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Luminescence chronology of the northeastern Bulgarian loess-paleosol sequences (Viatovo and Kaolinovo)

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Abstract

The key reference loess sequence of Viatovo in NE Bulgaria, lying within the lower Danube basin, records climatic and environmental changes over the last 800 ka. This 20 m thick loess sequence consists of seven loess units (L1 to L7) intercalated by six paleosols (S1 to S6) below the modern soil (S0). The Matuyama-Brunhes palaeomagnetic boundary has been identified in the lowest loess unit (L7). This reference loess sequence remains so far undated by radiometric techniques. Its chronostratigraphy relies only on pedostratigraphic and magnetostratigraphic evidence. This contrasts with the adjacent reference loess sequences of SE Romania (Tuzla, Mostistea, Mircea Vodă) whose luminescence chronology is now securely established up to MIS 8.

The aim of this study is to validate the chronostratigraphic framework of the Viatovo loess sequence using the luminescence dating method. For this purpose, the luminescence method is applied to the three upper loess units (L1, L2, L3) from two adjacent sites: the reference site of Viatovo and the nearby site of Kaolinovo, both located in the Ludogorie region (NE Bulgaria) and showing the same superposition of loess (L1 to L7) and interbedded paleosols. Luminescence dating (Infrared stimulated luminescence; IRSL) was carried out using the feldspar fine sand-size grain fraction (60-80 µm), applying the IRSL₅₀ and the pIRIR₂₉₀ dating techniques. For the equivalent dose (De) determination we used both the SAR post-infrared infrared (pIRIR₂₉₀) protocol on single aliquots and the multiple-aliquot additive dose (MAAD IRSL₅₀) protocol. The pIRIR₂₉₀ ages and the fading corrected MAAD-IRSL₅₀ ages are self-consistent. It demonstrates for the first time, that the three upper loess layers (L1, L2, L3) at Viatovo and Kaolinovo were deposited during the marine isotope stages (MIS) 2-4, 6 and 8, thus constraining the youngest well-developed paleosol (S1, a chernozem) to the Last Interglacial. These IRSL₅₀ and pIRIR₂₉₀ ages are in good agreement with the luminescence ages obtained for (1) the two upper loess layers (L1, L2) from the Harletz sequence in NW Bulgaria and (2) the three upper loess units (L1, L2, L3) from the adjacent SE Romanian loess sequences.

The luminescence chronology of the loess sequences is presented along with the pedogenic and magnetic property data. The pedostratigraphic records of the NE Bulgarian and SE Romanian loess sequences show the same palaeoclimatic trends, with progressive aridification during interglacial periods (from S6 to S0).

49 Key words: Loess, Pleistocene, IRSL dating, K-feldspars, Bulgaria, Romania
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1. Introduction

The loess-paleosol sequences from NE Bulgaria and SE Romania, both lying within the lower Danube basin (Fig. 1A) are among the thickest (Haase et al., 2007) and most complete southeastern European terrestrial palaeoclimatic archives. These 20 to 30 m thick loess-paleosol sequences record climatic and environmental changes over the last 800 ka. The Matuyama-Brunhes palaeomagnetic boundary (MBB) has been identified in the lowest loess unit both in NE Bulgaria, at Viatovo (Jordanova et al., 2008) and in SE Romania, at Tuzla (Balescu et al., 2003) and Zimnicea (Radan, 2012) (Fig. 1B). These loess deposits have an aeolian origin (Smalley and Leach, 1978; Evlogiev, 2007; Smalley et al., 2009) and their source material is mostly derived from the alluvium of the lower Danube valley, with additional dust transported from the outwash of the northern Fennoscandian ice sheet (Evlogiev, 2007; Buggle et al., 2008; Fitzsimmons et al., 2012; Kis et al., 2012; Jipa, 2014) and minor components from the Sahara (Stuut et al., 2009).

The reference loess sequences of Viatovo and Koriten in NE Bulgaria (Fig. 1B), well known for their most complete paleomagnetic records (Jordanova and Petersen, 1999a,b; Jordanova et al., 2008), consist of seven loess layers (named L1 to L7 from top to bottom, following the stratigraphic nomenclature adopted by Jordanova et al. (1999a,b) for the Bulgarian loess) and six intercalated paleosols (named S1 to S6) below the recent (Holocene) soil (S0) (Fig. 2). However, these remain so far undated by radiometric techniques. Their chronostratigraphy relies only on geomorphic, pedostratigraphic and magnetostratigraphic evidence (Jordanova and Petersen, 1999a,b; Jordanova et al., 2008). This contrasts with the adjacent SE Romanian reference loess sequences of Tuzla, Mostiștea and Mircea Vodă (Fig. 1B) whose chronology is now securely established by luminescence dating up to MIS 8 (Balescu et al., 2003, 2010; Timar et al., 2010, Timar-Gabor et al., 2011; Vasiliniuc et al., 2011, 2012, 2013).

The aim of the present study is to provide the first chronological framework for the Viatovo reference loess-paleosol sequence using the luminescence dating method in order to validate the magnetostratigraphy-based age model established by Jordanova and Petersen (1999a,b) and Jordanova et al. (2008) at Viatovo and Koriten.

For this purpose, the luminescence dating method (Infrared stimulated luminescence; IRSL) is applied herein to the three upper loess layers (L1, L2, L3) from the Viatovo loess-paleosol sequence (Jordanova et al., 2008; Kis et al., 2012) and the nearby long loess sequence of Kaolinovo (Fig. 2) which shows the same superposition of loess (L1 to L7) and interbedded paleosols. It could not be tested on the reference loess sequence of Koriten (Jordanova and Petersen (1999a,b) which is unfortunately no longer accessible.

90 Since the quartz OSL (*optically stimulated luminescence*) dating method applied to the
91 NW Bulgarian and SE Romanian loess is limited to the last 70-100 ka (Timar et al., 2010;
92 Timar-Gabor et al., 2011; Lomax et al., 2018), we focus herein on K-feldspars whose
93 luminescence signals saturate at much higher doses and thus have the potential to date much
94 older sediments. In the present study, we apply both the post-infrared infrared (pIRIR₂₉₀)
95 (Thomsen et al., 2008; Buylaert et al., 2012) and the IRSL₅₀ dating techniques to K-feldspar
96 fine sand-size (60-80 µm) grains from the three upper loess layers (L1, L2, L3). These ages will
97 be further compared with the luminescence ages obtained for (1) the two upper loess layers
98 (L1, L2) from the Harletz sequence (Fig. 1B) in NW Bulgaria (Lomax et al., 2018) and (2) the
99 three upper loess units (L1, L2, L3) from the adjacent SE Romanian loess sequences in order to
100 better ascertain the regional stratigraphic correlations of the Pleistocene loess deposits across
101 the lower Danube basin.

102 In this study, the luminescence chronologies of the NE Bulgarian and SE Romanian loess
103 sequences are presented along with the pedostratigraphic and magnetic susceptibility data
104 which provide important information on the palaeoclimatic evolution of the lower Danube
105 basin during the Pleistocene.

106 2. Study sites and sampling

107
108 The Viatovo and Kaolinovo loess sequences are located at the southern fringe of the Lower
109 Danube Basin (Fig. 1B). They are situated between the Danube and the Balkan Mountains, in
110 the Ludogorie region (Fig. 1B), which is characterized by a hilly relief with deeply incised river
111 valleys (Zagorchev, 2009). The average elevation of the region is 200-300 m asl. This region is
112 lying next to the Dobrogea plateau (Fig 1B) which extends from NE Bulgaria to SE Romania,
113 along the Black Sea coast, and culminates at 300 m asl. The reference loess sequence of
114 Koriten (Jordanova and Petersen, 1999a,b) located in this area is no longer accessible.

115
116 The Ludogorie and Dobrogea regions, both preserve some of the thickest and most complete
117 loess sequences of the lower Danube Basin. The Bulgarian loess deposits are the thickest (30-
118 40 m) close to the Danube and are thinning southwards, toward the Fore Balkan (Evlogiev,
119 2007). The grain-size of the loess is also gradually decreasing from the Danube alluvial plain
120 toward the Fore Balkan (Evlogiev, 2007).

121 The Viatovo and Kaolinovo loess-paleosol sequences are exposed on the walls of active
122 quarries where Albian - Early Eocene sandy kaolinitic deposits are being extracted (Tonov et
123 al., 2016). The sedimentary complex lying on top of these deposits is represented by a 6-8 m
124 thick Pliocene red clay formation (terra rossa) overlain by a loess-paleosol sequence of a 20 m

125 thickness, consisting of seven loess layers (L1-L7) and six interbedded paleosols (S1-S6)
126 (Fig. 2). The paleosols and loess layers recognized in the field are numbered and designated S
127 and L, respectively, according to the original stratigraphic nomenclature adopted by [Jordanova](#)
128 [and Petersen \(1999a,b\)](#) for the Bulgarian loess and later extended to the Romanian loess by
129 [Panaiotu et al. \(2001\)](#). This nomenclature complies with the unified Danube loess stratigraphic
130 model recently proposed by [Marković et al. \(2015\)](#).

131 The chronostratigraphy of the Viatovo and Kaolinovo loess sequences relies on geomorphic,
132 pedostratigraphic and magnetostratigraphic evidence. A chronostratigraphic framework has
133 been established on the basis of the detailed magnetic susceptibility (MS) records of the
134 Koriten and Viatovo loess-paleosol sequences ([Jordanova and Petersen, 1999a,b](#); [Jordanova et](#)
135 [al., 2007, 2008](#)). The MS signatures of these loess-paleosol sequences have been correlated
136 with the astronomically tuned ODP677 $\delta^{18}\text{O}$ record of [Shackleton et al. \(1990\)](#). The age of the
137 oldest loess deposit (L7) is constrained palaeomagnetically at Viatovo by the identification of
138 the Brunhes-Matuyama palaeomagnetic boundary (MBB: 772 ± 7 ka according to [Sugunama et](#)
139 [al., 2015](#); following the nomenclature system adopted by [Fitzsimmons et al., 2012](#) and
140 [Marković et al., 2015](#)) thus yielding a maximum age limit for these loess deposits ([Jordanova et](#)
141 [al., 2008](#)).

142 The **Viatovo** loess section (Fig. 2, Fig. S1) ($43^{\circ} 41' 18.6''$ N; $26^{\circ} 14' 48.6''$ E; 219 m
143 a.s.l) is a key reference site for the Bulgarian loess, being well-known for its most complete
144 palaeomagnetic record. The mineralogy, grain-size and magnetic mineral characteristics of this
145 loess-paleosol sequence were previously investigated ([Jordanova et al., 2007, 2008](#); [Kis et al.,](#)
146 [2012](#)). It has a thickness of 21 m. A short description of each loess unit is presented in [Table](#)
147 [S1](#). The recent (Holocene) soil (S0) is a dark gray soil (B-horizon). The first three paleosols
148 (S1-S3) are dark brown chernozem-like (steppe) soils associated with dry climatic conditions
149 ([Fotakieva, 1974](#); [Minkov, 1968](#); [Eckmeier et al., 2007](#)). The older paleosols (S4-S6) are red,
150 rubified, clay-rich forest soils that developed in a warmer and more humid climate ([Jordanova](#)
151 [et al., 2008](#)).

152 MBB has been identified at Viatovo, in loess L7 (Fig. 2). Moreover, two normal polarity
153 magnetozones have been discovered in the underlying red clay formation corresponding to the
154 Jaramillo and Olduvai subchronozons ([Jordanova et al., 2008](#)).

155 The **Kaolinovo** section (Fig. 2, Fig. S2) ($43^{\circ} 35' 52''$ N; $27^{\circ} 09' 26.7''$ E; 253 m asl) has a
156 total thickness of 20.3 m. A short description of each loess unit is presented in [Table S2](#). The
157 recent soil S0 is represented by a moderately leached Chernozem. The first three paleosols (S1-
158 S3) are dark brown chernozem-like (steppe) soils (Fig. S2). The older paleosols (S4-S6) are
159 reddish soils.

160 The stratigraphy and the IRSL sampling positions are shown in [Figure 2](#). The present
161 study focuses on seven IRSL samples collected within the three uppermost loess layers (L1, L2,
162 L3) both at Viatovo (VIA 1,2,3) and Kaolinovo (KA 1, 2, 3) and, in the oldest loess (L7) at
163 Viatovo, below the MBB (VIA 7). The IRSL technique is applied here to one sample per
164 stratigraphic unit, as previously performed by [Balescu et al. \(2003, 2010\)](#), [Timar et al. \(2010\)](#),
165 [Timar-Gabor et al. \(2011\)](#) and [Vasilinuc et al. \(2012, 2013\)](#) when testing their luminescence
166 dating techniques on the Middle Pleistocene Romanian loess (L2, L3). This sampling strategy
167 aims at obtaining the mean depositional age of each loess layer to validate the MS age model of
168 [Jordanova et al. \(2008\)](#).

169 **3. Methods**

170 *3.1. Sample preparation, equivalent dose (D_e) determination and fading corrections*

171
172
173
174 All dated samples were collected from homogeneous, unweathered and unworked loess. In
175 addition to field observations, sample positions within each loess horizon have been chosen on
176 the basis of the field MS variations, measured by field kappa meter KT-5 (SatisGeo s.r.o.,
177 Czech Republic).

178 The IRSL samples were wet sieved and subsequently treated with HCl (10%) to remove
179 calcium carbonate.

180 All IRSL measurements were performed on K-feldspar fine sand-size grains (60-80 μm). K-
181 feldspars (density < 2.58 g/cm^3 .) were separated using a heavy solution of sodium
182 polytungstate. The mineral grains were dispensed on aluminium disks or aluminium cups.

183 For the D_e determinations, two protocols were used ([Table S3](#)): the SAR pIRIR₂₉₀ technique on
184 single aliquots (monolayer of grains on disk; sample diameter of 4 mm) and the multiple-
185 aliquot additive- γ dose (MAAD IRSL₅₀) protocol (20 mg of grains in cups, 20 aliquots).

186 In the SAR pIRIR₂₉₀ protocol, single aliquots are exposed to increasing regenerated dose.
187 Interpolation of the natural signal on the dose response curve yields the D_e ([Fig. 3](#)). In the
188 standard MAAD protocol ([Aitken, 1998; Singhvi et al., 1982](#)), a series of increasing doses are
189 added to the natural sample, and the resultant dose response is extrapolated back to a negligible
190 light level to obtain the D_e ([Fig. 4](#)). The advantage of this protocol over SAR pIRIR is that
191 sensitivity changes are kept to a minimum and sensitivity corrections are therefore not usually
192 used.

193 This technique has recently been successfully re-explored in the context of the violet stimulated
194 luminescence signal of quartz from the Chinese loess ([Ankjaegaard et al., 2016](#)).

195 The MAAD protocol remains valid for well-bleached sediments such as loess whose wind-
196 blown nature ensures that their luminescence clock was completely reset prior to deposition.
197 Moreover, it should be noted that the MAAD protocol has previously been shown to provide a
198 good chronological discrimination between the MIS 8, MIS 6 and MIS 2-4 loess deposits of
199 SE Romania (Balescu et al., 2003, 2010) thereby providing a reliable and very useful
200 chronostratigraphic marker. In the present study, the MAAD approach applied in conjunction
201 with the pIRIR₂₉₀ technique, is aimed at obtaining an additional age control. It also allows
202 direct comparison with the MAAD IRSL₅₀ ages previously obtained for the three upper loess
203 layers (L1-L3) in SE Romania (at Tuzla, Mircea Vodă and Mostistea) (Balescu et al., 2003,
204 2010).

205 The pIRIR₂₉₀ and MAAD IRSL₅₀ measurements were performed respectively on a Lexsyg
206 Smart reader and an automated Daybreak 1100TL reader. Aliquots measured on the Lexsyg
207 Smart reader were irradiated with the internal ⁹⁰Sr /⁹⁰Y beta source (0.13 Gy/s). Those
208 measured on the Daybreak systems were irradiated with an external ¹³⁷Cs gamma source
209 (0.03 Gy/s). The luminescence of K-feldspars was detected with a 410 nm detection window
210 (combination of Schott BG39 glass and Semrock 414/46 Brightline HC interference filters) on
211 the Lexsyg Smart reader and through a blue-violet filter combination (Corning 7-59 and
212 Schott BG39) on the Daybreak systems. The growth curves of the K-feldspar samples were
213 fitted with a single saturating exponential function. The signal used for analysis was obtained
214 from the net initial signal (5 s), subtracted by the background signal taken at the end of the
215 IRSL decay (last 20 s). The Central Age Model (Galbraith et al., 1999) was used for the mean
216 D_e calculation.

217
218 All samples behave well in the pIRIR₂₉₀ protocol (in terms of recycling, recuperation and dose
219 recovery). Recuperation level was very low, being on average close to 0.47 %, and all less than
220 1 %. The recycling ratios of all samples range from 0.98 to 1.01, for an average of 1.00 ± 0.01.
221 The recycled dose point (last versus first) was chosen to be half of the equivalent dose. A dose
222 recovery test was conducted on K-feldspars from sample KA 2. These were bleached 1h with
223 an external solar simulator (Honlë SOL2) and given a dose of 500 Gy. We measured a
224 recovered to given dose ratio of 1.05 ± 0.03 (n=3).

225 In our pIRIR₂₉₀ protocol, we used a test dose of 133 Gy. Colarossi et al. (2018) have recently
226 shown that the magnitude of the luminescence response (Tx) to the test dose is dependent upon
227 the size of the luminescence signal (Lx) arising from the regeneration dose when using the
228 pIRIR₂₂₅ protocol on single grain (IRSL at 225°C for 2 s). They have demonstrated that when
229 the test dose is small (1-15% of the given dose) the carry-over of charge dominates the signal

230 arising from the test dose, whereas for larger test doses this impact is much lower. It should be
231 stressed however that in the present study, our pIRIR₂₉₀ protocol has been validated by a
232 recovery dose test. Also, we used a large test dose, between 15-80% of the De, as
233 recommended by Yi et al. (2016) who have shown that in this dose range, large given doses
234 were successfully recovered. But since we are using a higher stimulation temperature (290°C)
235 and a longer stimulation time (200 sec), our results cannot be directly compared with those of
236 Colarossi et al. (2018). Nevertheless, future application of the modified protocol of Colarossi et
237 al. (2018) designed to minimize the impact of the regeneration dose upon the measurement of
238 the test dose, might be useful to corroborate our results.

239 The K-feldspar IRSL₅₀ signal being affected by anomalous fading (i.e. an unexpected loss of
240 signal through time leading to an age underestimation), all our measured MAAD IRSL₅₀ ages
241 have been corrected for fading using the protocol of Mejdahl (1988, 1989) as previously
242 described in Balescu et al. (2003, 2010) (see Supplementary information). It relies on the IRSL
243 analysis of “infinitely old” K-feldspars, here collected from L7 at Viatovo (VIA 7), below the
244 MBB. All MAAD measurements have been performed on K-feldspar samples that have been
245 stored 1 year at room temperature after irradiation in order to reduce the effect of anomalous
246 fading (Spooner, 1992).

247 By contrast, the pIRIR₂₉₀ signal of the Bulgarian loess shows extremely low fading rates
248 suggesting in this case, that this is a stable, non-fading signal. Anomalous fading rates (*g*-value)
249 were measured using the SAR protocol of Auclair et al. (2003). Fading measurements were
250 carried out on about 12 aliquots per sample (Table S4). All loess layers show similar and
251 extremely low fading rates (*g*-values); the mean *g*-value ranging between -0.04 and
252 0.18 %/decade. Consequently, in the present study, the pIRIR₂₉₀ ages have not been corrected
253 for fading. It should be stressed that similar low and negative fading rates were also observed in
254 the NW Bulgarian loess of Harletz (Lomax et al., 2018) and in the Romanian loess (Vasiliniuc
255 et al., 2012) and have been assumed by these authors to be a laboratory artefact.

256 257 3.2. Dose rate determination

258
259 The concentrations of U, Th and K were measured through gamma spectrometry. For the
260 conversion to dose rate we used the absorbed beta dose coefficient given in Nathan (2010)
261 while we relied on Brennan et al. (1991) for the alpha absorption coefficient. The past water
262 content was estimated at $20 \pm 5\%$ (the average of the present-day natural water content and the
263 laboratory saturated water content). The cosmic dose rates were estimated using the present-
264 day burial depth of the sample (Prescott and Hutton, 1994). The dose rates were calculated
265 using the DRAC (Durcan et al., 2015). As suggested by Huntley and Baril (1997) we assumed

266 an internal K content of $12.5 \pm 0.5\%$ for the blue IRSL emission of the K-feldspars. The
 267 summary of the dose rates is reported in [Table S5](#).

269 4. Results and discussion

270 4.1. Dose rates and luminescence age estimates of the NE Bulgarian loess

271 All loess layers display similar dose rates, ranging between 2.94 and 3.43 Gy/ka ([Table](#)
 272 [S6](#)), except in the case of sample KA 1 collected within the loess L1 at Kaolinovo. This sample
 273 KA 1 with a much higher dose rate of 5.13 Gy/ka due to higher radionuclide concentrations,
 274 yielded a pIRIR₂₉₀ age of 45 ± 2 ka ([Table 1](#)). This higher dose rate could possibly be related to
 275 the presence of a tephra layer equivalent to the Campanian Ignimbrite/Y5 (39.28 ± 0.11 ka)
 276 reported at Rasova-Valea cu Pietre in SE Romania ([Fig. 1B](#)). [Anechitei-Deacu et al. \(2013\)](#)
 277 have shown that loess samples collected at that site, in L1 around this ash layer exhibited a
 278 similar radionuclide enrichment. This tephra layer has also been identified within L1 at
 279 Caciulatesti (Southern Romania; [Constantin et al., 2012](#)) and at Urluia (Dobrogea; [Fitzsimmons](#)
 280 [and Hambach, 2014](#)) ([Fig. 1B](#)). This is clearly a matter that requires further investigation.

281 Comparative dose response curves of the pIRIR₂₉₀ signal are shown in [Figure 3](#).
 282 Representative IRSL₅₀ additive growth curves are presented in [Figure 4](#). The luminescence
 283 results for the NE Bulgarian loess are reported in [Table 1](#). The pIRIR₂₉₀ ages and the fading
 284 corrected MAAD-IRSL₅₀ ages are self-consistent within the error limits, and both show
 285 stratigraphic consistency.

286 - For L1, these luminescence ages range between 41 and 69 ka and for L2 between 153
 287 and 213 ka. These results demonstrate for the first time that the two uppermost loess
 288 units, L1 and L2, at Viatovo and Kaolinovo, were deposited respectively during
 289 MIS 2-4 and MIS 6, thereby confirming the expected burial age previously inferred
 290 from the magnetostratigraphy age model established by [Jordanova et al. \(2008\)](#).

291 - For L3, the pIRIR₂₉₀ ages (293 ± 17 and 296 ± 17 ka) suggest a MIS 8 age. They
 292 should be considered as minimum ages since the natural ratios (L_n/T_n) of L3 are close
 293 to the laboratory saturation level of the growth curve ([Fig. 3](#)). Their measured D_e
 294 values are systematically higher than $2D_0$ ([Table 1](#)). The correlation of L3 with MIS 8
 295 is also supported by the corrected MAAD-IRSL₅₀ ages of L3 (285 ± 35 ka;
 296 264 ± 64 ka) whose corrected D_e values are lower than $2D_0$ ([Table 1](#)).

297 - The natural pIRIR₂₉₀ signal of the infinitely old sample VIA 7 (L7) is in saturation as
 298 expected for a non-fading signal ([Fig. 3](#)). Whereas the natural MAAD IRSL₅₀ signal of
 299 this infinitely old sample VIA 7 (L7) is not in saturation ([Fig. 4](#)) due to fading in the
 300
 301

environment (Mejdahl, 1988). It yields an apparent IRSL age of 296 ± 60 ka in good agreement with the apparent IRSL age of L7 at Tuzla in SE Romania (271 ± 39 ka; Balescu et al., 2003). The measured D_e value of VIA 7 (L7) is lower than $2D_0$ (Table 1). It is worth pointing out that infinitely old loess, lying below the MB boundary, in Romania, Bulgaria, Serbia and Hungary yield similar apparent ages, suggesting an upper age limit of around 250-300 ka, when using either the IRSL₅₀ or the pIRIR protocols: 271 ± 39 ka (IRSL₅₀) at Tuzla in Romania (Balescu et al., 2003), 296 ± 60 ka (IRSL₅₀) and >250 ka (pIRIR₂₉₀) at Viatovo in Bulgaria (this study), 239 ± 38 ka (IRSL₅₀) and >260 ka (pIRIR₂₉₀) at Stari Slankamen in Serbia (Murray et al., 2014), 316 ± 36 ka (IRSL₅₀) at Paks in Hungary (Frechen et al., 1997).

These luminescence ages may further be compared with those recently obtained for the Harletz loess sequence in NW Bulgaria (Fig. 1B). This sequence consists of three loess layers separated by two palaeosol complex (Lomax et al., 2018). The oldest loess unit rests on alluvial deposits of the Ogosta river (tributary of the Danube). Luminescence dating was carried out using the quartz fine grain fraction and a SAR protocol, and the feldspar coarser grain fraction (63-125 μ m), applying the MET-pIRIR protocol. The OSL and MET-pIRIR ages of the youngest loess unit (L1: 36-67 ka) are in good agreement with those obtained for L1 at Viatovo and Kaolinovo (Table S6). The underlying loess unit (L2) yielded underestimated OSL ages (84-110 ka). Whereas the MET-pIRIR ages of L2, ranging between 131 and 185 ka are in good accordance with those of L2 at Viatovo and Kaolinovo (Table S6). By contrast, the oldest loess unit, resting on alluvial sediments and overlain by an interglacial paleosol supposedly assigned to MIS 7, yielded an underestimated MET-pIRIR age of 131 ± 10 ka.

4.2. Comparison of the NE Bulgarian and SE Romanian loess-paleosol sequences

The NE Bulgarian and SE Romanian loess deposits belong to the same lower Danube loess area. They exhibit common characteristics and cover the same age range. In both regions, the MBB has been identified in the oldest loess L7 (Jordanova et al., 2008; Balescu et al., 2003). Moreover, the luminescence chronology of the three upper loess units (L1-L3) in SE Romania is now securely established thanks to the application of multiple dating protocols. Hence, it is worth comparing here their luminescence chronologies, along with their pedostratigraphic and magnetostratigraphic records.

337 4.2.1. Luminescence chronologies

338
339 So far, four different luminescence dating techniques have been applied to the SE Romanian
340 loess: the MAAD-IRSL₅₀ on K-feldspar silt-size grains (Balescu et al., 2003, 2010), the OSL on
341 fine and coarse quartz grains (Timar et al., 2010; Timar-Gabor et al., 2011; Vasiliniuc et al.,
342 2011; Constantin et al., 2012, 2014, 2015; Anechitei-Deacu et al., 2013; Timar-Gabor and
343 Wintle, 2013; Fitzsimmons and Hambach, 2014) and more recently, the SAR IRSL₅₀
344 (Vasiliniuc et al., 2013) and the pIRIR₂₂₅ on polymineral fine grains (Vasiliniuc et al., 2012).
345 These dating techniques have been tested on several long loess sequences showing a similar
346 superposition of loess and interbedded paleosols: at Tuzla (Balescu et al., 2003, 2010),
347 Costinești (Timar-Gabor and Wintle, 2013; Constantin et al., 2014), Mircea Vodă (Balescu et
348 al., 2010; Timar et al., 2010; Timar-Gabor et al., 2011; Vasiliniuc et al., 2012), Urluia
349 (Fitzsimmons et al., 2013; Fitzsimmons and Hambach, 2014), Mostiștea (Balescu et al., 2010;
350 Vasiliniuc et al., 2011) and Lunca (Constantin et al., 2015) (Fig. 1B). It is also worth pointing
351 out that the accuracy of the quartz OSL dating technique has been demonstrated on loess
352 samples bracketing a well-dated volcanic tephra layer (Campanian Ignimbrite/Y5:
353 39.28 ± 0.11 ka) within the Last Glacial (L1) loess, at Urluia (Fitzsimmons and Hambach,
354 2014), Caciulatești (Constantin et al., 2012), and Rasova-Valea cu Pietre (Anechitei-Deacu et
355 al., 2013; Zeeden et al., 2018) (Fig. 1B).

356 For the sake of comparison with the NE Bulgarian loess, we will focus here on the three most
357 investigated and better documented long loess sections from SE Romania (Tuzla, Mircea Vodă
358 and Mostiștea) where both the Upper and Middle Pleistocene loess deposits (L1, L2, L3), lying
359 in stratigraphic continuity, have previously been dated using multiple luminescence dating
360 protocols.

361 The Tuzla section is a cliff-exposure along the Black Sea shore comprising seven loess layers
362 (L1-L7) and six intercalated paleosols (S1-S6) (Fig. 2, Fig. S3). It is the longest loess sequence
363 in Romania spanning the last 800 ka. The Brunhes-Matuyama geomagnetic reversal has been
364 identified by J. Hus in loess L7 (Balescu et al., 2003). The Mircea Vodă section located on the
365 Dobrogea plateau and the Mostiștea section lying in the southern Danube plain show a
366 superposition of six (L1-L6) and four (L1-L4) loess layers, respectively (Fig. 2). Their
367 stratigraphy is reported on Figure 2. The comparative luminescence ages obtained for the three
368 uppermost loess units (L1, L2, L3) in NE Bulgaria and SE Romania are summarized in Table
369 S6. It should be pointed out that at Mircea Vodă and Mostiștea, a high-resolution sampling has
370 been performed within the upper loess layer L1 (Timar et al., 2010; Timar-Gabor et al., 2011;

371 [Vasiliniuc et al., 2012, 2013](#)) in order to estimate the depositional age range and loess
372 accumulation rate.

373 As shown in [Table S6](#), the OSL ages on fine quartz grains (4-11 μm) at Mircea Vodă provided
374 accurate ages for the Last Glacial loess (L1 unit) but they severely underestimated the expected
375 burial ages below the S1 paleosol ([Timar et al., 2010](#); [Timar-Gabor et al., 2011](#); [Vasiliniuc et al
376 2011](#)). The OSL ages on coarser quartz grains (63-90 μm), at Mostiștea, Mircea Vodă and
377 Costinești, are systematically higher than those obtained from fine quartz grains, both in L1 and
378 L2. The cause for this age discrepancy still remains under investigation; the differences
379 between the natural and the laboratory dose response for the two quartz fractions are believed
380 to be a cause for the observed age discrepancy ([Timar-Gabor and Wintle, 2013](#); [Constantin et
381 al., 2014, 2015](#)).

382 At Mircea Vodă, [Vasiliniuc et al. \(2013\)](#) applied the SAR IRSL₅₀ technique to polymineral fine
383 grains (4-11 μm) from loess L1 to L4 but their SAR IRSL₅₀ ages have a questionable accuracy
384 due to the poor dose recovery performances. These polymineral fine grain samples (from L1 to
385 L4) have also been dated using the pIRIR₂₂₅ technique ([Vasiliniuc et al., 2012](#)). Their fading
386 rate values are all close to 1 %/decade and assumed to be a laboratory artefact. Therefore,
387 [Vasiliniuc et al. \(2012\)](#) choose not to correct their pIRIR₂₂₅ ages for fading. For L1, they
388 measured pIRIR₂₂₅ ages ranging from 70 ± 11 ka to 19 ± 3 ka, up-section. For L2 and L3 and
389 L4, they obtained pIRIR₂₂₅ ages of respectively, 138 ± 22 ka, 245 ± 42 ka and 360 ± 71 ka,
390 consistent with the expected geological ages. However, the pIRIR₂₂₅ ages of L3 and L4 are
391 only minimum ages since the natural pIRIR₂₂₅ signals of L3 and L4 are respectively, close to
392 saturation and in saturation. The pIRIR₂₂₅ technique is therefore restricted at Mircea Vodă to
393 the three last glaciations. This is consistent with the behaviour that has been observed for L3 in
394 NE Bulgaria. This contrasts with the natural IRSL₅₀ signals of both the polymineral fine grains
395 ([Vasiliniuc et al., 2012, 2013](#)) and the K-feldspar silt grains ([Balescu et al., 2003, 2010](#)) of the
396 loess L3 in SE Romania and NE Bulgaria which have a much later saturation (higher D_0
397 values), thus opening the way for dating loess older than L3 (MIS 8). This is the object of
398 further investigation.

399 As shown in [Table S6](#), the MAAD IRSL₅₀ and pIRIR₂₉₀ ages of the Middle Pleistocene
400 NE Bulgarian loess (L2, L3) are consistent with the MAAD-IRSL₅₀ and pIRIR₂₂₅ ages obtained
401 in SE Romania. For the Last Glacial loess L1, in SE Romania and NE Bulgaria, there is a good
402 agreement among the feldspar luminescence ages obtained using multiple techniques (SAR
403 IRSL₅₀, MAAD IRSL₅₀, pIRIR_{225, 290} and MET-pIRIR). They range from 70 ± 11 ka to 19 ± 3
404 ka, covering the MIS 4-2 time interval.

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4.2.2. *Pedostratigraphic records*

The pedostratigraphic records of the Viatovo and Kaolinovo loess sequences (Fig. 2) lying in the Ludogorie region show the same gradual transition to increasingly cooler and drier interglacial climate conditions, up-section from S6 to S0. At both sites, the red paleosols (forest soils) preserved in the lower part of the loess sequences (S4-S6) are overlain by chernozem-like paleosols (S0-S3) developed in drier climatic conditions (steppic soils). This succession of soils reflects a regional trend of increased aridification in interglacial climate during the Middle Pleistocene (Jordanova et al., 2008). A similar climatic trend was observed at Koriten in the Bulgarian Dobrogea (Jordanova and Petersen, 1999a,b) and in SE Romania (Tuzla, Mircea Vodă, Mostiștea) (Conea, 1969; Panaiotu et al., 2001; Buggle et al., 2009; Balescu et al., 2010; Fitzsimmons et al., 2012; Marković et al., 2015). However, at Koriten and in SE Romania (Tuzla, Mircea Vodă, Mostiștea) (Fig. 2), the lower rubified interglacial soils (S3-S6) are overlain by steppe-forest soils (brown red soils; S1-S2) instead of Chernozems. This might suggest a shift towards drier climatic conditions westwards (in the Ludogorie region) during MIS 7 and MIS 5. It is worth also to emphasize that the transition from warm and humid towards cooler and drier climatic conditions occurred at Viatovo and Kaolinovo during MIS 9 with the first chernozem-like paleosol (S3), whereas further east, at Koriten and in SE Romania (Tuzla, Mircea Vodă, Mostiștea), it occurred during MIS 7 with the first steppe-forest soil (S2) (Fig. 2).

4.2.3. *Magnetostratigraphic records*

At Koriten and Viatovo, the magnetostratigraphic characterization of the loess-paleosol sequences provided the first chronostratigraphic framework for the Bulgarian loess (Jordanova and Petersen, 1999; Jordanova et al., 2007, 2008). The magnetic susceptibility (MS) of the loess-paleosol sequences has been widely used for interregional correlations of Pleistocene loess across the lower and middle Danube basin and has also been proved to be a valuable proxy for the reconstruction of past climate changes in this area during the Middle and Upper Pleistocene (Jordanova and Petersen, 1999a,b; Panaiotu et al., 2001; Jordanova et al., 2007, 2008; Hambach et al., 2008; Buggle et al., 2008, 2009; Marković et al., 2011, 2015; Fitzsimmons et al., 2012; Zeeden et al., 2016). Paleosols are identifiable by a characteristic increase in MS value; this enhancement with strongly magnetic minerals is related to pedogenesis. Hence, the MS variations clearly reflect here the pedostratigraphy of the loess-paleosol sequences.

442 On [Figure 5](#), the MS record of the new Kaolinovo loess sequence (unpublished data) is
443 compared to the previously published MS records of Koriten, Mostiștea and Mircea Vodă. As
444 shown on this figure, the loess-paleosol sequences from Koriten and SE Romania, lying
445 eastwards in the Dobrogea area and in the southern Romanian Danube Plain, exhibit the same
446 MS signatures. They register higher MS values in the older red paleosols (S3-S6) and lower
447 MS values in the upper forest-steppe soils (S1, S2). Interestingly, the first stratigraphic
448 correlation between the Romanian and Bulgarian loess sequences established by [Panaiotu et al.](#)
449 [\(2001\)](#) relied on the similarity of the Mostiștea and Koriten MS records.

450 By contrast the Viatovo and Kaolinovo loess-paleosol sequences, lying westwards in the
451 Ludogorie region, show different MS records. At Viatovo, the MS values of the younger
452 chernozem-like paleosols (S1-S3), formed in an arid context are comparatively higher than
453 those of the older red paleosols (S4-S6) developed under forest vegetation and more humid
454 conditions ([Hanesch and Scholger, 2005](#); [Jordanova et al., 2016](#)). This could be ascribed to the
455 predominance of strongly magnetic maghemite pedogenic minerals in the chernozem-like soils
456 and the smaller contribution of weakly magnetic hematite ([Jordanova et al., 1997](#)). While in the
457 older red paleosols a significant part of the pedogenic fraction is represented by hematite
458 resulting in a lower magnetic susceptibility enhancement. Also, the Kaolinovo MS record
459 exhibits a more variable picture. The S3 paleosol presents a lower magnetic susceptibility
460 enhancement as compared to the signal of the two younger paleosols (S2 and S1). This
461 difference is most probably a result of subsequent water logging conditions which would have
462 partially dissolved the finest pedogenic strongly magnetic iron oxides and established the
463 observed redoximorphic features.

464 The cause for this spatial variability in MS signatures among the Bulgarian loess palaeosol
465 sequences and the full palaeoclimatic implications of these MS results will be the subject of
466 another study.

467 **5. Conclusions**

470 This study is the first attempt to assess the luminescence ages to the key reference loess-
471 paleosol sequence of Viatovo in NE Bulgaria. It appears to provide a reliable chronological
472 framework for the Bulgarian loess over the last 300 ka.

473 The MAAD IRSL₅₀ and pIRIR₂₉₀ ages measured on K-feldspars from L1, L2 and L3 at Viatovo
474 and Kaolinovo (NE Bulgaria) are in good agreement with the burial ages expected from
475 stratigraphic and magnetostratigraphic evidence. Hence, it validates the chronostratigraphic
476 framework suggested by [Jordanova et al. \(2008\)](#) on the basis of the MS signature of the loess-

477 paleosol sequence. Moreover, the luminescence ages of the NE Bulgarian loess (L1-L3) are in
 478 good accordance with both the MAAD IRSL₅₀ and the pIRIR₂₂₅ ages of the three uppermost
 479 loess layers (L1-L3) from SE Romania. This IRSL investigation therefore provides new
 480 perspectives for confident interregional correlations of Pleistocene loess across the lower
 481 Danube basin.

482 The SE Romanian and NE Bulgarian loess-paleosol sequences record valuable
 483 pedostratigraphic and magnetic susceptibility data, which in the light of these new
 484 luminescence dates, should provide further insights into the palaeoclimatic evolution of the
 485 lower Danube basin during the Middle Pleistocene.

486 The pedostratigraphic records of the SE Romanian and NE Bulgarian loess-paleosol sequences
 487 indicate similar palaeoclimatic trends with progressive aridification during interglacials.
 488 However the transition from humid and warm to cooler and drier interglacial conditions
 489 occurred much earlier westwards, in the Ludogorie area (paleosol S3, MIS 9), than in the
 490 Dobrogea area (paleosol S2, MIS 7).

491

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690 Captions

691 Figure 1A. Loess distribution in Europe and location of the study area.

692
 693 Figure 1B. Location of the sampling sites and sites mentioned in the text.

694
 695 Figure 2. Loess-paleosol sequences from NE Bulgaria and SE Romania.
 696 Lithostratigraphy: L= loess deposit; S: soil and palaeosol.

697
 698 Figure 3. Comparative dose response curves of the pIRIR₂₉₀ signal observed for samples VIA2
 699 (L2), VIA3 (L3) and VIA7 (L7). The inset shows the natural decay curve of the pIRIR₂₉₀
 700 signal.

701
 702 Curves are fitted with a single saturating exponential function; VIA2 (L2): 2D₀=704; VIA3 (L3): 2D₀=718;
 703 VIA7 (L7): 2D₀=683.

704
 705
 706 Figure 4. IRSL_{50°C} additive growth curves obtained for samples VIA3 (L3) and VIA7 (L7).
 707 Curves are fitted with a single saturating exponential function; VIA3 (L3): 2D₀=1386; VIA7 (L7):
 708 2D₀=1466.

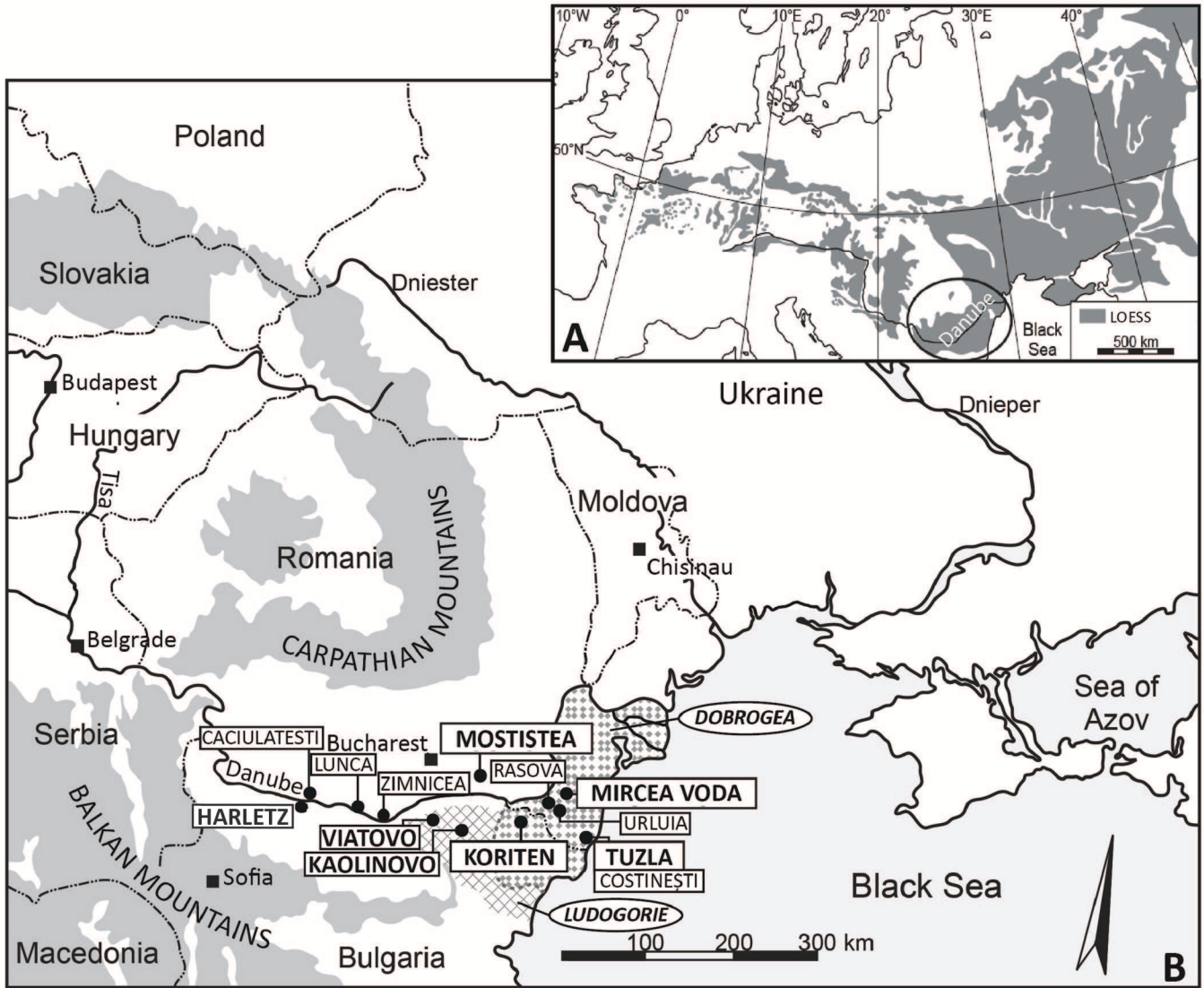
709
 710 Figure 5. Comparative magnetic susceptibility records of the SE Romanian and
 711 NE Bulgarian loess sequences.

712 Gray circles: IRSL samples from Balescu et al. (2010); white circles : IRSL samples from the
 713 present study.

714 References: Mostiștea (Panaiotu et al., 2001); Mircea Vodă (Timar et al., 2010); Koriten (Jordanova and
 715 Petersen, 1999a,b) ; Viatovo (Jordanova et al., 2008) ; Kaolinovo (unpublished)

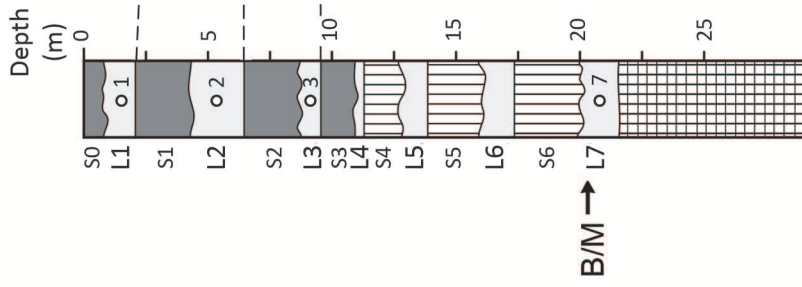
716
 717 Table 1. Luminescence data for the Bulgarian loess

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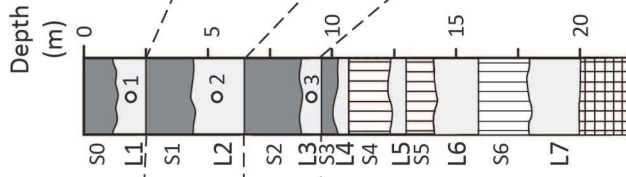


NE BULGARIA

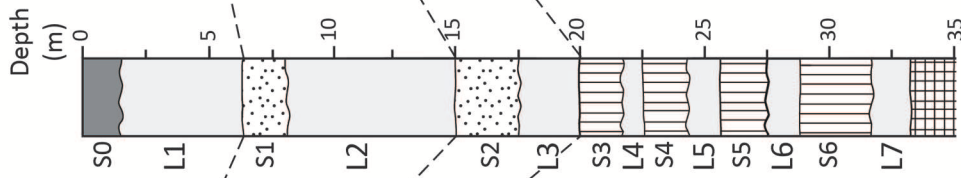
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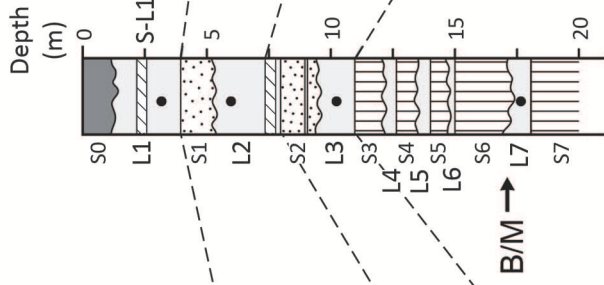
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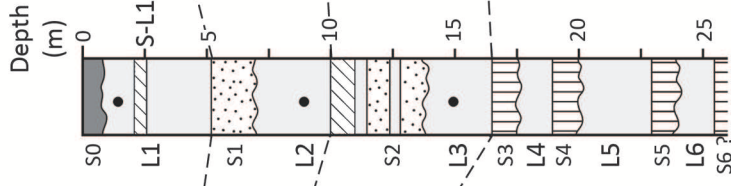
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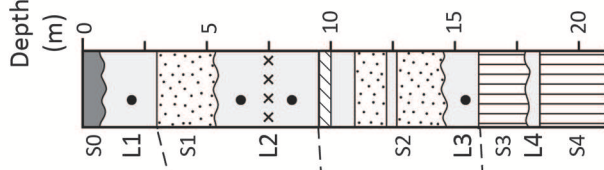
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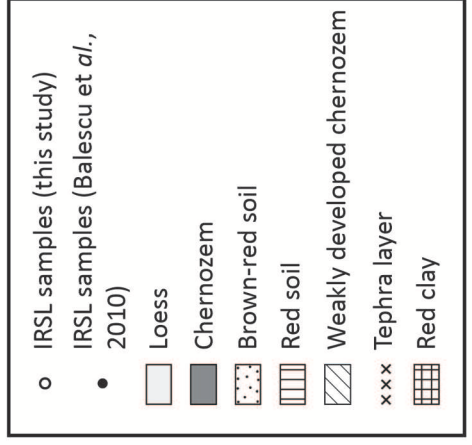
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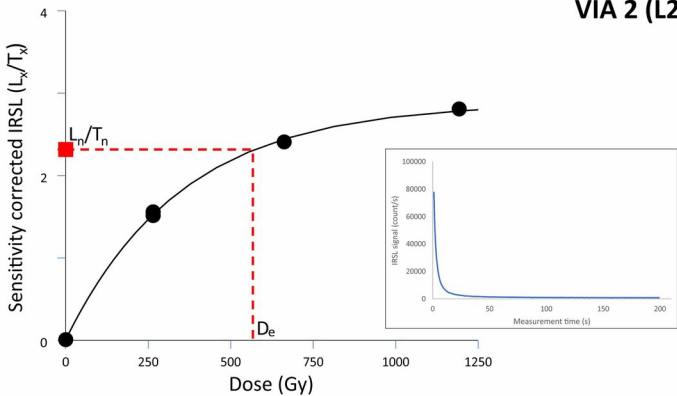
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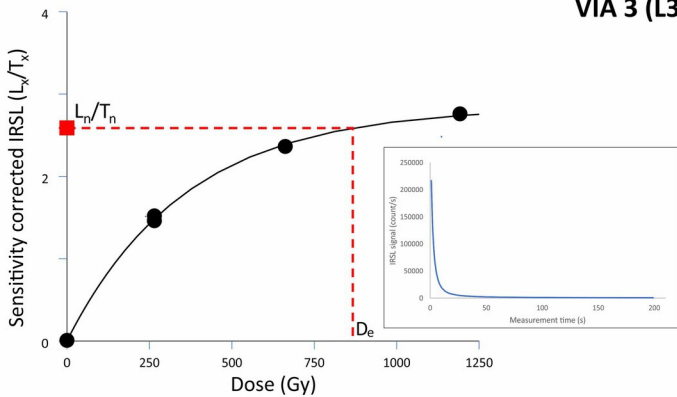
SE ROMANIA



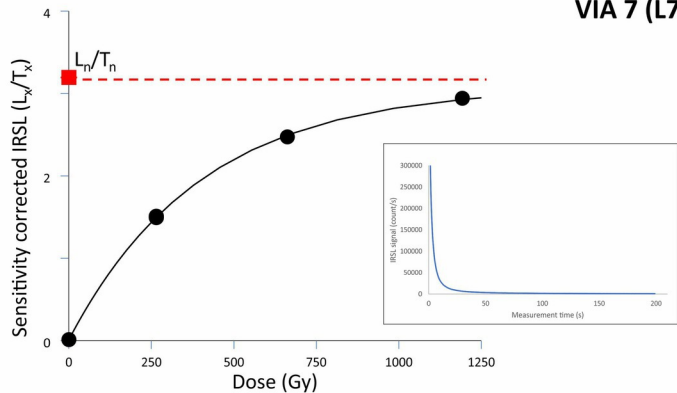
VIA 2 (L2)

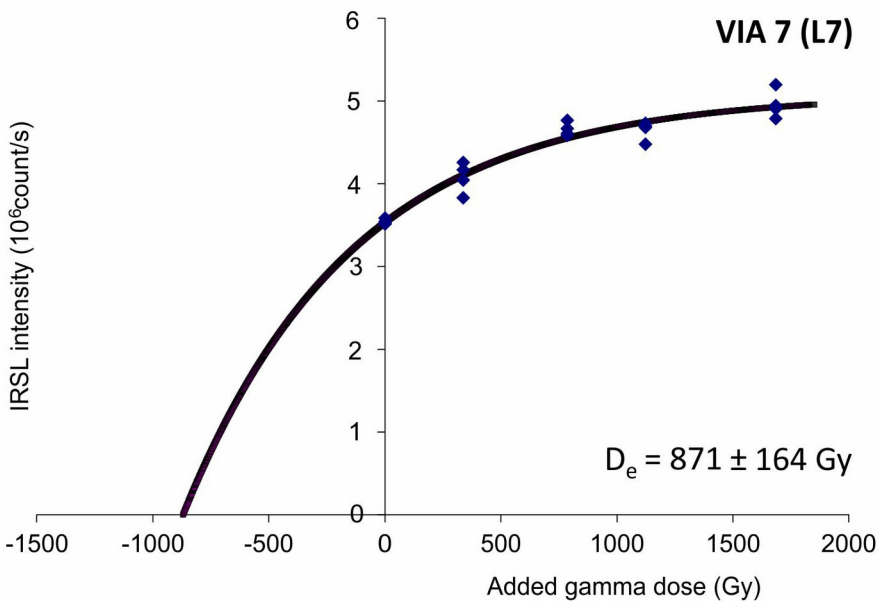
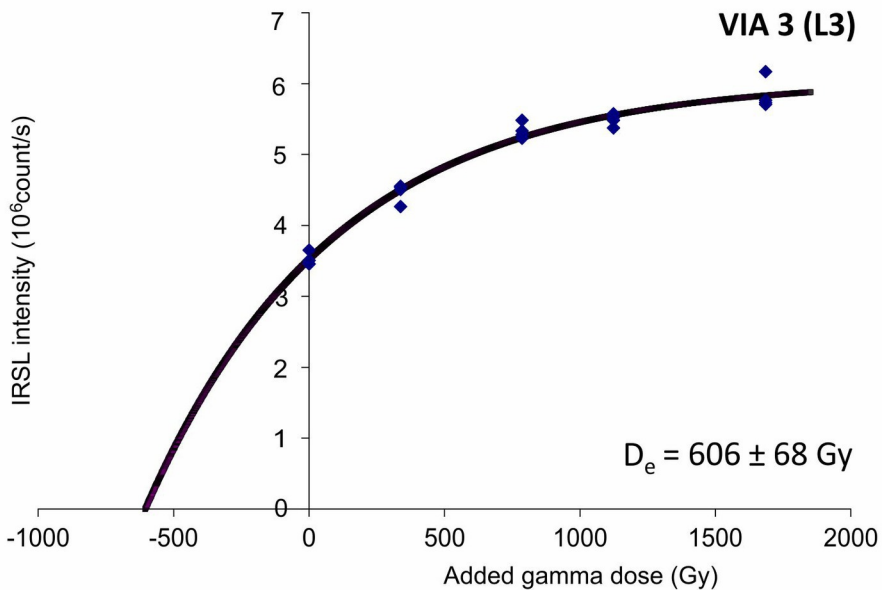


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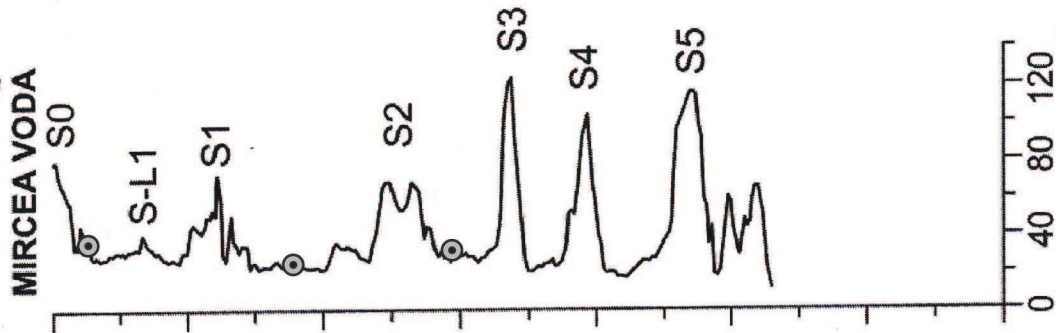
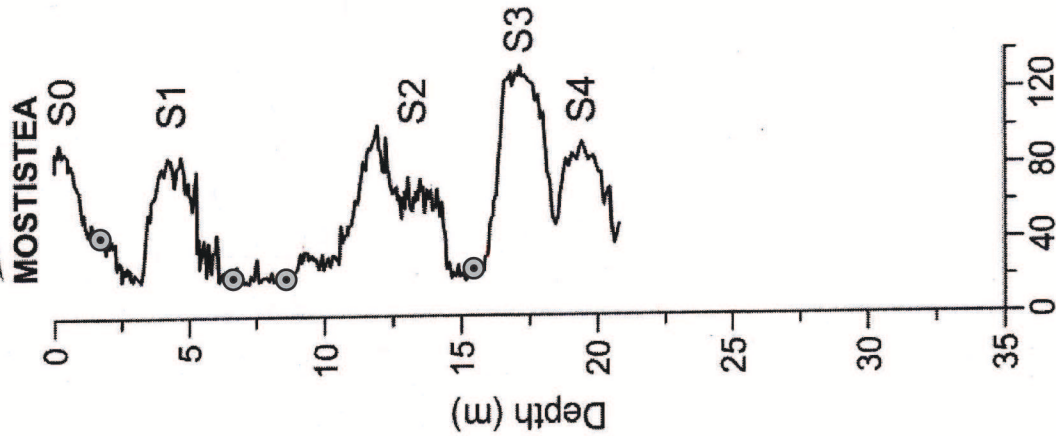


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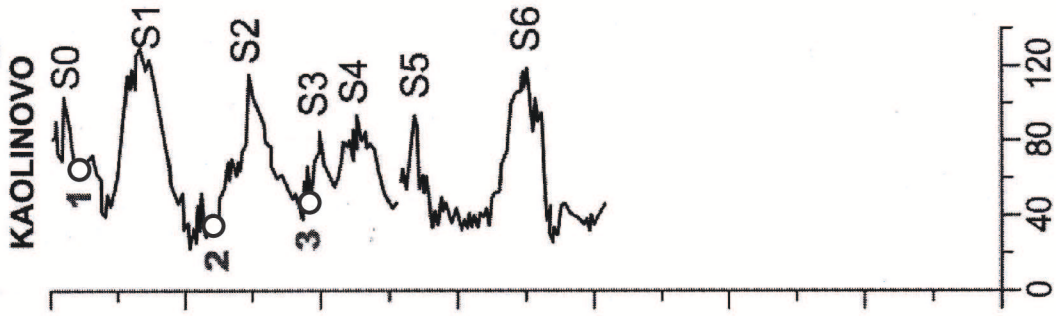
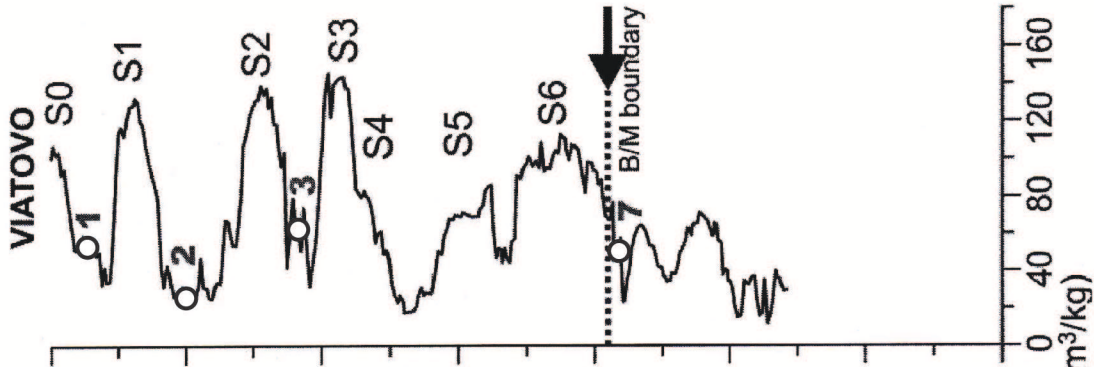
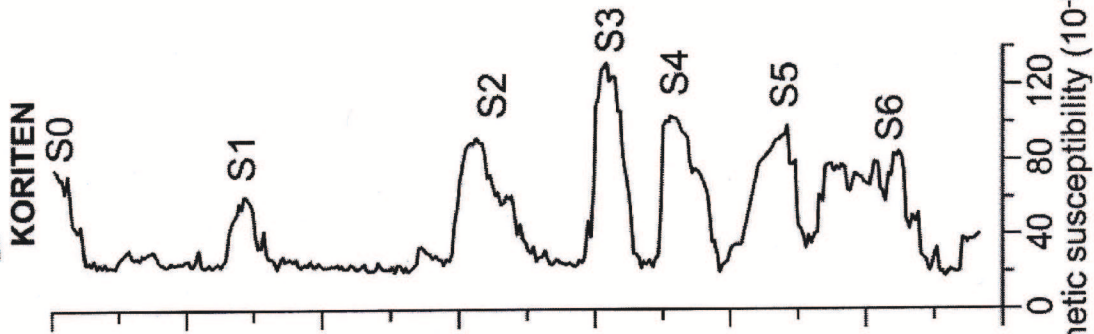




SE ROMANIA



NE BULGARIA



Site	Sample	Loess unit	Technique	Measured De (Gy)	Da (Gy/ka)	Measured ages: MAAD IRSL ₅₀ and PIR-IR ₂₉₀ (ka ±σ)	Corrected ages: MAAD IRSL ₅₀ (ka ±σ)	2Do	Chronostratigraphy
Viatovo	VIA 1	L1	MAAD-IRSL ₅₀	182 ± 8	3.22 ± 0.13	57 ± 5	60 ± 5	1136	MIS 2-4
			PIR-IR ₂₉₀	205 ± 5	3.22 ± 0.13	64 ± 5	-	741	
	VIA 2	L2	MAAD-IRSL ₅₀	442 ± 25	3.18 ± 0.13	139 ± 12	167 ± 14	1322	MIS 6
			PIR-IR ₂₉₀	572 ± 6	3.18 ± 0.13	180 ± 9	-	726	
	VIA 3	L3	MAAD-IRSL ₅₀	606 ± 68	2.97 ± 0.12	204 ± 26	285 ± 35	1386	MIS 8
			PIR-IR ₂₉₀	871 ± 37	2.97 ± 0.12	293 ± 17	-	694	
	VIA 7	L7	MAAD-IRSL ₅₀	871 ± 164	2.94 ± 0.12	296 ± 60	>1066	1466	> B/M ~780 ka
			PIR-IR ₂₉₀	saturated	2.94 ± 0.12	Infinity	-	736	
Kaolinovo	KA 1	L1	MAAD-IRSL ₅₀	218 ± 10	5.13 ± 0.18	42 ± 3	44 ± 3	1428	MIS 2-4
			PIR-IR ₂₉₀	228 ± 5	5.13 ± 0.18	45 ± 2	-	550	
	KA 2	L2	MAAD-IRSL ₅₀	516 ± 77	3.43 ± 0.13	150 ± 25	184 ± 29	1458	MIS 6
			PIR-IR ₂₉₀	573 ± 13	3.43 ± 0.13	167 ± 7	-	746	
	KA 3	L3	MAAD-IRSL ₅₀	638 ± 151	3.33 ± 0.13	192 ± 48	264 ± 64	1364	MIS 8
			PIR-IR ₂₉₀	986 ± 43	3.33 ± 0.13	296 ± 17	-	722	

Table 1