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Abstract
Purpose The science of sediment fingerprinting has been evolving rapidly over the past decade and is well poised to improve our understanding, not only of sediment sources, but also the routing of sediment through watersheds. Here, we discuss channel–floodplain processes that may convolute or modify the sediment fingerprinting signature of alluvial bank/floodplain sources and explore the use of nonconservative tracers for differentiating sediment derived from surface soil erosion from that of near-channel fluvial erosion.

Materials and methods We use a mathematical model to demonstrate the theoretical effects of channel–floodplain exchange on conservative and nonconservative tracers. Then, we present flow, sediment gauging data, and geochemical measurements of long- (meteoric beryllium-10, ^{10}Be) and short-lived (excess lead-210 and cesium-137, ^{210}Pb$_{ex}$ and ^{137}Cs, respectively) radionuclide tracers from two study locations: one above, and the other below, a rapidly incising knick zone within the Maple River watershed, southern Minnesota.

Results and discussion We demonstrate that measurements of ^{10}Be, ^{210}Pb$_{ex}$, and ^{137}Cs associated with suspended sediment can be used to distinguish between the three primary sediment sources (agricultural uplands, bluffs, and banks) and estimate channel–floodplain exchange. We observe how the sediment sources systematically vary by location and change over the course of a single storm hydrograph. While sediment dynamics for any given event are not necessarily indicative of longer-term trends, the results are consistent with our geomorphic understanding of the system and longer-term observations of sediment dynamics. We advocate for future sediment fingerprinting studies to develop a geomorphic rationale to explain the distribution of the fingerprinting properties for any given study area, with the intent of developing a more generalizable, process-based fingerprinting approach.

Conclusions We show that measurements of conservative and nonconservative tracers (e.g., long- and short-lived radionuclides) can provide spatially integrated, yet temporally discrete, insights to constrain sediment sources and channel–floodplain exchange at the river network-scale. Fingerprinting that utilizes nonconservative tracers requires that the nonconservative behavior is predictable and verifiable.

Keywords Beryllium-10 · Erosion · Fingerprinting · Lead-210 · Sediment

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1 Introduction

Sediment is routed through landscapes and river networks in an episodic and nonuniform manner (Allen 2008; Burt and Allison 2010). A critical gap in understanding the sediment routing system lies in the prediction of temporary storage and resuspension within the channel–floodplain complex (Pizzuto et al. 2014). Exchange of sediment between channels and their floodplains plays an important role in the complex patterns that we observe in sediment transport at the watershed scale (Leopold and Wolman 1957; Benda and Dunne 1997; Walling et al. 1999; Lauer and Parker 2008). Understanding this component of the sediment routing system is essential for developing mechanistic landscape evolution models over geologic timescales as well as rigorous predictive models of soil loss, carbon fluxes, sediment transport, and water quality over human timescales (Van Oost et al. 2007; Skalak and Pizzuto 2010; Pelletier 2012; Broothaerts et al. 2013). Moreover, the amount and mobility of sediment in a system influences the lag time between implementation of sediment reduction strategies in a watershed and observable improvements in water clarity at the watershed mouth.

A suite of physical, chemical, and biological factors govern the rates of erosion, transport, and deposition. However, a complete, predictive understanding of these factors, and the interactions between them, is out of reach at present (de Vente et al. 2013). Even simply monitoring the sediment routing system functioning at the watershed scale is exceedingly difficult because it is the sum of many small, often imperceptible, changes (mm of erosion or deposition), which occur infrequently and in localized hotspots, over enormous areas (Fryirs et al. 2007). High-resolution geomorphic change detection techniques using aerial or terrestrial light detection and ranging (lidar) are transforming our ability to monitor individual hotspots, but upscaling to an entire watershed in a meaningful way is nontrivial (Wheaton et al. 2010; Smith et al. 2011; O'Neal and Pizzuto 2011). Therefore, measurement techniques that are able to integrate over space and discretize over time are particularly useful to study the cascade of processes that comprise the sediment routing system.

The field of sediment fingerprinting has evolved over the past four decades with the primary goal of providing insight into the relative importance of sediment sources within a watershed (Davis and Fox 2009; Gellis and Walling 2011; Mukundan et al. 2012; Koiter et al. 2013; Walling 2013). Sediment fingerprinting approaches can be categorized into two broad categories, namely geographic fingerprinting and geomorphic fingerprinting (termed "spatial source" and "source type" fingerprinting, respectively, by Walling (2013). Geographic fingerprinting seeks to determine the relative importance of different geographic locations (e.g., two subbasins underlain by different rock types, the mineralogy of which can be used to determine provenance; Kelley and Nater 2000). Geomorphic fingerprinting seeks to identify which landforms (i.e., upland soils versus channel banks) or processes (i.e., landslides, sheet wash, etc.) contribute sediment that passes a given location (e.g., a gauging station).

Geographic fingerprinting invariably relies on some amount of serendipity that the landscape is naturally parsed into geochemically distinct units that align with study objectives. Geomorphic fingerprinting may also benefit from serendipity in that the landforms or processes of interest may or may not fractionate geochemical properties (or other distinguishing characteristics) in a useful or interpretable manner. Thus, the tracers used for fingerprinting must be selected carefully, and in the case of geomorphic fingerprinting, should be identified in the context of the geomorphic and pedogenic processes causing the differential signatures observed as well as alterations caused by past and current human activities (Smith and Blake 2014). The vast majority of fingerprinting studies focus on the fine sediment fraction that is primarily transported in suspension (typically <125 μm) and in this paper we likewise focus discussion on that fraction.

Distinguishing between sediment derived from upland soil erosion versus channel–floodplain erosion is of utmost importance for understanding watershed sediment dynamics. Here, we propose that sediment fingerprinting techniques utilizing a combination of geochemical tracers that exhibit conservative and nonconservative behavior over sediment routing timescales (10^2–10^4 years, e.g., long- and short-lived radionuclide tracers) are well poised to distinguish between these two fundamental sources of sediment. We define conservative tracers as those whose defining characteristic does not change appreciably over sediment routing timescales (e.g., concentration of a given element that does not change during erosion, transport, or deposition). Nonconservative tracers are defined as those whose distinguishing characteristic does change during sediment transport, deposition, and resuspension processes that comprise the sediment routing system. Useful nonconservative tracers are those whose defining characteristic changes in some predictable manner (e.g., decay of a radionuclide tracer during transport or storage). It is important to note that our definition of conservative and nonconservative tracers is specific to sediment routing timescales, typically multiple decades to millennia. Previous work has clearly explained the importance of demonstrating that all fingerprinting tracers are conservative over "event" timescales (i.e., days to years). We uphold this requirement for short-term observations, but demonstrate the usefulness of recognizing the nonconservative nature of some tracers over the longer timescales of sediment routing. While the particular suite of tracers to be used may vary from watershed to watershed, the notion that conservative and nonconservative tracers can be used in combination to inform our understanding of sediment routing is generalizable and may provide a framework from which more standardized sediment fingerprinting practices may develop.

This paper is organized as follows. In section 2.1, we discuss the importance of distinguishing between terrestrial and channel–floodplain sediment sources. Section 2.2 explains channel–floodplain exchange processes that may convolute or modify geochemical signatures of sediment. Section 2.3 discusses the use of conservative and nonconservative tracers in sediment fingerprinting. Section 3 applies the suite of beryllium-10 (^{10}Be), lead-210 (^{210}Pb), and cesium-137 (^{137}Cs) to the Maple River watershed in south-central Minnesota, USA, and discusses caveats for sediment fingerprinting with conservative and nonconservative tracers. Section 4 summarizes results and discusses generalizable insights from sections 2 and 3.

2 Theoretical context

2.1 Fingerprinting to distinguish between terrestrial and fluvial sediment sources

Determining the proportion of fine (i.e., suspended) sediment derived from upland (terrestrial) soil erosion versus channel–floodplain (fluvial) erosion is an intriguing topic for those engaged in basic research of understanding how sediment is routed through river networks. However, distinguishing between these two fundamental sources of sediment also carries great significance in applied geomorphology, which aims to implement effective sediment management strategies. These two fundamental source areas represent distinct transport regimes and the implications/options for sediment management differ substantially depending on the relative importance of each.

Sediment fingerprinting can distinguish between terrestrial and fluvial sources in a way that integrates space and discretizes time. Samples of suspended sediment collected at any given time represent a snapshot of what is contributed from active sources upstream. However, delivery of sediment to any given point in the catchment depends on travel distance, transport rates, and often variable amounts of storage between the source area and the suspended sediment sampling point of interest. This caveat is especially important in large watersheds, which typically contain many opportunities for short- or long-term storage. As one independent line of evidence, sediment fingerprinting can thus provide valuable insight to inform watershed modeling and sediment budgets.

Many other tools are useful for determining the relative importance of sediment sources at a local scale (<1 km^2), but few can make reliable predictions at larger scales. For example, watershed hydrology/erosion simulation models (e.g., Soil and Water Assessment Tool, SWAT; Water Erosion Prediction Project, WEPP), most of which are driven by the Universal Soil Loss Equation (USLE) or some derivative thereof, can be useful to identify the relative sensitivity of erosion at the local scale (<1 km^2). However, given the immense scatter in empirical relationships between measured erosion rates and the factors we know to be important (many of which are indeed included in the USLE), such approaches cannot necessarily produce accurate predictions of the absolute amount of sediment generated even at the local scale, despite widespread application of USLE for that purpose (Trimble and Crosson 2000; Montgomery 2007). More importantly, there exists no physical basis to predict sediment delivery at larger landscape scales (>100 km^2: Walling 1983; Trimble and Crosson 2000; de Vente et al. 2013), which limits the ability of USLE-based approaches to predict suspended sediment fluxes at the scale of moderate to large watersheds (>10^2 km^2). While it is tempting to circumvent this problem by simply calibrating a USLE-based model using the sediment flux measured at the watershed outlet (and thus tune soil erosion rates to observed sediment flux), this is done with the implicit assumption that the channel–floodplain network is in equilibrium (i.e., bank, floodplain, and channel bed erosion do not constitute a net supply or sink of fine sediment), an assumption that may be hard to defend in many watersheds over the simulation timescales and is nevertheless very rarely addressed. Instead, sediment fingerprinting techniques that distinguish between upland and fluvial sources can provide independent information to help calibrate terrestrial and/or fluvial erosion models to the appropriate proportions of the measured sediment loads. Having an independent line of evidence to distinguish between terrestrial and fluvial sources is especially important in landscapes experiencing systematic changes in precipitation or land and water management. Such systematic changes may shift the dominant sediment source without changing sediment flux, as shown by Belmont et al. (2011) and Schottler et al. (2014).

2.2 Floodplain processes that may convolute or modify the fingerprinting signal

Floodplains represent near-channel reservoirs where fluvial sediment can be temporarily stored for decades to millennia during transport from source to ultimate sink. Because sediment stored in the floodplain is necessarily a mixture of sediment derived from all sources upstream from that point, any conservative characteristics of that sediment should be expected to represent some weighted average of the relative contributions from upstream sediment sources. It should be noted that floodplains are not perfect integrators of upstream sediment sources because the sediment delivered to floodplains typically only represents sediment in transport during very large events when lateral and vertical accretion processes typically occur. Nevertheless, some have found floodplains and terraces to be useful integrators of upstream sediment sources and some have even used floodplains as archives documenting changes in sources over time (e.g., Owens et al. 1999; Bridgland 2000; Hesselink et al. 2003; Berner et al. 2012; Stout et al. 2014).

Channels primarily deposit sediment in floodplains by lateral and vertical accretion or infilling of channel cutoffs (Nanson and Croke 1992). Sediment is contributed back to the channel from the floodplains via bank erosion, sheet wash erosion over the floodplain surface, or channel incision through floodplain deposits following avulsion or a drop in local base level. We refer to the combination of all such processes simply as channel–floodplain exchange and note that the prevalence of such processes varies considerably from river to river and even within any given river system. Channel–floodplain systems for which mass erosion is equal to mass deposition over some defined space and time scales are said to be in equilibrium (Gilbert and Dutton 1877; Mackin 1948; Leopold and Wolman 1957). Aggrading channel–floodplain systems are those in which deposition outpaces erosion and degrading systems are those in which erosion outpaces deposition.

While it is well documented and accepted that such exchange processes occur, constraining the rates of channel–floodplain exchange has proven difficult. Even identifying the sign of the sediment mass balance (+ or −) is not feasible in some cases (Grams and Schmidt 2005; Erwin et al. 2011, 2012). Nevertheless, channels and floodplains exchange sediment, and therefore floodplains represent an imperfectly averaged reservoir within which conservative fingerprinting tracers are mixed. For this reason, it is rare to find conservative floodplain (i.e., channel bank) tracers that are independent of their upstream source areas. From a review of 48 fingerprinting studies, we identified 13 that isolated floodplain/bank as a distinct sediment source. Figure 1 shows that the vast majority of the conservative geochemical tracers used to fingerprint floodplain sediment exhibited concentrations that were intermediate between upstream sources (red dots in Fig. 1; see Electronic Supplementary Material, Table ES1). In many cases, geochemical tracers that are conservative over event timescales but nonconservative over sediment routing timescales (due to sediment transport, floodplain deposition, and resuspension processes) are measured as end-members in fingerprinting studies (black dots in Fig. 1; see Electronic Supplementary Material, Table ES1). This result is somewhat expected, given the role of floodplains in the sediment routing system. Statistical techniques can be used to identify which sediment source most closely matches the geochemical signature of a suspended sediment sample. However, if the tracers used are truly conservative over sediment routing timescales, such statistical approaches are unable to distinguish between in channel mixing of sediment directly eroded from upland sources versus mixing of sediment from various source areas and time periods via channel–floodplain exchange. This is problematic if a goal of the sediment fingerprinting is to distinguish bank/floodplain sources from other upland sources in the watershed.

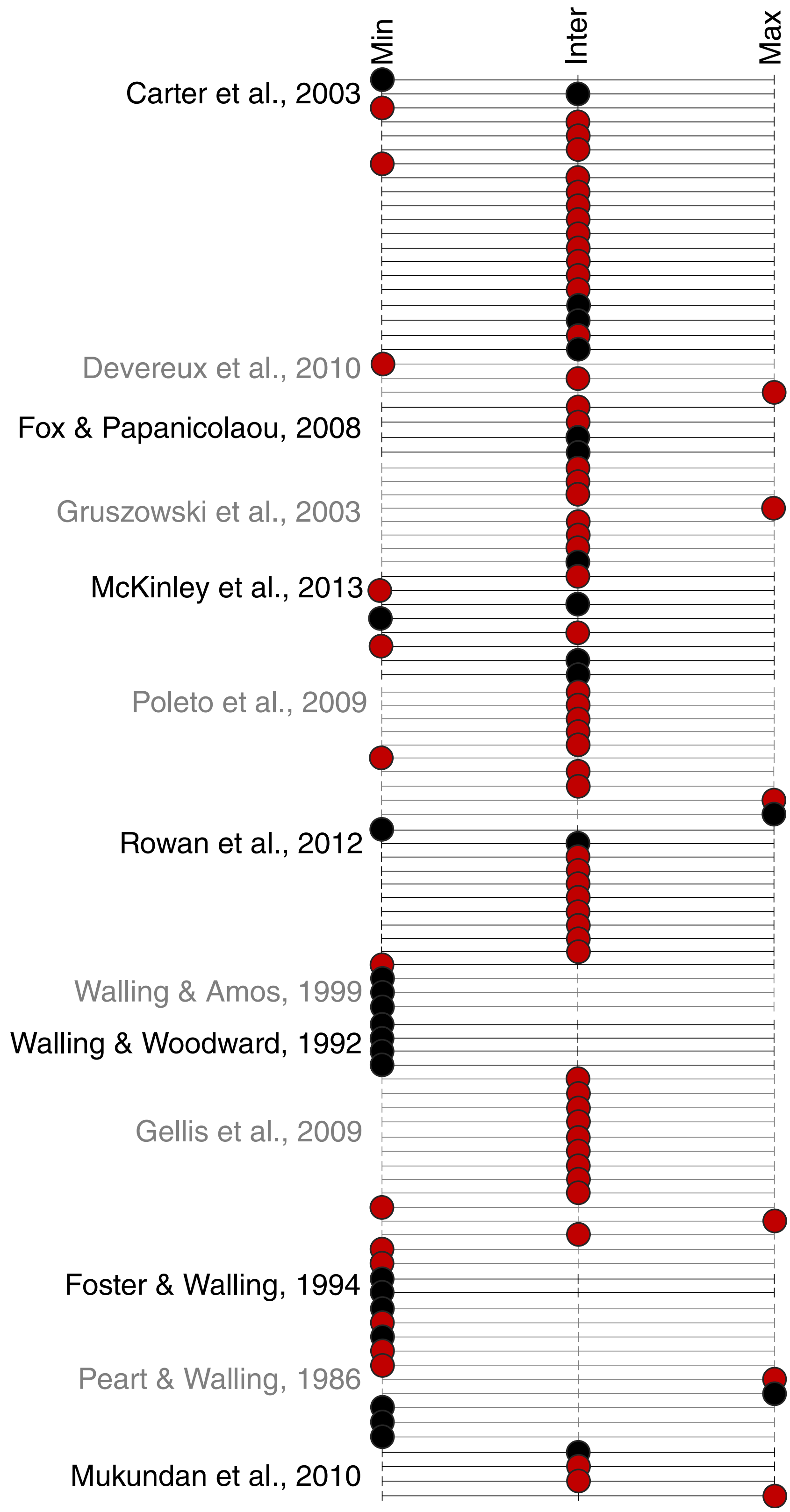
In contrast to the tendency for floodplains to mix conservative tracers associated with sediment derived from upstream sources, they also serve as long-term reservoirs where biological, chemical, and physical transformations may occur, thereby changing particular properties of the sediment stored within them. For example, the amount or type of organic matter associated with the sediment may change, or the concentration of a given element may change due to preferential uptake by biota or dissolution by chemical weathering, or, as discussed below, the concentration of a radionuclide (e.g., ^{7}Be, ^{210}Pb$_{ex}$, ^{137}Cs) may decrease due to natural decay over timescales much shorter than floodplain storage. Sediment fingerprinting conducted without consideration for processes that mix conservative tracers or alter the properties of nonconservative tracers may lead to erroneous interpretations, as discussed immediately below.

2.3 Using conservative and nonconservative tracers in sediment fingerprinting

A long-standing tenet of sediment fingerprinting has been the notion that investigators must verify that their tracers are conservative during transport over "event" timescales so that the fingerprinting properties can be directly compared between source area samples and target samples (e.g., suspended sediment: Motha et al. 2002; Walling 2013; Wilkinson et al. 2013). This prerequisite is important for discriminating direct sources of sediment, but may become problematic if one wants to quantify contributions from the floodplains themselves, due to the mixing effects of channel–floodplain exchange discussed above. So while we uphold the original impetus for demonstrating the conservative nature of tracers over "event" timescales, we posit that tracers exhibiting nonconservative behavior over sediment routing timescales (10^2–10^4 years) can also be immensely useful for sediment fingerprinting, provided that the nonconservative behavior is predictable and verifiable.

Fine sediment transported through a river network can experience a wide range of residence times, from days to millennia (Walling et al. 1998; Owens et al. 1999; Bonniwell et al. 1999; Skalak and Pizzuto 2010). Constraining the distribution of particle residence times for any given floodplain, let alone an entire channel–floodplain network, can be prohibitively costly and in many cases is simply not possible. However, for most single-thread, meandering rivers, it is reasonable to assume a bimodal distribution of residence times (Fig. 2) whereby most silt and clay particles are either transported out of the watershed quickly (days to a few years), or stored in a floodplain for a relatively long period of time (many decades to several millennia; Pizzuto et al. 2014). In such systems, measurement of both long- and short-lived radionuclide tracers associated with suspended sediment can provide insights into the proportion

Fig. 1 Results from 13 sediment fingerprinting studies that included floodplain/bank as a distinct sediment source. Studies are labeled on *left* and alternate in sets of *black* and *grey lines*. Number of lines under each study indicates the number of fingerprinting tracers used. Each line has been sorted from lowest to highest value. The location of the *dot* on each line indicates whether the floodplain/bank source was the minimum (*left*), maximum (*right*), or intermediate concentration relative to other source areas identified. *Red* and *black dots* are used for tracers that are expected to be conservative and nonconservative (N, C, ^{137}Cs, ^{210}Pb$_{ex}$, organic P, ^{7}Be, pyrophosphate iron, magnetic susceptibility), respectively, over sediment routing timescales. Original data available in the Electronic Supplementary Material, Table ES1



of sediment derived from different sources as well as the proportion exchanged between the channel and floodplain. The latter is made feasible because long-lived tracers are conservative over sediment routing timescales (i.e., reflect

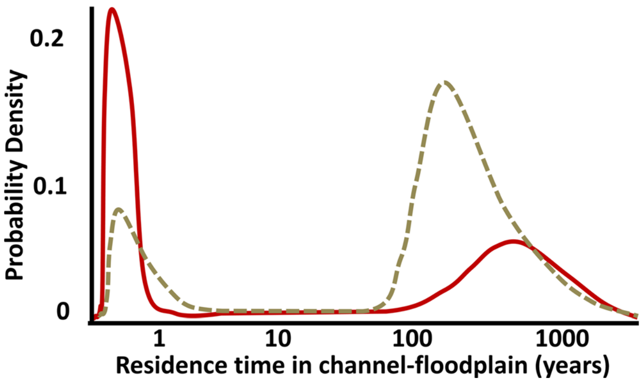


Fig. 2 Theoretical probability distributions for channel–floodplain residence time, expected to be bimodal for most single-thread meandering rivers. *Left peak* characterizes sediment that remains in channel. *Right peak* characterizes sediment that is stored/reworked from floodplain. Two different scenarios are depicted. *Red solid line* illustrates a case where most sediment remains in the channel. *Brown dashed line* represents a scenario with considerable channel–floodplain exchange, where a large proportion of the sediment is temporarily stored within the floodplain

the geochemical signature of the ultimate sediment source) and short-lived tracers decay during temporary storage, and thus gain a new geochemical signature if stored in the floodplain and reworked many decades or millennia later (Lauer and Willenbring 2010; Viparelli et al. 2013).

Conceptually, use of conservative and nonconservative tracers in fingerprinting simply requires that one understand the processes that impart the fingerprinting signature in source areas, the basic processes by which sediment is transported or stored within the landscape, and processes by which the fingerprinting signature is changed during transport or storage (Wallbrink and Murray 1993; Hancock et al. 2013). As a theoretical example, we consider a simple mass balance equilibrium (sediment mass in (Qs)=sediment mass out (Qs)), steady-state sediment routing system depicted in Fig. 3. We start with this simple, equilibrium system not because we believe it is directly representative of most real systems (many of which are likely out of equilibrium over decadal timescales), but rather because the assumptions, fluxes, and processes are easy to follow and yet the resulting sediment apportionment may be contrary to what some might expect.

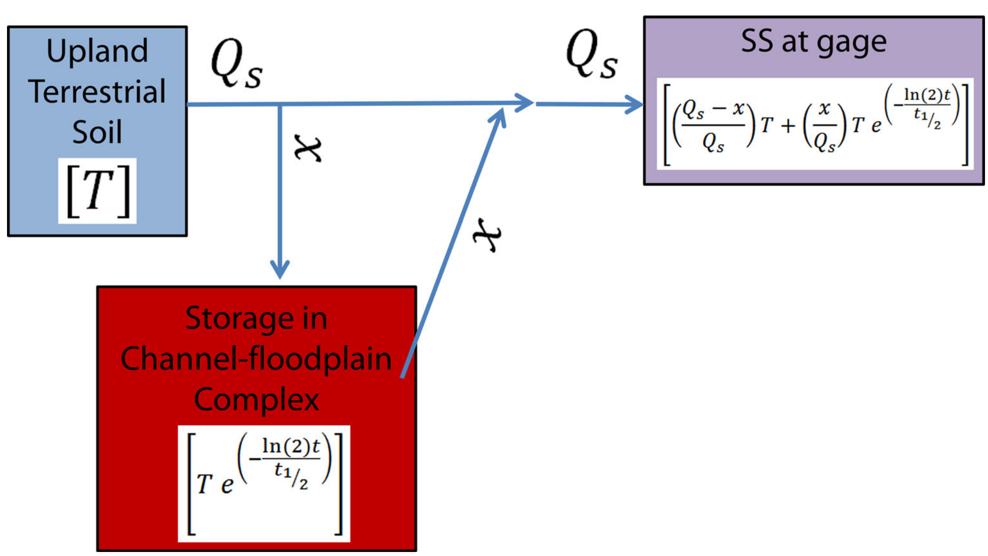
The conceptual model begins with an input (Q_s) of sediment derived from upland soils, which have a known and uniform tracer concentration ([T]). During transport downstream, some amount of sediment (x) is lost to floodplain storage. During floodplain storage, tracer concentration decreases according to a simple decay function (red box). However, note that under the equilibrium assumption floodplains cannot serve as a net source or sink for sediment, and therefore the mass deposited in the floodplain is equaled by the mass eroded from the floodplain (also shown as x). Thus, floodplains simply serve as a temporary reservoir where tracers associated with sediment can decay. Source apportionment from a suspended sediment sample collected at a given location (e.g., a gauge at the watershed outlet) is based on tracer concentration (defined by the equation in the purple box). The proportion of sediment interpreted to have come from the uplands using a simple unmixing model is described by Eq. 1:

$$\% \, Upland = \frac{\left[\left(\frac{Q_s - x}{Q_s}\right)T + \frac{x}{Q_s} T e^{\left(-\frac{\ln(2)t}{t_{12}}\right)}\right]}{[T]} \tag{1}$$

If floodplain residence time (t) is significantly longer than tracer half-life ($t_{1/2}$), the right term in the numerator of Eq. 1 goes to zero and sediment apportionment is driven by the amount of channel–floodplain exchange (x).

Below, we solve the model for two meteoric radionuclide tracers (e.g., meteoric ^{10}Be and ^{210}Pb$_{ex}$) associated with sediment for the simple scenario in which sediment is

Fig. 3 A model illustrates the importance of channel–floodplain exchange in interpreting geochemical fingerprinting results with radiogenic isotopes. For sediment tracers whose floodplain residence time (t) is much longer than the half-life ($t_{1/2}$), sediment apportionment based on tracer concentration measured in suspended sediment (*SS*) samples collected at a gauging station depends exclusively on the channel–floodplain exchange (x)



eroded from an upland soil and transported through a simple channel network with a floodplain component. This particular suite of tracers may be generalizable to many other systems and could be modeled in a more sophisticated and spatially distributed manner, as has been done by Lauer and Willenbring (2010) and Viparelli et al. (2013). Further, the conceptual model presented in Fig. 3 could be applied to any other suite of conservative and nonconservative tracers if the nonconservative behavior is quantifiable and verifiable. Challenges in applying the simple conceptual model presented in Fig. 3 include the fact that most channel–floodplain systems are not in a state of mass balance equilibrium, upland soil tracer concentrations are often not uniform, and there may be other geochemical transformations, grain size sorting, or additions of the tracer that must be accounted for. However, none of these complexities change the simple point for which this conceptual model was developed, namely to demonstrate that the interpretation of nonconservative tracer concentrations must be made with a firm understanding of the effect of temporary storage that occurs during sediment routing.

The ^{10}Be/^{210}Pb$_{ex}$ system has several advantages that facilitate its use for sediment fingerprinting to distinguish between terrestrial and channel–floodplain sediment sources. Both tracers are delivered to Earth's surface by rain and dry deposition, and adsorbed onto soil particles within the top ~10 and ~150 cm from the surface for ^{210}Pb$_{ex}$ and ^{10}Be, respectively (Noller 2000; Balco 2004; Graly et al. 2010). Delivery rates for both of these tracers are well constrained for many locations. The primary difference between these tracers is their disparate half-lives (22 years for ^{210}Pb$_{ex}$; 1,390,000 years for ^{10}Be). Because ^{10}Be is essentially conservative during sediment routing in many landscapes (Willenbring and von Blanckenburg 2010), it records the long-term (century+) erosional history of the landscape and can be used to identify locations that have experienced significant historical soil erosion (e.g., Stout et al. 2014). Sediment delivered directly via soil surface erosion exhibits high concentrations of both tracers. However, for sediment that experiences colluvial or floodplain storage, ^{10}Be concentration is essentially unaffected, but ^{210}Pb$_{ex}$ concentration decreases to below detection levels within ~75 years. Thus, any sediment resuspended from long-term floodplain storage would be expected to exhibit high ^{10}Be and low/nil ^{210}Pb$_{ex}$ concentrations. Assuming a bimodal distribution of fine sediment residence times in the channel–floodplain (Fig. 2, left peak remains in channel, right peak stored/resuspended from floodplain), the suspended sediment ^{210}Pb$_{ex}$ concentration depends on exchange between these two populations.

Figure 4 shows the results of the source apportionment unmixing model for ^{210}Pb$_{ex}$ over a range of floodplain residence times (0–400 years) and scenarios ranging from zero exchange to 100 % exchange between the channel and floodplain. Recall that floodplains cannot serve as a net source or

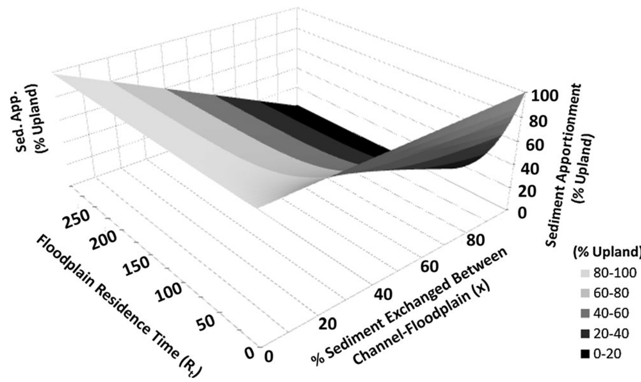


Fig. 4 Simple theoretical model demonstrating how sediment apportionment varies as a function of floodplain residence time (t) and channel–floodplain exchange rate (x) for a short-lived tracer (^{210}Pb$_{ex}$, $t_{1/2}$= 22.3 years) in a steady-state system wherein 100 % of sediment is initially derived from upland sources and floodplains cannot be a net source or sink. In contrast, the same model run for ^{10}Be would indicate ~100 % upland over this entire matrix of t and x values

sink for sediment in this system, thus any deviation from 100 % upland is erroneous. If floodplain residence time is 0 or the amount of channel–floodplain exchange is ~0, no bias occurs and apportionment based on suspended sediment tracer concentration correctly indicates 100 % upland sediment source. However, for floodplain residence times over 100 years (i.e., most real systems), the bias becomes a quasi-linear function of channel–floodplain exchange.

Many others have used ^{210}Pb$_{ex}$ to fingerprint sediment sources (Appleby and Oldfield 1983; He and Walling 1996; Kaste et al. 2007; Aalto and Dietrich 2005; Aalto and Nittrouer 2012; Smith et al. 2013). At a small watershed scale, this is a reasonable approach. However, as shown in Fig. 4, at larger watershed scales channel–floodplain exchange causes a bias in ^{210}Pb$_{ex}$ concentration and this bias can lead to erroneous interpretations for source apportionment. This potential bias has important real-world implications. For example, a water quality regulatory agency under the perception that 50 % of the sediment in our (equilibrium) river is derived from bank sources might implement an extensive (and expensive) bank protection program. In this (equilibrium) system, preventing erosion of cut banks would necessarily have the unintended effect of proportionally reducing deposition (maintaining equilibrium conditions). Thus, the mass of sediment moving through the reach would remain the same. No sediment reduction would be realized by the bank protection campaign. Under disequilibrium conditions or unsteady/nonuniform erosion and deposition, the model becomes more complicated (Lauer and Willenbring 2010; Viparelli et al. 2013), but the underlying potential for bias remains.

In contrast, decay of a long-lived tracer, such as meteoric ^{10}Be, is negligible over the range of floodplain residence times considered in this hypothetical system, or over the range of typical floodplain residence times in real river systems. Therefore, source apportionment would indicate 100 % upland for the ranges of x and floodplain residence times considered in Fig. 4. The ^{10}Be concentration may actually increase slightly above T if we were to consider additional delivery of our long-lived tracer to the channel and/or floodplain during storage (Wallbrink et al. 2002). Based on the assumed bimodal distribution of channel–floodplain residence times and negating other geochemical transformations or additions of tracer material (Fig. 2), the proportion of upland-derived suspended sediment that participates in channel–floodplain exchange can be approximated as the difference between the source apportionment estimates obtained using the two-end-member unmixing models for the long- and short-lived tracers independently (ΔU).

$$x \sim U_{10Be} - U_{210Pb} = \Delta U \qquad (2)$$

where x is the proportion of upland sediment that has spent many decades in floodplain storage, U_{10Be} and U_{210Pb} are the ^{10}Be and $^{210}Pb_{ex}$ source apportionment estimates, respectively, in terms of percent derived from upland (range 0–100 %). U_{210Pb} could be substituted with U_{137Cs} if ^{137}Cs is the short-lived radionuclide of interest. See additional explanations of Eqs. 1 and 2 in the Electronic Supplementary Material.

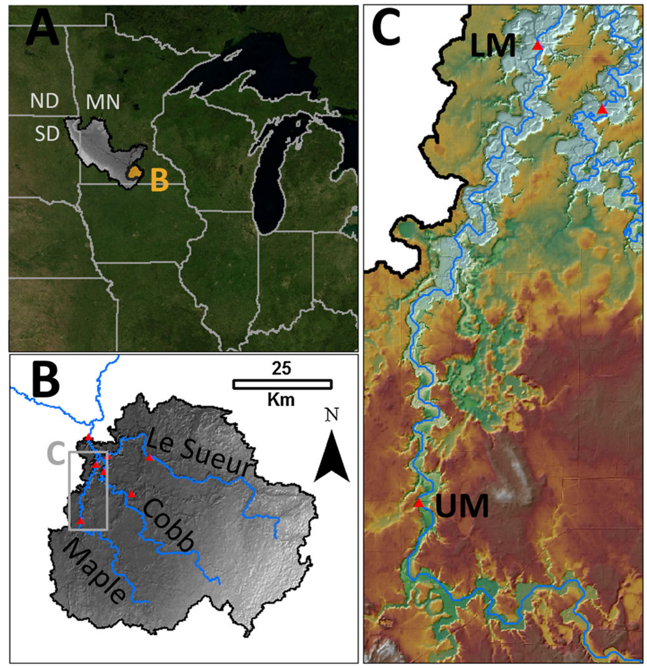


Fig. 5 Location of the Maple River, within the Le Sueur watershed (*orange* in **a**, enlarged in **b**), which is a tributary to the Minnesota River basin (*grey* in **a**), USA. *Red triangles* indicate locations of gauging stations distributed throughout the Le Sueur watershed, including the Upper Maple (*UM*) and Lower Maple (*LM*) gauges, overlain on a 3 m resolution lidar DEM in *panel C*

3 Application in the Maple River watershed

3.1 Study area

The Maple River is a tributary to the Le Sueur River in south-central Minnesota, USA (Fig. 5). The Le Sueur watershed provides a nearly unique natural experiment in which to study landscape evolution and sediment routing (Belmont et al. 2011; Gran et al. 2011, 2013). Approximately 13,400 years before present, base level for the Le Sueur River dropped approximately 70 m when Glacial Lake Agassiz catastrophically drained through, and incised, the proto-Minnesota River (Clayton and Moran 1982), to which the Le Sueur is a tributary. The knickpoint created at the junction between the Le Sueur and Minnesota River has propagated 40 km up through the Le Sueur network, causing rapid incision (3–5 m ky^{-1}) through a stack of interbedded tills and glaciofluvial sands (Belmont 2011). We refer to this rapidly incising reach that encompasses the lower portions of the Le Sueur and its two tributaries, the Big Cobb and Maple rivers, as the knick zone. The knick zone is characterized by relatively steep river channels (0.002 m m^{-1}) and rapidly eroding bluffs composed of fine-grained till. Above the knick zone, the landscape is remarkably flat with <5 m of relief per km^2. Beginning in the mid-19th century, Euro-American settlers began to convert what was a tall-grass prairie- and wetland-dominated landscape to farmland. Today, 75–80 % of the uplands are used for corn and soybean agriculture (Wilcock 2009).

The geologic and geomorphic history of the watershed has resulted in a fairly simple geochemical landscape comprised of three distinct sediment sources. Sediment derived from surface erosion on upland agricultural fields exhibits high concentrations of all three tracers. Sediment derived from bluffs, which are comprised of readily erodible till and therefore have very short exposure to the atmosphere, exhibit very low concentrations of all three tracers. Because of the disparate half-lives of our tracers, sediment stored in the floodplain for many decades contains little to no $^{210}Pb_{ex}$ and ^{137}Cs, while ^{10}Be acts as a conservative tracer, or becomes slightly enriched during floodplain storage due to additional atmospheric deposition over the uppermost 150 cm of the floodplain alluvium (see Electronic Supplementary Material, Tables ES2, ES3, ES4).

3.2 Methods and geochemical signatures of sources

Upland source area samples were collected from five sites, including cultivated and noncultivated land, within the top 5 cm of the soil profile. Upland meteoric ^{10}Be concentrations fall within a relatively narrow range ($2.02–2.91 \times 10^8$ atoms g^{-1}, see Electronic Supplementary Material, Table ES2). The small amount of variability in upland ^{10}Be concentrations is likely caused by small differences in local erosion, tillage history, and/or soil texture.

Samples collected from four bluff faces throughout the watershed exhibited remarkably consistent meteoric ^{10}Be concentrations (0.068–0.091×10^{8} atoms g^{-1}, see Electronic Supplementary Material, Table ES2). Therefore, we applied a simple two-end-member unmixing model to interpret ^{10}Be results for source apportionment (percent derived from upland vs. bluff), using averages from each of our sources to represent the end-members (uplands=2.48×10^{8} atoms g^{-1}, bluffs= 0.081×10^{8} atoms g^{-1}). Floodplain sediment ^{10}Be concentrations are reflective of the long-term average of upland vs. bluff contributions, which varies systematically in the downstream direction, as bluffs become increasingly dominant within the knick zone (see Electronic Supplementary Material, Table ES3). Additional meteoric ^{10}Be is delivered and adsorbed to floodplain sediment during storage (average fallout rate is on the order of 0.01×10^{8} atoms cm^{-2} years^{-1}, Willenbring and von Blanckenburg 2010). However, given the relatively short residence times of sediment in the floodplains (estimated 500 years based on floodplain width and meander migration rate; Belmont et al. 2011) this addition is likely small. Thus, we use ^{10}Be source apportionment as a maximum constraint on the percent derived from uplands.

Source fingerprints for ^{210}Pb$_{ex}$ and ^{137}Cs were determined by Schottler et al. (2010). Briefly, upland source fingerprints were obtained from a regional survey of sediment core samples from lakes draining upland areas, with no perennial fluvial inflows or outflows and sediment samples collected from edge of field runoff events. Average upland ^{210}Pb$_{ex}$ and ^{137}Cs activities were 66.6 (standard deviation (sd)=59.2) and 11.1 (sd=11.8) Bq kg^{-1}, respectively. Banks and bluffs were shown by Schottler et al. (2010) to contain no excess ^{210}Pb or ^{137}Cs because of their short exposure duration and geometric foreshortening, which reduces potential for deposition. Floodplain samples contain negligible ^{210}Pb$_{ex}$ and ^{137}Cs, consistent with our assumption that both tracers decay over much shorter time periods than the estimated floodplain residence time (see Electronic Supplementary Material, Table ES3). Ongoing delivery of ^{210}Pb$_{ex}$ via atmospheric deposition to the top 5 cm of floodplain alluvium is considered negligible compared with the 2–3-m thick bank that is eroded by the river via widening and meander migration. Lead-210 that is directly deposited in the river during storm events could potentially increase ^{210}Pb$_{ex}$ activity of suspended sediment, a potential violation of the assumption of conservation over “event” timescales, thereby potentially biasing sediment fingerprinting interpretations (Wallbrink et al. 2002; Owens et al. 2012). However, this problem is minimized in high sediment systems such as the Le Sueur, where there are many suspended sediment particles to adsorb the ^{210}Pb$_{ex}$ delivered in any given event.

Several other considerations are important to address with regard to the analysis of fingerprinting results in the Le Sueur watershed. Specific to this landscape, ravines, present within the lower watershed, complicate the unmixing model. These features are incipient tributary networks that connect the flat uplands with the incised knick zone. The geochemical signature of these features is indeterminate in our model because they exhibit ^{10}Be and ^{210}Pb$_{ex}$ concentrations between that of our end-member sources. However, previous work utilizing mass balance calculations and sediment gauging (Belmont et al. 2011) indicated that these features contribute a small proportion (~5 %) of the suspended sediment load, and therefore can be negated. More generally, comparison of multiple tracers requires measurements to be done on common source material. Preferably, source materials that are in transport would be sampled to reduce the differences in particle size and organic matter between mobilized sediment and parent material that may dilute or enrich tracer activity, though debate remains as to the conditions under which these factors may or may not be important (Smith and Blake 2014). Further, geochemical transformations during transport cannot necessarily be ignored as discussed extensively by Koiter et al. (2013), though we have no reason to believe that geochemical transformations significantly alter our tracer concentrations. Lastly, it is important to consider the possibility that tracers with such disparate half-lives may be influenced by erosional events from different time periods. For example, ^{10}Be may be depleted by significant erosional events that occurred over 100 years ago, while the current ^{210}Pb or ^{137}Cs inventory would not reflect this erosional event. While it has been determined that such an erosional event (stripping >10 cm of soil) has not occurred in the Le Sueur watershed (Belmont et al. 2011), such an event has been documented in the Root River watershed, complicating sediment apportionment in the study by Stout et al. (2014). Bearing these potential sources of error in mind, we discuss the reliability of our results and interpretations.

Suspended sediment samples were collected during storm events at the Upper and Lower Maple River gauges (UM and LM, respectively). Approximately 75 l of water was collected for each sample, which were allowed to settle 48–72 h and were concentrated by removing sediment-free water until the sample volume was <1 l. Samples were then freeze-dried, homogenized, and split for grain size analysis and measurement of ^{10}Be, ^{210}Pb$_{ex}$, and ^{137}Cs. Chemical processing and measurement of meteoric ^{10}Be by Accelerator Mass Spectrometry (AMS) was conducted by Purdue University Rare Isotope Measurement Laboratory (USA). Chemical processing and measurement of ^{210}Pb$_{ex}$ and ^{137}Cs was conducted by St. Croix Watershed Research Station, USA, by alpha and gamma spectroscopy.

4 Results and discussion

The hydrograph and hyetograph for the 2009 monitoring season (March 27–November 12) at the Lower Maple gauge (labeled LM in Fig. 5) are shown in Fig. 6a. Only one large rainfall–runoff event occurred during the year, on June 22–27. The portion of the annual hydrograph bounded by dashed lines is highlighted in Fig. 6b and c. Samples were collected at both gauging stations at each of the times indicated by triangles in Fig. 6b. Instantaneous total suspended sediment (TSS) concentrations and cumulative suspended sediment load (normalized by total event load) for each of the gauges are shown in Fig. 6c. During this event, a total of 1,350 and 2,170 Mg of suspended sediment was transported past the UM and LM gauges, respectively, representing 39 and 44 % of the total 2009 annual TSS load measured at each gauge. Notably, the 60 % increase in sediment loading occurs between the two gauges despite a mere 10 % increase in drainage area. Our samples cover the duration of the event that transported approximately 70 % of the event suspended sediment load.

Sediment fingerprinting results are shown in Fig. 7. Tracer concentrations are interpreted for source apportionment (shown as percent of sediment derived from soil erosion on upland agricultural fields) by deconvolving two-end-member unmixing models for each tracer independently. Because the Maple River is a net degradational system and therefore alluvial banks and till-bluffs both contribute sediment that is deficient in $^{210}Pb_{ex}$ and ^{137}Cs, we needed to adjust $^{210}Pb_{ex}$ and ^{137}Cs fingerprints to account for dilution caused by contributions of bluff sediment within the knick zone. To do this, we used ^{10}Be concentrations to initially apportion sediment between uplands and bluffs and then adjusted $^{210}Pb_{ex}$ and ^{137}Cs fingerprints accordingly using Eq. 3:

$$[T_i] = (U_{10Be}*[T_u]) + ((1-U_{10Be})*[T_B]) \quad (3)$$

where T_i is the adjusted fingerprint for $^{210}Pb_{ex}$ or ^{137}Cs after accounting for upland vs. bluff contributions, U_{10Be} is the fraction of sediment derived from upland sources as determined by ^{10}Be concentration (with 1-U_{10Be} being the fraction of sediment derived from bluffs), $[T_U]$ is $^{210}Pb_{ex}$ or ^{137}Cs concentration in upland-derived sediment, and $[T_B]$ is $^{210}Pb_{ex}$ or ^{137}Cs concentration in bluff-derived sediment (0 in this case). T_i is then used to compute U_{210Pb} or U_{137Cs} in Eq. 2.

Several observations can be made from Fig. 7 that support our simple but reasonable view of the system and provide insights into spatially integrated, temporally discrete sediment routing in this system. First, the unmixing model provides reasonable numbers that are consistent with our geomorphic understanding of the system. Specifically, above the UM gauge, upland agricultural fields and banks/floodplains are

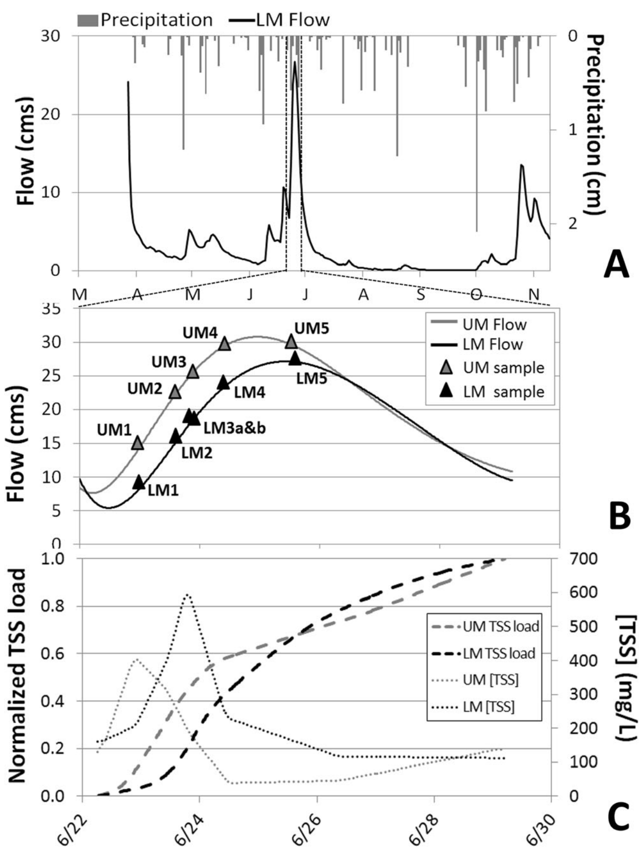


Fig. 6 Plot **a** shows the hydrograph and precipitation measured at the Lower Maple (*LM*) River gauge during the 2009 sampling season. Plot **b** is an inset of **a** showing the hydrographs and suspended sediment samples collected (*triangles*) throughout the June 22–29 flow event. Plot **c** shows instantaneous and cumulative suspended sediment concentrations over the course of the event

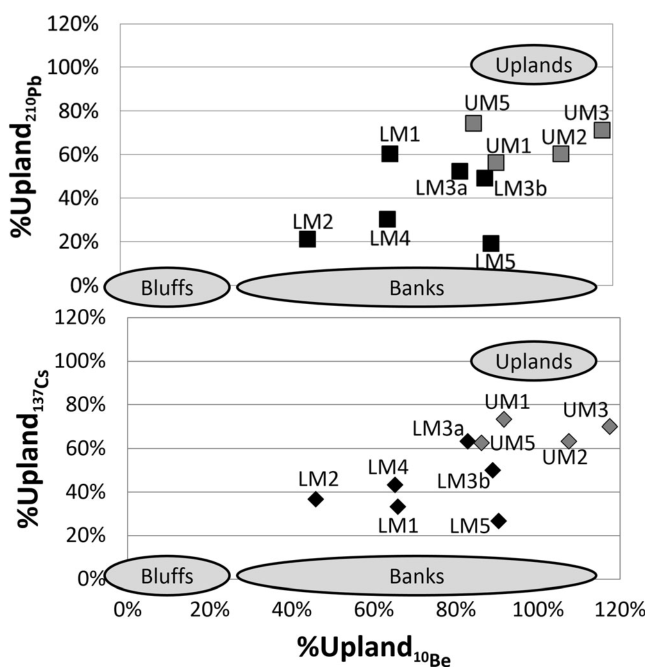


Fig. 7 Geochemical results interpreted for source apportionment as percent of sediment derived from upland for ^{210}Pb (*top panel*, *squares*) and ^{137}Cs (*bottom panel*, *diamonds*), both plotted against ^{10}Be results. Both combinations of tracers show a systematic increase in the relative contribution from nonupland sources from the Upper Maple (*grey*) to Lower Maple (*black*)

essentially the only source that can contribute sediment, aside from a few small bluffs. This fact is reflected in the ^{10}Be concentrations for suspended sediment collected at the UM gauge, which closely match concentrations measured in upland source areas. Two UM samples are interpreted as slightly exceeding 100 % upland, which could be caused by additional ^{10}Be delivery to floodplain alluvium during storage, slight differences in the grain sizes measured in source area and suspended sediment samples, and/or a slight underestimate of our upland source fingerprint. Between the UM and LM gauges, rapid Holocene incision has created many large bluffs that have been observed via air photo and terrestrial lidar analyses to be eroding rapidly (Belmont et al. 2011; Day et al. 2013a, b). The systematic decrease in both ^{10}Be and ^{210}Pb$_{ex}$ concentrations between the gauges is consistent with these observations.

As expected, the short-lived radionuclides exhibit systematically lower concentrations than ^{10}Be even upstream from the knick zone, indicating significant sediment exchange between the channel and floodplain. It is also important to note that ^{137}Cs provides slightly lower estimates of the percent of sediment derived from uplands than does ^{210}Pb$_{ex}$, possibly due to errors in comparing independently constrained upland fingerprints, direct deposition of ^{210}Pb$_{ex}$ to the river surface during the rainfall event, or due to transformations (e.g., desorption) that may deplete ^{137}Cs during fluvial transport (Parsons and Foster 2011). Nevertheless, the ^{210}Pb$_{ex}$ and ^{137}Cs results are quite consistent with one another at the coarse resolution of our analysis, especially when sediment loads are highest. Further, the systematic difference between ^{10}Be and the short-lived tracers confirms the caveat discussed earlier, namely, that short-lived tracers inherently underestimate upland contributions in large systems that have active channel–floodplain exchange processes causing storage and resuspension over multiple decades to millennia, as also shown theoretically in Fig. 4.

Disparity between the long- and short-lived radionuclide source apportionment estimates (ΔU) is a proxy measure of channel–floodplain exchange activity, assuming source fingerprints are accurately constrained. Figure 8 shows the disparity in source apportionment estimates (^{10}Be minus the short-lived tracer) over time, at each of the gauges. Note that ^{210}Pb$_{ex}$ and ^{137}Cs yield very similar results, each relative to ^{10}Be, for the time periods when sediment flux is high. The ΔU results suggest that an estimated 35–45 % of suspended sediment transported past the UM gauge spends a significant amount of time (>70 years) in storage within the channel–floodplain complex. During the period with high TSS load, approximately 15–35 % of suspended sediment transported past the LM gauge has spent time in long-term storage within the channel–floodplain complex.

Between the upper and lower gauges, the apparent reduction in the proportion of suspended sediment that has

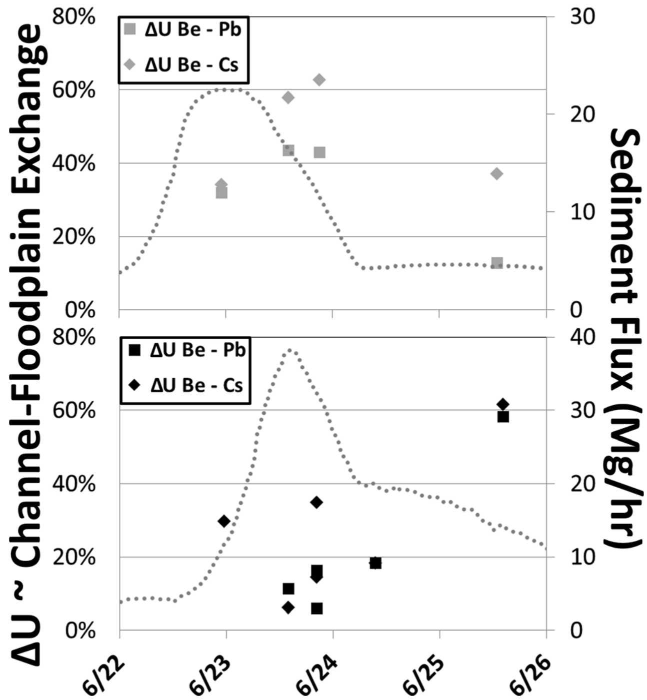


Fig. 8 The percentage of sediment that has participated in channel–floodplain exchange as estimated by ΔU for the Upper Maple (*top panel*, *grey*) and Lower Maple (*bottom panel*, *black*) sites over the course of the storm hydrograph. Sediment flux shown throughout the event as *dotted line*

experienced long-term storage may be the result of increased transport capacity and decreased floodplain storage within the steep knick zone (Belmont 2011). On the falling limb of the hydrograph, when bank erosion is expected to dominate (Simon et al. 2000), results at the upper and lower gauges suggest bank contributions are relatively high. These geochemical results are consistent with previous observations that meander migration rates, which primarily control floodplain reworking rates, are significantly faster within the knick zone, likely due to the increased loading of coarse sediment from bluffs (Belmont et al. 2011).

5 Conclusions

Measurement of geochemical tracers that exhibit conservative and nonconservative behavior over sediment routing timescales (e.g., long- and short-lived radionuclides) can provide spatially integrated, yet temporally discrete insights to constrain sediment sources and channel–floodplain exchange at the river network-scale. When utilizing nonconservative tracers in fingerprinting studies, the nonconservative behavior must be predictable and verifiable, accomplished here by modeling and measuring ^{210}Pb$_{ex}$ and ^{137}Cs decay during floodplain storage. Understanding the geomorphic processes that fractionate sediments with distinct geochemical signatures was essential for development and implementation of our sampling strategy and we encourage future fingerprinting studies to explain fingerprinting signatures with specific reference to geomorphic processes that cause differentiation of fingerprint properties between sources. In the Maple River watershed, we used three radiogenic tracers in combination with an unmixing model to observe a systematic shift in sediment sources from upland soil erosion to bluff and channel–floodplain erosion above and below a significant knick zone. Further, we demonstrated that differences in sediment apportionment from ^{10}Be, ^{210}Pb$_{ex}$, and ^{137}Cs measurements can be used to estimate the fraction of suspended sediment transported during the observed storm event that participates in channel–floodplain exchange.

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