Supplement 2 to

Geoarchaeological Investigation in a Domestic Iron Age Quarter, Tel Megiddo, Israel

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Materials and Methods

All reference to figures and tables in this section should be sought in the article cited above.

Micromorphology

Undisturbed monolithic sediment blocks were sampled using jackets of plaster of Paris. The blocks were dried in an oven (50 °C) prior to impregnation using an 8:2 mixture of polyester resin with acetone and 1% v:v MEKP. Pre-cut sample slices were commercially prepared into 30 μm thick thin sections by Quality Thin Sections Ltd., Tucson, Arizona, and Spectrum Petrographics Ltd., Vancouver, Washington. Thin sections were studied using polarizing light microscopes (Nikon Labophot2 and Nikon Eclipse 50iPOL) at various magnifications. Micromorphological descriptions follow the terminology of Marie-Agnes Courty, Paul Goldberg, and Richard Macphail (1989) and George Stoops (2003).

Fourier Transform Infrared Spectrometry (FTIR)

A few milligrams of sample were homogenized and ground in an agate mortar and pestle. About 0.3 mg were left in the mortar, mixed with about 40 mg of potassium bromide, and pressed into a 7 mm pellet using a hand press (Qwik Handi-Press, Spectra-Tech Industries Corporation) or a hydraulic press (Specac). Infrared spectra were obtained at 4 cm⁻¹ resolution for 32 scans. On-site analyses were conducted using a Nicolet IR200 and a Nicolet iS5 (Thermo), while in the Kimmel Center for Archaeological Science of the Weizmann Institute of Science, analyses were conducted using a Nicolet 380 (Thermo).

Interpretation of spectra was aided by an internal reference library (Weiner 2010). Two methods that were de-veloped at the Kimmel Center were regularly used to enhance interpretation: one relates to clay minerals and the other to the mineral calcite—both are quite common in the tell's sediments. Changes in the infrared spectrum of clay minerals provide means to determine whether clay minerals have been exposed to high temperatures (Berna et al. 2007; Shoval, Yadin, and Panczer 2011; Eliyahu-Behar et al. 2012). Due to site-specific differences in clay mineralogy of local soils, it was necessary to experimentally calibrate the spectral changes induced by heat to soils from the vicinity of Megiddo. Material from an unheated mud brick from Level Q-5 has been used for the experimental heating. About 1 g of mud-brick material was heated in a furnace oven (manufactured by Adam Mandel Furnaces and Systems) to various temperatures between 500 and 800 °C, each for 4 hours. The infrared spectra of the experimentally heated sediments were obtained upon cooling the sample to room temperature. **Figure 3** presents the results, showing that the composition of unheated mud brick (**Fig. 3a**) includes clay and calcite. Upon heating to 500 °C, the clay spectrum changes whereby the main clay absorption shifts from 1,034 to 1,038 cm⁻¹, bands related to the clay structural water—in the range of 3,600 cm⁻¹— disappears (circled in **Fig. 3b**), and the absorption at 519 cm⁻¹ (circled in **Fig. 3b**) lowers. The calcite spectrum is unchanged at this temperature. Further heating to 600 °C results in further shifting of the clay main absorption band and no change in the calcite spectrum (**Fig. 3c**), while a further heating to 650 °C results in the partial decomposition of calcite and the formation of calcium hydroxide identified by the absorption at 3,645 cm⁻¹ (**Fig. 3d**). At this point, it was necessary to remove cal-

cite from the mud-brick material (by dissolution using 1N hydrochloric acid) in order to observe the changes in the infrared spectrum of heated clay alone (**Fig. 3e, f**). Heating this decalcified mud-brick material to 700 and 800 °C resulted in pronounced shifts in the main absorption of the clay to 1,057 and 1,082 cm^{-1}, respectively. As mud bricks in antiquity did not burn after decalcification, we also heated pristine mud-brick material to 800 °C and obtained the infrared spectrum of sintering between clay and calcite (**Fig. 3g**), characterized by a broad main absorption and a small amount of calcium hydroxide.

FTIR analysis is also sensitive to heat-induced changes in the infrared spectrum of calcite. When calcite is heated to ca. 800 °C, it decomposes to calcium oxide that readily absorbs water to form calcium hydroxide. With time, carbon dioxide replaces the water molecule to form calcite again (Boynton 1980). This is the process of lime plaster/ mortar making. It has been shown that geogenic calcite (e.g., limestone, chalk, etc.) can be distinguished from pyrogenic calcite (e.g., plaster and mortar) by using the "grinding curve" method (Regev et al. 2010). Such changes were determined as follows: The mineral calcite has three characteristic infrared absorbance peaks in the 400–4,000 cm^{-1} range: ν_2 at 874 cm^{-1}, ν_3 at 1,420 cm^{-1}, and ν_4 at 712 cm^{-1}. The heights of those peaks were determined following Vikki Chu et al. (2008). To distinguish between geogenic and pyrogenic calcites, the height ratio of the ν_2/ν_3 peaks was plotted against the ν_4/ν_3 height ratio on a "grinding curve" plot (Regev et al. 2010; Poduska et al. 2011), using various samples: a calcite spar single crystal sample; limestone and chalk samples from Tel Megiddo itself and its vicinity; modern and archaeological ash samples; and modern lime plaster (**Fig. 4**). This shows distinct spectral differences between geogenic limestone and lime plaster (i.e., clearly separated curves), yet the wood ash and geogenic chalk curves overlap, indicating that these cannot be distinguished based solely on the parameters given in **Figure 4**. To differentiate between these materials, we relied on microscopy and micromorphology. Combining the methods of clay and calcite characterization using FTIR spectroscopy enhances interpretation by providing ranges that correspond to the intensity of heat to which these minerals have been exposed in the past.

A third mineral that is useful for determining human activities is the phosphate-containing mineral carbonated hydroxyl apatite (hereafter CHAP), readily identifiable using FTIR spectroscopy. This mineral originates either from bones and teeth or from the recombination of phosphate released during organic matter decomposition when calcium carbonate is already present within soils or sediments (Holliday and Gartner 2007). It has two characteristic absorbance bands at 605 cm^{-1} and 565 cm^{-1}. We carried out a small experiment by which we formed a quantitative calibration of calcium phosphate in geogenic and heated clay-rich sediments. The experiment was conducted as follows: 1N hydrochloric acid was added to clay-rich sediment until carbonates were completely dissolved (i.e., until fizzing stopped). The decalcified sediment was dried at 50 °C in an oven. Half of this sediment was heated in a furnace at 600 °C for 4 hours. Commercial calcium phosphate powder (Baker) was weighed in predetermined aliquots and mixed thoroughly with the dry decalcified sediment as well as with the heated sediment. The mixtures of calcium phosphate and sediment were pre-calculated so that the following weight percentages of phosphate within the prepared sediments were obtained: 0.5, 1, 2, 3, 5, and 10%. Each of the mixtures was then prepared for FTIR analysis using the potassium bromide method described above. The spectra obtained showed that the characteristic absorbance bands at 605 cm^{-1} and 565 cm^{-1} increase in height with the phosphate concentration (**Fig. 5**). This allowed us to define two broad categories that relate to the concentration of CHAP in sediment samples: "high," identified when both bands have clear maxima (above a phosphate concentration of 2–3%), and "low," when these bands appear as a shoulder rather than a clearly defined absorbance band (below a phosphate concentration of 2–3%).

Phytolith Analysis

Phytoliths have been extracted and quantified using the well-established rapid procedure of Ofir Katz et al. (2010), based on density separation using a 2.4 g/ml solution of sodium polytungstate. Results were available on a next-day basis during the excavation, and samples were further quantified in the laboratory after the excavation using the same procedure. A total of 113 sediment samples have been analyzed. Phytoliths were counted and identified at 200X and 400X using Nikon Eclipse 50i Pol and Olympus BX-41 optical microscopes. Phytolith identification was carried out using the standard literature (Twiss, Suess, and Smith 1969; Mulholland and Rapp 1992; Madella, Alexandre, and Ball 2005). To calculate the percentage of phytoliths identified in anatomical connection, phytoliths forming a single multicellular structure were counted individually and later added to the general count.

To evaluate whether grass-dominated phytolith assemblages represent whole plants or a selection of certain plant organs (i.e., a selection of inflorescence [chaff], on the one hand, or a selection of leaves and stems [hay, straw] on the other), seven common Mediterranean modern grass species were collected (with repetition on *Triticum aestivum* from two different localities in Israel that differ in annual precipitation). This new approach is preliminary, and future work will target more species. A quantitative

measure based on phytolith morphologies has been produced. Between three to five separate mature whole plant specimens were cut and mixed together from each species. The mixtures were washed in tap water, dried in the laboratory (50 °C for 3 days), and their dry weight recorded. Weighed aliquots from each plant mix were then dry-ashed at 500 °C for 4 hours. The resulting ash, composed primarily of phytoliths and small quantities of calcite, was pooled together and lightly washed with diluted acid (1N hydrochloric acid), dried, and weighed.

Three different slides from the phytolith assemblages were prepared for microscopic observation. The ratio of leaves/stems to inflorescence was calculated by counting phytolith morphologies that are distinctive for these plant organs (e.g., dendritic long cells from inflorescence, bulliform cells from leaves, etc.). Phytolith morphologies that are known to be present in all grass organs (e.g., short cells) were not included in this calculation. In each sample, around 500 phytoliths were counted in order to produce a statistically significant ratio. Thus, three ratios have been calculated per species. The averaged ratios, with a 1Σ standard deviation, are presented in **Table 1**. The data show that in all studied species, the averaged ratios are always above 1, indicating that whole plants produce more identifiable leaf/stem phytoliths than inflorescence phytoliths. Thus, selection of inflorescence grass parts will be evident by a ratio below 1.

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