[1]**Order the numbering of subheadings**

Paleomagnetic geochronology of Quaternary sequences in the Levant[1]

Ron Shaar[1], Erez Ben-Yosef[2]

[1] The Institute of Earth Sciences, The Hebrew University of Jerusalem, Jerusalem, 91904, Israel. ron.shaar@mail.huji.ac.il

[2] Department of Archaeology and Near Eastern Cultures, Tel Aviv University, Tel-Aviv, 6997801, Israel. ebenyose@post.tau.ac.il

X.1 Introduction

Paleomagnetic dating methods are based on comparing magnetic information from materials or sequences whose ages are at least partly unknown with the geomagnetic chronology. The global Quaternary geomagnetic chronology is continuously updated and includes at the moment 10 polarity reversals and at least 18 validated geomagnetic excursions that serve as chronological markers for sedimentary and volcanic sequences. In addition, short-term secular variations of the past several millennia can be used for the Holocene. Here, we review the principles underlying paleomagnetic chronology with emphasis on Quaternary rocks, sediments, and archaeological substances. We summarize a number of successful applications of paleomagnetic dating in the Levant, and provide insight into future possibilities of Quaternary paleomagnetism.

[1] This chapter is dedicated to the memory of our late mentor and dear friend Professor Hagai Ron who has pioneered paleomagnetic research in the region

Earth magnetic field is constantly changing at decades to millions of years time scales. The effort of reconstructing past geomagnetic variations have been motivated mainly by a geophysical interest to understand the behavior of Earth's magnetic field, its origin, and its impact on Earth's atmosphere and biosphere. The outcome of this research is a fairly robust geomagnetic record applicable as an independent chronological reference framework.

To apply paleomagnetism for dating, a robust geomagnetic chronology needs to be established first by compiling global paleomagnetic database into three independent variation records: 1) reversals, 2) excursions, and 3) short-term (secular) changes in direction and intensity. Then, on the basis of comparison with the appropriate reference chronology the magnetic polarity, direction, and intensity of materials or sequences are used obtain age estimates.

Paleomagnetism is a powerful chronological tool used for decades (Singer, 2014, Sternberg, 1997). Yet, it is important to emphasize that it involves a series of experimental procedures that require several stages of interpretations. Also, it is based on the reference geomagnetic chronology to date. Hence, when adopting paleomagnetism understanding of how it works is required. Here we review the principles underlying the paleomagnetic methods so that readers could adopt a realistic and critical view and adequately design paleomagnetic dating applications in their research. In Section X.2 we briefly explain how different materials record magnetic information. In Section X.3 we outline the basic paleomagnetic laboratory procedures. In Section X.4 we review the up to date geomagnetic chronologies. Finally, in Section 5 we review the application of paleomagnetic dating in selected geological and archaeological studies of the Levant. **X.2 Principles of natural magnetic recording**

Ferromagnetic minerals (for simplicity we group different types of magnetic minerals; ferro-, ferri-, anti-ferro, canted-, and defect- magnetism under one term, ***ferromagnetic***) are the fundamental building blocks of magnetic recording. Ferromagnetic minerals have an intrinsic spontaneous magnetization, which makes them act as small-scale "compass needles". The ferromagnetic minerals that are relevant to paleomagnetism are those whose magnetization is locked in space at room temperature. The most abundant groups of ferromagnetic minerals are Iron oxides: magnetite (Fe_3O_4), hematite (Fe_2O_3), and maghemite (γ-Fe_2O_3); Iron sulfides: greigite (Fe_3S_4), pyrrhotite, ($Fe_{1-x}S_x$), and Iron oxyhydroxides (e.g. Goethite, $\alpha FeOOH$). These groups can appear in their pure form or with substitutes. They are so abundant that they can be virtually found everywhere: in rocks, sediments, soils, dust, and various archaeological materials.

The mechanisms by which ferromagnetic minerals record and retain magnetic information (called hereafter ***Remanent Magnetization***) can be classified to: thermomagnetic (***TRM*** – magnetization acquired by cooling), depositional (***DRM*** - physical settling of magnetic particles in sedimentary environments), and Chemical (***CRM*** – chemical growth of magnetic minerals). Figure X.1 illustrates graphically these mechanisms. For further reading see Dunlop and Özdemir (2001) and Tauxe (2010). TRM (Figure X.1a) is acquired in igneous rocks and fired archaeological materials when ferromagnetic minerals are cooled through a critical temperature called the ***blocking temperature (T_B)***. In T_B the spontaneous magnetization becomes locked in space and a magnetization is gained in a direction parallel to the ambient field. DRM (Figure X.1b) can be acquired in marine, lake, and aeolian environments. DRM occurs when ferromagnetic particles align in the direction of the ambient field before settling. CRM (Figure X.1c) is acquired when new ferromagnetic grains grow and possess magnetization. In addition to the mechanisms shown in Fig. X.1 other processes contribute to the net magnetization of a material. The stability (or the 'decay' time) of the magnetization, which depends on the age, mineralogy, and size of the particles, may lead to an acquisition of a "viscous" magnetization (***VRM***). Also, various physical and chemical alterations may distort, erase, and even re-magnetize an existing magnetic signal through time.

To exemplify the concept and complexity of paleomagnetic recording we consider three lithologies relevant to Quaternary sequences as case studies: a lava flow, a lake varve, and a buried soil. Ideally, the lava flow would have a TRM recorded on the day the basalt cooled, the lake varve would have a pure DRM that got locked over a season, and the buried soil would have a CRM acquired over a specific polarity chron (see below for definitions of chrons). In these examples the original primary magnetization is tied directly to the age of the material, and is referred to hereafter as the Characteristic Remanent Magnetization (***ChRM***). In practice, the ChRM is often overprinted by many magnetic signals postdating the ChRM. For example: The basalt can acquire 'viscous' magnetization (VRM), can be demagnetized by chemical and physical weathering (CRM), and can even be struck by a lightning (that totally erases the original TRM); the lake varve can lose its DRM by bioturbation, oxidation, dissolution, and alteration (Figure X.1b), and can be masked by new ferromagnetic minerals growing in the sedimentary column (i.e. CRM); the buried soil can have a "soft" and unstable CRM that decay with time or be masked by new CRM that continues to form as the soil aged.

X.3 Basic paleomagnetic procedures

Given the complexity of the natural magnetic acquisition (Section X.2), there are some questions that should be properly addressed before one uses paleomagnetism: 1) "***Which*** minerals carry the magnetic signal?", 2) **"*What*** are the processes by which the magnetic signal was acquired?", and 3) **"*When*** was the magnetic signal acquired?". Also, obviously, we need a methodology for measuring the ChrM and isolating it from all possible overprints. Assessing these questions is the art of the paleomagnetic lab work.

There are different parameters and levels of paleomagnetic information that can be retrieved by paleomagnetic analyses. In some cases, the desired information would be only the polarity of the ancient field ('***Normal'*** or '***Reverse'***, Section X.4). In other cases, the precise direction and/or intensity of the ancient field should be precisely recovered.

Almost all types of paleomagnetic investigations require isolation of the ChRM. This is done by progressively demagnetizing the net magnetization (Natural Remanent Magnetization, ***NRM***) of the sample. Demagnetization can be done thermally, by heating the sample in zero field conditions, or magnetically, by exposing the sample to a strong Alternating magnetic Field (***AF***). After each demagnetization step a portion of the NRM vector is lost and the x, y, z components of the remaining vector are measured with a high sensitivity magnetometer.

Consecutive measurements of the remaining NRMs are plotted on an orthogonal plot where the length of the magnetization vector is normalized to the length of the initial NRM. An orthogonal plot that appears as a straight line converging toward the origin (Fig. X.2) means that the direction of the vector remained the same after all the demagnetization steps. In this case the ChRM is calculated through Principle Component Analysis (kirschvink, 1980) . A more complicated orthogonal plot should be interpreted with care. A robust demagnetization procedure would include both AF and Thermal techniques. An example is shown in Fig. X.2.

The paleomagnetic vector is described by the ***declination***, deviation from the geographic north, and ***inclination***, the angle between the vector and the horizontal plane. The paleomagnetic direction is usually calculated as a mean of several samples collected from the same 'site', where a 'site' is a unit representing the same point in time, e.g. a lava flow or a particular horizon in a sedimentary sequence. The site paleomagnetic mean is calculated via Fisher statistics (Fisher, 1953). Similarly, one can calculate the paleomagnetic mean of a locality by a Fisher mean of several sites.

Estimating the intensity of the ancient field (***paleointensity***) is by far more complicated than estimating the paleomagnetic direction. Experimental principles of paleointensity are beyond the scope of this chapter [see Valet (2003); Tauxe and Yamazaki (2007); Shaar et al. (2010); Shaar and Tauxe (2013)], but it is the basis of a relatively new archaeomagnetic chronological tool for the Holocene (Section X.4.2).

As mentioned above, a robust paleomagnetic analysis would not be complete without assessment of the “which”, “what”, and “when” questions concerning the ancient magnetization. Yet, this is far from being a trivial task. Above all, the fundamental data to consider are the geological setting, the detailed lithology, and the history of the system (formation and post-depositional). Then, there is a wide range of magnetic experimental procedures and microscopic observations that can assist in characterizing the magnetic mineralogy, grain size distributions, magnetization stability, and the origin of the magnetization (Dunlop and Özdemir, 2001, Liu et al., 2012, Tauxe, 2010). There is no simple recipe for this so-called “rock-magnetic analysis”, and its scope depends on the specific requirements of the study. Also, interpreting rock-magnetic data is not always simple. It is likely that some uncertainties concerning the rock-magnetic properties of the material will still remain open after rigorous analysis. However, we stress that paleomagentic interpretations based on inadequate assessment of the source, the age, and the stability of the magnetization should always be taken with extreme caution.

X.4 Geomagnetic chronology

X.4.1 Geomagnetic Polarity/Instability Time Scales

Geomagnetic reversals are one of the most prominent chronological markers in the Quaternary. During a reversal the polarity of the field rapidly switches between N/R states, where N (Normal) stands for northerly oriented field and R (Reverse) stands for southerly oriented field. The duration of a reversal is less than 10^4 years, perhaps even less than 10^3 years (Valet, 2003). The reversal chronology, known as the ‘Geomagnetic Polarity Time Scale’ (***GPTS***) started to develop in the early 1960s [e.g. Cox et al. (1964)], when Quaternary GPTS included only two polarity 'chrons', and one short-term event. The chrons were initially named after pioneers in geomagnetism (Brunhes, Matuyama, and Gauss, Cox et al. (1964), Fig. X.3). With time, more reversals were discovered and the nomenclature of GPTS became too complicated to maintain. The currently accepted terminology divides the Quaternary to Chrons and subchrons (Cande and Kent, 1995, Gee and Kent, 2007). Some of the subchrons are still known by their initial names (Jaramillo, Cobb Mountain, Olduvai, Reunion/Feni), and this nomenclature is still in widespread use. We note, that GPTS terminology in the published literature, especially for short-term events in the Matuyama, such as for example, Cobb Mountain, Gilsa, and Reunion, is not always consistent [for further reading see Gee and Kent (2007); Tauxe (2010); Singer (2014)]. As the age accuracy of the GPTS is of great importance in Quaternary research, it is continuously updated using new high quality data from sedimentary, oceanic and volcanic sources (Gee and Kent, 2007, Gradstein et al., 2012, Singer, 2014). Recently published Quaternary GPTS slightly differ from each other and display some minor discrepancies reflecting that the precise chronology is a work in progress.

Geomagnetic excursions are additional important detectable chronological markers. Excursions are brief (< 10^4 years) and substantial deviations of field direction from northerly / southerly oriented geomagnetic field [>45° declinational change Laj and Channell (2007)], accompanied with a significantly low field intensity (Roberts, 2008). As excursions are short events they are hard to detect, and therefore, there is controversy in the literature regarding some of the published

Quaternary excursions. We show in Fig. X.3 excursions validated by Laj and Channell (2007) and Roberts (2008).

X.4.2 Holocene geomagnetic secular variations

A relatively new addition to the paleomagnetic geochronology toolbox is regional curves of geomagnetic secular variations. Secular variations are short term (<10^5 years) changes in the direction and intensity of the field. If a regional secular variation curve is established for a given time interval then paleomagnetic dating can be applied by comparing direction and/or intensity of material/sequences whose age is unknown with the reference secular variation curve. While this concept is not new, its application requires a robust and accurate secular variations reference curves. Shaar and Ben-Yosef have compiled such a curve for the Levantine intensity using a range of well-dated archaeological materials. This is a work in progress, and its initial version is illustrated in Fig. X.4.

X.5 Applications of paleomagnetic geochronology in the Levant

Paleomagnetic chronology has been applied in a number of Levantine sequences where direct absolute ages could not be accomplished, or as a complementary method. Here, we briefly review some successful applications of paleomagnetic chronology in geological and archaeological contexts. Notably, the most densely sampled area is within Israel where the late Prof. Hagai Ron has established a laboratory in 1980 (Fig. X.5).

X.5.1 Paleomagnetic dating of geological units

<u>Volcanic sections</u>

In a pioneering application Freund et al. (1965) conducted one of the first worldwide paleomagnetic chronostratigraphic applications studying the Plio-Pleistocene volcanic field in Northern Israel. This volcanic field was later densely sampled for both paleomagnetic and radiometric analyses (Heimann and Ron, 1993, Heimann et al., 1996, Hurwitz et al., 1999, Ron et al., 1984, Ron et al., 1992). Magnetostratigraphy was used by these authors to refine K-Ar ages of successive lava flows (e.g. Nahal Orvim, Nahal Ashaf), infer ages of fluvial units (e.g. Nahal Hazor), and correlate different sections in the Golan Heights and Korazim block (Heimann and Ron, 1993). These two methods were combined by dating few of the flows in each sequence, including the uppermost and the lowermost ones, and then using them as anchors for GPTS correlation of all flows. The volcanic field outside of the Golan Heights was less studied. Vandonge et al. (1967) and Gregor et al. (1974) investigated volcanic units in Syria and Lebanon. Abou-Deeb et al. (1999) used paleomagnetic directions and polarities to obtain a chronological differentiation between independent volcanic units in Syria.

Loess and buried soils

Aeolian deposits and buried soils carry excellent magnetic signal. Yet, their net magnetization is a complex combination of different DRM, post depositional DRM (pDRM) and CRM mechanisms acting over a relatively long period (Evans and Heller, 2001, Liu et al., 2007). Thus, magnetostratigraphy analysis of loess and buried soils should take into account possible paleomagnetic delay (lock-in depth) in the order of 10^5 years. Despite this complexity, magnetostratigraphy was successfully applied on different types of buried soils, loess, and Hamra soil in Israel:

Evron quarry (Ron et al., 2003), Revadim Quearry (Gvirtzman et al., 1999, Marder et al., 1999), and Ruhama badlands (Laukhin et al., 2001, Ron and Gvirtzman, 2001).

Calcretes

Mashiah et al. (2009) recognized that calcrete (consolidated carbonate soil horizons) carry stable CRM. They used this phenomenon to date successive sections of calcretes in the base of Mt. Carmal via magnetostratigraphy as mean to put temporal constraints on the development of the Western Carmel escarpment. Zilberman et al. (2011) and Zilberman (2013) used similar approach to investigate calcretes overlying faults in Western Gallile and Modiin to constrain termination of the tectonic activity. Matmon et al. (2010) applied paleomagnetic methods to investigate the temporal evolution successive colluvial units in the Zurim escarpment, Bet-Kerem Valley.

Lacustrine deposits.

Paleomagnetism of the thick lacustrine sequence of the Dead Sea Group has been extensively studied, including the Sedom, Erk el Ahmar, 'Ubeidiya, Lisan, and Zeelim Formations (e.g., Matmon and Zilberman, this volume, Bar-Yosef and Belmaker , this volume; Stein, this volume) . Key paleomagnetic studies are reviewed here as examples.

Weinberger et al. (1997) employed paleomagetism for reconstructing tectonic evolution of the Sedom diapir. In their analysis they isolated a secondary Brunhes age normal polarity CRM. This CRM postdates the main tilt and constraints the termination of diapir emplacement.

The magnetostratigraphy of Erk el Ahmar (EEA) Formation was investigated in three different works (Braun et al., 1991, Davis et al., 2011, Ron and Levi, 2001). There are only minor differences between the paleomagnetic profiles and they all well agree with the initial profile of Braun et al. (1991). Yet, the paleomagnetic age interpretations given in the above publications are different. This exemplifies the dependency of magnetostratigraphy on absolute dates at tie points. Using an estimated biostratigraphic age of 1.5-2 Ma, Braun et al. (1991) correlated the EEA Normal polarity to either Olduvai or Reunion subchrons. Later, Ron and Levi (2001) obtained a more detailed inclination profile concluding that the subchron is more likely Olduvai. Recently, Davis et al. (2011) obtained eleven cosmogenic burial ages of 3.5-5.3 Ma, pushing the magetostraigraphy sequence to the Gauss or Gilbert Chrons. Magnetostratigraphy analysis of the younger 'Ubeidiya Formation, located few km north of Erk el Ahmar yielded four reversals events. Given faunal constraints of about 1.4 Ma, Sagi (2005) provided several possible age interpretations, all within the Matuyama Chron.

Sections of the late Pleistocene Lisan Formation near Nahal Prazim, Massada, and southern Sea of Galilee (Lake Kinneret) (Ohalo; Nadel, this volume) were studied for paleomagnetsim for recovering short-term secular variations of the geomagnetic field (Marco et al., 1998, Marco et al., 1999, Marco, 2002). Using AF and Thermal demagnetization procedures they isolated the paleomagnetic direction, and obtained detailed secular variations profiles revealing two excursions in Perazim section (Marco et al., 1998) and one excursion in Ohalo (Marco, 2002). Similar investigations targeted the Holocene in Lake Kinneret (Thompson et al.,

1985) and in the Dead Sea exposure and core data (Frank et al., 2007a, Frank et al., 2007b, Segal, 2003).

Despite the success of the studies above, questions regarding stability and origin of the magnetization in the Dead sea sediments were not raised until recently (Frank et al., 2007a, Frank et al., 2007b, Nowaczyk, 2011, Ron et al., 2006, Ron et al., 2007). Using thorough rock magnetic and microscopic investigation of the Lisan and Zeelim Formations they deciphered complex histories of magnetic acquisition. Ron et al. (2006) suggested a three-stage model for the complicated magnetization: 1) DRM acquisition of fine magnetite particles, 2) CRM acquisition of greigite accompanying the dissolution of the primary magnetite, and 3) Oxidation of the greigite following outcrop exposure. This model suggests that paleomagnetic data from Dead Sea rift lacustrine deposits should be cautiously examined.

Only few sedimentary Quaternary sequences outside the Dead Sea basin were studied using paleomagnetic methods. Frank et al. (2002) investigated Holocene paleomagnetic secular variations in Birkat Ram, the Golan Heights. Develle et al. (2011) used inclination anomalies derived Yammouneh basin core, Lebanon, to establish a paleomagnetic chronology based on excursion markers.

X.5.2 Archaeomagnetism

During the past several decades, Levantine archaeological research involved paleomagnetic research, predominantly for constraining ages of archaeological finds. The GPTS records obtained from sedimentary and volcanic sequences associated with archaeological material have been used to estimate ages of several important prehistoric sites, some of which are key for understanding early human evolution and migration routes. The age of 'Ubeidiya (Bar-Yosef and Belmaker, this volume), the earliest site in the Levant and one of the earliest outside of Africa, was further constrained by paleomagnetic study of the 'Ubeidiya sequence to specific time intervals [1.55-1.2Ma or 1.2-1Ma (Sagi, 2005)] . It has been suggested that a nearby site, Erk el-Ahmar, is even earlier [>2Ma, see above and Davis et al. (2011) Matmon and Zilberman, this volume]. However, the magnetostratigraphyhas no paleoanthropological material and the association of the Erk al-Ahmar Formation with such material is debated. Another key prehistoric site is Gesher Benot Ya'akov (GBY) (Goren-Inbar, this volume). The site, an accumulation of artefacts and ecofacts along a thick sedimentary sequence, was deposited through the Brunhes/Matuyama transition [~780 ka, (Goren-Inbar et al., 2000, Goren-Inbar et al., 1992)]. Similar to 'Ubeidiya, GBY is also in the Rift Valley of northern Israel, along which hominin expansion northward probably took place. The paleomagnetic stratigraphy contributes to our understanding of this process, and possibly also to the question of the timing of the first deliberate use of fire [see Goren-Inbar et al. (2004)]. Other Lower Paleolithic excavations successfully applying magnetostratigraphy are Evron Quarry [Middle Acheulian, Ron et al. (2003)], Bizat Ruhama (Laukhin et al., 2001, Ron and Gvirtzman, 2001), and Late Acheulian Revadim Quarry (Gvirtzman et al., 1999, Marder et al., 1999). However, considering the importance of the Levant in early prehistory and the substantial number of key sites subjected to systematic excavations, the number of reported sites applying paleomagnetism is surprisingly low.

While the GPTS is the basis for constraining the age of prehistoric sites, secular variations records are used for dating younger sites (usually not earlier than the Pottery Neolithic, the time when ceramic technology was introduced). However, the application of the latter is still limited as the resolution of the published records is currently low (Fig. X.4). Improvements of resolution and accuracy of the reference curves for the Holocene is an ongoing effort (Figs. X.4, X.5). Such efforts are taking advantage of the rich archaeological landscape of the Levant [based on well-dated heat-impacted archaeological materials, e.g., Ben-Yosef et al. (2008b) , Gallet et al. (2006), Shaar et al. (2011). Recently the availability of the Levantine Archaeomagnetic Compilation (LAC) data consisting primarily of intensity values was instrumental in constraining the age of important archaeometallurgical sites, some of which considered the earliest in the Levant if not anywhere. While some of these results corroborate the early age of smelting in the southern Levant(Ben-Yosef et al., 2008a), others(Ben-Yosef et al., 2010) contradict proposals for Neolithic copper smelting in the region. As the resolution of the LAC improves, its application in young archaeological contexts will become more effective, with potential to become a potent dating tool in the research of this archaeologically-rich region.

Acknowledgements

Working on this review exposed us to substantial amount of published and unpublished data produced by the meticulous research of our teacher and mentor, the late Prof. Hagai Ron, who passed away in 2012 after a short battle with a vicious disease. Hagai was one of the worldwide pioneers in applying paleomagnetism. His

profound and longstanding impact on the paleomagnetic research of the Levant goes far beyond this review chapter.

We thank A. Agnon, Shmulik Marco and Ofer Marder for comments and reviews.

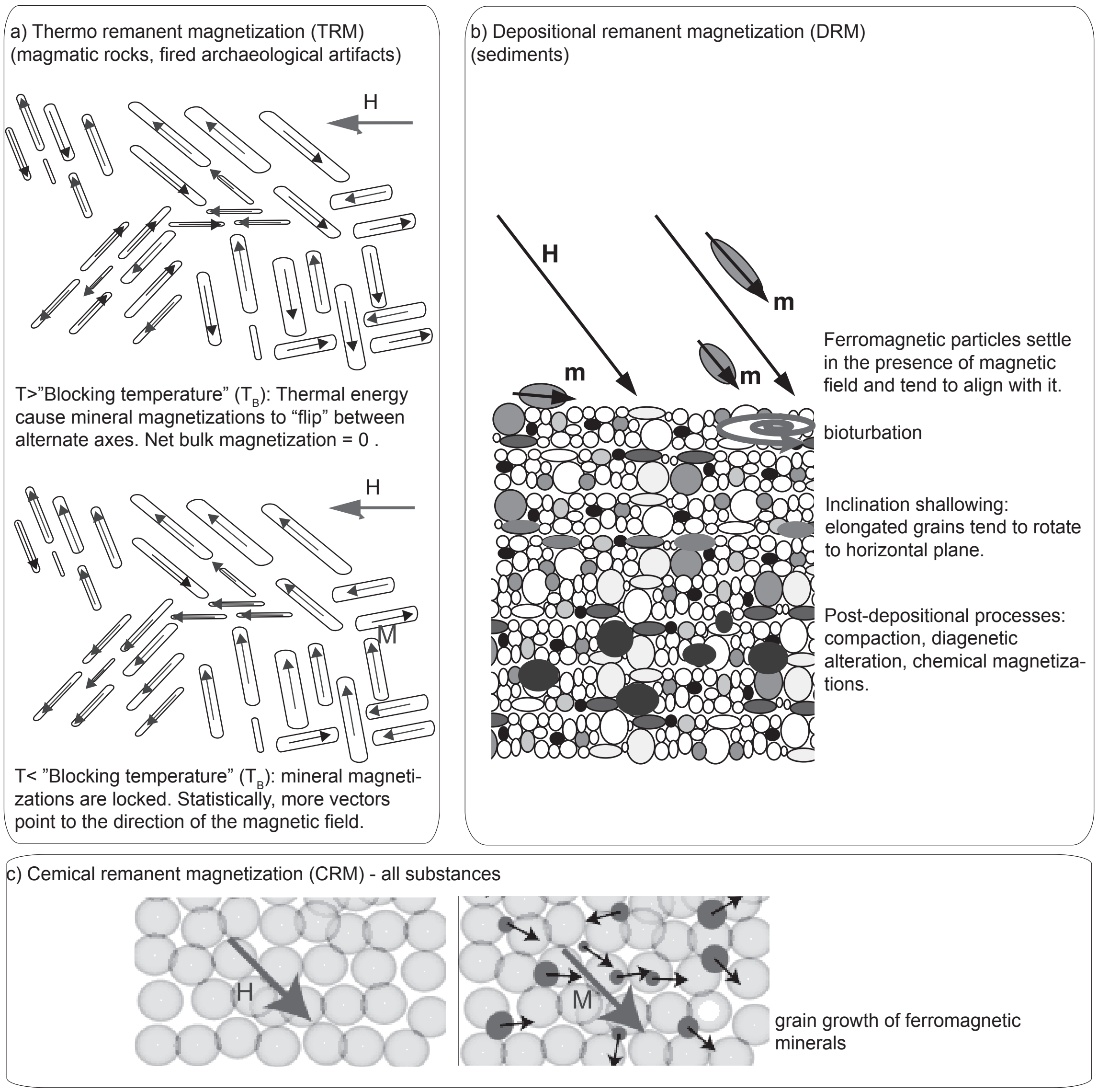

Figure X.1: Schematic illustration of the three main mechanism of magnetic remanence acquisition. Redrawn after Butler (1992) and Tauxe (2010)

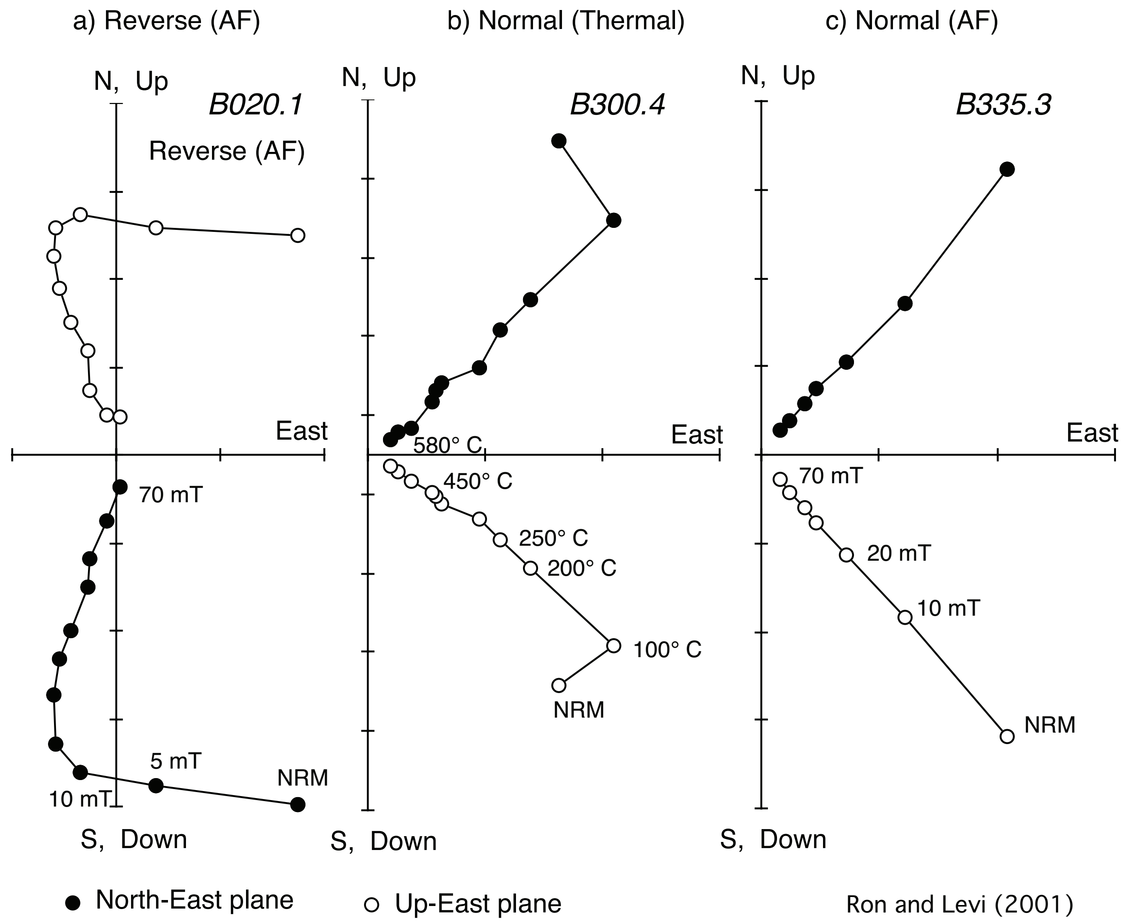

Figure X.2: Examples of demagnetization experiments displayed on orthogonal plots modified from Ron and Levi (2001) study of Erk el Ahmar Formation. a) AF demagnetization revealing stable Reverse polarity, b, and c) Thermal and AF experiments on sister specimens showing similar Normal directions.

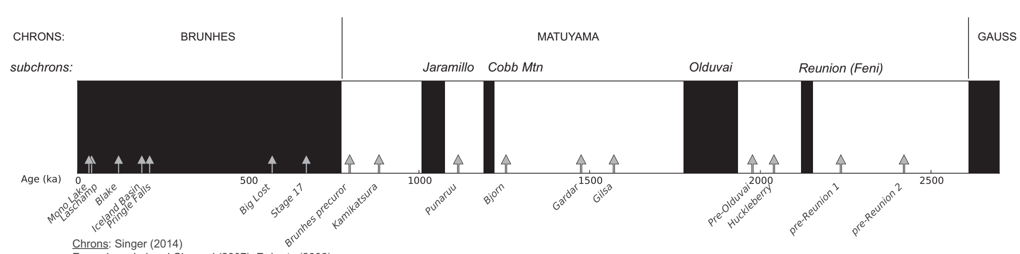

Figure X.3: The Quaternary Geomagnetic Polarity Time scale. Chrons and Subchrons following Singer (2014), excursions validated by Laj and Channel (2007) and Roberts (2008) are shown as arrows.

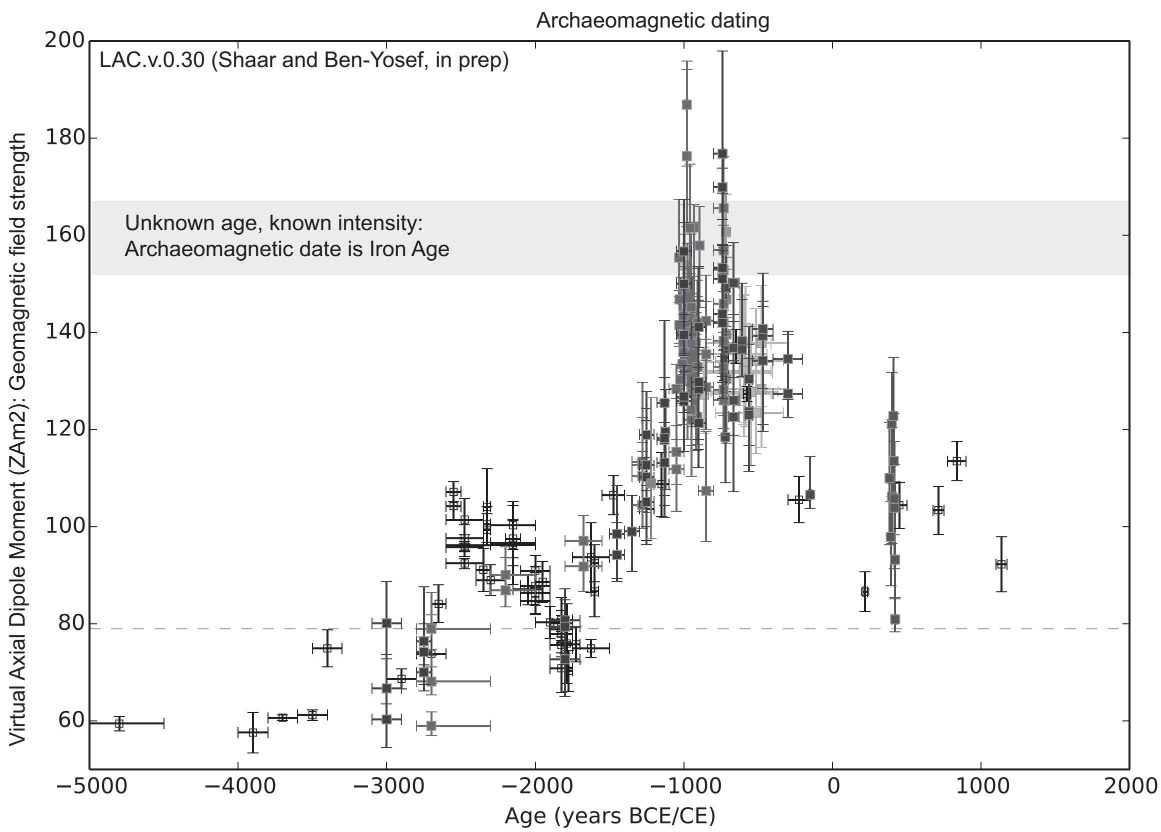

Figure 5.4: Archaeomagnetic ages of young materials using paleointensity. The Levantine Archaeomagnetic Compilation (LAC) is an in-progress effort to construct a new archaeo- and geo- chronology for young fired archaeological objects (Shaar and Ben-Yosef, unpublished) The gray stripe is paleointensity results from a sample whose age in unknown. A comparison with LAC suggests Iron-Age age. Similar declination/inclination curve of the Holocene may provide geochronology for young sediments.

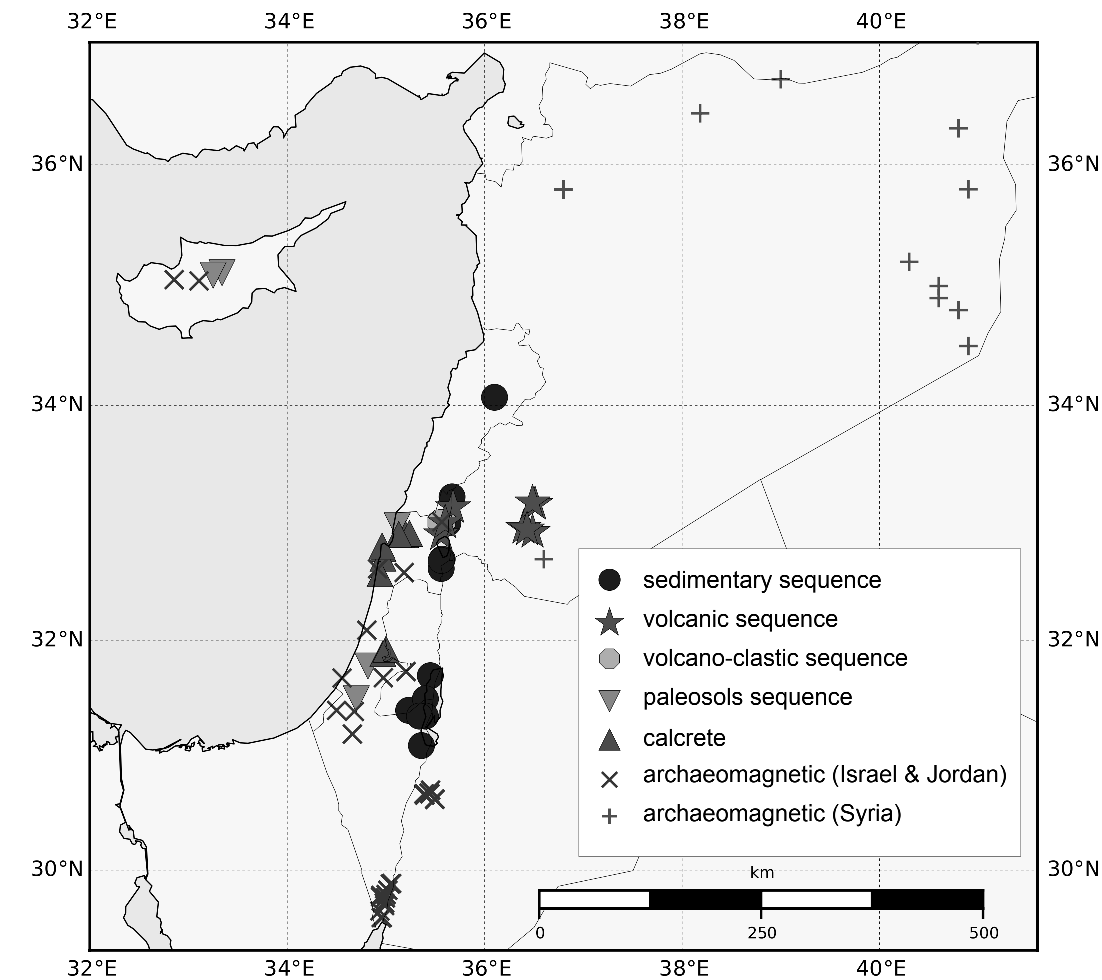

Figure X.5: Location map of the studies listed in Section X.5.

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