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Extending the area of investigation of fine versus coarse quartz optical ages from the Lower Danube to the Carpathian Basin

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Despite the general satisfactory performance of quartz in the single aliquot regeneration protocol (SAR), previous optically stimulated luminescence (OSL) dating studies of key loess sections in Romania, Lower Danube region, revealed a disturbing disagreement among the ages obtained on fine (4–11 μm) grains and coarse (63–90 μm) grains respectively. The current study aims at expanding these investigations, both by extending the area of study from the Lower Danube Basin to the Carpathian Basin and by applying time-resolved optically stimulated luminescence (TR-OSL) on quartz, in order to gain further insights into the above mentioned behaviour. The samples from the Orlovat loess paleosol section (Vojvodina, Serbia) showed a similar behaviour to that previously reported on Romanian loess. A marked difference between the dose saturation characteristics of fine and coarse quartz OSL signals is observed for both continuous wave (CW-OSL) and pulsed OSL (POSL), where the dose response (up to 1000 Gy) is well described by a sum of two saturating exponential functions. TR-OSL measurements show one single, characteristic quartz lifetime for both natural as well as regenerative signals in the entire dose range investigated. A general disagreement between the ages obtained on the two grain sizes for samples with equivalent doses higher than about 100 Gy is reported as in the case of Romanian loess, inferring that the age discrepancy between the two grain sizes might be more widespread than previously thought.

1. Introduction

The loess paleosol sequences of the Carpathian Basin and Lower Danube region (Serbia, Romania, Bulgaria) are thought to represent the most continuous and high resolution archives of regional climate and environmental change during the Late and Middle Pleistocene (Fitzsimmons et al., 2012), and a link between European and Asian loess deposits (Marković et al., 2012). Their significance however, can only be fully understood only once a reliable and absolute chronology is available.

Technological developments in the past years have resulted in significant improvements in the achievable precision and accuracy using luminescence dating. It is now considered one of the most important chronometric methods in the study of the late Quaternary (Wintle, 2008). However, dating applications recently performed on Romanian loess by the application of optically stimulated luminescence on quartz led to unexpected results.

A first study has reported optically stimulated luminescence (OSL) ages for the loess sequence near Mircea-Vodă (Dobrogea, SE Romania) using silt-sized (4–11 μm) quartz as dosimeter (Timar et al., 2010). An internally consistent set of optical ages was obtained; however, a comparison of these ages with a magnetic time-depth model based on magnetic susceptibility measurements suggested a systematic underestimation beyond the penultimate glacial period (the SAR OSL ages of the three samples below the S1 soil were interpreted as age underestimates). Interestingly, the OSL signals from these samples did not indicate any odd characteristics: the dominance of the fast component in the OSL signals was indicated by the decay shape of the CW-OSL and LM-OSL signals; and the samples passed the procedural tests of the single-aliquot regenerative-dose (SAR) protocol (i.e. recycling ratio, recuperation, dose recovery and preheat plateau tests) indicating that the protocol should provide reliable results, while the natural signals were found well below the saturation region of the laboratory dose response.

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It was concluded that optical dating of fine-grained quartz can be used to establish a reliable chronology for Romanian loess up to ~70 ka corresponding to an equivalent dose of ~200 Gy. Such a behaviour is consistent with results from old (>70 ka) Chinese loess (Buylaert et al., 2007), and with the more general suggestion that SAR may underestimate the true age in the older age range (Murray et al., 2007; Lowick et al., 2010a,b, 2011). Thus, apparently reliable OSL laboratory measurement procedure does not necessarily guarantee an accurate determination of the true burial dose. A subsequent study of coarse-grained (63–90 μm) quartz extracted from the same section (Timar-Gabor et al., 2011) revealed that equivalent doses obtained for coarse quartz grains were systematically larger than those for the fine grains; the observed difference was too high to be explained by partial bleaching or microdosimetric effects. Furthermore, both quartz fractions passed the procedural tests of the single-aliquot regenerative-dose (SAR) protocol and yielded an internally consistent set of optical ages.

Timar-Gabor et al. (2012) investigated into the shape of the dose response curve for the two grain size fractions in the high dose region of 5–10 kGy; they observed an age discrepancy in the Mostiştea section (Romanian plain) similar to that in the Mircea-Vodă. Work on Costinesti and Lunca sections (Dobrogea, SE Romania) (Constantin et al., 2015) confirms the same trend. From the study of Timar-Gabor et al. (2012) two additional issues of general importance to the SAR protocol emerged. Firstly the natural signal of an infinitely old sample was found not to be in saturation, perhaps implying that the dose response measured in the laboratory may not simulate trapped charge growth during burial; this aspect was further investigated by Timar-Gabor and Wintle (2013). Secondly, it was observed that the dose response curve for coarse grains (63–90 μm) is very different from fine grains (4–11 μm), the latter saturating at much higher doses. A similar observation has also been made by Constantin et al. (2012) where dose response curves of fine grains (4–11 μm) and coarse grains of different sizes (63–90, 90–125, 125–180 μm) were compared for Căciulăteşti site (SW Romania). This different dose response pattern was also reported for quartz extracted from loess in Western Europe by Kreutzer et al. (2012). This is mostly intriguing in correlation to the fact that the fine fraction underestimates the true ages earlier than the coarse fraction does. Also, the values obtained for the saturation characteristics for the fine quartz fraction are very similar to the values reported by other international studies (e.g. Lowick et al., 2010b) in their work on fine material, while the saturation characteristic doses obtained in this study for coarse grained quartz are close to the values previously reported by others on coarse material from other locations (e.g. Murray et al., 2007; Pawley et al., 2010).

In a geochemical characterisation study performed by Buggle et al. (2008) it was confirmed that both Serbian and Dobrudjan loess have a major component derived from Danube alluvium. As second material sources, loess in the Dobrudja region showed a significant contribution of a second loess source, probably the glaciofluvial sediments of the Ukraine, to which variable contributions from local sand dune fields can be considered. Although the different origin of the sedimentary grains in loess should not justify the difference in equivalent doses that leads to a systematic age offset between the coarse and fine grains ages, it should be taken into consideration whether the different properties of fine and coarse grains can be due to their possible different origin. In the present study we are extending the investigated area by analysing whether the above mentioned finds are more than a local feature of the eastern part of the Lower Danube basin or do apply to the Carpathian Basin loess as well.

2. Samples, instrumentation and measurement protocol

The present work focuses on eight loess samples collected form a loess–paleosol sequence exposed in a brickyard at the village of Orlovat, located in the Tamiš loess plateau (Fig. 1). Out of these, samples ORL-8 to ORL-4 have been collected from L1 unit, samples ORL 3 and ORL 2 have been sampled form S1 unit, while sample ORL 1 belongs to L2 unit, thus a total age span of more than 130 ka being covered (Fig. 2). The importance of the Orlovat section as a key archive for the late Pleistocene paleoclimate and paleoenvironment of the southeastern part of the Carpathian Basin has been emphasized by previous multi-proxi studies (Lukić et al., 2014; Marković et al., 2014) to which the reader is referred to for more information.

OSL dating of coarse (63–90 μm) quartz was also documented in Marković et al. (2014). Fine quartz extraction followed the conventional procedures described in our previous studies (Timar et al., 2010; Timar-Gabor et al., 2011; Constantin et al., 2014, 2015). All continuous wave optically stimulated luminescence measurements (CW-OSL) were made in the Cluj Luminescence Dating Laboratory with a Risø TL/OSL DA-20 reader equipped with blue light emitting diodes (LEDs) emitting at 470 ± 30 nm and IR LEDs emitting at 875 ± 80 nm; luminescence signals were observed through a 7.5 mm thick Hoya U-340 UV filter. Irradiations have been carried out using a ^{90}Sr–^{90}Y beta source, calibrated using fine and coarse quartz supplied by Risø National Laboratory. The dose rate, for the time of measurement, determined for coarse grains mounted on stainless steel disks was 0.147 Gy/s, while the dose rate for fine grains deposited on aluminium disks amounted to 0.118 Gy/s. Details of the measurement apparatus can be found in Thomsen et al. (2006). The OSL signal was collected in time intervals of 0.154 s. All samples have been analysed in a SAR protocol (Murray and Wintle, 2000, 2003). The OSL signal used for analysis was that obtained for the first 0.308 s of the decay curve minus a background derived from the signal measured between 2.464 and 3.080 s, as had been used in previous studies on Romanian loess (Timar et al., 2010; Timar-Gabor et al., 2011, 2012; Timar-Gabor and Wintle, 2013) and the recommendations of Cunningham and Wallinga (2010). Natural and regenerated signals were measured after a preheat of 10 s at 220 °C unless otherwise stated; the response to the test dose (16 Gy) was measured after a cutheat to 180 °C. The value of the test dose was kept constant through all measurements for both grain sizes of quartz. After the measurement of the response to the test dose, a high-temperature bleach was performed by stimulating with the blue diodes for 40 s at 280 °C (Murray and Wintle, 2003). Time resolved optically stimulated luminescence (TR-OSL) experiments have been carried out in Risø National Laboratory on a Risø TL/OSL-20 (their reader V) equipped with an integrated pulsing option to control the LEDs, and a Photon Timer attachment to record the TR-OSL described in detail in Lapp et al. (2009). All TR-OSL experiments have been carried out using the same measurement parameters previously used in CW-OSL. For stimulating and recording the TR-OSL signals the total measurement time was set to 100 s, with a pulse period of 500 μs consisting of an on time of 50 μs and an off time of 450 μs; this can be translated to a net stimulation period equivalent to 10 s in CW-OSL. A total of 500 data points were used for data collection in pulsed stimulation out of which the first and the last five channels were dead channels. Measurement of dose response curves using pulsed optically stimulated luminescence measurements have been specially designed in order to reproduce the CW-OSL measurements conditions as closely as possible, and to minimise any possible feldspar contamination to quartz, by: (i) selection of an on time of 50 μs (ii) gating the photomultiplier for counting only during the off period; (iii) ignoring the first 4 μs during the off time in TR-OSL OSL data analysis (Ankjærgaard et al., 2010).