

# Provenancing of archeological pumice finds from North Sinai

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**Abstract** Seven pumice samples from excavations in North Sinai have been investigated with respect to their geochemical composition. This type of volcanic rock has been used as an abrasive and thus has been an object of trade since antiquity. With the help of Instrumental Neutron Activation Analysis, six of these Bronze Age samples could be correlated to their volcanic sources on the islands of Santorini, Nisyros and Giali (Greece) using the typical element concentrations (“chemical fingerprint”). The source of one pumice sample remains unidentified excluding, however, the Santorini eruption as a possible source. The concluding section of this article discusses the possible contribution, however indirect, of the pumice from Sinai and elsewhere in the Eastern Mediterranean to the controversial issue of the accurate date of the “Minoan” eruption of Santorini.

**Keywords** Tephrochronology · Trade · Pumice · INAA · Chemical fingerprint

## Introduction

Pumice is a highly vesicular, foamy volcanic rock, primarily consisting of volcanic glass with a minor percentage of

crystalline components. Due to its structural characteristics, pumice has been used since antiquity as an abrasive and became a desired object of trade as early as the Bronze Age, as proven by its presence in excavated workshop installations (see, e.g., Faure 1971; Peltz and Bichler 2001; Huber and Bichler 2003; Steinhauser et al. 2006a, 2006b; Sterba et al. 2009). Moreover, pumice has a very low density that enables its floating on water over large distances before becoming soaked and sinking (Risso et al. 2002). In antiquity, this phenomenon was probably attributed to the divine properties of the pumice resulting with its ritual deposition in cult as well as funerary sites in the Mediterranean region (Steinhauser et al. 2006b).

Silica-rich, pumice-producing magma is generally very homogeneous due to well-balanced geological processes such as mingling and differentiation in the magma chamber. Although exceptions are known (Bouvet de Maisonneuve et al. 2009), pumice is often characterized by great homogeneity as well. This allows the application of “chemical fingerprint” techniques in order to reveal the volcanic source of an unknown pumice sample just by comparing significant element concentrations of the sample to a suitable database, see Steinhauser et al. (2006b) and references therein. Over the past decade, we have analyzed pumiceous products from the largest volcanic eruptions in the Mediterranean, which comprise our database. We focused on eruptions and eruption sequences that formed significant pumice deposits. In particular, the following volcanic centers have been sampled: Santorini, Milos, Kos, Nisyros, Giali (all Greece), Lipari (Italy), and Cappadocia (Turkey) (Peltz et al. 1999; Steinhauser et al. 2006a, 2007).

The present article focuses on the analysis of pumice samples from a number of sites in North Sinai. In the course of systematic archeological explorations in North Sinai, between the Suez Canal and Gaza (1972–1982), the Ben-Gurion University expedition recorded and investigat-

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ed more than 250 settlement sites with material remains of the New Kingdom (1550–1150 BCE, according to current Egyptian chronology). These sites, including military forts, administrative centers, and numerous campsites, enable us to fully comprehend the mechanism of Egypt's imperial administration along the ancient land bridge (Egyptian "Ways-of-Horus") between Egypt and Asia (Oren 1987).

## Materials and methods

The following pumice samples are included in the present analysis:

#1—site A-289, locus 134, stratum 3 (phase V): nineteenth dynasty. Site A-289 represents the best preserved military fortress along the Ways-of-Horus. The earliest remains in the fortress (stratum 4, phase VII) advocate its foundation in the mid-eighteenth dynasty, probably during the reign of Thutmose III (1479–1425 BCE). The large scale rebuilding of the structure (stratum 3, phase V) took place during the early reign of Sethos I, and the site was subsequently abandoned in the late Ramesside period during the reign of Ramesses VI or VII (ca. 1143–1125 BCE).

#42451—site A-289, locus 267, stratum 4 (phase VII): mid-eighteenth dynasty. #42453—site A-289, locus 418, stratum 2 (phase III): nineteenth dynasty. See above #1.

#2—site A-345, locus 186, phase V: mid-eighteenth dynasty. The major settlement site A-345, nearby El-Arish, served as an administrative center during the eighteenth dynasty. Its extensive architectural remains included a spacious granary and a potter's workshop. The occurrence of several pumice specimens in this industrial installation testifies of its use as a tool in the process of pottery manufacture, e.g., for burnishing vessels etc. Typological considerations advocate a mid-eighteenth dynasty date for the ceramic repertoire of phase V, during the reign of Thutmose III (1479–1425 BCE).

#42455—site A-345, locus 186, phase V: mid-eighteenth dynasty. See above #2.

#42452—site BEA-10B, locus 108: nineteenth dynasty. Located nearby the village of Bir el-Abd, site BEA-10B is represented by the remains of a sizeable fort and an excellently preserved mud-brick granary which is comprised of four circular silos. Once its dome-shaped roof had collapsed, the granary became a refuse place for the fortress. The diagnostic ceramic and other artifactual evidence from Silo B, where the pumice sample was also recorded, were assigned to the nineteenth dynasty (1295–1186 BCE).

#42454—site A-343, locus 10: second half of the eighteenth to early nineteenth dynasty. Located a short

distance from A-345, the small campsite A-343 is largely deflated. Surface remains, including local Egyptian ceramics and Cypriote imports imply a date not earlier than the mid-eighteenth dynasty. The terminal date in the early nineteenth dynasty is attested by the occurrence of Egyptian-type beer jars stamped with cartouches of Sethos I (1294–1279 BCE).

These samples were washed several times with distilled water in an ultrasonic bath and dried at 110°C. The first step in the analytical process was optical mineralogical investigation using a ZEISS™ STEMI SV8 stereomicroscope with variable magnification from 8 to 128. Crystallized minerals ("phenocrysts") in the glass matrix of the pumice, such as quartz, pyroxene, feldspar, ore-minerals, etc., can offer valuable information. The appearance of the mineral biotite (dark mica), for example, is of special interest, because it is a very characteristic constituent of pumice from certain eruptions. Other eruptions, producing biotite-free products, can be excluded as the source of a particular pumice lump at a first glimpse, if it contains biotite.

Instrumental Neutron Activation Analysis (INAA) was used to determine the major and trace element abundances in the samples. INAA is a perfectly suitable technique to simultaneously determine a large number of geochemically significant trace elements, and was therefore used for the identification of eruption products by their characteristic element distribution patterns, the chemical fingerprint. In particular, the elements Na, K, Sc, Cr, Fe, Co, As, Rb, Zr, Sb, Cs, Ba, La, Ce, Nd, Sm, Eu, Tb, Yb, Lu, Hf, Ta, Th, and U were determined.

Following the microscopical investigation, the pumice finds were sampled by carefully taking out a small amount of pumiceous material (approximately 0.1 g) from existing cracks in the pumice surface using steel tweezers. During this procedure, it was carefully avoided to include discernible phenocrysts into the sample. The pumice material was weighed into Suprasil™ quartz glass vials, sealed and irradiated for approximately 40 h together with internationally certified standard reference materials in the central irradiation tube (neutron flux density  $1 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$ ) of the TRIGA Mark II research reactor at the Atominstut in Vienna. The multielement standards used for the quantitative analysis were the CANMET reference soil SO1, NIST SRM 1633b Coal Fly Ash, BCR No. 142 light sandy soil, NIST SRM 2702 Inorganics in Marine Sediments and the MC rhyolithe GBW 07113.

After irradiation, the quartz glass sample vials were decontaminated and packed into PE vials fitting the automatic sample changer device of the Atominstut in Vienna. After a cooling time of 5 days, a first  $\gamma$ -spectrum was measured to obtain the activities of the short and

medium-lived activation products <sup>153</sup>Sm, <sup>239</sup>Np (decay product of <sup>239</sup>U), <sup>76</sup>As, <sup>24</sup>Na, <sup>42</sup>K, and <sup>140</sup>La. Three weeks later, a second measurement sequence was started to detect the long-lived activation products <sup>141</sup>Ce, <sup>169</sup>Yb, <sup>177</sup>Lu, <sup>233</sup>Pa (decay product of <sup>233</sup>Th), <sup>51</sup>Cr, <sup>181</sup>Hf, <sup>131</sup>Ba, <sup>147</sup>Nd, <sup>95</sup>Zr, <sup>134</sup>Cs, <sup>160</sup>Tb, <sup>86</sup>Rb, <sup>59</sup>Fe, <sup>65</sup>Zn, <sup>46</sup>Sc, <sup>60</sup>Co, <sup>182</sup>Ta, <sup>152</sup>Eu, and <sup>124</sup>Sb. The measuring times were 1,800s and 10,000s, respectively. All samples were measured in a fixed position at a distance of 4 cm beside the detector. The  $\gamma$ -spectrometry was performed with a 222 cm<sup>3</sup> HPGe-detector (1.78 keV resolution at the 1,332 keV <sup>60</sup>Co peak; 48.2% relative efficiency), connected to a PC-based multi-channel analyzer with a preloaded filter and a Loss-Free Counting system.

**Results**

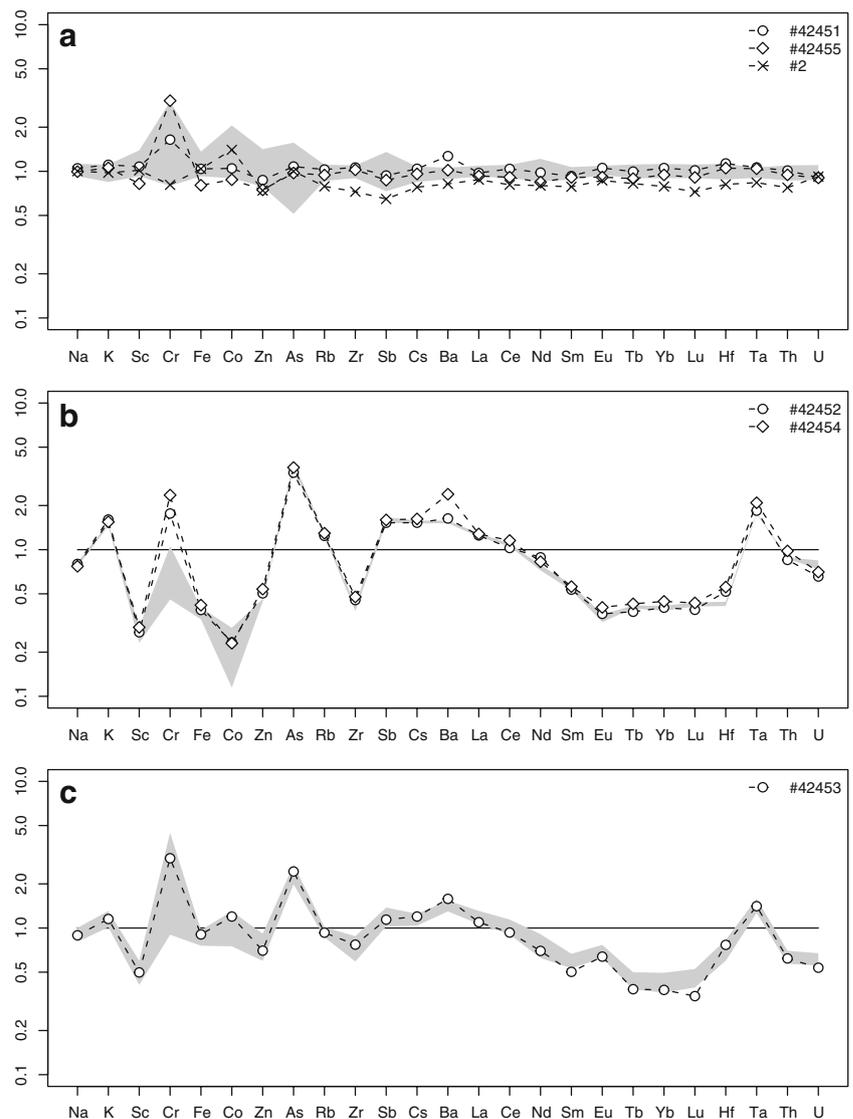
The results of the INAA are tabulated in Table 1. By comparison with the chemical fingerprints of Mediterranean eruptions in our database, we were able to identify the volcanic source of six pumice finds from North Sinai (see Fig. 1a-c; for the geographical setting, see the map in the Electronic Supplementary Material). The shaded areas in these figures represent the natural variation range of a certain element in the respective pumice deposit. All data were normalized to the average composition of Santorini Bo pumice (“Bo norm”), which is the product of the “Minoan” eruption of Santorini. Normalization of element concentrations to those in Bo pumice has been found to be the most efficient way of comparing all information gathered (Peltz et al. 1999; Steinhauser et al. 2007). The normalized data stay within a range of two orders of magnitude and can thus be plotted with the highest resolution. More widely used normalizations (continental crust, chondrite) can hardly resolve the small variations in the trace element concentrations with respect to the precision available by INAA. In agreement with the mineralogical diagnoses, the samples could be assigned to the following volcanic eruptions: #42451 and #42455 originate from the Minoan eruption of Santorini (Fig. 1a), #42452 and #42454 are Giali main pumice (Fig. 1b), #42453 originated from one of the caldera-forming eruptions of Nisyros (Fig. 1c). Pumice sample #2 originated from the Minoan eruption of Santorini as well. The element concentrations (cross signature in Fig. 1a) of this sample appear generally somewhat depleted. This phenomenon can be explained in the way that the sample was probably contaminated with quartz grains. Since the applied routine-INAA does not allow the determination of SiO<sub>2</sub>, only the weight of the sample appears to be shifted. In any case, the sample is definitely a member of the Upper Pumice deposit (“oberer Bimsstein”, Bo) and not of the Lower Pumice of Santorini (“unterer Bimsstein”, Bu), as

**Table 1** Element concentrations (mg/kg) of the archeological pumice samples investigated in this study

	Na	K	Sc	Cr	Fe	Co	Zn	As	Rb	Zr	Sb	Cs	Ba	La	Ce	Nd	Sm	Eu	Tb	Yb	Lu	Hf	Ta	Th	U
#42451	34900	26900	9.22	3.2	22700	4.1	61	3.0	111	308	0.28	3.0	720	31	65	25	5.7	1.06	1.01	5.3	0.84	8.77	0.85	20.2	5.4
#42452	26600	39000	2.34	3.5	8500	0.9	35	9.3	134	131	0.46	4.4	920	40	65	23	3.3	0.37	0.38	2.0	0.32	4.01	1.48	17.0	3.9
#42455	33100	25600	7.06	6.0	17500	3.4	52	2.7	102	297	0.26	2.8	570	29	57	22	5.6	0.91	0.90	4.8	0.75	8.14	0.84	18.9	5.3
#42454	25600	37600	2.53	4.6	9100	0.9	37	10.1	140	138	0.48	4.7	1350	41	73	21	3.5	0.40	0.43	2.2	0.36	4.31	1.68	19.6	4.2
#42453	29700	28000	4.27	5.9	19700	4.7	49	6.7	101	223	0.34	3.5	890	35	59	18	3.1	0.64	0.39	1.9	0.28	5.95	1.13	12.4	3.2
#1	13700	35200	1.98	8.5	11500	3.6	48	8.9	160	150	1.23	10.1	640	61	121	38	7.0	1.52	0.83	2.3	0.33	3.13	1.24	31.1	5.7
#2	33400	23700	8.71	1.6	22700	5.5	51	2.8	85	210	0.19	2.3	460	28	51	20	4.8	0.87	0.84	4.0	0.60	6.31	0.67	15.4	5.4

The analytical error is <10 rel.% for the elements Cr, Nd and Lu, and <5 rel.% for all other elements

**Fig. 1** Element concentrations of pumice samples normalized to the average element concentrations in Santorini Upper Pumice (Bo; “Minoan” pumice). **a** Samples #42451, #42455 and #2 compared to the natural variation range of elements in Santorini Upper Pumice (*shaded area*). **b** Samples #42452 and #42454 compared to the natural variation range of elements in Giali main pumice (*shaded area*). **c** Sample #42453 compared to the natural variation range of elements in Nisyros upper and lower caldera pumice (*shaded area*)



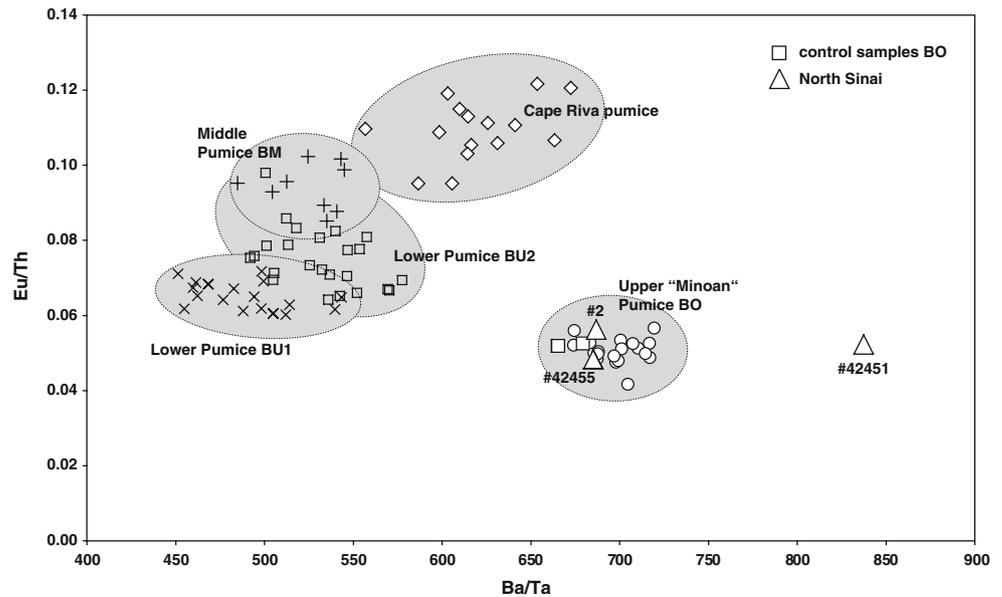
shown in Fig. 2. In this figure, the ratios of the concentrations of Eu/Th vs. Ba/Ta are plotted, thus any SiO<sub>2</sub> contamination is canceled out. Due to its barium anomaly, pumice sample #42451 is located outside the typical area of Minoan pumice samples. Nevertheless, the complete element pattern (Fig. 1a) evidences its provenance from Santorini. The source of sample #1 remains unknown—no volcanic pumice deposit in our database (Steinhauser et al. 2006b) indicated its provenance. Nevertheless, this sample definitely did not originate from Santorini, which is considered to have been the most significant volcanic eruption during the Bronze and Iron Ages in the Eastern Mediterranean region.

In general, the occurrence of pumice from a certain eruption can be used for dating purposes by providing a *terminus post quem* for its archeological context (Warren and Puchelt 1990). This is the case of the Minoan eruption of Santorini, which has been radiocarbon dated to 1627–1600 BCE (Friedrich et al. 2006) or more generally to ca.

1650–1600 BCE (Manning et al. 2006). On the debate of the <sup>14</sup>C dates, see also Wiener (2007), Bruins et al. (2009), and Warburton (2009b). Further, volcanic ash (“tephra”) from different archeological sites in the Mediterranean could be useful in establishing a chronological framework (“tephrochronology”) of datable ash samples in Bronze Age settlements (Doumas and Papazoglou 1980; Einarsson 1986; Pearce et al. 2002, 2004a).

It should be noted that all pumice samples under review show good agreement with the chemical fingerprint. Only some chromium and barium values appear slightly increased. For chromium, this is a very common phenomenon with archeological pumice samples and can be explained by the contamination with abraded particles from steel tools used in the excavation and sampling. Increased chromium values can even be regarded as the rule for archeological pumice finds. Positive barium anomalies are topic of recent investigations (Obenholzner et al. 2003; Moune et al. 2006;

**Fig. 2** Plot of the concentration ratios Eu/Th vs. Ba/Ta of all Santorini pumice samples of our database as well as pumice samples #1, #42451 and #42455. This type of presentation allows the clear distinction between “Minoan” and “Pre-Minoan” pumices (Cape Riva pumice, Lower Pumice Bu1 and Bu2, Middle Pumice Bm) of the Santorini volcano



Steinhauser and Bichler 2008; Steinhauser et al. 2008; Sterba et al. 2008). The glassy pumice surface effectively adsorbs barium ions from solutions. The source of the additional barium is probably the volcanic activity itself: barium-rich aerosols can be found in volcanic plumes. As an example, a study of cave sediments on the Greek island of Tilos showed significantly increased barium concentrations of the sediment layers that also contained traces of volcanic ashes (Steinhauser et al. 2008). Thus, both phenomena increased chromium and barium concentrations give no rise to doubt the validity of the assignments above.

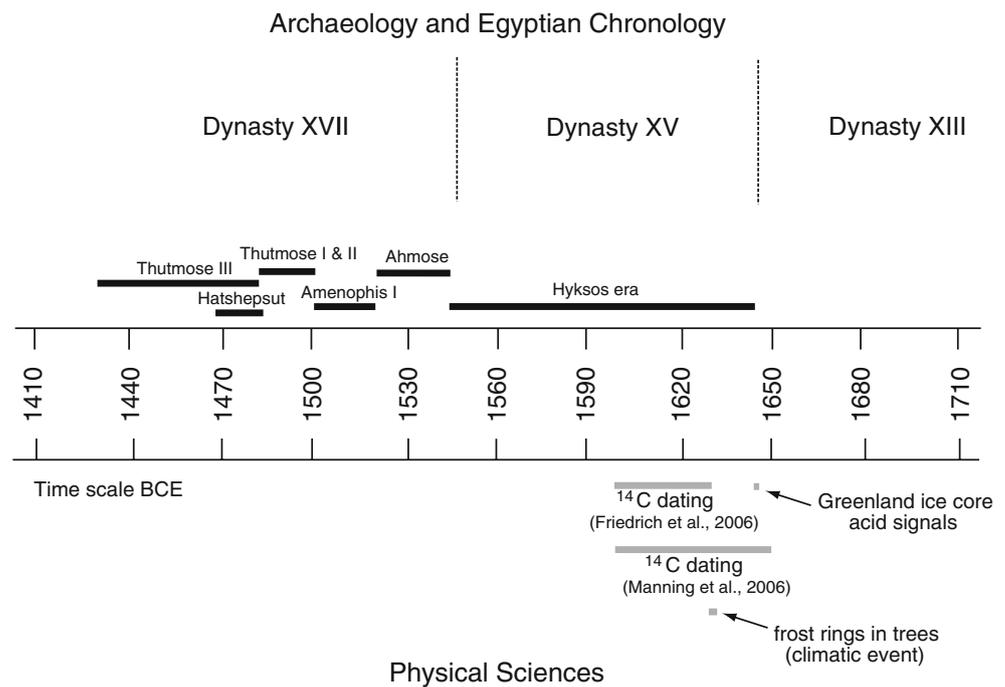
## Discussion

Six out of seven pumice samples from North Sinai could be correlated to their volcanic sources (Santorini, Giali, Nisyros). According to their structural characteristics these pumice lumps most likely were used as tools. This assumption has been corroborated at least in one case, i.e., site A-345, whereby nicely rounded pumice objects were recorded amongst the debris of the potters' workshop (see above sample #2). Pumice from volcanic eruptions could have reached the coast lines of the Eastern Mediterranean by wind and/or sea currents and then collected by the local inhabitants. Since large amounts of pumice are ejected into the sea only during the volcanic eruption itself, obviously the pumice samples from Giali and Nisyros which originated from geological deposits at least 30,000 years ago (Federman and Carey 1980; Limburg and Varekamp 1991; Hardiman 1999), trading connections between the Greek islands and the Sinai coast remain the only alternative explanation for their occurrence in the region. However, as we shall see

below, this is not necessarily the case for the Minoan Santorini pumice which presumably reached the shores of the eastern Mediterranean, including Sinai and the Nile Delta, at some point in the seventeenth or sixteenth century BCE.

In addition to the obvious cultural and economic significance of the occurrence of pumice in archeological excavations, the results of the present study take us back to the long-standing debate over the accurate dating of the Santorini eruption: physical versus archeo-historical dating (Manning 1999). For a simplified diagram of the time frame in question, see Fig. 3. Accordingly, various scientific methods have been applied thus far— $^{14}\text{C}$  dating (Friedrich et al. 2006; Manning et al. 2006), dendrochronological observations of frost rings in trees (LaMarche and Hirschboeck 1984), and Greenland ice core data (Hammer et al. 1987 and 2003). For the latter, only the sulfuric acid signals (Hammer et al. 1987) should be regarded as potentially relevant, since the origin of the tephra particles is still under dispute (Pearce et al. 2004b; Denton and Pearce 2008; Vinther et al. 2008). The more recent calibrated  $^{14}\text{C}$ -measurements dated the Santorini eruption to 1627–1600 BCE (Friedrich et al. 2006) or 1650–1600 (Manning et al. 2006). Archeological and historical considerations, however, argue for a much later date for this significant event of ca. 1500 BCE or even later. Amongst the various conventional archeo-historical arguments, the late dates are based on the occurrence of diagnostic Cypriote ceramics under the tephra deposits at Santorini and the evidence of their first appearance in Egypt (Tell el-Dab<sup>a</sup>) in settlement strata of the mid-eighteenth dynasty (Bietak and Höflmayer 2007). Such a serious discrepancy of 100–150 years between the radiocarbon dating and the archeo-historical dating (i.e., historical

**Fig. 3** Simplified diagram of the time frame for the possible eruption of the “Minoan” eruption of Santorini showing both an outline of the Egyptian chronology and the proposed natural-scientific dates. For reasons of clarity, parallel dynasties have been omitted. According to the archeo-historical dating, the eruption happened during the reign of Thutmose III. Physical dating suggests an eruption date in the late seventeenth century BC. See text for references



Egyptian chronology) cannot presently be resolved and must await further investigation.

Another archeological argument concerns pumice provenancing. It has been observed that Minoan pumice from Santorini is, to this date, absent from pre-eighteenth dynasty sites in Egypt and the eastern Mediterranean. Actually, the bulk of Minoan pumice specimens from archeological contexts (e.g., the Tell el-Dab<sup>a</sup> in the Nile delta) date, according to the present chronology, in the mid-eighteenth dynasty or the time of Thutmose III, 1479–1425 BCE. To date, 340 pumice samples from archeological contexts in the Egyptian Delta and various sites along the Mediterranean coast as well as Cyprus, Greece, and Crete have been analyzed in our work group and, categorically, not a single piece of Minoan pumice was recorded in pre-eighteenth dynasty or Middle Bronze Age context (Huber et al. 2003). It is interesting to note, however, that the number of excavated pumice samples from the eighteenth or nineteenth dynasty (314 samples, 250 Minoan) greatly exceeds the number from pre-eighteenth dynasty of Middle Bronze Age context (32 samples, 0 Minoan); hence, it remains possible that the apparent absence relates to the small number of pre-eighteenth dynasty samples and not necessarily the dating of the Santorini eruption. Moreover, it is an interesting phenomenon of this controversy that almost no “fact” is accepted by the entire community. Recently, the soundness of the stratigraphy established at Tell el-Dab<sup>a</sup> has been questioned (Warburton 2009a); however, the information provided by the excavators unsurprisingly stands in strong contrast to the criticism raised.

Oren (1997) reported that excavations of the Middle Bronze (MB) III (seventeenth to sixteenth century BCE) temple at Tel Haror in the Western Negev and some 20 km west of Tell el Ajjul, yielded imported Minoan pottery but no “Minoan” pumice. The piece of ceramics was a fragment of a large, handmade coarse pithos incised with Linear A graffiti. The sherd is too small for identification with a specific shape. However, petrographic analysis indicated an origin in the south coast of Crete near the important Middle Minoan site of Myrtos Pyrgos; a site which has already produced evidence of Linear A signs. The sherd was found at the Tel Haror sacred precinct in Stratum IVa dating to MBIII (seventeenth to early sixteenth century BCE, see Oren 1993; Oren and Olivier 1995; Day et al. 1999).

The pumice testimonies support the hypothesis that the eruption of Santorini must have occurred at some point in the eighteenth dynasty. On the other hand, assuming, that the Santorini eruption took place, according to <sup>14</sup>C determinations, in the second half of the seventeenth century BCE, it would indeed be most peculiar a phenomenon that pumice from the Minoan Santorini eruption were abundantly available along the shores of the Eastern Mediterranean, yet for some reason had been left unnoticed and unused by the local inhabitants for 100–150 years. In this respect, the North Sinai pumice samples are supportive of the emerging picture, especially since none originated from pre-eighteenth dynasty context. Three of the samples from North Sinai, #2, #42451, and #42455 which date in the mid-eighteenth dynasty and most likely during the reign of Thutmose III thus complement the results obtained elsewhere in the Mediterranean.

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