

New Late Neolithic (c. 7000–5000 BC) archeointensity data from Syria. Reconstructing 9000years of archeomagnetic field intensity variations in the Middle East

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1 New Late Neolithic (c.7000-5000 BC) archeointensity data from Syria. 2 Reconstructing 9000 years of archeomagnetic field intensity variations in the 3 Middle East 4 Yves Gallet^a, Miquel Molist Montaña b, Agnès Genevey ^c, Xavier Clop García b, Erwan 5 Thébault ^{a,d}, Anna Gómez Bach ^b, Maxime Le Goff ^a, Béatrice Robert ^e, Inga Nachasova f ^a 6 Institut de Physique du Globe de Paris, Sorbonne Paris Cité, Université Paris Diderot, UMR 7 7154 CNRS, F-75005 Paris, France ^b 8 SGR SAPPO. Prehistory Department, Facultat de Filosofia i Lletres, Edifici B, Universitat 9 Autònoma de Barcelona 08193 Bellaterra, Barcelona, Spain 10 ^c UPMC Université Paris 06, UMR CNRS 8220, Laboratoire d'Archéologie Moléculaire et 11 Structurale, LAMS, F-75005 Paris, France 12 ^d LPG, UMR CNRS 6112, Laboratoire de Planétologie et Géodynamique de Nantes, 13 Université de Nantes, 44322 Nantes cedex 03, France 14 e Université Lumière – Lyon 2, UMR CNRS 5133 ArchéOrient, Lyon, France 15 *Institute of Physics of the Earth, Russian Academy of Science, Moscow, Russia* 16

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21 ABSTRACT

22 We present new archeomagnetic intensity data from two Late Neolithic archeological 23 sites (Tell Halula and Tell Masaïkh) in Syria. These data, from 24 groups of potsherds 24 encompassing 15 different time levels, are obtained using the Triaxe experimental 25 protocol, which takes into account both the thermoremanent magnetization anisotropy 26 and cooling rate effects on intensity determinations. They allow us to recover the 27 geomagnetic intensity variations in the Middle East, between \sim 7000 BC and \sim 5000 BC, 28 i.e. during the so-called pre-Halaf, proto-Halaf, Halaf and Halaf-Ubaid Transitional 29 cultural phases. The data are compared with previous archeointensity results of similar 30 ages from Northern Iraq (Yarim Tepe II and Tell Sotto) and Bulgaria. We find that 31 previous dating of the Iraqi material was in error. When corrected, all northern 32 Mesopotamian data show a relatively good consistency and also reasonably match with 33 the Bulgarian archeointensity dataset. Using a compilation of available data, we 34 construct a geomagnetic field intensity variation curve for the Middle East 35 encompassing the past 9000 years, which makes it presently the longest known regional 36 archeomagnetic intensity record. We further use this compilation to constrain variations

37 in dipole field moment over most of the Holocene. In particular, we discuss the 38 possibility that a significant dipole moment maximum occurred during the third 39 millennium BC, which cannot easily be identified in available time-varying global 40 geomagnetic field reconstructions.

41

42 1. Introduction

43 Recent studies have been focused on the construction of time-varying global 44 archeomagnetic field models that cover most of the Holocene (e.g. Korte et al., 2011; 45 Nilsson et al., 2014; Pavón-Carrasco et al., 2014). These models have been developed 46 with the aim to decipher core dynamics over centennial and millennial time scales, the 47 evolution of the past solar activity and the interactions between geomagnetic field and 48 external processes (e.g. Korte et al., 2009; Licht et al., 2013; Usoskin et al., 2014). In all 49 these studies, however, the authors acknowledge the fact that for the more ancient 50 periods, i.e. beyond the first millennium BC, the reliability and accuracy of the 51 geomagnetic field models are strongly penalized by the low number and the poor 52 temporal and geographical distributions of the available archeomagnetic and volcanic 53 paleomagnetic data. To overcome this problem, often paleomagnetic data from 54 sediments have been included in the models reference dataset; nevertheless 55 sedimentary data do not significantly improve the accuracy of the models because a part 56 of them, difficult to estimate, may be biased by experimental errors and/or because 57 these data often lack precise dating (e.g. Valet et al., 2008; Nilsson et al., 2010). There is 58 therefore a critical need for new well dated archeomagnetic data dated with ages older 59 than the first millennium BC.

60 The Middle East, thanks to its rich archeological and historical heritage, offers the 61 possibility to travel back through the geomagnetic field history over most of the 62 Holocene, recovering what could be the longest known archeomagnetic field record. 63 Archeomagnetic studies conducted up to now were mainly focused on the Bronze and 64 Iron Age archeological periods, allowing a better characterization of the regional 65 geomagnetic field intensity behavior for the last 3 millennia BC (i.e. Genevey et al., 2003; 66 Gallet and Le Goff, 2006; Gallet et al., 2008, 2014; Ben-Yosef et al., 2008, 2009; Gallet and 67 Al Maqdissi, 2010; Thébault and Gallet, 2010; Shaar et al., 2011; Ertepinar et al., 2012; 68 Gallet and Butterlin, 2014). These studies have revealed significant field intensity

69 variations, and in particular a series of intensity maxima between \sim 2600 and 2500 BC, 70 between ~2300-2000 BC, around 1500 BC and at the very beginning of the first 71 millennium BC (e.g. Gallet et al., 2014). These studies have further shown that the 72 beginning of the first millennium BC was most probably marked by the highest 73 geomagnetic field intensity so far detected during the Holocene and perhaps even before 74 (Ben-Yosef et al., 2009; Shaar et al., 2011; Ertepinar et al., 2012; Livermore et al., 2014).

75 In contrast, for older periods between ~7000 BC and 3000 BC, i.e. during the Late 76 Neolithic (or Pottery Neolithic) and the Chalcolithic, the archeointensity data from the 77 Middle East remain relatively scarce, which prevents an accurate description of the 78 regional geomagnetic field intensity variations (e.g. Genevey et al., 2003; Ben-Yosef et al., 79 2008). However, several possibilities exist to sample pre-Bronze Age archeological sites. 80 This is particularly true for the 6th millennium BC, which saw the development of the 81 Halaf culture throughout the northern Mesopotamian region. This culture was named 82 after the Tell Halaf archeological site in northern Syria (Fig. 1a), which was discovered 83 and first excavated by the German diplomat Max von Oppenheim at the beginning of the 84 20th century. The Halaf culture is notably characterized by a plentiful pottery 85 production presenting a fine and light-colored clay paste, with brown or black 86 monochrome or polychromatic painted decorations (e.g. Akkermans and Schwartz, 87 2003; Nieuwenhuyse et al., 2013 and references therein). This well-fired ceramic 88 production thus constitutes a promising target for archeointensity investigations.

89 Going further back in time, archeomagnetic studies may benefit from recent 90 archeological studies conducted in Syria that focused on the 7th millennium BC, which 91 saw the emergence of the first pottery production in the Near East (e.g. Tsuneki and 92 Miyake, 1996; Le Mière and Picon, 1998; Nishiaki and Le Mière, 2005; Molist et al., 2007; 93 Nieuwenhuyse et al., 2010, 2013). At first rare, the pottery was sometimes of a 94 surprisingly elaborated conception with a painted decoration during a primitive phase 95 referred to as the "Initial Pottery Neolithic" (∼7000-6700 BC; e.g. Van der Plicht et al., 96 2011). By the middle of the 7th millennium BC, the use of ceramics spread over northern 97 Mesopotamia (Nieuwenhuyse et al., 2010, 2013 and references therein). This pre-Halaf 98 period is mainly represented by undecorated plant-tempered pottery with a coarse clay 99 paste shaped into baskets. The fineness of the clay paste improved at the end of the 7th 100 millennium BC during a period referred to as proto-Halaf (∼6050-5900 BC), just

101 preceding the Halaf period, with the use of mineral-tempered clay (e.g. Cruells and 102 Nieuwenhuyse, 2004). According to Akkermans and Schwarz (2003), the pre-Halaf 103 coarse pottery was produced in open fires, with heating temperatures of about 700- 104 750°C, while the elaborate Halaf ceramics were most probably heated at higher 105 temperatures in chambered kilns. For the pre- and proto-Halaf periods encompassing 106 the 7th millennium BC, archeointensity studies are thus still possible, but they may be 107 further complicated by the characteristics of the ceramic production.

108 To extend the Syrian geomagnetic field intensity record, which presently mainly 109 documents the Bronze and Iron Age periods, we conducted an archeomagnetic study on 110 the pre-Halaf, proto-Halaf, Halaf and Halaf-Ubaid Transitional archeological periods, a 111 time interval of nearly two millennia (∼7000 to ∼5000 BC) covering the Pottery 112 Neolithic, at the end of the Neolithic (e.g. Campbell, 2007; Campbell and Fletcher, 2010; 113 Van der Plicht et al., 2011; Nieuwenhuyse et al., 2013 and references therein). The new 114 archeointensity data reported in this study were mostly obtained from potsherds 115 collected from the archeological site of Tell Halula located in northern Syria (Fig. 1a). 116 These results were complemented by few data obtained from potsherds discovered at 117 the archeological site of Tell Masaïkh, located south-east along the middle course of the 118 Euphrates river (Fig. 1a). It is of interest to note that the longest and almost continuous 119 regional archeointensity record presently available was obtained from Bulgaria 120 (Kovacheva et al., 2014). It begins around 6000 BC, i.e. a date during the proto-Halaf 121 period, which means that some of the new data presented in this study are the oldest 122 archeointensity data recovered until now. Furthermore, we recall the recent effort of 123 data compilation of archeomagnetic, volcanic and sedimentary paleomagnetic results 124 that led to the construction of global archeomagnetic field models encompassing almost 125 the entire Holocene (Korte et al., 2011; Nilsson et al., 2014; Pavón-Carrasco et al., 2014). 126 Any new archeomagnetic intensity data dated to the Late Neolithic-Early Chalcolithic 127 period, now rather rare (e.g. Genevey et al., 2008; Knudsen et al., 2008), will therefore 128 allow us to test, at least regionally, the accuracy of the available models and in return 129 will better constrain these models. This point is particularly critical and we will also 130 report in this study on erroneous dating of a relatively large archeointensity dataset 131 previously obtained in the Middle East for the $7th$ and $6th$ millennia BC (Nachasova and 132 Burakov, 1995, 1998).

133

134 2. Archeomagnetic sampling

135 2.1. Tell Halula

136 Tell Halula ($\lambda = 36^{\circ}25'N$, $\varphi = 38^{\circ}10'E$) is located in the modern Syrian 137 administrative province of Raqqa, about 80 km west of the city of Raqqa and 85 km east 138 of the city of Aleppo. This archeological site, \sim 4 km west of the Euphrates, forms a sub-139 circular artificial mound (360 m x 300 m), with an archeological deposit thickness of $140 \sim 14$ m (Fig. 1b). Archeological excavations conducted since 1991 by a team of the 141 Universitat Autónoma de Barcelona revealed a total of 38 phases of occupation. From 142 the stratigraphic and archeological constraints (including chipped stone artefacts, 143 pottery typology, figurines and architecture), it has been determined that the site was 144 occupied continuously from the Middle Pre-Pottery Neolithic B (PPNB) to the Late Halaf 145 periods, i.e. from ~7800 to 5300 cal BC (Molist et al. 2007, 2013; Molist 1996, 2001). 146 The systematic archeological fieldwork at Tell Halula has brought significant knowledge 147 about the development of farming, especially in the final stages of the Neolithisation 148 process, when economic, technological and cultural changes were being consolidated.

149 The different phases of human occupation have been recovered in several 150 sectors, especially in the south, south-east and central parts of the settlement (Sectors 1, 151 2, 7, 14, 30, 44 and 45). The Neolithic ceramic horizon encompasses most of the seventh 152 millennium BC and part of the sixth millennium BC (Architectural Phases 20 to 38), 153 spanning the pre-Halaf (or Period 5 according to Lyon's School terminology; Hours et al., 154 1994), the proto-Halaf or Halaf Transitional and the Halaf (Early, Middle, Late) periods. 155 The archeological and stratigraphic data indicate the presence of a sedentary 156 population, with several large houses or architectural structures relatively dispersed 157 over a surface of \sim 6 ha, i.e. with large open areas between households and buildings for 158 domestic use. Furthermore, several structures for a collective use were discovered for 159 the pre-Halaf period, with a massive enclosing wall in Sector 1 and a drainage channel in 160 Sector SS7 (Molist, 1996, 1998; Molist and Faura, 1999; Molist et al., 2013).

161 The pottery assemblages analyzed in the present study were sampled following 162 the main chronocultural phases documented at Tell Halula (Fig. 2; see description in 163 Molist et al., 2013 and Supplementary Text1). Here, we used the same chronological

164 time scale as in Molist et al. (2013). For the first pottery production within the Early Pre-165 Halaf (Ceramic Phase I; ~7000-6600 BC), two groups of fragments collected from the 166 top of Sector 2 were analyzed. The first group of fragments (SY 127) was recovered from 167 a pit located in an open area and the second (SY 125), a little younger than the previous 168 one, was collected from an occupation level associated with a rectangular building. For 169 the intermediate pre-Halaf period (Ceramic Phase II; ~6600-6300 BC), three pottery 170 groups were collected from a large outdoor space between several domestic units. Their 171 age assignment was established via stratigraphy, with the group of potsherds SY96 172 being the most recent, SY97-129 intermediate and SY98-128 being the oldest. Finally, 173 pottery group SY130, comprising pottery fragments found in a pit from Sector 49, comes 174 from the late pre-Halaf (Ceramic Phase III; ~6300-6050 BC).

175 For the period referred to as proto-Halaf (~6050-5900 BC), corresponding to 176 Ceramic Phase IV defined at Tell Halula, two pottery groups of household artifacts were 177 collected in Sectors 44 (SY94-137) and 40 (SY95). A single group (SY91) lies within the 178 Early Halaf period (Ceramic Phase V; ~5900-5750 BC), which was recovered from a 179 multicellular house located in sector 44. Different pits discovered in the same area of 180 Sector 45 yielded four contemporaneous groups of pottery (SY87, SY88, SY89, SY90) 181 dated within the Middle Halaf period (Ceramic Phase VI; ~5750-5550 BC).

182 Nine groups of fragments were collected from the most recent chronological 183 phases at Tell Halula dated in the Late Halaf (Ceramic Phase VII; ~5550-5300 BC). This 184 relatively dense sampling was possible due to a relatively complete stratigraphic 185 sequence from Sector 49 (Gómez, 2011). For most of these groups, the fragments were 186 recovered from different pits excavated in a large open yard, that were used for the 187 disposal of ash and domestic waste. The stratigraphic data and the ceramic typology 188 distinguish five successive temporal intervals, each being documented by one or several 189 pottery groups (from older to younger: SY86-131; SY135; SY84 and SY138; SY82 and 190 SY83-136; SY80, SY81 and SY132).

191 In summary, 22 different pottery groups from 14 successive occupation levels 192 were thus sampled at Tell Halula, whose dates span \sim 1700 years, between \sim 7000 and 193 \sim 5300 BC. For displaying the results in a relative chronological framework for phases II 194 and VII, we made the rough approximation of an equi-temporal distribution for 195 respectively the three and five successive occupation levels (i.e. assuming a duration of

196 100 years for each intermediate pre-Halaf level between 6600 BC and 6300 BC and a 197 duration of 50 years for each Late Halaf level between 5550 BC and 5300 BC; dating 198 with * in Table 1).

199

200 2.2. Tell Masaïkh

201 The archeological site of Tell Masaïkh $(\lambda=34^{\circ}25^{\prime}N, \varphi=40^{\circ}01^{\prime}E)$ is located on a 202 river terrace in the middle Euphrates Valley (left bank), in the modern province of Deir 203 ez-Zor (eastern Syria). Discovered in 1996 by the Mission Archéologique Française de 204 Ashara/Terga led by O. Rouault, excavations at Tell Masaïkh $(\sim 4 \text{ km from Teraa})$, 205 conducted under the leadership of M.-G. Masetti-Rouault, have revealed several phases 206 of occupation starting with the Late Neolithic (Halaf). More recent periods include 207 significant Neo-Assyrian remains, with a citadel and a palace dated in the $9th$ - $8th$ 208 centuries BC (Iron Age period), which led the identification of Tell Masaïkh as the 209 Assyrian city named Kar-Assurnasirpal (see general discussion in Masetti-Rouault, 210 2010).

211 The discovery in the western sector D of Tell Masaïkh of an artisanal Halaf 212 settlement makes this site also quite unique. It is located away from most other known 213 Halaf archeological sites situated more to the North with rainfall above 250 mm/year 214 (while rainfall is below this isohyet in the Tell Masaïkh region; e.g. Masetti-Rouault, 215 2006; Robert, 2010), which opens discussion on farming systems and on the use of 216 irrigation at this time.

217 Excavations of the Halaf levels at Tell Masaïkh unearthed several occupation 218 levels in open areas with fire places (tannurs), several kilns probably for pottery 219 production and a 1.5 m-thick, \sim 20 m-long stone wall that supported a terrace. A rich 220 ensemble of Late Halaf potsherds was also recovered. The potsherds analyzed in the 221 present study were found in the uppermost layers dated in the Halaf-Ubaid Transitional 222 (~5300-5000 BC; e.g. Campbell and Fletcher, 2010) based on their typology and from 223 the painted decoration that used manganese pigments for black color. The youngest 224 Halaf pottery belongs to polychrome Late Halaf types associated with some Impressed 225 Ware known as Dalma types and Ubaid-style ceramics (Masetti-Rouault, 2005; Robert et 226 al., 2008; Robert, 2010). We sampled in Locus K171 two groups of these fragments with

227 fine mineral-tempered clay paste (pottery groups SY37, SY38), the first in the 228 occupation layer referred to as E2, and the second on floor E7 on top of layer E2.

229

230 3. New archeomagnetic intensity results

231 All the archeointensity measurements reported in this study were obtained using 232 the experimental protocol developed by Le Goff and Gallet (2004) for the Triaxe 233 magnetometer. The details of this experimental protocol can be found in Le Goff and 234 Gallet (2004) (see also Genevey et al., 2009; 2013; Hartmann et al., 2010; Gallet et al., 235 2014). We only recall here that it relies on magnetization measurements of a small 236 specimen $\left($ <1cm³ $\right)$ directly carried out at high temperatures and on a sequence of 237 measurements (with successive heating and cooling cycles) automatically performed 238 over a fixed temperature range between a low temperature referred to as T_1 (typically 239 of 150°C) and a high temperature referred to as T_2 (typically between 500°C and 530°C). 240 In the past few years, a relatively large collection of archeointensity data of different 241 ages and of different origins was obtained using the Triaxe, and comparative studies 242 with results derived from more classical methods (i.e. from the Thellier and Thellier's 243 (1959) method as revised by Coe (1967) or from the IZZI version of Thellier and 244 Thellier's (1959) method; e.g. Yu et al., 2004) demonstrated the reliability of the Triaxe 245 intensity data when quality criteria are taken into account. In our study, we use the 246 same quality criteria relative to the intensity determination for a specimen as those 247 described by Genevey et al. (2009) and Hartmann et al. (2010, 2011), and which were 248 also used more recently by Genevey et al. (2013), Gallet et al. (2014) and Gallet and 249 Butterlin (2014) (Supplementary Table 1). In particular, these criteria allow us to 250 eliminate the data that could be biased due to alteration of the magnetic minerals during 251 heating. Moreover, the temperature range over which the intensity determinations are 252 recovered from each specimen is precisely adjusted so that the analyzed magnetization 253 component is univectorial and corresponds to the magnetization acquired during the 254 manufacture of the pottery. Fig. 3 shows two examples of demagnetization behaviors. 255 After the removal of the viscous low-temperature component, the first behavior shows a 256 single magnetization component above \sim 200°C (SY89-08), while the second behavior 257 reveals two components (SY140-06). In these cases, the temperature range was 258 adjusted above \sim 200°C and \sim 340°C, respectively for obtaining intensity determinations

259 at the specimen level. Finally, the intensity data should not be affected by the presence 260 of multidomain magnetite grains and they take into account both the thermoremanent 261 magnetization (TRM) anisotropy and cooling rate effects on TRM acquisition (for a 262 thorough discussion on these aspects, see for instance in Le Goff and Gallet, 2004; 263 Genevey et al., 2008, 2009; Hartmann et al., 2010).

264 Our archeointensity analyses were complemented by hysteresis measurements 265 and by isothermal remanent magnetization (IRM) acquisition up to 0.8 T performed at 266 Saint Maur using a laboratory-built inductometer coupled with an electro-magnet. In 267 most cases, two fragments were analyzed for each group of fragments. IRM 268 measurements show very similar behaviors with saturation reached in relatively low 269 magnetic fields $\left(\sim 0.2 - 0.3 \text{ T}\right)$, indicating the absence of high-coercivity minerals (Fig. 4a). 270 We note that the hysteresis loops are generally not constricted (Fig. 4b-c). 271 Thermomagnetic low-field susceptibility curves obtained using a KLY-3 Kappabridge 272 coupled with a CS3 thermal unit show that the existing magnetic grains have maximum 273 unblocking temperatures below 600°C (Fig. 4d-g). All these magnetic properties indicate 274 that the magnetization of our specimens is most probably predominantly carried by 275 minerals of the (titano)magnetite family. Furthermore, the thermomagnetic curves 276 exhibit variable behaviors, independently of the age of the fragments, which suggests 277 the presence of (titano)magnetite with different titanium contents or different grain 278 sizes. We also observe a good reversibility between the heating and cooling 279 susceptibility vs. temperature curves, which constitutes a good marker of the stability of 280 the magnetic mineralogy on heating. We note that these magnetic properties are very 281 similar to those we previously obtained from Syrian fired-clay artifacts of younger ages 282 (e.g. Genevey et al., 2003; Gallet et al., 2008; 2014; Gallet and Butterlin, 2014).

283 Except for one case, the hysteresis parameters obtained for the fragments from 284 Tell Halula lie within the pseudo-single domain (PSD) range of magnetite defined by 285 Dunlop et al. (2002a) when projected on a Day plot (Day et al., 1977). Most M_{RS}/M_S and 286 H_{CR}/H_C ratios are concentrated inside a restricted area, with ~0.30>M_{RS}/M_S>~0.15 and 287 \sim 4>H_{CR}/H_C> \sim 2.5), above the theoretical mixing curves for mixture of SD and MD 288 magnetite grains but also well below the mixing curve of SD and superparamagentic 289 (SP) magnetite grains (Fig. 4h). According to Dunlop (2002b), this may reflect a large 290 distribution of grain sizes, including SP, SD and MD magnetite grains. In contrast, most

291 of the hysteresis parameters obtained from Tell Masaïkh (open blue triangles in Fig. 4h) 292 fall within the theoretical SD-MD mixing curves defined by Dunlop (2002a), therefore 293 indicating a coarser grain size distribution for those specimens. It is worth mentioning 294 that the evolution of the techniques (preparation of the clay paste, firing conditions) 295 used to produce ceramics at Tell Halula between the pre-Halaf and Halaf periods is 296 clearly not reflected in the hysteresis ratios, their dispersions being very similar 297 regardless of the age of the fragments (colored symbols in Fig. 4h). Further considering 298 the data from Tell Masaïkh and the previous ones obtained from Ebla/Tell Mardikh 299 (grey dots in Fig. 4h; Gallet et al., 2014), it appears that the distribution of the hysteresis 300 parameters obtained at a given archeological site constitutes a magnetic signature of the 301 clay source used to produce pottery at this site, and it may be used as an identification 302 tool complementary to more classical chemical analyses.

303 Fig. 5 shows the intensity results obtained from eight pottery groups. Each curve 304 from each panel shown in this figure exhibits the intensity data obtained for one 305 specimen over a temperature range often exceeding 200-250°C. In general, we only 306 analyzed one specimen per fragment. However, when the number of favorable 307 fragments was ≤ 5 (i.e. for pottery groups SY127, SY125, SY95, SY38), we analyzed three 308 specimens from each fragment and we first estimated a mean intensity value at the 309 fragment level before computing a mean value at the group level. The success rate of our 310 archeointensity analyses significantly varies according to the archeological periods. 311 While it is only 36% for the pre-Halaf period (54 fragments from 151 analyzed 312 fragments) and 56 % (22 from 39 fragments) for the proto–Halaf period, it increases up 313 to 70% for the sites dated in the Halaf period (133 from 191 fragments) and 67% for the 314 Halaf-Ubaid Transitional period (16 favorable fragments from 24 studied fragments). 315 The relatively low success rate for the pre-Halaf fragments is mainly due to the presence 316 of two magnetization components, which is likely related to the use of these ceramics for 317 cooking (hence preventing in many cases the clear isolation of a primary 318 magnetization). Examples of failed results are reported in Supplementary Fig. 1. Overall, 319 we analyzed a total of 405 fragments, among which 225 fragments (254 specimens) 320 yielded favorable archeointensity results, allowing us to determine 24 mean intensity 321 values at the pottery group level. Results obtained at the specimen/fragment level are 322 detailed in Supplementary Table 2, while Table 1 provides the group-mean intensity 323 values. These intensity values are generally well defined, with a number of fragments

324 analyzed per site larger or equal to 7 for 19 pottery groups (≥ 10 for 10 sites) and a 325 standard deviation always of less than 5 μ T, ranging between 1.8 % and 11.4 % of the 326 corresponding group-mean intensity values (≤ 5.0 % for 10 sites and ≤ 7.5 % for 21 327 among the 24 studied pottery groups). We note, however, that the mean intensity value 328 obtained for group SY125 (~6650-6550 BC) is only defined by two fragments (6 329 specimens), but it was kept for the discussion below because of the scarcity of such old 330 archeointensity data.

331

332 4. Late Neolithic archeointensity variations in the Middle East

333 The new archeointensity data are reported in Fig. 6 (see also Supplementary Fig. 334 2, where the results are averaged over the successive occupation levels). The new 335 results show that the time interval between ∼7000 BC and ~5000 BC was apparently 336 marked in the Middle East by an overall decreasing trend in geomagnetic field intensity. 337 This decrease was however not regular. In particular, a relative intensity minimum is 338 observed at the beginning of the Late Halaf period (pottery group SY86-131 with 20 339 favorable fragments), around the middle of the 6th millennium BC. An intensity peak 340 appears to have occurred during the Late Halaf period, between ∼5550 and ∼5300 BC. 341 This intensity peak is supported by the low geomagnetic field intensity values obtained 342 at Tell Masaïkh for the Halaf-Ubaid Transitional period.

343 We compared the new Tell Halula and Tell Masaïkh data with two other 344 archeointensity datasets of the same age previously obtained in relatively nearby 345 regions (Fig. 7). The first dataset includes results obtained at Yarim Tepe II and Tell 346 Sotto, two multi-level archeological sites from northern Iraq (Fig. 1a; Nachasova and 347 Burakov, 1995, 1998). In these two studies, the pottery fragments were selected and 348 dated according to their stratigraphic position within a sequence of archeological 349 deposits (with a total thickness of 780 cm at Yarim Tepe II and 280 cm at Tell Sotto), and 350 assuming a constant accumulation rate of archeological deposits. Although such a 351 sampling procedure may obviously introduce large uncertainties in the dating of the 352 studied fragments, it nevertheless appears that this approach can provide satisfactory 353 results (e.g. Nachasova and Burakov, 1998; Kostadinova-Avramova et al., 2014). 354 However, in both cases, the dating considered by Nachasova and Burakov (1995, 1998)

355 appears systematically shifted by several centuries relative to the most recent 356 chronological Pottery Neolithic time scale (see Campbell, 2007; Bernbeck and 357 Nieuwenhuyse, 2013). Indeed, the fragments from Yarim Tepe II are unambiguously 358 archeologically dated to the Middle-Late Halaf period (∼5750-5300 BC; e.g., Campbell, 359 2007; Robert, 2009; Bernbeck and Nieuwenhuyse, 2013 and references therein), but 360 their ages were mostly assigned in the 5th millennium BC. Similarly, the fragments 361 collected at Tell Sotto were dated to the middle of the 6th millennium BC by Nachasova 362 and Burakov (1998), but the studied ceramics are dated to the Late Pre-Halaf (Late 363 proto-Hassuna and Archaic Hassuna cultural phases), i.e. between ∼6400 and ∼6050 BC 364 (e.g. Bader, 1989; Bader and Le Mière, 2013; Le Mière pers. comm. 2014).

365 For these reasons, we assigned new ages to Yarim Tepe II and Tell Sotto 366 considering first, the stratigraphic position of the concerned fragments as provided by 367 the authors and second, assuming that the entire Middle-Late Halaf and Late Pre-Halaf 368 periods were represented in the Yarim Tepe II and Tell Sotto deposits (like the authors 369 considered but for two other time intervals). Finally, for displaying in Fig. 7a the data 370 obtained at Yarim Tepe II, with only a single specimen studied per fragment, and at Tell 371 Sotto we also performed intensity averaging over several fragments when the latter 372 come from the same stratigraphic intervals, i.e. each time there was a group of 373 fragments considered of the same age. We observe an overall good agreement with the 374 data obtained at Tell Halula and Tell Masaïkh. In particular, this agreement confirms the 375 occurrence in northern Mesopotamia of a relative intensity minimum around the middle 376 of the 6th millennium BC, which further strengthens the occurrence of an intensity peak 377 at the beginning of the second half of the 6th millennium BC.

378 The second archeointensity dataset comprises the results encompassing the 6th 379 millennium BC from Bulgaria that were recently updated by Kovacheva et al. (2014) 380 (Fig. 7b). From this new analysis, a century-scale intensity peak seems to be emerging 381 around the middle of the $6th$ millennium, which might coincide, within age uncertainties, 382 with that observed from the Syrian Late Halaf data. According to this interpretation, the 383 data available for the Halaf-Ubaid Transitional period would come prior to the 384 geomagnetic field intensity increase observed in the Bulgarian data at the end of the $6th$ 385 millennium BC. Constraining further this preliminary correlation will require the 386 acquisition of new archeointensity data in the Balkans and in the Middle East.

387

388 5. Discussion

389 We have undertaken the construction of a geomagnetic field intensity secular 390 variation curve in the Middle East during the Holocene. For this purpose, we selected all 391 the archeointensity data available inside a circle with a radius 1000 km around the 392 archeological site of Tell Halaf (λ=36°49'N, φ=40°02'E; Supplementary Fig. 3). The data 393 were retrieved from the ArcheoInt database (Genevey et al., 2008) and complemented 394 with the more recent studies (Ben-Yosef et al., 2009; Gallet and Al Maqdissi, 2010; Shaar 395 et al., 2011, 2014; Ertepinar et al., 2012; Gallet et al., 2014; Gallet and Butterlin, 2014). 396 They were obtained from the eastern part of Turkey, Cyprus, Syria, the Levant, Iraq, 397 from the western part of Iran and from the Caucasus. Note that the large dataset from 398 the Balkans and Greece (e.g. De Marco et al., 2008; Tema and Kondopoulou, 2011; 399 Kovacheva et al., 2014) has not been included to allow it to be compared to different 400 regional secular variation behaviors from elsewhere (e.g. between the Middle East, 401 Eastern Europe and Western Europe). Genevey et al. (2008) proposed a set of selection 402 criteria in order to distinguish between all available data those that meet minimum 403 quality criteria. This approach enabled the construction of two datasets referred to as 404 "Selected data" and "All data" in Genevey et al. (2008). Hereafter we have considered the 405 compilation of selected data to calculate the Middle East geomagnetic field intensity 406 variation curve, considering the new dating we estimated for Tell Sotto and Yarim Tepe 407 II and using, for these two sites, the mean intensity values computed from fragments 408 associated with the same stratigraphic level (Fig. 7a).

409 To calculate our curve, we first applied a method based on the use of sliding 410 windows of 200 years successively shifted by 10 years through the past 9 millennia. We 411 computed VADM values only for those time intervals containing at least 3 results. 412 Following Thébaut and Gallet (2010) and Licht et al. (2013), we also used the bootstrap 413 technique with 1000 runs by introducing random noise in the data within their 414 experimental and age uncertainties. This allowed us to compute 1000 intensity variation 415 curves. In Fig. 8a we displayed the averaged VADM (thick line) together with the 416 minimum and maximum VADM values obtained for the different sliding windows, hence 417 defining an envelope of equally possible VADM values. Due to the insufficient number of 418 archeointensity data spanning the 5th and 4th millennia BC, no averaged curve could be

419 determined between ∼4930 BC and ∼3650 BC, i.e. during the Ubaid and Uruk periods in 420 Mesopotamia. This time interval therefore constitutes a particularly important target for 421 future archeomagnetic studies in the Middle East. For other periods, the computed curve 422 appears very consistent with almost all the Syrian data (blue dots in Fig. 8a; Genevey et 423 al., 2003; Gallet and Le Goff, 2006; Gallet et al., 2006, 2008, 2014; Gallet and Al Maqdissi, 424 2010; Gallet and Butterlin, 2014 and this study). We observe the same variation trends, 425 with distinct intensity maxima during the second half of the first millennium AD, at the 426 beginning of the first millennium BC and around the middle of the third millennium BC. 427 Supplementary Fig. 4 also exhibits the averaged intensity curve computed without the 428 Syrian data, showing in particular that the latter data set allows us to better constrain 429 the curve during the third millennium BC (note that this curve takes into account the 430 new dating of the Tell Sotto and Yarim Tepe II data). The temporal resolution of 200 431 years of the regional averaged curve most probably prevents the recovery of distinct 432 century-scale intensity (VADM) maxima at ∼1500 BC, ∼2550 BC and ∼2300 BC clearly 433 observed from Syrian data at Ebla and Mari (Gallet et al., 2008, 2014; Gallet and 434 Butterlin, 2014), as well as the maximum in intensity between ∼5500 and ∼5300 BC 435 exhibited by the Tell Halula data or the spike events proposed by Ben-Yosef et al. (2009) 436 and Shaar et al. (2011) at the very beginning of the first millennium BC.

437 The second approach is similar to the method described above but relies on the 438 more complex cubic B-splines time parameterization and uses an iterative scheme to 439 identify and then to weight the data that are considered as outliers (Fig. 8b; modified 440 from Thébault and Gallet, 2010). The algorithm first proposes a set of possible spline 441 knots irregularly spaced. The spacing is designed to take full advantage of the varying 442 time resolution between epochs that arises from the uneven time distribution of the 443 reference archeomagnetic data. For instance, it is found that the maximum achievable 444 time resolution is about 150 years between 7000 BC to about 5000 BC and between 445 ∼3000 BC and 2000 AD, while searching for features with time resolution lower than 446 800 years makes little sense between ∼5000 BC and ∼3000 BC. Then, the data are as 447 before 1000 times randomly noised within their a priori error bars. For each curve, the 448 algorithm checks whether the maximum likelihood solution belongs to the a priori 95% 449 error bar of the data and weights accordingly the data that are systematically outside 450 this confidence interval. Fig. 8b displays the final solution with the maximum probability

451 in black and its 95% fluctuation envelope in light blue. This envelope contains 95% of 452 the maximum likelihood curves estimated by the bootstrap for the 1000 iterations, and 453 it highlights the variability between the different curves. This parameter is important for 454 testing the precision of the most probable curve and for identifying the fine time 455 variations that persist after resampling. Formally, however, the statistical significance of 456 a time variation can be assessed only after the computation of the 95% confidence 457 interval (in red) that is traditionally calculated a posteriori from the misfit function 458 between the data and the ensemble of models. Compared to the first approach, the 459 likelihood solution provided in supplementary Table 3 is generally smoother. This 460 feature is desired for testing whether the apparent fine time variation of the maximum 461 likelihood can be considered as robust. A striking feature emerging from the comparison 462 between Fig. 8a and Fig. 8b is that the final solution is independent of the chosen 463 modeling scheme. This is seemingly positive evidence that the observed magnetic field 464 intensity variations are well constrained (within the given time resolution) by the 465 available data in the chosen geographical area.

466 We then sought to constrain the variations in global geomagnetic dipole field 467 moment over the past 9 millennia. For this, we averaged the archeointensity data 468 available in the Middle East over sliding windows of 500 years, roughly assuming that 469 this rather long duration may suffice to average out most of the non-dipole 470 contributions (e.g. Hulot and Le Mouël, 1994; Genevey et al., 2008; Knudsen et al., 2008). 471 On the other hand, this averaging smoothes out the more rapid variations in dipole 472 moment over centennial time scales (Genevey et al., 2009; 2013). The curve constructed 473 using the same technique as in Fig. 8a is shown in Fig. 9a, together with the VADM 474 computed by Knudsen et al. (2008) using the global GEOMAGIA50 database (Korhonen 475 et al., 2008) and applying both temporal and geographical averaging to eliminate the 476 non-dipole components. As a general comment, the two curves exhibit the same dipole 477 behavior during the past three millennia (although the magnitude and the amplitude of 478 the variations are not strictly the same), characterized by two periods of stronger dipole 479 moment during the first millennium BC and during the second half of the first 480 millennium AD (see also Genevey et al., 2008; Hong et al., 2013). In contrast, these 481 curves are significantly different during the third millennium BC, with a smooth VADM 482 evolution in the case of the Knudsen et al. (2008) curve but with a distinct dipole 483 maximum in our Middle East curve. For older periods, there is again a good consistency

484 between the two curves, but we note the large error bars of Knudsen et al.'s (2008) 485 curve for the 7th-6th millennium segment. Thus the question remains as to the 486 significance of the dipole maximum observed in the Middle East during the third 487 millennium BC, which is well constrained by a significant number of data. Owing to the 488 rather good agreement between the two curves, especially during the past three 489 millennia, the VADM maximum we observe during the third millennium BC might well 490 be a global (dipole) geomagnetic feature that requires further confirmation. If true, it 491 would indicate that the dipole evolution varied more erratically than previously thought, 492 with an oscillatory behavior at least between ~3000 BC and 2000 AD of typical time 493 scale of about 1700 years (see also Burakov et al., 1998).

494 Fig. 9b compares our VADM variation curve with dipole moments derived from 495 global geomagnetic field modeling that was recently constructed using only 496 archeomagnetic and volcanic data (Pavón-Carrasco et al., 2014, in blue) and another that 497 also incorporated paleomagnetic data from sediments (Nilsson et al., 2014 in orange and 498 green; note that this latter reconstruction supersedes the previous field reconstruction 499 of Korte et al., 2011). The field models that partly rely on sediment data naturally show 500 time variations smoother than that of the models constructed using only the 501 archeomagnetic and volcanic data. Hence, the dipole moments derived by Nilsson et al. 502 (2014) during the 7th millennium BC are lower than the ones proposed by Pavón-503 Carrasco et al. (2014) and lower than the averaged VADM we estimated from the Middle 504 East. However, at the beginning of the first millennium BC, the VADM values from the 505 Middle East are much higher than the dipole moments from either models. Neither of 506 two reconstructions shows the distinct dipole maxima previously observed during the 507 past three millennia (Fig. 9a; Genevey et al., 2008; Knudsen et al., 2008), in particular the 508 one dated to the first millennium AD. This clearly poses the question of the consistency 509 between the VADM estimates and the time-varying dipole moment reconstructions. 510 Nevertheless, it could be argued that the field modeling of Pavón-Carrasco et al. (2014) 511 gives some support to the occurrence of a dipole moment maximum during the third 512 millennium BC (Fig. 9b). Such an agreement still needs to be confirmed because the 513 proposed field reconstruction shows numerous centennial-scale fluctuations with 514 similar amplitudes over the entire sequence, a feature whose geomagnetic origin is 515 questionable.

516 As a concluding remark, we point out that the different time-varying 517 archeomagnetic field reconstructions encompassing the 7th-5th millennium time 518 interval all suffer from the erroneous dating affecting the Yarim Tepe II and Tell Sotto 519 data. Together with the corrected Yarim Tepe II and Tell Sotto ages, the new 520 archeointensity data obtained in the present study dated to between 7000 BC and 5000 521 BC will help improve the reliability of the next generation of geomagnetic field models 522 spanning the Late Neolithic period. Besides implications for geomagnetism, this 523 improvement may be of particular interest in providing chronological time constraints 524 for archeological purposes, during a fascinating period (e.g. Berger and Guilaine, 2009) 525 that was marked by the beginning of the Neolithic expansion from the Middle East 526 toward Western Europe.

527

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777 Table caption

778 Table 1. Pottery group-mean intensity values obtained at Tell Halula $(\lambda=36^{\circ}25^{\prime}N,$ 779 $\omega = 38^{\circ}10'$ E; pottery groups SY127 to SY132) and Tell Masaïkh ($\lambda = 34^{\circ}25'$ N, $\omega = 40^{\circ}01'E$; 780 pottery groups SY37 and SY38). Information on the different archeological dating, 781 relative chronology and references are provided in the second, third and fourth 782 columns. See text for references on absolute dating (fifth column). * indicates that an 783 approximation was made on the dating (see text). The mean intensity values and their 784 standard deviations are provided in column 6. Column 7 shows the number Nb of 785 fragments (/n specimens) retained for computing the pottery group-mean intensity SC 786 values.

787

788 Figure captions

789 Fig. 1. (a) Location of the two Syrian archeological sites studied herein (Tell Halula and 790 Tell Masaïkh) and of three other sites discussed in the text (Tell Halaf, Yarim Tepe II and 791 Tell Sotto). ©Google Earth. (b) General view of the Tell Halula archeological site. © 792 Universitat Autónoma de Barcelona (UAB)/SAPPO.

793 Fig. 2. Examples of pottery sherds discovered at Tell Halula. These fragments are dated 794 to phases I, II and III of the pre-Halaf (photos 1-2, 3-4 and 5-6, respectively), to the 795 proto-Halaf (photos 7-8), and to the Early, Middle and Late Halaf (photos 9, 10-11 and 796 12-13, respectively). © Universitat Autónoma de Barcelona (UAB)/SAPPO.

797 Fig. 3. Triaxe intensity data obtained for two specimens from Tell Halula (SY89-08, 798 SY140-06). (a,c) Thermal demagnetization data; (b,d) Triaxe measurement series; (e) 799 Archeointensity results at the specimen level. See text and further explanations in Le 800 Goff and Gallet (2004).

801 Fig. 4. (a) Normalized IRM acquisition curves obtained for one fragment from each time 802 level. (b-c) Two examples of hysteresis loop. (d-g) Four examples of normalized 803 thermomagnetic low-field susceptibility (heating and cooling) curves obtained from 804 fragments collected at Tell Halula. These fragments are dated to the pre-Halaf (d,e), 805 Middle Halaf (f) and to the Late Halaf (g). (h) Hysteresis ratios $(M_{RS}/M_s \text{ vs. H}_{CR}/H_c)$ 806 obtained at Tell Mardikh/Ebla (grey color, Gallet et al., 2014), Tell Masaïkh (blue

807 triangles) and Tell Halula (see color code on the figure according to the archeological 808 periods of the fragments).

809 Fig. 5. Intensity data obtained from eight different archeomagnetic pottery groups (a-e, 810 Tell Halula; f, Tell Masaïkh). Each colored curve on each of these plots shows the 811 intensity data obtained for one specimen over the temperature range of analysis (for 812 further explanations, see in Le Goff and Gallet, 2004). Altogether, the results from 93 813 specimens are hence reported in this Figure.

814 Fig. 6. Archeomagnetic field intensity variations recovered from the new data obtained 815 at Tell Halula (blue circles) and Tell Masaïkh (blue triangles). All results are converted in 816 Virtual Axial Dipole Moments. The chronological time scale is provided in the text (see 817 also in Table 1).

818 Fig. 7. Comparison between our new archeointensity data (in blue) and previous results 819 obtained (a) from Yarim Tepe II and Tell Sotto (green circles and triangles, respectively), 820 two multi-level archeological sites located in North Iraq (Nachasova and Burakov, 1995, 821 1998) and (b) from Bulgaria (in red; Kovacheva et al., 2014). As discussed in the text, the 822 dating of the Yarim Tepe II and Tell Sotto data was modified from the original papers. 823 The solid vs. open circles indicate the intensity values obtained from several vs. one 824 specimen(s).

825 Fig. 8. Regional averaged geomagnetic field intensity variation curve in the Middle East 826 over the past 9000 years. The data were selected inside a 1000 km-radius circle around 827 the location $\lambda = 36^{\circ}49'N$, $\varphi = 40^{\circ}02'E$ (archeological site of Tell Halaf). All data were 828 transformed into VADM. Two different approaches were successively considered to 829 compute the curve. (a) We used sliding windows of 200 years shifted every 10 years and 830 the bootstrap technique for taking into account the experimental and age uncertainties 831 on the available intensity data. 1000 curves were hence computed and are shown here 832 the mean (thick black line), the minimum and the maximum VADM values obtained for 833 the different time windows. The Syrian data are also reported (blue dots) together with 834 all other available archeointensity data (grey dots) satisfying minimum selection criteria 835 (Genevey et al., 2008). (b) We used an iteratively reweighted least-squares algorithm, 836 combined with a bootstrap, modified from that of Thébault and Gallet (2010). The

837 continuous black line shows the maximum of probability, and the light blue lines its 95%

838 fluctuation envelope. The 95% confidence interval is displayed by the red lines.

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839 Fig. 9. Comparison between the geomagnetic field intensity (transformed into VADM) 840 variation curve in the Middle East, with averaging over sliding windows of 500 years 841 (black lines; see text), and previous dipole field moment reconstructions. The 842 comparison is made with (a) the VADM variation curve computed by Knudsen et al. 843 (2008) using temporal and geographic averaging (in red), (b) dipole moment 844 reconstructions derived from different time-varying global geomagnetic field modeling 845 (blue lines, modeling proposed by Pavón-Carrasco et al., 2014; orange and green lines, 846 the pfm9k.1b and pfm9k.1a modeling proposed by Nilsson et al., 2014).

MANU C

847

250 km

Figure 3

Figure 5

Figure 5

Figure 7

Figure 9

Table 1

