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Fluvial sediment pathways enlightened by OSL bleaching of river sediments and deltaic deposits

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Abstract. While a number of studies have investigated bleaching of the optical signals of sediments in rivers and deltas, unified trends and mechanisms for bleaching in these settings remain unresolved. Here, we explore the bleaching of the optically stimulated luminescence (OSL) signal of quartz sediments in a large fluviodeltaic system across time and space, by comparing residual doses of sand and silt from the modern Mississippi River channel with estimated residual doses of sand isolated from Late Holocene Mississippi Delta mouth bar and overbank deposits. Further insight is obtained from a comparison of burial ages of paired quartz sand and silt of Mississippi Delta overbank deposits. Contrasting some previous investigations, we find that the bleaching of the OSL signal is at least as likely for finer sediment as for coarser sediment of the meandering Mississippi River and its delta. In addition we find an unexpected spatiotemporal pattern in OSL bleaching of mouth bar sand deposits. We suggest this may be caused by changes in upstream pathways of the meandering channel belt(s) within the alluvial valley, or by distributary channel and coastal dynamics within the delta. Our study demonstrates that the degree of OSL bleaching of sand in a large delta can be highly time- and/or space-dependent. Silt is shown to be generally sufficiently bleached in both the modern Mississippi River and associated paleo-deposits regardless of age, and may provide a viable option for obtaining OSL chronologies in megadeltas. In addition to informing dating approaches, our work contributes to initiatives to use luminescence signals to fingerprint sediment pathways within river channel networks and their deltas.

1 Introduction

Optical dating (Huntley et al., 1985) and its subsequent methodological advances (e.g., Cunningham and Wallinga, 2010;Galbraith et al., 1999;Murray and Wintle, 2000, 2003;Cunningham and Wallinga, 2012) have enabled new chronologies of fluvial and deltaic systems, obtained from direct measurements of the burial time of clastic sediment. One primary requirement of optical dating is the complete resetting ("bleaching") of the luminescence signal by sunlight exposure of at least some of the grains within a sample upon burial; in other words, zeroing of the optical clock shortly before or at the time of the event of interest (e.g., Wallinga, 2002). In the absence of complete resetting, sediment grains retain a residual dose acquired during previous burial, which may lead to overestimation of the time of the most recent burial event. Assessing bleaching is especially important for dating of fluvial sediment deposited within the most recent millennium, because fluvial sediments are prone to incomplete bleaching and even small residual doses can produce highly inaccurate ages on young deposits (Wallinga, 2002). Populations of sediment grains (e.g., sediment samples) may be well/completely-bleached and contain only zeroed grains, or may be incompletely-bleached and contain at least some grains with residual doses (Duller, 2008). Here, we further classify incompletely-bleached sediment populations as heterogeneously- (containing both zeroed grains and grains with residual doses), or poorly-bleached (containing few to no zeroed grains).

A number of studies have investigated the degree of bleaching of river deposits and sediment entrained within modern river channels, and its relationship to grain size and geography. These have returned wide and varied results. Some studies have found that coarse sand is better bleached than fine sand (e.g., Olley et al., 1998, Murrumbidgee River, Australia;Truelsen and Wallinga, 2003, Rhine Meuse Delta, The Netherlands). Well-bleached silt has been identified in suspension in the Yangtze River (Sugisaki et al., 2015) and its Holocene deposits (Nian et al., 2018;Gao et al., 2018) as well as in recent (decades- to centuries-old) deposits of the Ganges-Brahmaputra Delta (Chamberlain et al., 2017). Yet, incomplete bleaching of silt has been shown for other fluvial systems such as fluvial terrace deposits of northwest China (Thompson et al., 2018) and the Elbe River, Germany (Fuchs et al., 2005).

The mechanisms that dictate the degree of bleaching of fluviodeltaic sediments are also not known to be absolute nor universal. For example, bleaching of sand may increase on average with transport distance due to sunlight exposure during temporary storage on bar surfaces (Stokes et al., 2001) or may decrease downstream due to the addition of poorly-bleached grains by tributaries or local bank erosion (McGuire and Rhodes, 2015). Sediment entrained within a river channel may be less well-bleached if temporary storage on the river banks is limited, because turbid water reduces the intensity of light exposure and restricts the light spectrum (Berger et al., 1990). Yet, in-channel transport offers an opportunity for sunlight exposure of sediment transported near the water surface, especially finer grains which are more evenly distributed in the water column (Fuller et al., 1998) or those in turbulent systems (Gemmell, 1988). Bleaching of entrained sediments may also occur during subaerial exposure of river bar surfaces under conditions of low water discharge (Cunningham et al., 2015;Gray and Mahan, 2015). Furthermore, sediment grains in-transit may experience different bleaching than those preserved in the stratigraphic record (Jain et al., 2004) because in-transit grains have not yet reached their final destination and therefore have not necessarily undergone the full range of bleaching opportunities.

Here, we aim to increase understanding of the degree and mechanisms of optical signal bleaching in fluviodeltaic sediments, through an investigation of residual doses of quartz sediments of the contemporary Mississippi River and associated Late Holocene deltaic deposits (Fig. 1). Using previously established methods (Chamberlain et al., in press) to analyze archival OSL data, we compare the residual quartz OSL doses of sand and silt sampled within the modern Mississippi River channel (Muñoz et al., 2018;Chamberlain et al., in press;Chamberlain, 2017) to those estimated from sand of multi-century to millennium-aged Mississippi Delta mouth bar (Chamberlain et al., 2018) and overbank (Shen et al., 2015) deposits. Further insight into OSL bleaching is obtained by reanalyzing the burial ages of paired quartz sand and silt of Mississippi Delta overbank deposits (Shen et al., 2015). All combined, these data allow us to investigate trends in OSL bleaching across time, space, grain size, and depositional environment within a large fluviodeltaic system. In addition to informing dating approaches, this work adds to ongoing initiatives to use luminescence signals to reconstruct the pathways and histories of sediment grains (e.g., Reimann et al., 2015;Reimann et al., 2017;Sawakuchi et al., 2018;Gray et al., 2017). Improved knowledge of waterborne sediment pathways is a key component to delta restoration initiatives, such as river diversions aiming to mitigate land loss in deltas through delivery of sediment to the delta plain (e.g., CPRA, 2017). These engineered outlets will siphon sediment from yet-to-be-determined depths and positions within the river channel, and so the availability and grain size of the utilized sediment will depend on its transport mode within the river (e.g., Esposito et al., 2017) as well as the location and geometry of the engineered feeder channel (e.g., Gaweesh and Meselhe, 2016).

2 Geologic setting

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2.1 Mississippi River hydrology

The Mississippi River is among the largest rivers in the world in terms of catchment size, sediment, and water discharge. Its catchment includes about 3.3×10^6 km^2 (Milliman and Syvitski, 1992) and drains about 41% of the continental United States (Fig. 1) (Milliman and Meade, 1983). Therefore, sediment grains arriving in the Mississippi Delta may originate as far as 2,400 linear kilometers upstream and have experienced lengthy and convoluted transport (with bleaching opportunities, e.g., Stokes et al., 2001), or as near as a few meters or less from nearby river cutbanks and have experienced minimal transport (and minimal opportunities for bleaching, e.g., McGuire and Rhodes, 2015) since their last major storage event.

The hydrograph of the Mississippi River is generally highest in the spring due to snowmelt and increased precipitation in the catchment, and has multiple spring peaks with an average discharge of 25,000 m^3/s or more (Supplementary File, Fig. S1) (Galler and Allison, 2008). The first springtime "freshet" serves to mobilize and flush sediment from the lower reaches of the Mississippi River channel that has accumulated during preceding autumn-time low flow (less than 8500 m^3/s) conditions (Galler and Allison, 2008). Historical discharge records (1964-2012) for the US Army Corps of Engineers gauge at Tarbert Landing (river km 492 above Head of Passes, Fig.1) show that cumulative annual discharge is highly variable between years, and can range from around 3×10^{11} m^3/yr to greater than 6×10^{11} m^3/yr (Allison et al., 2014). Mud (that is, silt and clay) is the primary material transported in suspension during low flow conditions in the lower reach of the river, and is generally evenly distributed throughout the water column at all discharges (Ramirez and Allison, 2013). The mass of suspended sand in the lower reach, thought to be mobilized from lateral bars on the river bed, is minimal during low flow events and becomes similar to that of fines during the highest flow events (Allison et al., 2014). This indicates that there is a seasonal opportunity for light exposure of sands, and a year-round opportunity for light exposure of silts during transport within the river channel.

In addition to being a major river with significant variance driven by natural sources, the Mississippi is presently one of the most highly engineered river systems in the world (Kesel, 2003;Allison et al., 2012). Flow within the contemporary Lower Mississippi River is generally contained by human-made levees, which limit the degree of interaction of the channel with its floodplain and decrease the cannibalization of banks by restricting river migration (Kesel, 2003). Throughout the Holocene and prior to modification, the Mississippi River meandered freely within a series of six channel belts of unknown absolute ages (Saucier, 1994). The construction of dams as well as flood and navigation control structures in the catchment has reduced the suspended sediment load reaching the delta by reported values of 50-70 % (Blum and Roberts, 2009;Kesel, 2003), although the effects of these structures on sand transport to and within the deltaic reach has been debated (Nittrouer and Viparelli, 2014;Blum and Roberts, 2014). Similar changes in hydrology and sediment transport due to engineering have been documented in other fluviodeltaic systems (e.g., Hobo, 2015;Erkens, 2009). An investigation into bleaching that considers the residual doses of sediments of both pre-anthropogenic and present-day conditions is therefore useful because the hydrology and related luminescence bleaching opportunities of grains in the Mississippi River and other major channels worldwide may have been quite different prior to human modification of rivers.

2.2 Mississippi Delta stratigraphy and OSL properties

The Holocene Mississippi Delta first emerged around 7 ka, as sediment delivery to the basin outpaced regional sea-level rise (Törnqvist et al., 2004), and is composed of a series of amalgamated sediment lobes (subdeltas) fed by discrete distributary networks (Fisk, 1944). This study mainly investigates deposits of the presently 10,000 km^2 Lafourche subdelta (Fig. 1), active from ~1.6 to ~0.6 ka (Törnqvist et al., 1996;Shen et al., 2015;Chamberlain et al., 2018), which has been extensively OSL dated for geologic research and therefore provides a valuable archive of data to explore bleaching of the quartz OSL signal. During its millennium of activity, the Lafourche distributary network constructed 6,000-8,000 km^2 of new land through progradation into a shallow bay (Chamberlain et al., 2018), while the upstream portion of the system aggraded via deposition primarily from episodically-active crevasse channel networks (Shen et al., 2015). These crevasse channels operated as shallow off-takes, siphoning suspended material from the axial distributary channels (Esposito et al., 2017). Relatively coarse and homogenous mouth bar sand deposits characterize the prograded portion of the subdelta (Chamberlain et al., 2018). A patchwork of generally finer-grain crevasse splay and natural levee deposits, which may overlie progradational facies or peat, characterize the near-channel overbank depositional environment (Törnqvist et al., 2008;Shen et al., 2015;Esposito et al., 2017;Mehta and Chamberlain, 2018). Discharge was shared between the Lafourche and Modern (Balize) subdeltas beginning with Modern subdelta initiation at ~1.4 - 1.0 ka and continuing until Lafourche subdelta abandonment at ~0.6 ka (Hijma et al., 2017), although the exact timing and nature of the discharge split is unknown.

Previous research applying OSL dating to Lafourche subdelta deposits mainly relied on the measurement of small-diameter aliquots of quartz sand in combination with the application of minimum age models (Galbraith et al., 1999;Cunningham and Wallinga, 2012) to extract paleodoses, because equivalent dose (D_e) distributions suggested that at least some of the fluvial deposits in this setting were not completely bleached (Shen et al., 2015;Chamberlain et al., 2018). These approaches were found to yield internally consistent OSL ages which agreed with radiocarbon constraints obtained from prior dating of underlying peat (Törnqvist et al., 1996), whereas the application of the Central Age Model (CAM) (Galbraith et al., 1999) was found to overestimate the OSL age of some samples (Chamberlain et al., in press). Shen and Mauz (2012) found that the subtraction of an early background interval (Cunningham and Wallinga, 2010) produced more accurate and younger OSL ages for contemporary deposits associated with the nascent Wax Lake Delta of the Mississippi Delta (Fig. 1), also suggesting incomplete OSL bleaching of Mississippi Delta grains. Results of their study were validated with independent chronology from historical records. Shen et al. (2015) employed late background subtraction for dating overbank sands and silts, and showed that late-background-subtracted luminescence ages for paired silt and sand fractions extracted from the same overbank samples agreed within 2σ, indicating that silt may be a viable option for OSL dating of Late Holocene Mississippi Delta deposits (e.g., Muñoz et al., 2018).

3 Methods

3.1 Compilation of archival data

This study uses quartz OSL data compiled from previous investigations of contemporary (Muñoz et al., 2018;Chamberlain et al., in press;Chamberlain, 2017) and prehistoric (Chamberlain et al., 2018;Shen et al., 2015) sediments of the Mississippi River and associated deltaic deposits (Fig. 1).

Modern Mississippi River bedload (Chamberlain et al., in press) and suspended load (Chamberlain, 2017;Muñoz et al., 2018) sediments were sampled at Bonnet Carre Upstream 2 (BCU2), a site 221 river kilometers above the Mississippi River mouth at Head of Passes (Fig. 1). This site corresponds to the AboveBC2 site in Allison et al. (2013). Sampling took place in the Mississippi River channel center during high-flow conditions of 18,320 m^3/s in May, 2014, when the channel depth at BCU2 was 21.9 m. Suspended sediment samples (n=5) were captured in 5 L Niskin bottles at 0% (0 m), 25% (5.5 m), 50% (11.0 m), 75% (16.4 m), and 90% (19.7 m) water depths and a bedload sediment sample (n=1) was captured with a grab sampler. All samples were covered during and following retrieval to prevent light-exposure. OSL results obtained from the suspended samples were previously fully documented in a doctoral dissertation (Chamberlain, 2017), and data from the fine (4-20 µm) silt fraction were published by Muñoz et al. (2018) without details regarding the sampling location and approach, and analytical aspects. Here, we present the full details of these five samples, including OSL data for coarse (45-75 µm) suspended silt (n=2).

To investigate bleaching of older sediments, we revisited samples of Late Holocene Mississippi Delta sediments previously collected and measured by Chamberlain et al. (2018) (mouth bar deposits, n=17) and Shen et al. (2015) (overbank deposits, n=23). The details of these samples are available in their primary publications.

3.2 OSL measurements, age, and residual dose calculations

Samples were prepared following standard procedures which are described in the primary publications and were generally consistent across datasets. Measurements of small-diameter (1-2 mm) sand aliquots and ~2 mg/disk silt aliquots were conducted using standard single-aliquot regenerative dose protocols (Murray and Wintle, 2000, 2003), also described in the primary publications. Relatively low preheat temperatures (200 - 220 ^{o}C) were adopted to avoid thermal transfer. To ensure consistency across the archival luminescence data repurposed here, the original output luminescence data (bin files) were reanalyzed using standardized approaches (Chamberlain et al., in press), which most-importantly included the subtraction of an early background interval from the integrated initial OSL signal to enhance the relative contribution of the most readily-bleached quartz fast component to the net OSL signal (Cunningham and Wallinga, 2010). Age modeling for the sand samples was revisited by Chamberlain et al. (in press) using the bootstrapped (Cunningham and Wallinga, 2012) Minimum Age Model (Galbraith et al., 1999) (bootMAM) and incorporating a novel approach to assign the input and uncertainty on the overdispersion (σ_b) parameter (Chamberlain et al., in press). Identical OSL signal determination approaches were applied to the modern river bedload and suspended load samples, and age modeling of these samples was conducted with the unlogged version (Arnold et al., 2009) of the bootMAM (bootMAMul) (Cunningham and Wallinga, 2012) for sand and a mean ± standard error for silt.

Estimating residual doses of in-transit modern sediments is fairly straightforward, because these should yield a zero D_e when completely bleached. We judged the bleaching of sand grains isolated from paleo-deposits using a novel technique that is described and vetted in Chamberlain et al. (in press). This estimated a residual dose for each sample as the difference in D_es obtained with the CAM ($D_{e,CAM}$) and the bootMAM ($D_{e,bootMAM}$) (Fig. 2). Here we consider the implications of these residual doses with regard to grain-size, depositional environment, and spatial and temporal distributions of the samples. Please see Chamberlain et al. (in press) for details of statistical approaches supporting the present study.

3.3 Dose rate and residual age estimation

Remnant doses preserved in grains upon burial have little direct relationship with the dose rate of the matrix from which the grains are ultimately isolated for luminescence dating, although the dose rate of this matrix is often used to determine residual age. The bulk sediment characteristics and geological context (e.g., radionuclide activity concentrations, cosmogenic exposure, water content) under which the residual doses were acquired are generally unknown. For this reason, we prefer to use residual dose rather than residual age to describe the bleaching of sediments. Approximations of residual age are also discussed, as it relates to potential inaccuracies in burial age estimates if poor bleaching is not adequately dealt with. These are informed by average dose rates of 2.43 ± 0.06 Gy/ka for sand, and 2.96 ± 0.05 Gy/ka for silt sampled within the Lafourche subdelta (Shen et al., 2015;Chamberlain et al., 2018).

Ages calculated for the comparison of sand and silt fractions isolated from the same sample used dose rates particular to those samples, presented in Shen et al. (2015). All dose rates from Shen et al. (2015) were updated here to use the radionuclide conversion factors of Guérin et al. (2011) (Supplementary File, Table S.1). Other dose rate details can be found in the original publications.

4 Results and interpretation

4.1 Residual doses of modern river sediments

Residual doses of all samples are provided in the Supplementary File, Table S1. Modern river sediments show a trend of increasing residual dose with both grain size and channel depth (Fig. 3). We found that residual doses of modern river silt, moving in suspension within the channel, are very low regardless of water depth (note the logarithmic scale of Fig. 3). These ranged from 0.027 ± 0.001 to 0.135 ± 0.013 Gy for the 4-20 µm grains, with a mean value of 0.078 ± 0.044 Gy, similar to the "bulk" D_e values reported by Muñoz et al. (2018). Our reported mean residual dose, obtained through subtraction of an early background interval plus other methods described in Chamberlain et al. (in press), corresponds to an estimated residual age of 12-41 years (1-sigma range).

Of the two coarse silt (45-75 µm) suspended load samples, only the deeper (19.7 m) sample (BCU2 I-5) produced a measurable quartz luminescence signal, while the coarse silt fraction of the shallower sample (BCU2 I-3, at 11.0 m depth) was not sufficiently luminescent and will not be discussed further. The residual dose of BCU2 I-5 was 0.227 ± 0.149 Gy (26-127 years), suggesting that bleaching of coarser silt transported deeper in the water column may be less complete than bleaching of finer silt moving in more shallow suspension (Fig. 3). However, this tentative suggestion will need further confirmation, as comparison is based on a single sample and the results on the fine and coarse silt fraction for this sample agree within errors.

Residual doses of modern river fine silt appeared to be slightly greater with depth in the channel (Fig. 3). This may suggest some stratification of the water column. Alternatively, and more likely, the apparent trend may reflect different grain-size distributions within the analyzed fraction. Although the same fraction (4-20 µm) was prepared for each of the suspended samples, the deeper samples are more likely than the shallow samples to contain relatively large silt grains within this range, because coarser grains are more likely to be transported near the river bed than finer grains. The lower samples are therefore more likely to contain material that experienced less light exposure. This interpretation is consistent with the observation that the 45-75 µm suspended sample (BCU2 I-5) appeared less completely bleached than all the finer, 4-20 µm suspended silt samples (BCU2 I-1,2,3,4,5) (Supplementary file, Table S1).

By contrast, the two grain size fractions of modern river bedload sand (BCU2 I-6) appeared to be heterogeneously-bleached. The residual dose of the 125-180 µm fraction of BCU2 I-6 was 1.617 ± 0.288 Gy. This corresponds to a 0.55-0.79 ka estimated residual age. A bootMAMul D_e of 0.027 ± 0.051 Gy indicated that this grain size fraction contained some well-bleached quartz grains capable of producing an accurate luminescence age. The residual dose of the 180-250 µm fraction of BCU2 I-6 was 10.507 ± 1.673 Gy, corresponding to a 3.63-5.02 ka estimated residual age. A bootMAMul D_e of 0.791 ± 0.534 Gy indicated that this grain size fraction contained very few, if any, well-bleached quartz grains. Nevertheless, the estimate agrees with expected zero dose at 2-sigma level, demonstrating the ability of the bootMAM model to provide accurate (yet imprecise) results for highly heterogeneously-bleached samples. We note that some aliquots of the coarse sand fraction provided D_es of more than 25 Gy (> 10 ka residual age), indicating that some coarser sand grains transported by the modern Mississippi River were eroded from Pleistocene deposits with limited light exposure during transport.

4.2 Residual doses of late Holocene deposits

Following the procedures outlined by Chamberlain et al. (in press), bleaching of each Late Holocene-aged sample was classified by its minimum residual dose, defined as the residual dose minus 1σ uncertainty. Sand isolated from mouth bar and overbank deposits ranged from well- to heterogeneously-bleached for both depositional environments (Fig. 4). This was indicated by residual doses, calculated as $D_{e,CAM}$ - $D_{e,bootMAM}$, ranging from zero to 2 Gy (and a single estimate of greater than 3 Gy). These values correspond to residual ages estimated to be in the range of 0 - 1 ka. Mouth bar deposits had a smaller proportion of well-bleached sand samples (29%), while overbank deposits contained a greater proportion of well-bleached sand samples (48%). Bleaching was more complete for samples with paleodoses less than about 2.3 Gy (Fig. 4). Above 2.7 Gy, mouth bar sand was found to be heterogeneously-bleached with considerable (>0.5 Gy) residual doses, while overbank sand of similar D_es ranged from well to heterogeneously bleached (Fig. 4).

4.3 Bleaching by grain size

Among all samples, we observed a trend of increasing residual dose with increasing median grain size (Fig. 5), suggesting that coarser sand may be the least likely grain size to be completely bleached in this system. Still, each sand grain size fraction also contains some well-bleached samples, indicating that sand grains of all investigated sizes could be bleached prior to preservation. As discussed above, the 180-250 µm fraction of the river bedload sample (BCU2 I-6) yielded an exceptionally high residual dose of more than 10 Gy. While results for this grain size fraction fit the observed trend of bleaching degree with grain size, they are informed by only one sample of sediment that was still in-transit in the river channel when captured and may not be representative of bleaching of these coarser grains, both moving in the channel and preserved in the stratigraphic record (e.g., Jain et al., 2004). For these reasons, we caution against over-interpreting the results of BCU2 I-6, and the coarser fraction of the modern river bedload sample is omitted from Fig. 5.

4.4 Temporal trends in bleaching

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Surprisingly, the bleaching of mouth bar sand showed a strong temporal trend (Fig. 6A). All mouth bar sand samples (n=7) dated to be older than ~ 1.2 - 1.1 ka possessed relatively high residual doses, ranging from about 1 to more than 3 Gy. Bleaching of sand isolated from mouth bar deposits younger than 1.1 ka (n=10) was much improved, with all samples yielding residual doses less than 0.1 Gy within uncertainty. Bleaching of overbank sand showed a trend of improvement with time, although there remained significant variability of the degree of bleaching, with both well- and heterogeneously-bleached overbank sands of all ages (Fig. 6A).

4.5 Spatial trends in bleaching

As the Lafourche subdelta expanded radially (Chamberlain, 2018), the temporal trend in bleaching of mouth bar deposits is also reflected spatially. Mouth bar deposits in the upstream reaches (above ~105 river km) are less well bleached, compared to those in downstream parts. To determine whether bleaching may occur during the overbank/crevasse process, residual doses of overbank sand at the PV, EF, and NV sites were plotted against distance to the present-day bank of Bayou Lafourche. This test revealed no spatial trends within the overbank sands (Supplementary File, Fig. S4), although we do note that overbank sand samples from different depths within the same borehole tend to have similar degrees of bleaching (Supplementary File, Table S1).

4.6 Bleaching of Late Holocene silt inferred from sand/silt pairs

Good agreement was found between the majority (n=5) of sand and silt pairs dated from the same overbank samples (n=7) (Fig. 7), broadly consistent with the findings reported by Shen et al. (2015). Silt ages (obtained from CAM) scattered both higher and lower than sand ages (obtained using bootMAM), indicating that these silts were generally sufficiently bleached for dating. Two samples, PV I-4 and PV I-5, produced silt ages that exceed sand ages by ~ 450 and 580 years respectively. The age overestimation by silt may be due to poor quartz bleaching (Shen et al., 2015). Alternatively, we identified evidence that OSL signals of these two samples contained a contribution from feldspar, which is less readily bleached than the fast-component signal of quartz (Godfrey-Smith et al., 1988;Wallinga, 2002). Despite strong luminescence signals, PV I-4 and PV I-5 had 20% and 17% of aliquots, respectively, rejected from our analysis for poor reproducibility. One additional aliquot (5%) of PV I-4 and 4 additional aliquots (13%) of PV I-5 were rejected from our analysis for not meeting infrared (IR) depletion criteria (Duller, 2003), indicating that etching may not have been entirely effective at removing feldspars for these two samples. In addition, PV I-4 did not exhibit a suitable 110 °C TL peak; rather, the TL signal increased from 110 °C onward (Supplementary File, Fig. S2). The finding that no aliquots of the other five samples, that produced agreeing sand/silt ages were rejected for IR depletion, corroborates our suggestion that age overestimation observed for silt samples PV I-4 and 5 is caused by feldspar contamination.

By coincidence, the samples selected by Shen et al. (2015) for the paired sand/silt analysis featured sand that we mainly classified as well-bleached, with little difference between sand ages obtained with CAM and bootMAM (Supplementary File, Fig. S3). It is possible that greater differences between bleaching of sand and silt could be identified if this test was performed on sediment pairs extracted from deposits with heterogeneously-bleached sand.

5 Discussion

5.1 Bleaching trends and implications to optical dating

It is little wonder that universal trends in bleaching of fluviodeltaic sediments have not yet been identified, considering the complex and numerous pathways river sediments may take prior to deltaic deposition and the natural variability among river systems in general. This study, which focused on one large meandering river and its deltaic deposits across time, identified lower average residual doses for finer sand grains than for coarser sand grains. This trend was observed both for in-transit sediment within the river and for sediments preserved within deltaic deposits (Figs. 3 & 5). Our findings with regard to grain-size-dependent sand bleaching are different from those of studies conducted in other systems (Truelsen and Wallinga, 2003;Olley et al., 1998), which featured smaller primary channels and included more samples of coarser grain sizes than investigated here.

We found that fine silt, moving in suspension within the modern river channel, was more completely bleached than sand moving as bedload (Fig. 3), and that bleaching of silt was also generally sufficient in river sediments deposited prior to human engineering of the system (Fig. 7). This is consistent with recent studies of other contemporary large river systems within their deltaic reaches (Sugisaki et al., 2015;Chamberlain et al., 2017), yet different again from studies of smaller and/or source-proximal rivers and their deposits (Fuchs et al., 2005;Thompson et al., 2018). Although many large rivers are well known to be turbid (e.g., the "Muddy" Mississippi, Morris, 2012;Gramling, 2012;Rutkoff and Scott, 2005), it is possible that turbulence within large and lengthy channels offers sufficient opportunities for bleaching of the finest material moving in suspension, via the constant upwelling of the sediment-laden river water.

Bleaching of mouth bar sand (75-125 and 125-180 µm), which generally includes the coarsest material transported by a distributary system (Wright, 1977), increased in time (Fig. 6A) and coastward (Figs. 6B & 8). Temporal and spatial trends coincide, due to radial growth of the delta through bayhead delta progradation (Chamberlain et al., 2018). The links between both make it difficult to parse the relationship of bleaching to distance versus time.

We offer a few plausible explanations for the spatiotemporal trend in bleaching of mouth bar sand within the Lafourche subdelta (Figs. 6 & 8):

1) The primary alluvial channel is known to have avulsed a number of times throughout the Late Holocene (Saucier, 1994;Chamberlain et al., 2018) and to have migrated via meandering within its channel belts, thereby occupying different pathways within the Lower Mississippi Valley (well upstream of the delta). The timing of channel belt avulsions and meander pathways is not well known. It is possible that a relatively landward avulsion (450-700 linear km inland, see Chamberlain et al., 2018) or divergence in a meandering pathway of the river within one channel belt circa 1.2 - 1.1 ka may have positioned the river in such a way that it mobilized younger deposits, for example by reworking late Holocene channel-belt deposits rather than eroding Pleistocene terrace deposits. Recently-bleached sediments would require less light exposure during transit in the river system to become well-bleached upon arrival and deposition in the delta.
2) Alternatively, the abrupt change in bleaching of mouth bar sand may be linked to hydrologic changes within the delta itself, associated with the activation of the Modern (Balize) subdelta circa 1.4 - 1.0 ka (Hijma et al., 2017). For example, after 1.2 - 1.1 ka much of the bedload may have been rerouted toward the Modern (Balize) subdelta, causing suspended-load transport during high-flow events to be the more dominant mode of sand-delivery to the lower reaches of Lafourche. Although under this scenario, the overbank deposits could be expected to be better bleached, because these are sourced to suspended material (Esposito et al., 2017).
3) Additionally, decreased discharge in Lafourche distributaries (due to partial avulsion to the modern route) or enhanced exposure as Lafourche channel tips prograded seaward and outside of the shelter of the pre-

Lafourche bay may have allowed marine processes to gain importance, potentially altering turbulence, turbidity, salinity, and/or suspension times of sediment at the mouths of Lafourche distributaries. Yet, the residual doses for overbank deposits also show a change in degree of bleaching around 1.1 ka. Although this trend is less clear compared to the mouth bar deposits, it suggests that sediment reworking at the river mouth is not the only explanation.

It is also plausible that these drivers operated in tandem; an avulsion of the alluvial channel may have driven delta-lobe switching circa 1.2 - 1.1 ka, and subsequent mobilization of younger sand upstream plus hydrologic changes at the Lafourche channel mouths that supported more complete OSL bleaching. There are not sufficient data at present to test these hypotheses. Bleaching of mouth bar sand was not found to correlate to depth within the deposit (Fig. 9A), suggesting that improved bleaching was not related to reworking of mouth bar surfaces nor bioturbation, which could be expected to produce greater bleaching for shallower deposits.

Bleaching of overbank deposits was also not found to be improved at shallower depths (Fig. 9B), or with proximity to the trunk channel (Supplementary File, Fig. S4 & S5). Other possible trends in bleaching of overbank sand merit further testing. The degree of bleaching of overbank sand may be linked to opportunities for bleaching during or immediately after deposition (e.g., Cunningham et al., 2011), or even to the time of year (and therefore water velocity and turbulence within the primary channel, e.g., Allison et al., 2014) that deposits formed.

Our study clearly shows that bleaching of deltaic sand is highly time- and/or space-dependent. Had our study only investigated a small subset of the data herein, we could have easily arrived at different conclusions with regard to bleaching by grain size. For example, a comparison of 75-125 µm overbank sands to only the 125-180 µm mouth bar sands younger than 1.1 ka would have indicated that coarse grains are the best bleached sand fraction in the Mississippi Delta, while a comparison to only the older mouth bar sands would have yielded the opposite finding. The complexity of our dataset demonstrates that caution is needed when modern analogues are used to infer the degree of bleaching of paleo-deposits.

5.2 Sediment fingerprinting and relevance to delta restoration

Despite the inherent complexity of river networks, strides are being made toward the use of luminescence signals to fingerprint fluvial sediments and reconstruct the routing of grains (e.g., Sawakuchi et al., 2018;McGuire and Rhodes, 2015). Our study demonstrates how luminescence signal bleaching may link to transport histories and/or the fluvial conditions under which grains are deposited, and gives insight into the last light exposure of sediment grains within a river channel. Such information is of high relevance to sustaining the Mississippi Delta and perhaps other deltas by engineered river diversions (e.g., CPRA, 2017), because the success of diversions will rely in part on their feeder channel's ability to mine sediment from suspended and/or bedload material within the river. For example, it has been proposed that locating diversions near sand bars on the river bed may maximize sand capture, thereby supplying the coarsest material needed to build a solid substrate of new land (e.g., Allison and Meselhe, 2010;Nittrouer et al., 2012;Meselhe et al., 2012). The residence times of river-bed bars and their ability to recharge are not well known, yet could be probed through estimates of the OSL residual doses of the bar sands. The methodology applied herein may thus provide a foundation for future work relevant to delta restoration.

6 Conclusions

- OSL bleaching of sand within a large delta can be highly temporally and/or spatially variable. Inferences about the degree and mechanisms of bleaching of fluviodeltaic sediments should therefore be drawn from large datasets. For dating purposes (e.g., establishing overdispersion of well-bleached samples for age model input), it is best if such datasets include samples from the time interval, depositional environment, and region of interest.
- Quartz silt extracted from late Holocene Mississippi Delta deposits and from suspension within the contemporary Mississippi River were generally well-bleached, consistent with previous findings in other large fluviodeltaic systems. The upwelling of turbid water may therefore play a significant role in bleaching of suspended sediment in large rivers, and quartz silt should be further tested as a viable option for luminescence dating in megadeltas.
- Although there are many unknowns with regard to drivers of luminescence signal bleaching in river sediment, our research demonstrates the potential of this rapidly-advancing tool to yield insight into the routing of sediments through fluvial systems, which is of relevance to delta restoration initiatives.

7 Code Availability

Bootstrap scripts for age modeling are available through the Netherlands Centre for Luminescence dating website (https://www.ncl-geochron.nl/en/ncl-geochron/Service.htm).

8 Data availability

The luminescence data may be accessed through the manuscripts in which they first published, which are referenced herein.

9 Author contributions

The research was designed by EC and JW. Analyses were conducted by EC, with input from JW. Both authors contributed to the interpretation of results and manuscript synthesis.

10 Competing interests

The authors declare that they have no conflict of interest.

11 Acknowledgements

We thank Mead Allison and Michael Ramirez for providing information regarding river conditions during sampling of the contemporary river sediments, and for enabling their collection. This manuscript was improved by comments on an earlier draft provided by Torbjörn Törnqvist, Steve Goodbred, Mead Allison, Barbara Mauz, and Zhixiong Shen, and by discussions with Tony Reimann and participants at the 2017 LED conference.

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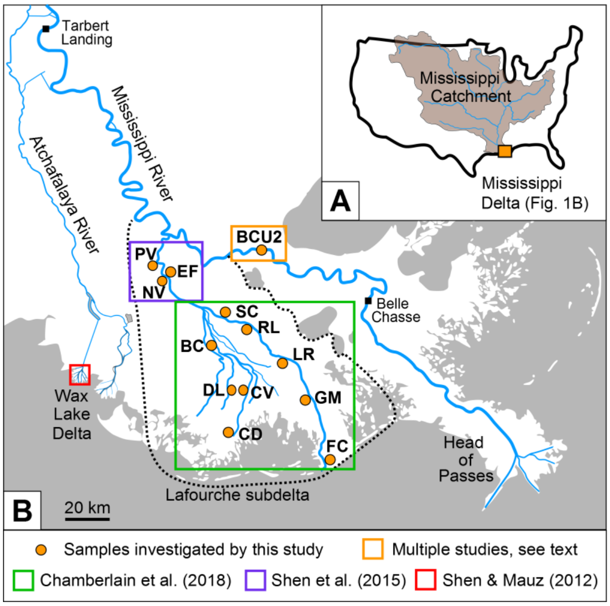

Figure 1. The Mississippi Delta and catchment (A), and locations of contemporary Mississippi River and Lafourche
660 **subdelta samples used for this study and their primary references, plus the locations of previous research in the Wax Lake Delta and of river gauge stations (B). See Sect. 3.1 Compilation of Archival Data for the primary references of the modern river samples (orange box). Modified from Chamberlain et al. (in press).**

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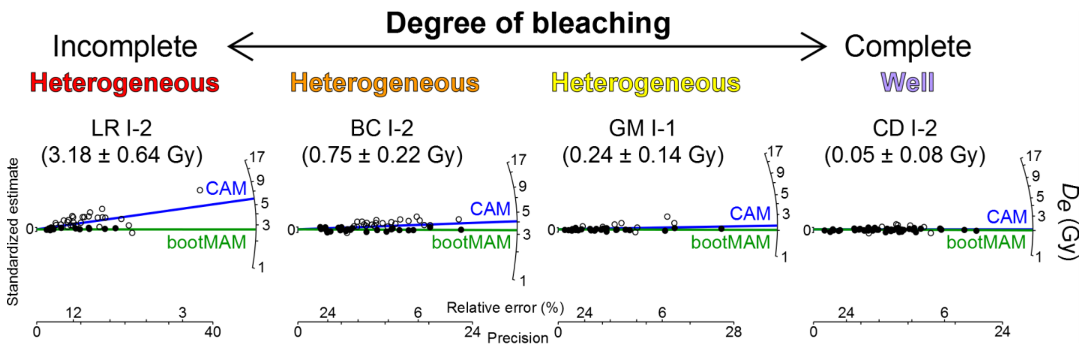
665 

Figure 2. Radial plots of four mouth bar sand samples provide an example of our approach to assessing the degree of bleaching of late Holocene deposits. Our assessment is based on the residual dose, given in parentheses, obtained from the difference in equivalent doses (*D_es*) estimated with the bootstrapped Minimum Age Model (bootMAM) and Central Age Model (CAM). Filled data points represent aliquots for which the *D_e* estimate agrees with the sample *De* obtained from bootMAM within 2σ uncertainty. Adapted from Chamberlain et al. (in press).

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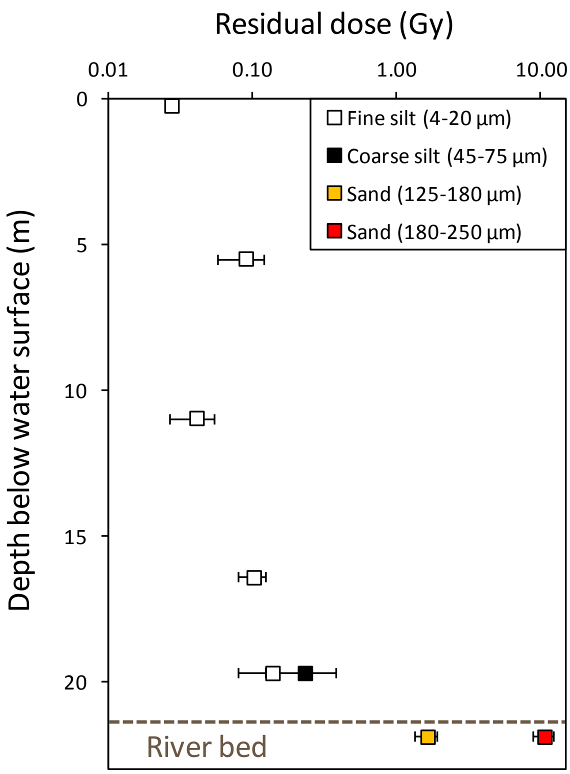

Figure 3. Residual doses of quartz silt and sand from sediments in transit in the modern Mississippi River, with sample depth in the river channel.

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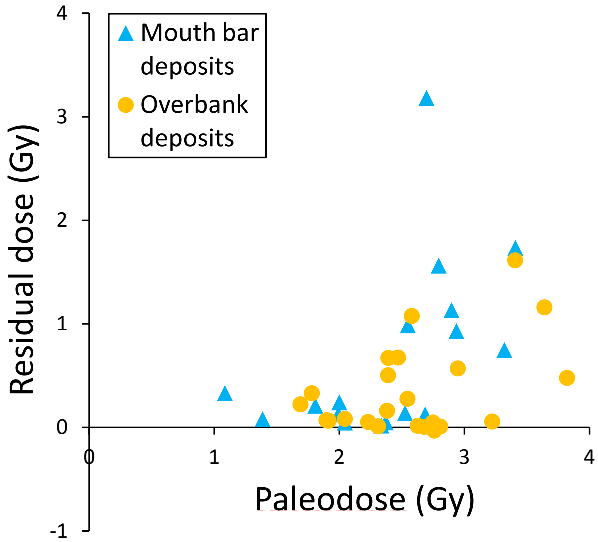

Figure 4. Residual doses calculated as $D_{e,CAM} - D_{e,bootMAM}$ versus the paleodose estimated as $D_{e,bootMAM}$ for mouth bar and overbank deposits of the Lafourche subdelta. Uncertainties, not shown here due to the high density of data points, are given in Table S1.

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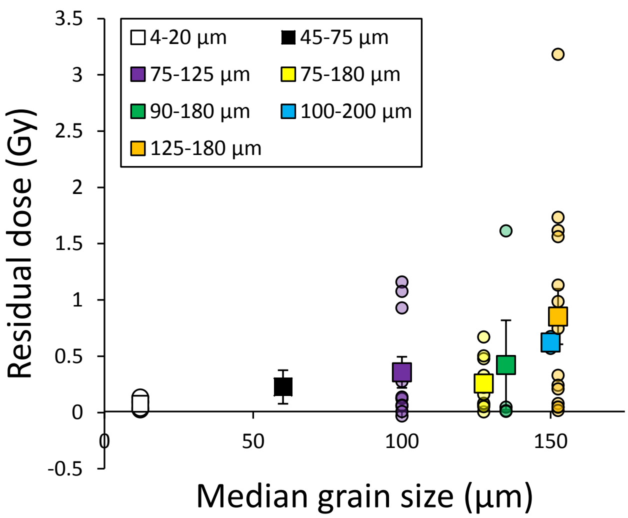

Figure 5. Mean (boxes) and individual (circles) residual doses by median grain size (see legend) for silt and sand samples of all depositional environments. Data are not shown for the 180-250 µm fraction, which consisted of only one sample and would plot outside the graph.

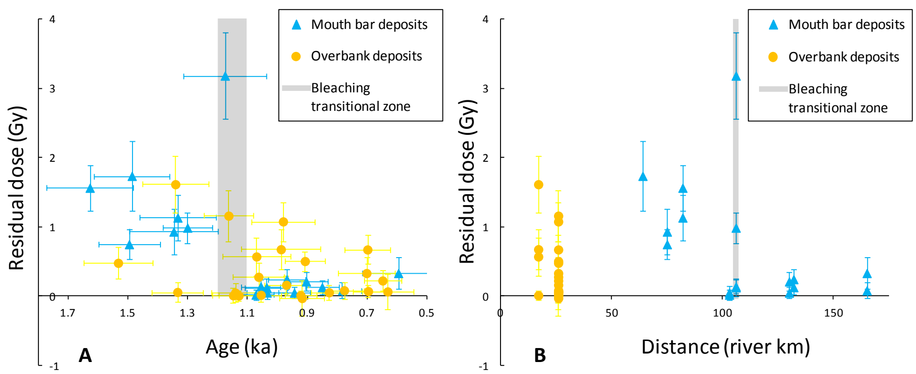

Figure 6. Bleaching of sands isolated from mouth bar and overbank deposits with (A) burial age obtained using the bootMAM OSL approach, and (B) distance seaward, in river kilometers relative to the junction of the modern river channel and Bayou Lafourche. The shaded region indicates the transition zone from heterogeneously- to well-bleached mouth bar deposits circa 1.2-1.1 ka, or around 100 river km seaward of the junction.

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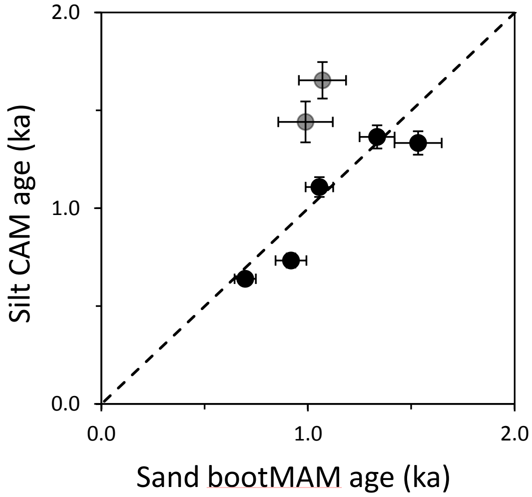

Figure 7. Comparison of ages of paired sand and silt fractions isolated from the same samples (n=7) of overbank deposits, collected by Shen et al. (2015) and reanalyzed here using early background subtraction plus other criteria. Gray circles indicate PV I-4 and PV I-5, two samples possibly affected by feldspar contamination or containing poorly bleached silt.

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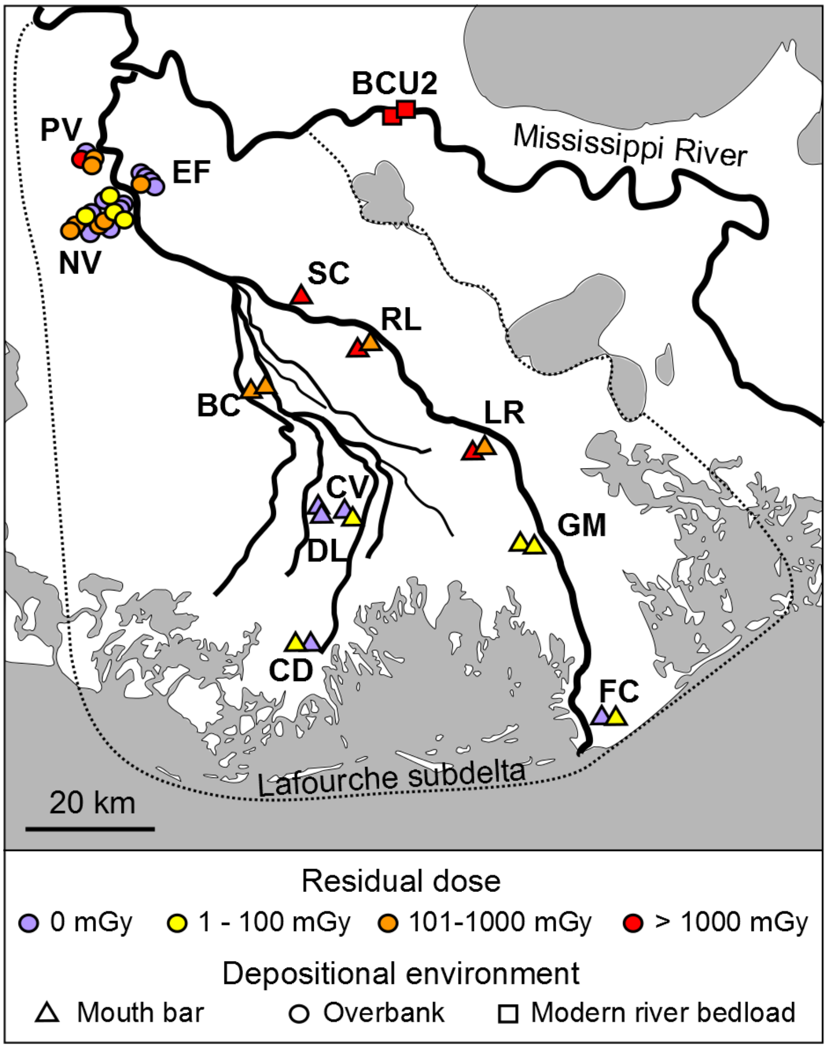

Figure 8. Geographic distribution of sands and their minimum residual doses, defined as the residual dose minus its uncertainty.

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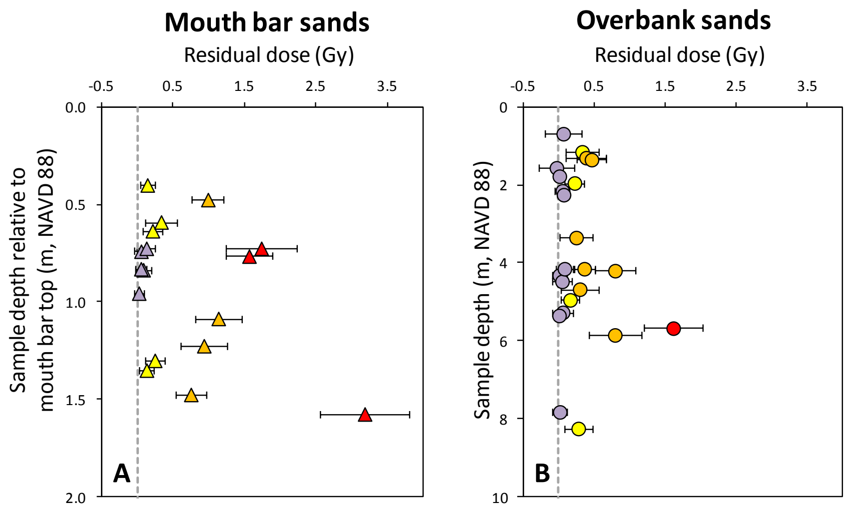

Figure 9. Bleaching of mouth bar (A) and overbank (B) sands with depth. Mouth bar sand depths are relative to the top of the mouth bar deposit, which formed at roughly sea level. Overbank sand depths are relative to mean sea level. Data points are color-coded by their minimum residual dose (see Fig. 8).