

## Palaeomagnetism of the loess/palaeosol sequence in Viatovo (NE Bulgaria) in the Danube basin

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Diana Jordanova, Jozef Hus, Jordan Evlogiev, Raoul Geeraerts. Palaeomagnetism of the loess/palaeosol sequence in Viatovo (NE Bulgaria) in the Danube basin. Physics of the Earth and Planetary Interiors, 2008, 167 (1-2), pp.71. 10.1016/j.pepi.2008.02.008 . hal-00532138

### HAL Id: hal-00532138 https://hal.science/hal-00532138

Submitted on 4 Nov 2010

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### Accepted Manuscript

Title: Palaeomagnetism of the loess/palaeosol sequence in Viatovo (NE Bulgaria) in the Danube basin

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PII:S0031-9201(08)00032-0DOI:doi:10.1016/j.pepi.2008.02.008Reference:PEPI 4900To appear in:Physics of the Earth and Planetary InteriorsDouble in the intervention of the second sec

 Received date:
 16-1-2007

 Revised date:
 12-2-2008

 Accepted date:
 13-2-2008

Please cite this article as: Jordanova, D., Hus, J., Evlogiev, J., Geeraerts, R., Palaeomagnetism of the loess/palaeosol sequence in Viatovo (NE Bulgaria) in the Danube basin, *Physics of the Earth and Planetary Interiors* (2007), doi:10.1016/j.pepi.2008.02.008

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1	Palaeomagnetism of the loess/palaeosol sequence in Viatovo (NE Bulgaria) in the
2	Danube basin.
3	
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5	
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11	
12	Abstract
13	The results of a palaeomagnetic investigation of a 27m thick loess/palaeosol sequence in
14	Viatovo (NE Bulgaria) are presented in this paper. The sequence consists of topsoil $S_0$ , 7 loess
15	horizons (L <sub>1</sub> -L <sub>7</sub> ) and 6 interbedded palaeosols (S <sub>1</sub> -S <sub>6</sub> ) overlying a red clay (terra rossa)
16	complex. Magnetic viscosity experiments, IRM acquisition, AMS analysis and NRM stepwise
17	alternating and thermal demagnetisation experiments of pilot samples were implemented for
18	precise determination of the characteristic remanence and construction of a reliable
19	magnetostratigraphical scheme. Analysis of IRM acquisition curves using the expectation -
20	maximization algorithm of Heslop et al. (2002) suggests that the best fitting is obtained by
21	three coercivity components. Component 1 corresponds to SD maghemite/magnetite, while
22	component 2 is probably related to the presence of oxidised detrital magnetites. The third
23	component shows varying coercivities depending on the degree of pedogenic alteration of the
24	samples and probably reflects the presence of detrital magnetite grains oxidised at different
25	degree.

The relevance of the Viatovo section as a key representative sequence for the loess cover in the Danube basin is confirmed by the presence of geomagnetic polarity changes in the lower part of the sequence. The youngest one recorded in the seventh loess unit  $L_7$  can be identified as corresponding to the Matuyama/Brunhes palaeomagnetic polarity transition. Two normal magnetozones were found in the red clay complex, probably corresponding to the Jaramillo and Olduvai subchronozones of the Matuyama chron.

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Key words: loess-palaeosol, viscous remanence, IRM acquisition, IRM component unmixing,
 magnetostratigraphy, Matuyama/Brunhes boundary, Jaramillo and Olduvai subchronozones,
 Bulgaria.

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#### 37 **1. Introduction**

38

39 Palaeomagnetism is one of the rare methods for chronostratigraphical correlation and 40 dating of Middle and Upper Pleistocene loess/palaeosol sediments, mainly because of the 41 time limits of other dating techniques and the lack of micro- and macrofaunal findings with 42 dating significance. The latter is particularly the case with the Viatovo sequence examined 43 below (Evlogiev, 2006). Indeed, other dating methods: amino acid racemification (Wehmiller 44 and Miller, 2000), thermoluminescence (Forman et al., 2000, Singhvi et al., 2001), Optically 45 Stimulated Luminescence (Huntley et al., 1985; Hutt et al., 1988) and radiocarbon (Trumbore, 46 2000) dating are limited in time. On the other hand, records of geomagnetic field reversals 47 such as the position of the Matuyama/Brunhes (M/B) polarity reversal and the possible 48 presence of "excursions" or short-term reversals within the Matuyama and Brunhes chrons are 49 important stratigraphic markers. Especially significant is the contribution of palaeomagnetism 50 to a reliable stratigraphic correlation of the East European loess deposits, in particular in the

51 Danube basin because of the varying sources of aeolian dust, differences in geomorphological 52 relief and climate gradients (Pye, 1987). Investigations of the loess-palaeosol sediments from 53 the Northern Black Sea coastal area by Dodonov et al. (2006) identified the 54 Matuyama/Brunhes boundary at the base of pedocomplex S<sub>7</sub>. Nawrocki et al. (2002) found 55 the M/B transition in the sequence of Zahvizdja (East Carpathian Foreland) in an interglacial 56 soil, equivalent to oxygen isotope stage 19. In the Western Black Sea area, Tsatskin et al. 57 (2001) and Gendler et al. (2006) discovered the same polarity boundary respectively between 58 pedocomplexes PK6 and PK7 in Roxolany and in the upper part of PK7 in Novaya Etuliya. 59 The aim of the present study is to establish a reliable magnetostratigraphy of the loess-60 palaeosol sediments in the low Danube area through detailed rock magnetic (Jordanova et al. 61 2007) and palaeomagnetic investigations of the Viatovo sequence. 62

- 63 **2.** Loess cover in Bulgaria and site description.
- 64

The recent geomorphological relief in North Bulgaria is blanketed by widespread loess deposits. Loess-palaeosol sequences of various thickness and age occur on certain palaeogeomorphological forms: the Pliocene Denudation Surface (PDS); the Old Abrasive-Accumulative Level (OAAL); Pleistocene river terraces  $T_6$ ,  $T_5$ ,  $T_4$ ,  $T_3$ ,  $T_2$ ,  $T_1$ ,  $T_0^1$  and the Holocene terrace  $T_0$  (Fig. 1a). Each of them has a typical lithology and stratigraphic position (Evlogiev, 1993).

The oldest palaeogeomorphologic form, PDS, in North Bulgaria was shaped during the Pliocene along the shore of the Dacian lake basin. It is characterised by an undulating to flat relief in lower Cretaceous limestones and sandstones at an absolute altitude of 200–250m. In the study area, in Viatovo, a number of palaeokarsts assumed to be of Albian age developed and were buried subsequently by kaolinitic sands. The complete sedimentary

76 sequence on the PDS form above the kaolinitic level in this region consists of red clays and a 77 loess/palaeosol sequence of 8 loess horizons and 7 palaeosols according to Evlogiev (2006). 78 The second geomorphologic unit, the OAAL, was formed at the beginning of the Biber glacial 79 about 2.59 Ma ago, by cold invasion and intensification of neo-tectonic movements. As a 80 result, significant hydrological changes occurred. In the realm of the Dacian basin these 81 changes were dominated by erosional incision that sculpted the OAAL relief. Its absolute 82 height is 70-90 m and it is sloping towards the Danube River. The OAAL consists of two 83 facies in the area of Russe: a basal gravel-sand complex (dated at 2.59 - 1.06 Ma) covered by 84 a gravel-clay complex (1.06 - 0.84 Ma). The ages mentioned are deduced from fauna, magnetostratigraphy and geochronology (Evlogiev, 2006). A complete loess/palaeosol 85 86 complex developed above the fluvio-lacustrine sediments, or the two gravel-clay complexes. The number of loess and palaeosol units on the seven river terraces  $T_6 - T_0^{-1}$  depends strongly 87 88 on the age of the terrace formation. Six loess units occur on T6 but only one on  $T_1$ .

89 The Viatovo loess/palaeosol sequence in NE Bulgaria is exposed in the walls of a quarry 90 that was opened for extraction of kaolin (Fig. 1b). Palaeosols and loess units that could be recognised in the field were numbered and are designated respectively  $S_1, S_2$ ...and  $L_1$ , 91  $L_2...$  The sequence consists of topsoil  $S_0$ , 7 loess horizons ( $L_1$  -  $L_7$ ) and 6 interbedded 92 palaeosols  $(S_1 - S_6)$  resting on a red clay (terra rossa) complex. In the sampling area, the 93 94 recent soil  $S_0$  was truncated by guarry activities and only the lower part was preserved, 95 characterised by its dark gray colour and prismatic structure. The upper three palaeosols  $(S_1 - C_1)$ 96  $S_3$ ) are dark brown in colour, becoming gradually more reddish towards the bottom part of the 97 units. Abundant reddish crotovinas (burrows) perforate the uppermost soil horizon. A 98 prismatic structure is typical for the middle part of the soil units. Their lower parts are rich in CaCO<sub>3</sub> nodules, reaching sizes of a few centimetres in the bottom of  $S_2$ . Palaeosols  $S_4 - S_6$ 99 100 show a brown-reddish to brown-pinkish colour, have a compact clayey prismatic structure

101 and contain CaCO<sub>3</sub> concretions of different sizes (up to 5cm) in their bottom parts. The first 102 loess unit L<sub>1</sub> shows signs of secondary pedogenic alterations. Its colour is dark brown and it 103 has a granular structure with CaCO<sub>3</sub> coatings. Crotovinas, several centimetres in size, are 104 visible over the whole unit and increase in number and size in its lower part. The least altered 105 loess unit is L2, yellowish in colour and with abundant CaCO<sub>3</sub> specks. Loess unit  $L_3$  is 106 affected by pedogenic alteration, brown in colour and with abundant CaCO<sub>3</sub> spots. The 107 thickness of unit  $L_4$  is only 50cm and it consists of brown-reddish crumbly loess with large 108 CaCO<sub>3</sub> blocks. Unit  $L_5$  is more powdery loess with CaCO<sub>3</sub> spots that are locally highly 109 concentrated and with red crotovinas in the bottom part. The sixth loess unit L<sub>6</sub> is brown-110 pinkish in its upper part and becomes brown and more clayey in depth. Dark brown 111 crotovinas are visible over the whole unit. Loess unit  $L_7$  shows also signs of severe 112 weathering. It is brown reddish to brown pinkish in colour, clayey and contains CaCO<sub>3</sub> spots. 113 Downward, it becomes yellowish with large brown reddish veins of clayey silt that occupy about 30% of the total surface in a vertical section. The red clays in the bottom part of the 114 115 sequence, reaching a total thickness of 5.6m, have a prismatic structure with shiny black Mn 116 coatings on the fractures. Horizontal CaCO<sub>3</sub> lenses (5 to 10cm thick and 20 to 40cm long) 117 occur at about 2.9 m below the top of the red clay complex, which are further down replaced 118 by red clay mixed with large irregular CaCO<sub>3</sub> blocks. The bottom part of the sequence, ending 119 at the kaolin level, is compact yellow sandy clay with some CaCO<sub>3</sub> lenses.

120

#### 121 **3. Methods**

The complete sequence has been sampled in four non-overlapping sections. Section 1 includes: topsoil  $S_0$ , first loess unit  $L_1$ , first palaeosol unit  $S_1$  and units  $L_2$ ,  $S_2$  and  $L_3$ . Section 2 covers units  $S_3$ ,  $L_4$ ,  $S_4$ ,  $L_5$ ,  $S_5$  and  $L_6$ . Section 3 contains palaeosol  $S_6$  and the underlying loess unit  $L_7$ . The fourth section cuts 5.60 m through the red clays down to the kaolin level. The

126 vertical sections were cut deeply in the walls of the pit in order to reach fresh, non exposed 127 material. As the main units could be followed easily along the walls of the pit (Fig. 1b) and 128 the whole sequence was sampled in only four vertical sections, the risk of overlapping and/or 129 sampling gaps could be avoided. Hence, the total thickness of the complete composite section 130 after adding the four sections is 27m. In total, 275 oriented cylindrical samples (sampling 131 interval of 10cm) for palaeomagnetic analysis were obtained with a hand-held piston driven, 132 split corer, specially designed for the purpose. The corer was driven horizontally in the 133 sediment and the cores oriented with a clinometer, solar and magnetic compass, in the same 134 way as it is commonly done for hard rocks. Mechanical disturbance was minimized as the 135 core (2.5cm in diameter and about 7cm long) can be recuperated by opening the two halves of 136 the corer. Close to section 1, another parallel section encompassing  $S_0$ ,  $L_1$ ,  $S_1$ ,  $L_2$  and  $S_2$  was 137 sampled additionally with a narrower sampling interval of 5cm for verification of the 138 presence (or absence) of the Blake event.

139 Moreover, two decimetre-sized oriented block samples were cut in all the units and loose 140 samples taken from the whole sequence with a sampling interval of 5cm for, respectively, 141 anisotropy of magnetic susceptibility (AMS) measurements and rock magnetic analysis. 142 Remanent magnetisation measurements were carried out in a three-axis cryogenic rock 143 magnetometer of 2G Enterprises, equipped with an in-line alternating field demagnetiser. 144 Isothermal remanent magnetisations were imparted to samples of each unit in a 2G pulse 145 magnetiser in fields up to 2.6T. Small one-inch cubes were cut from the oriented block 146 samples for AMS measurements in a KLY-3S magnetic susceptibility meter.

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#### 149 4. Experimental results

150 Rock magnetic properties and grain size analysis of all the units had been investigated in 151 detail before the palaeomagnetic examination started (Jordanova et al. 2007). In brief, 152 magnetic mineralogy deduced from thermomagnetic analysis of magnetic susceptibility, 153 induced magnetisation in strong fields and Mössbauer analysis suggest that magnetite, 154 oxidised magnetite and hematite are the main ferri- and antiferromagnetic phases present. 155 Mössbauer analysis of magnetic separates from S<sub>1</sub> and L<sub>2</sub> yielded more or less similar results. 156 The presence of maghemite was revealed in the strong magnetic fraction of  $S_1$  but not in  $L_2$ 157 The clay content along the sequence does not vary significantly and the mean percentage, 158 both in loess and soil horizons, is around 30. Exceptions are the less altered loess units L<sub>2</sub> and 159  $L_5$  with lower clay content. The observed high clay content but small differences in clay 160 content between soils and loesses are manifestation of the enhanced weathering and 161 pedogenesis, even during the glacials, particularly in the lower part of the section. All the 162 palaeosol units are characterised by high frequency dependent magnetic susceptibility ( $\chi_{FD}$ %) 163 with values up to 12%, pointing to a significant superparamagnetic (SP) fraction. A relatively 164 high content of SP Fe<sub>3</sub>O<sub>4</sub> grains can also be assumed for loess units  $L_1$ ,  $L_3$ ,  $L_6$  and  $L_7$ 165 (Jordanova et al. 2007). The presence in loesses of pedogenic magnetically soft grains in the 166 SP and viscous SP/SD (SD = single domain) state is also supported by the relatively low 167 coercivity (B<sub>c</sub>) and coercivity of remanence (B<sub>cr</sub>) values in these units, comparable to the 168 values attained in the palaeosols.

Additional rock magnetic experiments that are presented below yield further information
about the nature and magnetic stability of the remanence carriers as well as of the magnetic
fabric of all the units.

Fifteen samples were prepared from loose materials collected in all the units for a viscous remanent magnetisation (VRM) acquisition experiment. Mechanical redistribution of the grains, randomising any pre-existing remanence was chosen as a "demagnetisation"

175 technique. This option was preferred in order not to change the original domain state as is the 176 case during AF demagnetisation (Jordanova et al. 2007), or mineralogy that often happens 177 when heating loess and soil samples above their Curie-temperatures (Tc) followed by cooling 178 in zero field. Gently crushed and sieved material was pressed into PVC cylinders and sealed 179 with epoxy resin. Spontaneous VRM build-up in an ambient field of 48 µT was monitored over 2 months (Fig. 2). Samples from the upper part of the sequence  $(S_0 - L_4)$  show two 180 181 different linear segments in the VRM versus log(t) graphs, separated at the elapsed time of 182 about 6 weeks [corresponding to log (t) = 5.57]. Both palaeosols and loesses (respectively  $S_0$ -183  $S_3$  and  $L_1$ - $L_4$ ) show this peculiarity in contrast to the units from the lower part of the section 184  $(S_4 - L_7, red clay)$ . Calculation of viscous acquisition coefficients (Sa), taking separately the 185 two slopes of the VRM(log t) relationship (respectively Sa1 and Sa2), reveals the presence of 186 a fraction that acquires viscous magnetisation more slowly. In order to evaluate the 187 significance of an ultrafine grained fraction in the total viscosity, a plot of the Sa1 coefficients as function of  $\chi_{FD}\%$  (Forster et al., 1994) is given in Fig. 3. The leading role of the 188 189 concentration on the amount of viscous acquisition is revealed by the good linear correlation between Sa1 and  $\chi_{FD}$ % (with correlation factor R<sup>2</sup> = 0.91). 190

191 Stepwise IRM acquisition in pulse fields up to 2.6 T was monitored for 15 samples 192 representative for each lithological unit. Magnetic mineral components contributing to the 193 total remanence were unmixed, following the procedure by Heslop et al. (2002) and applying 194 the IRM Unmix software. The initial state of the samples was an AF-demagnetisation state, 195 which is most suitable when fitting log-normal distributions to IRM acquisition curves 196 (Heslop et al., 2004). The best fit was obtained with a 3-component model and some 197 representative examples are shown in Figure 4. Component 1, which is the main component 198 in the palaeosol samples, is characterised by a relatively low coercivity maximum  $B_{1/2}$  at 199 around 27mT in the palaeosol as well as in the loess samples (Fig.4 and Table 1). In the

200 palaeosol samples it contributes from 40 up to 70% (with an average of 51%) to the total 201 IRM. On the other hand, component 2 is the main component in the loess samples with an 202 average contribution of 53% to the total IRM and a coercivity peak around 70 to 80mT. A 203 distinct high-coercivity component, component 3, with quite varying maximum coercivities 204 between about 150 and 960mT can be identified in all samples (Table 1 and Table 2). In 205 general, maximum coercivities of component 3 are lower in palaeosol samples and most 206 weathered loess samples compared to less weathered loesses. Irrespective of the nature of the 207 samples it contributes around 13% to the total IRM (Table 1). A good linear correlation is 208 observed between the absolute intensity of component 1 and the viscous acquisition coefficient Sa and/or Sa2 (for two-slope curves) (Fig. 5). From the  $B_{1/2}$  versus relative 209 210 contribution graph in Figure 6 and from Table 2 it is clear that the higher the coercivity of 211 component 3, the smaller its relative contribution to the total IRM. Component unmixing for 212 the sample from the recent soil S<sub>0</sub> reveals quite different values for the unmix parameters 213 compared to the palaeosols and this is the reason to enter the data separately in Table 1.

214 Stepwise alternating field and thermal demagnetisations of IRM(2.6T) were applied to 215 several samples in order to gain information about the unblocking spectra of the remanence 216 carriers. The examples given in Figure 7 show a more or less similar behaviour for both loess 217 and palaeosol samples. Median destructive fields (MDF) of IRM(2.6T) are in the range of 218 15.8 - 16.9mT for palaeosols and in the range 17.5 - 21.2mT for loesses (Fig. 7a). This very 219 soft magnetic behaviour is accompanied by relatively high unblocking temperatures as can be 220 seen from the thermal demagnetisation curves (Fig. 7b). Half of the total IRM is 221 demagnetised between 270 and 300°C with a fairly continuous unblocking spectrum up to 222 680°C, above which complete demagnetization of the initial IRM is observed. The slightly 223 enhanced unblocking of IRM at about 300°C, especially in the palaeosol samples, is

accompanied by a strong increase at the same temperature in the room-temperature magneticsusceptibility measured after each heating-cooling step (Fig. 7c).

226 Anisotropy of magnetic susceptibility measurements on oriented one-inch cubes, cut 227 from the oriented block samples, reveal a normal sedimentary fabric with average maximum 228 (Kmax) and intermediate (Kint) susceptibility directions in the bedding plane (Fig. 8). The 229 scatter of principal directions is relatively high compared to other loess deposits (Matasova et 230 al., 2001; Hus, 2003; Lagroix and Banerjee, 2004; Zhu et al., 2004) but similar to the loess-231 palaeosol sequence of Roxolany in Ukraine (Hus, 2003). Only the principal susceptibility 232 directions of specimens from the least weathered loess unit L2 are much less scattered (Fig. 233 8b). Table 3 summarizes the average principal anisotropy parameters lineation (L), foliation 234 (F), corrected degree of anisotropy (P') and shape parameter T (Jelinek, 1981) for palaeosols, 235 loesses and red clay samples, respectively. No highly significant differences among the values 236 for the three groups can be observed. The low degree of anisotropy, less than 4%, is typical for loesses and palaeosols. A slightly higher degree of 7% is obtained for the red clays. 237

238 The best "cleaning" technique for the determination of the characteristic remanent 239 magnetisation (ChRM) was chosen after stepwise thermal as well as alternating field (AF) 240 demagnetisation test experiments on pilot samples. The AF demagnetisation with increasing 241 maximum amplitude up to 150 mT reveals the presence of a very soft remanence component 242 (Fig. 9). Values of the MDF of NRM (MDF<sub>NRM</sub>) are in the range 7.0 - 12.0mT for loess and 243 soil samples, consistent with the presence of a significant VRM in both. Moreover, there is a 244 negative relationship between  $MDF_{NRM}$  values and the percent relative contribution of the 245 first component from the IRM unmixing (Fig. 10). Figure 11 shows several examples of the 246 directional and intensity changes of loess and soil samples during stepwise thermal 247 demagnetisation. It is clear that besides a stable ChRM component; the samples contain an 248 important unstable component, probably of viscous origin. Cleaning temperatures of 300°C,

249 325°C and 350°C were finally chosen to isolate the ChRM of the whole collection of oriented 250 samples. The depth variation of the ChRM direction (declination D, inclination I) and 251 intensity along the composite section after partial demagnetization at 325°C is given in figure 252 12a. The declination is predominantly eastward and attains relatively high values in the upper 253 part of section 1 (S<sub>1</sub>, L<sub>2</sub> and upper part of S<sub>2</sub>), accompanied by relatively low inclination 254 values. One single sample in the upper part of S<sub>1</sub> reaches an anomalous intermediate 255 declination value. Below S<sub>2</sub>, the profile declination varies around zero, but remains 256 predominantly eastward down to loess unit  $L_7$  (Fig. 12a), where it becomes southward 257 accompanied by negative inclinations. This polarity change is interpreted as corresponding to 258 the Matuyama/Brunhes transition. Higher resolution of the directional changes in  $L_7$  was 259 obtained in a parallel section (Fig.12b). The switching of the inclination is observed within 260 the bottom part of loess unit L<sub>7</sub>, where the loess becomes yellowish with large brown reddish 261 veins suggesting strong leaching. Declination does not show any systematic behaviour and remains eastward when inclination becomes negative. Two reversed magnetozones can be 262 263 recognized in the red clay unit, although in a few samples no stable end-direction could be 264 reached after cleaning at 325°C. Rapid oscillations of declination within the upper part of the 265 red clays (between 22 and 24m depth) complicate the interpretation of the geomagnetic field 266 behaviour in this part of the record. The ChRM signal of a parallel section in the upper part of 267 section 1, sampled at 5 cm interval, did not reveal any indication of short term field reversals 268 or "excursions" similar to the Blake event (data not shown). Wiegank (1977) claimed to have 269 found the latter in unit L<sub>3</sub> of a loess/palaeosol section near Russe on terrace T<sub>6</sub> of the Danube 270 river.

271

#### 272 **5. Discussion**

273 5.1. Remanence carriers and magnetic stability

274 According to the results from previous detailed rock magnetic investigations, magnetite and 275 oxidised magnetites (maghemite) are the main remanence carriers in the palaeosol/loess units 276 of the Viatovo sequence. (Jordanova et al. 2007). Viscous acquisition experiments reported 277 this time contribute to a better understanding of their grain size distribution. The observed 278 two-slope VRM acquisition in samples from the upper units  $(S_0, L_1, S_1, L_2, S_2, L_3, S_3, L_4)$  and single-slope in the older units (Fig. 2), probably indicates different populations of viscous 279 280 grains in the upper part compared to the lower part of the sequence. This is consistent with the differentiation of palaeosol units according to the calculated background susceptibility for 281 282 each unit in Jordanova et al. (2007). Previous studies by Evlogiev (2006) indicate that the 283 recent soil  $S_0$  and the first three palaeosols are Chernozems, while the older palaeosols show 284 features of forest soils that developed in a more humid climate. Thus, it can be inferred that 285 the soil type (e.g. climate) is the main factor determining magnetic grain size distribution of 286 the pedogenically produced fine grained ferrimagnetic fraction. A possible explanation for the 287 single-slope behaviour is that in the older palaeosols a higher degree of grain surface low-288 temperature oxidation has led to a more uniform population of viscous maghemite grains. On 289 the other hand, in the dryer conditions of steppe vegetation, when Chernozems are formed, 290 grains are expected still to have a non-oxidised core with oxidised rim. Such grains, in which 291 the two phases are exchange coupled, are responsible for increased bulk coercivity (Van 292 Velzen and Zijderveld, 1995; Cui et al., 1994; Dunlop and Ozdemir, 1997). The good correlation between  $\chi_{FD}$ % and Sa coefficients, with R<sup>2</sup>= 0.91, suggests that the concentration 293 294 of fine viscous SP/SD grains is proportional to the degree of pedogenic enhancement both in 295 palaeosol and loess units (Fig. 3). The dominant role of the fine grained fraction in the 296 determination of magnetic viscosity of the loess/palaeosol samples is revealed by this linear 297 relationship. This implies also that the "in situ" formed pedogenic fraction has a broad grain size distribution, spanning the whole range from SP, viscous SP/SD to stable SD grains. 298

299 Analysis and modelling of IRM acquisition curves by a set of log-normal probability density 300 functions, applying an expectation – maximization algorithm and the IRMunmix program by 301 Heslop et al. (2002), confirms the above conclusion. The results suggest that the best fit of the 302 experimental data is obtained with a 3-component model (Fig. 4). Component 1 with  $B_{1/2}$  of 303 27 ( $\pm$ 3)mT on average and dispersion parameter DP of 0.24 ( $\pm$ 0.02) is identified in both loess 304 and palaeosol samples. It only varies in its relative contribution to the total IRM, with a 305 maximum contribution in the palaeosols and a minimum in the loess samples (Table 1). 306 Similar results have been reported by Spassov et al. (2003a) for the P-component in loess-307 palaeosol samples from the Lingtai section on the central Loess Plateau of China with  $B_{1/2}$  of 308 31 (±4)mT. The second component yields a  $B_{1/2}$  value of 35 (±8)mT or a slightly lower 309 coercivity compared to the second component of Eyre (1996). The aforementioned data, as 310 well as our data, are slightly higher than the reported  $B_{1/2}$  value of 20mT and DP of 0.25 to 311 0.38 by Geiss and Zanner (2006) for the pedogenic component in modern loessic soils from 312 mid-western United States. However, these authors analysed AF-demagnetisation curves of 313 IRM, which yield systematically lower estimations of the bulk coercivity, because the mean 314 interaction field in a sample is a demagnetising field that aids in AF demagnetisation but 315 opposes IRM acquisition (Dunlop and Ozdemir, 1997). On the other hand, we obtained a 316 good linear correlation between the estimated intensity of component 1 and the viscous 317 acquisition coefficients Sa (for single – slope samples) and Sa2 (for two-slope samples) (Fig. 318 5). It confirms our view that the population of grains above the critical SP/SD size, which 319 acquires an isothermal remanence, is a continuation of a common distribution of the 320 pedogenic fraction.

321 Component 2 is characterized by  $B_{1/2}$  of 70 (±12)mT and DP of 0.32 (±0.02) (Table 1). Its 322 relative contribution is highest in the loess samples with values up to 59% and most probably 323 corresponds to the presence of oxidised detrital magnetites.

324 Interpretation of the third component of the IRM unmixing should take into account the high 325 variability of the coercivity  $B_{1/2}$ , which is in close relation to the degree of secondary alteration (Table 2). Moreover, the negative linear relationship of  $B_{1/2}$  and the relative 326 327 contribution of component 3 in Figure 6 strongly suggest that this component represents 328 detrital SD-PSD grains with progressively higher degree of low-temperature oxidation of 329 magnetite to maghemite and, finally, hematite. But the observed very soft magnetic behaviour 330 of IRM against AF-demagnetisation (Fig. 7a) suggests that hematite does not contribute 331 significantly to the total remanence.

332 The enhanced unblocking of the isothermal remanence at around 300°C in Figure 7b can be 333 interpreted as being related to the inversion of maghemite (Evans and Heller, 1994). This 334 interpretation is supported by the obtained mean coercivity of the first component of the IRM 335 acquisition, corresponding quite well to the reported values for SD magnetites (Maher, 1988) 336 and maghemites (Enkin and Dunlop, 1987). At the same time, the strong increase in bulk 337 magnetic susceptibility measured after each heating step (Fig. 6c) is probably is due to 338 thermal transformation of the clay minerals and Fe-oxyhydroxides (lepidocrocite, ferrihydrite, 339 goethite) in the presence of reducing agents (organic matter) (Cornell and Schwertmann, 340 1996; Campbell et al., 1997) and partly caused by remanence unblocking. Thermal 341 demagnetisation of IRM(2.6T) (Fig. 7b) indicates also that there is still some remanence 342 demagnetised at temperatures well beyond 620°C. Similar parameters and thermal behaviour 343 have been observed for the detrital component D2 by Spassov et al. (2003a) and interpreted 344 by these authors as being carried by altered detrital magnetites.

345

346 5.2. Palaeomagnetism: ChRM and magnetostratigraphy

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348 The preservation of the original detrital fabric in weakly consolidated sediments, like loess 349 and palaesols, is some guarantee in magnetostratigraphical investigations that the directional 350 results are not entirely influenced by secondary alterations and mechanical disturbances. AMS 351 measurements of all the units of the Viatovo section (Fig. 8a) show that despite some high 352 scatter in the distribution of the principal susceptibility directions, a typical depositional 353 magnetic fabric is still preserved with Kmax on average in the horizontal (bedding) plane and 354 Kmin in the vertical plane. Shallower inclination values of Kmin for several samples are 355 obviously due to bioturbation and may be also to clay films (translocation) along vertical 356 cracks as it could be observed in the field. As shown in previous studies (Hus, 2003), "in situ" 357 formation of the pedogenic magnetic fraction does not influence very much the primary 358 magnetic fabric of loess sediments. The better grouping of the Kmin directions for samples 359 from unit L<sub>2</sub> (Fig. 8b), the loess unit least affected by secondary alterations and bioturbation, 360 confirms our assumption that mechanical disturbance due to bioturbation in the other units is 361 the main cause for the scatter in the principal directions. The low degree of anisotropy and 362 foliation (Table 3) indicate that the magnetic fabric, governed mainly by the ferrimagnetic 363 fraction because of relatively high mean susceptibility values, is quite isotropic.

364 Alternating field demagnetisation of NRM revealed very soft behaviour (Fig. 9) with less 365 than 20% of the original signal left after treatment in a 30mT field. This prevailing 366 contribution of the low-coercivity component (probably responsible for the VRM) led us to choose thermal treatment as the optimal technique for determining the characteristic 367 368 remanence direction. Furthermore, the observed correlation between MDF<sub>NRM</sub> and the relative 369 contribution of component 1 (identified by the IRM unmixing procedure in Figure 10 and 370 interpreted as pedogenic maghemite) convinced us that AF cleaning would be less successful 371 in isolating the characteristic remanence carried by the detrital magnetites (component 2) and 372 magnetites oxidised at different degree (component 3).

373 Stepwise thermal demagnetisation experiments revealed the presence of a secondary 374 component more or less in the present day magnetic field direction (Fig. 11), probably of 375 viscous or chemical origin. This component is demagnetised upon heating to  $200 - 250^{\circ}$ C, 376 which is also indicated by the change in slope of the NRM intensity versus temperature 377 curves. ChRM directions after partial demagnetisation at 300, 325 and 350°C were 378 determined by principal component analysis and show consistent behaviour (Fig. 12). Some 379 scattered directions and the two normal directions at the M/B polarity reversal interval may be 380 due some mechanical disturbance of the loess material and chemical overprinting after 381 deposition as suggested by the presence of large secondary carbonate concretions and clayey 382 reddish veins (see also Zhu et al., 2006). Another reason for the observed rapid directional 383 changes during polarity transitions may be the lock-in depth mechanism in loess-palaeosol 384 sediments and/or chemical overprinting as described in Spassov et al. (2003b). An earlier 385 palaeomagnetic investigation of the lower part of the loess complex near Russe, in the 386 Nikolovo section, located the M/B boundary between units S<sub>6</sub> and L<sub>7</sub> (Butchvarova, 1993). 387 The magnetisation of the red clay unit switches twice from reversed to normal polarity (Fig. 388 12). According to the results of the rock magnetic investigations (Jordanova et al, 2007), the 389 red clay unit may be considered as a thick pedocomplex that formed in subaerial conditions 390 with high weathering potential. Magnetic susceptibility variations suggest the presence of at 391 least two palaeosols (Fig. 12). The two periods of normal polarity correspond with an increase 392 in magnetic susceptibility in the two sub-units. The normal polarity magnetozone revealed in 393 the upper part of the red clay unit is assumed to correspond to the Jaramillo subchronozone (0.99 - 1.07 Ma) or spanning stages 27 to 31 of the  $\delta^{18}$  O chronology (Shackleton et al., 1990; 394 395 Tiedemann et al., 1994). The second normal polarity event is found in the middle of the red 396 clays. Because of its relatively long duration and the lower age estimate of the red clay 397 complex at 2.6 Ma (Evlogiev, 1993) it can be reasonably assumed that it corresponds to the

398 Olduvai subchron (1.78 - 1.94 Ma). Indeed, the short lived Cobb Mountain event identified in 399 deep-sea sediments (Biswas et al., 1999; Yang et al., 2004) is unlikely to have been recorded 400 or preserved in the record because of a low sedimentation rate, smoothing and post-401 depositional alterations. Indeed, this event is rarely found in loess/soil sequences (Rolph et al., 402 1989). The recently reported Kamikatsura and Santa Rosa short-lived geomagnetic excursions 403 in loess unit  $L_9$  of the Chinese loess (Yang et al., 2004) are probably absent in the Viatovo 404 section for the same reasons – the low sedimentation rate as well as the strong secondary 405 alteration of loess unit  $L_7$ . The absence of the Blake event in the Viatovo section is not 406 unusual, as discussed in other studies (Zhu et al., 1998; Stevens et al., 2006). Its presence 407 would have been of great importance for the Late-Pleistocene stratigraphy of the 408 loess/palaeosol sequence and for the identification of the last interglacial palaeosol. Indeed, in 409 many loess deposits in Western Europe (Hus and Geeraerts, 1986) the first palaeosol 410 encountered in depth is not an interglacial but an interstadial soil corresponding to stage 3 of 411 the oxygen isotope record (Shackleton et al., 1990). Therefore, the stratigraphy of the upper 412 part of the Viatovo sequence is deduced by correlation of its magnetic susceptibility signature with the astronomically tuned  $\delta^{18}$  O record from ODP site 677 (Fig. 13). According to the 413 414 previous established correlation between the magnetic susceptibility record of a core taken in 415 the loess/palaeosol sequence in Koriten in NE Bulgaria (Jordanova and Petersen, 1999), 416 palaeosol S<sub>1</sub> in Viatovo is the last interglacial (Eemian) soil (Fig. 13). Stage 3 is witnessed in 417 Viatovo by some high magnetic susceptibility values in the upper part of  $L_1$ , similar to the 418 Koriten record. With the two time constraints in the sequence, the Eemian soil and the 419 position of the M/B boundary, the one to one correspondence between magnetic susceptibility variations and the  $\delta^{18}$ O variations in the ODP 677 record strongly suggests that each palaeosol 420 421 that can be recognised in the Viatovo sequence corresponds to an interglacial.

422

#### 423 **6.** Conclusions:

424

425 1. Magnetic mineralogy (nature and magnetic domain state) of the loess-palaeosol units 426 from the Viatovo section deduced from unmixing IRM acquisition curves, VRM 427 acquisition experiments and thermal demagnetization of IRM(2.6T) indicate the 428 presence of pedogenic magnetite/maghemite particles spanning SP, viscous SP/SD 429 and stable SD sizes. Their contribution is especially enhanced in the palaeosols. 430 Detrital magnetic minerals are probably oxidised PSD magnetites, whereas the 431 contribution of hematite to the magnetic properties is insignificant. A three component 432 model fits best the experimental IRM acquisition curves. The third component shows 433 highly variable mean magnetic coercivity among the samples, reflecting the degree of 434 alteration of the original detrital magnetite grains with increasing oxidation.

The magnetic fabric of loess/palaeosol samples from the Viatovo section, revealed by
AMS measurements, points to an initial normal sedimentary fabric that has been
modified to some degree after deposition by pedogenic processes. Field evidence
shows that the scatter of the principal susceptibility directions can be mainly attributed
to mechanical disturbances due to bioturbation (presence of crotovinas) and eventually
clay migration along cracks.

- 3. The record of the Matuyama Brunhes polarity transition in loess unit L<sub>7</sub> displays
  several rapid directional changes of the characteristic remanence.
- 4. Notwithstanding strong pedogenesis, weathering and viscous overprints, a detailed
  magnetostratigraphy of the Viatovo section in North Bulgaria could be established.
  The Matuyama/Brunhes boundary has been recorded in the oldest loess unit L<sub>7</sub>
  present. Two normal polarity magnetozones, discovered in the underlying red clay
  formation, probably correspond to the Jaramillo and Olduvai subchronozones.

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450	Acknowledgements. This research benefited from a bi-lateral cooperation grant between the
451	Geophysical Institute of the Bulgarian Academy of Sciences and the Geophysical Centre of
452	the Royal Meteorological Institute of Belgium (project 1-12).
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- 584

585	Figure and Table caption	
586		
587	Fig. 1. a) Main geomorphological units in NE Bulgaria. Legend	: 1- Holocene, Upper- and
588	Middle Pleistocene river and sea terraces; 2- Lower Pleis	stocene river- and marine
589	terrace T6; 3- Old Abrasive Accumulation Level (OAAI	L); 4- Pliocene Denudational
590	Surface (PDS); b) View of the loess-palaeosol section in	n the eastern part of the kaolin
591	quarry in Viatovo.	
592	Fig. 2. VRM acquisition experiments for a) loess samples and b	) palaeosol samples. Time t is
593	in seconds.	
594	Fig. 3. Linear correlation between viscous acquisition coefficier	nts Sa1 (calculated from the
595	first slope in two-slope curves and from all points for sin	gle-slope curves) and percent
596	frequency dependent magnetic susceptibility $\chi$ fd%.	
597	Fig. 4. Gradient acquisition plot (GAP) for representative sample	les from loess and palaeosols
598	fit with a 3-components model using expectation – maxi	mization algorithm of Heslop
599	et al., 2002.	
600	Fig. 5. Correlation between viscous acquisition coefficients Sa a	and the intensity of component
601	1 obtained from the IRM unmixing procedure. In case of	f two-slope curves, the slope
602	of the second linear segment Sa2 is taken and plotted as	open diamonds. Full squares
603	denote Sa from single slope curves.	
604	Fig. 6. Dependence of the relative contribution of each component	ent with the corresponding
605	mean coercivity B1/2. Negative correlation is observed f	for component 3.
606	Fig. 7. Alternating field (a) and thermal (b) demagnetization of	IRM(2.6T) of loess and
607	palaeosol samples and the behaviour of room-temperature	re bulk magnetic
608	susceptibility, measured after each heating step (c).	

609	Fig. 8. Stereographic projections (on the lower hemisphere) of the maximum (Kmax) and
610	minimum (Kmin) susceptibility axes for loess and palaeosol samples from different
611	units (a) and b) principal susceptibility axes (Kmax, Kint, Kmin) for samples from
612	second loess unit L <sub>2</sub> only.
613	Fig. 9. NRM intensity changes during stepwise AF-demagnetization of loess and palaeosol
614	samples.
615	Fig. 10. Dependence between the $MDF_{NRM}$ and the relative contribution of component 1,
616	obtained after unmixing IRM acquisition curves for the same specimens.
617	Fig. 11. Thermal demagnetization behaviour (direction and intensity) of representative
618	samples with normal and reversed ChRM directions.
619	Fig. 12. a) Depth variation of bulk magnetic susceptibility, declination, inclination and
620	intensity of the characteristic remanence ChRM (after thermal cleaning at 325°C) in
621	the Viatovo section ; b) Declination, inclination and ChRM along unit $L_7$ in a parallel
622	section, sampled in more detail.
623	Fig. 13. Correlation of the magnetic susceptibility signatures of the loess/palaeosol sequences
624	in Koriten (Jordanova and Petersen, 1999) and Viatovo with the astronomically tuned
625	$\delta^{18}$ O changes in ODP 677.
626	Table 1. Mean values of parameters of the three components (log-normal Gaussian functions)
627	representing the best fit to experimental IRM acquisition curves for recent soil So,
628	loess units and palaeosols separately. Relative contribution of each component is given
629	in percent; B1/2 is the coercivity at which the gradient acquisition curve (GAP) is
630	centred; dispersion parameter DP is the width at half intensity of the GAP and
631	represents one standard deviation.
632	Table 2. Parameters of the third component obtained after fitting experimental IRM
633	acquisition curves for different samples.

- Table 3. Mean values of principal anisotropy of magnetic susceptibility parameters for
- 635 loesses, palaeosols and the red clay unit. N is number of samples; standard deviations
- 636 of the mean values are given between brackets.
- 637
- 638







Figure 2







Figure 4







Figure 6



Figure 7



Figure 8

AF demagnetization of NRM 4.5E-05 J 4.0E-05 - A01E020S02-S0 △ · · · A01E120S02-L1 3.5E-05 - A01E300S01-S1 -A01E490S02-L2 3.0E-05 - A01E780S02-S2 - \* AO2E060S02-L3 M (Am2/kg) 2.5E-05 - A02E130S02-S3 - A02E240S02-L4 2.0E-05 - A02E280S02-S4 o--- A02E500S02-L5 1.5E-05 -- A02E660S02-S5 - A02E790S01-L6 . . 1.0E-05 - A03E090S02-S6 -A03E370S02-L7 5.0E-06 • A04E250S02-RC 0.0E+00 -6 20 30 50 60 100 110 120 130 140 150 0 10 40 70 80 90 AF (mT) Figure 9 80 70 • So comp.1 contribution (%) 60 50 40 30 20 10 0 7 9 11 13 5 MDF (NRM) m T

Figure 10

Figure 11



3 of 39









#### **MANUSCRIP** ACCEPTED

Component 1				Comp	Component 2			Component 3		
		Rel.	B <sub>1/2</sub>	DP	Rel.	B <sub>1/2</sub>	DP	Rel.	B <sub>1/2</sub>	DP
		contr.	(mT)		contr.	(mT)		contr.	(mT)	
		(%)			(%)			(%)		
So		67	34	0.28	26	83	0.29	7	582	0.53
loe	esses	33±7	27± 2	0.24±0.	02 $53 \pm 6$	75±11	$0.3 \pm 0.02$	13±3	332±271	$0.83 \pm 0.13$
pa	laeosols	51±8	26± 1	0.23±0.	01 34.5±	8 57± 9	0.32 ±0.01	13±4	224±190	$0.73 \pm 0.13$
Al	l		27±3	0.24±.0	)2	$70 \pm 12$	$0.32 \pm 0.02$		332±225	0.75±0.14
	Table 1									
	sample	unit	compo	onent 3						
			contrib	ution (%)	B1/2 (mT)	DP(mT)				
	viaA01E120	) L1	1	6	179.6	0.97				
	1 = 101 - 100		4	0	075	0 70				

م م

sample	unit	component 3		
		contribution (%)	B1/2 (mT)	DP(mT)
viaA01E120	L1	16	179.6	0.97
viaA01E490	L2	12	275	0.72
viaA02E060	L3	16	349.4	0.97
viaA02E240	L4	17	176.2	0.83
viaA02E500	L5	13	332	0.83
viaA02E790	L6	10	402.3	0.63
viaA03E370	L7	12	966.3	0.75
viaA01E300	S1	13	198.8	0.83
viaA01E780	S2	14	163.9	0.73
viaA02E130	S3	16	149.1	0.88
viaA02E280	S4	10	513.9	0.62
viaA02E660	S5	22	224.4	0.81
viaA03E090	S6	10	590.6	0.72
viaA04E250	RC	10	489.6	0.49
viaA01E020	So	7	581.7	0.53

Table 2

unit	Ν	Kmean (x10 <sup>-6</sup> SI)	L	F	P	Т
palaeosols	21	1491 (377.7)	1.001 (0.001)	1.002 (0.002)	1.004 (0.002)	0.396 (0.463)
loesses	10	555.9 (143)	1.001 (0.001)	1.003 (0.003)	1.004 (0.004)	0.27 (0.486)
Red clay	8	523.7 (337.9)	1.003 (0.002)	1.004 (0.002)	1.007 (0.003)	0.284 (0.297)

### Table 3