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Spatial differences in the luminescence sensitivity of quartz extracted from Carpathian Basin fluvial sediments

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ABSTRACT

The luminescence sensitivity of a sample is the luminescence intensity measured in response to unit radioactive dose. Sensitivity is by no means a stable parameter, it might change during measurements, or in nature as well. The primary or natural magnitude of luminescence sensitivity is basically determined by mineralogical background (number of crystal impurities) and sedimentary prehistory (repeated exposure of the sediment to sunlight).

In the present study we have investigated the luminescence properties and sensitivity of coarse-grain (90–150 μm) quartz samples related to four major rivers of the Carpathian Basin (River Danube, Tisza, Szamos and Maros). In case of each region of interest 5 previously dated Late Pleistocene and/or Holocene samples were selected, each representing similar sedimentary environments, i.e. coarse grain channel deposits related to point bars and medial bars. Sensitivity was investigated using CW-OSL, LM-OSL and TL techniques using a multi-grain approach. By determining the normalised luminescence response to the same regeneration dose administered after bleaching, sensitivity base values were obtained for each sample. Using repeated cycles of dosing laboratory sensitivity change was also recorded. The base values and sensitivity change of the 20 investigated samples were then compared on a regional basis to identify potential differences, which might be used later for fingerprinting the sediments of the investigated rivers.

When considering mean sensitivity base values, calculated from several aliquots of the same sample, Danube related, mostly Alpine origin sediments exhibited 50–60% lower values compared to those with a Carpathian origin, and even at the considerable standard deviation obtained (coefficient of variation being 25–60%) they could be clearly separated using any of the measured luminescence sensitivity parameters. The discrimination of fluvial sediments with a Carpathian origin, but representing different catchments, is less straightforward, though, plotting against different sensitivity parameters can offer an opportunity to define fairly distinct groups of sample mean values. From this aspect total LM-OSL and fast component ratio seemed to be the best candidates, however, at the characteristic standard deviation and standard error separation can be unclear. No clear relationship was found in terms of sensitivity change, however some samples, related to River Maros showed practically no change during the laboratory sensitisation process. When plotting OSL ages against quartz sensitivity clear trends could be recognised, which can partly be explained by geomorphological reasons. Results in all, point to the possibility to differentiate Carpathian Basin fluvial sediments on the basis of their quartz luminescence sensitivity, a parameter that can be assessed easily during the routine dating process.

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1. Introduction

Quartz, being a natural dosimeter is an ultimately important mineral in luminescence dating. It occurs in almost every sedimentary environments and it has very advantageous properties in terms of trapped charge lifetimes and reproducibility of its luminescence signal (Aitken, 1998; Bøtter-Jensen et al., 2003). It is basically built up by SiO_4 tetrahedra, but natural quartz is by no means ideal in the sense that crystallographic defects, i.e. substitutions, interstitial elements and vacancies are very frequent in its lattice. For instance Si^{4+} can be substituted by isoelectronic and nonisoelectronic trace element cations, such as Ti^{4+}, Al^{3+}, Fe^{3+}, P^{5+}, which can enhance the incorporation of interstitial Li^{+}, Na^{+}, K^{+}, Cu^{+}, Ag^{+}, and at the same time O^{-} might miss from Si–O–Si bridge bonds, all leading to the presence of positively charged impurities (Krbetschek et al., 1997; Preusser et al., 2009). These impurities, developing in function of the temperature and pressure relevant during the crystallisation process, are responsible for the primary charge trapping capacity of the lattice and determine the magnitude of luminescence intensity measured in response to absorbed unit dose (Sharma et al., 2017), which can be termed with other words as luminescence sensitivity (Sawakuchi et al., 2011; Gray et al., 2019). Thus, subcrystalline properties inherited from the source rock can fundamentally determine TL and OSL behaviour of quartz recovered from various sedimentary environments (see e.g. Sawakuchi et al., 2011; Tsukamoto et al., 2011; Lü et al., 2014; Zular et al., 2015).

However, it was recognised early that during TL dating the luminescence response of quartz is not at all a constant parameter (Fleming, 1970), and for example artificial dosing can lead to the sensitisation of grains (Zimmerman, 1971). It was also observed that annealing or thermal treatment (Aitken, 1985; Bøtter-Jensen et al., 1995) and bleaching or exposure to light (Wintle, 1985; Li and Wintle, 1991; Li, 2002) can also lead to sensitivity changes, and can affect not only the TL but the OSL of quartz as well (Wintle and Murray, 1999). Due to these issues the monitoring and correction of sensitivity change has become a crucial point of single aliquot regeneration protocols (Murray and Wintle, 2000).

When studying the phenomena above, Li and Wintle (1992) have also raised that differences in luminescence sensitivity experienced in the laboratory might be related to the extension and degree of sunlight exposure prior to the deposition of grains. Namely, sensitivity can be enhanced in nature as well, most likely as a consequence of recurring sedimentary cycles and long transportation distance (Fitzsimmons, 2011; Wintle and Adamiec, 2017). For example, the high luminescence sensitivity of Australian sediments is usually explained by the extensive and the repeated reworking of quartz grains (Pietsch et al., 2008; Fitzsimmons et al., 2010; Fitzsimmons, 2011). Similarly, Preusser et al. (2006) claimed that the low luminescence sensitivity of sediments in New Zealand is primarily caused by their short sedimentary history. These findings also suggest that the farther the quartz grains get from their source the higher natural sensitivity they can have. Accordingly, when investigating the modern sediments of Australian rivers, Pietsch et al. (2008) and Gliganic et al. (2017) observed an increase of luminescence sensitivity with downstream transport distance. However, no such tendency was detected in terms of the Amazon River Basin (Sawakuchi et al., 2018), where differences and downstream change of quartz sensitivity could rather be related to the lithological background of subcatchments (Sawakuchi et al., 2012). Beside sediment cycling, the mode of sediment transport can also be of significance in determining the natural sensitivity of grains (Li and Wintle, 1992), though, in her study on Australian quartz from various depositional environments Fitzsimmons (2011) found no systematic sensitivity difference between aeolian and water-lain sediments.

Consequently, there is no uniform model for explaining the degree of sensitivity in terms of natural quartz, thus, sensitivity is determined by the combination of primary, i.e. petrological and mineralogical, and secondary, i.e. geomorphological and sedimentological parameters (Gray et al., 2019). In other words, the complexity of quartz crystals and the complexity of their deposition history both play a major role in determining luminescence sensitivity (Preusser et al., 2009; Fitzsimmons, 2011), however, the importance of the different driving factors seems to be site dependent. Still, based on several previous attempts, cited above, quartz luminescence sensitivity seems to be an adequate tool for sediment fingerprinting in sedimentary basins if 1) potential sources have a considerably different geological background, or 2) grains are transported at a different tempo or by different mechanisms to the deposition site.

Luminescence sensitivity and its change can be quantified either during the SAR dating process by recording the luminescence responses to uniform test doses applied after incrementally increasing regeneration doses (e.g. Fitzsimmons, 2011) or by applying repeated cycles of the same regeneration dose (e.g. Nian et al., 2019). Sensitivity can be assessed using different approaches: 1) simply by recording the first part of continuous wave (CW) OSL decay curves (Wintle and Murray, 2006), 2) by applying linearly modulated (LM) OSL (Bulur 1996), which also enables the breaking up of the OSL decay curve into components (fast, medium and slow) (Jain et al., 2003; Singarayer and Bailey, 2003), or 3) by using the 110 °C TL peak, since several studies observed a correlation between TL and OSL signals (Stoneham and Stokes, 1991; Stokes, 1994; Murray and Roberts, 1998; Chen et al., 2000; Wintle and Murray, 1999; Singhvi et al., 2011). Nevertheless, building up the relationship between TL and OSL can be complicated due to several reasons, such as differences in activated traps, thermal and phototransfer effects (Bøtter-Jensen et al., 2003). The 110 °C TL peak, however proved to be an adequate parameter to investigate the source area of quartz samples at some locations (Preusser et al., 2006; Pietsch et al., 2008; Lü and Sun, 2011; Sawakuchi et al., 2011, 2012; Thomsen et al., 2018). Furthermore, the TL 110 °C TL peak and the components separated by LM-OSL were also found to be in correspondence (Kiyak et al., 2007).

The assessment of luminescence sensitivity is complicated by the fact that it can have a rather significant grain to grain variability even within the same sample, as it was observed by several single grain studies earlier (e.g. Fitzsimmons, 2011; Chauhan and Singhvi et al., 2011). High variability is frequently attributed to the presence of super sensitive grains (Arnold et al., 2016), which are usually handled as outliers when calculating representative mean values (Zheng et al., 2009). However, as indicated by Fitzsimmons (2011), the presence or lack of such grains can also be an indicator of sediment provenance. Including these to the analysis by applying multi-grain measurements may increase the scatter of results (Fitzsimmons et al., 2010), but on the other hand, being the part of the quartz mixture, they can also contribute to the detection of differences in provenance. Consequently, the multi-grain method can also be applied successfully, if there are significant source dependant differences in sensitivity as shown in the studies of Lü et al. (2014) or Nian et al. (2019).

The present study focuses on the analysis of the luminescence sensitivity and the laboratory sensitivity change of quartz grains related to major rivers of the Carpathian Basin, draining water and depositing sediments coming either from the Alps or the Carpathians, being among the most extensive mountain systems in Europe. Concerning the luminescence sensitivity of these fluvial sediments only few data are available. The idea of this study has emerged from the observation that the luminescence sensitivity of modern coarse grain (90–150 μm) quartz along the Hungarian section of the Danube proved to be remarkably low in the study of Tóth et al. (2017a) and made the assessment of residual doses difficult. Similarly, samples from the Alps and also from the Tatra Mountains (Western Carpathians) exhibited low sensitivity values (Klasen et al., 2006; 2007; Moska and Murray, 2006; Trauerstein et al., 2017). However, no luminescence sensitivity data are available from the Eastern Carpathians and the catchment of the Tisza River, the largest tributary of the Danube, and no comparison has been made between the two major rivers of the basin before. Consequently, by applying several sediment samples dated earlier (Kiss et al., 2014; Sipos et al., 2016; Tóth

et al., 2017a, 2017b; Robu, 2018), OSL sensitivity and sensitivity change were assessed and compared systematically, using samples with similar geomorphological and sedimentary background, and applying identical measurement procedures. This way we aimed to reveal the spatial difference of sensitivity parameters on a basin-scale, and to investigate the potential of these parameters in sediment sourcing.

2. Study area and samples

The Carpathian Basin, located in East-Central Europe, and encompassed by the mountain chains of the Eastern Alps, the Carpathians and the Dinarides, is the largest intermountain basin of Europe. It was formed by the thrusting of the surrounding mountain systems as part of the Alpine orogeny starting from the Late Mesozoic. The catchment of its main river the Danube (catchment area: 801 000 km^2, river length: 2850 km, Q_{mean} at the estuary: 6500 m^3/s) and that of its largest tributary, the Tisza (catchment area: 157 000 km^2, river length: 960 km, Q_{mean} near the confluence: 820 m^3/s) span over a great variety of rock types, which can obviously affect the behaviour of quartz grains arriving to the basin.

The catchment of the Upper Danube, stretching upstream of the Carpathian Basin, belongs to the Schwab Alps, the Eastern Alps, the Bohemian Massif and the Western Carpathians (Fig. 1). In its southern part, with a drainage towards the Upper Danube, the Schwab Alps is built up by Jurassic limestone (Gwinner, 1976). The surface lithology of the Northern Calcareous Alps is also dominated by a Mesozoic cover (Fig. 1), however, in its contact zone to the Central Eastern Alps a 20–30 km wide stripe of Paleozoic metamorphosed sedimentary rock zone (Greywacke zone) is situated (Schmid et al., 2004). In relation with the Late Mesozoic thrusting of the Alps the Upper Austroalpine nappe complex of the Central Eastern Alps is comprised of metamorphic rocks of both low grade non-eclogitic and high grade eclogitic type (Kurz et al., 1999), though, their share from the Upper Danube catchment is not significant. The southern flank of the Bohemian Massif is characterised primarily by Paleozoic metamorphic and intrusive crystalline rocks (Medaris et al., 1995). Compared to the previous regions the Western Carpathians is the most complex in surface lithology (Hók et al., 2019; Kłapyta, 2020), thus, the weathering of a wide variety of sedimentary, igneous and metamorphic rocks can provide quartz grains to the sediment of the Danube.

The Tisza River drains the waters of the entire Inner Eastern Carpathians, and by its tributaries it also collects water and sediments from the Western Carpathians, the Southern Carpathians, the Apuseni Mountains and the Transylvanian Basin. The headwaters of the Tisza are located in the Northeastern Carpathians where surface lithology is characterised by flysch, composed of both coarse and fine grain siliciclastic marine sediments deposited in the Late Mesozoic and Paleogene, and thrusted during the late Alpine orogeny (Ślączka et al., 2006). Besides, Miocene volcanic/pyroclastic rocks, produced as a result of post-collisional volcanism (Harangi and Lenkey, 2017), and Miocene-Pliocene sedimentary rocks comprise the Upper Tisza catchment. The crystalline metamorphic units of the Eastern Carpathians (Munteanu and Tatu, 2003) and the Apuseni Mountains is connected to the Tisza through the Szamos, Körös and Maros rivers (Pál-Molnár et al., 2015), however, the largest amount of fluvial sediment originates from the Neogene sedimentary rocks of the Transylvanian Basin, and the predominantly high and medium grade metamorphic rocks of the Southern Carpathians (Iancu and Seghedi, 2017).

The Carpathian Basin lowlands has been filled up primarily by alluvial sediments since the end of the Miocene, and the fluvial network has undergone a complex development governed by tectonic, geomorphic and climatic forces. Consequently, the course of rivers changed significantly on the alluvial plains of the basin throughout the Pliocene and Pleistocene. This compound history has been addressed by several authors (e.g. Nádor et al., 2007; Vandenberghe et al., 2007; Gábris et al., 2012; Kiss et al., 2014), still there are some questions that can only be answered by the sourcing of sediments, especially in the southeastern part of the basin, where the main rivers meet.

Samples used for the general comparison of quartz sensitivity within the Carpathian Basin were chosen from previously dated samples representing the sediments of the Danube, Tisza, Szamos and Maros Rivers (Fig. 1 and Table 1). In total 20 samples were selected, and on a regional basis they can be classified into four groups, each comprising 5 samples. The first group represents the middle course of the Danube, where one modern sample and four Holocene palaeomeander and floodplain samples were subjected to the analyses (Tóth et al., 2017a, 2017b). The second group is located on the Upper Tisza catchment and contains three Tisza and two Szamos River samples, each being Holocene in age and

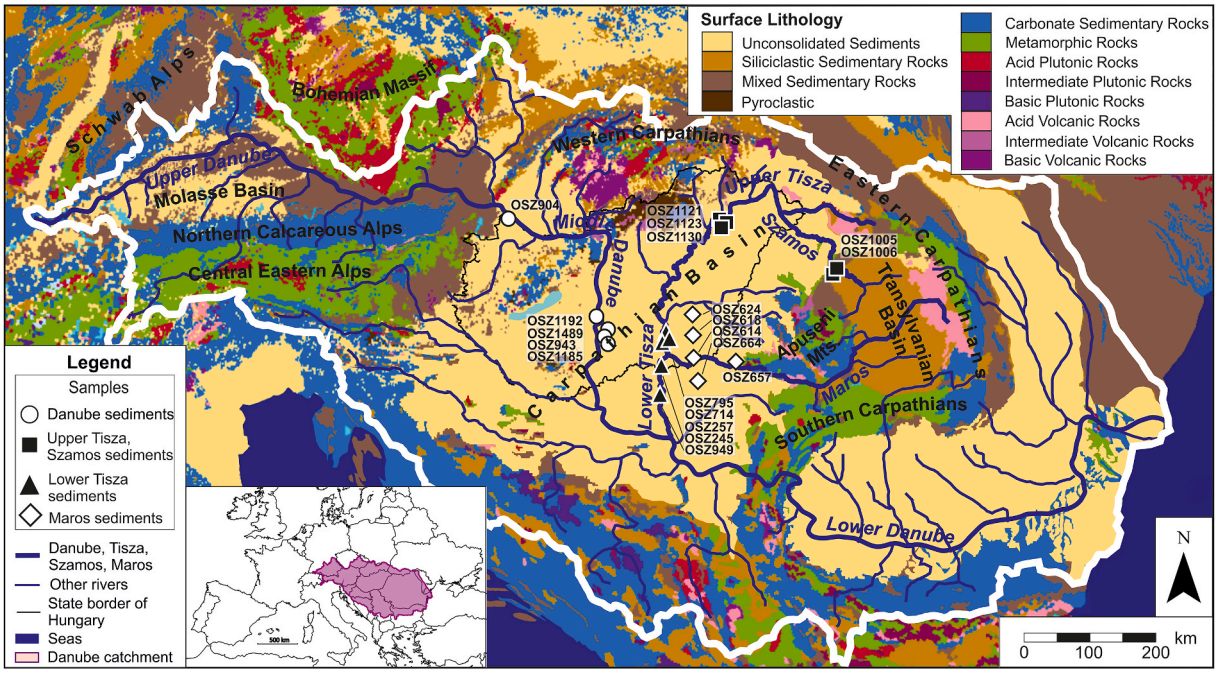

Fig. 1. The study area and the location of the analysed samples. Surface lithology is based on the map of Hartmann and Moosdorf (2012).

Table 1
Sample mean values of different quartz luminescence sensitivity parameters.

Region	Sample ID	Grain-size (μm)	OSL age (ka)	CW-OSL sensitivity mean ± SE (SD) (counts/mg/Gy)	TL 110 °C peak sensitivity mean ± SE (SD) (counts/mg/Gy)	LM-OSL fast component sensitivity mean ± SE (SD) (counts/mg/Gy)	Total LM-OSL sensitivity mean ± SE (SD) (counts/mg/Gy)	Fast component ratio mean ± SE (SD) (%)	CW-OSL sensitivity ratio (cycle 10./cycle 1.)
Danube	OSZ 904	90–150	modern sample	70.7 ± 8.0 (28)	897 ± 52 (180)	85.7 ± 14.7 (36)	3031 ± 173 (424)	2.83 ± 0.47 (1.14)	1.1 ± 0.04
	OSZ 943	90–150	6.61 ± 0.34	70.7 ± 14.4 (50)	932 ± 159 (551)	68.4 ± 11.8 (32)	2248 ± 100 (245)	3.04 ± 1.22 (3.00)	1.14 ± 0.02
	OSZ 1185	90–150	7.55 ± 0.28	53.0 ± 3.2 (11)	807 ± 128 (443)	71.8 ± 10.6 (26)	2326 ± 76 (187)	3.09 ± 0.45 (1.11)	1.75 ± 0.05
	OSZ 1192	90–150	6.84 ± 0.44	66.5 ± 6.8 (24)	890 ± 82 (284)	80.7 ± 24.7 (65)	2743 ± 357 (875)	3.90 ± 0.54 (1.2)	1.32 ± 0.02
	OSZ 1489	90–150	10.06 ± 0.43	61.5 ± 8.6 (30)	823 ± 58 (201)	64.0 ± 6.4 (12)	2178 ± 141 (344)	2.94 ± 0.38 (0.94)	1.06 ± 0.02
Upper Tisza	OSZ 1121	90–150	4.83 ± 0.25	193 ± 20 (69)	2118 ± 163 (564)	263 ± 43 (106)	5189 ± 685 (1677)	5.07 ± 1.5 (3.67)	1.04 ± 0.01
	OSZ 1123	90–150	4.23 ± 0.19	144 ± 16 (55)	1189 ± 117 (405)	208 ± 54 (132)	3039 ± 202 (494)	6.85 ± 1.16 (2.84)	1.06 ± 0.03
	OSZ 1130	90–150	4.03 ± 0.29	108 ± 9 (31)	1218 ± 179 (620)	139 ± 16 (38)	2648 ± 171 (418)	5.62 ± 0.36 (1.96)	1.08 ± 0.02
Szamos	OSZ 1005	90–150	12.45 ± 0.59	217 ± 41 (142)	1633 ± 254 (880)	232 ± 55 (191)	2939 ± 660 (1904)	7.04 ± 1.34 (3.27)	1.13 ± 0.03
	OSZ 1006	90–150	13.0 ± 1.22	170 ± 13 (45)	2471 ± 158 (547)	199 ± 14 (32)	3520 ± 126 (309)	5.43 ± 0.8 (0.89)	1.26 ± 0.04
Lower Tisza	OSZ 245	90–150	10.00 ± 1.4	186 ± 33 (114)	2597 ± 384 (1330)	198 ± 63 (155)	3304 ± 309 (757)	6.17 ± 1.97 (4.82)	1.21 ± 0.02
	OSZ 257	90–150	12.40 ± 1.4	117 ± 11 (38)	1728 ± 221 (766)	216 ± 35 (85)	3325 ± 114 (278)	6.48 ± 1.1 (2.7)	1.06 ± 0.01
	OSZ 714	90–150	4.35 ± 0.43	174 ± 19 (66)	2309 ± 147 (510)	182 ± 25 (61)	3158 ± 184 (450)	5.77 ± 0.7 (1.73)	1.15 ± 0.02
	OSZ 795	90–150	27.13 ± 1.54	205 ± 21 (73)	2364 ± 368 (1258)	250 ± 35 (87)	3528 ± 157 (384)	7.09 ± 1.03 (2.52)	1.16 ± 0.02
	OSZ 949	90–150	13.39 ± 0.70	203 ± 30 (104)	1783 ± 334 (1157)	201 ± 8 (19)	3374 ± 49 (121)	5.96 ± 0.3 (0.72)	1.08 ± 0.01
Maros	OSZ 614	90–150	15.50 ± 2.00	185 ± 16 (55)	1820 ± 195 (676)	167 ± 16 (40)	1446 ± 244 (598)	4.12 ± 0.49 (1.21)	0.98 ± 0.01
	OSZ 618	90–150	11.50 ± 1.90	158 ± 18 (61)	1587 ± 163 (565)	185 ± 31 (69)	3817 ± 194 (476)	4.84 ± 0.56 (1.24)	0.94 ± 0.02
	OSZ 624	90–150	14.3 ± 1.90	169 ± 19 (66)	2269 ± 428 (1484)	189 ± 21 (44)	4313 ± 153 (1341)	4.55 ± 0.76 (1.87)	1.14 ± 0.06
	OSZ 657	90–150	14.6 ± 1.17	181 ± 11 (38)	1991 ± 173 (599)	196 ± 17 (42)	4438 ± 153 (375)	4.46 ± 0.44 (1.09)	0.97 ± 0.02
	OSZ 664	90–150	7.1 ± 1.00	168 ± 9 (31)	2690 ± 34 (119)	177 ± 25 (61)	3402 ± 101 (248)	5.15 ± 0.59 (1.44)	1.29 ± 0.04

recovering pointbar sediments (Robu, 2018). The third group of samples were collected along the Lower Tisza River, where two main tributaries join the river. Samples were collected from pointbars located on the two levels of the floodplain and were previously dated to the Late Pleistocene and Holocene (Sipos et al., 2016). The fourth group is comprised of Late Pleistocene sediments collected from the mid channel bars of braided and the pointbars of meandering palaeo-channels on the Maros Alluvial Fan (Kiss et al., 2014), thus these samples can clearly be attributed to the Maros River.

3. Measurement procedures

When selecting the samples for the analyses, it was an important criterion that they should be of the same grain size, i.e. 90-150 μm. Consequently, the same sample preparation procedures were applied in case of each, mostly based on the steps given by Mauz et al. (2002). Samples were dried out and the desired grain size fraction was separated by sieving. In the next step the carbonate and organic material content was removed by treatment in 10% HCl and 10% H_2O_2. The separation of the quartz fraction was made using heavy liquid flotation (LST Fastfloat). Finally, a 45 min etching in 40% HF was performed to remove any remaining feldspar contaminations and the outer layer of quartz grains, which was followed by a HCl treatment to remove any remaining fluoride precipitations.

For the analyses approximately 6 mg of sample was used per aliquot, from each sample several aliquots were prepared by depositing the material on stainless steel cups placed and tared on an analytical balance with a ±0.1 mg accuracy. The weight of each aliquot was measured this way in order to perform mass normalization after the tests. The 6 mg sample equals to an approximately 200 pcs of 90–150 μm grains (same as the number of grains in case of a 2 mm mask), which can be spread easily on the cups without the undesired packing of grains. This was vital to ensure that each grain receives approximately the same treatment in the reader facility. On the other hand, lower amounts were not feasible to use either, since Mid-Holocene Danube samples dated previously by Tóth et al. (2017a) had such a low sensitivity when using 200 grains (2 mm mask) that natural OSL signals could hardly be distinguished from background in case of 60–70% of the aliquots. Though not on the Danube catchment, but still in an Alpine environment similar observations were made by Lowick et al. (2015), who found, based on their preliminary single grain measurements, that only <0.5% of grains had a signal significantly above the background, and used therefore 2 mm aliquots in the end for dating.

The luminescence response of aliquots to unit β-dose was investigated by applying continuous wave (CW) and linearly modulated (LM) OSL. During the necessary preheat of the aliquots the 110 °C TL peak was also recorded. Sensitivity measurements were performed using a RISØ TL-DA-20 luminescence reader, equipped with a $^{90}Sr/^{90}Y$ β-source

and an EMI ET9107-type photomultiplier. Optical stimulation was performed at 125 °C (heating rate: 5 °C/s) for 200 s by using 80 mW blue LEDs (470 ± 30 nm) set for 90% power release. Detection was made through a Hoya U-340 filter. Throughout the tests a uniform 210 °C preheat temperature along with a 160 °C cutheat were applied in case of each sample in order to increase the comparability of sensitivity measurements. The selection of these temperatures was based on combined preheat and dose recovery tests, yielding either 200 °C or 220 °C as the optimal preheat temperature to recover known doses (see e.g. Tóth et al., 2017b).

CW-OSL and 110 °C TL sensitivity parameters were determined following and slightly modified protocols of Zheng et al. (2009) and Nian et al. (2019). The applied measurement procedures, reader settings and steps of evaluation are summarised in Fig. S1. Prior to artificial dosing and sensitivity measurements the natural luminescence was bleached from the aliquots by applying two cycles of blue LED stimulation with a 3000 s relaxation time in between. Subsequent to bleaching, all aliquots were irradiated with a 24 Gy β-dose and the resulted luminescence response was recorded after applying a 210 °C preheat. In case of CW-OSL (100 s, 125 °C, 5 °C/s heating rate, 0.1 s/ch readout rate) the first 0.5 s of the decay curve was integrated to receive sensitivity values. Background was subtracted using the last 5 s of the decay curve. Results were then normalised by the masses measured during aliquot preparation. The normalised intensity determined at this first cycle of the measurements was termed as "sensitivity base value". Subsequently, the same procedure was repeated in nine further cycles of irradiation and readout to investigate the laboratory sensitivity change of samples. The result of the last cycle was compared to the sensitivity base value and a so called "sensitisation ratio" was calculated. TL intensities were also recorded in the meantime. As the 110 °C TL peak appeared sometimes at lower temperatures, the 80–120 °C temperature window was applied uniformly for signal integration. In case of three samples, representing different regions, 48 aliquots were measured in the way detailed above to assess how the distribution and scatter of sensitivity values are influenced by increasing aliquot numbers. The number of aliquots measured at the rest of the samples and used later for sample to sample comparison was determined on the basis of these tests.

During LM-OSL measurements the same bleaching, dosing and preheat parameters were used as in case of the CW-OSL measurements but on separate aliquots. The intensity of blue LEDs was increased from 0 to 90% in an interval of 1000 s. Intensity was recorded at every 0.1 s, background was determined by stimulating empty cups. The LM-OSL curves were resolved into components following Jain et al. (2003) and Singarayer and Bailey (2003) by applying the "Luminescence" package of the R software (Kreutzer et al., 2012). For further comparisons three parameters were calculated: 1) fast component intensity, obtained by integrating the first 45 s of the LM-OSL curve, 2) total LM-OSL intensity, i.e. the signal integrated from the entire LM-OSL curve and 3) fast component ratio, being the ratio of the two parameters above.

From the single aliquot values of sensitivity parameters, obtained by CW-, LM-OSL or TL, sample mean values were calculated, the uncertainty of which was given as the standard error (SE) and also as the standard deviation (SD) of single aliquot results. SD is indicated in parenthesis in the text and in the tables. Additionally, regional mean, SE and SD values were calculated from the aliquots of each sample representing a given area, in order to perform larger scale comparisons.

4. Results

4.1. Test measurements

During the first tests, 3 to 6 aliquots of each sample were subjected to sensitivity measurements, but then as the results showed a considerable scatter the number of aliquots was gradually increased to 48 in case of three samples (OSZ618, OSZ714 and OSZ1005) in order to determine the sufficient number of aliquots to obtain representative mean values for the samples (Fig. 2, Figs. S2 and S3). The results showed that the shape of the distribution curves may considerably change, i.e. in case of two samples negative skewness turned over positive when increasing aliquot number to 12 (Fig. 2 and Fig. S2). Sample mean values at different aliquot numbers stayed within the SE of the results. Variation was more significant at low aliquot numbers, and based on the three tests made, above 12 aliquots mean values remained stable with the exception of sample OSZ1005 (Fig. S3) as here some very sensitive aliquots appeared when increasing the number of measurements to 36 and 48. SD values either stayed similar throughout the tests, or in one case increased significantly at 12 aliquots (Fig. 2), thus the standard error of CW-OSL sensitivity values could not be significantly reduced by increasing the number of aliquots. Consequently, a maximum of 12 aliquots seemed to be an adequate compromise for further measurements and to perform meaningful comparisons between samples.

When increasing aliquot numbers it was also noticed, that the distribution of sensitivity values is not at all normal, as it could be expected on the basis of the number of grains on aliquots, but rather resembles the positively skewed distribution of equivalent doses characterising partially bleached samples (Fig. 2). Some of the aliquots yielded outstandingly high sensitivity values which might refer to the presence of 'supergrains' (Arnold et al., 2016) in some of the aliquots. However, if we disregard these aliquots, there is still a clear tendency of skewness, which might be in relation with the variable natural dosing of the grains, which in turn can have an effect on equivalent dose distributions as well. Consequently, the assessment of possible relationships needs a more systematic analysis in the future.

4.2. CW-OSL and 110 °C TL peak sensitivity

As outlying aliquots were not excluded from the calculation of sample means, CW-OSL sensitivity base values show 10–20% relative SE, corresponding to a 20–50% coefficient of variation (Table 1). If values from the same region are taken, then sediments related to the Danube and Maros River show the most even distribution (Fig. 3A), with a relative SE around 5%, whereas in case of the Tisza this value is around 10%. When regional mean values are considered, Danube sediments (64.4 ± 3.9 (30.0) counts/mg/Gy) can clearly be separated from the others, namely their CW-OSL sensitivity base value is approximately one third of Tisza and Maros values (Table 1 and Fig. 3A). The investigated Eastern Carpathian rivers can hardly be distinguished from each other concerning regional means at the present number of samples and measurements (Fig. 3A). Although the two sensitivity values related to River Szamos seem to be higher if their average is considered (193 ± 22 (106) counts/mg/Gy), but this difference is not significant and stays within the error of the Lower Tisza (176 ± 11 (88) counts/mg/Gy) and Maros (167 ± 9 (67) counts/mg/Gy) values. Accordingly, if Upper Tisza samples are taken separately (148 ± 11 (64) counts/mg/Gy) their sensitivity seems to be lower than any of the Carpathian values, but still significantly higher than that of Danube sediments (Fig. 3A).

The sensitivity values received on the basis of the 110 °C TL peak follow the pattern of CW-OSL results in general (Fig. 3B). Namely, the 110 °C TL peak sensitivity of the Danube is significantly lower in this case as well, i.e. roughly 40% of the values measured for sediments with a Carpathian background (Table 1), while samples related to River Tisza, Maros and Szamos can hardly be differentiated from each other (Table 2). If the distribution of sample means is considered within a region 110 °C TL peak values show a greater variation compared to CW-OSL values in terms of the Maros and Upper Tisza related sediments (Fig. 3B, Table 2).

4.3. LM-OSL sensitivity and fast component ratios

In general all 5 components (fast, medium, slow1-2-3) could be fitted to the measured LM-OSL curves. In case of sample (OSZ714) an ultrafast component could also be identified in four aliquots out of the twelve

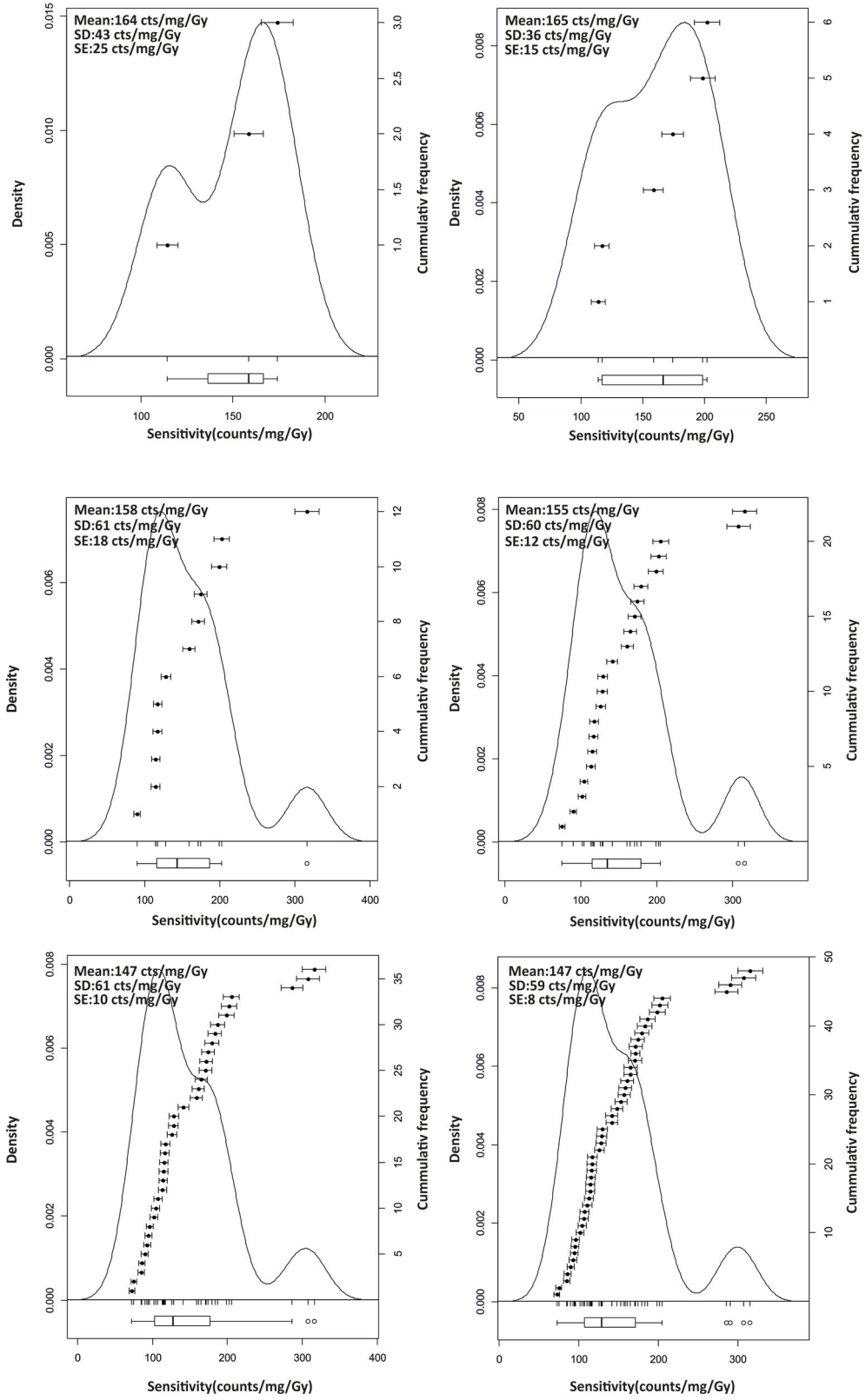

Fig. 2. Distribution of CW-OSL sensitivity results in case of sample OSZ618 (River Maros) by increasing the number of measured aliquots from 3 to 48.

measured, however, this component was not detected in any other samples. In relative terms LM-OSL fast component sensitivity results correspond slightly better to CW-OSL values than 110 °C TL peak values (Fig. 3C, Table 1), i.e. the relative SD of ratios calculated by dividing CW-OSL/110 °C TL peak sensitivity and CW-OSL/LM-OSL fast component sensitivity are 11.2% and 7.6%, respectively.

Another difference in comparison with CW-OSL values is that concerning the LM-OSL fast component sensitivity regional mean values of the Tisza are slightly higher than in case of the Maros (Table 1). However, due to the considerable uncertainty of sample mean values this

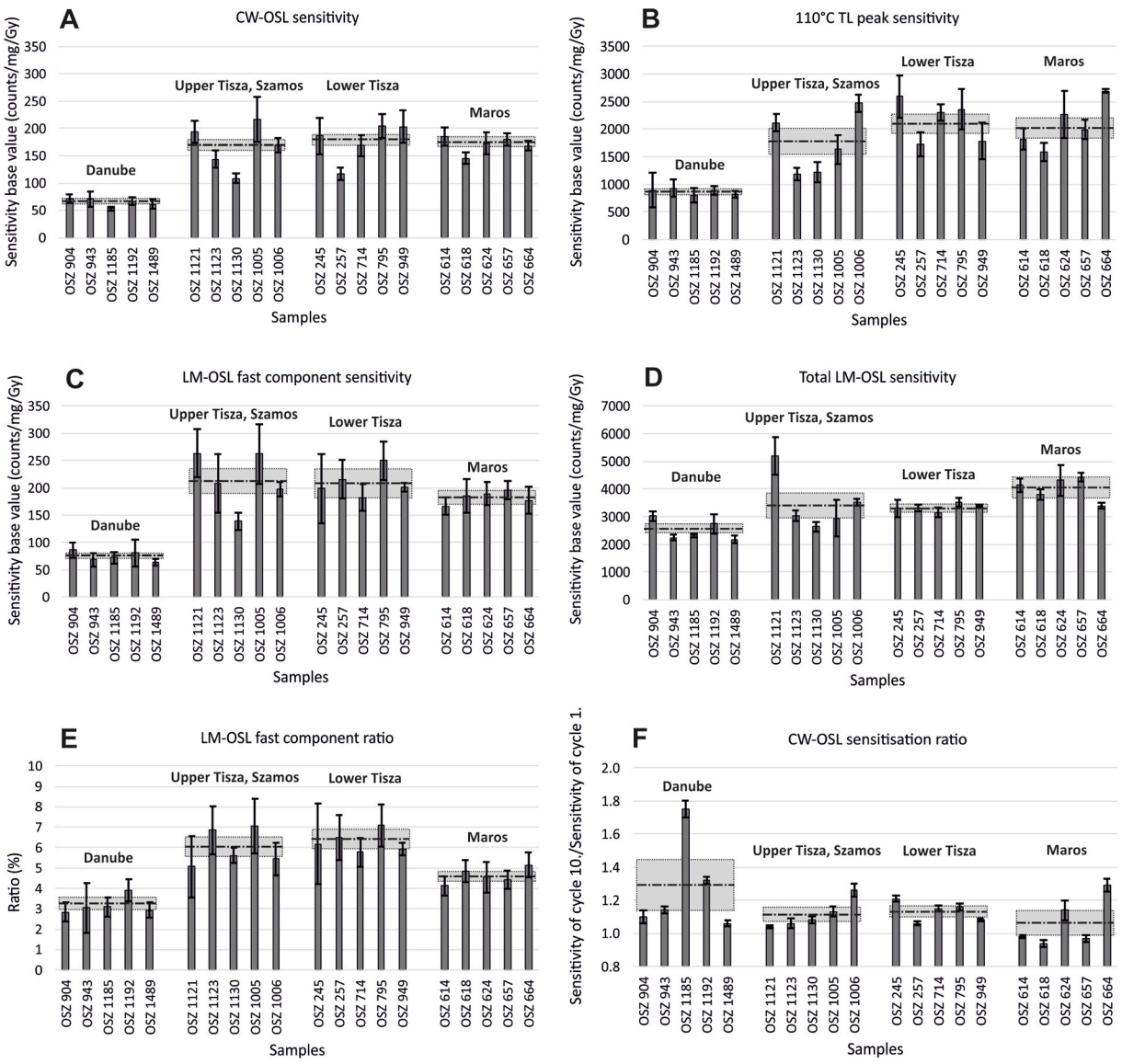

Fig. 3. Bar chart of mass normalised sensitivity base values determined with different techniques. Dashed lines indicate regional mean values, grey bands show the SE of the mean.

Table 2
Regional mean values of different quartz luminescence sensitivity parameters.

Region	CW-OSL sensitivity regional mean ± SE (SD) (counts/mg/Gy)	TL 110 °C peak sensitivity regional mean ± SE (SD) (counts/mg/Gy)	LM-OSL fast component sensitivity regional mean ± SE (SD) (counts/mg/Gy)	Total LM-OSL sensitivity regional mean ± SE (SD) (counts/mg/Gy)	Fast component ratio regional mean ± SE (SD) (%)
Danube	64.5 ± 3.9 (30)	869 ± 24 (161)	74.1 ± 4.0 (78)	2505 ± 164 (558)	3.16 ± 0.30 (1.60)
Upper Tisza, Szamos	166 ± 11 (85)	1726 ± 251 (583)	214 ± 23 (112)	3467 ± 453 (1434)	6.00 ± 0.48 (2.55)
Lower Tisza	176 ± 11 (88)	2156 ± 171 (570)	209 ± 26 (89)	3338 ± 60 (435)	6.30 ± 0.49 (2.68)
Maros	167 ± 9 (67)	2071 ± 191 (525)	183 ± 12 (103)	4023 ± 187 (768)	4.59 ± 0.25 (1.35)

difference is hardly detectable (Fig. 3C). Consequently, the differentiation of sediments based simply on this parameter is not feasible as indicated also by the large regional error terms received (Table 2).

However, it is common in CW-OSL and LM-OSL results that Danube related sediments are well distinguishable based on their very low sensitivity (Fig. 3A and C) but this difference decreases considerably if total LM-OSL intensities are compared (Fig. 3D). In this case the mean sensitivity value of Danube sediments reaches 60–75% of Tisza and Maros results. An obvious reason for this is that the proportion of the fast component is relatively low in Danube samples, whereas the contribution of medium and slow components to the total intensity is much higher than in the case of Carpathian origin quartz extracts.

Aliquot to aliquot variation of the fast component ratio showed similar values like in case of the sensitivity parameters examined above,

and with the exception of the Upper Tisza the relative SE of regional values also remained well below 10% (Table 1). The pattern of values is similar to that of other parameters (Fig. 3E), but an important difference is that the mean value of Maros related sediments (4.59 ± 0.25 (1.35) counts/mg/Gy)) this time lies slightly farther from that of the Upper and Lower Tisza regions (6.00 ± 0.48 (2.55) and 6.30 ± 0.49 (2.68) counts/mg/Gy), though, if the SD of regional values is taken then there is a considerable overlap, making the separation of samples still uncertain.

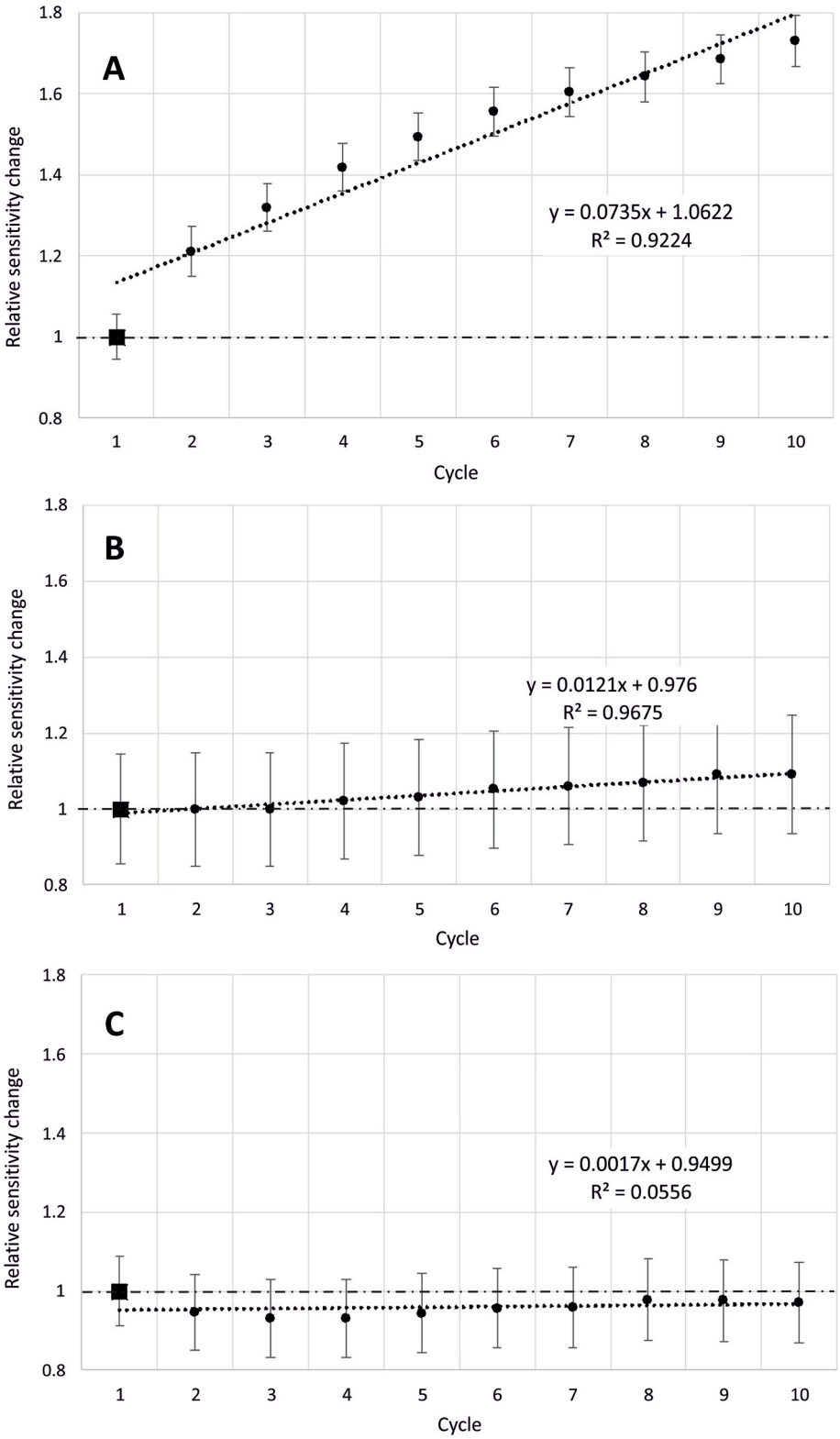

Fig. 4. Three typical sensitisation curves showing intensity change in response to the same dose repeated in 10 cycles. Sensitivity base value is marked with a square. A) Sample OSZ1185 (River Danube) with considerable sensitivity increase. B) Sample OSZ1130 (River Tisza) with slight sensitivity increase. C) Sample OSZ657 (River Maros) with no sensitivity change.

4.4. CW-OSL sensitivity change

Beside measuring sensitivity base values, samples were also sensitized in the laboratory to quantify their sensitivity change in response to repeated dosing and stimulation. Samples were compared using the CW-OSL sensitisation ratio. In general, three characteristic groups could be identified (Fig. 4). The first includes those samples that produced a continuous and considerable increase in sensitivity, i.e. their sensitisation ratio was above 1.20 (Figs. 4 and 3F.). In most of the cases sensitivity growth was linear, but in case of sample (OSZ1185), related to the Danube, a saturating exponential function could be fitted best to sensitivity values. This sample was also unique in the sense that its sensitisation ratio reached 1.75 ± 0.05, being well above the value of other samples (Fig. 3F). In total five samples were classified to this group, and actually all sampling regions were represented among these (Table 1).

The second group (eight samples) exhibited a moderate sensitivity increase with a sensitisation ratio between 1.05 and 1.20. Without any exception, sensitivity increase could be fitted with a linear function (Fig. 4). Again, all sampling areas were represented, however three out of the five Lower Tisza samples were falling into this group (Table 1, Fig. 3F). Finally, a very slight, or practically no sensitivity change could be detected in terms of the third group (seven samples), with sensitisation ratios between 0.95 and 1.05. This group was comprised again of samples representing each region, however three out of five Maros related samples, with the lowest ratios, could be attributed to this group (Table 1, Fig. 3F).

Based on the above, low and high sensitisation ratio samples can be identified at any of the studied regions, however regional mean values may show some difference (Fig. 3F). The highest mean value, 1.27 ± 0.13 is related to Danube sediments, but it is true that if the sample with extreme sensitivity change is disregarded, then the mean (1.16 ± 0.05) falls very close to that of the Lower Tisza samples (1.13 ± 0.03), thus differentiation between the quartz deposited by these rivers is not really possible using this parameter. In the meantime, the mean value attributed to the Maros River sediments was 1.06 ± 0.07, and if the seemingly outlying result of sample OSZ664 is excluded, then it falls to 1.00 ± 0.04, being well below of either Lower Tisza and Danube values (Fig. 3F). Some distinction can also be made between the Tisza and Szamos Rivers in terms of the Upper Tisza region, as far as the mean related to the later one is 1.20 ± 0.07, whereas the same value for Tisza is 1.06 ± 0.01.

5. Discussion

It is hard to compare sensitivity values obtained to the results of other studies, since the measurement and calculation of sensitivity parameters differ in most of the cases and the availability of fluvial data is also limited. However, if CW-OSL results of Haddadchi et al. (2015) or Nian et al. (2019) and TL 110 °C values reported by Zheng et al. (2009) are considered, each of them determined by using similar integration intervals, then we might claim that Carpathian Basin coarse grain quartz has a low (Danube) or moderate (Tisza, Maros) sensitivity in general. The dimness of Alpine quartz has also been demonstrated earlier by Klasen et al. (2006, 2007) and Trauerstein et al. (2017), though, the degree of sensitivity in their studies was not quantified.

Based on our data, it seems that low sensitivity is characteristic not only on the upstream catchment (Bickel et al., 2015; Lowick et al., 2015; Klasen et al., 2016), but far downstream as well, and provides a clear fingerprint for Danube related fluvial sediments in the Carpathian Basin. The spatial range of this observation cannot be thoroughly assessed on the basis of the present data, however, if the sensitivity value of sample OSZ904, being in the most upstream position along the Danube, is compared to more downstream samples, then actually no difference can be identified (Fig. 1 and Table 1). Besides, the mean sensitivity of Upper and Lower Tisza samples is also very similar, though the river collects several tributaries between the two regions investigated, and these can modify the sensitivity of the quartz mixture. After all, at the present spatial distribution of samples the observation of Pietsch et al. (2008), i.e. the downstream increase of quartz sensitivity, cannot be approved or disproved.

The generally low sensitivity of Danube quartz is also resembled by aeolian blown sands along the river (Novothny et al., 2010; Sipos et al., 2014), and since based on other proxies, a main source of Danubian loess is suggested to be fluvial silt (Újvári et al., 2010), the fingerprint of dim quartz might be also recorded in dust deposits. Although, several properties of fine grain quartz has been studied in detail (e.g. Timar--Gabor et al., 2015), no systematic analyses has been made in terms of its sensitivity so far.

Earlier research on different sensitivity parameters demonstrated a clear relationship between 110 °C TL and CW-OSL sensitivity (e.g. Li, 2002; Wintle and Murray, 2006). Based on the sensitivity values of Carpathian Basin fluvial sediments, a similar observation can be made. It is important to note, though, that the data at the higher end of the relationship show a considerable scatter, mostly because while 110 °C TL values can grow, CW-OSL sensitivity seems to have a limit at 200–250 ct/mg/Gy (Fig. 5A).

If optical type sensitivity values, such as LM fast component and CW-OSL sensitivities are plotted, which anyway should represent a closer relationship since the same traps are stimulated, then data show obviously a higher R^2, but still have a considerable scatter (Fig. 5B). The most probable reason for this is the variable contribution of fast and medium components to the first 0.5 s of the CW-OSL decay curve, which can also be an indication of the difference in sediment sources, just as found by Sawakuchi et al. (2018). Scatter may also be caused by the difference in the proportion of slow components, based on which the background was subtracted in during CW-OSL signal normalization. As shown by Fig. 5B optical sensitivity values not only in the case of the Danube, but also in terms of the Maros form one group, though, values significantly overlap with Tisza results, thus no clear distinction can be made between the two rivers. The geomorphic reason behind the much lower scatter of Danube and Maros values is probably the fact that on the measured sections and well upstream only minor (Danube) or even no (Maros) tributaries join these rivers, thus sediments from different sources are mixed better before deposition. On the other hand, in case of the Tisza optical parameters can be greatly affected by the adjoining major tributaries.

Based on the results, it was already suggested that LM-OSL sensitivity values may be the best indicators for distinguishing Carpathian Basin fluvial sediments. When fast component ratio and total LM-OSL sensitivity are plotted against each other (Fig. 5C), the quartzes of the main rivers can be separated better than in any other way. This means that beside the generally low sensitivity of Alpine Danube quartz, the relatively low proportion of the fast component makes Maros quartz also distinguishable from Tisza values.

The relationships found above, can help to clear up some geomorphic questions in the confluence zones of the basin's fluvial system. For example Popov (2012) attributed some hardly datable low sensitivity samples to the Tisza River in Serbia, possibly the sediments were rather deposited by the Danube. In the confluence zone of the Tisza and Maros Rivers a large meander was dated by Sipos et al. (2016), but the question remained which river had formed the channel. Robu (2018) reconstructed the development of the Tisza-Szamos Plain in Romania, where the complex arrangement of channel alignments and the variable flow direction of the Tisza did not allow to identify in all the case the channel forming river. Though in the present paper the Sava River was not investigated, Mitrinović et al. (2020) identified there highly sensitive quartz, offering a great potential to distinguish it from low sensitivity Danube sediments in the confluence zone near Belgrade.

Total LM-OSL sensitivity values were also plotted against the age of the samples (Fig. 6). In most of the cases this relationship is used to set up temporal differences in the provenance of sediments, see e.g. Lü and Sun (2011). In our case, the source of sediments hardly changed in the timespan of measured OSL ages, and only regional conclusions can be

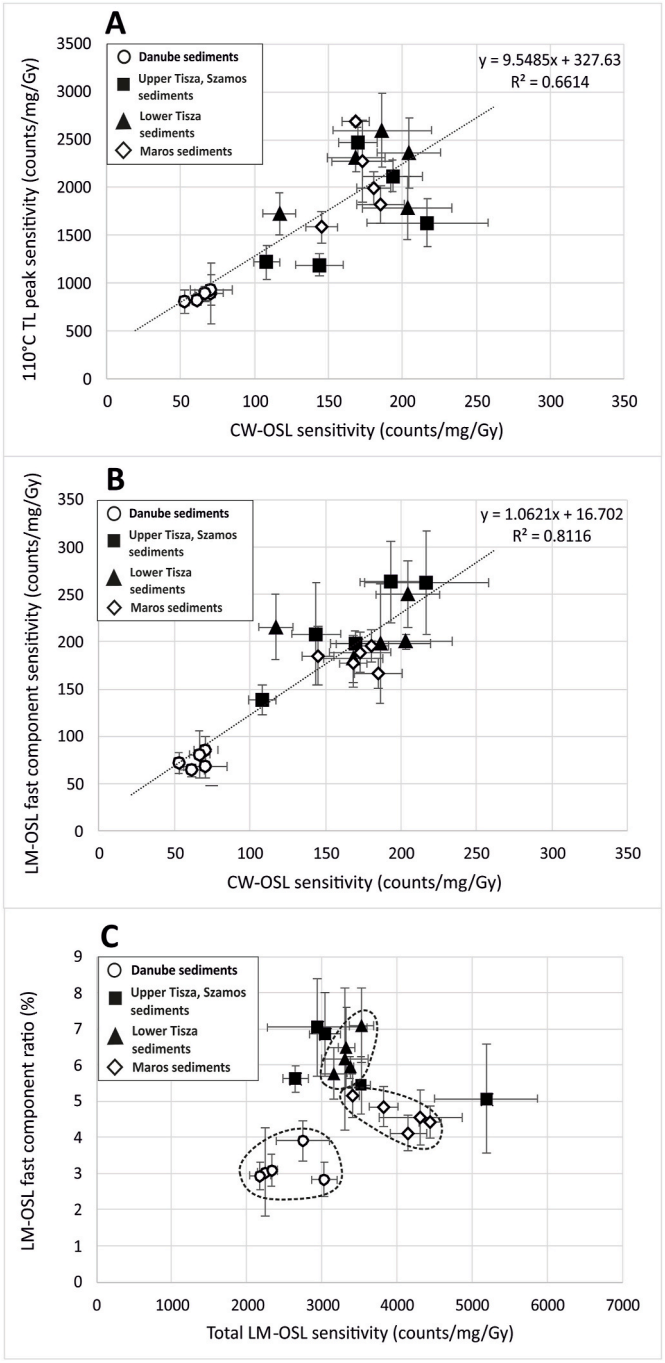

Fig. 5. The relationship of different sensitivity parameters.

drawn from the age plot due to the considerable differences in the sensitivity of Danube and Tisza related quartz extracts. This also means that a low number of data can be evaluated, still, some trends seem to appear. First of all, in case of the Danube total LM-OSL intensities show a slight but unambiguous increase as samples get younger (Fig. 6). With the exception of the modern sample the area from where these samples were taken was reworked by the Danube in the Holocene several times while a slight incision took place (Pécsi, 1967; Tóth et al., 2017a). Repeated reworking, therefore, may provide a geomorphological explanation to sensitivity growth as deposits get younger. On the other hand, in a depositional environment on the alluvial fan of the Maros River, where no significant reworking can be suspected due to returning avulsions at the apex of the fan (Kiss et al., 2014) an opposite trend can be observed (Fig. 6). The deposition age of Maros samples spans from the

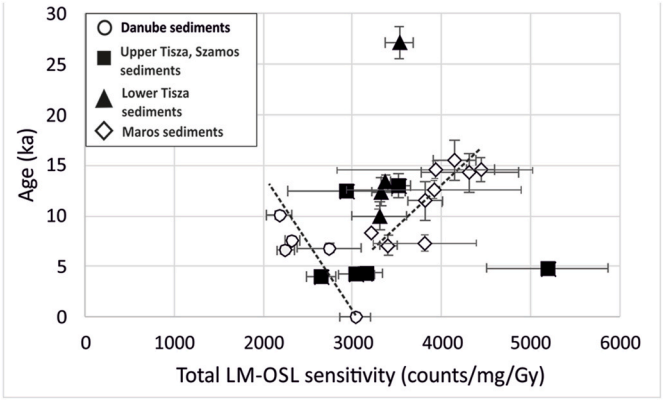

Fig. 6. OSL age versus total LM-OSL sensitivity plot.

Late Glacial to the Early Holocene. The discharge and the sediment transport capacity of the river increased in this time interval, meaning that sediment grains could reach their deposition site through less sedimentary cycles. Besides, by the deglaciation of the Southern Carpathian subcatchments of metamorphic lithology could provide more dim quartz grains to the sediments. Definitely, a larger set of data is needed to reinforce or disregard geomorphic and petrographic drivers behind temporal changes in quartz sensitivity, but trends in these cases seem to be evident.

Finally, it is quite hard to make any general conclusions if the results are matched to the surface lithology of the Danube Basin, as the analysed fluvial samples in the basin necessarily are a mixture of quartz grains with a variable origin (Fig. 1). However, if the Alpine and Carpathian part of the catchment is compared it is obvious, that the upland section of the Tisza and its tributaries gain a considerable amount of their sediment from Late Cretaceous–Palaeogene flysch and other, mostly Neogene siliciclastic sedimentary rocks (Fig. 1). Whereas in case of the Danube, the contribution of these to the sediment mixture is less important. This would support the observations of Fitzsimmons (2011), who emphasized the potential significance of "inherited" luminescence sensitivity in relation with sandstone sediment sources. On the other hand, the proportion of volcanic rocks is also greater on the surface of the Tisza catchment which may affect the luminescence sensitivity of quartz just in the opposite direction (Chithambo et al., 2007). Consequently, clear relationships can only be drawn after systematic sampling on subcatchments with a different lithology. This way, the Carpathian Basin could serve as an ideal place for understanding the interplay of intrinsic and extrinsic parameters of quartz luminescence sensitivity better.

6. Conclusions

In spite of the limitations of the present study, such as the averaging effect of multi-grain measurements and the complex lithological background of the sediments investigated, the detailed sensitivity analyses of fluvial samples from some major catchments of the Carpathian Basin have shown for the first time that the sensitivity of Alpine and Carpathian origin quartz extracts is remarkably different even in a lowland environment, several hundred km downstream of the upland catchments where the sediment is produced. Consequently, in the case of Danube related quartz each type of sensitivity parameter showed an outstandingly low value. Beside this major conclusion, several other observations were made.

Even though multi-grain measurements were conducted, the distribution of single aliquot CW-OSL sensitivity data show in most of the cases a positive skew, similar to the dose distribution of fluvial sediments. The relationship between natural predosing and sensitivity needs to be further elaborated by the systematic recording of normalised

sensitivity during the dating of fluvial sediments. The fact, however, that some samples showed almost no sensitivity increase in response to repeated artificial dosing may bring further complication to the assessment of the dose/sensitivity relationship.

When considering different types of sensitivity parameters, total LM-OSL intensities and fast component ratios showed the least variation within one group of samples, therefore it is claimed that in the Carpathian Basin these parameters can be used best for sediment provenancing. This was also supported by the fact that using a total LM-OSL vs. fast component ratio plot the samples of the investigated regions could be separated relatively well. Results therefore point on the fact that beside CW-OSL sensitivity the proportion of components within the OSL signal can provide a further tool for distinguishing sediments of different lithological background.

Based on the results, geomorphological drivers might also affect the sensitivity parameters of coarse grain quartz samples, e.g. by the better mixing of grains the distance of sediment transport can have a positive effect on the scatter of sample mean sensitivity values. More systematic analysis needs, however, to determine whether age vs. CW-OSL relationships have a geomorphic meaning or not. At the present number of samples this cannot be unambiguously stated.

Nevertheless, the findings presented above can help to make distinctions between the origin of sediments in the complex fluvial landscape of Carpathian Basin lowlands, especially at the confluence zones of major rivers. Moreover, by taking into consideration that extensive aeolian deposits in the region, which have been blown out from fluvial sediments, the present relationships may help to reconstruction their sources as well.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.quageo.2021.101166.

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