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Radiogenic fingerprinting reveals anthropogenic and buffering controls on sediment dynamics of the Mississippi River system

Samuel E. Munoz[1,2,3*], Liviu Giosan[3], Jurek Blusztajn[3], Caitlin Rankin[4], and Gary E. Stinchcomb[5,6]

[1]*Department of Marine & Environmental Sciences, Northeastern University, Boston, Massachusetts 02115, USA*

[2]*Department of Civil & Environmental Engineering, Northeastern University, Boston, Massachusetts 02115, USA*

[3]*Department of Geology & Geophysics, Woods Hole Oceanographic Institution, Woods Hole, Massachusetts 02543, USA*

[4]*Department of Anthropology, Washington University, Saint Louis, Missouri 63130, USA*

[5]*Watershed Studies Institute, Murray State University, Murray, Kentucky 42071, USA*

[6]*Department of Geosciences, Murray State University, Murray, Kentucky 42071, USA*

*E-mail: *s.munoz@northeastern.edu*

ABSTRACT

Radiogenic isotopes of strontium ($^{87}Sr/^{86}Sr$) and neodymium ($^{144}Nd/^{143}Nd$) are widely used to trace sediment across source-to-sink networks, with samples typically collected from outcrops at basin headwaters and sediments along the channel margin, floodplain, and/or sea floor. Here we establish Sr-Nd isotope systematics of recent (< 1k yrs.) Mississippi River (USA) basin alluvial sediments, evaluate the sensitivity of these isotope systems to the presence of artificial impoundments that trap sediments behind

them, and test their ability to provenance mixed sedimentary records. Sediment cores collected from floodplain depressions and oxbow lakes along the Mississippi and its major tributaries, where an extensive lock and dam system was constructed during the mid-20th century, show that the isotopic signatures of major tributaries are distinct, and that some of these signatures shift following dam closure. We then use mixing models to demonstrate that, near the confluence of major tributaries, Sr and Nd isotope signatures can be used to provenance sediments deposited in floodplain lakes during overbank floods. Further downstream, where sediments are well mixed, the provenance of overbank deposits is more challenging to evaluate using Sr-Nd isotope systematics. Given the global pervasiveness of artificial impoundments on rivers, our findings imply that widely employed sediment fingerprinting techniques based on modern conditions may not be representative of conditions from as recently as a century ago.

INTRODUCTION

Sediment transport from source to sink is a fundamental process that shapes landscapes and the geological record (Allen, 2008). Radiogenic isotopes of strontium and neodymium are widely used as sediment tracers to identify source area and patterns of erosion and deposition within and across drainage networks (e.g., McLennan et al. 1989; Clift et al., 2008; Padoan et al., 2011). Basin-wide systematics of Sr and Nd isotopes are typically evaluated by collecting samples from outcrops at basin headwaters (i.e., source rocks), along channel margins, the floodplain surface, and/or offshore (i.e., mixed sediments). Over the last century, artificial impoundments that trap sediments have been built on many of the world’s major rivers and their tributaries, profoundly altering the downstream delivery of sediments (Syvitski et al., 2005). The potential for these artificial

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impoundments to influence Sr and Nd isotope systematics has previously been recognized (e.g., Padoan et al., 2011) but, to our knowledge, the magnitude of this influence has yet to be evaluated.

Here, we examine the Sr and Nd isotope systematics for the Mississippi River system, USA (Fig. 1). The Mississippi is the largest river in North America and one of the world's most heavily engineered drainage networks (Knox, 2007), where dams constructed primarily during the mid-20th century on the Missouri, Upper Mississippi, and Ohio Rivers (Fig. DR1) have reduced average annual sediment fluxes to the Gulf of Mexico by 50%–70% (Horowitz, 2010). We measured isotopes of Sr and Nd on sediment samples collected from the floodplains of the Missouri, Upper Mississippi, and Ohio Rivers to evaluate the isotopic variation among the major tributaries of the Mississippi River system before and after dam closure. Using these data, we constructed Bayesian mixing models (Parnell et al., 2013) to evaluate the provenance of flood deposits preserved in oxbow lakes previously used in paleoflood reconstructions of the Upper Mississippi (Munoz et al., 2015) and Lower Mississippi Rivers (Munoz et al., 2018). Together, our analyses establish Sr-Nd isotope systematics for the largest drainage network in North America, test their ability to provenance mixed sedimentary records, and allow us to evaluate the sensitivity of these radiogenic tracers to artificial impoundments.

MATERIALS AND METHODS

We collected a sediment core using a gouge auger in October 2016 from three in-filling depressions in the floodplains of the Missouri (38.664869°N, 90.702690°W; core length: 97 cm), Upper Mississippi (39.112535°N, 90.695270°W; core length: 137 cm), and Ohio Rivers (37.166491°N, 89.064583°W; core length: 96 cm) that are periodically inundated by floodwaters and are downstream of most dams (Fig. 1; Fig. DR2). To establish chronological control on these cores, we measured ^{137}Cs activity on bulk sediment samples at 5 cm resolution on the upper 60 cm of each core in a Canberra GL2020RS well detector for low-energy germanium radiation (Fig. DR3). We collected five to seven samples from each core that were stratigraphically >20 cm below the base of ^{137}Cs activity (i.e., AD 1954) and above the peak of ^{137}Cs activity (i.e., AD 1963) to represent the depositional environment before and after the closing of the majority of artificial impoundments on the Mississippi River system (Fig. DR1 in the GSA Data Repository[1]). To test the significance of difference between pre- and post-dam samples, we used Welch's *t*-test implemented in R.Studio v.1.1.423 (*t.test* function) because this provides a conservative test of difference in the mean of two samples that is appropriate for small sample sizes with unequal variance (Welch, 1947).

To minimize grain-size and hydraulic sorting effects, the <63 μm filtrate was used for grain size and isotopic analysis. For grain-size analysis, organics were removed by burning at 360 °C in a muffle furnace, then dispersed in water before 5 s. sonication and analysis in a Beckman Coulter LS 13 320 laser diffraction particle-size analyzer. Nd and Sr chemistry was performed with conventional ion chromatography. Strontium was separated and purified from samples using Sr-Spec (Eichrom) resin. Nd chemistry was measured with LN resin (Eichrom) following method described by Scher and Delaney (2010). Sr and Nd analyses were conducted on a NEPTUNE multi-collector ICP-MS with internal precision around 10–20 ppm (2σ). The $^{143}Nd/^{144}Nd$ and $^{87}Sr/^{86}Sr$ ratios for unknowns were normalized by the offset between our average measured value of the Nd

La Jolla and Sr NBS987 standards during the analytical session and the preferred $^{143}Nd/^{144}Nd$ of 0.511847 (White and Patchett, 1984) and $^{87}Sr/^{86}Sr$ of 0.710240 (Jackson and Hart, 2006), and external precision is estimated to be 15–25 ppm (2σ). $^{143}Nd/^{144}Nd$ isotopic composition is expressed further as ε_{Nd} (DePaolo and Wasserburg, 1976) units relative to $(^{143}Nd/^{144}Nd)CHUR$ = 0.512638 (CHUR—chondritic uniform reservoir). We tested the influence of grain-size on Sr and Nd isotopes (McLennan et al., 1989) using regression to find a significant linear relationship between ε_{Nd} and the mode of grain-size on Missouri River and oxbow lake samples ($R^2 = 0.7711$, $p < 0.001$) but not the other tributaries (Fig. DR4) nor between grain size and $^{86}Sr/^{87}Sr$ ratios. We thus used linear regression to normalize εNd of Missouri River and oxbow lake samples to the pooled mode of grain-size measurements (Table DR1).

To compare the isotopic composition of the mixed sediments collected as part of this study with that of the source rocks that underlie the Mississippi River basin, we extracted the Sr and Nd isotope measurements within 500 km of the basin (*n* = 1356) from the GeoRoc database (http://georoc.mpch-mainz.gwdg.de/georoc/; Sarbas and Nohl, 2008) and assigned each sample to its corresponding geological unit based on its location. To evaluate the potential of Sr-Nd isotopes to provenance sedimentary paleoflood records from oxbow lakes on the Mississippi River, we collected two samples from previously dated cores at Horseshoe Lake, Illinois (HRM) ~20 km below the confluence of the Missouri River (Munoz et al., 2015) and four samples from Lake Mary, Mississippi (MRY) ~1000 km below the confluence of the Ohio River (Munoz et al., 2018), and performed grain-size and isotope analysis on these samples using the same approach as described above (Fig. 1). Chronologies of these oxbow paleoflood records were developed using a Bayesian age model informed by ^{137}Cs, ^{210}Pb, ^{14}C, optically-stimulated luminescence (OSL) and stratigraphic markers; additional details of their age models can be found in their original publications (Munoz et al. 2015; 2018). At HRM, one sample is from a fine-grained deposit dated to A.D. 1160 ± 90 yr (reported as median age ± 2σ confidence interval) interpreted by Munoz et al. (2014) as the suspended load from a large Mississippi River flood event, and one sample from the same core dated to A.D. 1450 ± 160 yr that was not associated with a flood. At MRY, we collected samples of fine-grained sediment immediately overlying prominent flood deposits dated to A.D. 1917 ± 13 yr, 1934 ± 12 yr, 2011 ± 7 yr, and 2014 ± 6 yr interpreted by Munoz et al. (2018) to represent major historic floods in A.D. 1927, 1937, 2011, and 2016, respectively. To evaluate the provenance of the oxbow lake sediments, we constructed Bayesian mixing models using the *simmr* package (Parnell et al., 2013) in RStudio using pre- and post-dam means and standard deviations for each tributary as end members, with only the Missouri and Upper Mississippi for deposits at HRM and all three sampled tributaries for deposits at MRY.

RESULTS AND DISCUSSION

Mississippi River Sr-Nd Isotope Systematics

Mississippi River system floodplains sediment Sr and Nd isotopic compositions exhibit a wide range of values that group into three distinct clusters, where these differences are associated with the major tributaries of the Missouri, Upper Mississippi, and Ohio (Fig. 2). The εNd values and $^{87}Sr/^{86}Sr$ ratios of all floodplain sediments range from −14.75 to −11.55 and 0.716688–0.729936, respectively. Floodplain samples from the tributaries are highly differentiated over the $^{87}Sr/^{86}Sr$ range, with the lowest values for the Missouri River (mean, $\bar{x}$ = 0.717807, standard deviation, σ = 0.000095) and the highest values from the Ohio River ($\bar{x}$ = 0.728123, σ = 0.000062) and no overlap among the three tributaries sampled. Over the range of εNd values, samples from the Missouri and Upper Mississippi exhibit high variability (range 1.73 and 1.95, respectively), with less variability in samples from the Ohio River (range of 0.98). All of the floodplain samples fall within the εNd range for the Archean shield and Paleoproterozoic orogens that have been subsequently incorporated into Paleozoic through Cenozoic age marine and terrestrial sedimentary rocks covering much of the Mississippi basin (Peucker-Ehrenbrink et al., 2010).

In contrast to Nd, strong radiogenic Sr enrichment is evident in Mississippi basin sediments relative to source rocks (Fig. 2a). For the Sr system, older sedimentary cover exhibits higher $^{87}Sr/^{86}Sr$ ratios relative to source rock composition (e.g., Peucker-Ehrenbrink et al., 2010). Incongruent weathering of source rocks is the primary factor leading to this enrichment as it elevates $^{87}Rb/^{86}Sr$ and favors ingrowth of radiogenic $^{87}Sr/^{86}Sr$ in the resulting sedimentary rocks; this effect is enhanced when sedimentary rocks are recycled multiple times (e.g., Taylor et al., 1983). Sedimentary carbonates are rich in Sr and less radiogenic than siliciclastic rocks, but they are roughly evenly distributed across all three basins examined (King and Beikmanm, 1974). In comparison to Missouri River sediments, lower εNd and enriched $^{87}Sr/^{86}Sr$ of upper Mississippi River sediments likely reflect erosion of glacial till and loess originating from the Precambrian Canadian Shield (e.g., Peucker-Ehrenbrink et al., 2010). The lack of radiogenic enrichment for Nd but high $^{87}Sr/^{86}Sr$ ratio for Ohio sediments suggest instead erosional inputs from old unglaciated sedimentary rocks outcropping in this sub-basin (e.g., Oh and

Raymond, 2006). In short, the three major tributaries of the Mississippi River system exhibit distinct Sr-Nd isotopic signatures as a result of differences in underlying parent rock material, sedimentary cover and weathering rates of each basin.

Floodplain sediments deposited prior to and after the closure of artificial impoundments sometimes exhibit significant differences in their isotopic composition (Fig. 2b). On the Missouri and Upper Mississippi River, $^{87}Sr/^{86}Sr$ ratios increase from pre- to post-dam samples, with significant shifts ($p < 0.05$; unpaired t-tests) in their means (Table DR2). This enrichment of radiogenic Sr may be related to sorting of the sediment load by dams that keep lighter radiogenic minerals (e.g., muscovite) suspended but trap the less radiogenic heavier fraction (Garçon et al., 2014). Alternatively, these shifts in the isotopic composition of Missouri and Upper Mississippi River sediments may be due to trapping of sediments from the upper reaches of their basins behind dams (Meade and Moody, 2010) assuming these regions provide less radiogenic Sr. Sediments from the Ohio River, in contrast, do not exhibit a significant shift in the either $^{87}Sr/^{86}Sr$ ratio or εNd values ($p > 0.1$). The insensitivity of Ohio River sediments to the establishment of dams may reflect the placement of the lock and dam system on that tributary relative to the glaciated and unglaciated erodible lithologies. Overall, our data demonstrate that the isotopic composition of sediments transported by the Mississippi River system has shifted markedly over the last century, implying that dams can alter isotope systematics of a continental drainage network.

Provenance of Mixed Sediments

The isotopic composition of sediments deposited in floodplain lakes below the confluence of major tributaries reflect both sediment provenance and the degree of mixing (Fig. 3). The Missouri River is the dominant source of suspended sediment to the lower Mississippi River (Knox, 2007), and this is reflected in the isotopic signatures of sediments in oxbow lakes, which tend to have large contributions from the Missouri River. In sediments from Horseshoe Lake (HRM), situated on the Upper Mississippi River, a prehistoric deposit that was interpreted by Munoz et al. (2014) to be the result of an overbank flood is composed primarily of Missouri River sediment ($\bar{x}$ = 80%, σ = 10%; Fig. 3a). Background sediment deposited in HRM, in contrast, is more similar in composition to material from the Upper Mississippi River ($\bar{x}$ = 71%, σ = 13%; Fig. 3b) confirming the extra-local provenance of the presumed flood deposit in HRM as hypothesized by Munoz et al. (2014; 2015). Further downstream, sediments in Lake Mary (MRY) associated with the 1937 and 2011 floods are also composed primarily (~60%) of Missouri River sediments and moderate contributions from the Upper Mississippi (~25%) and Ohio (~15%), with only minor compositional differences between the two events (Figs. 3c and d).

The 1937 and 2011 floods rank among the largest historical floods by discharge on the lower Mississippi, but these events differed in their hydrometeorological properties. The 2011 flood was triggered by large spring rainstorms over the lowermost part of the basin, while the 1937 event was a winter flood caused by rainfall falling primarily over the Ohio River basin (Smith and Baeck, 2015). Given the differences in water source between the 1937 and 2011 floods, we expected the 1937 event to have a larger contribution from the Ohio River, but mixing models demonstrate that the sediments deposited by these two events are compositionally indistinguishable (Figs. 3c and 3d). The compositional homogeneity of these sediments likely reflects buffering (i.e., mixing and reworking of sediments) along the lower Mississippi alluvial plain constructed with sediment shed from the continental interior during the Cenozoic (Knox, 2007). We note that homogeneity in sediment composition was also observed in other samples from the lower Mississippi River (Table DR1), implying that buffering is a persistent effect along the lowermost reaches of the river. These findings imply that isotope systems used to trace sediments are most useful to provenance fluvial deposits when geological contrasts are not overwhelmed by buffering (e.g., near a tributary with distinctive sediment geochemistry). Over longer timescales than those considered here, more pronounced shifts in sediment composition may be distinguishable downstream or offshore using the Sr-Nd isotope system.

CONCLUSIONS

Sediments of the three major tributaries of the Mississippi River system—the Missouri, Upper Mississippi, and Ohio Rivers—exhibit strong contrasts in their Sr-Nd isotopic signatures as a result of their underlying geology. Dam closure can significantly affect this isotopic composition via selective trapping and/or sorting of the sediment load. We also demonstrate that these isotopes can provenance recent (< 1 kyr B.P.) mixed sediments deposited in floodplain lakes, and show how they are sensitive to alluvial plain buffering that increases downstream from confluences of major tributaries. The data and analyses presented in this study will be useful for establishing the dynamics of erosion and sedimentation for the largest river system in North America, and has implications for similar studies on other large regulated rivers—including the Nile, Yellow, and Indus Rivers—with sharply reduced sediment loads as a result of dams. Our findings imply that dams act as valves that regulate tributary contributions to the main stem, and can generate measurable shifts in sediment provenance. These findings indicate that widely employed sediment fingerprinting techniques for current conditions may not be representative of past conditions as recently as a century ago.

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FIGURE CAPTIONS

Figure 1. The Mississippi River system and the geological provinces of its basin (Hoffman, 1989), showing locations of tributary floodplain sediment samples collected for this study, oxbow lake sediment samples (Munoz et al. 2015; 2018), and artificial impoundments (dams on the Missouri River; lock and dam structures on the Upper Mississippi and Ohio Rivers). More detailed maps of floodplain sediment sampling locations are available in Fig. DR2.

Figure 2. Radiogenic isotopes of Sr ($^{87}Sr/^{86}Sr$) and Nd (ε_{Nd}) of parent material and sediments of the Mississippi River system. A: Envelopes ('loops' of bag plots excluding outliers) of samples from source rocks surrounding the Mississippi River basin (Sarbas and Nohl, 2008) and fluvial sediment samples collected in this study. B: Detail of fluvial sediments identifying floodplain samples by tributary and timing (i.e., pre- or post-dam), and oxbow lake samples by site and age.

Figure 3. Density plots describing the proportion of a floodplain lake sample contributed by each tributary derived from Bayesian mixing model *simmr* (Parnell et al., 2013). A: A.D. 1160 flood in at Horseshoe Lake, Illinois (HRM). B: A.D. 1450 background in HRM. C: A.D. 1937 flood in Lake Mary, Mississippi (MRY). D: A.D. 2011 flood in MRY.

[1]GSA Data Repository item 2018xxx, supplemental data tables (Tables DR1 and DR2) and figures (Figs. DR1–DR4), is available online at http://www.geosociety.org/datarepository/2018/, or on request from editing@geosociety.org.