understanding of trade and economy in the broader Southern Levantine region during this period.

True purple in antiquity

True purple was produced primarily from three species of sea mollusks of the Muricidae family, which were common in the Mediterranean Sea: *Hexaplex trunculus (Murex trunculus)*, Bolinus brandaris (Murex brandaris) and Stramonita haemastoma (Thais haemastoma) (Fig 1). This is in contrast to the "imitation purple" dye that was manufactured using various techniques that were based on much cheaper materials than the dyes from the sea snail [7,8]. True purple does not fade easily (as described by the Roman Period author Plutarch [9]). Together with the high complexity of the dyeing process, the small quantity of dyestuff that each snail contains (0.9 g on average in *H. trunculus*, for example [10]), it is easy to understand how purple textiles turned into an object of desire and the color itself into a symbol of nobility [11-13]. The true purple colors range in shades from purplish-red to violet-blue. Dyeing is based on material extracted from the snail's hypobranchial gland (located under the mollusc's mantle [14]), and the exact shade depends on different parameters, such as the chemical precursors compounds of each snail species, the dyeing process, and the levels of oxygen and light that the dye is exposed to [15]. The dyeing process using murex was a sophisticated, multi-stage process [15]. It was more complex than dyeing techniques based on plants: The snail-harvest [15,16] and the extraction of the gland from the snail required knowledge of biology and much more efforts than collecting plants in the field [17-19]; murex-based dyeing must take place close to the site from which the snails originate, because the freshness of the material has a significant effect on the results [19-22]; and finally, the dyeing process is long and involves biochemical, enzymatic and photochemical reactions, and requires reduction and oxidation processes that probably took several days [19].

Because the purple dyeing technology is no longer practiced [10], our knowledge is based on textual sources like the description of Pliny the Elder (Plinius, NH: IX; see also [19,23]),



Fig 1. Shells of the three species of murex snails. left to right: S. haemastoma, H. trunculus and B. brandaris (Photo by Shahar Cohen).

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and inferences from traditional indigo dyeing, which although simpler, involves similar processes (such as the ones related to redox and alkalinity conditions) and is still practiced today in different parts of the world [18,24]. Experimental dyeing also provides insights on the ancient dyeing technology. Although most experiments use strong chemical reagents to shorten the process to a few minutes (like in the hydrosulfite vat method, see S1 Appendix), successful dyeing has been achieved also in experiments based on materials that were available in antiquity [17–19,23,25]. The murex dye belongs to the 'vat dye' group that is not soluble in water, thus it requires an alkaline environment. This was achieved by adding various materials such as natural natron, or wood ash of alkaline plants [19,26,27], urine, or even broken shells (S1 Appendix). Reconstructing the reduction stage, which in modern vats is achieved by sodium hydrosulfite and requires non-aerobic conditions, is much more complicated [17]. A number of options have been raised, including the material of the vessel itself (tin or lead, [27]), honey, or madder roots with bran (known as indigo reduction agents [28]), but all failed in experiments. The only successful reconstruction is based on the bacteria in the flesh of the snail itself [17-19]. In this reconstruction, the dye-solution was heated at a moderate temperature (around 50°C) for a few days, until it turned yellow, indicating a leuco (soluble) form, and that reduction was completed. In the last stage, after about three days, the fleece or textile was

dipped in the reduced dye-solution and then left to oxidize in the air. Once oxidation took place, the dye turned purple.

It is not entirely clear when murex was first used for dyeing, bur the first archaeological evidence that regards the purple dyeing industry are indirect evidence which includes heaps of murex shells, that are dated to around the 19th century BCE. In most cases, this early evidence is associated with the Minoans [29-32]. However, recent archaeomalacological analyses raised the possibility of a multiregional origin of the purple dyeing industry, including regions outside the Aegean [33]. Direct evidence to the use of shellfish purple was identified in the islands of Santorini and Rhodes, where purple pigments from murex snail were found in wall paintings dated to the 17th century BCE [34,35]. The earliest textual evidence for the use of true purple in the Near East is attribute to the murex shells according to textual context is from Akkadian tablets from Nuzi, Mesopotamia, dating to 1425 BCE (this identification is based on contextual and philological arguments [36,37]). Other early written sources are the Amarna letters dated to the 14th century BCE, in which items made of wool dyed with the prestigious purple are described among other merchandise [38]. According to some scholars, the Phoenicians received their name because of their important role in the dyeing industry (assuming the original meaning of φοῖνιξ was "red-purple", [32,39]). The first use of the term 'royal purple' is found in a clay tablet from Knossos in Crete dated to the 13th century BCE [40,41]. In the Hebrew Bible, the words argaman and tekhelet appear side by side and most probably represent a purple hue and a violet-blue hue respectively (similar to the Akkadian words argamannu and takiltu [42]). It is widely accepted that both colors were based on murex snails, while tekhelet is specifically linked to Hexaplex trunculus by most scholars [43-45] (although it should be noted that the specific hue is still enigmatic [45]) Both hues are mentioned in the Hebrew Bible in relation to clothing of people of the highest social rank (for example, the garments of the High Priest, Exodus 28:6 [46], and the Midianite kings, Judges 8:26) and religious activities (for example, activities at the tabernacle, Exodus 35:35). Wool dyed with argaman and tekhelet was mentioned in Sennacherib's Cylinder among other expensive materials like talents of gold and silver that Hezekiah king of Judah gave him from his palace and the temple in Jerusalem [47]. Most of the textual evidence relating to royal purple comes from the Classical era. It also indicates that purple textiles were highly valued and served as a symbol of prestige, social status and power. Among the various sources are Pliny the Elder [48], Aristo [22] and Vitruvius [49].

In contrast to the many sources that mention purple textiles and the dyeing industry, archaeological evidence is still extremely limited. Common evidence of the purple-dye industry in archaeological sites are the heaps of murex shells, from which the dye-glands were removed [25,50–52]. Such heaps, however, may be evidence of other uses of the snails, including food, lime productions, ceramic temper material, decoration and more [14,33,53,54]. Their association with the dyeing industry is typically based on archaeological indicators such as industrial facilities, working surfaces, indicative tools and alike [55]. Large quantities of crushed shells with a hole in the area of the hypobranchial gland also suggest a deliberate cracking activity as part of the dyeing industry [53]. Still, in some cases crushed murex-shell deposits are difficult to interpret [33]. Some researchers propose a recycling hypothesis, according to which the waste from the purple-dye industry was used for other purposes [33,55].

Important direct evidence of the purple industry can be found in stains of purple on pot sherds. The dye remains are found in most cases on the upper part of ceramic basins, on the inside surface, the areas in which the reduced dye-solution was exposed to air, and underwent oxidation that turned it purple [19,56]. Pottery sherds stained with true purple were found in Sarepta in Lebanon— 14th–13th centuries BCE [57], Ugarit (Minet el-Beidha)— 15th–13th centuries BCE [52], Tel Keisan— 11th century BCE [19], Tel Shikmona— 10th–9th centuries BCE

[58] and Tel Kabri— 7th century BCE [19]. These finds indicate the important role of coastal sites in this industry.

Only a small number of textiles dyed with true purple were found in excavations, and until this study, the few that were found in Israel were all dated to the Roman period: two textiles from Masada [59,60] and three from a cave in Wadi Murabba'at, in the Judean Desert [61]. It should be emphasized that the rarity of true purple textiles predominantly reflects the limited preservation of textiles in the archaeological record; nevertheless, within the aforementioned assemblages (Masada, Murabba'at and now Timna) true purple textiles are the rarest.

Similar picture also emerges from research in other regions of the ancient Old World, which yielded only scant remains of pre-Roman textiles dyed with shellfish-based dyes. The earliest true purple textiles were found in Syria and are dated to the early second millennium BCE. These include finds from a burial at Chagar Bazar (18th–16th centuries BCE, [15]), and samples preserved in gypsum found in Qatna's palace [62]. The latter were found together with other precious artefacts, including gold beads and jewelry, and were associated with the royal elite [62]. Early evidence of true purple textiles also includes samples from a burial in Stamna (Aetolia, Western Greece) dated to the 12th–11th centuries BCE [63].

The archaeological finds and their context

Three of the many fragments of dyed textiles and fibers from Timna that were analyzed, yielded results compatible with true purple dyestuff (nos. 004, 017, 018; see Figs 2–4). They were found in the excavations of Area 13, Site 34 ("Slaves' Hill")—one of the largest copper smelting camps in the region [64]. The finds came from a context close to bedrock, defined as a "mixed" locus that represents a shallow deposition of waste embedded in reddish sediments and crushed sandstone (Locus 515, Basket 5644).

The site was tightly dated to the late 11th-first half of the 10th century BCE (early Iron Age) based on the results of excavations in several areas. These include a dozen of radiocarbon dates from short-lived materials (mostly date seeds [64]), the study of pottery typology [65] and other considerations [66]. In addition, as part of the current study we sent a fragment from one of the dyed items (no. 017) to be directly dated by radiocarbon. The result (Table 1) is in perfect agreement with the previously published chronology of the site.

Sample no. 004 was selected from a small group of pink-purplish wool fibers (Fig 2), which were probably ripped from a textile that did not survive. A microscopic examination indicated that the fibers were dyed before they were spun into threads, a standard procedure in the dyeing of wool [10,67]. Sample no. 017 was selected from two threads with a Z-ply (s2Z), 63.8 mm long and 4.9 mm wide, with a pink-purple edge (Fig 3). It is difficult to know the specific function of this string but we can assume that it does not come from a rough textile, but from a decorative fringe or a tassel, both common in the Timna textiles assemblage [68,69] and in the ancient world in general [70,71]. Sample no. 018 (Fig 4) was taken from a textile fragment (1.2 X 0.4 cm). The textile shows a weft-faced tabby weave with uncolored warp threads and decorative pink-purple weft threads. The threads are S-spun. We can assume that this fragment is part of decorative textile bands that are common in the Timna textiles [69]. It is difficult to say with certainty whether all the samples are related to the same textile, but this should not be ruled out (especially samples nos. 004 and 018 that gave similar chemical results-see later). The fiber was identified as wool on the basis of the morphology of its internal layer as observed under a polarizing microscope (Zeiss Axioscope 5) at x500 magnification. Wool fibers (sheep in this case) are characterized by a unique cellular appearance of overlapping scale-like structure (Fig 5), which is unlike any other fiber [72,73], including linen (also found in the Timna assemblage together with goat hair [6]). Wool played an important role in the dyeing industry

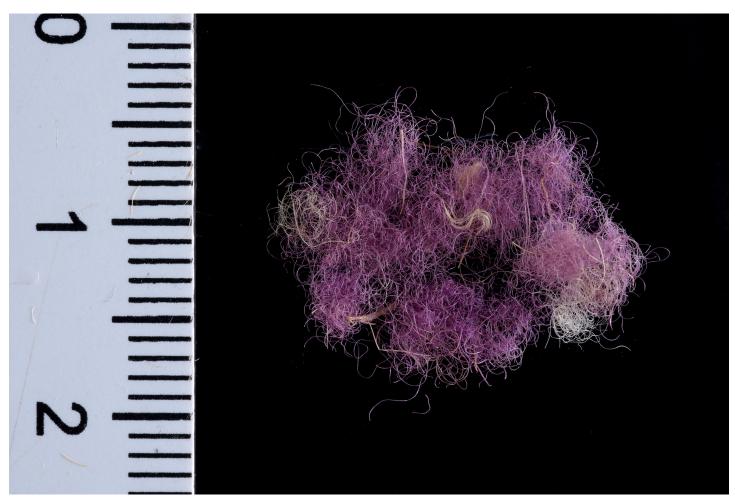


Fig 2. Sample no. 004. Wool fibers dyed pink-purple hue (photo by Dafna Gazit, courtesy of the Israel Antiquities Authority).

and was the best raw material for dyeing in the Levant prior to the introduction of cotton and silk [67,74–78]. This is because the protein composition of the fiber allows better absorption of the dye than that of the linen fiber [79,80]. The coarse goat hair on the other hand, was not used for delicate or embellished textiles.

Methods

Although the composition of the murex dye is not completely clear [27], it is obvious that the final shade is affected by certain colorant components (chromatogram), which can be identified by HPLC analysis in a single measurement: indigotin (IND) and indirubin (INR). The two are found in plant sources such as woad (*Isatis tinctoria L*.) and the indigo plant (*Indigofera tinctoria L*), as well as in several species of shellfish [81–83]. Other components that confer the purple color are present only in mollusk dyes: 6-monobromoindigotin (MBI), monobromoindirubin (MBIR), 6,6-dibromoindigotin (DBI), and 6,6- dibromoindirubin (DBIR), which are the ones that confer the purple color [62,68,83,84]. Other minor components, such as isatinoids and indirubinoids, were also recognized [82], but since they are often absent from archeological textiles, will not be discussed here.

Identifying the organic dye colorants in archaeological textiles is a complex task due to the low concentration of the molecules in the fibers and the limited amount of material available



Fig 3. Sample no. 017. A wool string made of two strands plied in Z-direction, with a pink-purple edge (photo by Dafna Gazit, courtesy of the Israel Antiquities Authority).

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for destructive analysis. In this study we used HPLC analysis, which is considered the most appropriate and reliable method for identifying dyes in archaeological textiles since 1985 [80,85–89]. This method, which is based on chromatography, is efficient in separating and identifying a mixture of compounds and is commonly used in analytical chemistry and biochemistry. It relies on pumps to pass a pressurized liquid solvent containing the sample mixture through a column filled with a solid adsorbent material. Each component in the sample interacts slightly differently with the adsorbent material, causing different flow rates for the different components, and leading to the separation of the components as they flow out of the column, then to be detected by a UV-Visible absorbance detector that quantifies the components in the solution [80,90]. Although the analysis is destructive, it can identify components that are present in minute amounts, and thus only very little quantities of the tested substance are required, ensuring minimal damage with high degree of accuracy and separation capability, qualities that are crucial for the identification of minor components in archeological textiles.

The identification of dyes was based on a database containing known chemical components and wool that was dyed with known dyestuffs and analyzed by the authors prior to the current Timna research [68], and on a series of analyses that were done by the authors in a previous study of wool that was dyed with each of the murex species (*Hexaplex trunculus, Bolinus brandaris* and *Stramonita haemastoma*), and analyzed under the same conditions as the archaeological samples ([58,61]; S1 Appendix)

Extracts of dyed textiles were analyzed by the HPLC-DAD (High-Performance Liquid Chromatography with Diode Array Detector) system (Hitachi LaChrom Elite Chromatography), at the HPLC Unit of the Mina and Everard Goodman Faculty of Life Sciences at Bar-Ilan University. HPLC system running EZ Chrom Elite v. 3.2.1 software consisted of an L-2130 binary pump, an L-2200 autosampler, an L-2300 column oven (column temperature of 30°C was used for all analyses), and an L-2455 Diode Array Detector, set to obtain chromatogram spectra in the range of 200-700nm, with extracted chromatograms at 254nm, 454nm, and 554nm. The chromatographic column was a GraceSmart RP18, 5 μ m, 250mm × 4.6mm ID. In each test, a characteristic chromatogram was obtained, and the color components were identified by the particulars of their retention time (R_t) and their characteristic absorbance spectra, including the wavelengths of the absorbance peak in the UV-visible spectrum (λ max).

Extraction method

The dyes were extracted from the fibers with DMSO (Dimethyl sulfoxide) solvent, which was considered appropriate for indigoid dyes [35,91]. Samples weighing 3 mg each were placed in an Eppendorf test tube (1.5 mL), along with 150 µl of DMSO. Each sample was heated to 95 °C in a water bath for 10 minutes. By that time, the solution turned blue, and was then separated from the settled sediment and transferred to a clean Eppendorf test-tube. Finally, the sample was subjected to centrifugation for 5 minutes at 12,000 g., and 20 µl of the supernatant was injected into the HPLC system' HPLC.

HPLC analysis protocol

The mobile phase for the protocol that was used in this study was made up of A: phosphoric acid 0.5% (w/v); B: methanol; and C: H_2O . The flow rate was held at 1 ml/min, and the injection size was 25 µL. Gradient elution conditions are tabulated in Table 2.



Fig 4. Sample no 018. A wool textile fragment decorated with pink-purple weft threads (photo by Dafna Gazit, courtesy of the Israel Antiquities Authority).

Table 1. Radiocarbon date for sample no. 017*.

Laboratory number	Locus/basket	Material	Age BP	Cal. BCE	
Beta-562244	515/5644	Wool	2850 ± 30 BP	1114–924 cal. BCE (2σ)	
				1053–933 cal. BCE (1σ)	

* Dating was done at the Beta Analytic Radiocarbon Dating Laboratory; calibration based on IntCal20 atmospheric curve.

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Results

According to the analysis result at 554 nm (Table 3), the archaeological samples were dyed with true purple. They contain two molecules that are principal markers of this color, namely MBI and DBI. The chromatogram of sample no. 004 shows only two pronounced peaks at 554 nm: MBI was detected at 10.75 R_t with a typical spectrum (242nm, 288 nm, 347, 608nm λ_{max}), and main peak at 14.40 R_t of DBI with a typical spectrum (292 nm, 307nm, 353 nm, 596nm



Fig 5. A single fiber of sample no. 004 under a polarizing microscope (photo by Naama Sukenik).

Time (min)	% A	% B	% C	
0	10	50	40	
3	10	80	10	
20	10	80	10	
25	10	90	0	
30	10	90	0	
33	10	50	40	
40	10	50	40	

Table 2. Linear gradient elution for HPLC-DAD analysis of the extracted purple dyes.

 λ_{max}). The other components did not appear in a significant amount. The chromatogram of sample no. 017 has a few peaks at 554 nm (Fig 6): IND was detected at 8.46 R_t and with a typical spectrum (242nm, 286 nm, 339 nm, 613nm λ_{max}), MBI was detected at 10.68 R_t (242 nm, 289 nm, 346 nm 608 nm λ_{max}), DBI at 14.23 R_t with a typical spectrum (292 nm, 304nm, 350 nm, 603nm λ_{max}) and DBIR was detected at 20.3 R_t with typical spectrum (304nm, 368nm, 553 nm λ_{max}). The chromatogram of sample no. 018 similar to sample 004 shows main peak of DBI at 14.40 R_t with a typical spectrum (291 nm, 302nm, 353 nm, 601nm λ_{max}) and minor peak at 10.7 of MBI (240nm, 347nm 610nm λ_{max}). It can therefore be determined with certainty that the fibers in all three samples were dyed with genuine shellfish dye.

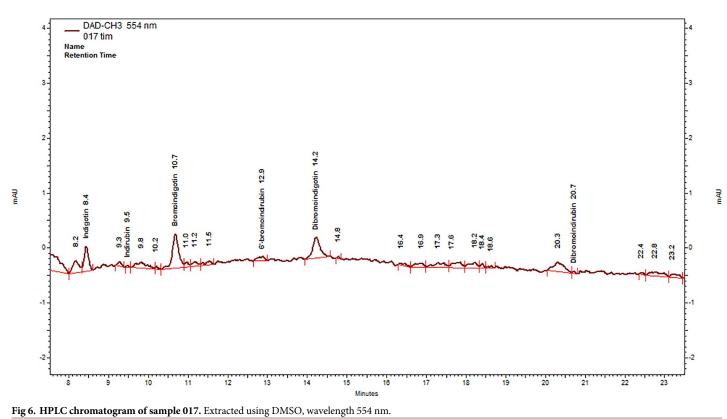
Distinguishing between the three Mediterranean molluscan species, *H. trunculus*, *B. brandaris* and *S. haemastoma*, is not easy because numerous variables (*e.g.* geographical location, age and sex of the snails, the dyeing process, analytical methods employed to record the composition etc.) may affect the reported composition of the purple dye [10,84,92]. Nevertheless, it is possible to see a clear trend with respect to the ratios of the dye substances in each species [27,35,58,61,84,91,93], that helps to identify the species of the murex used in the dyeing of the archaeological textiles with a high level of certainty.

Examination of the percentage of the colorants and the proportions of the components in chromatograms that were done by the authors in pervious analyses (Fig 7 and S1 Appendix; [58,61,94]), which is in agreement with other previous studies (see for example: [35,84,91]), shows that in most cases *H. trunculus* contains relatively high concentration of IND (indigotin) and high concentration of MBI (monobromoindigotin) compound, while DBI (*6,6-dibromoindigotin*) is present in relatively small quantities; in *B. brandaris* and *S. haemastoma* on the other hand, DBI compound is the most abundant (over 60%), but the proportion of IND in dyes derived from these two mollusks is low (less than 5%) and it cannot always be detected in the chromatography (Fig 7; S1 Appendix: Table 1; see also [61,84,93,95,96]).

According to the chromatogram of samples nos. 004 and 018 only two components were detected. The most abundant colorant is DBI (85%-86%), followed by MBI (14%-15%), while

IAA Number		IND MBI		DBI	DBIR			
004	Retention time (minutes) (\lambda max nm)	8.5	10.7 (242, 288, 347, 608)	R _t 14.4 (292, 307, 353, 596)	20.6			
	Relative percentages % of the component	1%<	14%	86%	1%<			
017	Retention time (minutes) (\lambda max nm)	R _t 8.45 (242, 286, 339, 613)	R _t 10.68 (242, 289, 346, 608)	R _t 14.2 (292, 304, 350, 603)	R _t 20.3 (304, 368 553)			
	Relative percentages % of the component	11.10%	15.60%	63.87%	9.40%			
018	Retention time (minutes) (\lambda max nm)		10.7 (240, 347, 610)	14.4 (291, 302, 353, 601				
	Relative percentages % of the component	1%<	14.28%	85.71%	1%<			

Table 3. Analytical results, including retention time, spectral wavelengths identification and the relative percentage of the components that were identified at 554 nm.



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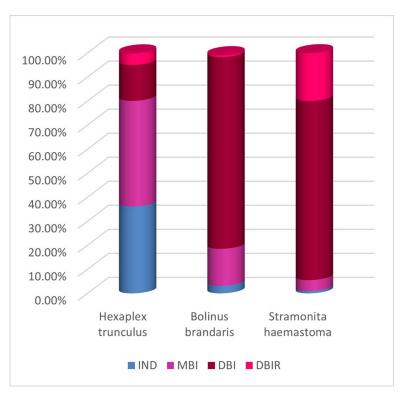


Fig 7. Relative proportions of dye components (IND; MBI; DBI; DBIR) at 554 nm in modern fleeces dyed with the three species of sea mollusks (the graph shows the average results of all the modern samples).

no IND and DBIR components were found in significant amounts (Table 3). Hence, it can be determined that samples were colored with a dye substance obtained from *B. brandaris* or *S. haemastoma*, which are characterized by a relatively high concentration of DBI compound and low concentration of IND (see also [91]). The experimental dyeing with this species strengthens our conclusion, because while *H. trunculus* yielded more violet-blue hues due to the indigo compound, the two other species produce a more pink-purple hue (Fig 8; S1 Appendix), due to the high concentration of the combined DBI and the DBIR components which are responsible for the characteristic red/pink hue (Table 3 [97]). Although a separation of *B. brandaris* and *S. haemastoma* species is not possible [35,91], it should be noted that according to the experimental dyeing results, *S. haemastoma* is a little easier to use than *B. brandaris* due to the difference in gland-size (an average of 0.6 gram in *S. haemastoma*, an average of 0.2 gram in B. *brandaris*), and it produces a hue of reddish-purple similar to that of the archaeological fiber (see S1 Appendix).

According to the chromatogram of sample no. 017, the DBI compound appears in a relatively high concentration (63.87%), which presents the relatively high peak areas measured at 554 nm, followed by MBI (15.60%), IND (11.10%) and DBIR (9.40%) (Table 3, Fig 7). Although all the components that are known in each of the three species of murex were found in the same sample (include DBIR components, see Fig 6), the proportions that are seen in the chromatogram, with high concentration of DBI but still relatively large concentrations (above 8% each) of MBI, IND and DBIR, do not fit any of the typical murex species. This composition could result, according to Koren (e.g., [19,84]), from numerous variables in H. trunculus, including a type of *H. trunculus* that is indigo-poor and DBI- rich. However, in our opinion it is more likely to be the outcome of a double dyeing method using either B. brandaris or S. haemastoma in combination with H. trunculus, a method that was mentioned by Pliny the Elder for obtaining reddish-purple colors (Plinius, NH: IX, 62, 137). A series of dyeing experiments that was conducted by the authors shows that although B. brandaris or S. haemastoma produce very beautiful color, the quantity obtained is small. Adding a few glands of *H. trunculus* helps in increasing the amount of color that is absorbed into the fibres (S1 Appendix). Doumet [98] already noted the importance of the enzymes (purpurase) existing in H. trunculus for the dyeing process, and showed that a better result was achieved when 10% of H. trunculus glands were added to a *B. brandaris* or *S. haemastoma* bath dye (see also [21,99]). However, it is important to refer to our proposal with caution. More information about the origin of the purple pigments and the impact of the variables, is necessary before a clear conclusion can be drawn in the future.

Discussion

Observations on the purple dyeing industry of the early iron age

The dyed textiles from Timna constitute a corpus of paramount importance, as no parallels are known to us today from the Southern Levant. Their analyses enhance our knowledge of garment and textile production technologies in the early Iron Age (~1100–900 BCE). The results of the present study indicate that the pigments present in three different samples of fiber are remnants of true purple dye. While pre-Roman evidence of the use of this dye in the Southern Levant exists in the form of murex shells heaps (*e.g.*[52,100] or stained ceramics (*e.g.* [19,56,58,84]), until now none of the surviving pre-Roman textile fragments have tested positive for a true purple dye.

According to our results the dyeing process of all samples was based on *B. brandaris* or *S. haemastoma* (the analytical results did not allow distinguishing between these two species). In one sample (no. 017) *H. trunculus* was most probably also used, and this sample therefore

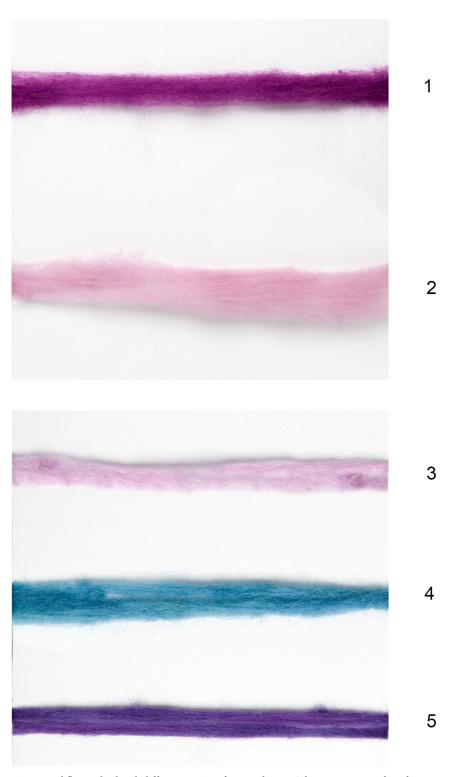


Fig 8. Wool fleeces dyed with different species of sea snails. 1–2: *S. haemastoma*; 3. *B. brandaris*; 4–5: *H. trunculus* (Dyeing—Zohar Amar and Naama Sukenik. Photo by Shahar Cohen).

represents the double-dyeing process for the production of the expensive dye of Tyrian purple, which was described by Pliny the Elder (NH: IX, 62, 137). According to Pliny, in this method the fiber was dipped into a first bath of '*pelagium*' and then into a second bath of '*bucinum*'. Although the identification of these species is not certain, it is agreed by all researchers that Pliny refers to two different species of murex [10,101–103]. It is important to emphasize that the two species were added to enhance the reddish color [19], as seems to be the case for the Timna sample (here probably *H. trunculus* with *B. brandaris* or *S. haemastoma*).

It is reasonable to suggest that the Timna samples are a product of the dyeing industry in the Phoenician Coastal Plain. The three murex species discussed above are present in the Mediterranean Sea and are not found in the Red Sea [104,105]. In the Eastern Mediterranean, the largest concentration of Iron Age sites with evidence of purple industry is located along the northern coast of the Southern Levant [15] (Fig 9). They include Tyre, Shikmona, Tel Kabri and Tel Keisen [16,52,56,58,100,106]. The only other sites with possible evidence of Iron Age purple industry currently known in the wider Eastern Mediterranean are Hala Sultan Tekke in Cyprus [52] and Tel Garisa in modern day Israel [16].

Another species of murex—*Virgin murex* (*Chicoreus virgineus*, also from the family *Murici-dae*)—is found in the Red Sea [107]. Although there is no direct evidence of dyeing with this species, a few of its shells were reported from the Roman sites of Myos Hormos (Egypt) and the port of Hafun (Somalia), raising the possibility that it was used for dyeing [108]. However, the scarcity of the finds, coupled with the absence of historical evidence makes the possibility of the use of this snail in the production of the Timna samples much less likely.

It is important to emphasize that in Egypt, only one site related to the purple industry has been found so far; it is dated to the Late Bronze and the Roman periods (Marsa Matruh, [52]). Additionally, the identification of mollusk purple in Egyptian textiles is rare prior to the Roman Period [52,109–111]. For example, in the Amarna textiles no indication of true purple was found [112], and the same is the case for other assemblages of high quality textiles (related to the pharaohs and high officials) from Middle Kingdom and New Kingdom sites [113]. The exclusion of Egypt as the source of the purple dyed textiles of Timna is in accord with our assumption that all wool textiles of this assemblage were not obtained from Egypt, which specialized in growing flax and the production of linen [68,69]

Observations on the early iron age society of Timna Valley

The find of fibers dyed with true purple in Timna adds to a growing body of evidence for the complexity of the society operating the copper mines during the early Iron Age [64,68,69]. This period, which followed the collapse Egypt and the end of its control over the Southern Levant [114], is typically considered an era of limited inter-regional connections, although this is starting to change with the advance of the application of archaeological sciences [115]. The societies are assumed to be quite fragmented, and the timing of the emergence of local polities of historical significance, such as the kingdoms of ancient Israel, Edom and Moab, is hotly debated, as it has implications on our understanding of the historicity of the Old Testament (e.g. [116]).

The identification of the early Iron Age society of Timna Valley and the nearby regions with biblical Edom is discussed elsewhere ([117] and references therein)]. The discovery of true purple-dyed textiles renders further support to the suggestion that this society was part of a *kingdom* (the Edomite Kingdom) already in this period [117–119], as it provides strong evidence for the presence of elite (see also a study based on diet [120], and direct evidence for long-distance trade connections (see also [68,118]). There is little doubt that the expensive true-purple garments were used as a marker for social status, and had a role in establishing a

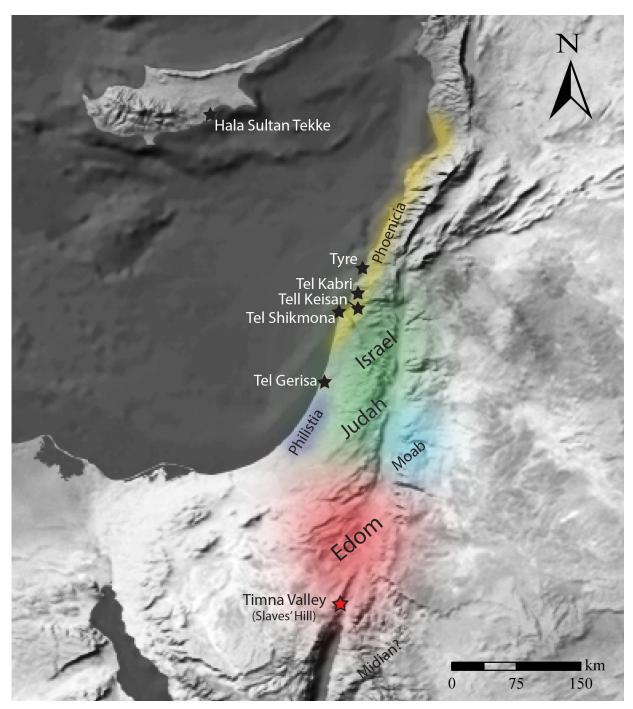


Fig 9. Location of "Slaves' Hill" (Timna Valley) and other iron age sites with evidence of the true purple industry. (Base map made with Natural Earth, free vector and raster map data @ naturalearthdata.com).

hierarchical social structure in the fledgling kingdom. In that regard, it is interesting to note the biblical description of the *kings* of Midian–a neighboring kingdom to Edom (although without archaeological identification)–who wore "purple [ארגמן] garments" (Judges 8:26). This reference is traditionally dated to the period under discussion, in which tribal nomadic societies consolidated into strong polities, typically without leaving any conspicuous

archaeological remains [121,122]. The uniqueness of the case of Edom is twofold, as it involves both an exceptionally rich archaeological record of nomads (the result of mining and smelting activities) and an unprecedented preservation of organic materials (the result of the extreme arid climate). The latter provides a window into various aspects of life of the early Edomites, and-by proxy—of other peoples and cultures of the broader region in this period.

The true-purple dyed textiles were found within industrial waste, in association with smelting activities. This further stresses previous observations on the important role of metalworkers within this society; the smelters, holding knowledge of one of the most sophisticated crafts of the time, were part of—or in close association with—the elite of the society, with access to excellent and exotic foods [120,123], and luxurious commodities, including those represented by the new finds. The new evidence also provides insights into elite "fashion"—in early Iron Age Timna and probably beyond. In addition to the decorations in red and blue dyes from plant sources [68], we can now conclude the use of decorations with the threads of the prestigious true purple. Colored clothing was the preference of elite in the Ancient Near Eastern societies [70]; yet, we know very little about the variety of decoration choices. In this study, sample no. 018, a decorated textile, and sample no. 017 that seems to be an element of a fringe or tassel, were probably part of a garment of a prestigious wardrobe [69,70]. It is interesting to note that in all known cases, the archaeological textiles with evidence of purple (from the Roman period) were not completely dyed with this expensive color, but incorporated purple threads as only one element of the textile weaving (usually in the weft), and sometimes only in un-woven decorative elements [124]. In most cases these textiles are very delicate and represent a high quality of weaving [15]. The correlation between high-quality textiles and expensive dyestuff is in accord with the textual evidence (see details in the Introduction above).

To conclude, this is the first time that a 3000 years old wool textile was found in the Southern Levant with evidence of shellfish-based dyes. This find helps researchers to reconstruct the complete *chaîne opératoire* of the true purple industry of the Iron Age, starting with the archaeological evidence of shell heaps in various sites along the Mediterranean coast, through finds of the dyeing vessels (stained ceramic basins), to the final product of textiles decorated with the most expensive dyestuff in the ancient world. Only three fragments survived, but they open a window into a wide range of social aspects, from the lives of Iron Age elite, to the means of establishing social status in the kingdoms of the region, including those based on nomads [121].

Supporting information

S1 Appendix. (DOCX)

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References

- Andersson Strand E, Frei KM, Gleba M, Mannering U, Nosch M, Skals I. Old Textiles–New Possibilities. Eur J Archaeol. 2010; 13: 149–173. https://doi.org/10.1177/1461957110365513
- Quillien L. The Economic Role of Coloured Textiles in Babylonia (1st Millennium BC). In: Thavapalan S, Warburton DA, editors. The Value of Colour: Material and Economic Aspect in the Ancient World. Berlin: Berlin Edition Topoi; 2019. pp. 201–230. https://doi.org/10.12968/prps.2008.1.89.38150
- Good I. Archaeological Textiles: A Review of Current Research. Annu Rev Anthropol. 2001; 30: 209– 226. https://doi.org/10.1146/annurev.anthro.30.1.209
- Ben-Yosef E. The Central Timna Valley Project: Research Design and Preliminary Results. In: Ben-Yosef E, editor. Mining for Ancient Copper: Essays in Memory of Professor Beno Rothenberg. Tel Aviv: Tel Aviv University. Institute of Archaeology; 2018. pp. 28–63.
- Ben-Yosef E. The Central Timna Valley Project (CTV). Available: <u>https://archaeology.tau.ac.il/ben-yosef/CTV/</u>.
- Workman V, Shamir O, Sukenik N, Gleba M, Ben-Yosef E. The Central Timna Valley Project: 5 Years of Ongoing Textile Research. ATR. 2017; 59: 67–70.
- Sukenik N. A 2000 Years Old Counterfeiting Industry. In: Meiri O, Bloch Y, Kaplan Y, editors. Out of the Blue. Jerusalem: Bible Lands Museume; 2018. pp. 100–113.
- Sukenik N, Iluz D, Varvak A, Amar Z. A Re-Evaluation of The Textile Dyes In The Cave Of Letters. In: Ortiz J, Alfaro C, Turell L, Martínez MJ, editors. Ancient Textiles and Dyes in the Ancient Mediterranean World. Valencia: Publicacions de la Universitat de València; 2016. pp. 263–273.
- 9. Plutarch A. Plutarch Lives (B. Perrin tran. 1959). Cambridge, Mass., London: Harvard University Press; W. Heinemann.
- 10. Cardon D. Natural Dyes, Sources, Tradition, Technology and Science. London: Archetype; 2007.
- 11. Reinhold M. History of Purple as a Status Symbol in Antiquity. Collect Latomus. 1970; 116: 1–73.
- 12. Ziderman I. "BA" Guide to Artifacts: Seashells and ancient purple dyeing. Biblic Archaeol. 1990; 53: 98–101.
- Alfaro Giner C. Purple and Aristocracy: Colour, Blood and Luxury as Social Identifiers in Antiquity. In: Alfaro Giner C, García JO, Martinez Garcia MJ, editors. luxury and Dress, Political Power and Appearance in the Roman Empire and its Provinces. Valencia: Universitat de valencia; 2013. pp. 75–98. https://doi.org/10.1017/CBO9781107415324.004
- Spanier E, Karmon N. Muricid Snails and the Ancient Dye Industies. In: Spanier E, editor. The Royal Purple and the Biblical Blue Argaman and Tekhelet. Jerusalem: Keter Publishing House; 1987. pp. 179–192.

- Marín-aguilera B, Iacono F, Gleba M. Colouring the Mediterranean: Production and Consumption of Purple-dyed Textiles in Pre-Roman Times. J Mediterr Archaeol. 2018; 31/2: 127–154.
- 16. Amar Z. The Argaman (Purple): Porphura and Arjawan in Jewish Sources and Further Inquiries into Tekhelet. Har Bracha. Har Bracha; 2014. (Hebrew).
- 17. Edmonds J. The Mystery of Imperial Purple Dye. Hist Dye Ser. 2002; 7: 1–41.
- 18. Boesken Kanold I. Tyrian Purple Dyeing: An experimental approach with fresh Murex trunculus. Dye Hist Archaeol. 2005; 20: 26–29.
- Koren ZC. New Chemical Insights into the Ancient Molluskan Purple Dyeing Process. ACS Symp Ser. 2013; 1147: 43–67. https://doi.org/10.1021/bk-2013-1147.ch003
- Spanier E. Cannibalism in Muricid Snails as a Possible Explanation for Archaeological Findings. J Archaeol Sci. 1986; 13: 463–468. https://doi.org/10.1016/0305-4403(86)90015-4
- Alfaro GC, Mylona D. Fishing for Purple Shellfish (Muricidae) in Ancient Greece: Acquisition Technology and First Steps in Purple Dye Production. In: Alfaro GC, Tellenbach y M, Ortiz J., editors. Production and Trade of Textiles and Dyes in the Roman Empire and Neighbouring Regions Producción (Purpureae vestes IV Textiles and Dyes in antiquity) Production). valència; 2014. pp. 149–233.
- 22. Aristotal. Historia Animalium, A.L. Peck (tran.). Cambridge; 1965.
- Koren ZC. The First Optimal All-Murex All-Natural Purple Dyeing in the Eastern Mediterranean in a Millennium and a Half. Dye Hist Archaeol. 2005; 20: 136–149.
- Li S, Cunningham AB, Fan R, Wang Y. Identity Blues: The Ethnobotany of the Indigo dyeing by Landian Yao (Iu Mien) in Yunnan, Southwest China. J Ethnobiol Ethnomed. 2019; 15: 1–14. https://doi.org/ 10.1186/s13002-018-0283-y PMID: 30611288
- Ruscillo D. Reconstructing Murex Royal Purple and Biblical Blue in the Aegan. In: Bar-Yosef Mayer D, editor. Archaeomalacology: Molluscs in Former Environment of Human Behavior. Oxford: Oxbow Books; 2005. pp. 99–106.
- Michel RH, McGovern PE. The chemical processing of Royal purple dye: ancient descriptions as elucidated by modern science. Archaeomaterials. 1987. pp. 135–143. Available: http://www.penn. museum/sites/biomoleculararchaeology/wp-content/uploads/2010/04/archeomat2.pdf.
- 27. Michel RH, McGovern PE. The Chemical Processing of Royal Dye: Ancient Descriptions as Elucidated by Modern Science, Part II. Archeomaterials. 1990; 4: 97–104.
- Elsner O, Spanier E. Dyeing with Murex Extracts. An Unusual Dyeing Method of Wool to the Biblical Sky Blue. Proc 7th Int Wool Text Res Conf. 1985; 118–130.
- 29. Stieglitz RS. The Minoan Origin of Tyrian Purple. Biblic Archaeol. 1994; 57: 46–54.
- Gillis C. The Color Purple. In: Faegersten F, Wallensten J, Östenberg I, editors. Tankemönster En Festskrift till Eva Rystedt. Lund: Fanni Faegersten; 2010. pp. 84–91.
- **31.** Reese DS. Palaikastro Shells and Bronze Age Purple-Dye Production in the Mediterranean Basin. Annu Br Sch Athens. 1987; 82: 201–206.
- Soriga E. Mari(ne) purple: western textile technology in Middle Bronze Age Syria. In: Landenius Enegren H, Meo F, editors. Treasures from The Sea: Sea Silk and Shellfish Purple dye in antiquity (Ancient Textiles Series 30). Oxford: Oxbow Books.; 2017. pp. 79–95.
- 33. Carannante A. Archaeomalacology and Purple-Dye: State of the art and New Prospects of Research. In: Cantillo JJ, Bernal D, Ramos J, editors. Molusco y púrpura en contextos arqueológicos atlánticomediterráneos: nuevos datos y reflexiones en clave de proceso histórico. Cádiz: Universidad de Cádiz; 2014. pp. 273–282. https://doi.org/10.1128/AAC.00103-14 PMID: 25070110
- Sotiropoulou S, Karapanagiotis I. Conchylian Purple Investigation in Prehistoric Wall Paintings of the Aegean Area. In: Meijer L, Guyard N, Skaltsounis L, Eisenbrand G, editors. Indirubin, the Red shade oF Indigo. Roscoff; 2006. pp. 71–78.
- Karapanagiotis I, Mantzouris D, Cooksey C, Mubarak MS, Tsiamyrtzis P. An Improved HPLC Method Coupled to PCA for the Identification of Tyrian Purple in Archaeological and Historical Samples. Microchem J. 2013; 110: 70–80. https://doi.org/10.1016/j.microc.2013.02.008
- Murphy E. Wool in the Nuzi Texts. In: Breniquet C, Michel C, editors. Wool Economy in the Ancient Near East and the Aegean (Ancient Textiles Series vol 17). Oxbow and Philadelphia: Oxbow Books; 2014. pp. 283–309.
- 37. Rendsburg GA. A Further Note on Purple Dyeing. Biblic Archaeol. 1991; 54: 121.
- Moran WL. The Amarna Letters. W L.M., editor. Baltimore and London: Johns Hopkin University Pr.; 1992.
- **39.** Astour MC. The Origin of the Terms "Canaan" "Phoenician", and "Purple." J Near East Stud. 1965; 24: 346–350.

- 40. Palaima TG. Maritime Matters in the Linear B Tablets. In: Laffineur R, Basch L, editors. Thalassa L'Egée Préhistoire et la Mer Actes de la Troisième Rencontre Egéenne Internationale de L'Université de Liège, Station de Recherches Sous-Marines et Océanographiques (StaReSO), Calvi, Corse (23–25 avril 1990). Liège; 1991. pp. 273–310.
- 41. Ventris MG., Chadwick J. Documents in Myceaean Greece. Cambridge; 1973.
- 42. Thavapalan S. Purple Fabrics and Garments in Akkadian Documents. J Anc Near East Hist. 2018; 3: 163–190. https://doi.org/10.1515/janeh-2017-0007.
- **43.** Ziderman II. Revival of Biblical Tekhelet Dyeing with Banded Dye-Murex (Ph. trunculus): Chemical Anomalies. Dye Hist Archaeol. 2001; 16/17: 87–90.
- 44. Ziderman II. The Biblical dye Tekhelet and its Use in Jewish Textiles. Dye Hist Archaeol. 2008; 21: 36–44.
- Sagiv G. Deep Blue: Notes on the Jewish Snail Fight. Contemp Jew. 2015; 35: 285–313. <u>https://doi.org/10.1007/s12397-015-9138-1</u>
- MacDonald N. The Priestly Vestments. In: Berner C, Schäfer M, Schott M, Schulz S, Weingärtner M, editors. Clothing and Nudity in the Hebrew Bible and the Ancient Near East. London: T&T Clark; 2019. pp. 435–448.
- 47. Holladay JS. Long-Distance Trade and the Wealth of Nations ca. 1000–600 B.C.: A New Perspective ("Poor Little [Agrarian] Judah" at the End of the Eighth Century B.C.: Dropping the First Shoe). In: Gitin S, Wright EJ, Dessel LP, editors. Confronting the Past: Archaeological and Historical Essays on Ancient Israel in Honor of William G Dever. Winona Lake, Indiana: Eisenbrauns; 2006. pp. 309–331.
- **48.** Plinius. Naturalis Historia (H. Rackham and W.H.S. Jones trans. 1963–1938). London: W. Heinemann.
- 49. Vitruvius. On Architecture, F. Granger (tran.) 1931–1934. Cambridge, Mass: Harvard University Press.
- Alfaro Giner C, Ribas BC. Methodological Aspects of Purple Dye Production on Ibiza: The New Site of Cala Olivera. In: Alfaro Giner C, Karali L, editors. Purpureae Vestes: Symposium Internacional sobre Textiles y Tintes del Mediterráneo en el mundo antiguo. 2008. pp. 195–208.
- Gillis C. The Color Purple. In: Faegersten F, Wallensten J, Östenberg I., editors. Tankemönster En Festskrift till Eva Rystedt. Lund; 2010. pp. 84–91.
- 52. Reese DS. Shells from Sarepta (Lebanon) and East Mediterranean Purple-Dye Production. Mediterr Archaeol Archaeom. 2010; 10: 113–141.
- Alberti ME. Murex Shells as Raw Material: the Purple-Dye Industry and its By-Products. Interpreting the Archaeological Record. Kaskal. 2008; 5: 73–90.
- Susmann NM. Preliminary Approaches for the Identification and Classification of Mediterranean Murex Dye Production Sites. Archaeol Text Rev. 2015; 57: 89–103.
- Alberti ME. Murex Shells as Raw Material: The Purple-Dye Industry and Its By-Products. Interpreting the Archaeological Record. Kaskal. 2008; 5: 73–90.
- Karmon N, Spanier E. Remains of a Purple Dye Industry Found at Tel Shiqmona. Isr Explor J. 1988; 38: 184–186.
- McGovern PE, Michel RH. Royal Purple Dye: Tracing Chemical Origins of the Industry. Anal Chem. 1985; 57: 1514A–1522A.
- Sukenik N, Iluz D, Amar Z, Varvak A, Bar S. New Evidence of the Purple-Dye Industry at Tel Shiqmona, Israel. Arcaeometry. 2017; 59: 775–785. https://doi.org/10.1111/arcm.12290
- 59. Koren ZC. The Unprecedented Discovery of The Royal Purple Dye On The Two Thousand Year-Old Royal Masada Textile. Am Inst Conserv Text Spec Gr Postprints. 1997; 7: 23–34. https://doi.org/10. 1017/CBO9781107415324.004
- **60.** Koren ZC. Dyeing with Sea Snail for the Production of Tekhelet and Argaman in Antiquity. In: Meiri O, Bloch Y, Kaplan Y, editors. Out of the Blue. Jerusalem: Bible Lands Museume; 2018. pp. 87–97.
- Sukenik N, Varvak A, Amar Z, Iluz D. Chemical Analysis of Murex-Dyed Textiles from Wadi Murabba'at, Israel. J Archaeol Sci Reports. 2015; 3: 565–570. https://doi.org/10.1016/j.jasrep.2015.08.003
- **62.** James MA, Reifarth N, Mukherjee AJ, Crump MP, Gates PJ, Sandor P, et al. High Prestige Royal Purple dyed Textiles from the Bronze Age Royal Tomb at Qatna, Syria. Antiquity. 2009; 83: 1109–1118. https://doi.org/10.1017/S0003598X00099397
- 63. Kolonas L, Sarri K, Margariti C, Vanden Berghe I, Skals I, Nosch M. Heirs from The Loom? Funerary Textiles from Stamna (Aitolia, Greece). A Preliminary Analysis. In: Fotiadis M, Laffineur R, Lolos Y, Vlachopoulos A, editors. The Aegean Seen from the West: Proceedings of the 16th International Aegean Conference, University of Ioannina, Department of History and Archaeology, Unit of Archaeology and Art History, 18–21 May 2016. Leuven: Peerers; 2017. pp. 533–544.

- Ben-Yosef E. Back to Solomon's Era: Results of the First Excavations at "Slaves' Hill" (Site 34, Timna, Israel). Bull Am Sch Orient Res. 2016; 376: 169–198.
- 65. Kleiman S, Kleiman A, Ben-Yosef E. Metalworkers' Material Culture in the Early Iron Age Levant: The Ceramic Assemblage from Site 34 (Slaves' Hill) in the Timna Valley. Tel Aviv. 2017; 44: 232–264. https://doi.org/10.1080/03344355.2017.1357313
- Ben-Yosef E, Liss B, Yagel OA, Tirosh O, Najjar M, Levy TE. Ancient Technology and Punctuated Change: Detecting the Emergence of the Edomite Kingdom in the Southern Levant. PLoS One. 2019; 14: 1–16. https://doi.org/10.1371/journal.pone.0221967 PMID: 31532811
- 67. Yadin Y. The Finds from the Bar Kokhba Period in the Cave of the Letters. Jerusalem; 1963.
- Sukenik N, Iluz D, Amar Z, Varvak A, Workman V, Shamir O, et al. Early Evidence (Late 2nd Millennium BCE) of Plant-Based Dyeing of Textiles from Timna, Israel. PLoS One. 2017; 12: e0179014. https://doi.org/10.1371/journal.pone.0179014 PMID: 28658314
- 69. Workman VA. The Fabric of Copper Prodection: The Textile and The Cordage Artifacts From Iron Age Timna. MA, Thesis. Tel-Aviv University. 2016.
- 70. Thomason A. Clothing and Nudity in the Ancient Near East: Iconographic and Archaeological Aspects. In: Berner C, Schäfer M, Schott M, Schulz S, Weingärtner M, editors. Clothing and Nudity in the Hebrew Bible and the Ancient Near East. London: T&T Clark; 2019. pp. 87–126.
- Bertman S. Tasseled Garments in the Ancient East Mediterranean. Amrican Sch Orient Res. 1961; 24: 119–128.
- 72. Sukenik N. A Linen Wick from the Northern Church at Shivta, Israel Naama. In: Motsianos I, Garnett KS, editors. Glass, Wax and Metal: Lighting Technologies in Late Antique, Byzantine and Medieval Times. Oxford: Archaepress Publishing; 2019. pp. 203–209. https://doi.org/10.2307/j.ctvndv5k4
- 73. Kvavadze E, Bar-Yosef O, Belfer-Cohen A, Boaretto E, Jakeli N, Matskevich Z, et al. 30,000-Year-Old Wild Flax Fibers. Science (80-). 2009; 325: 1359. <u>https://doi.org/10.1126/science.1175404</u> PMID: 19745144
- 74. Sheffer A, Granger-Taylor H. Textiles From Masada—A Prelminary Selection. In: Aviram J, Foerster G, Netzer E, editors. Masada IV. Jerusalem: Israel Exp.; 1994. pp. 153–256.
- 75. Shamir O, Baginski A. The Earliest Cotton Ikat Textiles from Nahal 'Omer Israel 650–810 CE. In: Nosch M-L, Feng Z, Varadarajan L, editors. Global Textile Encounters. Oxford & Philadelphia: Oxbow Books; 2014. pp. 65–73.
- 76. Shamir O, Baginski A. Textile Hoard from Jericho Cave 38 in the Qarantal Cliff. In: Guri-Rimon O, editor. Hoards and Genizot as Chapters in History. Haifa: Hecht Museum, University of Haifa; 2013. pp. 76*–87*.
- 77. Shamir O. Cotton textiles from the Byzantine period to the Medieval period in ancient Palestine. Rev d'ethnoécologie. 2019; 0–39. https://doi.org/10.4000/ethnoecologie.4176
- Shamir O. Textile Trade to Palestine in the Roman Period. In: Droß-krüpe K, Nosch M, editors. Textiles, Trade and Theories from the Ancient Near East to the Mediterranean. Münster: Ugarit-Verlag; 2016. pp. 231–245.
- 79. Brunello F. The Art of Dyeing in the History of Mankind. Hickey B (tran., editor. Vinenza; 1973.
- **80.** Hofenk de Graaff JH. The Colourful Past: Origins, Chemistry and Identification of Natural Dyestuffs. London: Archetype Publications; 2004.
- Clark RJH, Cooksey CJ, Daniels MAM, Withnall R. Indigo, Woad, and Tyrian Purple: Important Vat Dyes from Antiquity to the Present. Endeavour. 1993; 17: 191–199. <u>https://doi.org/10.1016/0160-9327</u> (93)90062-8
- Koren ZC. HPLC-PDA Analysis of Brominated Indirubinoid, Indigoid, and Isatinoid Dyes. In: Meijer L, Guyard N, Skaltsounis L, Eisenbrand G, editors. Indirubin, the Red shade of Indigo. Roscoff: Roscoff; 2006. pp. 45–53.
- Wouters J, Verhecken A. High-Performance Liquid Chromatography of Blue and Purple Indigoid Natural Dyes. J Soc Dye Color. 1991; 107: 266–269. https://doi.org/10.1111/j.1478-4408.1991.tb01351.x
- Koren ZC. Archaeo-Chemical Analysis of Royal Purple on a Darius I stone Jar. Microchim Acta. 2008; 162: 381–392. https://doi.org/10.1007/s00604-007-0862-4.
- 85. Taylor GW. Natural Dyes in Textile Applications. Rev Prog Color Relat Top. 1986; 16: 53–61.
- Koren ZC. An Efficient HPLC Analysis Scheme for Plant and Animal Red, Blue, and Purple Dyes. Dye Hist Archaeol. 1995; 13: 27–37.
- Koren ZC. Non-Destructive vs. Microchemical Analyses: The Case of Dyes and Pigments. Proceedings of ART2008, 9th International Conference, Non-Destructive Investigations and Microanalysis for The Diagnostics and Conservation of Cultural and Environmental Heritage. Jerusalem; 2008. pp. 37.1–37.10.

- Vajanto K, van Bommel MR. Dyed Textiles from Late Iron Age Finland. Fennoscandia Archaeol. 2014; XXXI: 61–78.
- **89.** Vasileiadou A, Karapanagiotis I, Zotou A. Determination of Tyrian Purple by High Performance Liquid Chromatography with Diode Array Detection. J Chromatogr A. 2016; 1448: 67–72. <u>https://doi.org/10.1016/j.chroma.2016.04.046</u> PMID: 27125189
- Pollard MUDO, Batt CUDB (Reino U, Stern BUDB (Reino U, Young SMM. UT (Estados U. Analytical Chemistry in Archaeology. Cambridge: Cambridge University Press; 2006. <u>https://doi.org/10.1017/ CBO9780511607431</u>
- Karapanagiotis I, Sotiropoulou S, Vasileiadou S, Karagiannidou E, Mantzouris D, Tsiamyrtzis P. Shellfish Purple and Gold Threads from a Late Antique Tomb Excavated in Thessaloniki. Arachne. 2019; 5: 64–77.
- Cooksey CJ. Tyrian Purple: 6,6'-Dibromoindigo and Related Compounds. Molecules. 2001; 6: 736– 769.
- Michel RH, Lazar J, McGovern PE. Indigotin Dyes in Peruvian and Coptic Textiles of the University Museum of Archaeology and Anthropology. Archeomaterials. 1992; 6: 69–83.
- 94. Sukenik N. Dyes in Textiles from the Early Roman Period Found in the Judean Desert Caves: Chemical, Historical and Archaeological Aspects. PHd, Thesis. Bar-Ilan University (Hebrew). 2013.
- Wouters J. A New Method for the Analysis of Blue and Purple Dyes in Textiles. Dye Hist Archaeol 10th Annu Meet. 1991; 17–21.
- Mantzouris D, Karapanagiotis I. Identification of Indirubin and Monobromoindirubins in Murex Brandaris. Dye Pigment. 2014; 104: 194–196. https://doi.org/10.1016/j.dyepig.2014.01.011
- 97. Karapanagiotis I. A Review on the Archaeological Chemistry of Shellfish Purple. Sustainability. 2019; 11. https://doi.org/10.3390/su11133595
- 98. Doumet J. Study of the Ancient Purple Colour. Beirut: Imprimerie Catholique; 1890.
- 99. Meiers F. Historical Outline and Chromatic Properties of Purpura Rubra Tarentina and its Potential Identification with Purple dye Extracted from Bolinus brandaris. In: Landenius Enegren H, Meo F, editors. Treasures from The Sea: Sea Silk and Shellfish Purple dye in antiquity (Ancient Textiles Series 30). Oxford: Oxbow Books; 2017. pp. 138–144.
- 100. Karmon N, Spanier E. Archaeological Evidence of the Purple Dye Industry from Israel. In: Spanier H, editor. The Royal Purple and the Biblical Blue Argaman and Tekhelet. Jerusalem: Keter Publishing House; 1987. pp. 147–158.
- 101. Born W. Purple in Classical Antiquity. Ciba Rev. 1937; 1/4: 111–118.
- 102. Jidejian N. Tyre: Through the Ages. Dar el-Mas. Beirut; 1969.
- **103.** Graves DE. What is the Madder with Lydia's Purple? A Reexamination of the Purpurarii in Thyatira and Philippi. Near East Archaeol Soc Bull. 2017; 62: 3–29.
- 104. Rusmore-Villaume ML. Seashells of the Egyptian Red Sea: The Ilustrated Handbook. Cairo: American University in Cairo; 2008.
- Bosch DT, Dance SP, Moolenbeek RG, Oliver PG. Seashells of Eastern Arabia. Dubai: Motivate publishing; 1995.
- **106.** Koren ZC, Verhecken-Lammens C. Microscopic and Chromatographic Analyses of Molluskan Purple Yarns in a late Roman Period Textile. e-PreservationSciences. 2013; 10: 27–34.
- 107. Ramasamy S. Incidence of Imposex in the Muricid Gastropod Chicoreus Virgineus from Tuticorin, Southeast Coast of India. J Mar Biol Assoc India. 2003; 45: 208–213.
- Schörle K. From Harbour to Desert: An Integrated Interface on the Red Sea and its Impact on the Eastern Egyptian Desert. Boll di Archeol line. 2010; 1: 44–53.
- 109. Cardon D, Nowik W, Granger-Taylor H, Marcinowska R, Kusyk K, Trojanowicz M. Who Could Wear True Purple in Roman Egypt? Technical and Social Considerations on Some New Identifications of Purple from Marine Molluscs in Archaeological Textiles. In: Alfaro GC, Brun J-P, Borgard PH, Pierobon Benoit R, editors. Purpureae Vestes IIi: Textiles y Tintes en la Ciudad Antigua. Valencia: Universitat de València—Centre Jean Bérard; 2011. pp. 197–214.
- Kalaitzaki A, Vafiadou A, Frony A, Reese DS, Drivaliari A, Liritzis I. Po-Pu-Re: Workshops, Use and Archaeometric Analysis in Pre-Roman Central Eastern Mediterranean. Mediterr Archaeol Archaeom. 2017; 17: 103–130. https://doi.org/10.5281/zenodo.160967
- 111. Verhecken A. Relation Between Age and Dyes of 1st Millennium ad Textiles Found in Egypt. In: de Moor A, Fluck C, editors. Methods of Dating Ancient Textiles of the 1st Millennium AD from Egypt and Neighbouring Countries. Tielt; 2007. pp. 206–213.

- 112. Kemp BJ, Vogelsang-Eastwood G. The Ancient Textile Industry at Amarna. Excavation memoir 68th. London: Egypt Exploration Society; 2001. Available: http://scholar.google.com/scholar?hl=en&btnG= Search&q=intitle:The+Ancient+Textile+Industry+at+Amarna.#0.
- 113. Pfister R. Les Textiles du Tombeau de Toutankhamon. Rev des arts Asiat. 1937; 11: 207–218.
- 114. Cline EH. 1177 B.C.: The Year Civilization Collapsed. Princeton: Princeton University Press; 2014.
- 115. Ben-Yosef E. Archaeological science brightens Mediterranean dark age. Proc Natl Acad Sci. 116: 5843–5845. https://doi.org/10.1073/pnas.1901885116 PMID: 30824590
- **116.** Levy TE, Higham T. The Bible and Radiocarbon Dating—Archaeology, Text and Science. London: Equinox Publishing Ltd.
- 117. Ben-Yosef E. A False Contrast? On the Possibility of an Early Iron Age Nomadic Monarchy in the Arabah (Early Edom) and Its Implications to the Study of Ancient Israel. In: Lipschits O, Sergi O, Koch I, editors. in press, From Nomadism to Monarchy? "The Archaeology of the Settlement Period" Thirty Years Later. Winona Lake.
- 118. Ben-Yosef E, Langgut D, Sapir-Hen L. Beyond Smelting: New Insights on Iron Age (10th C. BCE) Metalworkers Community from Excavations at A Gatehouse and Associated Livestock Pens in Timna, Israel. J Archaeol Sci Reports. 2017; 11: 411–426. https://doi.org/10.1016/j.jasrep.2016.12.010
- 119. Levy TE, Najjar M, Ben-Yosef E. New Insights into the Iron Age Archaeology of Edom, Southern Jordan. Los Angeles: Cotsen Institute of Archaeology Press, University of California; 2014.
- Sapir-Hen L, Ben-Yosef E. The Socioeconomic Status of Iron Age Metalworkers: Animal Economy in the 'Slaves' Hill ', Timna, Israel. Antiquity. 2014; 88: 775–790.
- Ben-Yosef E. The Architectural Bias in Current Biblical Archaeology. Vetus Testam. 2019; 69: 361– 387.
- **122.** Ben-Yosef E. forthcoming. Rethinking early Iron Age nomads. In: Garfinkel Y, Faust A, editors. State Formation Processes in the Early Iron Age Southern Levant.
- 123. Sapir-Hen L, Lernau O, Ben-Yosef E. The Diet of Ancient Metal Workers: The Late Bronze and Early Iron Ages in The Arabah Valley (Timna and Faynan). In: Ben-Yosef E, editor. Mining for Ancient Copper: Essays in Memory of Professor Beno Rothenberg (Monograph series of the Sonia and Marco Nadler Institute of Archaeology; no 37). Winona Lake, Indiana; Tel Aviv: Eisenbrauns; Emily and Claire Yass Publications in Archaeology; 2018. pp. 64–80.
- 124. Gleba M, Vanden Berghe I. Textiles from Strozzacapponi (Perugia/Corciano). New Evidence of Purple Production In Pre-Roman Italy. In: Alfaro Giner C, Ortiz-García J, editors. Production and Trade of Textiles and Dyes in the Roman Empire and Neighbouring Regions. valència; 2014. pp. 167–173.