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RESEARCH ARTICLE

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The "Flint Depot" of prehistoric northern Israel: Comprehensive geochemical analyses of flint extraction and reduction complexes and implications for provenance studies

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Abstract

Recent research has demonstrated that the Eocene Timrat formation in northeastern Israel, which appears as an extensive land "strip" west of and parallel to the Rift Valley, was a major source of prehistoric flint. This supposition is supported by three large-scale extraction and reduction (E&R) complexes identified within this region, which offer direct evidence of intense Lower and Middle Palaeolithic exploitation and limited Neolithic/Chalcolithic activities. Here, we present a first comprehensive overview of this "industrial strip" and of its E&R complexes (Nahal Dishon, Mt. Achbara, and Sede Ilan), demonstrating that these production areas were used mainly for the manufacture of large-volume items such as Lower Palaeolithic hand axes, Middle Palaeolithic Levallois cores, and Neolithic/Chalcolithic axes/adzes. Furthermore, we integrate information from recently published field studies and lithic analyses with new intercomplex and intracomplex inductively coupled plasma mass spectrometry (inductively coupled plasma (ICP)-MS) analyses of flint debitage. The relatively large number of analysed samples ($n = 69$) constitutes the first robust reference database for provenance studies of this E&R "strip." The potential contribution for provenance studies is demonstrated by a detailed ICP-MS comparison drawn between specific extraction and reduction localities within the Dishon complex and flint tools found in six occupation sites located up to 20 km from the sources. The detailed geochemical study also yielded methodological insights regarding challenges associated with flint heterogeneity and patination effects.

KEYWORDS

flint extraction and reduction, Galilee, inductively coupled plasma mass spectrometry, Palaeolithic, provenance studies

1 | INTRODUCTION

The Eastern Upper Galilee and the northern Jordan Valley host many important prehistoric sites, including the Lower Palaeolithic site of Gesher Benot Ya'aqov (Goren-Inbar et al., 2000), and the Middle Palaeolithic site of Nahal Amud Cave (Hovers, Rak, Lavi, & Kimbel, 1995; Suzuki & Takai, 1970; see Figure 1), and the archaeologically rich Neolithic/Chalcolithic sites of Beisamoun (Barkai, 2005; Bocquentin, Barzilai, Khalaily, & Kolska Horwitz, 2011; Lechevallier, 1978; Rosenberg, Assaf, Getzov, & Gopher, 2008) and Hagoshrim (Barkai, 2005, p. 76; Getzov, 2008; Rosenberg, Getzov, & Assaf, 2010; Rosenberg et al., 2008). All of these sites and many more (mentioned below) contain extensive flint assemblages; however, research efforts aimed at locating flint sources are inconclusive and only point to geological outcrops in the area (Delage, 1997, 2007a, 2007b). Recent research focusing on the Middle Palaeolithic Amud Cave provides important but geographically limited geochemical data of the Eocene flint within an ~8 km radius around the cave (Ekshtain, Ilani, Segal, & Hovers, 2016).

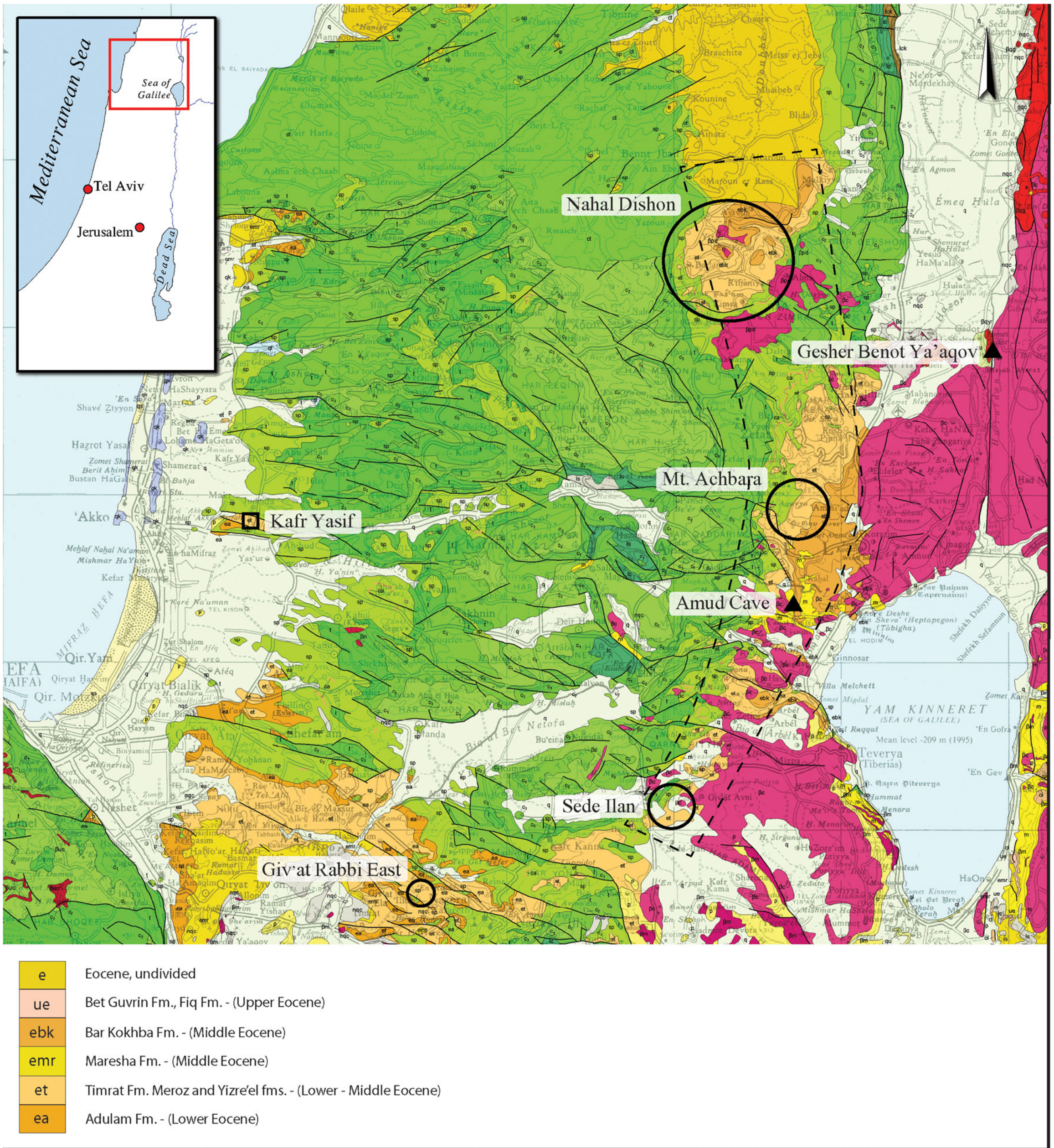


FIGURE 1 The geology of the Galilee and the East Eocene E&R "strip" (within the dashed line), with Nahal Dishon, Mt. Achbara, and Sede Ilan E&R complexes. The Giv'at Rabbi East E&R complex is also located within the Eocene formation. Kafr Yasif (in the square), a small outcrop of the Timrat formation, was sampled by Nathan et al. (1999) (Geological map: Sneh, Bartov, & Rosensaft, 1997). E&R: extraction and reduction [Color figure can be viewed at wileyonlinelibrary.com]

Recently, identified extensive open-air flint extraction and reduction (henceforth E&R) complexes (the term *complex* is used here to denote the entire area in which E&R activities took place; subareas are termed E&R *localities*) in the central Dishon Basin, the Eastern Galilee (Finkel, Gopher, & Barkai, 2016), and Mt. Achbara (Finkel, Gopher, Ben-Yosef, & Barkai, 2018) combined with data from the Sede Ilan E&R complex (Barkai & Gopher, 2009; Barkai, Gopher, & LaPorta, 2006; Figure 1) provide fresh insights on the origins of prehistoric Eocene flint in northern Israel. Lithic assemblages from tailing piles documented in field surveys of the Dishon complex (the largest and richest) indicate late Lower Palaeolithic and Middle Palaeolithic procurement (Finkel et al., 2016), and in a specific site (Mt. Reihan) a large Neolithic/Chalcolithic bifacial workshop has been found (Finkel, Gopher, Ben-Yosef, & Barkai, 2017). Findings from the Achbara complex attest mainly to Middle Palaeolithic activity and to a lesser extent to Neolithic/Chalcolithic activity (Finkel et al., 2018). The most southern and smallest complex in the "strip" is Sede Ilan with late Lower Palaeolithic and Middle Palaeolithic E&R activity (Barkai & Gopher, 2009; for an elaboration of E&R Phenomena see the paragraph on the Dishon E&R complex below). The following question is raised: Was the Eocene flint extraction and reduction "strip" of north Eastern Galilee the source for all of northern Israel (and possibly parts of southern Lebanon) in the Lower and Middle Palaeolithic, and later in the Neolithic and Chalcolithic periods when large-volume flint items were in demand? Identifying indicative characteristics of flint found in this "strip" may enable cross-referencing with flint artifacts in Palaeolithic and Neolithic/Chalcolithic occupation sites of the Galilee and provide answers to the above question. It is important to note that the E&R complex of Giv'at Rabbi in the Lower Galilee (Ekshtain, Barzilai, Inbar, Milevski, & Ullman, 2012; Yaroshevich, Shemer, Porat, & Roskin, 2017) is also situated within a flint bearing Eocene formation (Figure 1) and was recently suggested to constitute one of the sources of the Middle Palaeolithic Qafzeh Cave.

Inductively coupled plasma mass spectrometry (ICP-MS) and mostly nondestructive laser ablation (LA) ICP-MS have been used more frequently in flint provenance research (Pereira, Terrada, & Bicho, 2017) in recent years. The method is based on establishing the geochemical signature of flint outcrops for comparisons with the results of artifacts analyses of occupation sites. As ICP-MS can detect trace elements in very low concentrations (ppb-range) and is considered more accurate than the second most common analytic technique of X-ray fluorescence (Högberg, Hughes, & Olausson, 2012; Hughes, Högberg, & Olausson, 2010, 2012), its application to provenance research is indeed promising whereas presenting some methodological challenges (see Section 2).

1.1 | Flint in the Galilee

The current knowledge of flint sources in the Galilee is presented at three levels: (a) the flint bearing formations of the entire Galilee; (b) the Eocene Timrat formation "strip" of the Eastern Galilee; and (c) the northern part of the "strip"—the Dishon E&R complex. In relation to the latter, six occupation sites in the vicinity of the flint source, which were selected for a provenance study, are also described.

1.1.1 | Flint bearing geological formations of the entire galilee

There are several chert/flint-bearing geological formations in the Galilee (Figure 1). (Note that in research on this region both terms are used to describe the same thing. While Delage et al. [see below] preferred the term chert, we follow Ekshtain et al. [see below] and use the term flint.) They can be divided into two distinct groups that vary considerably in the quality of their flint as observed in the field (this major division is relevant to the entire southern Levant with some regional differences in field appearance and quality). Lower quality flint is found in Upper Cretaceous formations (cf., Delage, 2007a, Figure 1), including the Yagur formation (Cenomanian) with Kamon and Karkara members; the Deir Hanna formation (Upper/Late Cenomanian) with Ya'ara and Rosh members and the Sakhnin formation (Upper/Late Cenomanian); these are all characterized by relatively rare flint exposures of quite small nodules (e.g., 5–10 cm in dimension in the Yagur and Sakhnin formations and 10–20 cm in the Deir Hanna formation) of relatively poor quality flint. Upper Cretaceous formations also include the Yanuh formation (Upper/Late Cenomanian), which contains small patches of flint bearing rocks with very small nodules; the Yirka formation (Lower/Early Turonian), which for the most part lacks silicification; the Bi'na formation (according to Delage flint is almost "absent from the lower member of the Bi'na formation (Turonian), but some geologists mentioned their presence in the upper member" (Delage, 2007a, p. 38); and the Mishash formation (Campanian), which is composed predominantly of bedded chert but is present in the Galilee in only a few places as rather thin, easily extracted horizons of 10–20 cm thick.

Higher quality flint appears in the Eocene formations (Delage, 2007a). According to Delage (2007a, p. 39), "The Zor'a Formation (Lower Eocene) in [the] Galilee is characterized by an abundance of silicifications." This formation contains five different members all bearing flint, of which Member C holds the largest nodules of up to 50 cm in length (Delage, 2007a, p. 40; Figure 4). The entire formation is described as of high quality and high density and as easy to extract. Delage (2007a, p. 39) also notes that "Several major chert-bearing source areas stand out in the region: West Zomet Yasif, North Zomet Yasif, Nahal Evlayim (Western Galilee), and Nahal Dishon (Eastern Galilee)." On the Nahal Dishon area, Delage (2007a, p. 41) writes that "[this] mountainous area [...] is the main location of the Zor'a formation on the eastern slope of the Galilee. A wide range of siliceous rocks have been observed in this zone. Embedded in either limestone or chalk, chert nodules of various dimensions (up to 40 cm long) can be easily extracted. These rocks are not always suitable for flintknapping." Delage's reservations on the quality of Eocene flint in the Dishon area have been refuted by Finkel et al. (2016, 2017) who demonstrate its strong suitability for tool production. Another area of relatively broad exposure in this formation not surveyed by Delage is located around Mt. Achbara (Finkel et al., 2018). Furthermore, recent geological work has resulted in a more detailed documentation of the Eocene sedimentary sequence of the Galilee, and consequently the broadly defined Zor'a formation does not appear in new geological maps. Instead, five formations are identified (Sneh, Bartov, & Rosensaft, 1997), including Adulam (Lower Eocene), Timrat (Lower–Middle Eocene), Maresha (Middle Eocene), Bar Kokhba (Middle Eocene), and Beit Guvrin/Fiq (Upper Eocene). Uplifting and erosion that took place in the Galilee since the end of the Eocene removed Eocene layers from the Central Galilee (Figure 1; Segev, Schattner, & Lyakhovsky, 2011 and references therein), leaving small exposed patches of the Maresha and Adulam Eocene formations on the upper western side (Figure 1 and see the more detailed geological map, Sneh, 2004) and wider areas of Timart exposure in the Lower western Galilee (Sneh, 2008).

1.1.2 | The Eocene Timrat formation "strip" in the Eastern Galilee

A "strip" of Timrat and Bar Kokhba formations appears in the Eastern Upper and Lower Galilee. This south-to-north "strip" is 2–3 km wide in Sede Ilan, is 6–8 km wide around Mt. Achbara and the Dishon Stream and is wider in Lebanon (Figure 1 and see detailed maps—Levitte & Sneh, 2013; Sneh & Weinberger, 2006). In the detailed geological maps, the Timrat formation is described as bearing a large number of flint nodules (Figure 2) while the younger Bar Kokhba bears less. All of the recently found E&R areas are located on the Timrat formation while the nearby Bar Kokhba formation was found through intensive surveys conducted by our team to be almost flint-less (except in the vicinity of Amud Cave where relatively small nodules are abundant). Based on Delage's descriptions and the geological maps we suggest that Dalage's Zor'a C member bearing large flint nodules should be identified with the Timrat formation. It is also worth noting that although not on the "strip," the E&R complex of Giv'at Rabbi East is also located on the Eocene Timrat (and Adulam) formations (Ekshtain et al., 2012; Yaroshevich et al., 2017).

A detailed description of the geography and ecology of each of the E&R complexes along the Eastern Galilee Eocene "strip" is provided in previous publications (Finkel et al., 2016, 2018). General observations of the entire "strip" show that all areas form part of what botanists have termed the "Desert of the Galilee/The Arid Galilee" with relatively low vegetation coverage (Rabinovitch-Vin, 1986, p. 166). Additionally, two of the E&R complexes, the Dishon, and Achbara, share similar geomorphological and geographical features: they are both perched on high plateaus above the nearby Dishon and Amud Streams. General geological maps (Figure 1) indicate that the Eocene "strip" continues northward into Lebanon. Unfortunately, we could not find any detailed geological study of this area. However, based on the higher topographic elevation of Eocene outcrops there (in areas up to 900 m above sea level [masl], cf., 700 masl for the Dishon E&R complex) and similarities in geological structures, it is reasonable to assume that the younger Bar Kokhba formation dominates the landscape. Accordingly, therefore, the likelihood of finding any significant prehistoric E&R complexes there is rather low. That said, more cautious models for potential prehistoric flint procurement systems should consider the entire extent of this geological strip as a potential source of high-quality raw materials.

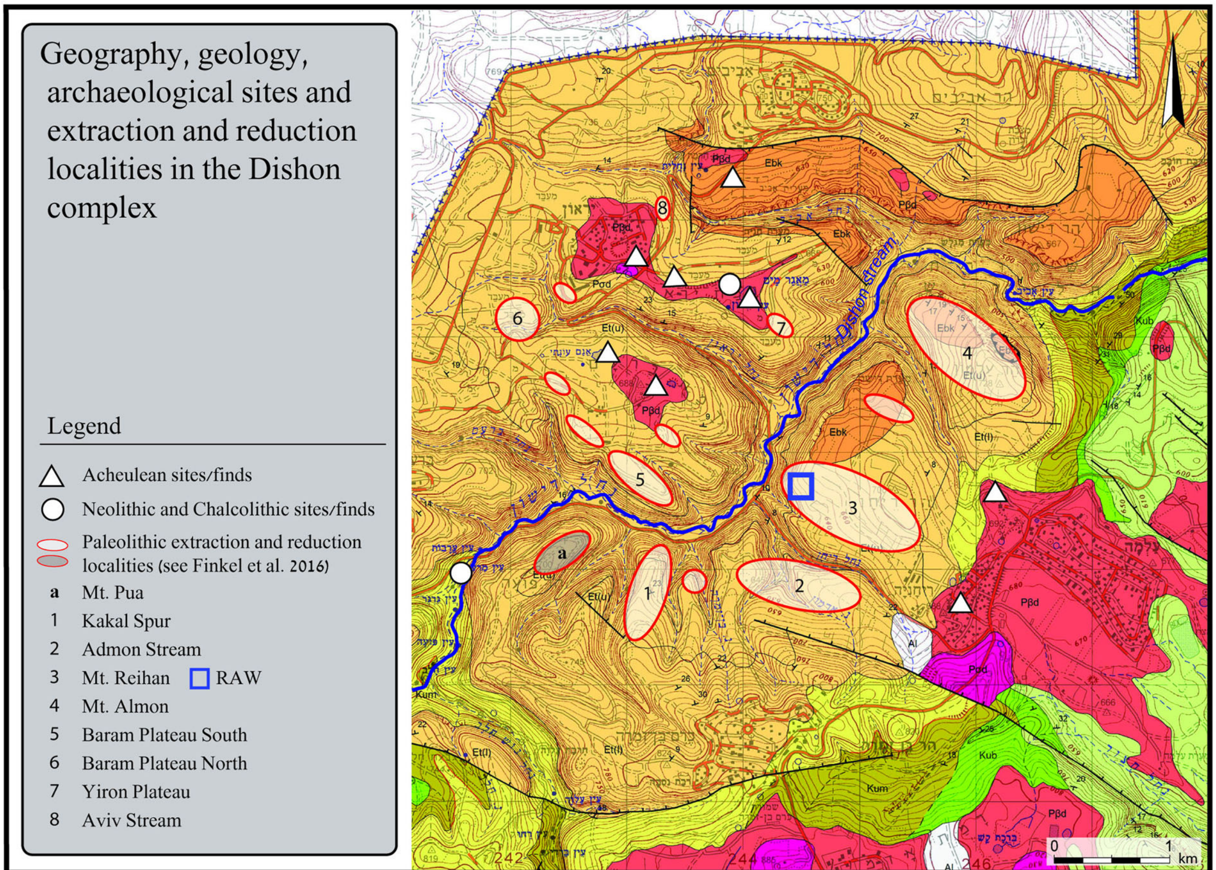
The geochemical characterization of flint from different formations of the Galilee is only in its early stages. Frachtenberg and Yellin (1992) presented a preliminary study of flint sources in Israel by neutron activation analysis, which covered two locations of the Lower Galilee. Nathan, Segal, and Delage (1999) presented the first ICP-MS and ICP-optical emission spectrometry (OES) geochemical study of flint in Israel. In this study, geological samples from six formations in the Western Galilee were analysed (Deir Hanna, Yanuh, Yirka, Zor'a, Bi'na, and Mishash) as well as archaeological samples from Hayonim Terrace (Epi-Palaeolithic Natufian culture). In a later geochemical study, Segal, Nathan, Zbenovich, and Barzilay (2005), correlated archaeological items from the Modi'in area (central Israel) with geological samples of the Mishash (Upper Cretaceous) and Zor'a (Eocene) formations, part of these drawn from the Galilee. Ekshtain 's et al. (2016) work on flint sources of the Amud Cave is the most relevant to our research. It focuses on the Eocene flint of the central Eastern Galilee (in close vicinity to the Amud Cave) and provides data concerning the Cenomanian flint of the central Galilee. The data presented here are the result of the first thorough ICP-MS analysis of flint from the Timart formation ($n = 69$ [total]). Contrary to previous research on the Galilee, which focused only on natural geological outcrops, we (based on a survey covering the Eocene formation outcrops of an extensive geographical area from close to the modern border between Israel and Lebanon in the north to the area west of the Sea of Galilee to the south representing ~90 sq. km of Eocene exposure) sampled flint only from contexts of prehistoric E&R complexes, all of which represented waste generated from quarrying activities.

In the following sections we will use the Dishon E&R complex as an example to illustrate characteristics of the E&R process, natural flint, and other features of the E&R phenomenon within the Eocene Timrat formation "strip."

1.2 | The Dishon E&R complex and the six occupation sites selected for provenance study

Nahal Dishon (the "Dishon Stream") is located on the western mountain flank of the Eastern Upper Galilee. The 96 sq km drainage basin runs 32 km east from Mt. Meron to the Hula Valley. The area is an erosive surface divided into mountainous plateaus at altitudes of 650–750 masl and peaks of up to 830 masl—the Baram, Yiron, Dalton, and Alma Plateaus (Figure 2). The surface is dissected by the Dishon Stream and its tributaries running between the plateaus at elevations of 500-400 masl in deeply incised, V-shaped gorges with occasional cliffs. Geologically, the study area consists of Eocene limestone and chalk. The Lower Eocene Timrat formation is 400 m thick and is characterized by limestone and chalk karrens which contain large numbers of flint nodules (Delage, 2007a; Levitte & Sneh, 2013). The Dishon area underwent erosion and by the Late Pleistocene reached an advanced stage that differs only slightly from that of the current topography (Yair, 1962, p. 195). For the purposes of our research (following M. Ohel, 1991, p. 161; Brosh & Ohel, 1981, p. 25), we assume that at the end of the Lower Palaeolithic and during the Middle Palaeolithic periods the topography of the plateaus was similar to that observed today although the depth of the incision of the Dishon Valley and its tributaries must have been shallower.

Recently, nine E&R localities were identified within the Dishon E&R complex, all within the Timrat formation's outcrops (Finkel et al., 2016). Among the localities, which were titled "Localities 1–8" and Mt. Pua (Figure 2; for the latter see, Barkai & Gopher, 2011; Barkai, Gopher, & LaPorta, 2002), we selected three for the geochemical study and defined them as sampling sites Bn, Bs, and R (Localities 6, 5, and 3, respectively, in Finkel et al., 2016). The chosen localities are positioned 1 to 2 km from each other on a west-northwest–east-southeast line that crosses the E&R complex. We provide a cross-section of the entire complex in Figure 3. Every sampling site includes a specific tailing pile. Although described in detail in our previous work, it is important to sketch the E&R phenomenon we are dealing with—not only in the Dishon, but along the entire Eocene Timrat formation "strip." All of these complexes are characterized by tailing piles purposely created in these industrial areas during long-term periods of flint extraction and reduction as part of the management of the extraction landscape (Gopher & Barkai, 2014). How were



STRATIGRAPHY

SYSTEM		SERIES - STAGE	SYMBOL	THICK. m	LITHOLOGY	LITHOSTRATIGRAPHY: MAPPING UNITS	GROUP
QUATERNARY		HOLOCENE	Al	2+		Alluvium, colluvium, soil	
TERTIARY	NEOGENE	PLIOCENE	Pαd			Dalton Scoria & Tuff	
			Pβd			Dalton Basalt	
	PALEOGENE	EOCENE	Ebk	100+		Bar Kokhba Formation	AVEDAT
			Et(u)	100-150		Timrat Formation Upper Memeber	
			Et(l)	100-200		Timrat Formation Lower Memeber	
		PALEOCENE	KTmgt	42-110		Mishash (chalk), Ghareb & Taqiye fms.	MOUNT SCOPUS
		SENONIAN	Kum: Kuma	61		Menuha Fm. – Ahihud Mbr. (incl. Kabri Marl)	
			Kum: Kumh			Menuha Fm. – Har Zefat Mbr.	
		TURONIAN	Kub	0-30		Bina Formation	
			Kuyi	0-20		Yirka Formation	
			Kuy	0-40		Yanuh Formation	
			Kusa	142		Sakhnin Formation	

FIGURE 2 Geography, geology, Palaeolithic, and Neolithic/Chalcolithic occupation sites, and E&R localities in the Dishon research area (Geological map: Levitte & Sneh, 2013). E&R: extraction and reduction [Color figure can be viewed at wileyonlinelibrary.com]

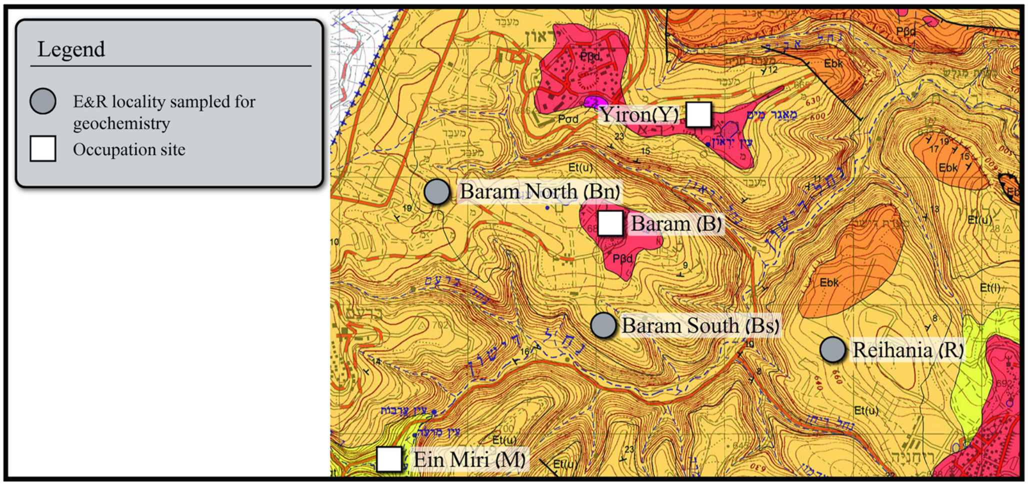


FIGURE 3 Three E&R localities (Bn, Bs, and R, cf., Localities 6, 5, and 3, respectively, in Figure 2) and occupation sites (Y, B, and M) in the Dishon E&R complex area. Bn: Baram Plateau North, Bs: Baram Plateau South, R: Mt. Reihan. B: Baram; Bn: Baram north; Bs: Baram south; E&R: extraction and reduction; M: Ein Miri ; R: Mt. Reihan; Y: Yiron [Color figure can be viewed at wileyonlinelibrary.com]

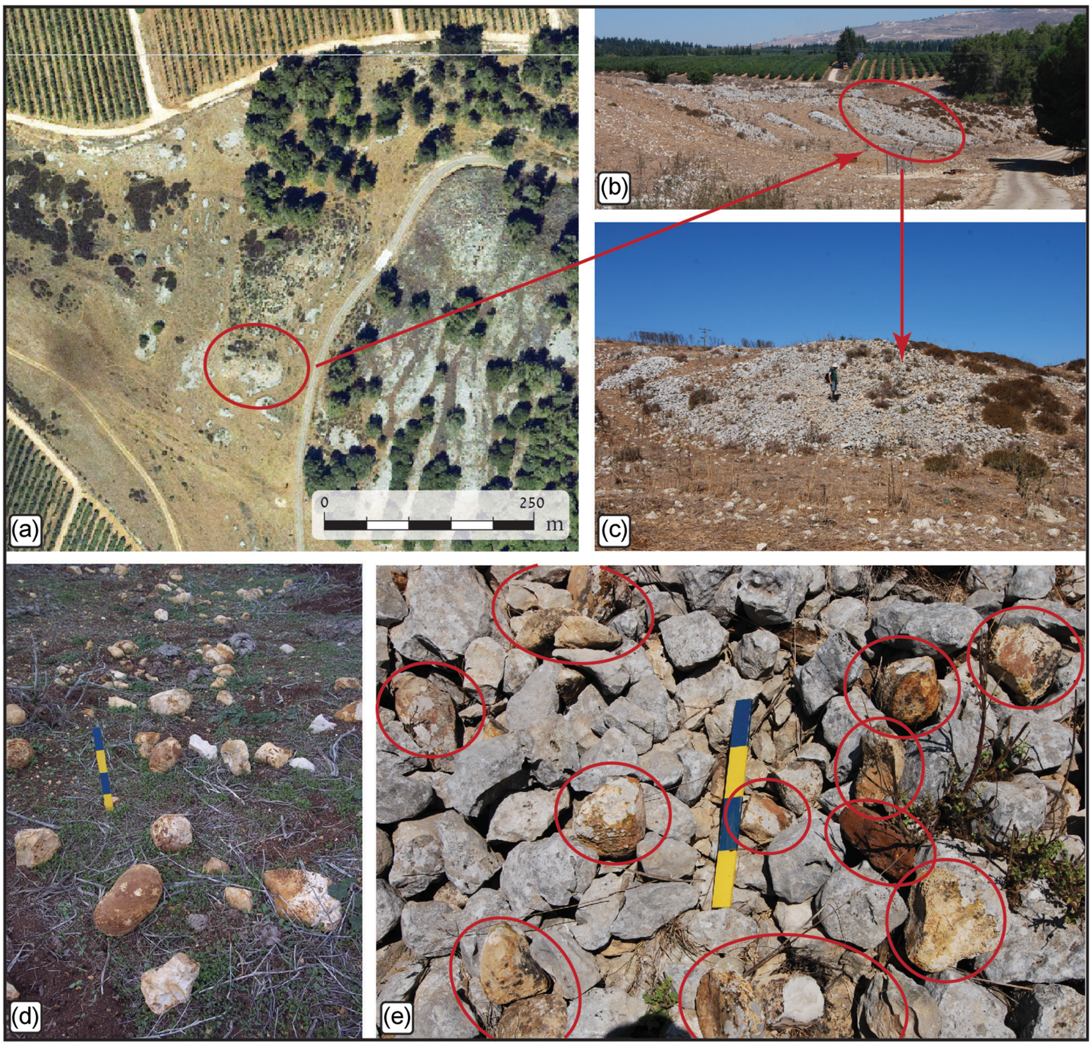


FIGURE 4 (a) Aerial photo of extraction and reduction locality No. 6—Baram Plateau North (the circle marks the surveyed pile); (b) ground photo from the south (the circle marks the surveyed pile); (c) close-up image of the surveyed pile; (d) flint nodules near the surveyed pile; (e) flint (circled) and limestone on the surveyed pile [Color figure can be viewed at wileyonlinelibrary.com]

these complexes actually formed? Surveys of E&R localities conducted in recent years combined with data from small-scale excavations conducted in tailing piles of Mt. Pua and Sede Ilan (Barkai & Gopher, 2009; Barkai et al., 2002, 2006; Gopher & Barkai, 2006, 2014) suggest the following sequence of operations:

- locating specific desired flint extraction fronts;
- extracting flint sometimes by employing limestone or basalt tools;
- creating stone waste piles (backfill piles) from large amounts of broken limestone during or immediately after flint extraction;
- aligning backfill piles on top of and between exhausted extraction fronts, leaving unexploited flint extraction fronts free for further use; and
- flint knapping—core shaping for later use; blank production and/or tool making conducted over top of backfill piles that form the tailing piles known today (e.g., in Mt. Pua [the Dishon complex]—a $2 \times 2\,m^2$ excavation in pile PW3 yielded 1,146 flint items; In Sede Ilan, a $2 \times 2\,m^2$ excavation in pile SE3 yielded 480 flint items (Barkai et al., 2006).

Similarities between the Dishon, Mt. Achbara, and Sede Ilan complexes in geology, topography, and major E&R features—i.e., tailing piles—enable us to use those limited excavations and a wide array of surface observations to suggest the abovementioned process.

The tailing piles at those sites vary in size from small (1–2 m in diameter and up to 1 m in height) to large (tens of m in diameter and 3–5 m in height; see Figure 4 for an illustration). It is important to note that the quantities of lithic materials are massive (an average of 286.5 artifacts in $1\,m^3$ in tailing pile PW3 [350 sq. m] and 441 artifacts in $1\,m^3$ in tailing pile PW100 [20 sq. m] at Mt. Pua—arkai et al., 2002), and further fieldwork must be planned accordingly; if this is not conducted, storage facilities will be overwhelmed by lithic material originating from extraction sites and workshops (see Gopher and Barkai, 2014, for further discussion; and Elston, 1992 and McBryde, 1984, for information on similar ultrarich extraction and reduction complexes). The piles are concentrated within relatively restricted areas, which creates a highly visible extraction landscape that appears as an artificial (usually conspicuous) mark on the landscape.

There are two main causes of the minimal postdepositional processes observed along the E&R "strip." The rugged karstic landscape combined with grassland vegetation of the area termed by botanists as the "Arid Galilee" (rather than common oak forests of the broader region) due to the local soil's limited water carrying capability (Rabinovitch-Vin, 1986) explain why the area was used in history mainly for pastoral agriculture and not for plant cultivation, which would heavily damage the E&R complexes. Natural post-depositional processes were minimal due to the plateau/moderate slope topography, which characterize the three E&R complexes. As the tops of the piles are positioned 1–5 m above the surrounding ground, we can deduce that all flint artifacts found on their surfaces are in situ (i.e., they were not washed downhill). That said, a better understanding of postdepositional processes will hopefully be gained through future excavations of various contexts at these sites. One of the most common features present is flint-knapping waste with flint artifacts found on the surfaces of and within the piles. Basalt wedges, probably used to enlarge natural fissures in the limestone karrens, are also found on and within the tailing piles and sometimes in considerable numbers (e.g., at Sede Ilan, Lower Galilee, Barkai et al., 2006) but also in the Dishon and Achbara (Finkel et al., 2016, 2018, respectively). The caching of flint artifacts underneath one of the piles has been reported at Mt. Pua (Barkai & Gopher, 2011).

We note above that we prefer to assign Delage's Zor'a formation Member C to the Timrat formation. This flint was characterized by Delage as bearing the best combination of characteristics for flint knapping (as we suggest—aimed at high volume artifacts), that is high knapping quality; 5–50 cm in dimension, high density within outcrops, and ease of extraction (Delage, 2007a, p. 54: appendix 1; see also Yaroshevich et al., 2018, for a description of the Givat Rabi East E&R site (Figure 1) situated at the contact point between the Adulam formation with the Timrat formation as bearing high quality "flint nodules, 5–50 cm in diameter, finely crystalline, with homogenous texture […] but difficult to extract"). In Finkel and Gopher (2018, see figures within) we focused our research on nodule sizes in the Dishon E&R complex. We measured 50 loose nodules in Locality 3 (Mt. Reihan —R, see Figure 2), resulting on average flint nodules of 17.7 kg. We measured 20 nodules in Locality 2 and Baram north (Bn) in the Dishon complex (see Figure 2) with similar results (average nodule weights of 20.1 and 17.7 kg, respectively, see some of the nodules measured in Figure 4d). This indicates that in terms of size, the Eocene flint of the Timrat formation is suitable for the production of high volume artifacts. Nodules at the Mt. Achbara E&R complex look the same as those in the Dishon (every E&R location's description for the Achbara, Finkel et al., 2018, and Dishon, Finkel et al., 2016, complexes include a figure of natural nodules found in its vicinity). Regarding visual characteristics of the flint, to the naked eye knapped flint items found in the Dishon and Achbara E&R complexes look very similar; they have a homogenous texture, beige-gray coloring, and varied patination of mainly gray and yellow. This can be seen when observing rejected tools found on E&R piles (Figure 5—all patinated to various extents; Item 3 is extraordinary with its reddish patina), flint debitage from E&R piles (Supporting Information Figure 1, 30 items and Supporting Information Figure 2) and more than 90 rejected tools and cores presented in figures of our previous works (Finkel et al., 2016, 38 items [some with stronger signs of patination with breaks revealing a beige-gray color]; Finkel et al., 2017, 29 items; Finkel et al., 2018, 26 items) and compared with flint tools from a few sites, which have similar attributes and which were, therefore, chosen for geochemical analysis (Figures 6 and 13). This visual gray patination is also visible in Ekshtain's "debitage classified as Eocene" from the Amud Cave (Ekshtain et al., 2016; Figure 5, artifacts a–d). Yaroshevich et al. (2018) described the Givat Rabi East E&R site flint as having "a gray-beige shade, often having orange-pink concentric veins." Ekshtain described the Timrat flint north of the Amud Cave (in the vicinity of our Achbara E&R complex) as gray with a few variations (Ekshtain et al., 2016; Supporting Information Table 3, Location 31). It is important to note that based on these common textures and colors,

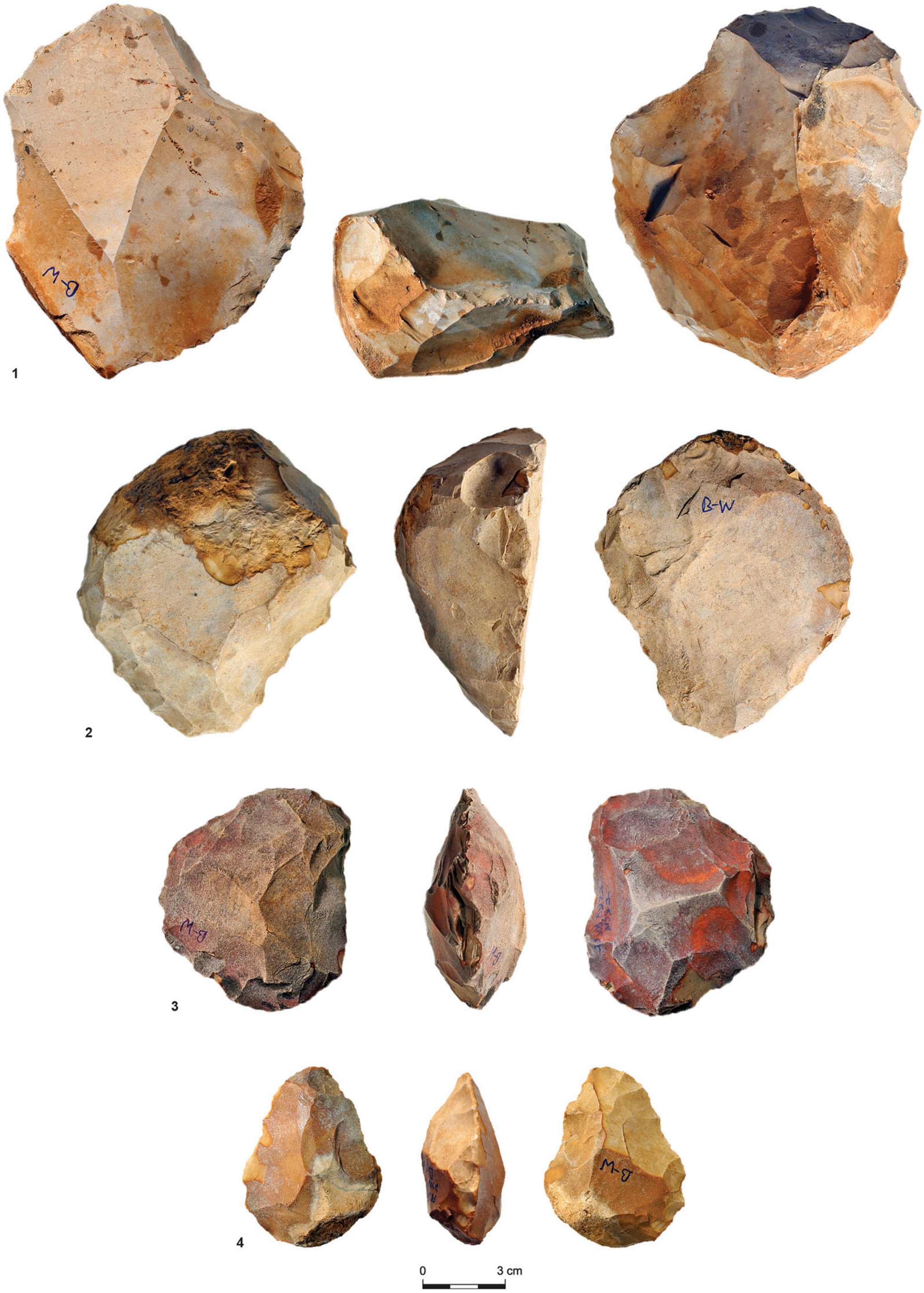


FIGURE 5 Artifacts from the surveyed pile (Figure 4c). (1) Levallois core; (2) core; (3, 4) biface [Color figure can be viewed at wileyonlinelibrary.com]

many researchers in Israel have attributed the flint items they have found to the Eocene without conducting any geochemical or in-depth visual research. Of the abundant examples of this practice it is worth mentioning a few from northeastern Israel: Ma'ayan Baruch (Hamara; Ronen, Ohel, Lamdam, & Assaf, 1980, p. 19; Stekelis & Gilead, 1966, p. 7), Beisamoun (Khalaily, Kuperman, Marom, Milevski, & Yegorov, 2015, p. 12), and Rasem Harbush (Noy, 1998, pp. 270–271). Examples of analytic data in support of the visual link in this region include those for the Amud Cave (Ekshtain et al., 2016) and 'Ein Qashish (Ekshtain, Malinsky-Buller, Ilani, Segal, & Hovers, 2014).

The following is a description of these localities and piles:

- Sampling tailing pile R (within E&R Locality No. 3 in Figure 2; Figures 3 and 6): This pile is named for Mt. **R**eihan located on the eastern bank of Nahal Dishon and flanked by the Reihan Stream to the south and Mt. Almon to the north. The entire locality covers a

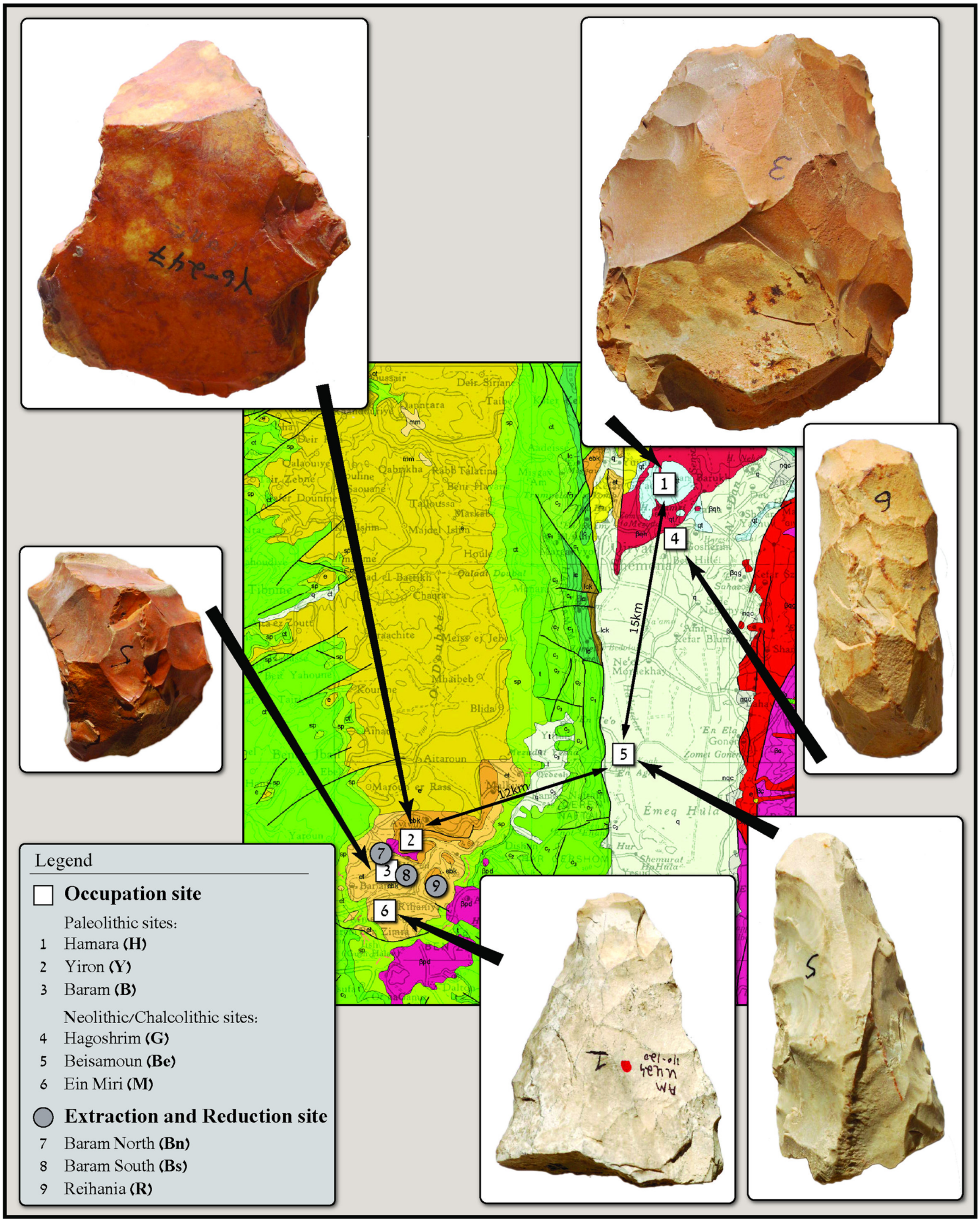
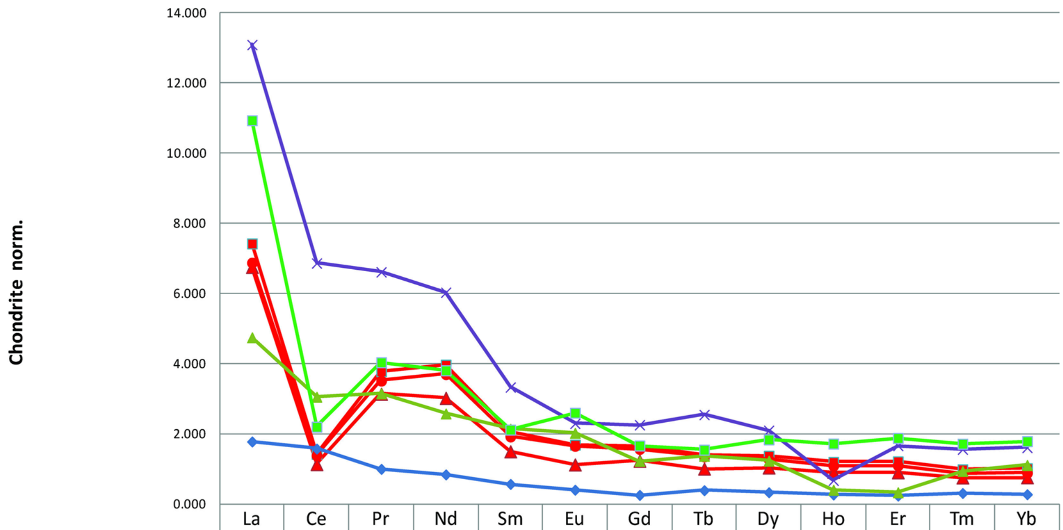


FIGURE 6 The three E&R localities in the Dishon area and the six occupation sites sampled for geochemical analysis with an example of one artifact from each site (Geological map: Sneh, Bartov, & Rosensaft, 1997). E&R: extraction and reduction [Color figure can be viewed at wileyonlinelibrary.com]

2 × 1 km area that slopes gradually from 700 masl in the northeast to 610 masl in the southwest. It includes three areas of large tailing piles with virtually none that are moderately sized or small. Areas in the center and to the east of the locality are characterized according to lithic technology as Late-Lower/Middle Palaeolithic. The most western area of piles is different and constitutes part of a 40,000 sq. m Neolithic/Chalcolithic Bifacial workshop (Finkel et al., 2017). Flint debitage used for the ICP-MS analysis was taken from the largest E&R pile (RAW 100; see in Finkel et al., 2017). Artifacts from the surveyed pile include mainly adze and axe bifacial roughouts and Levallois cores attesting to human activity in the Middle Palaeolithic and Neolithic/Chalcolithic.

- Sampling tailing pile Bs (within E&R Locality No. 5 in Figure 2; Figures 3 and 6): This pile is named for the **B**aram Plateau (**S**outh)



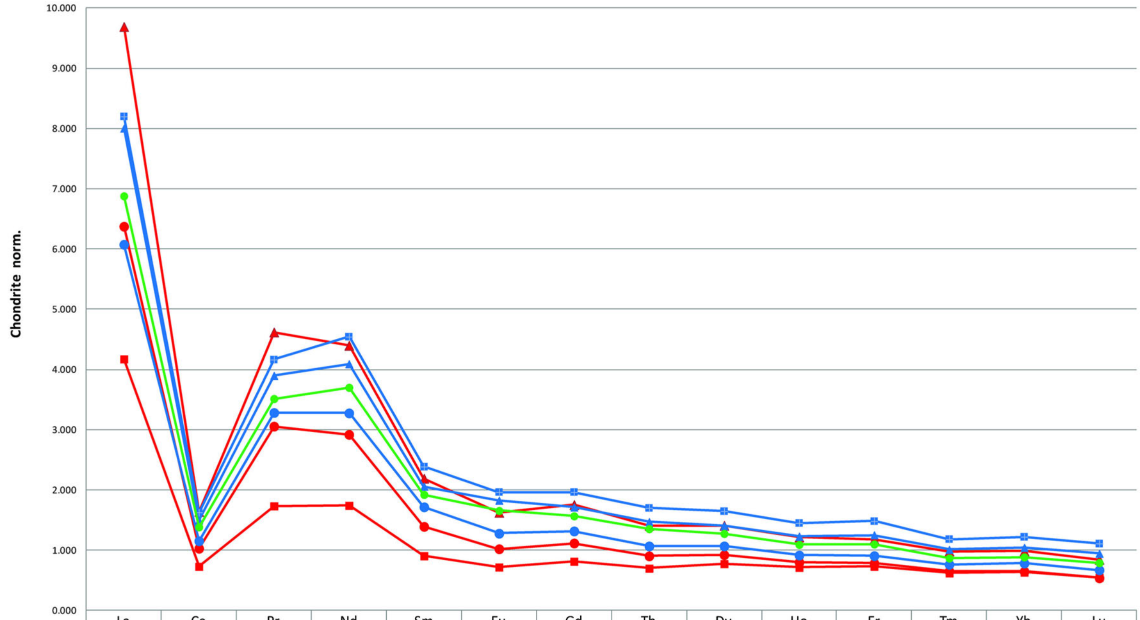
	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb
Timrat: Dishon	6.758	1.138	3.139	3.024	1.501	1.126	1.233	1.009	1.040	0.915	0.904	0.756	0.760
Timrat: Achbara	7.427	1.428	3.783	3.971	2.055	1.691	1.666	1.414	1.375	1.200	1.215	0.987	1.018
Timrat: Sede Ilan	6.880	1.387	3.515	3.701	1.924	1.657	1.569	1.356	1.276	1.094	1.101	0.869	0.888
Deir Hanna	1.781	1.578	1.000	0.842	0.571	0.405	0.258	0.392	0.333	0.270	0.238	0.313	0.278
Yanuh	4.750	3.056	3.154	2.579	2.143	2.027	1.226	1.373	1.233	0.405	0.333	0.938	1.111
Yirka	13.094	6.867	6.615	6.035	3.333	2.297	2.258	2.549	2.100	0.676	1.667	1.563	1.611
Zor'a	10.938	2.203	4.038	3.811	2.119	2.601	1.667	1.548	1.842	1.723	1.881	1.719	1.792

FIGURE 7 A comparison of REE concentrations (after chondrite normalization) in flint from main flint-bearing formations of the Galilee. Data for the Timrat formation are the result of the current study; they are presented separately for each of the three main E&R complexes of the Eastern Galilee (Dishon, *n* = 29; Achbara, *n* = 30; Sede Ilan, *n* = 10). Data for the Zor'a formation (from a natural outcrop near Kafr Yasif identified as the Timrat formation in new maps, see Figure 1) are taken from Nathan et al. (1999) excluding an outlier (Sample 4-BG-13, following advice by C. Delage). Data for the Deir Hanna, Yanuh and Yirka formations (Cenomanian–Turonian) are drawn from Nathan et al. (1999). E&R: extraction and reduction; REE: rear earth element [Color figure can be viewed at wileyonlinelibrary.com]

located on the western bank of Nahal Dishon. The entire locality is a spur on the eastern edge of the plateau 1-km long and 300-m wide sloping slightly from 650 masl in the northwest to 620 masl in the southeast. Several hundreds of medium-sized tailing piles were found in this locality (covered by pine trees). Flint nodules were found embedded within the limestone near the piles. Artifacts from the sampled pile include probable Levallois cores, bifacial rough-outs, and adze roughouts (possibly Chalcolithic), attesting to human activity during the Lower and Middle Palaeolithic and possibly, to a much lesser extent, in the Neolithic/Chalcolithic (Finkel et al., 2016, p. 236 and figures).

- Sampling tailing pile Bn (within E&R Locality No. 6 in Figure 2; Figures 3 and 6). This pile is named for the Baram Plateau (north) located on the western bank of Nahal Dishon. The locality, placed on the northwest part of the Baram Plateau, is 500 × 500 m in extent and is at an elevation of 675 masl. It is situated in a shallow upper basin of one of the tributaries of the Yiron Stream and includes 10 large or elongated tailing piles interspersed by a few medium-sized tailing piles. Flint nodules were found on the ground near the piles (Figure 4d). Artifacts from the sampled pile include cores, Levallois cores, bifacial roughouts, and bifaces, attesting to human activity during the Lower Palaeolithic (Acheulean) and Middle Palaeolithic (Mousterian) periods (Figure 5, Finkel et al., 2016, p. 238). In complementing the detailed geochemical study of the three localities

in the Dishon E&R complex, we also sampled six prehistoric occupation sites (Figure 6). Three sites are located in the immediate vicinity of these flint sources and include the Lower Palaeolithic Baram and Yiron sites, the Neolithic/Chalcolithic site of Ein Miri (see also Figure 3), and three others in the Hula valley (located up to 20 km from the Dishon E&R complex), including the Lower Palaeolithic Ma'ayan Barukh (Hamara) and the Neolithic sites of Beisamoun and Hagoshrim (Figure 6). These sites were chosen because of the relative abundance of bifacial tools retrieved from each (thousands; Barkai, 2005), which not only testifies to their important role in the lives of the sites' inhabitants, but which also allowed us to conduct a destructive analysis of single archaeological tools (complete and broken). The three occupation sites in the Dishon Basin were discovered as part of previous studies on the prehistory of the region that demonstrate a rich occupation history and a variety of site types. Regarding earlier research, it is important to note Turville-Peter et al.'s (1927) identification of a "factory" with bifaces in the Baram Plateau and Ronen's 1970s surveys and Ohel's of the 1980s. These surveys discovered Acheulean sites on the basalt caps of the Yiron, Baram, and Riehania Plateaus (M. Ohel, 1986a, 1986b, 1991; Ronen, Gilead, Bruder, & Meller, 1974). Earlier archaeological studies regarding the Neolithic and Chalcolithic periods for the Nahal Dishon Central Basin include Chalcolithic open-air sites on the Yiron Plateau (see Figure 3), where a few adzes and chisels were found (Khalaily,



	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
Dishon Bn	9.689	1.641	4.619	4.399	2.191	1.626	1.755	1.412	1.410	1.217	1.182	0.983	0.984	0.846
Dishon Bs	4.172	0.730	1.734	1.743	0.906	0.719	0.815	0.700	0.774	0.714	0.732	0.622	0.636	0.548
Dishon R	6.375	1.031	3.057	2.920	1.394	1.022	1.118	0.904	0.924	0.801	0.785	0.652	0.648	0.540
Achbara E	8.003	1.501	3.899	4.094	2.059	1.827	1.715	1.472	1.407	1.229	1.253	1.011	1.046	0.947
Achbara S	8.201	1.627	4.165	4.542	2.387	1.965	1.966	1.701	1.648	1.453	1.483	1.184	1.223	1.110
Achbara W	6.075	1.155	3.284	3.277	1.718	1.280	1.318	1.069	1.069	0.918	0.908	0.765	0.785	0.672
Sede Ilan	6.880	1.387	3.515	3.701	1.924	1.657	1.569	1.356	1.276	1.094	1.101	0.869	0.888	0.792

FIGURE 8 A comparison of REE concentrations (after chondrite normalization) in flint from seven E&R localities from the three E&R complexes (for each $n = 10$, except for R [$n = 9$]; cf., Figure 1). All samples represent the Eocenian Timrat formation. E&R: extraction and reduction; REE: rear earth element [Color figure can be viewed at wileyonlinelibrary.com]

Marder, & Shaked, 2000), Nahalit Cave (see Figure 2; Frankel, Getzov, Aviam, & Degani, 2001, pp. 41–42 and 96–97), Riehania Pool (Shalem, 2008), and a cave designated as Aviv_12, where an adze was found (Ullman, 2014). The three occupation sites in the Dishon Basin, sampled as part of the current study, include:

- The Yiron Site ("Y" in Figures 3 and 6): The site was discovered by Ohel in his 1979 survey of the area of the Yiron Plateau. Ohel described nine Acheulean sites (most of them located on the basalt cap and some 200–300 m from the basalt cap boundaries; M. Ohel, 1986a). In this study, we used samples from three different locations examined in the framework of Ohel's survey (see Table 1).
- The Baram Site ("B" in Figures 3 and 6): The Baram Plateau survey discovered three sites around water bodies on the basalt cap (M. Ohel, 1991): two Acheulean sites from which altogether just over 500 hand axes were retrieved (M. Ohel, 1991, p. 77) and one Mousterian site. In this study, we sampled items from one of the Acheuleans sites (see Table 1).
- Ein Miri ("M" in Figures 3 and 6): The site of Ein Miri contains EpiPalaeolithic and Neolithic/Chalcolithic materials (Shimelmitz, Barkai, & Gopher, 2004) and is situated on the Dishon Valley floor. Findings from this site include a significant number of axes with smaller numbers of adzes and chisels (Yerkes & Barkai, 2013). Prausnitz (1959) identified this site as Khirbet Kharruba and dated it to the Neolithic/Chalcolithic. Neolithic flint extraction sites are located in rock shelters and karstic cavities on the Dishon Valley floor (Gopher & Barkai, 2006) near the Ein Miri site. In this study we sampled broken axes and retouched flakes from lithic assemblages retrieved from an excavation carried out in 2001.

The three sites in the Hula Valley sampled as part of the current study are key sites of the Neolithic/Chalcolithic and Lower Palaeolithic archaeology of the entire region. They include:

- Ma'ayan Barukh/Hamara ("H" in Figure 6 located 20 km northeast of the Dishon complex): A broad, open Lower Palaeolithic site situated in the northern part of the Hula Plain (Stekelis & Gilead, 1966) where by the end of the 1970s approximately 6,000 hand axes had been found (Ronen et al., 1980). The greyish flint was assumed by the authors to have originated from Eocene outcrops, and they identified its source ~6 km north of the site in modern-Day Lebanon (on current geological maps it is located 9 km away). This identification was based on proximity; however, as we note above, it is most likely that the Eocene formation exposed there is the flintless Bar Kokhba formation. There is also a narrow strip of Eocene limestone 4 km to the east (Figure 6), but a field survey conducted by our team demonstrates that it does not contain a significant flint source. In this study, we sampled broken hand axes.
- Beisamoun ("Be" in Figure 6): This is a Prepottery Neolithic B and a Pottery Neolithic village site located near the northwestern shore of

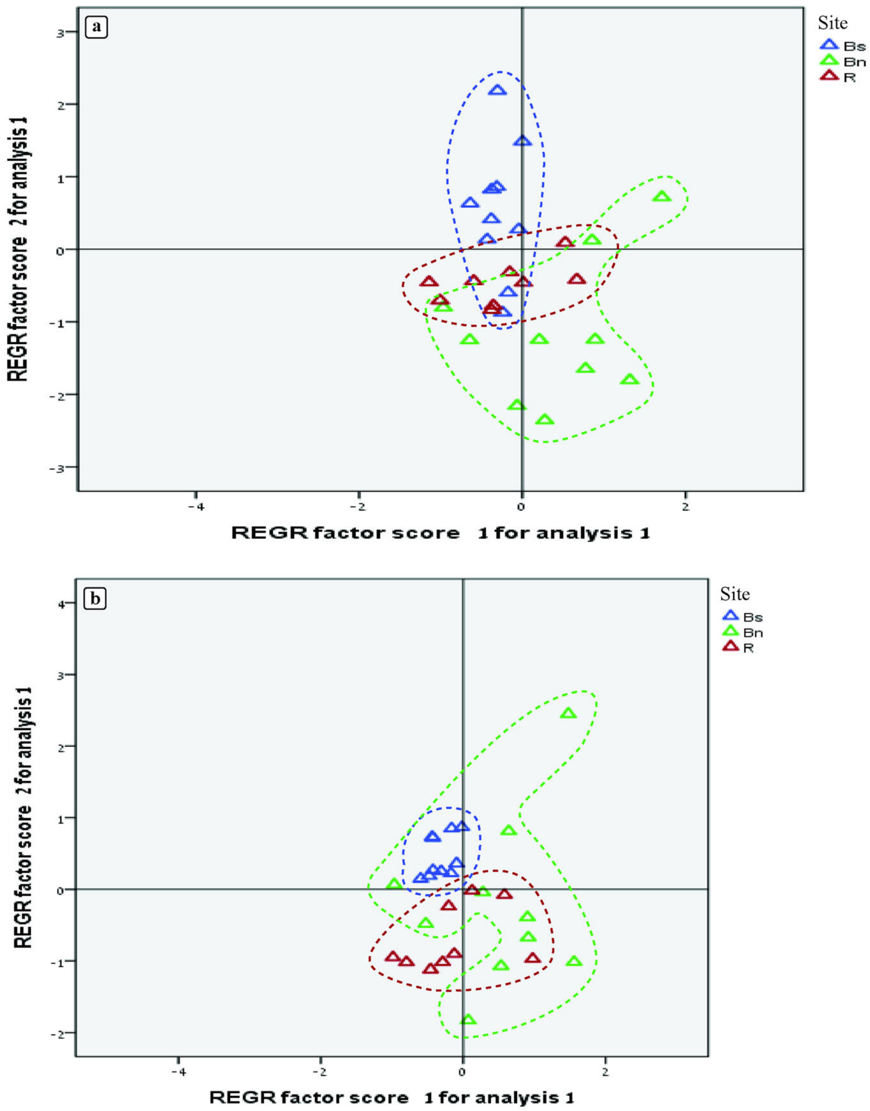


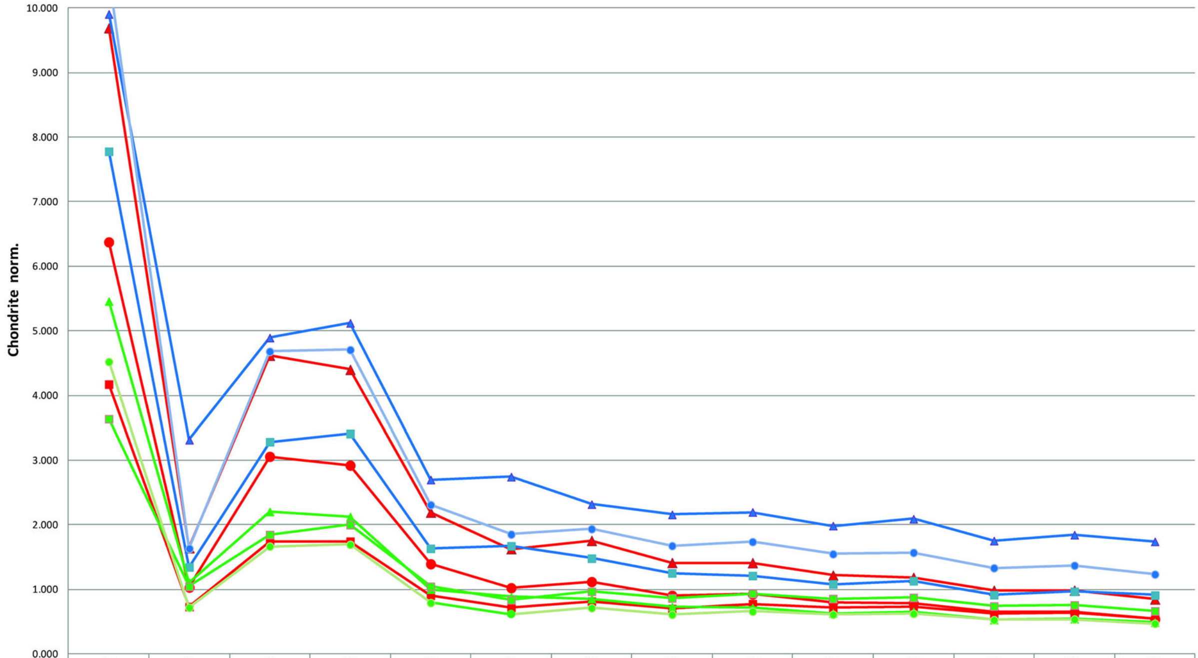
FIGURE 9 (a) A PCA of 39 elements (total variance explained by components 1 + 2– 58.23); (b) A PCA of elements with communalities of >0.9 after PC extraction was analysed again (Na, K, Ca, Mn, Fe, Rb, Y, La, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, and Pb; total variance explained by components 1 + 2– 77.76). PCA: principal component analysis [Color figure can be viewed at wileyonlinelibrary.com]

Lake Hula 12 km east of the Dishon complex (Barkai, 2005; Bocquentin et al., 2011; Lechevallier, 1978; Rosenberg et al., 2008). Lithic findings include many bifaces classified mainly as axes but also as adzes and chisels. Barkai (2005, p. 76) reported that 5,894 bifacial tools were collected from the surface. Khalaily et al. (2015, p. 12) suggested that "The immediate vicinity of the site offers various flint sources, mainly from the Eocene formation in the Naftali Hills. Most of the flint originates from these sources." The Naftali Hills are located west of Beisamoun and have no Eocene formations. The Dishon area 10 km to the west of the Naftali Hills is the closest Eocene formation. In this study, we sampled broken axes.

- Hagoshrim ("G" in Figure 6 located 17 km northeast of the Dishon complex): This is a Neolithic (Prepottery Neolithic C and Pottery Neolithic) and Chalcolithic site that yielded 6,854 bifacial tools through a survey (Barkai, 2005, p. 76) and more were recovered during excavations (Getzov, 2008; Rosenberg et al., 2008, 2010). In this study, we sampled adzes.

2 | METHODS

The main goal of the current research was to establish a robust geochemical signature of the Eocene flint of northern Israel as a basis for provenance studies and to test if regional geochemical differences can be detected in flint sources from the same geological formation. As demonstrated above, the principal (and almost exclusive) source of Eocene flint in this region is the Timrat formation, and accordingly all samples were collected from its outcrops and more specifically from debitage of prehistoric extraction and reduction activities (see above). In addition, flint tools from six occupation sites were sampled to test whether their visual identification as "Eocene flint" was correct and whether their provenance can be pinpointed to a specific E&R Locality within the Timrat formation. Items from the occupation sites were of the colors and textures mentioned above, which are considered to be characteristic of Eocene flint by researchers working in this region.



	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
Dishon Bn	9.689	1.641	4.619	4.399	2.191	1.626	1.755	1.412	1.410	1.217	1.182	0.983	0.984	0.846
Dishon Bs	4.172	0.730	1.734	1.743	0.906	0.719	0.815	0.700	0.774	0.714	0.732	0.622	0.636	0.548
Dishon R	6.375	1.031	3.057	2.920	1.394	1.022	1.118	0.904	0.924	0.801	0.785	0.652	0.648	0.540
Yiron	9.906	3.313	4.893	5.120	2.697	2.743	2.317	2.157	2.196	1.984	2.090	1.752	1.844	1.740
Baram	7.780	1.343	3.282	3.409	1.632	1.671	1.480	1.253	1.210	1.081	1.130	0.913	0.966	0.909
Hamara	10.497	1.632	4.680	4.705	2.309	1.852	1.935	1.674	1.737	1.547	1.569	1.331	1.370	1.233
Ein Miri	5.458	1.105	2.205	2.125	1.001	0.883	0.854	0.729	0.714	0.631	0.653	0.527	0.542	0.488
Hagoshrim	3.636	1.045	1.847	1.998	1.044	0.835	0.967	0.865	0.933	0.855	0.872	0.740	0.756	0.664
Beisamoun	4.520	0.718	1.665	1.693	0.791	0.616	0.712	0.608	0.657	0.609	0.623	0.526	0.536	0.470

FIGURE 10 Chondrite normalization of REE from three E&R localities and six occupation sites (*n* = 10 for each except for R [*n* = 9]). E&R: extraction and reduction; REE: rear earth element [Color figure can be viewed at wileyonlinelibrary.com]

2.1 | Sampling

To build a comprehensive geochemical data set, we designed a three-step sampling strategy: (a) Ten samples from the same tailing pile (from its center) were analysed to present one E&R location (a single tailing pile); (b) in the Dishon and Achbara areas, three E&R locations located 1–2 km from each other were sampled, analysed, and compared to test the intercomplex variation; (c) based on the two previous steps, a comparison was conducted of the three different E&R complexes located 15 km from each other.

Sixty-nine samples of flint flakes (reduction debitage) from seven E&R localities in the three E&R complexes were analysed (10 samples from each tailing pile except for the R locality in the Dishon complex):

- Twenty-nine samples from three tailing piles in the Dishon E&R complex, including Bn (co-ordinates: 241910E/774949N; all co-ordinates are given in the Israel Transverse Mercator), Bs (co-ordinates: 243048E/773846N), and R (co-ordinates: 244547E/773838N, nine samples). For their description, see above and Figures 3 and 6.
- Thirty samples from three tailing piles in the Mt. Achbara E&R complex (designated in Finkel et al., 2018) as E (east; co-ordinates: 247581E/758871N), W (west; co-ordinates: 247246E/759047N), and S (south; co-ordinates: 246576E/758002N).
- Ten samples of flint flakes (reduction debitage) from a tailing pile in the Sede Ilan E&R complex (designated as "Pile 3" in Barkai & Gopher, 2009; Barkai et al., 2006).

It is important to note that while researchers who focus on natural outcrop sourcing employ "geological" flint to identify outcrops, our focus is on E&R sites; this makes it reasonable and more accurate to represent the local source by flint debitage ("archaeological" flint) and not by nearby unexploited geological flint. The flint items were selected according to three criteria: the absence of cortex (to avoid biases caused by differences in chemical compositions between the calcareous cortex and the inner part of the flint item); the absence of inclusions and a minimal weight of 50 g to have a representative sample (after grinding) of otherwise heterogeneous sample. In Luedtke's (1979) paper on the identification of sources of chert artifacts she defines three types of possible errors. A Type 1 error involves the identification of an unknown material as a member of one source when it actually belongs to another source in a study; a Type 2 error occurs when an unknown material is determined to be from a source not included in the study when it is actually from one of the sources considered in the study; a Type 3 error occurs when an unknown material is identified as a member of one of the sources considered in the study when it actually belongs to a source not considered in the study. Our research is prone mainly to Type 1 errors (flint debitage items found on a tailing

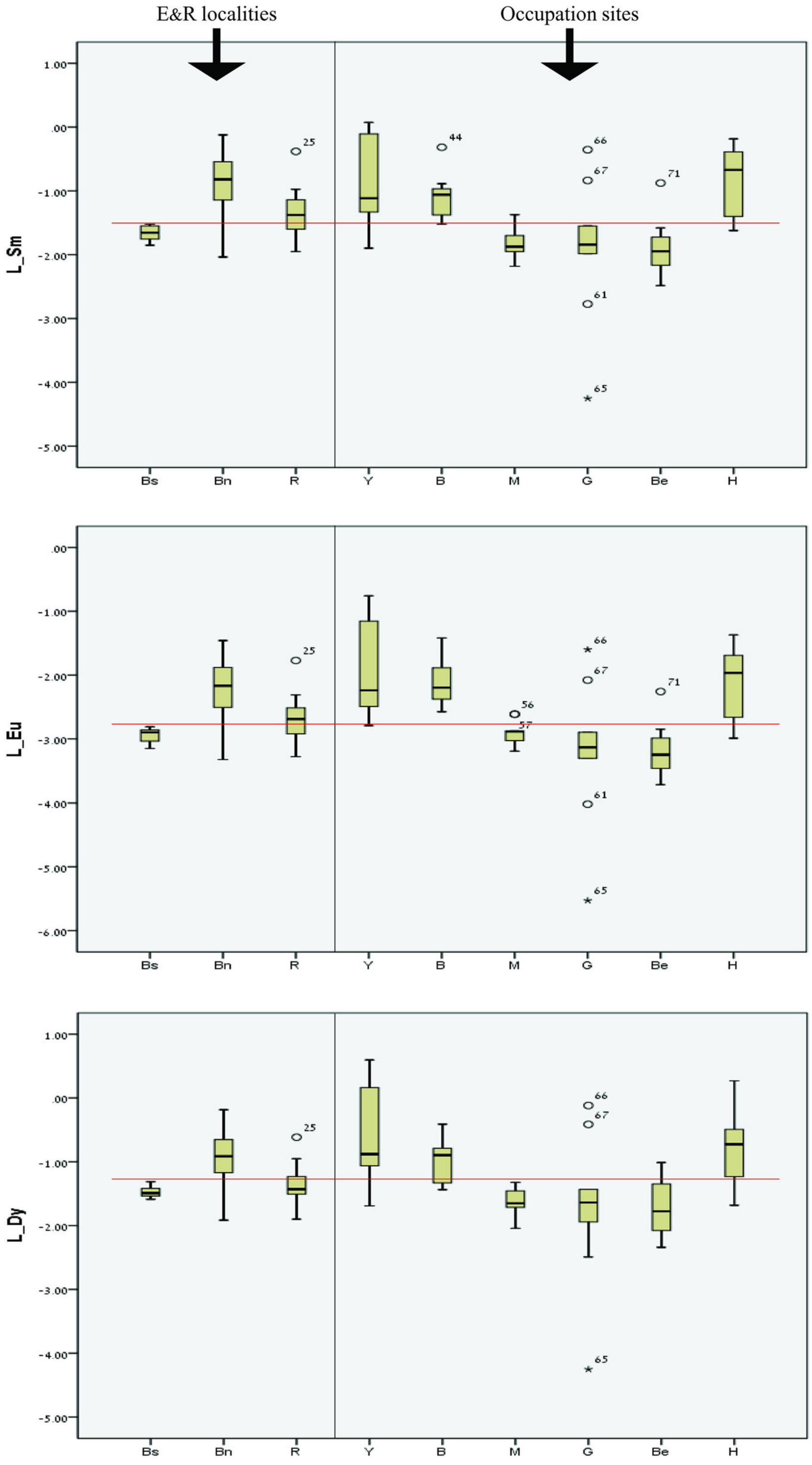


FIGURE 11 Boxplots presenting three REE (Samarium, Europium, and Dysprosium) log values. The red line highlights the pattern and has no statistical meaning. REE: rear earth element [Color figure can be viewed at wileyonlinelibrary.com]

pile within the Dishon complex originating from another locality within the Dishon complex) and less to Type 3 errors because the possibility of having a knapped item brought into the Dishon complex from an unexploited natural outcrop not considered in the study seems to be minimal. Analysing 10 samples from each pile enabled us to identify rare but potential cases in which an artifact found on a pile's surface was transported from another flint formation (i.e., not of Eocene origin), but not when a Timrat Eocene item was transported from a E&R location positioned 1 km away within the Dishon complex. With this in mind, we believe that one such transported item from a different

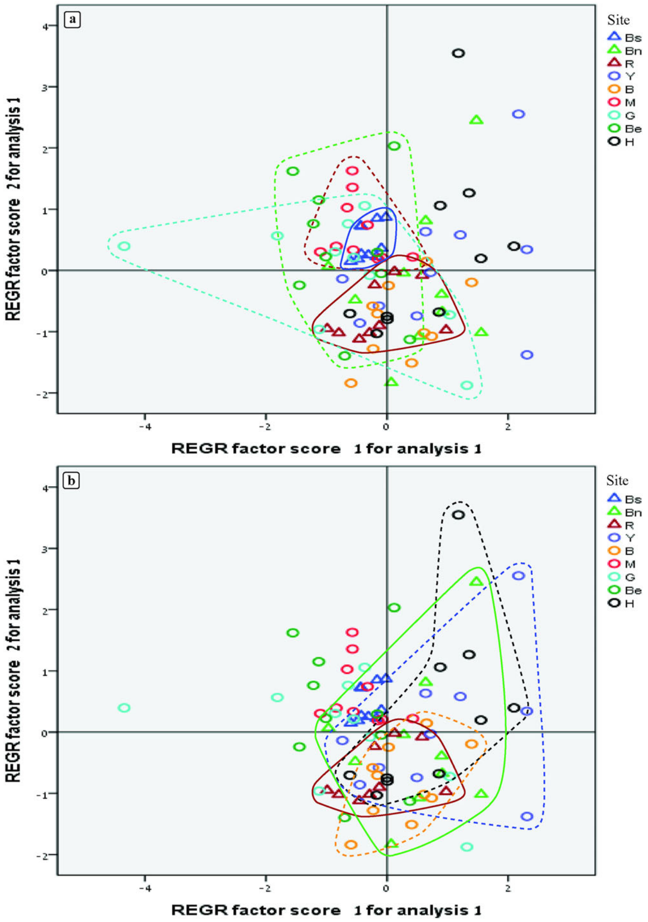


FIGURE 12 A PCA of elements with communalities of >0.9 after PC extraction was analysed again (Na, K, Ca, Mn, Fe, Rb, Y, La, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, and Pb; total variance explained by components 1 + 2 – 77.76). (a) A comparison of Neolithic/Chalcolithic sites M, G, and Be (dotted lines) and E&R localities R and Bs. (b) A comparison of Palaeolithic sites B, Y, and H (dotted lines) and E&R localities Bn and R. PCA: principal component analysis [Color figure can be viewed at wileyonlinelibrary.com]

location in our data would not have significantly altered our 10-sample average and thus the conclusions based on those averages.

Sixty samples of flint items (mostly broken) from six occupation sites were analysed (ten samples from each site)—see Table 1 for a description.

2.2 | Analysis

The geological samples were analysed geochemically using an ICP-MS. Each sample weighed ~50–100 g. Grinding was applied in two phases using a: (a) Heavy duty grinder (model: Retsch BB 100 Jaw Crusher; Tungsten Carbide; full object, 50–100 g) and (b) Tungsten Carbide-based mill (model: Retsch RS 200; ~10 g out of the 50–100 g; 700 rpm, 1 m). The analysis was conducted on 0.5 g (out of ~10 g) treated through the following process: it was dissolved in 9 ml HNO_3 (70%); after 1 hr, 3 ml of HF (50%) was added; then, was heated at 80°C for 24 hr; evaporation was facilitated; 2 ml of HNO_3 was added; evaporation was facilitated; 10 ml of HNO_3 (1%) was added; and finally an ICP-MS analysis was conducted using Agilent 7500 Cx. First, average results for each E&R locality or archaeological occupation site were normalized according to Chondrite norms and were compared with results for flint from other geological formations in the Galilee (data from Ekshtain et al., 2016; Nathan et al., 1999) to establish the Eocene origins of the items. The results were then analysed using statistical software SPSS version 23.0. A principal component analysis (PCA) of 39 elements (out of 40, Tl was excluded due to its marginal values) identified by the ICP-MS was applied to examine the level of correspondence between geochemical signatures of the E&R localities and the archaeological sites. Following this, elements with communalities of >0.9 (see Gluhak, Rosenberg, & Ebeling, 2016) were analysed again (Na, K, Ca, Mn, Fe, Rb, Y, La, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, and Pb), Most of them were rear earth elements (REE), which are usually used to create flint geochemical signatures. On both PCAs, eigenvalues exceeded 1 (5 for all elements and 2.5 for those with communalities of >0.9). We believe that this two-step analysis is rigorous enough to show clear differences between sites. Boxplot graphs were created to visually present differences in values between sites. Before the PCA and Boxplot the data were log-transformed to ensure the independence of the compositional data.

As mentioned in the introduction, ICP-MS still faces analytical and methodological challenges such as geochemical heterogeneities within the same flint nodule/seam (see Luedtke, 1992, p. 54); possible effects of patination (Hughes et al., 2012; Moreau et al., 2016 and references therein; Moroni & Petrelli, 2005); and differentiating flint sources of the same geological age or formation (Luedtke, 1992, p. 54; Moroni & Petrelli, 2005; Rey-Solé, Scherstén, Naeraa, Olausson, & Mangado, 2017; Speer, 2014). In this paper, we report an ICP-MS analysis of a robust collection of flint debitage samples drawn from selected localities in the three E&R complexes (total $n = 69$; 9–10 from each E&R locality) and of flaked flint items of the Palaeolithic and Neolithic/Chalcolithic occupation sites of northern Israel (total $n = 60$; 10 from each site). The results shed new light not only on the provenance of flint in the region but also on methodological issues concerning the method itself, including the ability—in certain cases—to differentiate between outcrops of the same geological formation (age). Based on our results we will compare our method (ICP-MS) to inductively coupled plasma mass spectrometry with laser sampling (LA-ICP-MS; which we did not use) and we discuss their relative advantages and disadvantages. We will also address the effect of patination on the geochemical analysis of flint items.

3 | RESULTS

The results are presented in the following three parts: (a) through a comparison between the Timrat Eocene flint and other flint bearing formations in the Galilee; (b) through a comparison between different outcrops within the Timrat formation along the E&R "strip" (an interarea comparison of the Timrat Eocene flint [Dishon–Mt.

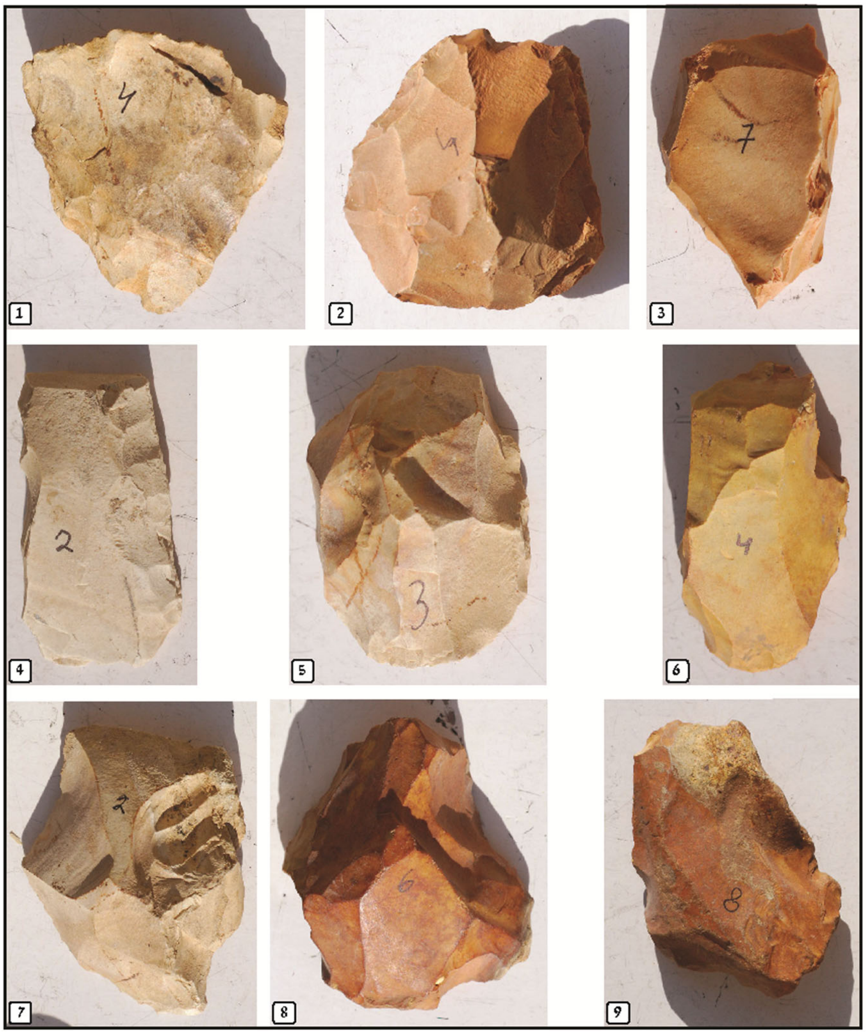


FIGURE 13 Varying patination of flint items from three occupation sites used for the ICP-MS analysis. (1–3) Hamara (H)—Lower Palaeolithic (in Supporting Information Table 1: H4, H5, and H7). (4–6) Hagoshrim (G)—Neolithic (in Supporting Information Table 1: G2, G3, and G4). (7–9) Yiron (Y)—Lower Palaeolithic (in Supporting Information Table 1: Y2, Y6, and Y8) [Color figure can be viewed at wileyonlinelibrary.com]

Achbara–Sede Ilan] and an intra-area comparison of the Dishon and Mt. Achbara areas); and (c) through a Dishon provenance case study—a comparison between the three E&R localities within the Dishon area and six occupation sites in the Eastern Upper Galilee and Hula Valley (three Lower Palaeolithic and three Neolithic/Chalcolithic).

Altogether, 40 elements were identified through the IPC-MS analysis of the 119 samples (39 were analysed, see above; Supporting Information Table 1).

TABLE 1 Description of items from six occupation sites analyzed with inductively coupled plasma mass spectrometry

Site	Description of items
Baram (B)	Cores and retouched flakes
Yiron (Y)	Cores and retouched flakes
Ma'ayan Baruch (Hamara; H)	Broken handaxes
Hagoshrim (G)	Adzes
Beisamoun (Be)	Broken axes
Ein Miri (M)	Broken axes and retouched flakes

3.1 | Comparison of the Timrat Eocene flint to other flint bearing formations in the Galilee

To separate the Eocene flint of the Timrat formation from other geological flint sources found in the Galilee, we normalized our data with the Chondrite Norm (values according to Piper & Bau, 2013) and compared them to previously published data from the Galilee. The comparison (Figure 7) shows that Timrat Eocene flint in the Dishon, Achbara, and Sede Ilan E&R complexes can be differentiated from other geological flint sources owing to its unique pattern of REE concentrations (in particular, Ce depletion). It is worth noting that although the latter is not a rare feature of flint (especially in deep-sea cherts; Shimizu & Masuda, 1977), it does not occur in non-Eocene sources of the region under study.

3.2 | Comparison of different outcrops within the Timrat formation along the E&R "strip"

Overall, when comparing the seven E&R localities within the three E&R complexes, a similarity between them emerges (Figure 8). Differences between the three E&R localities within the Dishon E&R complex are more pronounced than the differences between the three E&R localities within the Mt. Achbara E&R complex: there is a slight rise from Pr to Nd in the Mt. Achbara complex and Sede Ilan compared with a slight decrease observed in the Dishon localities.

3.3 | Dishon provenance case study—Comparison of the three E&R localities within the Dishon area and six occupation sites in the eastern Upper Galilee and Hula Valley

As shown above, it is impossible to differentiate between flint from the three E&R complexes (Sede Ilan, Achbara, and Dishon) by geochemistry alone. However, in taking into account archaeological and geographical considerations and in working at a higher resolution using a procedure such as the PCA of all of detectable elements, patterns can be detected, including, nuanced differences between different localities within the same E&R complex, which in turn can be used for correlations with tools from *relevant* occupation sites. This is best demonstrated by results for the Dishon E&R complex and their comparison with artifacts from the occupation sites sampled as part of this study. These sites are chronologically and geographically related to the Dishon area: three of them are located within the Dishon area itself and the Dishon is most likely the nearest flint source for the three others. Moreover, the chronology of the sites best matches remains at the Dishon E&R localities, as Achbara (15 km south) shows evidence of mainly Middle Palaeolithic and very limited Neolithic/Chalcolithic activities and as Sede Ilan (30 km south) shows mostly evidence of late Lower Palaeolithic and Middle Palaeolithic activities.

A PCA of geochemical data from the three E&R localities of the Dishon complex (Baram north [Bn], Baram south [Bs], and Mt. Reihan [R]) is presented in Figure 9. The analysis together with the REE comparisons presented in Figures 10 and 11 show a clear difference between raw material localities Bn and Bs, a distinction between localities Bs and R and a partial overlap between Bn and R. We can state with caution that interformation differences can be identified in the case of the Dishon extraction and reduction area.

Figure 10 shows similar values for the Bs and R E&R localities and for Neolithic/Chalcolithic occupation sites of Hagoshrim (G), Ein Miri (M), and Beisamoun (Be). The Baram north (Bn) E&R locality flint contains higher levels of REEs and values more similar to those of Palaeolithic sites (mainly Hamara [H] and Yiron [Y]) in La, Pr, Nd, and Sm. Figure 11 presents the same observation with boxplots. Based on the ability to generally differentiate between Lower Palaeolithic occupation sites and the Neolithic/Chalcolithic occupation sites (Supporting Information Figure 3), Figure 12a presents similarities between E&R localities Baram south (Bs) and Mt. Reihan (R) and the three Neolithic/Chalcolithic occupation sites, and Figure 12b shows a similarity between the E&R locality Baram north (Bn) and the three Lower Palaeolithic occupation sites (Yiron [Y], Baram [B], and Hamara [H]; see also Supporting Information Figure 4). It is important to note that the similarity presented in Figure 12 (and in Supporting Information Figures 3 and 4) is based on the visual proximity of the ellipses and not on statistical measures. Altogether, these observations suggest that

- within the Dishon area, the geochemistry of flint items from the Neolithic/Chalcolithic site of Ein Miri (M) can be distinguished from that of items from the Palaeolithic site of Baram (B) situated 1.5 km to the northeast;
- the geochemistry of flint items from the Neolithic site of Hagoshrim (G) is somewhat different from that of items from the Palaeolithic site of Ma'ayan Baruch (Hamara [H]) situated 3 km north; and
- the geochemistry of flint items from the Palaeolithic site of Ma'ayan Baruch (Hamara [H]) is more analogous to that of items from the Palaeolithic site of Baram (B) located 20 km southwest than to that of items of the closer Neolithic sites of Hagoshrim (G) and Beisamoun (Be).

Table 2 presents chronocultural data on the three sampled tailing piles (artifacts methodically collected from a 2 × 2 sq. m area on the surface at the center of the three tailing piles and from indicative items of the entire surface of the piles). The comparison shows that pile Baram north (Bn) was utilized in the Lower–Middle Palaeolithic; pile Baram south (Bs) mostly during the Lower–Middle Palaeolithic but also in the Neolithic/Chalcolithic; and pile Mt. Reihan (R) mostly in the Neolithic/Chalcolithic and during the Middle Palaeolithic. These finding support the geochemical analysis

TABLE 2 Comparison of flint items from a 2 X 2 sq m area in the center of the three piles in the three E&R localities (Finkel et al., 2017)

Category of items	*Bn*	*Bs*	*R* (RAW 100)
Core	14 (11%)	7 (3.6%)	12 (3.7%)
Levallois core	1 (0.7%)		
Big flake	6 (4.7%)	2 (1%)	48 (14.8%)
Retouched big flake	2 (1.6%)	7 (3.6%)	5 (1.5%)
Flake	22 (17.2%)	50 (25.6%)	182 (56.2%)
Retouched flake	17 (13.3%)	58 (29.8%)	39 (12%)
Bifacial roughout	1 (0.7%)—early	1 (0.5%) —late	16 (4.9%)
Blade	4 (3.1%)	4 (2%)	9 (2.8%)
Chunk	61(47.7%)	66 (33.9%)	6 (1.9%)
Special waste			7 (2.2%)
Total	128 (100%)	195 (100%)	324 (100%)
Indicative items from the whole pile's surface (including the 2 × 2 sq. m)	**2** Early bifacies **2** Early bifacial roughouts **3** Levallois cores	**1** Early bifacial roughout **4** Levallois cores **3** Late bifacial roughouts	**38** Late bifacial roughout (adzes/axes) **7** Levallois cores

results concerning the correlation found between these localities and the Palaeolithic and Neolithic/Chalcolithic occupation sites.

The geochemical results of the current study provide a robust new data set (see Supporting Information) for the Eocene flint of the Timrat formation, complementing previously published data. The geochemistry of flint items from occupation sites further confirms their identification as Eocene flint from visual attributes. Moreover, based on geographic considerations and a PCA of 39 elements, we demonstrate that in specific cases it is possible to distinguish between spatially close outcrops within the same geological formation and to correlate them with occupation sites.

4 | DISCUSSION

We will discuss the implications of this study for prehistoric flint provenance in northern Israel and then methodological issues.

4.1 | Flint provenance in northern Israel: The importance of the Timrat Eocene "strip" and the Dishon E&R complex

In combining (a) evidence from the three E&R complexes along the Timrat E&R "strip"; (b) the broad use of "visually Eocene" flint in Palaeolithic and Neolithic/Chalcolithic northern Israel; and (c) the similar geochemistry of the Timrat flint outcrops, we suggest that the Timrat Eocene "strip" was a well-known prime source for quality flint over long periods of time in prehistoric northern Israel. Located along the rift valley 8–10 km west of the Sea of Galilee—Jordan River—Hula lake line, the Timrat E&R "strip" probably supported both Palaeolithic hunter–gatherer groups moving along the valley following the seasonal Middle Pleistocene movements of large herbivores (Devès, Sturdy, Godet, King, & Bailey, 2014) and the inhabitants of Neolithic/Chalcolithic settlements.

We combined three different lines of evidence to reach our conclusion: First, from the identification of Eocene flint items at archaeological sites as indicated by researchers of the Lower Palaeolithic Ma'ayan Baruch (Ronen et al., 1980; Stekelis & Gilead, 1966), of the Neolithic Beisamoun axes (Khalaily et al., 2015) and of Palaeolithic Baram and Yiron (M. Ohel, 1986a, 1991); second, through a geochemical analysis of archaeological items of Timrat Eocene formation origin; and third, from the presence of a wide and extensive Eocene flint E&R area in which lithic technologies in use correlate well with those found at occupation sites. Altogether, while keeping in mind that other E&R complexes may be found north of the Galilee in Lebanon, we can suggest that the Central Dishon Basin E&R complex was used as a regional "industrial" area for northeastern Israel and not only for the occupation sites we specifically examined but also for other sites whose assemblages contain flint items visually attributed to Eocenean origins as in the case of the Lower Palaeolithic site of Gesher Benot Ya'aqov (Type 01 artifacts suggested to originate from the Dishon area, Delage, 2007b, p. 224). Within the Dishon complex, different locations were preferred in different periods. The common thread connecting these very different periods, from a flint procurement point of view, was the need for relatively high-quality large flint nodules that could provide initially large cores/flakes for the production of either early hand axes and Levallois cores or late axes and adzes. Timrat flint served as the best material for this purpose.

Is the phenomenon described here the result of embedded or direct procurement (Binford, 1979, 1980; or "special purpose": Frahm et al., 2015)? We would like to note that all E&R complexes include an extensive area used for flint extraction from a single geological formation that was visited repeatedly in the Lower and Middle Palaeolithic and much later in the Neolithic/Chalcolithic periods to procure high quality stone from primary geological sources needed for the production of large-volume flint items. Although we cannot prove that direct procurement rather than embedded procurement was being practised, evidence from ethnographic accounts of recent hunter–gatherer communities presents a model of specific extraction and reduction parties arriving at flint sources for rather short but intensive working sessions (e.g., Australia, Gould & Saggers, 1985, p. 121; Jones & White, 1988, p. 76; Papua, New Guinea, Burton, 1984, p. 237; Hampton, 1999, pp. 235 and 239). It is important to note that although direct procurement is not intuitively related to Palaeolithic hunters–gatherers, accumulating evidence from North Africa (Foley & Lahr, 2015), the Arabian Peninsula (Groucutt et al., 2017; Jennings et al., 2015), Israel (Barkai & Gopher, 2009; Barkai et al., 2002; Finkel et al., 2016; Gopher & Barkai, 2006, 2011, 2014), India (Paddayya et al., 2000, 2002; Paddayya, Jhaldiyal, & Petraglia, 2006; Petraglia, LaPorta, & Paddayya, 1999; Shipton, 2013), and other areas suggests that this was the case at least in certain regions of the old World.

4.2 | Methodological issues

The nondestructiveness of LA-ICP-MS has rendered the method a first choice in many geochemical studies (see, e.g., studies presented in Pereira et al., 2017) and the only choice when dealing with museum collections. However, it is hampered by its sensitivity to the relatively high heterogeneity of flint and to the debatable effects of patination, which are usually compensated for by trying to represent fully natural heterogeneities that appear on the item's outer parts (Andreeva, Stefanova, & Gurova, 2014; Brandl et al., 2011; Gurova et al., 2016; Moreau et al., 2016; Moroni & Petrelli, 2005; Pettitt, Rockman, & Chenery, 2012; Speer, 2014). This still does not guarantee a reliable representation of the composition of a complete object, as inner parts of the object may contain other concentrations of elements than the outer parts. Here, we overcame both challenges by pulverizing relatively large samples (50–100 g of debitage [flakes] and broken hand axes, axes, and adzes) and through the use of ten items from each site (either E&R locations or occupation sites). Our results shed new light on the methodological challenges and debates of current research.

4.2.1 | Visual and geochemical links

Flint interformation color and texture differences are a common phenomenon worldwide. This is clearly visible in Ekstain's macroscopic observations of different flint formations in the Galilee (Ekshtain et al., 2016) and the Mt. Carmel area (Ekshtain et al., 2014). Ekshtain et al. (2016) acknowledged that although Eocene flint exposures in the Eastern and Central Galilee are similar in many visual characteristics to Eocene exposures near the Amud Cave, "[…] Still, various areas can be distinguished geographically […]" (Ekshtain et al., 2016). However, there is no correlation between visual variability in Ekshtain et al.'s studies and the different Eocene formations, and visual variability was observed also within each of the formations (i.e., Timrat and Bar Kochba). In the case of the Amud Cave, the definition of "East Eocene" REE values was based on averaging 25 samples from a few locations of the 207 sq km area surrounding the Amud Cave and based on combined samples from both the flint-rich Timrat and the relatively flint-poor Bar Kokhba formations (*ibid.* Table II; note that the flint of the latter is also poorer in quality). This poses a certain problem assuming that these inter-Eocene formations are geochemically different. Notwithstanding intraformation variability, the attempt to establish geochemical and visual links between archaeological artifacts and geological exposures of Eocene flint was successful in both of Ekshtain et al.'s cases. The results presented in this study correlate visually and geochemically artifacts from six occupation sites with debitage from three Eocene flint E&R sites (all from the Timrat formation), thus fortifying Ekshtain's finds. Furthermore, for many years researchers assigned bifacial flint tools in almost all Palaeolithic and Neolithic/Chalcolithic sites of northern Israel to Eocene flint based on visual attributes—Lower Palaeolithic bifaces of Ma'ayan Baruch (Hamara; Ronen et al., 1980; Stekelis & Gilead, 1966); Baram (M. Ohel, 1991, pp. 24–37 and 121–149); Yiron (M. Ohel, 1986a, pp. 169–173); Gesher Benot Ya'aqov Type 01 artifacts (Delage, 2007b); Chalcolithic bifaces of the Golan Heights (mainly from Rasem Harbush; Noy, 1998, pp. 270–271); Neolithic Axes of Hanita (Ronen, 1968, p. 12); and Neolithic Beisamoun (Khalaily et al., 2015, p. 12). Middle Palaeolithic items from the Amud Cave have been visually and geochemically shown to be of Eocene origin (Ekshtain et al., 2016). This assignment is not only justified now by our new geochemical data but may also relate most of these items specifically to the E&R Timrat Eocene flint "strip."

4.2.2 | Overcoming flint heterogeneity

Flint heterogeneity constitutes a well-known challenge to establishing a geochemical fingerprint. Luedtke wrote that "Intraformational variability occurs on more than one scale. On the smallest scale, that visible in thin sections, it is evident that many impurities are not evenly spread throughout a fragment of chert but rather are clumped into discrete crystals, localized around fossils, or concentrated along laminations, and so forth. Thus, chemical variability will usually be great if tiny portions of a fragment of chert are sampled but should decrease if larger samples are analysed or if samples are pooled" (1992, p. 54).

As is frequently shown, focal LA-ICP-MS measurements result in inter-item variations of element values (Gurova et al., 2016—Supporting Information; Moreau et al., 2016—Supporting Information and references within; Moroni & Petrelli, 2005). This is treated by averaging measurements (Speer, 2014, averaged 60 measurements to represent one item—10 spots × 6 readings per spot), but as stated above, this still does not guarantee a reliable representation of an object's total composition. Some researchers attempt to overcome this challenge by chipping small pieces from an archaeological item artifact (1–5 mg; Olofsson & Rodushkin, 2011) that do not damage it but at the same time are still sensitive to the same bias. ICP-MS-based works usually do not specify the size/weight of flint items used as samples, but relatively small-flaked items are typically used (Pettitt et al., 2012—Points, endscrapers, burins; see Figures 1 and 2; Olofsson & Rodushkin, 2011—flakes, scrapers; see Table 1; Ekshtain et al., 2016—Levallois points, blades, Levallois and non-Levallois flakes; see Table 2). In our case, the archaeological items were more voluminous bifaces or cores (Table 1), enabling us to begin the process with a relatively heavy item (50–100 g), to fully grind it and then to analyse 0.5 g, which represented the entire item. Our relatively low interlocation standard deviation (~50% of the average for REEs) compared to much higher standard deviations (a few hundred) observed in some cases where a small number of samples were pooled together from few different geographical locations (Nathan et al., 1999, table 3; Vallejo Rodríguez, Urtiaga Greaves, & Navazo Ruiz, 2017, table 1) attests to the advantages of this approach.

4.2.3 | Patination effects on geochemical fingerprints

Differences in patination were observed between various E&R localities (Baram north [Bn] showed darker patination than Baram south [Bs], and Mt. Reihan [R] as well as between Palaeolithic occupation sites [Baram, Yiron, and Hamara], which showed darker patination than Neolithic/Chalcolithic occupation sites [Hagoshrim, Beisamoun, and Ein Miri]). The intrasite variability of patination (albeit smaller) was found in every site, whether in E&R localities or occupation sites (see Figure 13). For Ma'ayan Baruch (Hamara), for example, the researchers state that "The flint tools are covered by a predominantly red-brown patina of various shades: dark red-brown, light red-brown, red-yellow, and so forth; there are also tools covered by a light yellow patina" (Stekelis & Gilead, 1966, p. 7) and that "On 25 of the hand axes a different patination appears on each face: one face is generally brown and the other is either yellow, white, or other colors. Some exhibit multi-colored bands" (Ronen et al., 1980, pp. 19–20). At the Acheulean sites of Yiron and Baram, Ohel basically found the same phenomenon (M. Ohel, 1991, p. 149).

Hughes et al. (2012), using energy-dispersive X-ray spectroscopy (EDXRF), compared both sides of one flint item and claimed that "in relation to the unpatinated surface, patinated surface shows SiO_2 depletion and dramatic enhancement in Al_2O_3, K_2O, and Fe—in particular, the Fe, Cl, and Ti values are also elevated in the patinated sample [and supported] the position that postfracture weathering and the deposition environment do introduce chemical changes in the affected surfaces (e.g., Shepherd, 1972, 114 ff.; Luedtke, 1992; Högberg and Olausson, 2007, pp. 67–69)." Moreau et al. (2016) suggested otherwise. They collected a series of eight LA-ICP-MS aligned test measurements from the transversal section of a patinated lithic artifact made from Spiennes flint collected on the surface and concluded that "…trace element contents detected in the patinated artifact appear to be consistent with the natural variability of elemental concentrations documented for the geological samples of Spiennes flint" (Moreau et al., 2016, p. 234).

In our geochemical analysis, darker patination did not result in any detectable difference in chemical compositions compared to light colored patination, which supports Moreau et al.'s (2016) claim (see Figure 13 and Supporting Information). Using samples based on the grinding of large complete items (50–100 g) as the basis for our geochemical analysis and not using a near-surface sample only minimizes the problem, as the volume of the patinated surface is negligible compared with the volume of the entire item. Furthermore, elements that are the most soluble and that, therefore, move from inner parts of the artifact to the surface are Fe and Mn and not REEs, which "are quite stable during diagenesis and weathering, therefore they reflect the chemistry of the original sediment as derived from primary accumulation" (Moroni & Petrelli, 2005).

4.2.4 | Microscale (1–2 km) intracomplex geochemical variability

Identifiable geochemical differences between outcrops of flint of the same geological age but different formations are quite common (Luedtke, 1992, pp. 53–56) and this is known for our region too, with studies showing differences between Eocene age flint from different parts of Israel (e.g., Central Israel, Carmel, and Galilee) polling together a few different formations (Ekshtain et al., 2014, 2016; Segal et al., 2005). Identifiable differences between geographically close flint outcrops within the same formation are a rare phenomenon (see Moroni & Petrelli, 2005; Rey-Solé et al., 2017; Speer, 2014). In our case such variability was found in the Dishon E&R complex (especially between Bn and Bs located 1.5 km from each other), but not in the Achbara complex. As Luedtke pointed out, varied conditions in the depositional environment (e.g., water salinity and pH, fauna and flora in the water, the presence of other types of rocks and volcanoes in the deposition area, etc.) result in impurity variability (Luedtke, 1992, p. 36). One or more of these variables in the deposition environment is likely responsible for the intracomplex variations observed. As our work exemplifies, the need for intraoutcrop resolution is critical when dealing with large-scale E&R complexes.

5 | CONCLUSIONS

The present study provides the first robust geochemical data set for one of the major sources of raw materials of prehistoric northern Israel—Eocene flint of the Eastern Galilee. In contrast to previous studies of northern Israel's flint, which were focused on natural geological exposures, our data were extracted from three areas identified as large-scale prehistoric flint E&R complexes, and a chemical analysis (ICP-MS) was conducted on flint items identified as a direct product of prehistoric extraction activities. The new data set, which represents the geochemical "fingerprint" of the Timrat formation, is clearly distinct from that of flint from any of the other (not-Eocene) flint-bearing formations of northern Israel. This is most readily evident in the composition diagram of the 13 REEs and especially in the distinct "trough" pattern observed in the change between La, Ce, and Pr in flint of the Timrat formation.

In addition, we found that while it is yet impossible to distinguish between the three major E&R complexes identified in this region (when all data are averaged per complex) and even between the Dishon and Sede Ilan E&R complexes separated by more than 30 km, a high resolution study of a specific E&R complex may enable the distinction of specific localities (small and distinct areas of extractions). With the study presented above we were able to separate three localities within the Dishon E&R complex based on a PCA of all 39 elements detected through the geochemical analysis. Furthermore, we used this information to match these specific localities to flint tools found in six occupation sites (three Lower Palaeolithic and three Neolithic/Chalcolithic) located up to 20 km from the complex. The results show that in each period, a different E&R locality was utilized.

The relatively large quantity of analysed samples used ($n = 69$) also yielded methodical insights, most importantly addressing challenges of chemical heterogeneity observed in the original flint samples and the debated effects of patination. In our study we deliberately avoided LA-ICP-MS and demonstrated that pulverizing 50–100 g samples is sufficient in both averaging a sample's chemistry and in coping with any possible effects of patination.

We hope that in following a similar procedure more high-quality geochemical data will be generated by future studies of this and neighboring regions. While more data for the Eocene flint of the Timrat formation might help facilitate differentiation between intraformation sources, there is a dire need to produce robust data sets for other flint-bearing formations and especially for those of the Upper Cretaceous, which were another significant source of raw materials in prehistoric southern Levant, and our current mapping of their geochemistry is rather limited.

Lastly, the present study further emphasizes the importance of Eastern Galilee flint in prehistoric times: recently discovered E&R complexes together with previously identified locations should now be understood as part of a vast region delineated by the exposure of the flint-bearing Timrat formation, which is shaped as an elongated, north-south "strip" along the western margins of the Rift Valley. This major source of high quality flint used primarily for large-volume items such as Lower Palaeolithic hand axes, Middle Palaeolithic Levallois cores, and Neolithic/Chalcolithic axes/adzes is now one of the better-studied prehistoric flint sources in the Levant; it undoubtedly contributed to several aspects of human activities in the area such as the selection of site locations. A rich flint source such as this should be taken into account in future models of human prehistory both locally and as a key region of human dispersals.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of the article.

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