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关于以色列国的基巴拉和哈约尼姆两洞穴的微矿物的研究：对于考古纪录中灰烬沉积的认识

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关键词 火，灰烬，矿物，红外光谱学，成岩作用

火的考古纪录对于认识早期人类的技术能力是十分有趣的。通过研究烧骨、炭屑、石器、以及高温对于烧过的泥土的古地磁特性的影响，我们可以间接地推断古人对火的使用问题。关于用火的直接证据则是保存下来的燃烧后的残留物，即炭屑和灰烬。它们能够提供古人类用火的方式，以及其它的许多信息。所以它们的潜在价值是巨大的。关于遗址的居住者能不能制造火和控制火的问题，目前很难明确地回答。最可能的是从灰烬和炭屑中能看出点什么。

灰烬在大多数环境中都是不稳定的，但古代的灰烬都经历过一系列的成岩作用的变化。它们往往在遗址中被冲散，其结果是火堆的典型的宏观面貌（有炭屑和红色土壤的卵圆形透镜体）常常消失了。如果发生了这种情况，用光学显微镜在薄切片中所见的灰烬特有的微形态特征也将按照保存状态而有所改变。基巴拉洞穴通常具有保存良好的火堆，用肉眼就可以认识它们。在本文中我们利用这一事实以论证与真正火堆有联系的矿物的多样性及成岩作用的过程。我们将上述研究成果应用于研究哈约尼姆洞穴。在哈约尼姆洞穴，火堆的保存是不佳的。

新鲜的木头烧成为灰烬以后，其中的矿物包括两个来源。它们既可分开，又互相有关。

(a) 它们原是木头中的坚硬部分，燃烧后在灰烬中或原封不动，或为火烧所改变，常见的例子是方介石。它通常是新鲜灰烬中的主要矿物成分，它被认为是木头中的草酸钙晶体，经过高温氧化，然后变成氧化钙，最后转化为方解石。在木头中，另一种东西是由非晶质的硅质组成的植物石。在通常的营火温度中（低于950℃）其矿物成分不会改变。硅酸体也是存在于许多种树木内的复杂构造，也是木头灰烬的一种成分。这种硅酸体富含硅、铝、钾和铁。

(b) 在树木中还有许多离子。经过燃烧后，由此生成的含离子的矿物多数溶于水，所以与考古记录不大有关系。它们是方钙石、羟钙石、斜硅钙石和钾石盐。

总之，可以认为木头烧成的灰烬几乎全由方解石组成，植物石和硅酸体占很小的百分比（以色列的7种树木内占0.3%—3.7%）。应当想到，用来烧火的干木头总是粘附了一些土壤和灰尘，那么这些土壤和灰尘也会存留在灰烬内。

在基巴拉洞穴内有许多保存良好的火堆。它们为白、灰或黄色，圆一椭圆形，直径约

30—150厘米，在剖面中呈透镜体状，厚度在几到几十厘米。它们通常位于薄层黑褐色富含炭屑的沉积物上面。黑色的炭屑常常局限于其上覆的灰烬的中央部。这使人想到一种可能性，即灰烬本身扩散到了原生火堆的范围之外。

对于基巴拉洞穴中残留的烧木的初步研究表明，它们之中的85%是由两种橡木所组成的。这两种植物正是今天该地林带中的主要种类。表1列举了基巴拉洞穴中几个火堆的编号以及它们的主要矿物的名称。表2则列出表1所列举的矿物的分子式，同时也列出本文所涉及的各种矿物的分子式。这些矿物主要是用红外光谱仪鉴定的。由于新鲜的灰烬主要由方解石组成。所以我们可以假定，7个主要由方解石组成的火堆乃是保存得最好的。在这些火堆的顶部，具有以碳羟磷灰石或以碳氟磷灰石为主要成分的壳。它意味着在这个环境中，方解石与富含磷酸盐的地下水起化学反应后，溶解，作为碳羟磷灰石或碳氟磷灰石再沉淀。据研究，反应的顺序如下：碳羟磷灰石或碳氟磷灰石被斜磷钙铝石所取代，而后者又被磷铝镁钙石和羟磷钾铁矿所替代。在上述序列中，我们不知道在什么时候生成磷钙铝石。但是有一点是可以想象的，即碳酸盐矿物或磷酸盐矿物可以在地下水中溶解，并被搬运到洞外。在浓度为6N的盐酸中，它们比硅酸体和植物石易溶解。这使我们可以想到，仅由后者组成的火堆曾经历过最长期的和/或最严重的溶解状态。图3显示基巴拉洞中研究过的许多火堆的分布图。所有以硅酸体为主的火堆都在界线之南，这也是没有保存为骨化石的区域。所以观察到的这种分布情况表明：新鲜灰烬中相对稳定的成分乃是硅酸体。

从1965年起，哈约尼姆洞穴被断断续续地进行着发掘。但到目前为止，根据宏观的形态特征而识别出来的火堆很少。洞内几乎没有发现炭屑。但是洞内有不少结核，它们由磷铝镁钙石、羟磷钾铁矿和磷酸铝石等组成。这一事实意味着，含磷矿物在某些地方被溶解了，却在另一些地方重又以结核的形式沉积下来。在哈约尼姆洞穴内，火堆的保存状况很差，可能是由于上述这些因素。

观察哈约尼姆洞穴中矿物的分布，最引人注目的是：洞穴中央的最丰富的矿物是硅酸体。它们呈层状出现，为乳白色或灰色。它们的颜色虽然不同，但其红外光谱却是相同的。还不清楚这些颜色变化的原因是什么。在某些部位，它们可厚达数米。这种情况意味着，在哈约尼姆洞穴中，木头的灰烬在沉积物中占很大的比例。因为可溶性的由方解石衍生的矿物在原始的灰烬体积中超过90%，在溶解过程中沉积物的体积一定极大地减少了。

近代的研究表明，灰烬可以许多不同的矿物形态而存在着，如方解石，碳羟磷灰石，碳氟磷灰石，纤磷钙铝石，磷铝镁钙石，羟磷钾铁矿，以及硅酸体等。如果上述矿物不是呈结核状，也不是呈反应边的形式出现在剖面内岩石的表面，而是呈层状出现的话，那么它们应被认为可能是由灰烬转化而来的。硅酸体或植物石在地层中的存在，也可提供强有力的证据，表明地层乃由木灰所形成。

已知硅酸体存在于树木中，但不知道是否所有树木均形成硅酸体，或者只存在于树木中，如果其它植物也形成硅酸体，那么它们成层地存在于考古遗址中，那可能是由于其它的过程。

总之，我们要在考古记录中识别灰烬是不简单的，特别是灰的矿物成分在这方面是有用的。识别灰烬乃是辨认火堆的前提。火堆是人类有意识用火的结果，无疑地是人类进化

史上一种重要的文化属性. 如果炭屑保存得好，可以从中研究出更多的东西。希望上述研究成果能对增进了解人类的用火有所贡献。

表 1 基巴拉洞穴中分析的几个火堆的主要矿物成分

矿 物	火堆编号	矿 物	火堆编号
方解石	7	羟磷钾铁矿	6
碳羟磷灰石/ 碳氟磷灰石	13	磷钙铝石	2
磷铝镁钙石	14	硅酸体	17
纤磷钙铝石	1		

表 2 在文中涉及的矿物及其化学分子式

碳酸盐	
方解石 (C)	$CaCO_3$
白云石	$(Ca, Mg)CO_3$
磷酸盐	
碳羟磷灰石 (D)	$Ca_5(PO_4, CO_3)_3(OH)$
碳氟磷灰石 (F)	$Ca_5(PO_4, CO_3)_3(F)$
纤磷钙铝石 (CR)	$CaAl_3(PO_4)_2(OH) \cdot 5H_2O$
磷铝镁钙石 (M)	$Ca_4MgAl_4(PO_4)_6 \cdot (OH)_4 \cdot 12H_2O$
羟磷钾铁矿 (L)	$K_2(Fe^{3+}, Al)_4(PO_4)_4 \cdot (OH)_2 \cdot 4H_2O$
磷钙铝石 (T)	$H_6K_3Al_5(PO_4)_8 \cdot 18H_2O$
磷铝石	$AlPO_4 \cdot 2H_2O$
其它	
石英	SiO_2
方钙石	CaO
羟钙石	$Ca(OH)_2$
斜硅钙石	$Ca_2(SiO_4)$
钾石盐	KCl
硅酸体	参看 Schiegl *et al.* (1995)

图 的 说 明

图1 哈约尼姆和基巴拉洞穴的地理位置图

图2 在基巴拉洞穴中保存良好的火堆的照片，它们在剖面中呈透镜体状。其中颜色较暗的富含炭屑的层位可由灰烬转化而来的，颜色较浅的矿物层的下面

图3 在基巴拉洞穴的中央部位和北部，表示出许多由灰烬转化而来的矿物体的位置. 此图还显示了发掘的区域（其边界与发掘用的方格相平行），含磷的沉积物的位置（黑色圆圈）和不含磷的沉积物的位置（白色圆圈）. 两者之间的界线以虚线表示。方解石的灰烬层以卵圆圈表示，内有字母“C”。以上所有信息均来自 Weiner *et al.* (1993) 的论文. 灰烬层中每种矿物都由表 2 中规定的英文字母来表示. 每一方格的面积是

1平方米。罗马数字表示地层的层号。这里的注释与即将发表的 Weiner *et al.* (1995) 的注释相同

图4 哈约尼姆洞穴中由灰烬转化而来的地层的照片，硅酸体是它们的主要成分

(徐钦琦 节译)

RECOGNIZING ASH DEPOSITS IN THE ARCHAEOLOGICAL RECORD: A MINERALOGICAL STUDY AT KEBARA AND HAYONIM CAVES, ISRAEL

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Abstract

Preserved ash in the archaeological record constitutes an important source of information on the manner in which fire was used by hominids. Recognizing fossil ash deposits is difficult, because ash is not stable in most environments, but undergoes a series of diagenetic alterations. Here we review the mineralogical transformations that ash deposits underwent in two caves in Israel. We note that a relatively stable component of ash, at least in these environments, is the siliceous aggregate fraction. Its presence in bedded layers within the stratigraphic sequence at other sites may constitute good evidence for the presence of fossil ash deposits.

Key words Fire, ash, minerals, infrared spectroscopy, diagenesis

Introduction

The archaeological record of fire is of much interest in understanding the technological capabilities of early humans. The use of fire can be inferred indirectly from the presence of burned bones (e.g. Shipman *et al.*, 1984; Stiner *et al.*, 1995), charcoal (e.g. Oakley 1970), lithic artifacts (e.g. Purdy and Brooks 1971) or from the effects that high temperatures have on the paleomagnetic properties of burned soils (e.g. Barbetti *et al.*, 1980). Direct evidence for fire at a site can be obtained from the preserved remnants of the fire itself – the charcoal and the ash (van Riet Lowe 1954). These are potentially of much value as they can also provide information on the manner in which fire was used and its spatial context vis-a-vis other activities at the site.

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The important question of whether or not the site occupants were able to ignite and control fire is very difficult to answer unequivocally, but if any insight can be gained, it will most likely be derived from the ash and charcoal components. For a comprehensive review of the evidence of fire in the archaeological record, see James (1989).

In reality the major difficulty in documenting and understanding the archaeological record of fire results from the facts that both wood ash and charcoal are not stable in most environments, but undergo a series of diagenetic changes. They are also likely to be physically dispersed around the site and consequently the classic macroscopic features of hearths (oval-shaped lenses associated with charcoal and reddened soil) are often lost. If this occurs the characteristic micromorphological features of ash seen in thin-sections studied by optical microscopy (see Courty *et al.*, 1989 for details) will also vary according to the state of preservation. In this study, we exploit the fact that Kebara cave generally has remarkably well preserved hearths, that are easily recognized by the naked eye, in order to document the mineralogical diversity and diagenetic processes associated with bona fide hearths. We then apply this knowledge to Hayonim cave, where hearths are generally not well preserved. Figure 1 shows the geographic locations of Kebara and Hayonim caves.

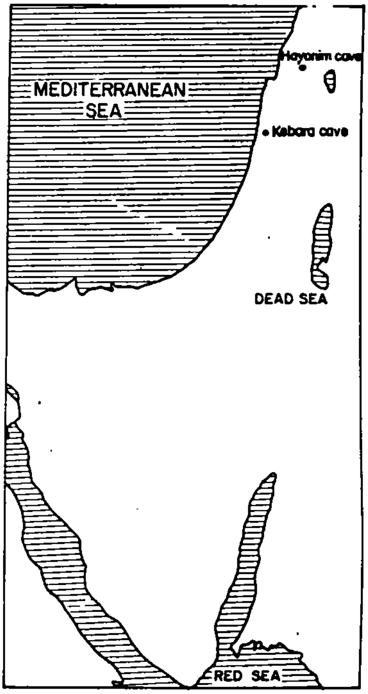

Figure 1. Map showing the geographic locations of Hayonim and Kebara caves

Mineralogical Characteristics of Fresh Wood Ash

The minerals present in fresh wood ash can have two separate, but related sources. (a) They are present in the original wood as a solid phase. After burning they reside in the ash intact or are altered by the heating process. A common example is calcite, which is usually the major mineral component of fresh ash. It is thought to be formed primarily from the oxidation at high temperatures of calcium oxalate crystals present in the wood to CaO, followed by hydration and carbonation to calcite (Wattez and Courty 1987; Etiégni and Campbell 1991). Phytoliths composed of amorphous silica are present in the wood and are usually unaffected mineralogically by heating at normal campfire temperatures (up to 950℃, Stiner *et al.*, 1995). Siliceous aggregates are also present in the wood of many species of trees (Sangster and Parry 1981), and are components of wood ash (Schiegl *et al.*, 1994). The siliceous aggregates are complex structures found in the wood and bark of many trees. They are composed of a variety of crystalline minerals, presumably derived from the local environment in which the tree grew, embedded in an amorphous mineral matrix rich in Si, Al, K and Fe. For more details, see Schiegl *et al.* (1994). It is not yet known if they are altered by the heating process. (b) Ions that are present in a soluble or bound form in the wood undergo oxidation reactions in the fire to form a wide variety of mineral phases, most of which are very soluble and of little relevance to the archaeological record. Examples are lime, portlandite, larnite, and sylvite (Etiégni and Campbell 1991). Thus in terms of the archaeological record, freshly formed wood ash can be considered as being composed almost entirely of calcite, except for a few weight percent (0.3–3.7 weight percent as measured in 7 species of trees in Israel (Schiegl *et al.* 1994) of amorphous silica in the form of phytoliths and siliceous aggregates. In our experience the latter is much more abundant than the former. It should also be borne in mind that dry wood used for burning always contains adhering soil and dust, which will also be present in the ash.

The Major Mineral Components of the Hearths Analysed in Kebara Cave

The well preserved hearths in Kebara cave (Courty *et al.*, 1989; Bar–Yosef *et al.*, 1992) (Fig.2) are usually white, grey or yellow in color, oval– to circular–shaped with diameters of roughly 30 to 150 cm, and in section are lens–shaped with thicknesses varying from a few centimeters to tens of centimeters. They are usually underlain by a thin layer of charcoal–rich

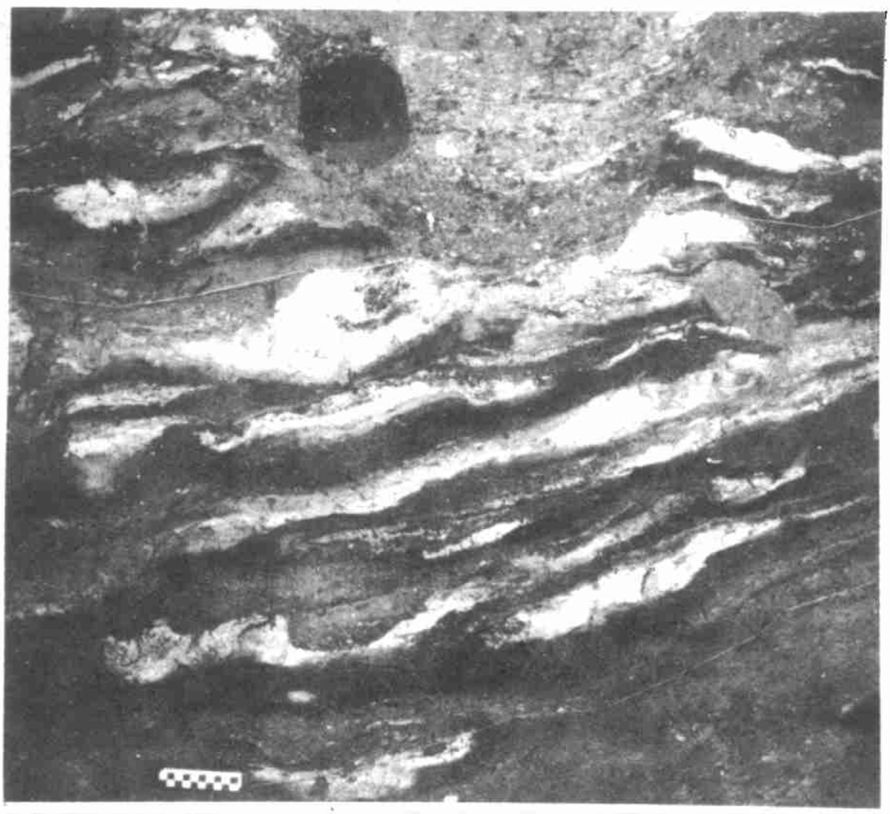

Fig 2. Photograph of the well preserved hearths in Kebara cave, showing their lens shape in section and the presence of dark charcoal–rich layers beneath the lighter colored ash–derived mineral layer

brownish black sediment, which interestingly is often limited only to the central region of the overlying ash remnants. This raises the possibility that the ashes themselves were dispersed over a larger area than the original hearth location.

The micromorphological characteristics of three of the Kebara cave hearths are described in detail in Courty *et al.* (1989). Two of the hearths are similar in texture, being dominated by a fine-grained groundmass of globular, light yellow to yellow brown nodules. The third contains as a major phase micritic calcite, which in part has altered to isotropic golden-yellow patches. Charcoal, red clay and quartz grains are present in fair abundance. Siliceous plant remains as well as burned bone fragments were also observed. A preliminary study of the charred wood remains from Kebara cave showed that two species of oak (*Quercus calliprinos* and *Quercus ithaburensis*) constitute 85% of the analyzed material. These two species are also the dominant elements of the present day arboreal vegetation (Baruch *et al.* 1992).

Table 1. Major Mineral Components of the Hearths Anslyzed in Kebara Cave

Mineral	Number of Hearths	Mineral	Number of Hearths
Calcite	7	Leucophosphite	6
Dahllite/ Francolite	13	Taranakite	2
Montgomeryite	14	Siliceous aggregates	17
Crandallite	1		

Table 2. The Minerals and their Chemical Formulae Discussed in the Text

Carbonates	
Calcite (C)*	$CaCO_3$
Dolomite	$(Ca, Mg)CO_3$
Phosphates	
Dahllite (D)	$Ca_5(PO_4, CO_3)_3(OH)$
Francolite (F)	$Ca_5(PO_4, CO_3)_3(F)$
Crandallite (CR)	$CaAl_3(PO_4)_2(OH) \cdot 5H_2O$
Montgomeryite (M)	$Ca_4MgAl_4(PO_4)_6 \cdot (OH)_4 \cdot 12H_2O$
Leucophosphite (L)	$K_2(Fe^{3+}, Al)_4(PO_4)_4 \cdot (OH)_2 \cdot 4H_2O$
Taranakite (T)	$H_6K_3Al_5(PO_4)_8 \cdot 18H_2O$
Variscite	$AlPO_4 \cdot 2H_2O$
Others	
Quartz	SiO_2
Lime	CaO
Portlandite	$Ca(OH)_2$
Larnite	$Ca_2(SiO_4)$
Sylvite	KCl
Siliceous aggregates (SA)	See Schiegl *et al.* (1995) for a detailed description

Table 1 lists the major mineral components found in hearths in Kebara cave and the number of hearths in which each of the minerals is the most abundant component. Table 2 lists the chemical formulae of the minerals in Table 1, as well as all the others mentioned in the text. The minerals were identified mainly by FTIR (see Schiegl *et al.* (1994) for more details of methodologies used). As fresh ash is composed primarily of calcite, we can assume that the seven calcite–dominated hearths are the best preserved. Dahllite or francolite are also present in these hearths, usually as crusts on the top surface, implying that in this environment calcite reacts with the phosphate–rich groundwaters and dissolves and reprecipitates as carbonated apatite(dahllite) or fluoridated carbonated apatite(francolite). Careful analyses of the other associated phosphate minerals in hearths and in the zones of reaction of dolomitic rocks buried in the Kebara cave sediments (Weiner *et al.*,1993; Weiner *et al.*,1995) suggest that the "reaction" sequence is as follows: dahllite or francolite is replaced by crandallite, which in turn is replaced by montgomeryite and finally leucophosphite. We do not know when taranakite forms in this

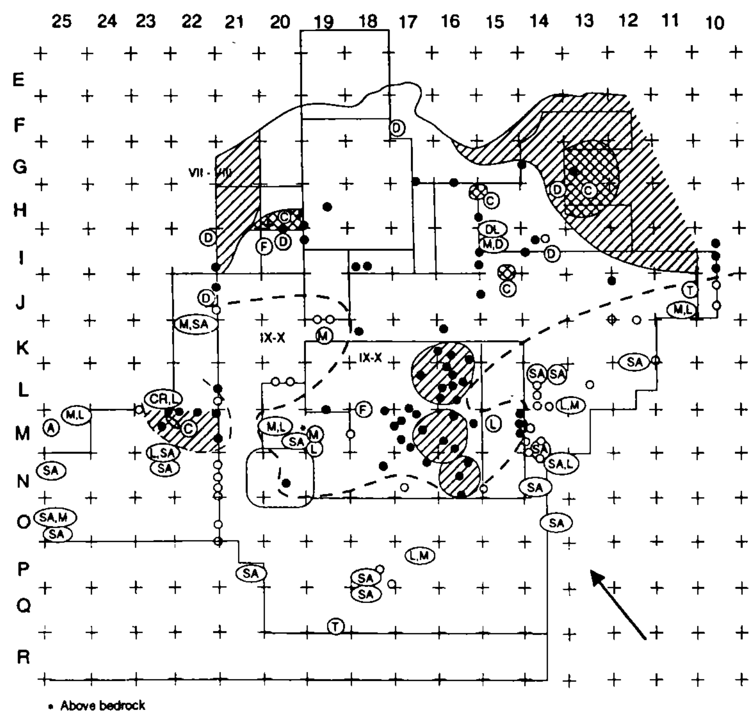

Figure 3. Map showing the mineralogical variations of the ash layers in the central and northern parts of Kebara cave. It also shows the boundaries of excavated areas (straight lines parallel to grid squares), the areas of dense bone accumulations (parallel–lined hatch) and the locations of sediments that contain traces of phosphate (filled circles) and those that do not (empty circles). The boundary between them is shown as a dashed line. The calcite ash layers are designated by the areas of cross–hatching and the letter "C". All the above information was reported by Weiner *et al.* (1993). The mineralogy of each of the ash layers is indicated by a one–letter code defined in Table 2. Each grid square is 1 m^2. The Roman numerals indicate some of the stratigraphic layers. Note that a very similar figure was published previously by Weiner *et al.* (1995)

sequence. It is also conceivable that at any stage the carbonate or phosphate minerals may dissolve in the ground water and be transported out of the cave. They are all more soluble than the siliceous aggregates and phytoliths in 6N HCl(Schiegl *et al.*,1994), suggesting that hearths composed only of the latter have been subject to the longest and/ or severest dissolution conditions.

Figure 3 shows a map of the distribution of many of the hearths analyzed in Kebara cave in relation to the apatite dissolution boundary mapped by Weiner *et al.* (1993). Note that the map combines information from stratigraphic units Ⅵ－Ⅷ, Ⅸ－Ⅹ and Ⅺ－ⅩⅢ. All the hearths in which siliceous aggregates are the major component are indeed south of the boundary, which is also the area in which no bones are preserved. The one exception (excavation square M20) is from a series of hearths located just above bed rock in Unit ⅩⅢ in the deep sounding. This observed distribution, therefore, strongly supports the notion that a relatively stable component in fresh ash is the siliceous aggregate fraction.

Ash Remnants in Hayonim Cave

Hayonim cave has been excavated intermittently since 1965 (Bar-Yosef 1991). The current ongoing excavation is aimed at investigating the Mousterian layers. To date, however, it contains very few hearths which can be recognized as such based on their macroscopic morphological characteristics. Charcoal is also almost absent in Hayonim cave and, in general, there is much less quartz in the sediments as compared to Kebara cave. There are also many areas containing abundant nodules composed of montgomeryite, leucophosphite and variscite,

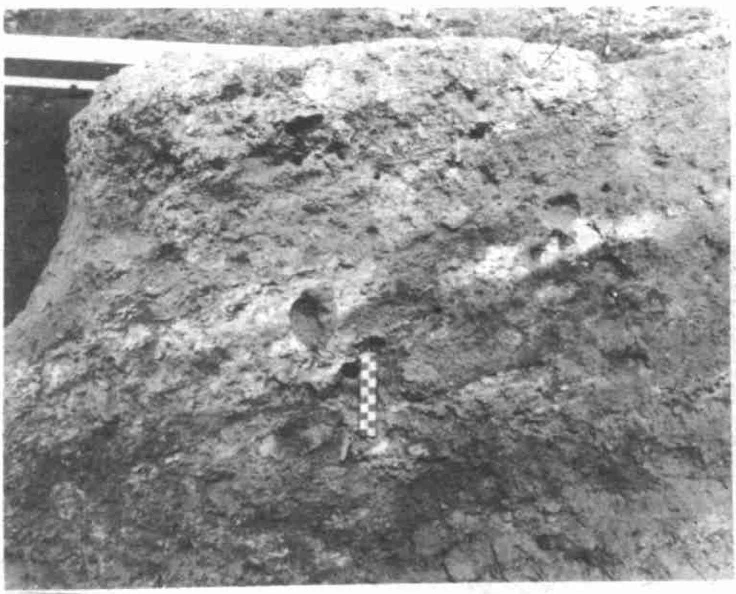

Figure 4.　Photograph of the ash-derived layers in Hayonim cave which are composed predominantly of siliceous aggregates

implying that phosphate-containing minerals have dissolved in one location and reprecipitated in another in the form of nodules. All the above factors probably contribute to the fact that hearths, as such, are poorly preserved in Hayonim cave.

A most surprising observation in terms of the mineral components in Hayonim cave, is that in the central part of the cave, the single most abundant component of the sediments is siliceous aggregates (Schiegl *et al.* 1994). These appear in the form of bedded layers that are either milky-white or grey in color (Fig. 4). Both varieties provide more or less the same infrared spectrum and it is not clear what is responsible for the color variation. The bedded sequence is a few meters thick in some places.

This observation implies that wood ash is an abundant component, if not the most abundant component, of the sediments that have accumulated in Hayonim cave. Furthermore, as only the siliceous aggregates are preserved in the central part of the cave, we can infer that the more soluble calcite-derived minerals have all dissolved and been removed from that area of the cave. As the latter constitute well over 90% by volume of the original ash (Schiegl *et al.* 1994), there must have been a huge reduction in volume of the sediments during this dissolution process. This, and other archaeological implications of these observations, will be discussed in detail elsewhere.

Recognizing Ash Deposits in the Archaeological Record

Very few prehistorically occupied caves have preserved ash deposits that can be unequivocally recognized as such by the naked eye. In contrast many caves contain burned flints and bones, clearly implying that fire was used at the site. One possible explanation for this is that the ash remains have been totally destroyed by diagenesis. Another possibility is that they are preserved, but have not been recognized as such. It is of interest to note that in an earlier micromorphological study of Hayonim cave, Goldberg (1979) observed the unusual nature of the sediments in the central part of the cave, but did not associate them with being ash-derived.

The current study demonstrates that ash can be found in a variety of different mineral forms, such as calcite, dahllite, francolite, crandallite, montgomeryite, leucophosphite and the so-called siliceous aggregates. If these minerals are found as bedded layers within the stratigraphic section (as opposed to concretions, nodules, or as reaction rims on the surfaces of rocks buried in the section), they should be considered as possibly being derived from ash. The presence of siliceous aggregates and/ or wood-derived phytoliths in these layers, would constitute strong evidence that the layers indeed originate from wood ash.

As always in archaeology, several words of caution are in order. Siliceous aggregates are known to be present in wood. It is not yet known, however, whether they are formed by all trees, or for that matter whether they are present exclusively in wood. If they are formed by other plants, their presence in the form of bedded layers in an archaeological site may be due to other processes. Siliceous aggregates themselves do most likely undergo diagenetic degredation, about which very little is known as yet. This could complicate the use of these materials for recognizing ash deposits. It is also conceivable that they may dissolve completely, and hence their absence does not prove that ashes were not present at a site. Based on our current knowledge, it seems unlikely that siliceous aggregates will dissolve before the other carbonate– or phosphate–containing ash–derived minerals, and hence can be expected to be associated with the latter in bedded layers.

Concluding Comments

Recognizing ash in the archaeological record is by no means simple, especially when the ash has been dispersed and subjected to diagenesis. The mineral components of ash can be useful in this regard. Identifying the presence of prehistoric ash is a prerequisite for recognizing hearths. As the latter are the products of deliberate human–made fires, they are undoubtedly an important cultural attribute in human evolution. Much too can be learned from the charcoal, provided that it is preserved in a reasonable state. It is hoped that this information will contribute to an improved understanding of the use of fire by hominids.

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