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1 **The last 30,000 to 700,000 years ago: unravelling the timing of human settlement for the**  
2 **Palaeolithic site of Kozarnika**

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4 Maryam Heydari<sup>1,2\*</sup>, Guillaume Guérin<sup>2,3</sup>, Nikolay Sirakov<sup>4</sup>, Philippe Fernandez<sup>5</sup>, Catherine  
5 Ferrier<sup>6</sup>, Aleta Guadelli<sup>7</sup>, Jean-Claude Leblanc<sup>8</sup>, Stanimira Taneva<sup>4</sup>, Svoboda Sirakova<sup>4</sup> and  
6 Jean-Luc Guadelli<sup>6</sup>

7  
8 <sup>1</sup>Institute of Earth and Environmental Sciences, University of Freiburg, Albertstr. 23b, 79104  
9 Freiburg, Germany

10 <sup>2</sup>IRAMAT-CRP2A, UMR 5060, CNRS-Université Bordeaux Montaigne, Maison de  
11 l'archéologie, Esplanade des Antilles, 33607 Pessac cedex, France

12 <sup>3</sup>Université de Rennes, CNRS, Géosciences Rennes, UMR 6118, 35000 Rennes, France

13 <sup>4</sup>National Institute of Archaeology with Museum-Bulgarian Academy of Sciences, 2, Saborna  
14 Street, 1000 Sofia, Bulgaria

15 <sup>5</sup>CNRS, Aix Marseille Univ, Minist Culture, LAMPEA UMR 7269, Maison Méditerranéenne  
16 des Sciences de l'Homme, 5 Rue du Château de l'Horloge BP 647, F13094, Aix-en-Provence,  
17 France

18 <sup>6</sup>PACEA-UMR5199 CNRS, Université de Bordeaux, Allée Geoffroy Saint Hilaire, Bâtiment  
19 B18, CS50023, 33615 Pessac Cedex, France

20 <sup>7</sup>National Institute of Immovable Cultural Heritage, Latchezar Stanchev Street 7, 1125 Sofia,  
21 Bulgaria

22 <sup>8</sup>TRACES-UMR5608 CNRS, Université de Toulouse le Mirail, Maison de La Recherche, 5,  
23 Allées Antonio Machado, 31048 Toulouse Cedex 1, France

24

25

26

27 \*Corresponding author: [mariheyd@gmail.com](mailto:mariheyd@gmail.com)

28

29 **Abstract**

30 Kozarnika cave is a renowned prehistoric site in the Balkans, which contributes significantly  
31 to our understanding of the human past due to its rich assemblages associated with the Lower  
32 to Upper palaeolithic periods. To unravel the timing of human occupations in Kozarnika,  
33 various dating methods have been employed in the past. Radiocarbon dating was used to unfold  
34 the timeframe for the Kozarnikian lithic tradition uncovered in the Upper Palaeolithic sequence  
35 of the cave, and paleomagnetic dating assigned the Brunhes–Matuyama reversal to the layer  
36 beneath the Lower Palaeolithic assemblages.

37 In this study, we employed luminescence-dating methods including a body of different signals  
38 to date sediment layers from the top to the bottom of the sequence covering the period of ca 30  
39 to 700 ka. Our investigations revealed that the Kozarnikian tradition in layers 5a-c falls between  
40 30–35 ka. Following that, we suggest that the Middle Palaeolithic period initiated between  
41 250–309 ka and lasted until 40–53 ka. More importantly, we have updated the age of the  
42 Neanderthal radius discovered in the Mousterian assemblages to 201±17 ka.

43 Our dating of the Lower Palaeolithic assemblages in the cave rendered the period of ca 300-  
44 700 ka. Taking into account the timeframe suggested by the paleomagnetic sequence <780 ka  
45 it is likely that we have reached the threshold of the luminescence dating method and the age  
46 range may represent the minimum age for this period.

## 47 **1. Introduction**

48 The Balkans are renowned for witnessing different hominin and human species of inhabitants  
49 due to the location between the Near East and the Western Europe during the long period of  
50 the Palaeolithic. Human remains associated with the Neanderthals or anatomically modern  
51 humans (AMH) have been discovered in the Middle and Upper palaeolithic assemblages in  
52 various sites (e.g., Galanidou, 2004; Glen and Kaczanowski, 1982; Hoffecker, 2009; Hublin et  
53 al., 2020; Strait et al., 2016; Tillier et al., 2017). Additionally, the high possibility of interaction  
54 between two human species (Fu et al., 2015; Hajdinjak et al., 2021) renders this region unique  
55 for further investigations.

56 Establishing a reliable temporal framework disclosing the timing of hominin species'  
57 settlements, the demise of one (e.g., Neanderthal), and domination of another (e.g., AMH) is a  
58 fundamental study, and it can be revealed through applying numerical dating methods. Among  
59 the whole body of techniques, radiocarbon dating plays a major role to track dynamics of the  
60 human peopling during the Late Middle to Upper Palaeolithic periods generally for Europe  
61 (e.g., Higham et al., 2014) and in particular for the Balkans (e.g., Alex et al., 2019; Devière et  
62 al., 2017; Fewlass et al., 2020; Higham et al., 2006.; Trinkaus et al., 2003).

63 Whilst numerous temporal frameworks have been constructed to date the humans' fossils or  
64 the artefacts attributed to the Middle/Upper Palaeolithic, the number of available datasets is  
65 relatively scarce for the Lower to Middle Palaeolithic. This could be explained due to the  
66 shortage of the well-preserved Lower Palaeolithic sequences in eastern Europe (Doronichev,  
67 2008). Having said that, the Palaeolithic site of Kozarnika has been a special place all along  
68 not only because of its rich archaeological remains related to the Lower Palaeolithic but also  
69 for offering the first Bulgarian Neanderthal's remain, a radius associated with a rich Mousterian  
70 assemblage (Tillier et al., 2017). Furthermore, the Upper palaeolithic industry found in the cave  
71 stands out from the other lithic productions and is known as *Kozarnikian* (Guadelli et al., 2005;  
72 Tsanova, 2006; Sirakov et al., 2007).

73 During the last decades, different dating methods have been employed to establish reliable  
74 chronologies for the sequences in the cave. The Upper Palaeolithic sequence was dated using  
75 the radiocarbon method (Guadelli et al., 2005, in press). Moreover, preliminary luminescence

76 ages attributed to the neanderthal radius were previously published (Tillier et al., 2017). In  
77 another study, a palaeomagnetism sequence was established for stratified layers in Kozarnika  
78 (Muttoni et al., 2017). Finally, a timeframe for the Lower and Middle Palaeolithic assemblages  
79 was demonstrated on mammal biochronology (Guadelli et al., 2005, in press; Fernandez and  
80 Cregut-Bonnoure, 2007; Popov and Marinska, 2007; Sirakov et al., 2010).

81 In this study, we applied multiple luminescence-dating methods not only to elucidate the  
82 timeframe of humans resided during the Middle and Upper Palaeolithic but to shed light on the  
83 timing of the inhabitants at the Lower Palaeolithic and provide a full chronology covering the  
84 complete sequence through sediment samples. This will allow us to improve our understanding  
85 about the timing of humans' settlements, particularly for Kozarnika and more widely for the  
86 Balkans.

87

### 88 **1.1 Site location**

89 Kozarnika cave is located in the northwest of Bulgaria, 30 km away from the Serbian border  
90 (Fig. 1). The site is situated in the northern slopes of the Balkan Mountains, merging to the  
91 Danubian Plain, which is extended to the Danube River to the north. The cave is positioned at  
92 an altitude of 481 m on the mountainside of Bashovishka Bara valley. The streams in the valley  
93 drain into the Skomlia River irrigating the region, which feeds the Danube River ca 30 km  
94 away.

95 The first report about Kozarnika dates back to 1933 (Popov (Попов), 1933) and years after in  
96 1996, the site was excavated systematically by Bulgarian-French researchers (Guadelli et al.,  
97 2005). Following years of collaborative work, 21 geological layers were discovered in a  
98 sequence of about 9 m containing assemblages attributed to the Lower, Middle, Upper  
99 Palaeolithic as well as Holocene.

100

Fig. 1.

101

### 102 **1.2 Stratified geological layers including archaeological discoveries**

103 The Bashovishka Bara valley has cut 185 m deep into the northern foothills of the Balkan  
104 through three main geological formations. The top formation belongs to the Early Jurassic  
105 which at the middle is replaced by the Middle Jurassic and finally at the bottom is superimposed  
106 on the Late Jurassic. The Kozarnika cave consists mainly of limestone, which is formed in the  
107 Upper Jurassic geological formation (Guadelli et al., 2005; Sirakov et al., 2010). The cave is  
108 part of a larger karst landscape with other caves discovered in the north-western region of  
109 Bulgaria.

110 The Pleistocene layers in the cave are generally well-preserved. However, we observed  
111 truncations in the cave for some layers containing Middle and Upper Palaeolithic assemblages  
112 (Guadelli et al., 2005; Sirakov et al., 2010). These truncations are likely stemming from the  
113 cave's subsequent use in modern times, when it was used as a mushroom farm and goat farm,  
114 and finally as a bunker in the 1960s.

115 The geological layers in the cave were categorised into four main groups covering the top to  
116 the bottom of the excavation profile (Fig. 2). Their general characteristics are explained below.

117

118

Fig. 2.

119

120

### 121 **1.3 Geological layers containing the archaeological finds**

122 **Layers 3a-4.** These layers with a thickness of 1 m to 1.4 m form the top of the sequence. Most  
123 of the sediment is of aeolian origin. Nevertheless, a considerable amount of calcareous clasts  
124 gravels in colours from light brown to whitish silty medium can be found in this part in addition  
125 to debris from the ceiling and the walls of the cave. The archaeological assemblages belong to  
126 the middle and late Kozarnikian tradition (the indigenous eponymous bladelets industry  
127 mentioned above) associated with the Upper Palaeolithic (Guadelli et al., 2005).

128 **Layer 5a-10a.** These layers show thicknesses of 1.2-1.5 m and were deposited through a  
129 combination of aeolian transport (recognised by beige silty-fine grains), and sediment  
130 fragments from the ceiling and walls (identified with coarse grains and calcareous gravel and  
131 pebble). Both groups of sediments are distributed irregularly over the section. Layer 5b  
132 contains some volcanic ashes superimposed on layers 6 and 7, which both were composed of  
133 silty sediment varying from light grey (6) to light brown (7). Due to the similarity between the  
134 origin of these two layers, they both were considered as one layer named 6/7. The calcareous  
135 gravels and charcoal also were sparsely observed in this layer (Ferrier et al., 2009). Generally  
136 speaking, the formation of layers 5a, 6/7, and 10a is dominated by aeolian processes. Layers  
137 10a-9a contain archaeological elements related to the Middle Palaeolithic period. The  
138 archaeological finds in layer 6/7 are consistent with the Middle and Initial Upper Palaeolithic  
139 assemblages. The upper layer (5a) contains the blade industry known as the Kozarnikian  
140 tradition.

141 **Layer 10b-10c.** These layers originated from roughly weathered calcareous gravel and  
142 pebbles, with a combination of loamy sand with dark brown colour that could be attributed to  
143 human activity in the past. The sediment thicknesses of these layers vary from 15 cm to 50 cm

144 and contain archaeological artefacts associated with the very early Middle Palaeolithic with the  
145 Mousterian characteristics.

146 **Layer 11a-13.** These layers were located at the bottom of the sequence having a thickness of  
147 ca 2.5 m. The sediments in this section consist of orange loamy fine and coarse fractions with  
148 flint, limestone, and debris from the ceiling and the walls of the cave. In particular calcareous  
149 pebbles and blocks with various concentrations were observed in layer 11a as a common feature  
150 of this layer in a different part of the cave. The same pattern with a combination of a dark  
151 fraction of silty material with quartz origin applies for layers 11b, 11c, 12, and 13. It is likely  
152 that this sequence of layers is a result of mudslides according to Ferrier et al. (2009). All of  
153 these layers comprise findings from an industry affiliated with the Lower Palaeolithic period.

## 154 **2. Material and Methods**

155 Luminescence dating estimates energy (radiation dose) deposited in natural minerals after the  
156 last sunlight exposure. This quantity is known as the equivalent dose (estimated in the  
157 laboratory), which is built up by energy emitted by natural radioelements in the sediment over  
158 the burial time (dose rate). Dividing the equivalent dose by the corresponding dose rate, results  
159 in the burial time (age). We used various techniques to determine the equivalent doses of the  
160 samples. First, we began with the blue optically stimulated luminescence (OSL) for quartz  
161 (Huntley et al., 1985), followed by post infrared stimulated luminescence at the elevated  
162 temperature (Post-IR IRSL, Thomsen et al., 2008) to completely cover the Middle to the Upper  
163 palaeolithic sequences. In addition, the infrared stimulated luminescence (IRSL, Hütt et al.,  
164 1988) signals for some samples were measured at 50 °C to crosscheck with the two other age  
165 series.

166 It is well-known that signal saturation is a key parameter hampering the application of  
167 luminescence dating for the Lower to Middle Palaeolithic. Given that, we attempted to tackle  
168 this problem by combining various luminescence signals with different saturation degrees to  
169 extend the age limit. We switched to violet stimulated luminescence (VSL) (Ankjærgaard et  
170 al., 2016, 2013; Jain, 2009) on quartz accompanied by infrared radiofluorescence (IR-RF)  
171 (Trautmann et al., 1999b, 1999a, 1998, for review see Murari et al., 2021) on K-feldspar to  
172 estimate ages for the older samples.

173 The dose rates were estimated by means of both *in situ* and laboratory measurements for all  
174 samples (further details below).

### 175 **2.1 Sample preparation**

176 We present ages for 18 sediment samples extracted from the profile, using opaque tubes (see  
177 Table 1). Our sample preparation followed routine procedures applied in luminescence dating  
178 (Preusser et al., 2008). After conducting the grain size analysis (see Fig. S1 in the  
179 supplementary material) coarse grains with 180  $\mu\text{m}$  to 250  $\mu\text{m}$  diameters were extracted using  
180 wet sieving. To eliminate carbonates, the obtained fraction of sediments was rinsed in HCl  
181 (30%) over a couple of hours until no further reaction was observed. Following that, they were  
182 soaked in  $\text{H}_2\text{O}_2$  (30%) to remove organic compounds. Later, density separation was carried out  
183 through lithium hetero-polytungstates (LST) heavy-liquid. First, quartz grains were separated  
184 from the bulk sample using a density of  $2.62 \text{ g cm}^{-3}$ . Then, K-feldspar grains were extracted  
185 from the remaining material by applying the LST with a density of  $2.58 \text{ g cm}^{-3}$ . We removed  
186 the outer layers of quartz and K-feldspar grains affected by alpha particles by etching them  
187 with HF; (40% for 40 min on quartz and 10% for 10 min on K-feldspar). Etching was followed  
188 by resting the grains in HCl (15%) overnight to eliminate unwanted fluorides. Finally, all grains  
189 were sieved for the second time with the smaller mesh size ( $<180 \mu\text{m}$ ) to remove any remaining  
190 fragmented grains.

191 Table 1

### 192 **2.3 Instrumentation**

193 Various instruments were employed for measuring luminescence signals using different  
194 minerals and techniques. Blue OSL was measured on Freiberg Instrument lexsysg SMART  
195 TL/OSL systems (Richter et al., 2015) equipped with five blue LEDs ( $458 \Delta 3 \text{ nm}$ , max.  $100$   
196  $\text{mW cm}^{-2}$ ) and five infrared LEDs ( $850 \Delta 3 \text{ nm}$ , max.  $300 \text{ mW cm}^{-2}$ ) as well as on a Freiberg  
197 Instrument lexsysg research reader (hereafter L1, (Richter et al., 2013) equipped with five blue  
198 LEDs ( $458 \Delta 10 \text{ nm}$ , max.  $100 \text{ mW cm}^{-2}$ ) and five infrared LEDs ( $850 \Delta 3 \text{ nm}$ , maximum  $300$   
199  $\text{mW cm}^{-2}$ ). The stimulation powers for lexsysg research and lexsysg SMART systems were set  
200 to  $100 \text{ mWcm}^{-2}$  and  $40 \text{ mWcm}^{-2}$ , respectively. Signals were detected using a UV filter  
201 combination (Hoya U340, 2.5 mm and Delta BP 365/50 EX) in both systems.

202 Post-IR IRSL (pIR<sub>50</sub>IRSL<sub>290</sub>) signal was measured using a blue-violet filter combination Schott  
203 BG 39 (3 mm) and Chroma D410/30x in Freiberg Instruments lexsysg research reader with a  
204 stimulation power of  $100 \text{ mWcm}^{-2}$ .

205 IRSL signals were measured using Risø TL/OSL DA-15 and DA-20 readers equipped with five  
206 blue LEDs ( $470 \Delta 20 \text{ nm}$ , max.  $105 \text{ mW cm}^{-2}$ ) and five infrared LEDs ( $870 \Delta 40 \text{ nm}$ , max.  $145$   
207  $\text{mW cm}^{-2}$ ). Signals were detected through a combination of Schott BG39 and Corning 7-59  
208 (Bøtter-Jensen et al., 2003).

209 VSL signals were measured using Freiberg Instruments lexsyg research reader containing  
210 violet-emitting LEDs (405  $\Delta$  3nm, max. 100 mW cm<sup>-2</sup>). The stimulation power was set to 100  
211 mWcm<sup>-2</sup> and the signal was detected using a combination of a Hoya U340 (2.5 mm) and an  
212 AHF-BL HC 340/26 interference filter.

213 All systems were operated with a <sup>90</sup>Sr/<sup>90</sup>Y- source which was calibrated using Risø calibration  
214 quartz (Hansen et al., 2015). The dose rates available for each sample in lexsyg research and  
215 SMART as well as Risø TL/OSL systems were ca 8 Gy min<sup>-1</sup>, 11 Gymin<sup>-1</sup>, and 7 Gy min<sup>-1</sup> at  
216 the measurement dates, respectively.

217 IR-RF was measured through another Freiberg Instrument lexsyg research system, which was  
218 equipped with a Hamamatsu H7421-50 PMT in conjunction with a Chroma D850/40  
219 interference filter. A calibrated <sup>90</sup>Sr/<sup>90</sup>Y- source delivered a dose rate of ca 3 Gy min<sup>-1</sup>. For the  
220 bleaching of the natural IR-RF, we used the internal solar simulator of the machine. All samples  
221 were prepared using stainless steel cups with a diameter of 8 mm.

222 Approximately, a hundred grains were mounted in diameter of 3 mm using silicon oil for  
223 measuring blue OSL, pIR<sub>50</sub>IRSL<sub>290</sub>, and IRSL. The number of grains was increased for VSL  
224 and IR-RF; in these cases, the cups were fully covered by grains, representing several thousand  
225 grains.

#### 226 **2.4 Dose rate**

227 The dose rate available to quartz and feldspar consists of external and internal parts due to the  
228 activity of the radioelements outside and inside of the grains. For the external part, we have  
229 only estimated beta and gamma dose rates because the etching removed the outer layer of the  
230 grains affected by alpha-radiation.

231 We estimated the radionuclide concentrations of U, Th, and K using high-resolution low  
232 background gamma-ray spectrometry (Guibert and Schvoerer, 1991). The obtained  
233 concentrations were then turned into beta-dose rates using conversion factors after Guérin et  
234 al. (2011) and corrected using grain size attenuation factors following Guérin et al. (2012). The  
235 effect of water on dose rate was corrected after Nathan and Mauz (2008). We estimated the  
236 saturation water content based on the grain size distribution as suggested in Nelson and  
237 Rittenour (2015). The sample-specific percentages of the sand, silt, and clay fraction are  
238 displayed in Table S1 in the supplementary material. We have used the fraction of 0.5 $\pm$ 0.2 (of  
239 the saturation value) as a representative value to approximate the past water content. We  
240 increased the obtained value to 20 $\pm$ 8% for samples (Koz16 to Koz21) due to the mudflow  
241 origin of layer 11b downwards.

242 Gamma-dose rates were estimated using artificial dosimeters ( $\text{Al}_2\text{O}_3:\text{C}$  chips; see Kreutzer et  
243 al., 2018 for further details) for all samples. The chips were first heated up for 10 min at  $350^\circ$   
244 C. Three chips were inserted in each aluminium tube and remained in the sediment for 271  
245 days. After retrieving, the stored energy was measured using Daybreak 2200 OSL reader  
246 system (Bortolot, 2000). The chips were stimulated through green light (Nichia NSPG10) and  
247 the emissions were recorded through a 7.5 mm Hoya U-340 filter. For comparison, the gamma-  
248 dose rate was also estimated using radioelement concentrations derived from the gamma-ray  
249 spectrometry. The concentrations were converted to the dose rate after convention factors  
250 tabulated in (Guérin et al., 2011) and corrected for water content following (Guérin and  
251 Mercier, 2012).

252 The cosmic-dose rate was calculated using the coordinate of the site and the overburden depth  
253 after the procedure in Prescott and Hutton, 1994; Prescott and Stephan (1982). The uncertainty  
254 of the cosmic dose rate was estimated at 10% after Prescott and Hutton (1994).

255 The internal dose rate was estimated using K and Rb concentrations of  $12.5 \pm 0.5\%$  (Huntley  
256 and Baril, 1997) and  $460 \pm 20$  ppm after Mejdahl (1987) respectively. The internal dose rate for  
257 quartz coarse grains was taken from Mejdahl (1987).

## 258 **2.5 Luminescence signal measurements**

259 We applied several protocols to both quartz and feldspar grains and accordingly the  
260 corresponding signals were used for dating. We have measured blue-OSL and VSL for quartz  
261 grains. In addition, IRSL at  $50^\circ\text{C}$ , and  $\text{pIR}_{50}\text{IRSL}_{290}$  at an elevated temperature ( $290^\circ\text{C}$ ) were  
262 measured to establish ages based on feldspar grains. Nevertheless, this approach potentially  
263 could disclose the history of grains' bleaching, which is a worthy matter of consideration since  
264 the accuracy of the obtained ages heavily depends on it. Below, the setting for each protocol is  
265 explained.

### 266 **Blue optically stimulated luminescence (blue-OSL)**

267 We used the single aliquot regenerative (SAR) dose protocol (Murray and Wintle, 2000) to  
268 estimate the equivalent dose ( $D_e$ ) for each sample.

269 First, the dominance of the fast-decaying signal favourable for relatively young sediment  
270 samples was tested. We simply compared the blue-OSL signal from one of Kozarnika's  
271 samples with calibrated quartz Risø batch 90 (Hansen et al., 2015), which is known to be  
272 dominated by the fast decaying luminescence signal. In addition, a linearly modulated OSL  
273 (LM-OSL, Bulur, 1996) technique was applied to deconvolute the signal components for this  
274 sample to be compared with the calibration quartz.  $D_e$  estimation was carried out using Analyst

275 software v. 4.53 (Duller, 2015). A single saturating exponential function was used for fitting  
276 the dose-response curve.

277 The regenerative dose steps were followed by a null dose delivered to monitor charge transfers.  
278 This step was followed by a repetition of the first regenerative dose to verify significant  
279 sensitivity changes. In the end, the same dose as the first regenerative was delivered to the  
280 samples and the corresponding signal was measured after stimulating with IRSL followed with  
281 blue OSL to check any significant contamination by feldspar grains after Duller (2003).

282 The luminescence signal was measured at 125°C for 40 s after preheating at 260°C for 10 s.  
283 The preheat plateau test was applied to sample Koz2 to select an appropriate temperature before  
284 measuring the signal in SAR protocol (Murray and Wintle, 2000). In this test,  $D_e$ s were  
285 measured at various degrees starting from 200°C and reaching 300°C (ramping up by 20°C at  
286 each step). The cut heat was 40°C lower than the corresponding preheat temperature. Dose-  
287 recovery tests were conducted on some samples to monitor the reliability of the SAR protocol's  
288 parameters. Samples were bleached for 100 s using blue light, followed by a pause of 10,000  
289 s. For the second time samples were bleached using blue light and then they received a beta  
290 dose inside the OSL reader approximately equivalent to their natural doses (Koz2, 4, 7, 10).

#### 291 **Infrared stimulated luminescence (IRSL<sub>50</sub> and pIR<sub>50</sub>IRSL<sub>290</sub>)**

292 K-feldspars were used for estimating the  $D_e$ s for some of the samples to obtain independent  
293 ages, also revealing the grains' bleaching histories. Particularly, K-feldspar is favourable when  
294 the expected temporal range is higher than what the blue-OSL signal of quartz can achieve.  
295 Due to this higher upper age limit, this mineral has been used to unravel the temporal  
296 frameworks attributed to the Middle and also Lower Palaeolithic periods of the cave. Having  
297 said that, the drawback of using feldspar is athermal anomalous fading (loss of signal over time  
298 (Wintle, 1973)) that impedes the proper estimation of the true  $D_e$ . To overcome this problem,  
299 we determined the fading rate (Huntley and Lamothe, 2001) and corrected its effect on  $D_e$ . In  
300 this study, for three samples Koz4, Koz8, and Koz11, IRSL signals were measured (six aliquots  
301 for each sample) following fading measurement after a pause of approximately 1 h and 4 h.  
302 The fading rate was calculated using the function  
303 *Luminescence::analyse\_FadingMeasurement()* implemented in the R (R Core Team, 2021)  
304 'Luminescence' (Kreutzer et al., 2012; Kreutzer et al., 2021) package after methodological  
305 work by (Huntley, 2006). These values were then used to correct the  $D_e$  estimates using  
306 *Luminescence::calc\_Huntley2006()* after Kars et al., 2008 and King et al., 2018 also embedded  
307 in the R 'Luminescence' package. A single saturating exponential function was used to  
308 generate a dose-response curve.

309 Furthermore, we measured the IRSL signal at an elevated temperature following the IRSL at  
310 50°C. This method was suggested after (Buylaert et al., 2011; Li and Li, 2011; Thiel et al.,  
311 2011; Thomsen et al., 2011, 2008) and it is proposed that the signals do not suffer from the  
312 fading, or at least the fading rate can be considered as insignificant (Buylaert et al., 2012, 2011).  
313 The parameters' setting for measuring this signal was derived from Buylaert et al., (2012) and  
314 was applied to a large number of samples containing 4 to 12 aliquots.

### 315 **Infrared radiofluorescence (IR-RF)**

316 Aside from the IRSL signal, K-feldspar emits another signal during ionising radiation, which  
317 is detected in the infrared wavelength range, known as IR-RF (for a recent review see Murari  
318 et al., 2021). The advantage of employing this signal is the larger age limit, which presumably  
319 could cover the temporal range attributed to the Lower Palaeolithic sequence of the cave. For  
320 IR-RF measurements, we have employed the parameters' setting suggested by Frouin et al.  
321 (2017). The regenerative signals were measured after bleaching of ca 3 h (10,800s). To ensure  
322 this time sufficiently reset the natural IR-RF signal, we also carried out a bleaching test.

323 The  $D_{eS}$  for samples Koz11, Koz14, Koz16, Koz18, Koz20, and Koz21 were estimated through  
324 the approach of horizontal and vertical sliding (Kreutzer et al., 2017; Murari et al., 2018)  
325 implemented in *Luminescence::analyse\_IRSAR.RF()*.

### 326 **Violet stimulated luminescence (VSL)**

327 Unlike the relatively early saturation of the fast decaying blue-stimulated luminescence signal,  
328 some slow decaying components in quartz are deemed to be of higher saturation dose (Jain,  
329 2009). This makes them attractive for dating older sediment like the Lower Palaeolithic  
330 sequences. Thus, we have applied the SAR protocol suggested by Ankjærgaard et al., (2016)  
331 to estimate the  $D_e$  based on deep traps. This protocol was designed in a way that first the fast-  
332 decaying component is depleted using blue light stimulation and violet stimulation is then  
333 applied to probe the slow decaying signal with a higher saturation dose. We tested this  
334 technique on several samples (Koz8, 14, 16, 18, 21). However, the outcome of this application  
335 was not promising (all signals were in apparent saturation), therefore, we proceeded with the  
336 multiple aliquot additive dose (MAAD) protocol as an alternative solution to the standard SAR  
337 protocol offered in (Ankjærgaard et al., 2016). We used one of the youngest samples (Koz4)  
338 for which an independent age using the blue OSL technique was estimated and for which we  
339 had enough grains available to prepare a large number of aliquots.

340 The prepared samples were categorised into several fractions (each group comprising six  
341 aliquots) which were exposed to various amounts of beta irradiations on top of the natural dose.  
342 The first irradiative dose was set to 117 Gy and gradually it was increased to the dose of 2760

343 Gy. In the next step, following the approach of Ankjærgaard et al. (2016) we measured only  
344 the natural luminescence signal ( $L_n$ ) which was corrected by the test dose signal ( $T_n$ ) for each  
345 set of irradiated samples to minimise the impact of sensitivity change arising from repeated  
346 irritations and heating of the samples. Then the obtained L/T values were subsequently used to  
347 construct the MAAD dose-response curve fitted using a Lambert W function (Pagonis et al.,  
348 2020). Afterwards, the  $L_n/T_n$  ratios measured from the natural samples were used (six aliquots  
349 for each natural sample) to estimate the  $D_{eS}$  (see result section).

350

### 351 **3. Results**

#### 352 **3.1 Dose rate**

353 Table 2 shows the obtained concentrations of U, Th, and K. The equivalent U concentration  
354 was determined from the top (pre- $^{226}\text{Ra}$ ) and the bottom of the chain (post- $^{226}\text{Ra}$ ). This allows  
355 checking potential disequilibria in the U chain (Guibert et al., 2009). For samples Koz6, Koz7,  
356 Koz18, radioactive disequilibria were observed. To minimise this effect on the dose rates,  
357 instead of using the obtained values from the bottom of the chains (which were smaller than  
358 the top values), the average of the top and bottom concentrations was calculated and employed  
359 to estimate the dose rates for those samples (Table 3).

360

Table 2

361 The beta-dose rate for all samples ranged from  $0.86 \pm 0.04$  Gy/ka to  $1.70 \pm 0.08$  Gy/ka. The *in*  
362 *situ* and laboratory (i.e. from the concentrations of K, U and Th) gamma-dose rates ranged  
363 from  $0.19 \pm (<0.01)$  Gy/ka to  $0.72 \pm 0.01$  Gy/ka and from  $0.49 \pm 0.02$  Gy/ka to  $0.97 \pm 0.03$  Gy/ka,  
364 respectively. We have illustrated the two distinct series of gamma dose rates in Fig. 3. As  
365 shown in this figure, most *in situ* gamma-dose rates were significantly smaller than the  
366 corresponding gamma-dose rates obtained through means of gamma-ray spectrometry. It  
367 should be noted that we did not have a reliable estimation of the water content at sampling data,  
368 thus, it is likely that our estimation of the past water contents (varying between  $14 \pm 6\%$  to  $20 \pm 8$   
369  $\%$ ) is not in close alignment with the current water content. However, this does not explain the  
370 substantial discrepancies observed between the two dose rate results. It is more likely that the  
371 limestone pebbles and gravels that can readily identify in Fig. 2 caused a heterogeneous  
372 environment in which the *in situ* gamma dose rate values are smaller than the laboratory  
373 measurements. Therefore, we considered the *in situ* values more representative of the gamma  
374 dose rates received by the samples and used those values to determine the final dose rate.  
375 Particularly, because they cover gamma rays from larger distances and hence provide more

376 accurate estimates. The internal dose rate for quartz was set to  $0.06\pm 0.03$  Gy/ka and a value of  
377  $0.90\pm 0.14$  Gy/ka was obtained for K-feldspar. Cosmic dose rates varied between  $0.06\pm 0.01$   
378 Gy/ka to  $0.09\pm 0.01$  Gy/ka. In the end, the final dose rate for quartz and K-feldspar grains vary  
379 between  $1.21\pm 0.05$  Gy/ka and  $2.38\pm 0.09$  Gy/ka, and between  $2.05\pm 0.15$  Gy/ka and  $3.22\pm 0.16$   
380 Gy/ka, respectively.

381 Table 3

382 Fig. 3.

383

### 384 **3.2 Equivalent dose ( $D_e$ ) determination**

#### 385 **Blue-OSL**

386 Fig. S2 in the supplementary material illustrates with pseudo-LM OSL (Bos and Wallinga,  
387 2012) the presence of a fast-decaying signal at the beginning of the curve, but it also highlights  
388 at least contamination with another signal, at the end of the curve. Fig. S3 (true LM-OSL)  
389 revealed that presumably, the slow decaying signals (slow components) are also contributed to  
390 the luminescence signal.

391 We employed late background subtraction; the signal was measured using channels 1 to 3 (0.3  
392 s) and the background was set to 300 to 399 (10 s). The recycling ratios, (a representative value  
393 for expressing the sensitivity change after administrating the cycles of heating and irradiating)  
394 were within 20% of the unity for all samples. Furthermore, the recuperation signal ratio  
395 following the received zero dose was less than 5%. An IR depletion ratio test Duller (2003)  
396 was carried out at the end of the SAR protocol and the results were within 10% for all samples.  
397 Fig. S4 in the supplementary material depicts the outcome of the preheat test for sample Koz2.  
398 We considered  $260^\circ\text{C}$  and  $220^\circ\text{C}$  as acceptable temperatures for the preheat and cut-heat in  
399 our SAR protocol; using such elevated temperatures allows subtracting a late background by  
400 removing less stable components than the fast component (Jain et al., 2003). Dose recovery  
401 ratios for samples Koz2, Koz4 (given dose ca 48 Gy), Koz7 and Koz10 (given dose ca 83 Gy)  
402 resulted in  $1.02\pm 0.04$ ,  $1.01\pm 0.03$ ,  $0.96\pm 0.08$ ,  $0.91\pm 0.07$ , respectively. These values are  
403 consistent with 1 within uncertainties and fulfil the standard criterion (commonly accepted is  
404 within 10% of unity), indicating the suitability of the SAR protocol's parameters for our  
405 samples. However, the smooth reduction in these values alongside increasing the  $D_e$ s, indicates  
406 that reaching the non-linear part of the dose-response curve and may give rise to  $D_e$   
407 underestimation.

408 Fig. 4a and Fig. 5a show typical dose-response curves, and a visual representation of the  $D_e$   
409 distribution for sample Koz4 as an example. We have estimated the central dose for each

410 sample using the arithmetic average (as suggested for multi-grain OSL by (Guérin et al.,  
411 2017)), and its uncertainty was determined through the standard error of the mean. The  
412 estimated central doses ranged from  $48\pm 2$  Gy to  $117\pm 9$  Gy for samples Koz1 to Koz10 (see  
413 Table 4). We also measured blue OSL signals for subsequent samples, however, since we  
414 reached the saturation limit of the signal, further results were considered untrustworthy or at  
415 least underestimated.

#### 416 **IRSL<sub>50</sub> and pIR<sub>50</sub>IRSL<sub>290</sub>**

417 IRSL<sub>50</sub> signals for three samples were measured by integrating the first 6 s and subtracting the  
418 last 56 s as the background. The obtained *g*-values (measured for six aliquots per sample) for  
419 samples Koz4, Koz8 and Koz11 resulted in  $3.25\pm 0.04$  ( $3.52\pm 0.04$ , norm. 2 days) %/decade,  
420  $3.31\pm 0.04$  ( $3.58\pm 0.04$ , norm. 2 days) %/decade,  $3.35\pm 0.07$  ( $3.63\pm 0.07$ , norm. 2 days) %/decade  
421 (see Fig S5). The fading corrected *D<sub>e</sub>*s for these three samples were  $109\pm 1$  Gy,  $171\pm 4$  Gy,  
422  $512\pm 72$  Gy, respectively. The typical dose-response curves and *D<sub>e</sub>* distribution for sample  
423 Koz4 are illustrated in Fig. 4c and Fig. 5c, respectively.

424 The pIR<sub>50</sub>IRSL<sub>290</sub> signals were measured using the first 8 s and the background was subtracted  
425 using the last 60 s of the shine-down curves. The recycling ratios were within 10% of unity and  
426 recuperation ratios were less than 5%. The dose-response curves and the *D<sub>e</sub>* distribution for  
427 sample Koz4 are depicted in Fig. 4b and Fig. 5b, respectively. The *D<sub>e</sub>* representing the central  
428 equivalent dose for all samples was determined using the arithmetic average of *D<sub>e</sub>* values and  
429 its uncertainty was estimated using the standard error of the mean. The obtained *D<sub>e</sub>*s for our  
430 samples varied between  $95\pm 3$  (Koz2) to  $671\pm 36$  Gy (Koz14) (see Table 4). Due to the lack of  
431 K-feldspar grains for Koz15, we could not measure any signal, however, for the subsequent  
432 samples, the corresponding signals were mainly in or close to saturation and thus the outcome  
433 was not considered trustworthy.

434 Fig. 4.

436 Fig. 5.

#### 438 **Infrared-radiofluorescence (IR-RF)**

439 The result of the bleaching test on sample Koz11 is displayed in Fig. S6. This figure shows  
440 that a bleaching time of 10,800 s appears to be sufficient, lying within the plateau. The IR-RF  
441 signal was measured using a channel resolution of 10 s. The observed scatter in the points after  
442 reaching the plateau is probably induced by negligible movement of grains on the sample

443 carrier. Typical IR-RF signals for samples Koz11, Koz18, Koz21 are shown in Fig. 6. The  
444 natural and regenerative signals are illustrated with red and green, respectively. The horizontal  
445 and vertical sliding method was successfully applied to natural signal and an excellent match  
446 between natural signal and regenerative is observed. The  $D_e$  distributions for samples Koz11,  
447 Koz18, and Koz21 are represented in Fig. 7 using kernel density estimate (KDE) plot. The  $D_e$   
448 representing the central equivalent dose for each sample is estimated using the arithmetic  
449 average and its uncertainty was determined using the standard deviation.

450 Fig. 6.

451 Fig. 7.

### 454 **Violet stimulated luminescence (VSL)**

455 A typical dose-response curve alongside the natural and regenerative signals observed after  
456 applying the VSL (SAR) protocol is shown in Fig. S7 for one sample. Fig. S8 shows a smooth  
457 increase in the obtained  $L_n/T_n$  values as samples' depths are increased along with the profile.  
458 This gradual rise suggested that VSL has the potential to yield chronological information, as  
459 was already observed in (Ankjærgaard et al., 2016). Fig. 8 illustrates the L/T distribution  
460 obtained for each set of six aliquots prepared originally from Koz4; while the first set  
461 corresponds to the natural dose, all other sets were irradiated in the laboratory with increasing  
462 doses on top of the natural. We tested several fitting functions on our data set. Finally, as  
463 depicted in Fig. 8, we fitted a dose-response curve using a Lambert W function to our dataset,  
464 which seems to be more justified than other commonly fitting dose-response curve functions  
465 (Pagonis et al., 2020). Subsequently, the  $L_n/T_n$  obtained from the natural samples were  
466 projected onto the fitted curve (Fig. S9). Because of the nature of the MAAD approach, which  
467 adds doses on a natural sample that already has a dose, the dose-response curve necessarily  
468 intersects with the y-axis. Theoretically, one could attempt to estimate the equivalent dose for  
469 the first sample from the intersection of the curve with the x-axis. Due to the exponential  
470 character of the curve, this extrapolation comes at the cost of very large uncertainties; hence it  
471 was not used here. However, the  $L_n/T_n$  signal of Koz4 project onto the MAAD dose-response  
472 curve should result in an equivalent dose around 0. We estimated a value of  $11 \pm 12$  Gy for this  
473 sample, which is consistent with the zero dose within uncertainty. To estimate the equivalent  
474 dose of all other samples, the natural equivalent dose of sample Koz4 based on quartz blue  
475 OSL was added to all estimated results to determine the final  $D_e$  for each sample.

476

477

Fig. 8.

478 **3.3 Age estimation**

479 The obtained ages based on the various signals are presented in Table 4. The OSL dating for  
480 the uppermost sample (Koz1) belonging to layer 5a, exhibited an age of  $32\pm 2$  ka (one sigma).  
481 Gradually, the OSL ages increase smoothly with depth until layer 10b (sample Koz11), for  
482 which two aliquots out of ten were in saturation. Thus, we considered this age ( $141\pm 12$  ka) to  
483 be a minimum age. The corresponding ages for samples Koz11 and Koz12 estimated based on  
484  $pIR_{50}IRSL_{290}$  signals led to  $161\pm 13$  ka and  $199\pm 12$  ka (one sigma). Subsequently, only the OSL  
485 age for layer 10a ( $59\pm 4$  ka) and younger samples above were considered reliable (Fig. 9).

486 As shown in Fig. 9 (inset) (between layer 5a to layer 10a), the  $pIR_{50}IRSL_{290}$  ages were  
487 determined for a number of samples. Generally, at first glance, it can be concluded that the  
488  $pIR_{50}IRSL_{290}$  ages are older than the corresponding OSL ones. However, apart from one sample  
489 (Koz2 from layer 5b), for which no overlapping between the OSL and  $pIR_{50}IRSL_{290}$  ages is  
490 observed within two sigma ( $26\text{-}33$  ka for OSL and  $35\text{-}43$  ka for  $pIR_{50}IRSL_{290}$ ), the rest overlap  
491 and the age outcome from the two approaches can be generally evaluated consistently.

492 The IRSL ages which were fading-corrected were estimated for three samples. All of them are  
493 older compared to both, OSL and  $pIR_{50}IRSL_{290}$  ages. The obtained IRSL ages resulted in  $45\pm 3$   
494 ka and  $66\pm 4$  ka, respectively, for samples Koz4 (layer 6/7) and Koz8 (layer 9c). At 95%  
495 confidence, these two ages exhibit timeframes of  $39\text{-}51$  ka and  $58\text{-}75$  ka that overall are in  
496 alignment with the periods of  $33\text{-}41$  ka and  $48\text{-}74$  ka ascertained from  $pIR_{50}IRSL_{290}$ .  
497 Nevertheless, the IRSL age of sample Koz11 (layer 10b)  $250\pm 40$  ka is significantly larger than  
498 the corresponding  $pIR_{50}IRSL_{290}$  age of  $161\pm 13$  ka. The IR-RF age for this sample resulted in  
499  $201\pm 17$  ka; thus, it seems that the  $pIR_{50}IRSL_{290}$  age for this sample could be underestimated.  
500 The  $pIR_{50}IRSL_{290}$  ages increase in depth up to layer 10c. Afterwards, we have a clear indication  
501 of signal saturation (four aliquots out of six) for sample Koz16 (layer 11b). From this layer  
502 down to the lowermost layer, only IR-RF and VSL ages are available.

503 Besides the VSL ages for the oldest part of the sequence, several ages were also determined  
504 for the youngest samples. Although at first glance, the VSL ages for the subsequent samples  
505 Koz5, Koz6, and Koz7 do not seem consistent with the outcome of other dating techniques,  
506 their corresponding age intervals overlap one another (at two sigma).

507 Getting back to the older part of the sequence, we can see an excellent match between IR-RF-  
508 and VSL- based results,  $279\pm 29$  ka and  $324\pm 94$  ka, respectively for sample Koz14, layer 10c  
509 as an example. This picture does not hold for the older ages, however, still within two sigma,

510 both ages can be considered consistent, given the large error inherent for the VSL ages observed  
511 for all samples and related to the shape of the dose-response curve.

512 The IR-RF ages are more precise (i.e., they show smaller analytical error bars) and are less  
513 scattered. The VSL age estimated for the oldest sample Koz21 belonging to layer 13c in the  
514 profile was estimated at  $704 \pm 168$  ka. This age seems to be greater than the corresponding age  
515 based on IR-RF ( $387 \pm 28$  ka), although the large uncertainty obtained for the VSL age, makes  
516 a reasonable comparison difficult.

517 Table 4

518 Fig. 9.

519

## 520 4. Discussion

### 521 4.1 How confident are we about the obtained chronologies?

522 In the previous section, we presented ages based on different luminescence dating techniques.  
523 Since the age discrepancies observed between the various signals were not always negligible,  
524 it is worthwhile to critically reflect the ages and discuss reasons for these differences.

525 The IRSL ages estimated for all three samples led to older ages compared to other results. King  
526 et al. (2018) argued that employing a single saturating exponential fit can result in  
527 overestimated  $D_e$  dependent on the characteristic of the dose-response curve. Nevertheless, it  
528 should be noted that a single saturating exponential function perfectly described the dose-  
529 response curve for these three samples in this study. A general-order kinetic (GOK, Guralnik  
530 et al., 2015) also was tried for fitting sample Koz11 for which the  $D_e$  was not located in the  
531 linear part of the dose-response curve, and hence varying fitting functions could potentially  
532 lead to different outcomes. Applying this fitting resulted in a larger value compared to the  
533 single-saturating exponential function.

534 It is worth mentioning that the GOK can result in more reliable estimation only if the higher  
535 part of the dose-response curve is thoroughly defined by incorporating relatively large  
536 regenerative points (King et al., 2018). The largest regenerative dose used for generating the  
537 dose-response curve for Koz11 was only 600 Gy, while the individual  $D_e$  s of ca 600-700 Gy  
538 were obtained for this sample. This could justify why applying GOK resulted in a larger  $D_e$ ,  
539 and thus it was not a suitable function for our dataset. Having said that, even if the shape of the  
540 dose-response curve is considered problematic for Koz11, for younger samples, Koz4 and  
541 Koz8 this is not the case. Therefore, we can argue that it is more likely that the obtained  $g$ -  
542 values are not displaying the true measures and they are possibly overestimated. The proper

543 estimation of the fading rate in percentage per decade (the  $g$ -value) is a challenging task and  
544 minute variations in temperature and conditions can result in a very different estimate (cf.  
545 Kadereit et al. (2020) and the related public discussion). Although the reason behind this is  
546 not clear, it is likely that our estimation of the  $g$ -value, which is based on maximum pausing of  
547 only 4 h, was insufficient to properly determine the fading rate in the laboratory.

548 Previously in section 3.3, it was stated that  $pIR_{50}IRSL_{290}$  ages for the upper part of the sequence  
549 seemed to be slightly older than the OSL ages. However, the  $pIR_{50}IRSL_{290}$  signal may contain  
550 a residual dose (hard to bleach signal, Buylaert et al., 2011) contributing to the measured signal  
551 and giving rise to overestimated age. Due to the scarce amount of K-feldspar grains, we did  
552 not measure residual dose for our samples, however, in the literature, the amount of 5-20 Gy  
553 (e.g., Buylaert et al. 2011) was suggested as a reasonable measure. We calculated the residual  
554 doses for the upper part of the sequence assuming that the OSL and  $pIR_{50}IRSL_{290}$  ages agree  
555 with one another. The values ranged from 9 Gy to 29 Gy, consistent with the values suggested  
556 by Buylaert et al. (2011). Subtracting this range from the obtained  $pIR_{50}IRSL_{290}$   $D_e$  resulted in  
557 the ages in excellent accordance with the corresponding quartz OSL ages. Thus, the  
558 discrepancies observed earlier between these two series of ages might be related to  
559 ‘unbleachable’ residual doses.

560 Furthermore, taking into account the slow bleaching rate of the  $pIR_{50}IRSL_{290}$  signal compared  
561 to the OSL signal (Murray et al., 2012), we can safely conclude that the sediment samples were  
562 well-bleached before burial. This inference is also supported from the sedimentological  
563 perspective since the original nature of the sediments in the cave was identified as loess-  
564 derivatives and wind-blown loess (Muttoni et al., 2017). Although some scholars have argued  
565 that these sediments should be categorised as loess-like and not (primary) loess (Jipa, 2014).  
566 In addition, the bleaching rate of IR-RF is also considered similar to  $pIR_{50}IRSL_{290}$  (Frouin et  
567 al., 2017), hence the IR-RF ages do not suffer from an age overestimation as already observed  
568 by an overall concordance between the  $pIR_{50}IRSL_{290}$  and IR-RF ages.

569 The OSL and  $pIR_{50}IRSL_{290}$  ages increase gradually along the profile until layer 10a.  
570 Afterwards, due to the saturation, no OSL age was reported. Heydari and Guérin, (2018)  
571 demonstrated that the Bayesian models implemented in BayLum (Guérin et al., 2021; Philippe  
572 et al., 2019) can lead to a more accurate dose estimation when the signal is close or in  
573 saturation, compared to ‘classical analyses’ using the Analyst software. Thus, in the future, it  
574 might be worth applying Bayesian modelling on single grain measurements for samples older  
575 than layer 10a and comparing the outcomes with other independent ages available based on  
576 different signals.

577 The  $pIR_{50}IRSL_{290}$  ages for layer 10b resulted in  $161\pm 13$  ka (Koz11) and  $199\pm 12$  ka (Koz12).  
578 The corresponding ages for sample Koz11 using IRSL, VSL and IR-RF yielded  $250\pm 40$  ka,  
579  $321\pm 77$  ka,  $201\pm 17$  ka, respectively, all older than  $pIR_{50}IRSL_{290}$ . Thus, the  $pIR_{50}IRSL_{290}$  age  
580 for sample Koz11 appears to be underestimated. Albeit the first argument which immediately  
581 springs to mind is the possibility of fading, such conjecture was not derived from the rest of  
582 the samples, and it does not appear plausible here.

583 For the next layer (10b), the  $pIR_{50}IRSL_{290}$  age ( $199\pm 12$  ka) with a  $D_e$  of  $416\pm 16$  Gy is in  
584 excellent accordance with the corresponding age using the IR-RF signal ( $201\pm 17$  ka). The same  
585 trend is observed for layer 10c where  $pIR_{50}IRSL_{290}$  resulted in  $281\pm 19$  ka whereas IR-IF  
586 exhibited  $279\pm 29$  ka.

587 The  $pIR_{50}IRSL_{290}$  ages estimated for layer 10c ( $D_e$  of 659-671 Gy) were the oldest age obtained  
588 using this signal. Afterwards, for the lower layer (11b), measurements clearly show evidence  
589 for signal saturation (four aliquots out of six). On the other hand, IR-RF measurement for layer  
590 11b resulted in  $D_e$  of  $888\pm 23$  Gy. Therefore, the interval between  $671\pm 36$  Gy (the highest  $D_e$   
591 estimated based on  $pIR_{50}IRSL_{290}$ ) and  $888\pm 23$  Gy can be considered as a saturation level for  
592 the  $pIR_{50}IRSL_{290}$  signal which is consistent with other findings (e.g., Yi et al., 2015). Although  
593 in the literature ca. 1,000 Gy was suggested as a threshold for IR-RF (Erfurt and Krbetschek,  
594 2003), comparing this age with the VSL might imply that the signal saturation for IR-RF is  
595 around  $923\pm 23$  Gy ( $D_e$  of oldest sample in layer 13c). By looking at the chronological results  
596 from layer 11b to the lowermost layer (13c), it is not inconceivable to speculate that we already  
597 gradually reached a saturation level IR-RF signal at even above layer (11b) with the  $D_e$  of  
598  $888\pm 23$  Gy. Comparing VSL ages determined for these layers with IR-RFs, could even  
599 reinforce this possibility although the large errors presented for VSL ages do not allow for a  
600 very precise and decisive conclusion.

601 More importantly, Muttoni et al. (2017) established a palaeomagnetism sequence for the  
602 Kozarnika cave suggesting the layer immediately after 13c lies in the Brunhes-Matuyama  
603 boundary with the timing of 780 ka. They also attributed layers 13a-c to layers L6 to L7 in the  
604 stratigraphical Danube loess-palaeosol presented in Marković et al. (2015) (their Fig. 16) and  
605 the link allows them to speculate that a period of 600-750 ka (early Brunhes) might represent  
606 the timeframe for layers 13a-c. Indeed, the silt fraction becomes more and more abundant from  
607 layers 11a to 13b, and the Kozarnika area is included in Pontic East European loess (domain  
608 V) (see Lehmkuhl et al., 2021). On the other hand, there is no loess near Kozarnika (see  
609 Evlogiev, 2007). The closest loess deposit is reported in distances from 60 km to 100 km of

610 the cave location. Furthermore, silt remobilized in the karst might be considered loess-like  
611 (alluvial and aeolian), which extends more in the west than the loess (see for example Jipa,  
612 2014 or the “lössartige und andere Lehme” of Fotakieva & Minkov, 1966). The sediment  
613 accumulation of the lower layers of Kozarnika is endocarstic and therefore discontinuous, with  
614 gaps posing difficulties to any quantification. Therefore, we cannot invalidate the hypothesis  
615 of sedimentary lacks under the magnetic “event” at the lower part of layer 11b (around -6.50  
616 m) as identified by Kovacheva et al. (in Guadelli et al., 2005) and Muttoni et al. (2017). This  
617 might place the Brunhes-Matuyama limit at the base of layer 11b and not in layer 13, which is  
618 in agreement with the macro-mammals assemblages of layer 11b (Guadelli et al., 2005, in  
619 press; Fernandez and Crégut-Bonnoure, 2007; Sirakov et al., 2010).

620 In either way, this independent chronological data (palaeomagnetism sequence) can be seen as  
621 a strong argument supporting the saturation assumption of IR-RF for layer 13. The VSL age of  
622  $704 \pm 168$  ka for layer 13c, agrees with the period of 600-750 ka, however, only due to the very  
623 large uncertainty. Apart from that, the agreement of the VSL ages with other independent ages  
624 obtained for the Kozarnika sequence tends to confirm the overall accuracy of the VSL ages,  
625 which is consistent with the observation of Ankjærgaard et al. (2016).

626 These large uncertainty margins, resulting from the leverage effect of the signal uncertainty  
627 caused by the flat dose-response curve, are the main drawback of the VSL application.  
628 However, this might be ameliorated by increasing the number of measured samples to specify  
629 a more precise probability density distribution after projecting  $L_n/T_n$ s onto the dose-response  
630 curve. This may lead to more confidence in the  $D_e$  estimates in future studies.

#### 631 **4.2 Unravelling the timing of humans in the cave**

632 The Kozarnika cave contains three main assemblages associated with the Lower, Middle, and  
633 Upper Palaeolithic periods. For the Upper Palaeolithic sequence, some radiocarbon dates are  
634 available (Guadelli et al., 2005, in press). Although some of these results were provided nearly  
635 two decades ago (new procedures have been suggested since then for sample pre-treatments to  
636 improve the accuracy and precision of radiocarbon dates e.g., Wood, 2015), comparing these  
637 dates as independent ages with luminescence-based ages is still advantageous. The uppermost  
638 luminescence-age based on OSL technique resulted in  $32 \pm 2$  ka for layer (5a), and  $30 \pm 2$  ka for  
639 the layer below (layer 5b). The available radiocarbon age for layer 5b exhibited 31.2 ka cal.  
640 BP (original data from Guadelli et al. (2005) and calibrated from Muttoni et al. (2017))  
641 showing an excellent concordance with OSL ages within uncertainties. New radiocarbon ages

642 fall in the range of 29,219-30,030 ka cal. BP to 31,919-33,130 ka cal. BP for layer 5a and  
643 36,152-37,276 ka cal. BP to 36,987-39,875 ka cal. BP for layer 5b (Guadelli et al., in press).  
644 Following that, layer 5c represents the radiocarbon range of 40-44 ka cal. BP (original data  
645 from Guadelli et al., (2005). These results agree with the new radiocarbon ages suggesting a  
646 range of 39,998-41,592 ka cal. BP to 42,085-44,541 ka cal. BP (Guadelli et al., in press).  
647 Although there is no luminescence age available for this layer, the age of the layer immediately  
648 beneath (6/7) yielded  $33\pm 2$  ka and  $35\pm 2$  ka. Sequentially the radiocarbon dates for this layer  
649 ranged from 42,194-45,025 ka cal. BP to 43,243-54,492 ka cal. BP (Guadelli et al., in press)  
650 which is significantly older than the OSL ages.  
651 The OSL ages for the Upper Palaeolithic assemblages (Kozarnikian) uncovered in layer 5a is  
652 in good concordance with the radiocarbon age, however the discrepancies between the OSL  
653 and radiocarbon ages for layers 5b and 5c (Guadelli et al. in press) are significant. One possible  
654 scenario that could explain the discrepancies between the outcome of two distinct dating  
655 methods could be the scatter between OSL  $D_e$ s for each sample (see Fig. 5a as an example). In  
656 such a case implementing signal grain measurements which allows for properly applying a  
657 broad body of statistical models could be preferable.  
658 Layer 6/7 with OSL ages of  $33\pm 2$  ka and  $35\pm 2$  ka and radiocarbon age range of 42-54 ka cal.  
659 BP was considered an intermediate level between the Middle and Upper Palaeolithic  
660 (Kozarnikian) containing artefacts associated with both affinities. Still, this does not  
661 necessarily imply the mixture of Middle and Upper affinities but rather a concatenation of  
662 successive and/or alternately rapid occupations (see Guadelli et al., in press for more details).  
663 Although such a situation seems infrequent, it has been already observed for example at  
664 Mandrin cave (France) or at Buran Kaya III (Crimea) (Silmak et al., 2017).  
665 The layer after 9ab is attributed to the termination of the Middle Palaeolithic period rendering  
666 the Mousterian Levallois elements. The OSL age of  $43\pm 3$  ka established for layer 9ab within  
667 uncertainties agrees with the range of 41,030–39,260 ka cal. BP representing the termination  
668 of the Mousterian industry after Higham et al. (2014). Heydari et al. (2020) used Bayesian  
669 modelling specifically designed for treating OSL samples to reduce age uncertainties attributed  
670 to Palaeolithic assemblages. In particular, they combined OSL ages with radiocarbon dates for  
671 one Palaeolithic site and obtained chronologies that were more precise (Heydari et al., 2021).  
672 Thus, applying Bayesian modelling to treat luminescence ages in combination with  
673 radiocarbon dates seems advantageous in this context to improve the precision for the  
674 continuous or discontinuous transition timeframe from the Middle to Upper Palaeolithic period.

675 A remarkable finding in layer 10b was a radius of an infant attributed to a Neanderthal, which  
676 earlier was dated between  $128 \pm 13$  ka and  $183 \pm 14$  ka using OSL and pIR<sub>50</sub>IRSL<sub>290</sub> techniques  
677 (Tillier et al., 2017). Nevertheless, in the present study, we acknowledge that the OSL age of  
678  $128 \pm 13$  ka is underestimated, and we suggest an age of  $201 \pm 17$  ka, based on IR-RF, revealing  
679 older deposit of the Neanderthal infant's skeleton. This result confirms the new  
680 biochronological interpretation of the middle sequence of Kozarnika cave (Guadelli et al., in  
681 press). Nevertheless, a stark contrast for the oldest layers exists between the biochronological  
682 framework according to micromammals (Popov and Marinska, 2007) and the one based on  
683 large mammal species (Guadelli et al., 2005; Fernandez and Crégut-Bonnoure, 2007; Sirakov  
684 et al., 2010).

685 The Lower Palaeolithic industries were found from layer 11a downwards. The luminescence  
686 dating suggested the onset of the Middle Palaeolithic period in the timeframe of  $279 \pm 29$  ka  
687 (IR-RF)  $324 \pm 94$  ka (VSL), which is perfectly in line with what we expect from other  
688 chronologies established for Middle Palaeolithic assemblages (Richter, 2011).

689 The youngest ages attributed to the Lower Palaeolithic assemblages in layer 11b were  $337 \pm 23$   
690 ka (IR-RF), and  $419 \pm 137$  ka (VSL) and the oldest ages for layer 13c exhibited  $387 \pm 28$  ka (IR-  
691 RF) and  $704 \pm 168$  ka (VSL). Although here luminescence dating is nearly at its age limit, taking  
692 into account the smooth increase in the obtained ages along with the profile, does not seem to  
693 support a period of 1,400-1,600 ka estimated using large mammal biochronology (Sirakov et  
694 al., 2010) for layers 13a-c or even the revisited biochronological estimation of 1,200/1,000-  
695 1,400/1,200 ka for the layer 12 to layers 13a-c suggested by Guadelli et al. (in press).  
696 Surprisingly, the maximum age of 780 ka also questions the biochronology result for the Lower  
697 Palaeolithic sequence of the Kozarnika cave (Brunhes-Matuyama boundary) proposed for layer  
698 13c using palaeomagnetic dating (Muttoni et al., 2017). At this stage, it seems that further  
699 chronological work is required to reach a decisive conclusion for the oldest layers of the  
700 sequence.

## 701 **5. Conclusions**

702 A complete chronology for the Kozarnika cave was established using various luminescence-  
703 dating techniques. Combining different approaches allowed us to successfully date sediments  
704 from ca 30 ka to 700 ka unravelling the timing of human occupations during a large part of the  
705 Palaeolithic. Nevertheless, at this stage, we cannot rule out the possibility of underestimated  
706 timeframes for the layers containing the Lower Palaeolithic assemblages.

707 The obtained ages rely on the measurement of different signals for each sample to increase our  
708 confidence about the reliability of the outcomes. The overall accordance between the OSL with  
709 the pIR<sub>50</sub>IRSL<sub>290</sub> ages for the upper part of the sequence makes it likely that both signals were  
710 completely bleached before mineral deposition. The same conclusion was deduced for the IR-  
711 RF signal, negating the possibility of age overestimation due to the slow beaching rate that  
712 often impedes the application of this method.

713 This study indicates that the saturation dose for the pIR<sub>50</sub>IRSL<sub>290</sub> signal (at around 700 Gy to  
714 900 Gy) led to ages of 291±22 ka and 281±19 ka as the oldest ages obtained in Kozarnika using  
715 this signal. On the other hand, the IR-RF saturated later, which enabled the age limit to be  
716 extended until 387±28 ka for this sequence. Moreover, the IR-RF doses measured on samples  
717 from layers 11a to 13c are indistinguishable, which probably indicates signal saturation. In that  
718 case, a dose of ~900 Gy might correspond to the field saturation limit of IR-RF for our samples.  
719 The IRSL ages were deemed to overestimate other ages presumably due to an inaccurate *g*-  
720 value estimate. Applying the VSL technique in combination with MAAD protocol resulted in  
721 ages that were consistent with other signal-based ages and implied the trustworthiness of  
722 outcomes. The drawback of this application was large uncertainty margins, which could be  
723 ameliorated by increasing the number of measured samples, and hence improving the precision.  
724 More crucially, Muttoni et al. (2017) assigned the Brunhes–Matuyama reversal (780 ka) to the  
725 layer beneath 13c using Palaeomagnetic dating. Subsequently, a timeframe of 600–750 ka was  
726 suggested for layers 13a–c (Muttoni et al., 2017) by correlating layers 13a–c to the Danube loess  
727 sequence layers L6–L7 (Marković et al., 2015). Thus, the obtained VSL age for layer 13c within  
728 error is consistent with that period. However, the large uncertainty on this VSL age renders a  
729 precise interpretation difficult. In addition, the independent age available for layers 13a–c (with  
730 the range of 600–750 ka) also might indicate that the IR-RF ages represent the minimum age  
731 for these layers.

732 Employing various luminescence signals in this study led to determining the timing of the  
733 human inhabitants of Kozarnika cave. The Kozarnikian assemblages discovered in layers 5a–c  
734 dates back to 32±2 ka and 33±2 ka. Generally, radiocarbon dates (Guadelli et al., 2005, in  
735 press) provided for Kozarnikian assemblages in layers 5a–c and 6/7 are older compared to  
736 luminescence dating.

737 This study revealed that the onset of the Middle Palaeolithic period happened at 279±29 ka  
738 (IR-RF) 324±94 ka (VSL) and it lasted until between 49±4 ka and 43±3 ka. The lower  
739 Palaeolithic assemblages in Kozarnika cave dated back to between 337±23 ka (IR-RF) and

740 704±168 ka (VSL). However, due to the supposed limit of luminescence dating in this dose  
 741 range (setting our upper datable age limit), it is likely that they present minimum ages.  
 742 Therefore, further chronological investigations using dating methods with extended age limit  
 743 are required to shed light on the reliability of these ages and to further constrain the timing of  
 744 the humans who lived during the Lower Palaeolithic period in the Kozarnika cave.

745 Table 1. Location of the sediment samples taken from the site.

Sample name	Squares	X	Y	Z	Geological layers
Koz1	H7	199.000	1498.900	-4.259	5a
Koz2	H7	199.000	1498.580	-4.570	5b
Koz3	H7	199.000	1498.333	-4.609	5c
Koz4	H6	199.000	1497.632	-4.695	6/7
Koz5	H8	199.000	1499.560	-4.549	6/7
Koz6	H8	199.000	1499.590	-4.744	9ab
Koz7	H7	199.000	1498.580	-4.787	9ab
Koz8	H7	199.000	1498.730	-4.954	9c
Koz9	H8	199.000	1499.560	-4.896	9c
Koz10	H7	199.000	1498.610	-5.078	10a
Koz11	F8	196.000	1499.119	-5.232	10b
Koz12	F7	196.000	1498.325	-5.310	10b
Koz13	H7	199.000	1498.770	-5.259	10c
Koz14	F8	196.000	1499.122	-5.396	10c
Koz16	F8	196.000	1499.107	-5.880	11b
Koz18	G11	197.419	1503.000	-7.437	12
Koz20	G11	197.473	1503.000	-7.551	13a
Koz21	H11	198.000	1502.000	-8.000	13c

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749 Table 2. The concentration of radionuclides obtained through gamma-ray spectrometry.

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Sample name	K (%)	U pre- <sup>226</sup> Ra (ppm)	U post- <sup>226</sup> Ra (ppm)	Th (ppm)
Koz1	1.58±0.02	1.93±0.08	1.97±0.02	6.83±0.07
Koz2	1.48±0.02	1.17±0.07	1.24±0.02	4.75±0.05
Koz3	1.54±0.02	1.19±0.07	0.99±0.02	4.95±0.06
Koz4	1.47±0.02	1.13±0.07	1.33±0.02	5.26±0.06
Koz5	1.44±0.02	1.11±0.07	1.29±0.02	4.72±0.06
Koz6	2.15±0.02	2.08±0.08	1.64±0.02	7.92±0.07
Koz7	2.06±0.02	1.92±0.07	1.55±0.02	7.63±0.06
Koz8	1.41±0.02	1.14±0.05	1.32±0.01	4.84±0.05
Koz9	1.37±0.02	1.24±0.07	1.13±0.02	4.78±0.05
Koz10	1.35±0.02	1.21±0.07	1.17±0.02	4.95±0.05
Koz11	1.13±0.01	0.88±0.06	0.89±0.01	4.08±0.05
Koz12	1.20±0.01	0.92±0.06	0.91±0.01	4.30±0.05
Koz13	1.29±0.02	1.16±0.07	1.11±0.01	5.64±0.05
Koz14	1.34±0.01	1.25±0.05	1.17±0.01	5.75±0.05

Koz16	1.41±0.01	1.67±0.06	1.39±0.01	6.97±0.06
Koz18	1.60±0.02	1.61±0.07	1.06±0.01	6.47±0.06
Koz20	1.47±0.02	1.75±0.08	1.59±0.02	7.78±0.07
Koz21	1.50±0.02	1.78±0.07	1.76±0.02	8.25±0.07

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Table 3. The estimated dose rates for sediment samples in this study.

Sample name	Water content (WF %)	Laboratory Gamma (Gy/ka)	<i>in situ</i>			Total D <sub>r</sub> Quartz (Gy/ka)	Total D <sub>r</sub> K-Feldspar (Gy/ka)
			Gamma (Gy/ka)	Beta (Gy/ka)	Cosmic (Gy/ka)		
Koz1	15±6	0.81±0.03	0.31±0.02	1.31±0.06	0.09±0.01	1.77±0.07	2.61±0.15
Koz2	14±6	0.63±0.02	0.34±0.01	1.13±0.06	0.09±0.01	1.62±0.06	2.45±0.15
Koz3	14±5	0.63±0.02	0.39±0.02	1.17±0.06	0.09±0.01	1.70±0.07	2.54±0.15
Koz4	15±6	0.66±0.02	0.31±0.02	1.14±0.06	0.09±0.01	1.60±0.07	2.44±0.15
Koz5	14±5	0.63±0.02	0.38±0.01	1.12±0.05	0.09±0.01	1.65±0.06	2.48±0.15
Koz6	14±5	0.97±0.03	0.54±0.01	1.70±0.08	0.09±0.01	2.38±0.09	3.22±0.16
Koz7	15±6	0.92±0.03	0.59±0.01	1.61±0.08	0.09±0.01	2.34±0.09	3.18±0.16
Koz8	14±5	0.63±0.02	0.50±0.00	1.11±0.05	0.09±0.01	1.75±0.06	2.59±0.15
Koz9	14±6	0.60±0.02	0.39±0.00	1.06±0.05	0.09±0.01	1.60±0.06	2.44±0.15
Koz10	15±6	0.60±0.02	0.33±0.01	1.05±0.05	0.09±0.01	1.53±0.06	2.36±0.15
Koz11	15±6	0.49±0.02	0.20±0.00	0.86±0.04	0.09±0.01	1.21±0.05	2.05±0.15
Koz12	15±6	0.52±0.02	0.19±0.00	0.92±0.05	0.09±0.01	1.25±0.06	2.09±0.15
Koz13	14±5	0.62±0.02	0.25±0.01	1.03±0.05	0.08±0.01	1.43±0.06	2.26±0.15
Koz14	14±6	0.64±0.02	0.35±0.00	1.06±0.05	0.08±0.01	1.56±0.06	2.39±0.15
Koz16	20±8	0.68±0.03	0.57±0.01	1.09±0.07	0.08±0.01	1.80±0.07	2.64±0.15
Koz18	20±8	0.70±0.03	0.72±0.01	1.19±0.08	0.07±0.01	2.04±0.08	2.87±0.16
Koz20	20±8	0.74±0.03	0.50±0.03	1.16±0.07	0.07±0.01	1.79±0.08	2.62±0.16
Koz21	20±8	0.78±0.04	0.23±0.01	1.20±0.07	0.06±0.01	1.55±0.08	2.39±0.16

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Table 4. The D<sub>e</sub> and corresponding ages within uncertainties estimated using various luminescence signals.

Sample names	OSL		pIR <sub>50</sub> -IRSL <sub>290</sub>		IRSL		VSL		IR-RF	
	D <sub>e</sub> (Gy)	Age (ka)	D <sub>e</sub> (Gy)	Age (ka)	D <sub>e</sub> (Gy)	Age (ka)	D <sub>e</sub> (Gy)	Age (ka)	D <sub>e</sub> (Gy)	Age (ka)
Koz1	57±2	32±2								
Koz2	48±2	30±2	95±3	39±2						
Koz3	56±3	33±2								
Koz4	53±1	33±2	90±3	37±2	109±1	45±3				
Koz5	58±2	35±2								
Koz6	117±9	49±4	176±5	55±3			101±35	42±15		
Koz7	101±6	43±3	162±6	51±3			96±21	41±9		
Koz8	92±4	53±3	158±15	61±6	171±4	66±4	166±119	95±68		
Koz9	82±4	51±3	138±3	57±3						
Koz10	90±5	59±4	168±4	71±4						

Koz11	330±22	161±13	512±72	250±40	390±91	321±77	411±18	201±17
Koz12	416±16	199±12						
Koz13	659±39	291±22						
Koz14	671±36	281±19			504±144	324±94	668±56	279±29
Koz16					755±245	419±137	888±23	337±23
Koz18					768±227	377±113	835±20	290±18
Koz20							906±17	346±23
Koz21					1092±253	704±168	923±23	387±28

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#### 790 **Author Contributions**

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792 Conceptualization: Maryam Heydari, Guillaume Guérin, Jean-Luc Guadelli

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794 Data curation: Maryam Heydari

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796 Formal analysis: Maryam Heydari

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798 Funding acquisition: Maryam Heydari, Jean-Luc Guadelli

799

800 Investigation: Maryam Heydari, Guillaume Guérin, Nikolay Sirakov, Philippe Fernandez,

801

802 Catherine Ferrier, Aleta Guadelli, Jean-Claude Leblanc, Stanimira Taneva, Svoboda

803

804 Sirakova, Jean-Luc Guadelli

805

806 Methodology: Maryam Heydari, Guillaume Guérin

807

808 Project administration: Jean-Luc Guadelli, Guillaume Guérin

809

810 Software: Maryam Heydari, Jean-Luc Guadelli

811

812 Supervision: Jean-Luc Guadelli, Guillaume Guérin

813

814 Visualization: Maryam Heydari, Jean-Luc Guadelli

815

816 Roles/Writing - original draft: Maryam Heydari

817

818 Writing - review & editing: Maryam Heydari, Jean-Luc Guadelli, Guillaume Guérin, Philippe

819

820 Fernandez

#### 808 **Figure captions**

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810 Fig. 1. The Kozarnika cave is located in the northern hills of the Balkan Mountains, ca 30 km  
811 south of the Danube River, in the northwest of Bulgaria.

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813 Fig. 2. Profile picture of the cave where sediment samples were taken from. The obtained  
814 luminescence ages according to different techniques are presented here.

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Fig. 3. Comparison between the *in situ* and laboratory (gamma-ray spectrometry) gamma dose rate estimates. The *in situ* results are significantly lower compared to the laboratory measurements due to the heterogeneous environment observed in the sediment layers. Since limestone fragments from the bedrock are the source of radiation heterogeneities (limestone is generally poor in radioelements), this systematic discrepancy is expected. Thus, *in situ* measurements are more representative of the sediment burial conditions.

Fig. 4. Illustrating the dose-response curves for the OSL, pIR<sub>50</sub>IRSL<sub>290</sub>, and IRSL measurements derived from sample Koz4.

Fig. 5. D<sub>e</sub> distributions generated from OSL (A) pIR<sub>50</sub>IRSL<sub>290</sub> (B) and IRSL<sub>50</sub> (C) measurements for sample Koz4 as an example using Abanico plot (Dietze et al., 2016).

Fig. 6. Typical IR-RF curves were obtained for three samples. Each plot represents results for one multiple grain aliquot. The natural signal is indicated by red-filled circles and the regenerated signal by green crosses. The horizontal and vertical sliding was used to estimate the D<sub>e</sub> for each subsample.

Fig. 7. The kernel density estimate (KDE) plots for three samples as examples to show typical D<sub>e</sub>s distributions.

Fig. 8. VSL MAAD dose-response curve. The L/T s for each set of irradiated samples are shown in this figure. For the fitting, we applied a dose-response curve function based on the Lambert W function. The value of 11±12 Gy was obtained using this fitting for sample Koz4 (not irradiated), which within uncertainties is consistent with zero.

Fig. 9. The age-depth plot for all samples in this study covering the period of ca 30 ka to 700 ka using various luminescence signals. To provide better insight into the ages from Koz1 to Koz10 they are depicted in the inset.

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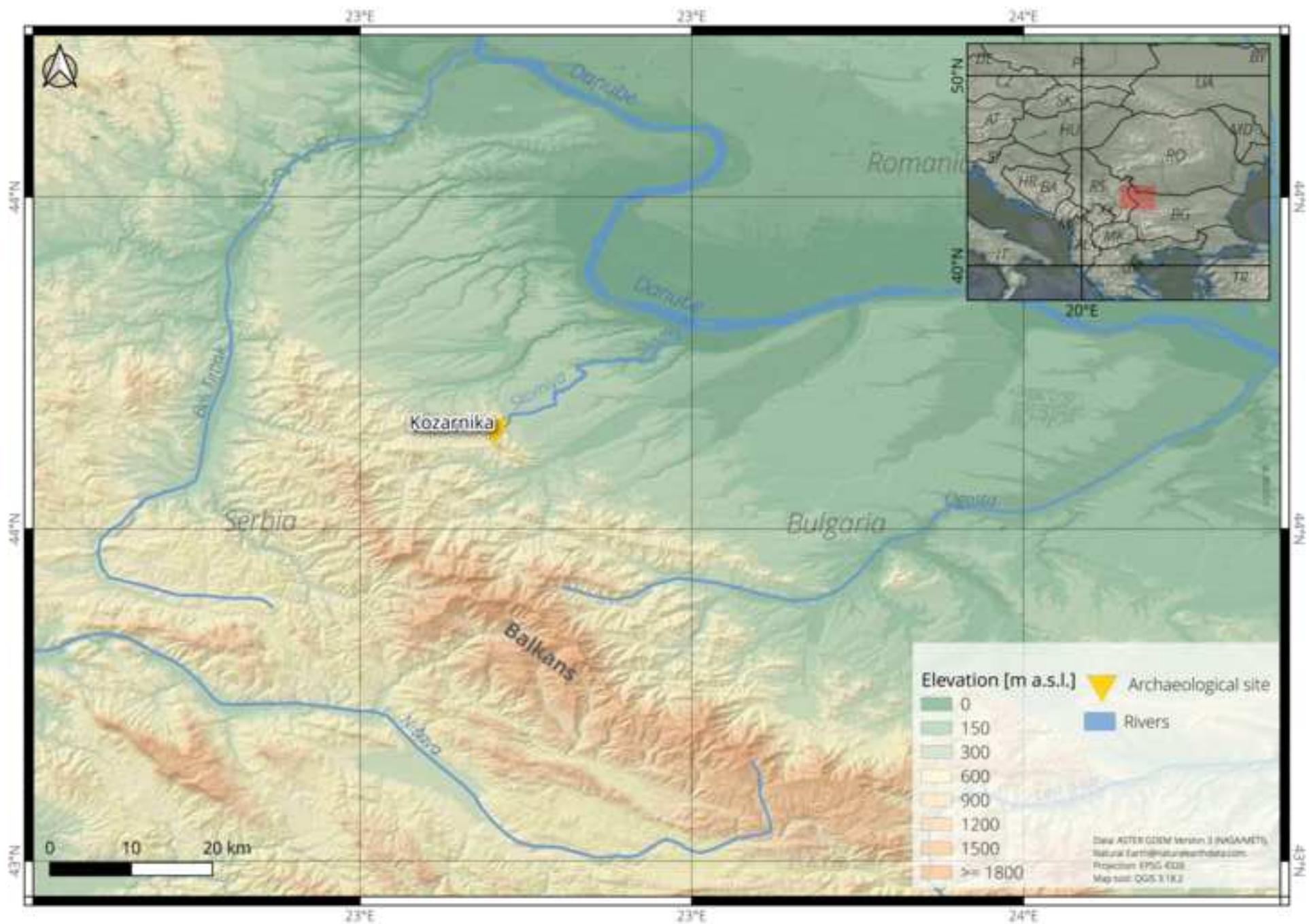
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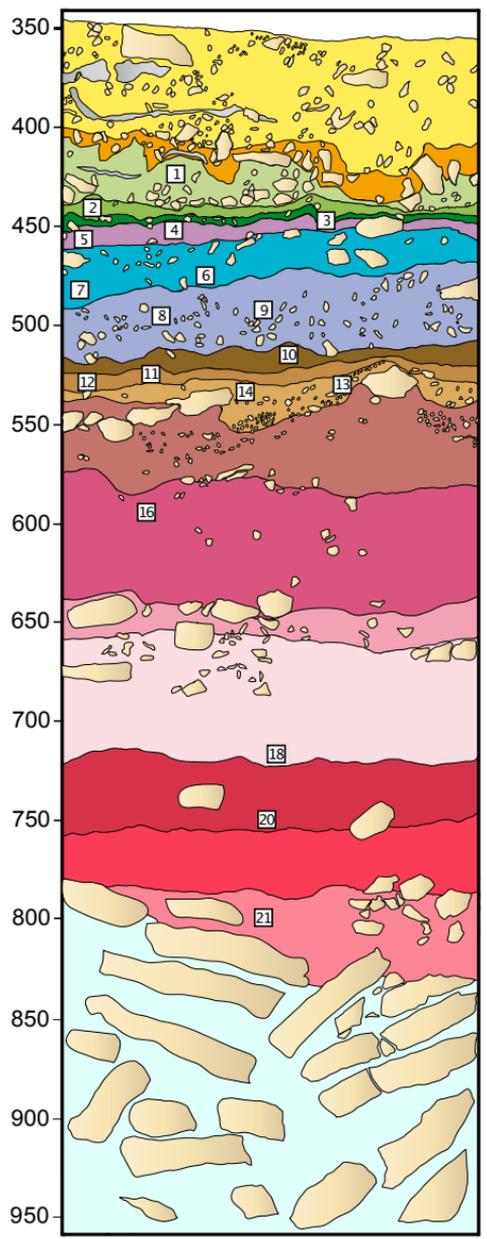
Figure

[Click here to access/download;Figure;Figure1.png](#)



Figure

Click here to access/download:Figure;Figure2.pdf



**OSL**

- Koz [1] 32±2 ka
- Koz [2] 30±2 ka
- 3a Koz [3] 33±2 ka
- 3b Koz [4] 33±2 ka
- 4 Koz [5] 35±2 ka
- 5a Koz [6] 49±4 ka
- 5b Koz [7] 43±3 ka
- 5c Koz [8] 53±3 ka
- 6/7 Koz [9] 51±3 ka
- 9ab Koz [10] 59±4 ka
- 9c
- 10a
- 10b
- 10c

**IR-RF**

**VSL**

**pIRIR**

- Koz [11] 201±17 ka
- 11a Koz [12] 161±13 ka
- Koz [13] 199±12 ka
- Koz [14] 279±29 ka
- 11b Koz [16] 337±23 ka
- 11c
- 12
- Koz [18] 290±18 ka
- 13a Koz [20] 346±23 ka
- 13b
- 13c Koz [21] 387±28 ka

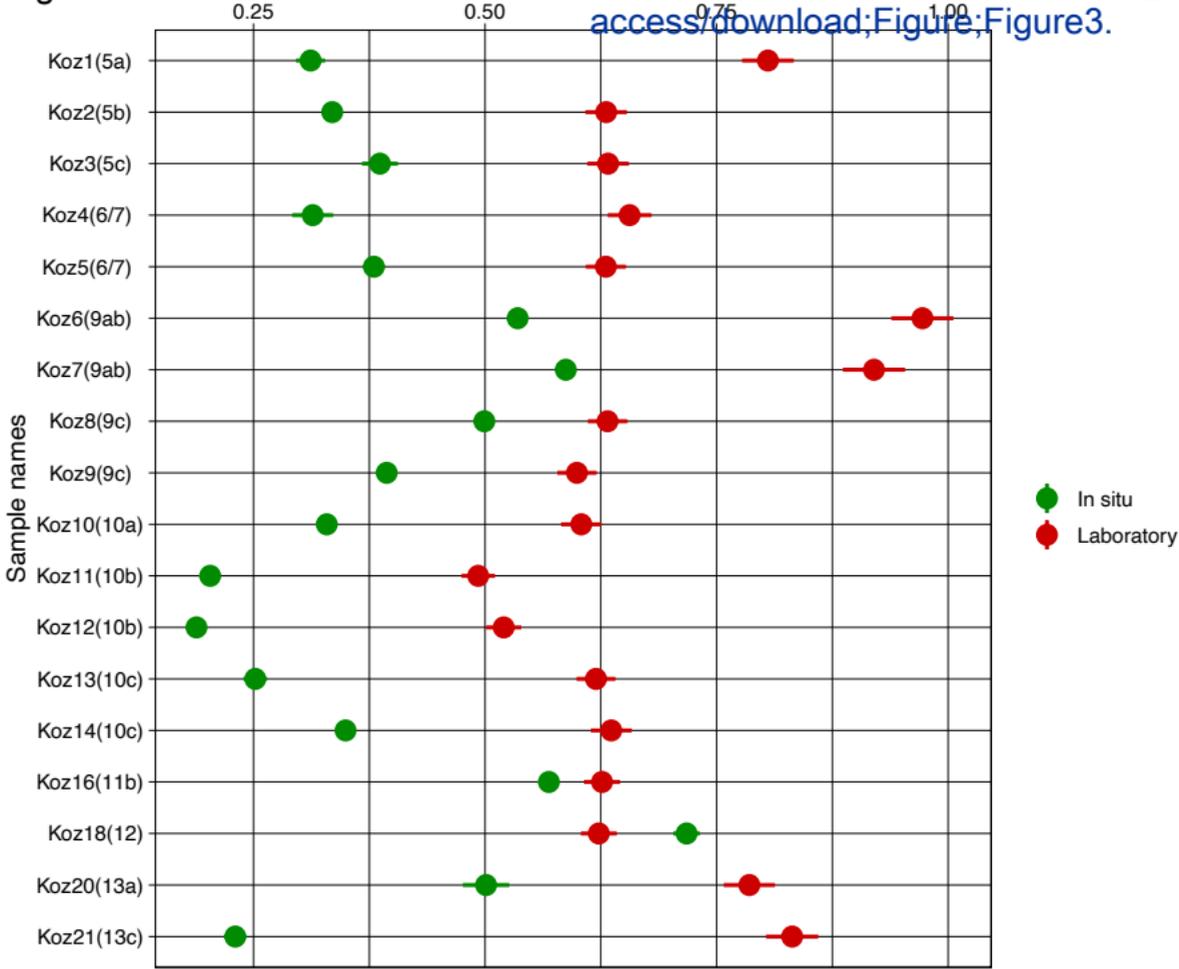
- 321±77 ka
- 161±13 ka
- 199±12 ka
- 291±22 ka
- 281±19 ka
- 419±137 ka
- 377±113 ka
- 704±168 ka

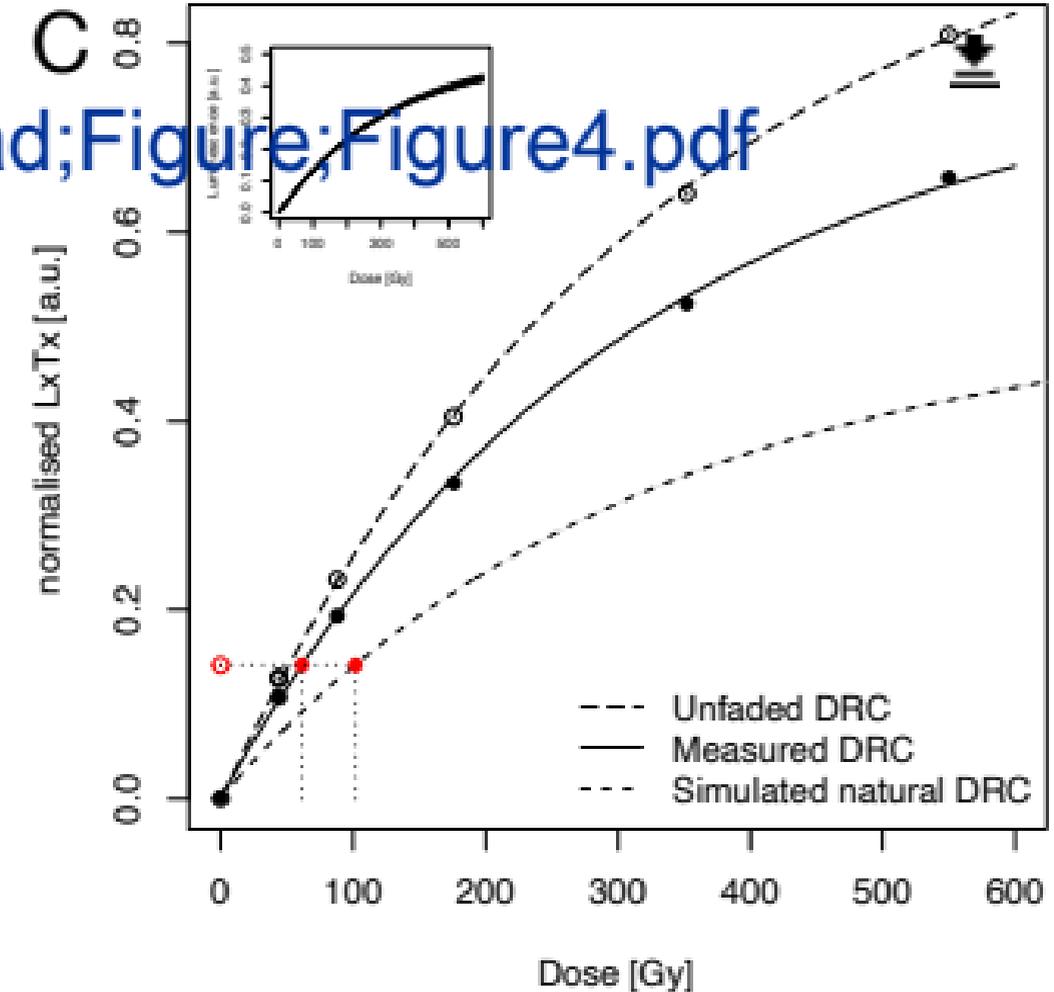
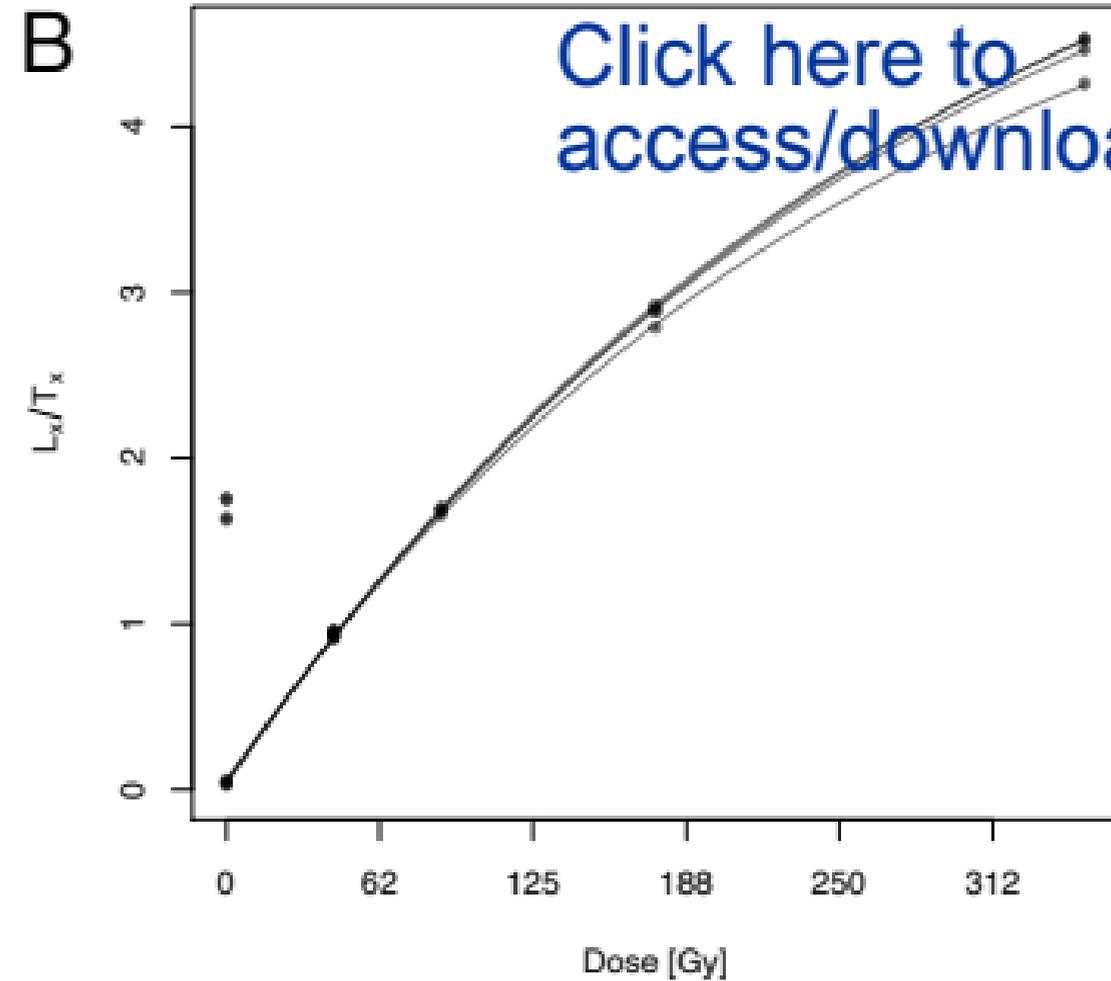
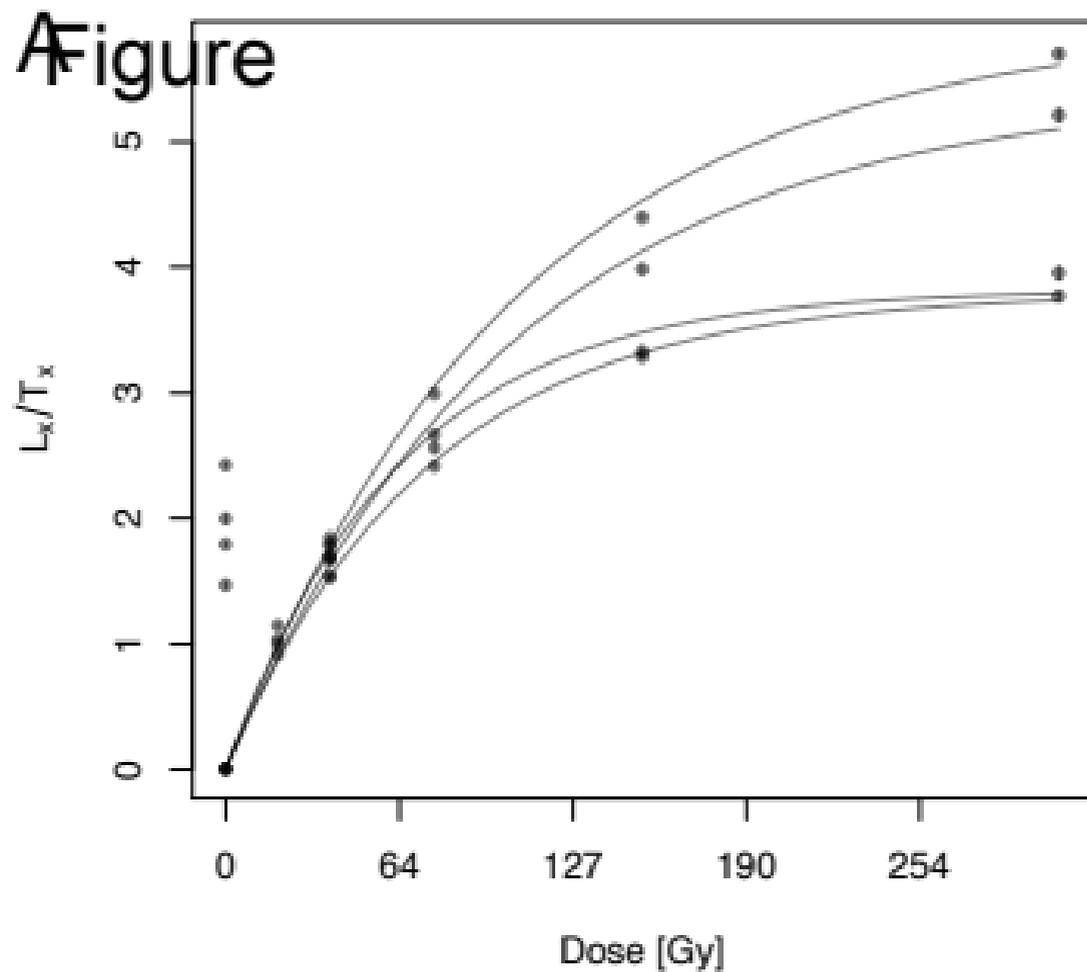
14, 15 & 16

Charcoal and burnt bones X Sediment samples  
Limestone blocks

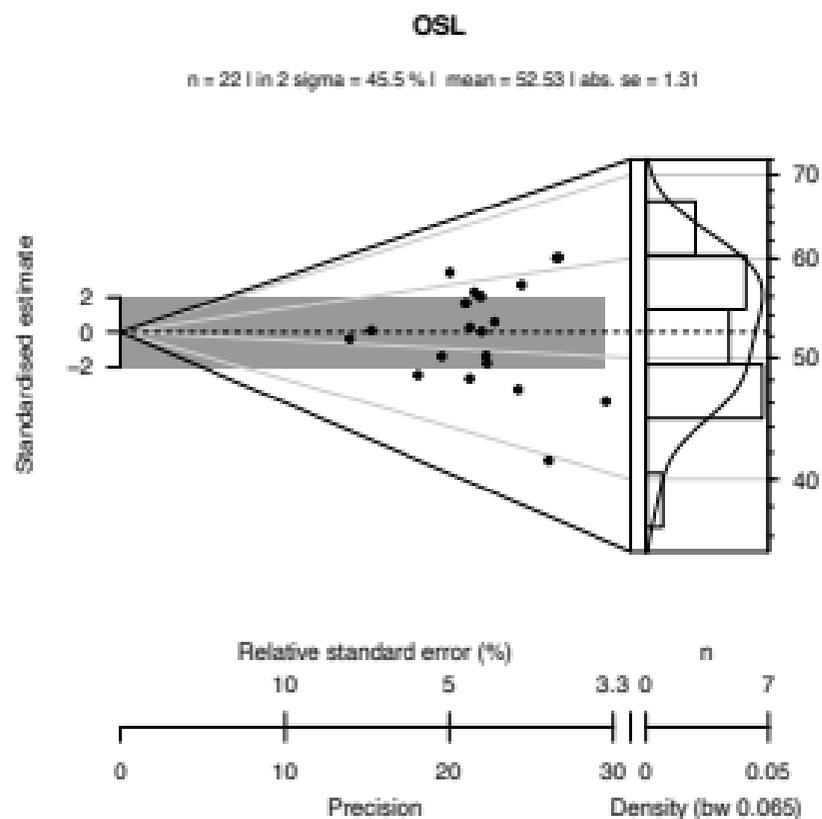
# Figure

Gamma dose rate (Gy/ha)

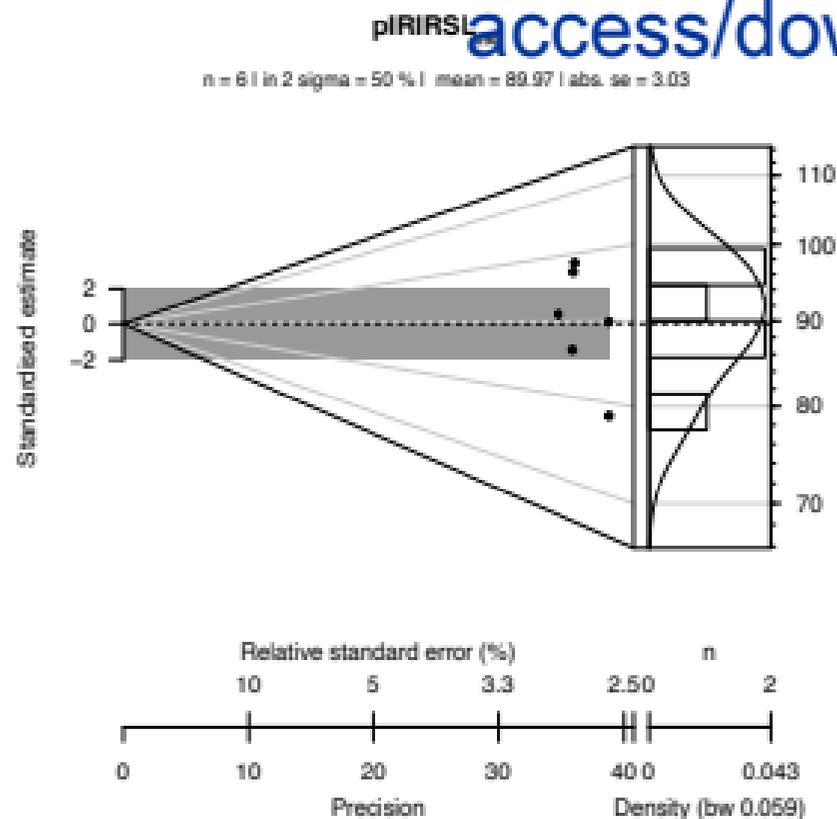




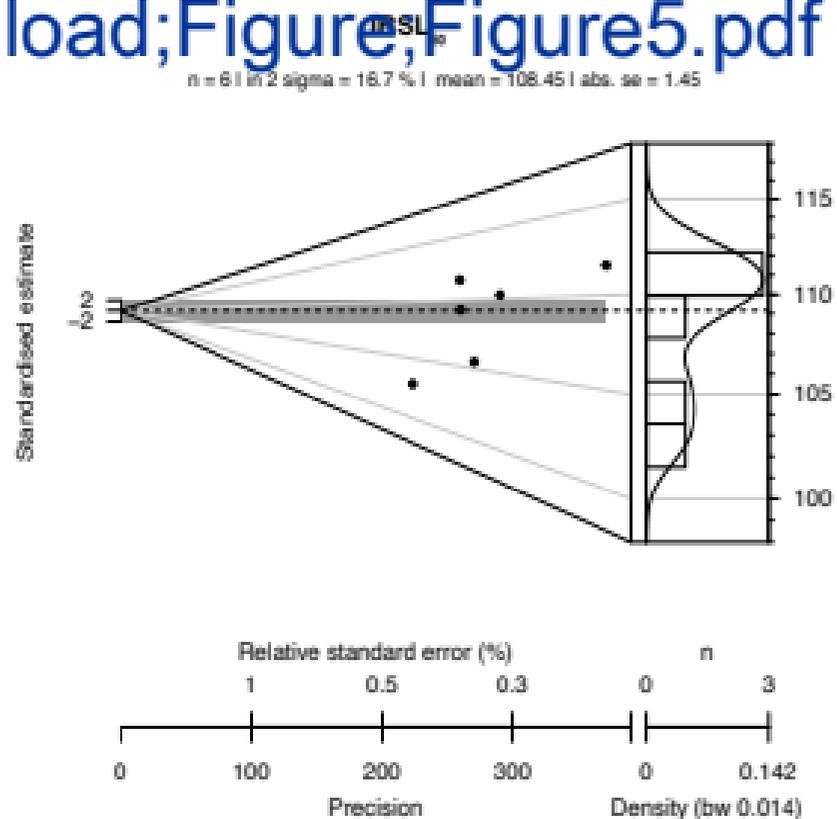
# Figure



# B

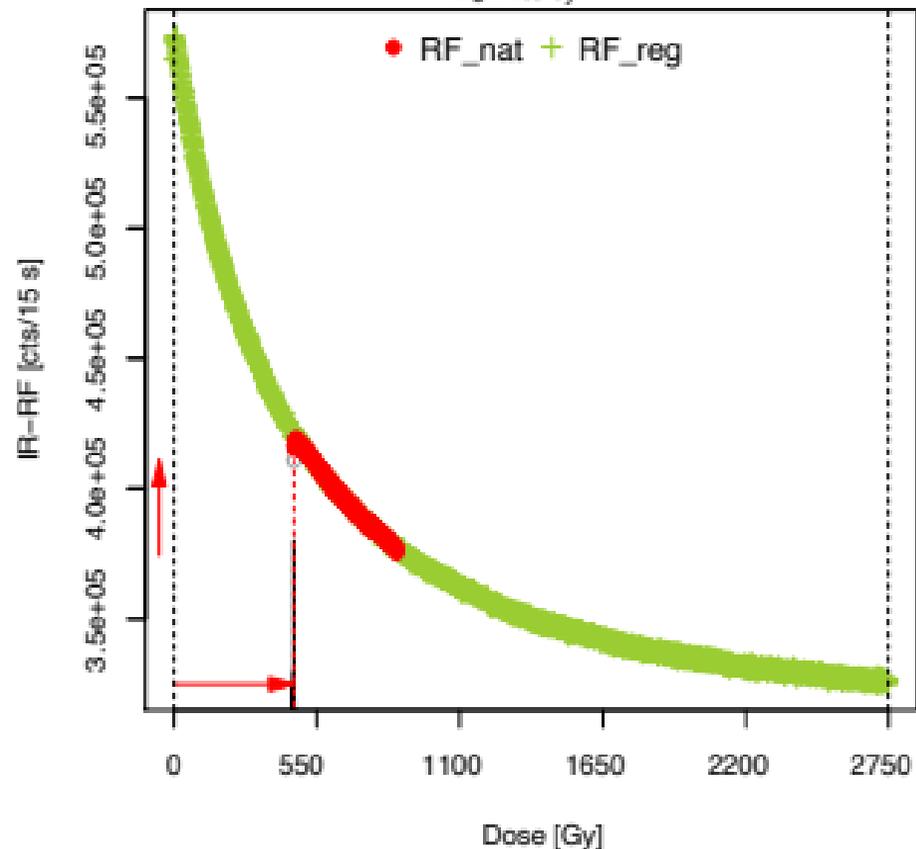
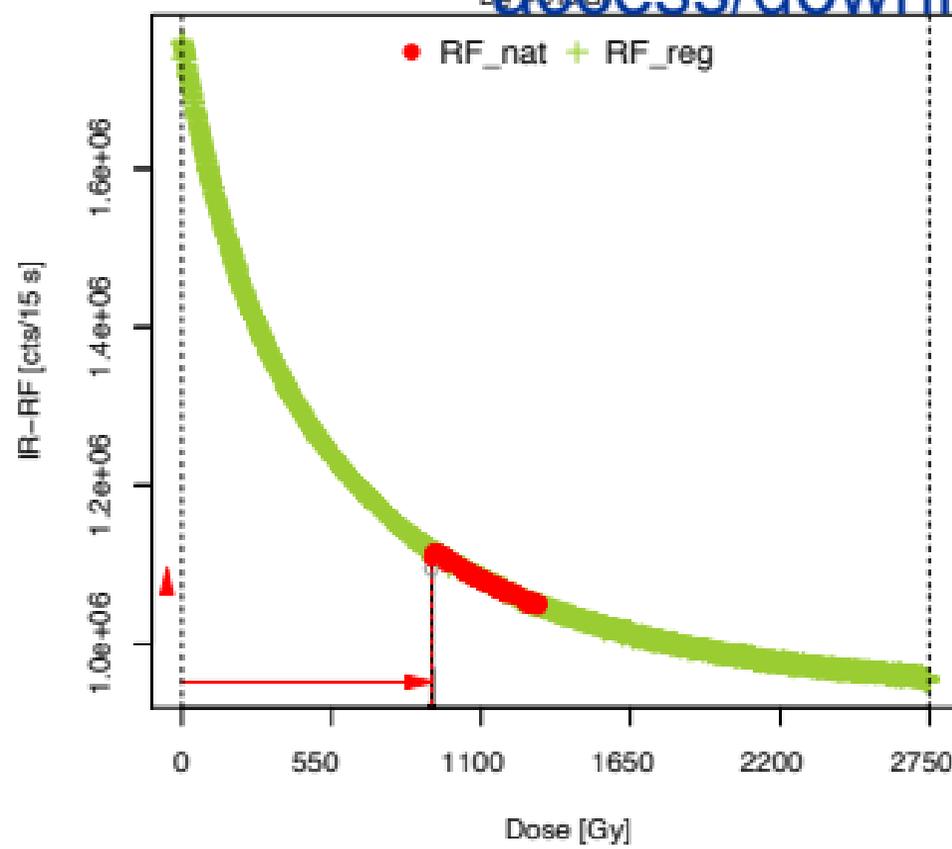
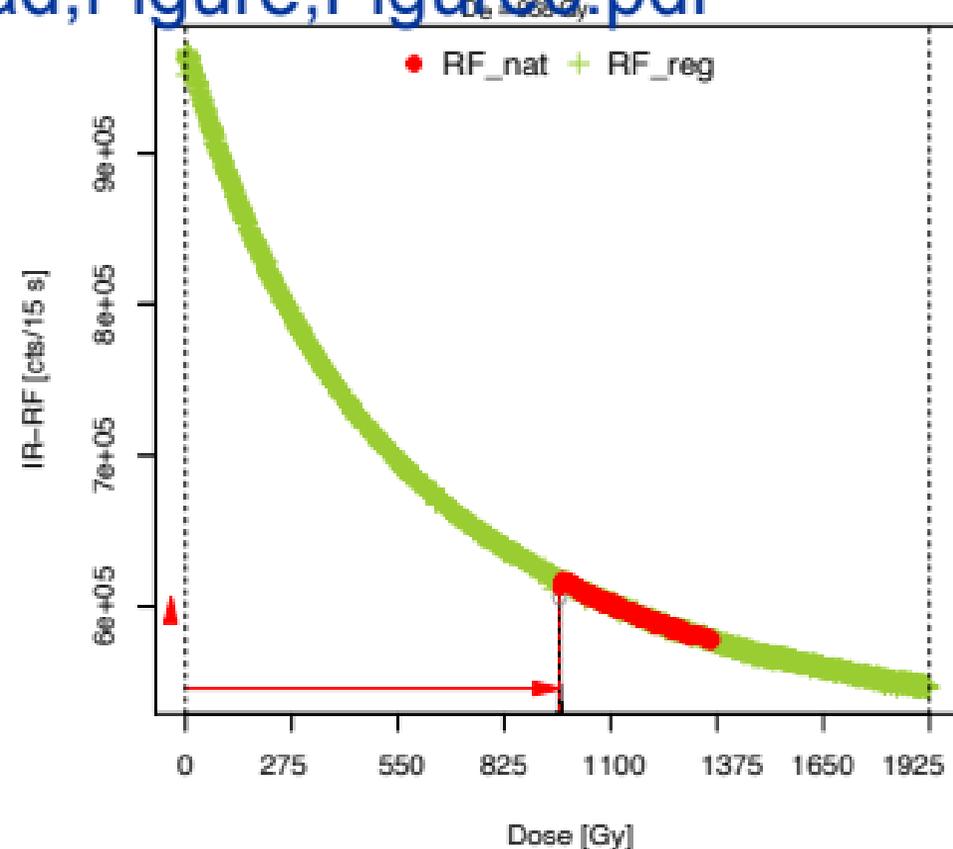


# C



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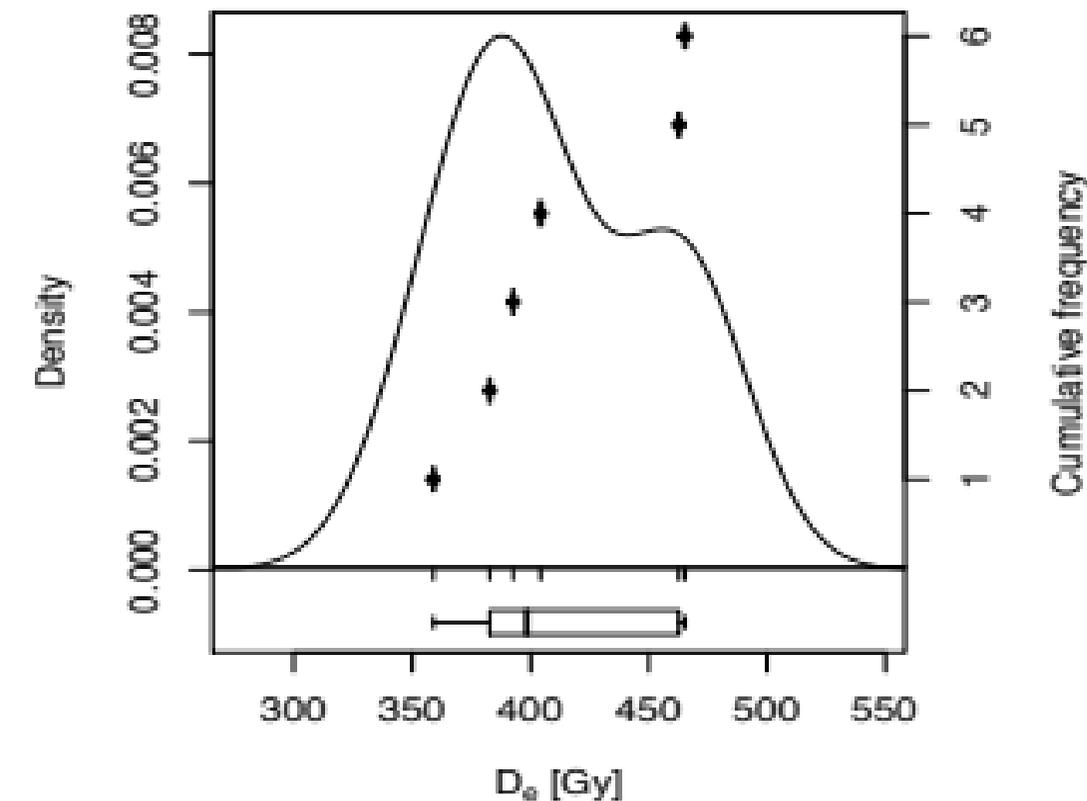
**Figure****Koz 11** $D_e = 463 \text{ Gy}$ **B****Koz** $D_e = 818 \text{ Gy}$ **C****Koz 21** $D_e = 920 \text{ Gy}$ 

[Click here to access/download;Figure;Figure6.pdf](#)



**A** Figure Koz 11

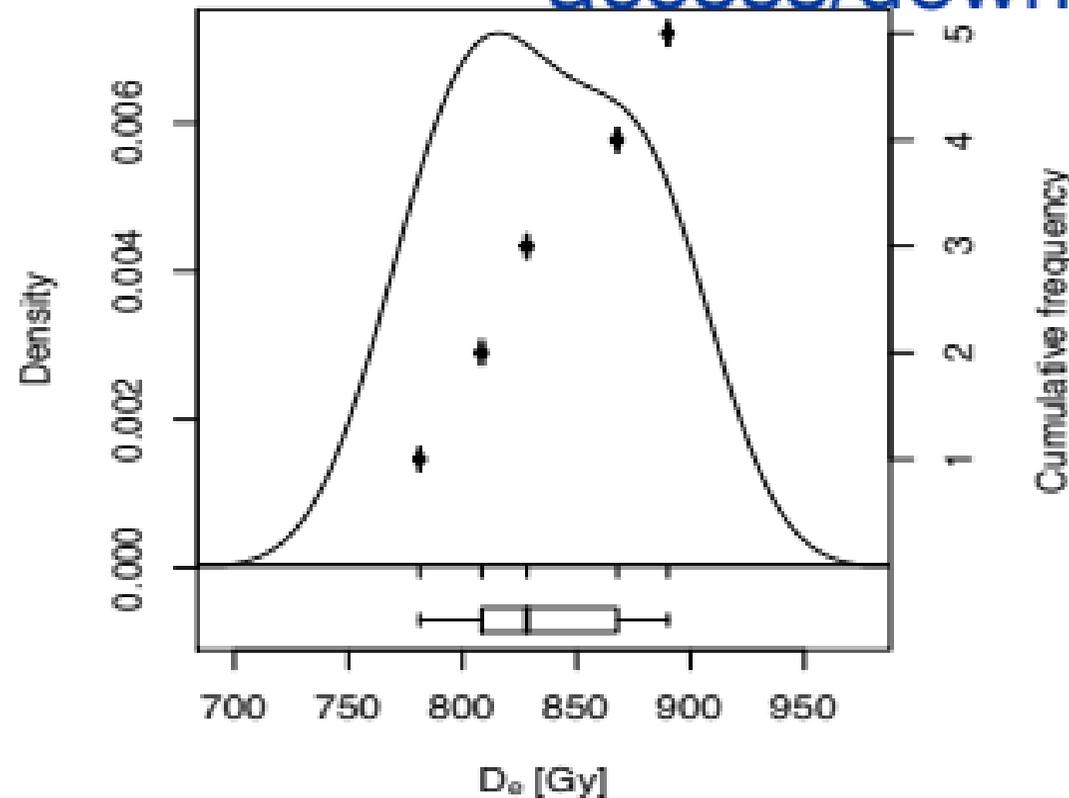
n = 6 | mean = 411.12 | abs. sd = 43.65



**B**

Koz 18 [Click here to access/download;Figure7.pdf](#)

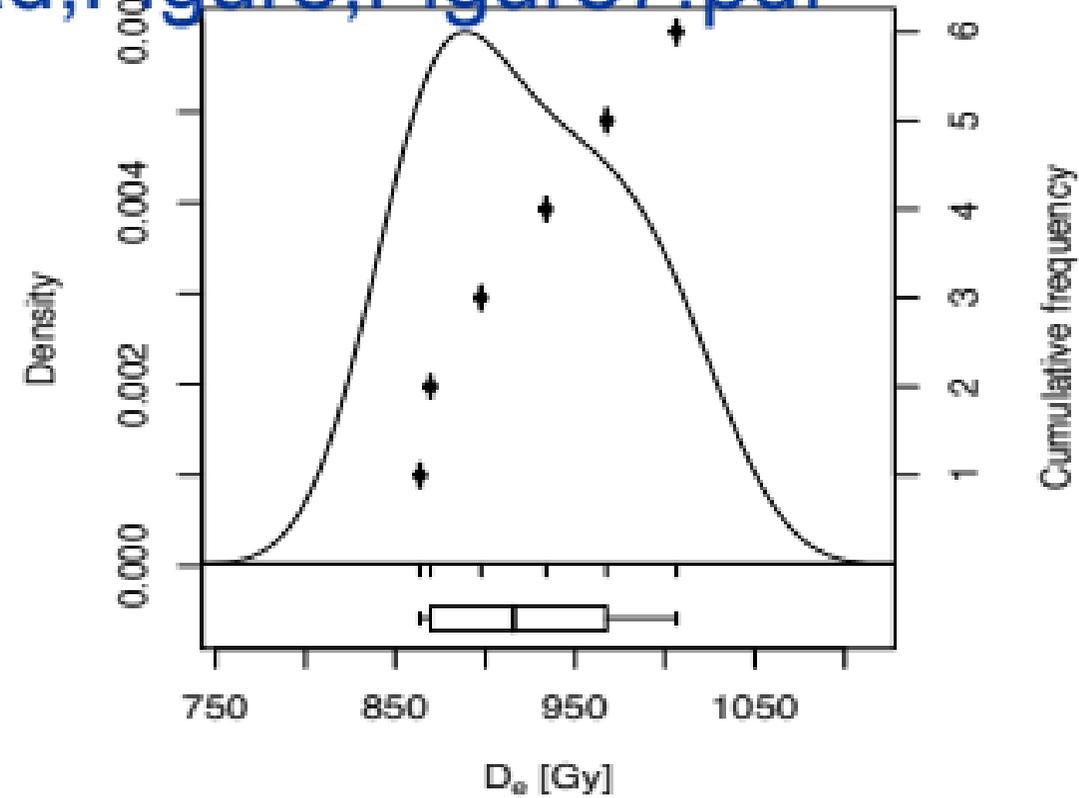
n = 5 | mean = 835.22 | abs. sd = 44.05

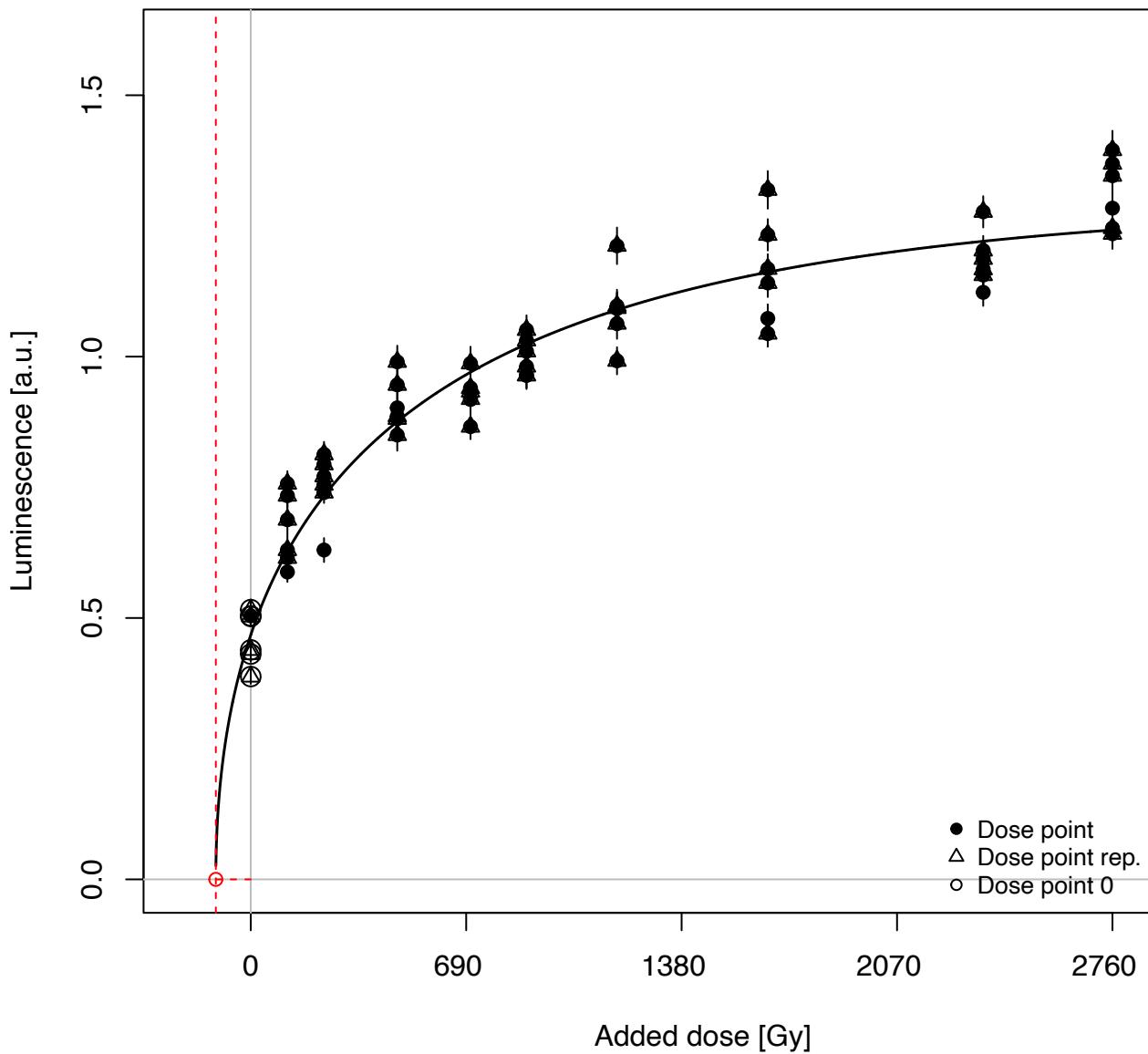


**C**

Koz 21 [Click here to access/download;Figure7.pdf](#)

n = 6 | mean = 923.77 | abs. sd = 56.75





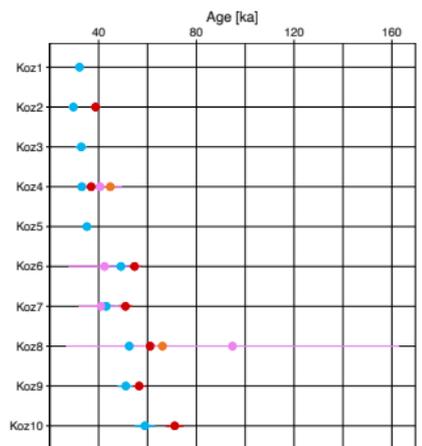
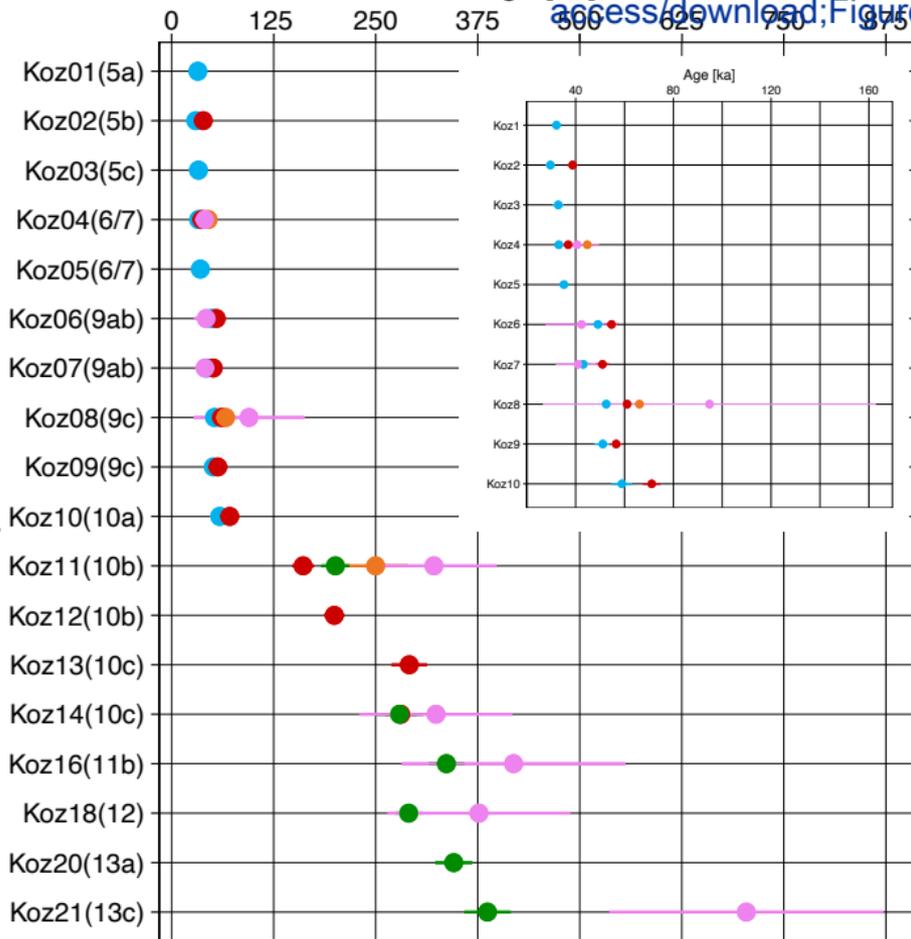
# Figure

Age [ka]

[Click here to access/download;Figure;Figure9.p](#)



Sample names



Signal

- OSL
- IRSL
- pIRIRSL
- IR-RF
- VSL