Testing polymineral post-IR IRSL and quartz SAR-OSL protocols on Middle to Late Pleistocene loess at Batajnica, Serbia

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The loess–palaeosol sequence of Batajnica (Vojvodina region, Serbia) is considered as one of the most complete and thickest terrestrial palaeoclimate archives for the Middle and Late Pleistocene. In order to achieve a numerical chronology for this profile, four sets of ages were obtained on 18 individual samples. Equivalent doses were determined using the SAR protocol on fine (4–11 μm) and coarse (65–90 μm) quartz fractions, as well as on polymineral fine grains by using two elevated temperature infrared stimulation methods, pIRIR225 and pIRIR250. We show that the upper age limit of coarse quartz OSL and polymineral pIRIR290 and pIRIR225 techniques is restricted to the Last Glacial/Interglacial cycle due to the field saturation of the natural signals. Luminescence ages on coarse quartz, pIRIR225 and pIRIR290 polymineral fine grains are in general agreement. Fine quartz ages are systematically lower than the coarse quartz and pIRIR ages, the degree of underestimation increasing with age. Comparison between natural and laboratory dose response curves indicate the age range over which each protocol provides reliable ages. For fine and coarse quartz, the natural and laboratory dose response curves overlap up to ~150 and ~250 Gy, respectively, suggesting that the SAR protocol provides reliable ages up to c. 50 ka on fine quartz and c. 100 ka on coarse quartz. Using the pIRIR225 and pIRIR290 protocols, equivalent doses up to ~400 Gy can be determined, beyond which in the case of the former the natural dose response curve slightly overestimates the laboratory dose response curve. Our results suggest that the choice of the mineral and luminescence technique to be used for dating loess sediments should take into consideration the reported limited reliability.

The Middle Danube basin comprises several high-resolution Middle to Late Pleistocene loess–palaeosol sequences (LPS) that document clear similarity in the long-term palaeoclimate response between European and Asian loess deposits (Buggle et al. 2009, 2013; Marković et al. 2015 and references therein; Sümeği et al. 2018; Perić et al. 2019). This similarity allowed for inferring common variability in regional atmospheric circulation patterns and dust dynamics driven by glacial-to-interglacial climate variability (e.g. Marković et al. 2015; Zeeden et al. 2018a). However, reliable numerical age constraints for Middle Danube LPS are rare (Stevens et al. 2011; Thiel et al. 2011a; Murray et al. 2014; Böskén et al. 2017; Zhang et al. 2018; Perić et al. 2019). Therefore, the most common approach in deriving chronological frameworks for Middle Danube LPS has relied on corelative techniques based on variability in magnetic susceptibility data (e.g. Marković et al. 2006, 2015, 2018a; Basarin et al. 2014; Zeeden et al. 2018a). Magnetic susceptibility, a proxy for pedogenetic intensity in loess (Schäelti et al. 2018; Zeeden et al. 2018b), allows the tentative alignment of loess records onto the benthic isotope stack (Lisiecki & Raymo 2005) or the insolation curve (Laskar et al. 2004).

Luminescence dating is the most applicable method in directly dating the emplacement time of loess-forming mineral particles (Roberts 2008). To date, luminescence dating has been applied on quartz and feldspars extracted from Serbian LPS (Fuchs et al. 2008; Stevens et al. 2011; Timar-Gabor et al. 2015; Perić et al. 2019) mainly for records covering the Last Glacial Cycle (LGC). Beyond the LGC, only Murray et al. (2014) have reported minimum pIRIR290 ages for samples found to be in field and laboratory saturation. They determined minimum ages derived from minimum equivalent dose estimates based on an upper limit of 86% of saturation of the laboratory dose response curves.

At the Batajnica LPS (Fig. 1) discussed here, five clearly distinguishable loess and palaeosol units are exposed, reaching to Marine Isotope Stage (MIS) 16 and possibly beyond (Marković et al. 2009). Despite the importance of the Batajnica LPS as a terrestrial palaeoclimate record for the Middle Danube loess field, no numerical ages have been reported so far.
Here we (i) provide a detailed multi-method luminescence chronology for the Batajnica profile with the focus on the last glacial loess unit; (ii) explore the upper dating limit of the single-aliquot regenerative dose (SAR) protocol applied on 4–11 and 63–90 µm quartz OSL, as well as pIRIR<sub>290</sub> and pIRIR<sub>225</sub> applied on 4–11 µm polymineral fine grains from samples collected from L1 (c. MIS 5–2) loess unit and at the lower boundaries of S1 (c. MIS 5), S2 (c. MIS 7) and S3 (c. MIS 9) palaeosols; and (iii) compare the natural and laboratory generated quartz SAR-OSL and post-IR IRSL<sub>225</sub> and IRSL<sub>290</sub> dose response curves and thus assess what is the dose range over which each protocol provides reliable ages.

**Sampling and site description**

Batajnica LPS is located along the Danube, near Batajnica settlement, 15 km northwest of Belgrade (latitude 44°55′29″N, longitude 20°19′11″E; Fig. 1). The composite Batajnica LPS previously investigated by Marković et al. (2009) reaches a thickness of 40 m. The section comprises at least five loess–palaeosol couplets, extending to or beyond 600 ka (Fig. 2). Two visible dark-brown (altered) volcanic ash layers have been documented in loess unit L2 (Fig. 2). Previous studies on Batajnica include pedostratigraphical correlations, rubification index and environmental magnetic data analyses (Marković et al. 2009). A relative chronology has been obtained by correlating the magnetic susceptibility record with Lingtai (Chinese Loess Plateau) and the SPECMAP oxygen isotope record (Marković et al. 2009).

Luminescence investigations were carried out on 18 individual samples collected in stainless steel tubes from the same outcrops studied by Marković et al. (2009). Seven samples were taken from the L1 loess unit. Doublet samples were collected from the lower boundaries of the S1, S2 and S3 palaeosols, and one sample from the upper boundary of the S2 palaeosol. Additionally, four samples were taken to assess the chronological range of the L2 tephra layers (Fig. 2).

**Methodology**

**Sample preparation and analytical facilities**

The luminescence samples were prepared under low-intensity red light conditions. Gamma spectrometry and water content measurements were carried out using material from the ends of each sample tube, whereas fine quartz (4–11 µm), coarse quartz (63–90 µm) and polymineral fine grains were extracted from the inner part. A treatment with hydrochloric acid (10% concentration) was employed for calcium carbonate removal followed by a treatment with hydrogen peroxide (10% concentration followed by 30%) for organic matter removal. Finer (<63 µm) and coarser (>63 µm) grains were separated through wet sieving. Fine grains (<11 µm) were obtained by settling using Stokes’ law. The quartz fraction was enriched by etching with hydrofluorosilicic acid for 10 days. The 4–11 µm quartz fraction was obtained by centrifugation in distilled water. Polymineral fine grains (4–11 µm) were separated from
the finer polymineral fraction obtained after Stokes’ law settling and centrifuging in distilled water (Frechen et al. 1996; Lang et al. 1996). The coarser (63–90 μm) grain fraction was separated through dry sieving. Since this fraction consists of a polymineral mixture, a density separation using heavy liquid was performed (2.62 and 2.75 g cm\(^{-3}\)). To isolate the quartz grains from the plagioclase feldspars, a treatment with hydrofluoric acid (40% concentration) was applied for 40 min. A rinse with hydrochloric acid (10%) for 60 min removed the precipitated fluorides. For measurement, fine quartz and polymineral grains were mounted on aluminium discs whereas for coarse grains stainless steel discs were used.

Luminescence measurements were performed using TL/OSL Rise DA-20 readers, equipped with a classic or automated detection and stimulation head (DASH) (Lapp et al. 2015). Luminescence signals were detected by EMI 9235QA and PDM 9107Q-AP-TTL-03 (160–630 nm) photomultipliers. 7.5-mm-thick Hoya U-340 UV filters were used for the detection of the quartz signals while for polymineral fine grain signals a blue filter combination (Schott BG39 + Corning 7-59, with transmission between 320–460 nm) was used. Laboratory irradiations were carried out using \(^{90}\)Sr–\(^{90}\)Y radioactive sources that were calibrated with gamma-irradiated calibration quartz (Hansen et al. 2015) for both fine and coarse grain fractions. The dose rate used for measurements was 0.11 Gys\(^{-1}\) for fine grains and 0.13 Gys\(^{-1}\) for coarse grains.

The bleaching experiments on polymineral fine grains were carried out under controlled laboratory conditions using an array of TL29D16/09N lamps, which deliver a power of approximately 1000 W m\(^{-2}\) with similar spectral characteristics to natural sunlight (Petrušić et al. 2011). We have assumed that 1 day of exposure to this lamp corresponds to 1 day of exposure to natural daylight during summer time.

**Equivalent dose determination**

Equivalent doses of fine (4–11 μm) and coarse (63–90 μm) quartz grains were determined using the single-aliquot regenerative dose (SAR) procedure (Murray & Wintle 2000, 2003). Full details on each protocol are given in Table S1. Optical stimulation was carried out with blue light-emitting diodes for 40 s at 125 °C. The
net CW-OSL signal used for analysis was integrated over the first 0.308 s of the decay curve and employing an early background subtraction from 1.69–2.30 s interval. Sensitivity changes were corrected by using the OSL response to a test dose of 17 Gy throughout the whole set of measurements. A preheat temperature of 220 °C for 10 s and a cutheat of 180 °C were employed. A high-temperature bleach of 280 °C for 40 s was performed at the end of each SAR cycle (Murray & Wintle 2003). The robustness of the SAR protocol was checked by the intrinsic performance tests (recycling and recuperation; Murray & Wintle 2003) included in every measurement. OSL IR depletion tests were performed in order to investigate the purity of the quartz luminescence signals on all aliquots measured (Duller 2003). For equivalent dose determination, only the aliquots that yielded recycling and OSL IR depletion ratios within 10% deviation from unity were accepted. Recuperation ratio was considered suitable if it reached <2% of the natural signal.

Two elevated temperature infrared stimulation methods based on the SAR procedure were used for measuring the polymineral fine grains. We first applied a post-IR IRSL protocol (pIRIR$_{290}$) on all samples from the Batajnica LPS (Table S1b; Buylaert et al. 2011b, 2012; Thiel et al. 2011a). After a preheat of 320 °C for 60 s, the samples were exposed to IR diodes for 200 s at 50 °C in order to reduce the signal susceptible to fading by allowing recombination of near-neighbour trap and centre pairs. The signal of interest was recorded during a subsequent IR stimulation at 290 °C for 200 s. A test dose of 17 Gy was used in all measurements, unless otherwise stated. The net signal used for analysis was integrated from the initial ~2.5 s of stimulation minus a background evaluated from the last 50 s. At the end of every cycle, a high-temperature bleach was performed for 100 s at 325 °C to remove residual charge.

A further post-IR IRSL protocol (pIRIR$_{225}$) (Table S1c; Roberts 2008; Buylaert et al. 2009; Wacha & Frechen 2011; Vasiliniuc et al. 2012) was employed to determine equivalent doses on samples previously measured with pIRIR$_{290}$, in order to check the reliability of the pIRIR$_{290}$ ages. After a preheat of 250 °C for 60 s, the aliquots were stimulated with IR diodes, firstly at 50 °C for 200 s and then at 225 °C for 200 s. The signal from the latter stimulation was used in equivalent dose determination. The response to a test dose of 17 Gy was determined in the same way. At the end of each cycle of the protocol, a bleach was performed at 290 °C for 100 s.

Annual dose determination

High-resolution gamma spectrometry was applied to determine the radionuclide activity concentrations using a well-type HPGe detector. In order to achieve $^{226}$Ra–$^{222}$Rn equilibrium, the samples were stored for 1 month. The activity of $^{238}$U was determined indirectly by measuring the $^{234}$Th emissions of 92.3 and 92.8 keV peaks. The $^{228}$Ra activity was determined from the emissions at 295 and 351 keV of $^{214}$Pb and the emission at 609 keV of $^{214}$Bi. The activity of the $^{322}$Th was determined indirectly by measuring the following peaks: 338 keV ($^{228}$Ac), 911 keV ($^{228}$Ac), 238 keV ($^{212}$Pb) and 583 keV ($^{208}$Tl). For determination of the $^{40}$K activity, the 1461 keV peak was used. The annual dose rates were determined using the conversion factors tabulated by Guérin et al. (2011). The beta attenuation and etching factor for 63–90 μm quartz fraction was assumed to be 0.94±0.05 (Mejdahl 1979). For the 4–11 μm quartz fraction, an alpha efficiency factor of 0.04±0.02 was taken into account whereas for the polynuclear fine grains a value of 0.08±0.02 was used (Rees-Jones 1995). The time averaged water content was assumed to be 15% with a relative error of 25% based on the average humidity of the loess samples in Vojvodina. The external contribution from beta and gamma radiation (additionally alpha radiation for 4–11 μm grains), as well as from the cosmic rays, was included in the total dose rates. For each sample, the dose rate of the cosmic rays was estimated as a function of depth, altitude and geomagnetic latitude (Prescott & Hutton 1994). The internal dose rate contribution for the coarse quartz fraction was assumed to be 0.010±0.002 Gy ka$^{-1}$ (Vandenberghhe et al. 2008). Given the size of fine grains (4–11 μm), it is assumed that any dose rate derived from internal alpha activity is negligibly small.

Additionally, the total $^{210}$Pb content was determined for 10 samples by measuring the concentration of $^{210}$Po (5.304 keV peak, alpha energy) by means of alpha spectrometry. A chemical separation was performed on the loess matrix to isolate the Po isotopes from the other alpha emitters (Data S1). The measurements were carried out using an ORTEC SOLOIST 450 mm$^2$ PIPS detector (19 keV resolutions) and an ASPEC-92 Dual Multichannel analyzer.

Determination of expected ages for the major palaeosol units based on magnetic susceptibility correlation

Marković et al. (2009) described the characteristic stratigraphical pattern in magnetic susceptibility for the uppermost soil and various palaeosol units at Batajnica and how it can be correlated with similar records throughout Eurasia. It is well known that major palaeosols within Eurasian LPSs exhibit a distinctive magnetic enhancement pattern, which is also reflected in grain size and colour proxies, that denotes the degree of pedogenesis, which in turn is hydroclimatically controlled. These patterns and their stratigraphical superposition allow for secure identification of major palaeosols and serve as independent chronostratigraphical markers that constitute the backbone of the loess chronostratigraphy in the wider Danube Basin and beyond (Marković et al. 2015;
Necula et al. (2015). Starting from this well-established base, Basarin et al. (2014) correlated the astronomically tuned magnetic susceptibility record of the Titel/Stari Slankamen composite profile to the benthic oxygen isotope stack (Lisiecki & Raymo 2005) for the last c. 800 ka.

As such, the major loess–palaeosol transitions identified in Batajnica according to the magnetic susceptibility record were correlated to the corresponding ones in the Titel/Stari Slankamen composite profile (Fig. 2). This approach (see Basarin et al. 2014) further enabled a correlation to the benthic oxygen isotope stack (Lisiecki & Raymo 2005). As the luminescence samples were collected from the same outcrop where the magnetic susceptibility samples were taken, previous field marks allowed for secure correlation of our samples and the magnetic susceptibility samples. Expected age estimates have been assigned to samples collected from the Holocene soil and palaeosol boundaries (BAT-1.0, 1.11, 1.12A, 1.16, 1.17 and 1.19A) based on the data reported by Basarin et al. (2014) and their correlation to benthic oxygen ages of Lisiecki & Raymo (2005) (Fig. 2, Table S2).

This method of determining expected ages relies on the assumption that no large erosional gaps occur at the loess–palaeosol transitions for Batajnica and Titel/Stari Slankamen. We are confident that this assumption is valid for the investigated sites as the regional Danube Basin stratigraphy is very well constrained by integrating many profiles; in case a significant erosional gap occurs (e.g. Stari Slankamen L2), it can be clearly identified (Marković et al. 2009, 2015). Smaller erosion gaps cannot be excluded, but they are not relevant at the time scale of the uncertainties in the expected depositional ages. Furthermore, in order to account for possible uncertainties, a 10% error has been associated with the boundary ages between loess and soil or palaeosols.

Results and discussion

Annual doses and calculation of expected equivalent doses

Tables 1 and S3 contain relevant information on dosimetry and specific radionuclide activities. The annual doses were derived from the activity concentrations of the $^{238}\text{U}$, $^{232}\text{Th}$ $^{40}\text{K}$ and $^{210}\text{Pb}$ measured using gamma spectrometry.

We have investigated the secular equilibrium assumption in the $^{238}\text{U}$ chain by assessing the ratio between the activity concentration of $^{210}\text{Pb}$ measured directly using the 46 keV line and $^{220}\text{Ra}$ indirectly measured using the $^{214}\text{Pb}$ (352 and 295 keV) and $^{214}\text{Bi}$ (609.3 keV) energy peaks. The average ratio calculated for all samples is $0.48\pm0.03$ (Fig. S1). This is caused either by the escape of $^{222}\text{Rn}$ during the burial time or by inaccurate gamma spectrometry measurement of $^{210}\text{Pb}$. Thus, the activity concentration of $^{210}\text{Pb}$ was determined indirectly using alpha spectrometry by measuring $^{210}\text{Po}$ for 10 samples. As seen in Fig. S2, the ratios between the alpha and gamma spectrometry results are consistent with unity, showing that the activity concentrations of $^{210}\text{Pb}$ obtained by gamma spectrometry are accurately determined. This indicates that ~50% radon loss occurs in the investigated loess samples. Thus, the annual doses that consider the measured $^{210}\text{Pb}$ were used in to determine the luminescence ages.

By multiplying the expected age estimates (Table S2) by the corresponding environmental dose rate, expected equivalent doses and their associated uncertainties were determined (Table S2).

Luminescence properties – quartz

For both quartz fractions the net signals displayed a rapid decay during optical stimulation as documented for sample BAT-1.10 in Fig. S3. The patterns of the natural and regenerative decay curves were found to be similar to that shown by the luminescence signal of the calibration quartz, which is recognized as being dominated by the fast component (Hansen et al. 2015). Representative laboratory growth curves are shown in Fig. S3, displaying a good behaviour in the SAR protocol. The dose response curves were well fitted using the sum of two saturating exponential functions. The recycling and IR depletion ratios were within 10% from unity, demonstrating that the sensitivity changes during repeated SAR cycles are accurately corrected for, and the quartz signals are pure (Table S4).

Preheat plateau. – The dependency of the equivalent doses on the preheat temperature was investigated for sample BAT-1.13B (fine quartz). Different preheat temperatures ranging from 180 to 280 °C were applied on sets of five aliquots for each sample, in combination with a test dose cutheat of 180 °C. Figure S4 shows that over the investigated interval of preheat temperatures, equivalent doses do not display systematic variation. The recycling ratio and recuperation were satisfactory for all the aliquots measured.

Dose recovery. – Further investigation was focused on analysing whether the SAR protocol can successfully measure a known irradiation dose prior to any thermal treatment by applying the dose recovery test (Murray & Wintle 2003). Sets of five aliquots were prepared from 4 to 11 μm quartz samples (BAT-1.9, 1.11, 1.12B, 1.13B) and 63–90 μm quartz samples (BAT-1.1, 1.9, 1.11, 1.12B). The aliquots were irradiated with a beta dose, chosen to approximate the equivalent doses, after their natural signals were bleached by a repeated exposure to the blue light-
Table 1. Summary of the OSL, pIRIR<sub>290</sub> and pIRIR<sub>235</sub> ages considering the residual dose estimated based on the modern analogue sample. * indicates the set of polymer al ages considering the residual dose measured artificially. The data were determined following Atkin & Allred (1972). The uncertainties associated with the luminescence and dosimetry data are random; the uncertainties mentioned with the optical ages are the overall uncertainties. The systematic errors taken into account include; 2% beta source calibration, 3% conversion factors, 5% attenuation and etching factors, 3% gamma spectrometer calibration, 1% cosmic radiation, 25% water content. All uncertainties represent 1σ. Specific activities were measured on a well detector by gamma spectrometry and the ages were determined considering 15% water content; beta attenuation and etching factors used for 63<sup>11</sup>mL fine grains (Rees-Jones 1995). The total dose rate consists of the contribution from the beta and gamma radiations for coarse grains as well as the contribution from alpha radiation in the case of fine grains. The contribution of cosmic radiation was taken into account and calculated accordingly to Prescott & Hutton (1994). For coarse quartz grains an internal dose rate of 0.01±0.02 Gy ka<sup>-1</sup> was considered (Vandenberghe et al. 2008). In italics are given the equivalent doses calculated by interpolating backgrounds.

| Sample code | Stratigraphical unit | Depth (cm) | 4-11 Gy (E/Gy) | 65-90 µm quartz | pIRIR<sub>290</sub> (pg) | pIRIR<sub>235</sub> (pg) | Total random error (%) | Total systematic error (%) | Total dose rate (Gy ka<sup>-1</sup>) | Age (ka) | Age (ka)* |
|-------------|---------------------|------------|----------------|-----------------|---------------------|---------------------|--------------------------|--------------------------|---------------------------|---------|---------|
| BAT 1.0     | S0/L1               | 97         | 31±11          | 20±11           | 96±9               | 57±6                | 4.1                      | 46±25                    | 16.5                      | 8.8±6   | 7.4±6   |
| BAT 1.1     | L1                  | 103        | 37±14          | 40±12           | 45±10              | 45±10               | 4.1                      | 59±23                    | 13.7                      | 6.7±6   | 8.3±6   |
| BAT 1.7     |                     | 214        | 74±11          | 90±16           | 89±10              | 91±17               | 2.7                      | 70±15                    | 8.0±6                     | 8.6±6   | 8.6±6   |
| BAT 1.8     |                     | 425        | 160±11         | 150±16          | 121±10             | 105±11              | 2.6                      | 60±7                     | 10.7                      | 9.2±6   | 8.8±6   |
| BAT 1.9     |                     | 641        | 145±15         | 136±16          | 185±18             | 162±10              | 4.7                      | 66±10                    | 7.6±6                     | 8.8±6   | 8.5±6   |
| BAT 1.10    |                     | 121        | 176±12         | 160±17          | 217±16             | 189±10              | 3.2                      | 51±8                     | 6.1±6                     | 7.6±6   | 8.3±6   |
| BAT 1.11    | S0/L1               | 935        | 217±14         | 211±18          | 320±14             | 292±15              | 3.1                      | 46±5                     | 8.3±6                     | 8.4±6   | 8.4±6   |
| BAT 1.12A   | S2/L2               | 1200       | 253±15         | 224±12          | 490±23             | 377±19              | 3.3                      | 62±5                     | 5.9±6                     | 8.9±6   | 8.9±6   |
| BAT 1.12B   |                     | 1200       | 206±16         | 262±16          | 490±23             | 290±15              | 4.3                      | 72±5                     | 5.9±6                     | 9.0±6   | 9.0±6   |
| BAT 1.13A   | L2                  | 1500       | 275±16         | 670±13          | 789±12             | 572±17              | 4.5                      | 112±6                    | 4.6±7                     | 8.6±6   | 8.6±6   |
| BAT 1.13B   |                     | 1500       | 365±19         | 357±16          | 745±16             | 741±17              | 4.2                      | 133±6                    | 5.6±7                     | 8.3±6   | 8.3±6   |
| BAT 1.14A   |                     | 1450       | 354±17         | 290±12          | 765±13             | 542±17              | 3.6                      | 104±8                    | 4.3±7                     | 8.7±6   | 8.7±6   |
| BAT 1.14B   |                     | 1450       | 359±11         | 214±16          | 649±68             | 661±42              | 4.4                      | 63±8                     | 7.1±7                     | 8.8±6   | 8.8±6   |
| BAT 1.15A   | S2/L2               | 1500       | 580±16         | 366±13          | 1171±129           | 690±28              | 3.5                      | 148±11                   | 3.0±7                     | 8.7±6   | 8.7±6   |
| BAT 1.16    |                     | 1500       | 386±19         | 377±12          | 1031±89            | 525±35              | 3.6                      | 76±8                     | 4.9±7                     | 8.1±5   | 8.1±5   |
| BAT 1.17    | L2/L2               | 1900       | 365±19         | 377±27          | 1031±89            | 525±35              | 3.6                      | 76±8                     | 4.9±7                     | 8.1±5   | 8.1±5   |
| BAT 1.19A   | L4/L4               | 2000       | 465±59         | 454±57          | 1170±86            | 718±66              | 2.7                      | 72±7                     | 6.1±7                     | 7.1±5   | 8.9±5   |
| BAT 1.19B   |                     | 2000       | 475±27         | 464±27          | 1240±62            | 53±8                | 3.5                      | 85±5                     | 5.9±7                     | 9.1±5   | 7.2±5   |

* indicates the set of polymer al ages considering the residual dose measured artificially.
emitting diodes for 100 s at room temperature with a pause of 10 ks. These doses were then determined using the SAR protocol. As seen in Fig. 3A, for the quartz samples, the SAR protocol can successfully recover laboratory doses up to 320 Gy on fine quartz and 260 Gy on coarse quartz.

Quartz equivalent doses. – Equivalent doses were measured on at least 10 aliquots of each grain size. Tables 1 and S4 summarize the equivalent dose results, as well as the results from the SAR-intrinsic tests (Recycling, IR depletion and Recuperation).

Equivalent doses measured on fine quartz aliquots range from $31^{+6}_{-1}$ Gy for sample BAT-1.0 collected from the Holocene soil to $486^{+5}_{-6}$ Gy for sample BAT-1.19A from below the S3 palaeosol. The natural signal emitted by this sample interpolates well below the laboratory saturation threshold. Yet, the measured fine quartz equivalent dose reaches only 50% of the expected burial dose (Table S2). Coarse quartz equivalent doses range from $28^{+1}_{-1}$ Gy for the sample from modern soil to $262^{+16}_{-16}$ Gy for a sample collected below the S1 palaeosol. For older samples, the natural signal was in field saturation and close to laboratory saturation (see below).

Luminescence properties – polymineral fine grains

Figure S3C, D shows representative growth curves for sample BAT-1.10 constructed using pIRIR$_{290}$ and pIRIR$_{225}$ protocols. The dose response curves were well fitted using the sum of two saturating exponential functions. The growth curves pass very close to the origin demonstrating that the recuperation is insignificant. As can be seen in insets of Fig. S3C, D the decays with stimulation time of the natural and regenerative signals have similar shapes.

Effect of test dose size on equivalent dose. – The effect of test dose size on the measured equivalent dose using the pIRIR$_{290}$ protocol was investigated on samples BAT-1.9, 1.12A and 1.16 (Fig. 4). Sets of three to five aliquots were used for each test dose (except for the 17 Gy test dose). As shown in Fig. 4 for samples BAT-1.9 and BAT-1.12A no systematic trend in equivalent dose values with test dose magnitude can be seen. Although not so clear, this is also valid for sample BAT-1.16, which yielded pIRIR$_{290}$ equivalent doses of $1172^{+129}_{-84}$ and $996^{+64}_{-64}$ Gy by employing test dose sizes of 2 and 50% of the $D_e$, respectively. The expected equivalent dose for sample BAT-1.16 is $617^{+84}_{-84}$ Gy (Table S2). It is important to note that the natural signal emitted by sample BAT-1.16 normalized to a test dose of 17 Gy was interpolated in the saturation region of the dose response curve. The results from the Batajnica loess samples indicate that the dependence of the equivalent dose on the size of test dose, supported by investigations in Chinese loess (Yi et al. 2016), might not be a general phenomenon.

Determination of residual doses. – One of the most important assumptions of the luminescence dating method is that the luminescence signal of the mineral has been fully reset in nature by light exposure prior to deposition. The aeolian nature of loess deposits suggests us that the mineral constituents have experienced considerable daylight exposure. Buylaert et al. (2012) showed that PIRIR signals bleach at a much slower rate than the quartz OSL signal. For pIRIR$_{225}$ signals emitted by K-feldspar residual doses not exceeding 2 Gy were reported (e.g. Buylaert et al. 2009). But later on, hard-to-bleach (or un-bleachable) pIRIR signals amounting to $\sim$6 Gy have been reported for K-feldspar extracts from Chinese loess after extensive exposure in a solar simulator (Yi et al. 2016). Bleaching experiments conducted by Stevens et al. (2011) on polymineral fine grains from loess collected from the Carpathian Basin.
sugar that the value of the residual is influenced by the exposure time and conditions as well as the equivalent dose of the sample with residual values. For pIRIR290 signals ranging from 5.5 Gy to 3.5 Gy, the dose recovery ratio improved from 1.36 to 0.87. For sample BAT-1.8, a dose recovery ratio of 1.31 was obtained with 2 Gy. The residual dose was determined for each sample. These residual doses were subtracted from all the pIRIR D_e values prior to calculation of the ages.

Dose recovery. – Dose recovery tests (Murray 1996; Wallinga et al. 2000) were performed on five aliquots from samples BAT-1.8 to 1.11 and BAT-1.12B. The natural signal was bleached by exposing fresh aliquots to a lamp for 2 weeks. Laboratory known doses were chosen to match the measured equivalent dose. The residual doses were subtracted from the recovered doses before calculating the dose recovery ratios. Unless otherwise stated, a test dose of 17 Gy was used throughout the measurements. As shown in Fig. 3B, the pIRIR250 protocol can successfully recover laboratory doses up to 316 Gy, while the pIRIR290 protocol systematically overestimates the given laboratory doses. Following the suggestions of Yi et al. (2016), we carried out pIRIR290 dose recovery tests using higher test doses. For sample BAT-1.9, the dose recovery ratio improved from 1.36±0.06 to 1.07±0.03 using a 17 Gy test dose (7% of D_e) and a 50 Gy test dose (23% of D_e), respectively. However, for sample BAT-1.11, the dose recovery ratio of 1.31±0.04 and 1.41±0.10 were obtained using test doses of 17 Gy (5% of D_e) and 150 Gy (41% of D_e), respectively. As shown above, the size of the test dose does not influence the measured equivalent dose (Fig. 4) or the saturation characteristics of the pIRIR290 dose response curves (see below). Thus, in this paper we will discuss the equivalent doses measured with a test dose of 17 Gy.

Unsatisfactory dose recovery tests for pIRIR290 have also been reported by Murray et al. (2014) for Serbian loess, by Veres et al. (2018) for Ukrainian loess and by Thiel et al. (2011b) for Japanese loess. On fluvial and lacustrine sediments from Heidelberg Basin, Germany, Li et al. (2018) reported poor pIRIR290 dose recovery and good pIRIR225 dose recovery. However, such behaviour is not general since Thiel et al. (2011a), Yi et al. (2016), Böskens et al. (2017), Stevens et al. (2018) and Zhang et al. (2018) reported good pIRIR290 dose recovery ratios for the loess in the Middle Danube basin and the Chinese Loess Plateau.
Polyminal fine grains equivalent doses. – The pIRIR\textsubscript{290} and pIRIR\textsubscript{225} equivalent doses after the subtraction of residual doses are displayed in Table 1 while the measured equivalent doses are presented in Table S4. The pIRIR\textsubscript{290} equivalent doses vary from 36±9 Gy (BAT-1.0) to 490±21 Gy (BAT-1.12A). The smallest pIRIR\textsubscript{225} equivalent dose is 37±6 Gy (BAT-1.0), whereas the maximum is 377±19 Gy (BAT-1.12A).

The natural pIRIR\textsubscript{225} and pIRIR\textsubscript{290} signals yielded by older samples scatter around or above the saturation threshold of the dose response curve (see below).

Fading rate measurements for high doses. – While pIRIR\textsubscript{225} fading rates >1%/decade are reported in the literature (Thomsen et al. 2008; Buylaert et al. 2009; Zhang et al. 2018), Vasliniu et al. (2012) calculated fading rates ranging from 0.6 to 1.3%/decade on samples collected from the Last Glacial loess in Mircea-Voda, Romania, and concluded they are laboratory artefacts. Stevens et al. (2011), Thiel et al. (2011a) and Murray et al. (2014) reported fading rates considered to be laboratory artefacts for polynimal pIRIR\textsubscript{290} signals on Serbian and Austrian loess sites along the Danube valley. Balescu et al. (in press) determined extremely low fading rates (<0.18%/decade) for pIRIR\textsubscript{290} signals emitted by K-feldspars extracted from Bulgarian loess.

Fading rates (percentage of the signal lost per decade of time; Aitken 1985) were measured on fresh 4–11 µm polynimal aliquots from samples BAT-1.11 and BAT-1.19A (Fig. S5, Table S7). The aliquots were bleached by exposure to a lamp for 2 weeks and irradiated with beta doses approximating the equivalent doses for BAT-1.11 (317 and 364 Gy) and BAT-1.19A (400 Gy). A test dose of a magnitude of 17 Gy was used throughout all fading measurements. Four consecutive prompt read-outs were carried out before fading measurement. The aliquots were preheated before storage. Following the read-out after different storage times, two consecutive prompt read-outs were carried out.

Figure S5A–L shows a significant scatter in the pIRIR sensitivity-corrected luminescence signals registered during prompt read-outs. It is interesting to note that the signal intensity recorded after the first instantaneous read-out always lies above the values recorded during the subsequent prompt read-outs. We consider that no systematic variation with delay time is identified for the pIRIR sensitivity-corrected luminescence signals.

The luminescence stability of quartz is well known, and previous studies reported that the quartz signal is not thought to suffer from anomalous fading (Aitken 1998). However, Thiel et al. (2011a) reported fading rates of 1.3±0.3%/decade for fine quartz extracted from Chinese loess. Fading tests were also performed by Lowick & Valla (2018) on fine and coarse quartz. They determined quartz g-values ranging from 0 to 6%/decade while for calibration quartz g-values of 1–2%/decade were measured.

We performed fading measurements on 4–11 µm quartz and 63–90 µm quartz extracted from sample BAT-1.11 in order to test whether the calculated g-values on polynimal material are fitting artefacts caused by large scatter in the data set. The aliquots were bleached twice with blue LEDs for 100 s at room temperature. Between the two stimulations, a pause of 10 ks was used. After bleaching, the g-values were measured using the SAR-OSL protocol in the same manner as for the polynimal fine grains. Figure S5M–S shows that the scatter in the quartz data is comparable with the polynimal data set. We report average g-values of 2.49±0.44 and 2.92±0.61%/decade for 4–11 and 63–90 µm quartz, respectively (Table S7). Such g-values cannot reflect real fading as this would imply a significant signal loss during burial. This is in contradiction with our observation that the natural signals emitted by 63–90 µm quartz are close to full saturation, as discussed below.

We interpret the presented data set as follows. There is no solid evidence that quartz and pIRIR\textsubscript{225} and pIRIR\textsubscript{290} polynimal signals fade; rather it appears that by using this procedure on the investigated samples, one cannot measure reliable g-values. Since we cannot confidently measure g-values using the short-term standard fading measurement procedures in the following we discuss the uncorrected pIRIR\textsubscript{225} and pIRIR\textsubscript{290} ages for samples BAT-1.0–BAT 1.12B. One should also note that fading corrections should not be attempted if the natural signal interpolates beyond the linear range of the dose response curve, as is the case with most of the natural signals emitted from the Batajnica samples (Huntley & Lamotho 2001).

High laboratory doses given on top of naturally accrued doses. – Previous studies reported a mismatch between SAR dose response curves and the dose response curve constructed when large doses were added on top of natural ones for fine and coarse quartz, indicating that the SAR-OSL protocol is problematic in the high dose range. Anechitie-Deacu et al. (2018) showed that in the case of fine quartz extracted from aeolianites the sensitivity-corrected luminescence signal after a large dose was added on top of the natural one was not found to be at the same level to the laboratory saturation region, reaching only 86% of SAR laboratory full saturation intensity, while in the case of coarse quartz 96% of full saturation was attained. For quartz extracted from Ukrainian loess, Veres et al. (2018) showed that after adding 3500 Gy on top of the natural signal of 4–11 µm quartz (De ~550 Gy), the sensitivity-corrected signal reached 96% of the luminescence level induced by a 4000 Gy beta dose. In the case of the 63–90 µm fraction identical luminescence levels were measured after adding a 500 Gy beta dose on top of the natural signal (De ~500 Gy) and after a 1000 Gy beta dose.

If large laboratory doses are added on top of natural signals found in field saturation, an increase of the signals
may suggest a degree of fading that may have been undetected during standard fading rates measurements. On the other hand, if a large dose is given on top of the naturally accrued doses, one normally expects the magnitude of the measured signal to be the same as the saturation value measured in the SAR protocol.

We chose to perform such an experiment on sample BAT-1.19A, collected below the S3 palaeosol. As can be seen in Fig. 7A, for a sample with an expected age of more than 300 ka, the pIRIR225 natural signal reaches 84±1 % of saturation level, while the pIRIR290 natural signal lies above the laboratory dose response curve (105±3%). For quartz, natural sensitivity-corrected signals are at 95±6% of laboratory saturation levels in the case of 63–90 μm grains, while in the case of fine grains only 56±1% of saturation level is reached.

Ten aliquots of these samples were prepared for each measurement protocol. The first five aliquots were used as control aliquots for recording the natural signals (L/Tn), while the rest of the aliquots were irradiated to a beta dose added on top of the naturally accrued dose. The magnitude of this dose was chosen to be large enough for the signal to reach full saturation (Fig. 7A). The sensitivity-corrected luminescence signal (denoted as (L/Tn)*) was recorded after the large dose was added. The same aliquots were subsequently measured in a standard SAR protocol after an irradiation to a dose of 5000 Gy, chosen to equal the equivalent dose plus the given dose. The value of this normalized signal is hereafter denoted as (L/Tn).

If (L/Tn)* is greater than (L/Tn) there is the possibility that during burial time some loss of the signal may have occurred. In addition, if (L/Tn)* is greater or smaller than (L/Tn), issues regarding unknown measurement problems during the first cycle of the measurement or other as yet unknown causes can also be suspected.

From Table S8 it can be seen that for quartz the ratio between (L/Tn)* and (L/Tn) is 1.90±.02 in the case of fine grains and 0.96±0.08 in the case of coarse grains. For polymineral fine grains the ratio of (L/Tn)* to (L/Tn) is 1.35±0.03 in the case of pIRIR225 and 1.20±0.04 in the case of pIRIR290. The ratio between (L/Tn)* and (L/Tn) in the case of pIRIR225 is 1.10±0.03 while for pIRIR290 is 1.15±0.05. In the case of fine and coarse quartz corresponding ratios of 0.95±0.02 and 1.03±0.11, respectively, were obtained (Table S8).

This indicates that in the case of all signals excepting coarse quartz OSL the maximum light levels attained during prolonged natural irradiation can be increased by additional laboratory irradiation. We choose to restrain ourselves from interpreting this observation as an indicator of signal fading during burial because our results here as well as those obtained in fading tests indicate that it is likely that the signals measured during the first measurement cycle in pIRIR protocols slightly overestimate the response obtained for the same dose later given in the SAR protocol. The cause of this behaviour needs further investigation.

Luminescence ages

The ages calculated using the measured activity concentrations of 210Pb are generally 10 to 20% older than those calculated assuming secular equilibrium in the 238U chain, but the two sets of ages are consistent within error limits (Fig. S6). Assuming a constant radon loss over the whole sample burial time, the set of ages calculated using the measured activity concentrations of 210Pb is discussed in the following section.

Table 1 displays the 4–11 μm quartz, 63–90 μm quartz, pIRIR290 and pIRIR225 polymineral ages assumed to be unaffected by fading and by considering two possibilities for residual dose correction, namely by using the values obtained by laboratory bleaching experiments and by considering residuals derived through the use of a modern analogue. Only the ages obtained by using the latter correction procedure are discussed further on.

For Batajnica, the luminescence ages are in agreement with the previous chronological framework based on magnetic susceptibility correlation to other records (Buggle et al. 2009; Marković et al. 2009), placing the L1 loess unit into the Last Glacial.

Except for sample BAT-1.9, all fine quartz ages are systematically lower than coarse quartz ages and the difference between them increases with depth (Fig. 5), with the fine quartz underestimating the expected depositional ages based on magnetic stratigraphy (Fig. 6). At the neighbouring Titel loess section, Perić et al. (2019) reported unreliable fine quartz ages beyond 30–35 ka (~100 Gy).

The coarse quartz, polymineral pIRIR225 and pIRIR290 ages largely agree (Fig. 5) and within errors are consistent with the expected ages (Fig. 6).

Immediately below the S1 palaeosol, the coarse quartz, pIRIR225 and pIRIR290 yielded ages up to 116±12 ka (262±16 Gy), 130±14 ka (377±19 Gy) and 169±17 ka (490±21 Gy), respectively. However, it is difficult to interpret these results obtained immediately below the S1 palaeosol since the doublet samples BAT-1.12A, B, differ by 23% on fine quartz, 10% on coarse quartz, 38% on pIRIR225 and 28% on pIRIR290. This causes a difference of 17 and 11 ka on fine and coarse quartz, respectively, and of 36 ka on polymineral fine grains. As the dosimetry data for the doublet samples are consistent, with the exception of 210Pb, the age discrepancies seem to originate from the difference between the equivalent doses. More investigations are required to explain the different equivalent dose results yielded by doublet samples.
For samples taken from deeper units (BAT-1.13A to 1.19A, B), except for fine quartz, the natural signal is in the region of laboratory saturation. Interpolation of the natural signal onto the high curvature region of the dose response curve results in large and asymmetric uncertainty in the equivalent dose. Consequently, the obtained luminescence ages for samples BAT-1.13A to 1.19A, B have low precision and their reliability is questionable in terms of accuracy, as is shown below.

### Accuracy of the reported luminescence ages

In order to identify the factors that control the upper limit of luminescence dating we investigate the saturation characteristics of the natural and laboratory signals emitted by both quartz and polymineral grains.

#### Laboratory dose response curves.

Laboratory dose response curves up to 5000 Gy were constructed on sample BAT-1.19A collected from below the S3 palaeosol (>300 ka according to Marković et al. 2009) using 4–11 and 63–90 µm quartz as well as polymineral fine grains employing the pIRIR225 and pIRIR290 protocols (Fig. 7A). All dose response curves were best fitted with the sum of two saturating exponential functions:

\[ I = I_0 + a \left(1 - e^{-bD}ight) + c \left(1 - e^{-dD}ight) \]  

where \( I \) is the intensity of the signal for a given dose \( D \), \( I_0 \) is the intercept, \( a \) and \( b \) are the amplitudes of the exponential functions while \( D_{01} \) and \( D_{02} \) are the doses that characterize the onset of saturation of each exponential function, also named characteristic doses in the literature. Following the suggestion of Wintle & Murray (2006) we consider the saturation threshold at 85% of the laboratory dose response curve. We assess the closeness to saturation of natural signals by calculating the ratios between the average sensitivity-corrected natural signals (\( L_{\text{nat}}/T_{\text{nat}} \)) and the corrected luminescence signals measured for the 5000 Gy regenerative dose (\( L_x/T_x \)).

The laboratory dose response curve on 63–90 µm quartz saturates much earlier than fine quartz and polymineral samples and has characteristic saturation doses of \( D_{01} = 21\pm28 \) Gy and \( D_{02} = 153\pm46 \) Gy. The coarse quartz natural signal reaches laboratory saturation. Fine quartz on the other hand, yields a laboratory luminescence signal that continues to grow to doses beyond 5000 Gy and the natural signal is interpolated well below the saturation region of the dose response curve. However, the equivalent dose measured for this sample underestimates severely the expected equivalent dose (Table S2). The characteristic doses for fine quartz dose response curves are \( D_{01} = 178\pm17 \) Gy and \( D_{02} = 1635\pm164 \) Gy. Timar-Gabor et al. (2012) also found the natural fine quartz signal of an infinitely old sample from Costinesti LPS (Romania) not to be in saturation. Timar-Gabor et al. (2017) determined similar saturation characteristic doses for worldwide loess and samples of various sedimentary origins.

It is important to note that pIRIR225 and pIRIR290 laboratory dose response curves have lower characteristic doses than the fine quartz. We report \( D_{01} = 142\pm13 \) Gy and \( D_{02} = 795\pm36 \) Gy for pIRIR225 while for pIRIR290 \( D_{01} = 193\pm32 \) Gy and \( D_{02} = 764\pm145 \) Gy. The natural pIRIR225 signal of sample BAT-1.19A is 84±0.7% of laboratory saturation while the natural pIRIR290 signal is 105±3% and lies slightly above the light level corresponding to the 5000 Gy dose (Fig. 7A, Table S9).
Previous studies document the influence of the magnitude of the test dose on the pIRIR290 laboratory growth curves up to high doses (e.g. Murray et al. 2014; Yi et al. 2016; Colarossi et al. 2018). Figure 7B shows pIRIR290 laboratory dose response curves up to 5000 Gy on sample BAT-1.19A using different test doses: 200 Gy (11% of De), 400 Gy (22% of De) and 800 Gy (44% of De). The test doses can be expressed as a fraction of light saturation derived from the growth curve, and ratios of 0.37±0.01, 0.53±0.01 and 0.72±0.01 are determined for 200, 400 and 800 Gy, respectively. The growth curves were fitted with the sum of two saturating exponential functions using Equation 1. The corrected luminescence signals were normalized to the sum of the values of the amplitudes of the exponential functions obtained by fitting (a+c). The saturation parameters (D01; D02) do not display a significant increase with increased magnitude of test dose (Table S10). Based on our finds for Batajnica, we conclude that the dependence of the saturation parameters on the size of the test dose reported for Chinese loess (Yi et al. 2016) or fluvial sediments from South Africa (Colarossi et al. 2018), might not represent a general feature of pIRIR290 laboratory signals.

In order to quantify the closeness of the natural sensitivity-corrected luminescence signals to laboratory saturation for all samples collected from the deeper units (BAT-1.12A-BAT-1.19B), we calculated the ratio between the natural sensitivity-corrected luminescence signal and the average maximum corrected luminescence signals induced by a dose of 5000 Gy. The latter was measured after constructing laboratory dose response curves up to 5000 Gy on samples BAT-1.17 and BAT-1.19A. The ratios are given in Table S11.

**Natural dose response curves** – To construct natural dose response curves, the average sensitivity-corrected natural luminescence signals (Lnat/Tnat) of the samples listed in Table S2 were plotted against the expected equivalent doses determined above. The natural growth curves are further compared to average laboratory dose response curves in order to assess the reliability of results (Fig. 8). Fine grained quartz natural and laboratory growth curves overlap until ~150 Gy (Fig. 8A). This suggests that fine quartz natural signals resulting from doses up to ~150 Gy (c. 50 ka) could result in reliable ages. For higher natural doses the SAR protocol underestimates the expected fine quartz equivalent doses. Over the entire dose range investigated here, neither the natural nor the laboratory dose response curves reach full saturation. This may be due to the dose range being lower than 1200 Gy. This indicates that the fine quartz signals are outside field and laboratory saturation for the entire dose range investigated (Figs S7, 7A).

Timar-Gabor & Wintle (2013) reported similar results for fine quartz extracted from the Costinesti LPS in Romania. The natural and laboratory dose response curves overlap up to ~200 Gy. Coarse-grained quartz natural and laboratory growth curves overlap until ~250 Gy, corresponding to an upper age limit of c. 100 ka for the Batajnica samples (Fig. 8B). This implies that for natural doses up to 250 Gy (c. 100 ka), the SAR protocol provides reliable coarse quartz...
OSL ages. Figs 9A, S7 show that the natural signals emitted by coarse quartz samples with measured equivalent doses higher than \(~\sim 250\) Gy (BAT-1.13A to 1.19A) reach field saturation but underestimate the full laboratory saturation by roughly 15\%. Similar results have been reported for ‘infinitely old’ loess samples from Mircea-Voda LPS in Romania (Timar-Gabor et al. 2012) where the laboratory signals overestimate the natural field saturation by 10–15\%.

The results obtained on 4–11 and 63–90 \(\mu\)m quartz from Batajnica confirm also the findings of Chapot et al. (2012) for 35–63 \(\mu\)m quartz extracted from Chinese loess. They recommend a maximum limit for OSL dating for an equivalent dose of 150 Gy after observing the overlap between the natural and laboratory dose response curves.

The uncorrected pIRIR\(_{225}\) natural and laboratory growth curves appear to generally overlap over the dose range investigated (up to \(~\sim 1200\) Gy; Fig. 8C). However, the overlap beyond \(~500\) Gy should be interpreted with caution because of the large errors of the data points and their spread along with our observation that the natural signals are in field saturation (Fig. S7).

The natural pIRIR\(_{290}\) signals are consistent with the laboratory signals for doses up to \(~400\) Gy (Fig. 8D). For higher doses, the natural pIRIR\(_{290}\) signals overestimate the laboratory signals for the Batajnica samples. Figs 9B, S7 show that the pIRIR\(_{225}\) and pIRIR\(_{290}\) natural signals are in both field and laboratory saturation for samples collected from L2 loess, starting with BAT-1.13A. Similar results have been reported for the pIRIR\(_{290}\) protocol by Murray et al. (2014) on Stari Slankamen LPS, Serbia.

Regional chronostratigraphical correlations

Loess records in the southern Middle Danube basin have been investigated intensively for palaeoclimate reconstructions (e.g. Marković et al. 2006, 2007, 2014, 2015;
Bokhorst et al. 2009; Buggle et al. 2009; Stevens et al. 2011; Obreht et al. 2019). In particular, trends observed in environmental magnetic proxies have allowed for insights into the regional past long-term climatic and environmental variability, as well as attempts at chronos-

trigraphical correlations amongst different sections (Marković et al. 2015). The magnetic susceptibility records from Batajnica (Marković et al. 2009), Stari Slankamen (Marković et al. 2011), Rogulić gully and other sections at the Titel loess plateau (Bokhorst et al. 2009; Basarin et al. 2014; Perić et al. 2019), Surdük (Antoine et al. 2009) and Crvenka (Marković et al. 2011; Marković et al. 2018b) display similar variability for LGC (Fig. 2). Nevertheless, despite the available lumi-

cence data for Irig (Marković et al. 2007), Surdük (Fuchs et al. 2008), Stari Slankamen (Schmidt et al. 2010; Murray et al. 2014), Titel (Perić et al. 2019), Crvenka (Stevens et al. 2011) and Orlovat (Timar-Gabor et al. 2015), the coherence in palaeoclimate reconstructions amongst different LPS from the southern Middle Danube basin has only been evaluated by astronomical tuning (Basarin et al. 2014) and has not been discussed and tested in the framework of absolute luminescence dating.

By comparing the existing loess chronologies for individual sites in Serbia with available magnetic susceptibility records (this study and Antoine et al. 2009; Buggle et al. 2009; Schmidt et al. 2010; Stevens et al. 2011; Perić et al. 2019; Orlovat is not considered due to very strong influence of local conditions (Obreht et al. 2015)), it can be stated that similarities in magnetic susceptibility variations are also chronologically coherent. Below the Holocene S0 soil, intervals with low magnetic susceptibility values are consistently dated to between 20–35 ka. In particular, this period is characterized by similarly high dust accumulation rates in the whole region. As an illustrative example, samples at ~2 m depth in all sections with comparable sedimentation rates (Batajnica, Stari Slankamen, Titel and Crvenka) show an age range between 20–30 ka, while samples at ~4.5 m are consistently dated to c. 35 ka (Schmidt et al. 2010; Stevens et al. 2011; Perić et al. 2019, and this study).

Beyond this interval (that can be tentatively related to MIS 2), all existing records in the southern Carpathian Basin are dated at considerably lower resolution (with the exception of the Titel LPS dated by Perić et al. (2019) but with fine quartz ages older than 35 ka considered unreliable), and comparison is more challenging. However, it is evident that the interval 35–40 ka reflects increased variability in magnetic susceptibility values pointing to pedogenesis phases that resulted in weak palaeosol formation (i.e. the L1SS1 according to Marković et al. 2008, 2015). The following interval is characterized by a decrease in magnetic susceptibility observed at all compared sections, but dated only at the Batajnica (this study) and Crvenka (Stevens et al. 2011) sections at c. 50 ka. Stevens et al. (2011) linked this interval to the strong local influence of Heinrich Event 4. Although this needs further confirmation, it could also reflect to some degree the impact of the Campanian Ignimbrite/Y-5 ash fall, similarly to what has been observed in the Lower Danube loess records (Veres et al. 2013; Obreht et al. 2017; Zeeden et al. 2018b). The next interval comprising loess and very weakly expressed palaeosols is characterized by another increase in magnetic susceptibility that probably represents the early MIS 3. However, no direct age estimates are available for this interval and we cannot provide further chronological constraints. Between this layer and the S1 palaeosol, a typical loess layer (L1L2) with low magnetic susceptibility has been tentatively related to MIS 4 by pedos-
tratigraphical correlation techniques (Marković et al. 2015). For the upper boundary, luminescence ages from Batajnica place L1L2 between c. 50 and 58 ka, thus rather within late MIS 4. The L1/S1 transition is placed around 74 and 84 ka based on coarse quartz and pIRIR$_{225}$ ages. Interestingly, in almost all other sections with available age control over L1/S1 (Orlovat, Crvenka, Stari Slankamen), this transition is placed around 60–70 ka. This would suggest that the S1 palaeosol as developed in loess may encompass most of MIS 5 (Marković et al. 2008, 2009, 2015), and not only the Last Interglacial optimum (MIS 5e) as previously Bronger (2003) proposed. An exception is the Surduk section where the S1/L1 transition has been placed between 75–90 ka (Fuchs et al. 2008).

Furthermore, establishing secure chronological constraints beyond the LGC is very challenging considering limitations in numerical dating, as indicated in this study. However, a more secure regional correlation can be achieved using a tepro stratigraphical approach (Veres et al. 2013; Insinga et al. 2014; Leicher et al. 2016). In southeastern Europe, regional correlation has been widely applied relying on the L2 tephra recorded in many LPS (e.g. Vandenberghe et al. 2014; Marković et al. 2015, 2018a; Obrecht et al. 2016). Samples BAT-1.13A, B and BAT-1.14A, B (Fig. 2) were collected in order to attempt constraining the depositional time of the two visible tephra layers identified within loess unit L2 (equivalent to MIS 6; Marković et al. 2015). Although glass shards were identified in both tephra layers (Fig. S8) they are altered and microprobe analyses did not allow for diagnostic geochemical fingerprinting. However, as seen in Fig. 2, the upper L2 tephra is clearly visible in the magnetic susceptibility data, and similar features have been reported in several MIS 6 loess records from the Adriatic coast to the Black Sea area (Laag et al. 2018). Based on a similar tephra report in Bulgaria, Antoine et al. (2019) proposed a correlation with Vico B-ignimbrite dated at 162±8 ka but without reporting any verifiable geochemical data in support. Therefore, it shall be stated that the upper L2 tephra could potentially be the equivalent of one of the tephra layers found in Lake Ohrid (e.g. OH-DP-0617-Vico B at 162±6 ka and OH-DP-0624-CF-V5-Pigliano Tuff at 163±22 ka; Leicher et al. 2016) and in the Fucino basin (e.g. TF-15, TF-16, TF-17; Giaccio et al. 2017) and dated to 150–160 ka. The Mediterranean Sea tephra data also document several major tephra layers between 140–170 ka (Insinga et al. 2014). Moreover, as seen also at Batajnica, at least two tephra layers are preserved regionally within the L2 loess; this highlights the risk in deriving ages without proper geochemical control, especially when at the limit of reliable luminescence dating (Table 1).

Conclusions

Luminescence dating of 63–90 µm quartz and 4–11 µm polymineral grains using the pIRIR$_{225}$ protocol provided age estimates for the LGC at Batajnica that generally agree with the previously established correla- tive chronological scheme proposed by Marković et al. (2009). The 63–90 µm quartz ages are reliable up to 100 ka before exhibiting field and laboratory saturation. The 4–11 µm quartz yields reliable ages only for MIS 2 loess samples and increasingly underestimates the true depositional time for older samples. The pIRIR$_{225}$ and pIRIR$_{290}$ protocols applied on 4–11 µm polymineral grains provide reliable ages over the LGC, beyond which they are in field and laboratory saturation. Interpretation of the pIRIR$_{225}$ and pIRIR$_{290}$ ages obtained immediately below the Last Interglacial palaeosol is hampered by different results yielded by doublet samples.

The upper limit of the luminescence dating methods at Batajnica is imposed by saturation of the natural signals starting with samples collected from L2 loess (BAT-1.13A-1.19A, B).

The observed similarities in pacing of magnetic susceptibility records in the majority of sites in northern Serbia are chronologically coherent with the Batajnica section and match global records. This allowed us to construct natural dose response curves. The comparison between the natural and laboratory dose response curves predicts the dose range over which reliable luminescence ages are obtained for each signal investigated. For 4–11 and 63–90 µm quartz, the natural and laboratory dose response curves overlap until ~150 and ~250 Gy, respectively, while for pIRIR$_{225}$ there is an apparent overlap of the natural growth curve up to at least 500 Gy. The pIRIR$_{290}$ natural and laboratory growth curves are consistent with the laboratory dose response curve up to doses of 400 Gy beyond which the natural signals overestimate the laboratory signals.

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Marković, S. B., Bokhorst, M. P., Vandenbergh, J., McCoy, W. D., Oches, E. A., Hambach, U., Gavrilov, T., Jovanović, M., Zoller, L., Stevens, T. & Machalet, B. 2008: Late Pleistocene loess-paleosol sequences in the Vojvodina region, north Serbia. Journal of Quaternary Science 23, 73–84.

Marković, S. B., Hambach, U., Catto, N., Jovanović, M., Buggle, B., Mačalet, B., Zoller, L., Glaser, B. & Frechen, M. 2009: Middle and Late Pleistocene loess sequences at Batina, Vojvodina, Serbia. Quaternary International 198, 255–266.

Marković, S. B., Hambach, U., Stevens, T., Kukla, G. J., Heller, F., McCoy, W. D., Oches, E. A., Buggle, B. & Zoller, L. 2011: The last million years recorded at the Stari Slankamen (Northern Serbia) loess-paleosol sequence: revised chronostratigraphy and long-term environmental trends. Quaternary Science Reviews 30, 1142–1154.

Marković, S. B., Oches, E., McCoy, W. D., Frechen, M. & G. A. 2007: Malacological and sedimentological evidence for “warm” glacial climate from the Irig loess sequence. Vojvodina, Serbia. Geochemistry, Geophysics, Geosystems, 8, Q09008, https://doi.org/10.1029/2006GC001065.

Marković, S. B., Oches, E., Sümegi, P., Jovanović, M. & G. A. 2006: An introduction to the Upper and Middle Pleistocene loess-paleosol sequences of Ruma section (Vojvodina, Serbia). Quaternary International 149, 80–86.

Marković, S. B., Mačalet, B., Kukla, G. J., Hambach, U., Fitzsimmons, K., Gibbard, P., Buggle, B., Zech, M., Guo, Z., Hao, Q., Wu, H., O’Hara-Dhand, K., Smalley, I., Ujvari, G., Sümegi, P., Timár-Gabor, A., Veres, D., Sirocko, F., Vasiljevíc, M. & Szírvecse, Z. 2015: Danube loess stratigraphy – Towards a pan-European loess stratigraphic matrix. Earth-Science Reviews 148, 228–258.

Marković, S. B., Stevens, T., Mason, J., Vandenbergh, J., Yang, S., Veres, D., Ujvari, G., Timár-Gabor, A., Zeeden, C., Gos, Z., Hao, Q., Obreht, I., Hambach, U., Wu, H., Gavrilov, M. B., Rolf, C., Tomić, N. & Lehmkuhl, F. 2018a: Loess correlations – Between myth and reality. Palaeogeography, Palaeoclimatology, Palaeoecology 509, 4–23.

Marković, S. B., Sümegi, P., Stevens, T., Schäetzl, R. J., Obreht, I., Chu, W., Buggle, B., Zech, M., Zech, R., Zeeden, C., Gavrilov, M. B., Perić, Z., Szírvecse, Z. & Lehmkuhl, F. 2018b: The Cretaean loess-paleosol sequence: a record of continuous grassland domination during the Late Pleistocene, for the southern Carpathian Basin. Palaeogeography, Palaeoclimatology, Palaeoecology 509, 33–46.

Marković, S. B., Timár-Gabor, A., Stevens, T., Hambach, U., Popov, D., Tomić, N., Obreht, I., Jovanović, M., Lehmkuhl, F., Keh, H., Marković, R. & Gavrilov, M. B. 2014: Environmental dynamics and luminescence chronology from the Orlóvat loess–paleosol sequence (Vojvodina, northern Serbia). Journal of Quaternary Science 29, 189–199.

Mejdahl, V. 1979: Thermoluminescence dating: beta-dose attenuation in quartz grains. Archaeometry 21, 61–72.

Murray, A. S. 1996: Developments in optically stimulated luminescence dating: application to a 2000-years of flood deposits. Quaternary Science Reviews 89, 563–576.

Murray, A. S. & Wintle, A. G. 2000: Luminescence dating using an elevated temperature post-IR IRSL protocol. Quaternary Science Reviews 19, 498–520.

Obreht, I., Zeeden, C., Schulte, P., Hambach, U., Eckmeier, E., Timár-Gabor, A. & Lehmkuhl, F. 2015: Aeolian dynamics at the Orlóvat loess–paleosol sequence, northern Serbia, based on detailed textural and geochemical evidence. Aeolian Research 18, 69–81.

Perić, Z., Adophi, E. L., Buylaert, J. P., Stevens, T., Ujvari, G., Marković, S. B., Hambach, U., Fischer, P., Zeeden, C., Schmidt, C., Schulte, P., Huayu, L., Shuangwen, Y., Lehmkuhl, L., Obreht, I., Veres, D., Vott, A. & Zoller, L. 2019: Quartz OSL dating of late Quaternary Chinese and Serbian loess: a cross Eurasian comparison of dating results and mass accumulation rates. Quaternary International 592, 30–44.

Petrusić, Z., Jovanović, U., Jovanović, I. & Mancić, D. 2011: Realization and calibration of the wireless UV radiation measurement system. Contemp Or Cos (Renewable energy sources) 2, 164–170.

Prescott, J. R. & Hutton, J. T. 1994: Cosmic ray contributions to dose rates for luminescence and ESR dating: large depths and long term variations. Radiation Measurements 22, 497–500.

Rees-Jones, J. 1995: Optical dating of young sediments using fine-grain quartz. Ancient TL 13, 9–13.

Roberts, H. 2008: The development and application of luminescence dating to loess deposits: a perspective on the past, present and future. Boreas 37, 483–507.

Schäetzl, R. J., Betts, E. A., Crouvie, O., Fitzsimmons, K. E., Grimley, D. A., Hambach, U., Lehmkuhl, F., Marković, S. B., Mason, J. A., Owczarek, P., Roberts, H. M., Rousseau, D. D., Stevens, T., Vandenberghen, J., Zate, M., Veres, D., Yang, S. L., Zech, M., Conroy, J. L., Dave, A. K., Faust, D., Hao, Q. Z., Obreht, I., Prud’homme, C., Smalley, I., Trpali, A., Zeeden, C. & Zech, R. 2018: Approaches and challenges to the study of loess - introduction to the Loess-Fest. Quaternary Research 89, 563–618.

Schmidt, E., Mačalet, B., Marković, S. B., Tsukamoto, S. & Frechen, M. 2010: Luminescence chronology of the upper part of the Stari Slankamen loess sequence (Vojvodina, Serbia). Quaternary Geochronology 5, 137–142.

Stevens, T., Buylaert, J.-P., Thiel, C., Ujvari, G., Yi, S., Murray, A. S., Frechen, M. & Lu, H. 2018: Ice-volume-forced of the Chinese Loess Plateau global Quaternary stratotype site. Nature Communications 9, 983, https://doi.org/10.1038/s41467-018-03329-2.

Stevens, T., Marković, S. B., Zech, M., Hambach, U. & Sümegi, P. 2011: Dust deposition and climate in the Carpathian Basin over an independently dated last glacial-interglacial cycle. Quaternary Science Reviews 30, 662–681.

Sümegi, P., Gulyás, S., Molnár, D., Sümegi, B. P., Almond, P. C., Vandenberghen, J., Zhou, L., Pál-Molnár, E., Töröcsik, T., Hao, Q., Smalley, I., Molnár, M. & Marsi, I. 2018: New chronology of the best developed loess/paleosol sequence of Hungary capturing the past 1.1 ma: implications for correlation and proposed pan-Eurasian stratigraphic schemes. Quaternary Science Reviews 191, 144–166.

Thiel, C., Buylaert, J.-P., Murray, A., Terhorst, B., Hölzer, I., Tsukamoto, S. & Frechen, M. 2011a: Luminescence dating of the Stratzing loess profile (Austria) – Testing the potential of an elevated temperature post-IR IRSL protocol. Quaternary International 234, 23–31.

Thiel, C., Buylaert, J.-P., Murray, A. S. & Tsukamoto, S. 2011b: On the applicability of post-IR IRSL dating to Japanese loess. Geochronometria 38, 369–378.

Thomsen, K. J., Murray, A. S., Jain, M. & Better-Jensen, L. 2008: Laboratory fading rates of various luminescence signals from feldspar-rich sediment extracts. Radiation Measurements 43, 1474–1486.

Timár-Gabor, A. & Wintle, A. G. 2013: On natural and laboratory generated dose response curves for quartz of different grain sizes from Romanian loess. Quaternary Geochronology 18, 34–40.

Timár-Gabor, A. & Buylaert, J.-P., Guralnik, B., Trandafir-Antohi, O., Constantin, D., Anecheit-Deacu, V., Jain, M., Murray, A. S., Porat, N., Hao, Q. & Wintle, A. G. 2017: On the importance of grain size in
luminescence dating using quartz. *Radiation Measurements* 106, 464–471.

Timar-Gabor, A., Constantin, D., Marković, S. B. & Jain, M. 2015: Extending the area of investigation of fine versus coarse quartz optical ages from the Lower Danube to the Carpathian Basin. *Quaternary International* 388, 168–176.

Timar-Gabor, A., Vandenberghe, D. A. G., Vasiliniuc, S., Panaiootu, C. E., Dimolfte, D. & Cosma, C. 2011: Optical dating of Romanian loess: a comparison between silt-sized and sand-sized quartz. *Quaternary International* 240, 62–70.

Timar-Gabor, A., Vasiliniuc, S., Vandenberghe, D. A. G., Cosma, C. & Wintle, A. G. 2012: Investigations into the reliability of SAR-OSL equivalent doses obtained for quartz samples displaying dose response curves with more than one component. *Radiation Measurements* 47, 740–745.

Vandenberghe, D. A. G., De Corte, F., Buylaert, J.-P., Ku Timar-Gabor, A., Vasiliniuc, S., Yi, S., Buylaert, J.-P., Murray, A. S., Lu, H., Thiel, C. & Zeng, L. 2016: A review of quartz optically stimulated luminescence ages from the Lower Danube to the Carpathian Basin. *Quaternary International* 494, 17–33.

Wintle, A. G. & Murray, A. S. 2012: Investigations into the reliability of SAR-OSL equivalent doses obtained for quartz samples displaying dose response curves with more than one component. *Radiation Measurements* 47, 740–745.

Vandenberghe, D. A. G., De Corte, F., Buylaert, J.-P., Ku Timar-Gabor, A., Vasiliniuc, S., Yi, S., Buylaert, J.-P., Murray, A. S., Lu, H., Thiel, C. & Zeng, L. 2016: A review of quartz optically stimulated luminescence ages from the Lower Danube to the Carpathian Basin. *Quaternary International* 494, 17–33.

Vandenberghe, J., Marković, S. B., Jovanović, M. & Hambach, U. 2014: Site-specific variability of loess and palaeosols (Ruma, Vojvodina, northern Serbia). *Quaternary International* 334–335, 86–93.

Vasiliniuc, S., Vandenberghe, D. A. G., Timar-Gabor, A., Panaiootu, C. & Cosma, C. 2012: Testing the potential of elevated temperature post-IR IRSL signals for dating Romanian loess. *Quaternary Geochronology* 10, 75–80.

Veres, D., Lane, C. S., Timar-Gabor, A., Hambach, U., Constantin, D., Szakacs, A., Fülling, A. & Onac, B. P. 2013: The Campanian Ignimbrite/Y5 tephra layer – A regional stratigraphic marker for Isotope Stage 3 deposits in the Lower Danube region, Romania. *Quaternary International* 293, 22–33.

Veres, D., Tesa, V., Gerasimenko, N., Zeeden, C., Hambach, U. & Timar-Gabor, A. 2018: Short-term soil formation events in last glacial east European loess, evidence from multi-method luminescence dating. *Quaternary Science Reviews* 200, 34–51.

Wach, L. & Frechen, M. 2011: The geochronology of the “Gorjanović loess section in Vukovar, Croatia. *Quaternary International* 240, 87–99.

Wallinga, J., Murray, A. & Duller, G. 2000: Underestimation of equivalent dose in single-aliquot optical dating of feldspars caused by preheating. *Radiation Measurements* 32, 691–695.

Wintle, A. G. & Murray, A. S. 2006: A review of quartz optically stimulated luminescence characteristics and their relevance in single-aliquot regeneration dating protocols. *Radiation Measurements* 41, 369–391.

Yi, S., Buylaert, J.-P., Murray, A. S., Lu, H., Thiel, C. & Zeng, L. 2016: A detailed post-IR IRSL dating study of the Niuyangzigu loess site in northeastern China. *Boreas* 45, 644–657.

Zeeden, C., Hambach, U., Obrecht, I., Hao, Q., Abels, H., Veres, D., Lehmkuhl, F., Gavrilov, M. & Marković, S. B. 2018a: Patterns and timing of loess-palaeosol transitions in Eurasia: constraints for palaeoclimate studies. *Global and Planetary Change* 162, 1–7.

Zeeden, C., Hambach, U., Veres, D., Fitzsimmons, K., Obrecht, I., Böskén, J. & Lehmkuhl, F. 2018b: Millennial scale climate oscillations recorded in the Lower Danube loess over the last glacial period. *Palaeogeography, Palaeoclimatology, Palaeoecology* 509, 164–181.

Zhang, J., Rolf, C., Wach, L., Tsukamoto, S., Durr, G. & Frechen, M. 2018: Luminescence dating and palaeomagnetic age constraint of a last glacial loess-palaeosol sequence from Istria, Croatia. *Quaternary International* 494, 19–33.

**Supporting Information**

Additional Supporting Information may be found in the online version of this article at http://www.boreas.dk.

**Fig. S1.** The ratio between $^{210}$Pb concentration determined by gamma spectrometry and $^{226}$Ra/$^{214}$Pb and $^{214}$Bi peaks) concentrations determined by gamma spectrometry.

**Fig. S2.** The ratio between $^{210}$Pb concentration determined by gamma spectrometry and $^{210}$Po concentration measured by alpha spectrometry.

**Fig. S3.** Representative sensitivity-corrected dose response curves constructed for sample BAT-1.10 using one aliquot of (A) fine (4–11 μm) quartz grains, (B) coarse (63–90 μm) quartz grains and (C, D) polymineral fine (4–11 μm) grains.

**Fig. S4.** Equivalent dose dependence on preheat temperatures for fine quartz fraction from sample BAT-1.13B.

**Fig. S5.** Results of the fading rate measurements on individual aliquots of 4–11 μm polymineral material and 4–11 and 63–90 μm quartz.

**Fig. S6.** Luminescence ages of quartz and polymineral fine grains.

**Fig. S7.** Average values for the natural sensitivity-corrected luminescence signals for all the investigated aliquots.

**Fig. S8.** Glass shards identified in the upper tephra layer (A) and lower tephra layer (B) in the L2 unit.

**Table S1.** Flowchart of the SAR-OSL (Murray & Wintle 2000, 2003), pIRIR290 (Buylaert et al. 2011b, 2012; Thiel et al. 2011a) and pIRIR225 (Buylaert et al. 2009; Wach & Frechen 2011; Vasiliniuc et al. 2012) protocols applied in this study.

**Table S2.** Information on the samples used for the construction of the natural dose response curves.

**Table S3.** Summary of dosimetry data.

**Table S4.** The measured equivalent doses along with the performance parameters of the SAR-OSL protocol as well as for pIRIR290 and pIRIR225 for each quartz grain-size and polymineral fine grains.

**Table S5.** The measured residual doses along with the performance parameters of the pIRIR290 procedure for fine (4–11 μm) polymineral grains of the 10 samples analysed.

**Table S6.** The measured residual doses along with the performance parameters of the pIRIR225 procedure for fine (4–11 μm) polymineral grains of the nine samples analysed.

**Table S7.** $g$-values measured on polymineral fine grains using the pIRIR290 and pIRIR225 protocols as well as on fine and coarse quartz using the SAR-OSL protocol.

**Table S8.** The effect of adding large doses on top of the naturally accrued dose for sample BAT-1.19A using SAR-OSL, pIRIR225 and pIRIR290 protocols.
Table S9. Maximum corrected luminescence signals in the SAR-OSL and pIRIR protocols used and natural corrected luminescence signals for the sample BAT-1.19A.

Table S10. Saturation characteristics for dose response curves up to 5000 Gy on BAT-1.19A using different test doses.

Table S11. Natural corrected luminescence signals for the samples BAT-1.12A–BAT-1.19B and maximum corrected luminescence signals recorded in the SAR-OSL and post-IR IRSL protocols.

Data S1. Sample preparation for alpha spectrometry.