Sources of alluvium in a coastal plain stream based on radionuclide signatures from the $^{238}$U and $^{232}$Th decay series

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1. Introduction

Discerning alluvial sources and their change over time or distance is a fundamental question in hydrology and geology, often critical in identifying impacts of human and natural perturbations on fluvial systems. Surfaces of upland interfluves and subsoils, sources of alluvium in the lower Loco Bayou basin, Texas, were distinguished using the isotope ratios $^{226}$Ra/$^{232}$Th, $^{226}$Ra/$^{230}$Th, and $^{230}$Th/$^{232}$Th. Channel alluvium indicates a transition from interfluve surface to subsoil sources during flood (subsoil ~34% to ~91%, over about 8 km) and bank-full stages (subsoil ~9% to ~74%, over about 12 km), with distance downstream. These results indicate strong coupling between hillslope and channel processes, reflecting land use change from forested to agricultural, concentrated in lower Loco Bayou. This methodology shows that sediment sources can be differentiated based upon landscape placement where lithologic contrast is absent. The geochemistry, long half-lives, and fractionation of $^{238}$U and $^{232}$Th decay series radionuclides during pedogenic and fluvial processes in humid climates suggest that these methods are applicable in a wide variety of fluvial systems.

INDEX TERMS: 1040 Geochemistry: Isotopic composition/chemistry; 1803 Hydrology: Anthropogenic effects; 1815 Hydrology: Erosion and sedimentation; 1824 Hydrology: Geomorphology (1625); KEYWORDS: natural radionuclides, fingerprinting, alluvium, source apportioning


1.1. Previous Research

Sources of alluvium to rivers have been assessed using a variety of tools including soil and sediment mineralogy [Phillips, 1992; Woodward et al., 1992], combinations of lithogenic and atmospheric radionuclides [Olley et al., 1993, 1997], heavy metals [Passmore and Macklin, 1994; Lecce and Pavlowsky, 2001], petrology [Schneiderman, 1995], and mineral magnetics [Caitcheon, 1998], and combinations of fallout radionuclides and geochemistry (C and N) [Nagle and Ritchie, 1999]. While this list is more illustrative rather than exhaustive, the common theme is that sediment source areas within a watershed have different physicochemical, mineralogical and other properties that may allow for an estimate of the relative contribution of these sources to stream sediments.

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morphology, including provenance determination of coastal sediments [Roberts and Plater, 1999], fluvial overbank sedimentation rates [Murray et al., 1990], and resolving fluvial sediment sources [Olley and Murray, 1994] and source fluxes [Olley et al., 1993]. These radionuclides have been used either solely [Olley, 1994; Olley et al., 1997] or together with fallout radionuclides [Olley et al., 1993; He and Owens, 1995] to address fluvial source and transport problems. Additional examples can be found in the compilation on this subject by Ivanovich and Harmon [1992].

1.2. Secular Disequilibrium

[5] Use of radionuclides as tags to fingerprint sediment sources to fluvial systems is based on the application of secular disequilibrium, where the daughter concentration at any time t is

\[ {N_d} = {N_p} + \left( {{N_d} - {N_p}} \right)e^{-\lambda_d t}, \]

where \( {N_d} \) and \( {N_p} \) are daughter concentrations at time t and \( t = 0 \), respectively, \( {N_p} \) is the initial parent concentration, \( \lambda_d \) is the daughter decay constant, and \( e \) is the Naperian constant (2.718) [Olley et al., 1997].

1.3. Thorium and Radium: Chemistry and Fractionation

[6] A principal assumption in tracer studies is that the marker(s) are conservative over a wide range of conditions, in this case, moving with sediment, while retaining the original source signature. Thorium is essentially insoluble (<1 \times 10^{-4} \text{ ppb}) [Kaufman, 1969], in surface water and does not undergo changes in oxidation state under normal Eh-pH conditions [Ivanovich and Harmon, 1982]. It has a +4 charge, tends to strongly sorb onto suspended particulate matter, and thus it is almost exclusively transported on particulate matter in rivers [Ivanovich and Harmon, 1992]. Radium exists in one oxidation state in nature, Ra\(^{2+}\). Its mobility is limited by coprecipitation in barite, calcite [Tanner, 1964], and hydrous oxides of Fe and Mn, adsorption (clays, quartz, and Fe (III) oxhydroxides [Riese, 1982; Ames et al., 1983]), and ion exchange. Radium in freshwater is strongly adsorbed to particulates.

[7] The relatively short-lived radionuclides such as \(^7\text{Be} \) (53.5 days), \(^{210}\text{Pb} \) (22 years), and \(^{137}\text{Cs} \) (30 years), which are often used to assess geomorphic processes, may decay before reorganization and transport by pedogenic processes. Longer-lived radionuclides in the \(^{238}\text{U} \) and \(^{232}\text{Th} \) decay series can be profoundly fractionated and re-organized within surface environments over geologic timescales, especially where chemical, physical and biological processes are intense. Significant fractionation of radium isotopes from other U-series nuclides during weathering has been documented [Rosholt, 1982; Muhs et al., 1990]. Michel [1984] showed recent additions of radium to surface soils based on radium excesses over \(^{230}\text{Th} \), without addressing the mechanism(s) responsible. Vegetation has a significant affinity for radium [Popova et al., 1964; Taskaev et al., 1977; Linsalata, 1994] and has been shown to be directly responsible for enrichment of radium in surface soils [Greenman et al., 1999]. In contrast, thorium depletion in surface soils has been observed in a variety of settings and locations, including soils in California [Hansen and Huntington, 1969], soils in the Mississippi River drainage basin [Rosholt et al., 1966], acidic soils observed by Gueniot [1983], and soils throughout the eastern United States [Greeman et al., 1999]. Greeman et al. [1999] determined from soil extractions that depletion of thorium in surface horizons results in part from depletion of radionuclide-rich Fe-oxides via some combination of colloidal particle transport and solubilization of this phase. An additional possibility is that thorium is leached from the surface as dissolved organic complexes, as suggested by Langmuir and Herman [1980].

1.4. Experimental Approach

[8] This research is focused on delineation of two primary alluvial source compartments, surfaces of upland interfluves, and subsoils. Specifically, the surface component coincides with the upper ~20 cm of the soil profile, where organic content and vascular plant activity is greatest. Subsoil then is that portion of the soil profile which is found from ~20 cm depth to bedrock. Bedrock, where exposed, can also serve as a source of sediment to channel alluvium and is therefore also considered a part of the “subsoil” component. When related to alluvium in transient storage, these compartments allow the dominant erosion processes, sheet wash versus gully development at the watershed scale to be distinguished. \(^{230}\text{Th} /^{232}\text{Th} \) values in soils and parent material have been shown to be equivalent [Olley and Murray, 1994], suggesting that these isotopes can be used to fingerprint fluvial sediment sources [Olley et al., 1997]. It is expected that this ratio may also be useful in distinguishing contributions from surfaces of upland interfluves versus subsoil compartments. \(^{226}\text{Ra} /^{232}\text{Th} \) has also been used as a source marker for soils and sediment [Murray et al., 1990, 1991; Olley et al., 1997]. \(^{226}\text{Ra} /^{230}\text{Th} \) is also utilized here as it should mirror \(^{226}\text{Ra} /^{232}\text{Th} \) results as the ratios of these elements are chemically equivalent, differing only in the half-life of the thorium nuclide and decay series origins for each set of ratios. Ra/Th ratios are anticipated to delineate surface and subsoil contributions, if surface radium enrichment coincides with thorium depletion.

[9] Given that these radionuclide signatures evolve in soils in response to external factors and in situ biogeochemical and pedogenic processes, and yet remain conservative over the relatively short timescales of erosion and fluvial transport, the radionuclide signatures of alluvial sediments reflect their original source component. Thus the primary objective of this case study is to test the use of isotope ratios of radium and thorium to fingerprint alluvial source compartment as opposed to source lithology or discrete spatial location in a developed and hydrologically managed basin.

[10] The specific objectives of this study are to (1) use \(^{238}\text{U} /^{232}\text{Th} \) series radionuclides to fingerprint sediment source compartments within the lower Loco Bayou basin, (2) examine the relative importance of sheet wash and gully development on sediment erosion as a function of space and hydrologic flow, and (3) examine geomorphic and anthropogenic causes for systematic change in sediment sources of channel alluvium as a function of distance.

2. Materials and Methods

2.1. Study Basin

[11] The field site for this case study is the lower section of the Loco Bayou watershed, below Lake Nacogdoches in Nacogdoches County, Texas (Figure 1). The entire basin...
has an area of 265 km², 9 km² of which is occupied by Lake Nacogdoches, with the lower section covering 37 km². Lake Nacogdoches provides the bulk of the drinking water for the city of Nacogdoches. When last surveyed in 1994, the lake had a storage capacity of 48,731,859 m³, representing a 6.6% reduction in capacity since completion in 1976 (see the List of Lake Surveys Completed by The Texas Water Development Board Volumetric Survey Program at http://www.twdb.state.tx.us/assistance/lakesurveys/compsurveys.htm).

Loco Bayou is in the Pineywoods region of the east Texas coastal plain. The climate is subtropical, with mean annual precipitation of 1200–1500 mm [National Oceanic and Atmospheric Administration, 2001]. While variable over the latter half of the 1990s because of consecutive droughts in the state, the distribution of precipitation is commonly bimodal in this region. The primary wet season here corresponds to January through April, often with a secondary peak in precipitation in early to late fall, with the summer months being the driest. The lower basin is predominantly forested (Figure 1); the main land use is agriculture, with minor portions of the basin utilized for residential areas, gravel mining, and forest clear-cuts, which vary spatially year to year. Total relief here is ~60 m; slopes prevail bordering the dam and valley walls in the north where forests dominate land cover. The southern half of the basin widens into a gently undulated floodplain, much of which has been cleared and utilized for agriculture.

The bedrock geology of the lower basin is dominated by the Eocene Weches Formation, a glauconitic marine sand...
This unit is exposed over ~0.5 km down reach of a culvert on Moccasin Creek (LSUPM2), a primary tributary of Loco Bayou that enters ~1 km south of the dam (Figure 2). Isolated patches of the up section, Eocene Sparta Sand, a limonitic coastal unit, can be found mainly in the headlands near the dam, but no outcrops were discovered. The channel itself is underlain by Quaternary alluvium, and the lowermost portion of the basin, near the confluence with the Angelina River, is bordered by Quaternary fluvial terrace deposits. The main channel can be characterized as degradational just south of the dam, where basal scour and tilted hardwoods indicate erosion and downcutting. Similar observations are made on the lower half of Moccasin Creek (Figure 2), south of the culvert. The Weches Formation is exposed here and incised; as one continues downstream, bedrock is quickly buried, but evidence of downcutting persists, including steep channel walls over 3 m high at base flow, tilted hardwoods, and large rotational slumps. Moving south from confluence with Moccasin Creek, the channel meanders through cohesive floodplain loams with some evidence of lateral scour and spatially constrained channel degradation most usually associated with cattle crossings and bridges. The iron-rich rocks are expressed directly at Loco Bayou in the soils, which are categorized into three associations: the upland ridge, red land belt loams of the Nacogdoches-Trawick, sandier, hill slope soils of the Cuthbert-Tenaha, and terrace loams of the Attoyac-Bernaldo-Besner associations [Dolezel, 1980].

2.2. Particle Size Variability

Differences in particle surface area have been shown to influence radionuclide adsorption [Megumi et al., 1982], which is particularly important in fluvial systems, where transport results in sorting of materials by particle size and density [Paola et al., 1992]. While an individual radionuclides activity concentration is a function of substrate surface area, daughter/parent ratios generally remain constant within analytical uncertainty [Murray et al., 1990, 1991; Olley et al., 1997]. This approach has been used when considering fallout radionuclides including $^{210}\text{Pb}$, $^{137}\text{Cs}$ and $^7\text{Be}$ [Bonniwell et al., 1999], and lithogenic radionuclides including radium and thorium [Olley et al., 1993, 1997] in fluvial systems. We have attempted to minimize the effects of particle size variations by both analyzing a specific range

Figure 2. Lower Loco Bayou watershed geology and sample locations. The bedrock geology is digitized from the 1:250,000 scale Palestine sheet [Shelby et al., 1968].
of particle sizes (<0.5 mm) and employing daughter/parent ratios.

2.3. Sampling

[15] Samples of channel alluvium, floodplain sediment and source area soils were collected from the near surface (0–2 cm). Source area samples were focused at actively eroding sites throughout the basin, concentrating on surfaces of upland interfluves and exposed subsoils on slopes (Figure 2). All samples were combined in the field, consisting of 8–10 subsamples collected over an approximately 10 m² area. Five samples of “fresh” bedrock were collected from the Weches Formation, where it outcrops in the channel of Moccasin Creek; these were combined and analyzed. Two soil cores were also collected. Alluvium is assumed to be derived from (1) soil erosion, (2) redistribution of sediments in storage, or, most likely, (3) a combination of processes 1 and 2, as the dam effectively limits the quantity of sediment available from the upper watershed [Phillips, 2001]. All source compartment and floodplain samples were collected from winter of 1999 through spring of 2001; sampling of alluvium was concentrated during flood (January – February 2001) and bank-full (April 2001) stages. Three water samples were collected at alluvial sampling sites during flood stage to assess radium in solution coinciding with annual maximum discharge and sediment transport capacity. Water samples were sequentially filtered in the field through 50, 20, 5, and 0.2 μm filters to remove suspended sediments and finally passed through two MnO₂ impregnated 0.5 μm filters to efficiently scavenge Ra in solution, as described by Baskaran et al. [1993]. MnO₂ filters were subsequently ashed and gamma counted.

2.4. Sample Processing and Radiochemistry

[16] Bulk samples were dried at 70°–80°C for 24 hours, then gently disaggregated with mortar and pestle and passed through 2 mm and 0.5 mm sieves. High-resolution gamma spectrometry was employed to resolve ²²⁸Ra (t₁/₂ = 5.75 years, via ²²⁸Ac Eγ = 911 keV) and ²²⁶Ra (t₁/₂ = 1602 years, via ²¹⁴Pb Eγ = 552 keV), using Canberra HPGe well detectors and a multichannel analyzer, model 747. Samples were contained in plastic test tubes (inner diameter 1.3 cm and height 9.4 cm) and sealed with epoxy for 20 days in storage, or, most likely, (3) a combination of processes 1 and 2, as the dam effectively limits the quantity of sediment available from the upper watershed [Phillips, 2001]. All source compartment and floodplain samples were collected from winter of 1999 through spring of 2001; sampling of alluvium was concentrated during flood (January – February 2001) and bank-full (April 2001) stages. Three water samples were collected at alluvial sampling sites during flood stage to assess radium in solution coinciding with annual maximum discharge and sediment transport capacity. Water samples were sequentially filtered in the field through 50, 20, 5, and 0.2 μm filters to remove suspended sediments and finally passed through two MnO₂ impregnated 0.5 μm filters to efficiently scavenge Ra in solution, as described by Baskaran et al. [1993]. MnO₂ filters were subsequently ashed and gamma counted.

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[17] Alpha spectrometry was employed to resolve ²³²Th (t₁/₂ = 1.39 × 10¹⁰ years), ²³⁰Th (t₁/₂ = 7.52 × 10⁹ years), and ²²⁶Ra (t₁/₂ = 1.91 years), using a Canberra alpha spectrometer, model 7404, mated to a Canberra multichannel analyzer, model 8224. Thorium samples were spiked with ²³⁹Th tracer and completely digested (HF, HCL, and HNO₃) over heat. The solution was passed through two sets of anion exchange columns to selectively isolate thorium isotopes, as described by Buessler et al. [1992]. The elution was acidified with H₂SO₄ and plated onto stainless steel planchets via sulfate electrodeposition prior to counting, according to methods described by Hallstadius [1984] and Buessler et al. [1992]. Chemical recoveries for thorium isotopes averaged >70%.

[18] Relative source contributions were determined using a simple mixing model [Olley et al., 1993];

\[ AX + BY = C, \]

where A and B are the Ra/Th values for subsoil and interfluval surface sources, respectively, C is the Ra/Th ratio of the output mix, and X and Y are the relative contributions of each, where \( X + Y = 1 \). This model does not consider nonlinearity in the ratios of the two independently varying activities [Faure, 1986; Olley et al., 1993; Kendall and McDonnell, 1998]. The deviation from linearity is small for the measured range of ²²⁶Ra, ²³⁰Th, and ²³²Th values and resultant ratios.

3. Results and Discussion

3.1. Sediment Sources

[19] Data for all samples are shown in Table 1, summary statistics and applied ratios are shown in Table 2, and end-member mixing results for alluvial samples are shown in Table 3. Core data (Figure 3) support Ra/Th fractionation in the soils of lower Loco Bayou, suggesting radium depletion coinciding with thorium enrichment with depth. Mean ²²⁶Ra/²³⁰Th and ²²⁶Ra/²³²Th signatures for interfluve surfaces versus subsoil compartments are statistically distinguishable, passing equal variance t tests, where \( \alpha = 0.05 \) (p₁ = 0.003 and p₂ = 0.001) (Figures 4a and 4b). While the difference between ²³⁰Th/²³²Th signatures for these compartments is not statistically significant, Figure 4c does graphically reflect Th enrichment at depth and depletion at the surface. Some signature overlap of these compartments is observed and was expected because of their contiguous relationship in the field.

[20] All three river water samples yielded no appreciable dissolved radium concentration during high discharge. A maximum value for ²²⁶Ra of 0.001 Bq/L was determined, which falls at the lower range of observed concentrations in U.S. rivers [Kraemer and Genereux, 1998]. Also, examining particulate ²²⁶Th/²²⁸Ra data shows that although relatively enriched with ²²⁸Th, the ratios are consistent within the mean error in alluvial samples (Figure 5). This ratio would change down-reach if recent adsorption of ²²⁸Ra (t₁/₂ = 5.75 years) had occurred, as ²²⁸Th (t₁/₂ = 1.91 years) would have insufficient time to grow into equilibrium with the recently adsorbed parent [Olley et al., 1997]. These data support radium immobility in freshwater and a closed system following removal from the soil profile.

[21] These data unanimously show that subsoil materials become the dominant source of alluvium with increasing distance down reach from the dam (Table 3). Both ²²⁶Ra/²³⁰Th and ²²⁶Ra/²³²Th (Figures 6a and 6b) are in reasonable agreement, with ²³⁰Th/²³²Th data supporting the same trend (Figure 6c), while exhibiting linearity reflective of equilibrium. Differences in alluvial signatures between flood and bank-full stages are also observed. While no clear trend is evident for floodplain samples, values for their ratios are constrained by source end-members (Table 2).

3.2. Human Agency and Source Flux: Cause and Effect?

[22] Coupling of channel and hillslope processes can be strong, where material is transferred from hillslope to
channel rapidly and continuously, resulting in a perturbation sensitive channel. Conversely, a buffered system has significant floodplain or valley fill deposits, protecting hillslopes from basal erosion, while isolating the river from hillslope sediment supply. Upon initial inspection, lower Loco Bayou would appear buffered based on field observations. Exceptions are not spatially extensive and can be found immediately south of the dam and of a highway culvert that bisects Moccasin Creek at LSUPM2 (Figure 2). At these locations, channel degradation, basal scour, and tilted hardwoods all indicate erosion and downcutting, typical observations immediately downgradient from a dam or culvert. Moving south from the dam, Loco Bayou quickly spreads out into a meandering floodplain stream moving through loamy soils bordered by gentle terrain.

Phillips [2001] and Phillips and Marion [2001] make similar observations, characterizing the floodplain of the lower basin as aggradational, with no apparent lack in sediment supply, estimating sedimentation by soil stratigraphic and dendrogeomorphic methods to be 44 mm/yr at the same location where the northernmost alluvial samples were collected for this research.

[21] Radionuclide data, however, suggest that hillslope and channel processes are strongly coupled. Radionuclide signatures indicate that alluvium is increasingly derived from subsoil with distance down reach for both flood and bank-full stages. The bulk of the northern half of the basin above the first set of alluvial samples is densely forested, and save for the areas immediately around the dam effluent and Moccasin Creek culvert, subsoil sediment sources are

Table 2. Radionuclide Activity Ranges and Mean Isotope Ratios in Loco Bayou Soil and Sediment Samples

<table>
<thead>
<tr>
<th>Compartments</th>
<th>n</th>
<th>230Th</th>
<th>228Th</th>
<th>226Ra</th>
<th>226Ra</th>
<th>232Th</th>
<th>226Ra/232Th</th>
<th>226Ra/228Th</th>
<th>226Ra/228Th</th>
<th>226Ra/228Th</th>
</tr>
</thead>
<tbody>
<tr>
<td>Interfluvial</td>
<td>11</td>
<td>22.9–77.2</td>
<td>20.8–41.0</td>
<td>15.9–41.1</td>
<td>20.6–45.1</td>
<td>0.82 ± 0.09</td>
<td>0.73 ± 0.09</td>
<td>0.85 ± 0.09</td>
<td>1.03 ± 0.11</td>
<td>1.50 ± 0.19</td>
</tr>
<tr>
<td>Subsoil</td>
<td>14</td>
<td>29.7–95.9</td>
<td>27.8–77.9</td>
<td>15.3–84.8</td>
<td>19.6–47.1</td>
<td>0.81 ± 0.09</td>
<td>0.75 ± 0.09</td>
<td>0.55 ± 0.05</td>
<td>0.69 ± 0.07</td>
<td>1.56 ± 0.17</td>
</tr>
<tr>
<td>Alluvium</td>
<td>6</td>
<td>24.7–70.5</td>
<td>21.2–69.1</td>
<td>21.7–41.7</td>
<td>20.6–47.6</td>
<td>0.90 ± 0.10</td>
<td>0.69 ± 0.08</td>
<td>0.71 ± 0.07</td>
<td>0.80 ± 0.07</td>
<td>1.54 ± 0.18</td>
</tr>
<tr>
<td>Floodplain</td>
<td>5</td>
<td>35.1–90.3</td>
<td>29.1–77.8</td>
<td>23.3–59.0</td>
<td>24.4–52.4</td>
<td>0.92 ± 0.10</td>
<td>0.83 ± 0.09</td>
<td>0.84 ± 0.08</td>
<td>0.93 ± 0.09</td>
<td>1.50 ± 0.19</td>
</tr>
</tbody>
</table>

*a* Radionuclide activity ranges are given in Bq/kg.
probably equal to or more likely of secondary importance relative to interfluvial surface supplies. This suggestion is supported by observations of consistent maximum values for interfluvial contribution at the northernmost sampling point during flood (~66%) and bank-full (~88%) stages. This implies an increase, moving north in lower Loco Bayou, in the importance of sheet and shallow rill erosion from uplands and a corresponding decrease in the importance of channel and gully erosion in the supply of channel alluvium. Land use significantly changes from dominantly forested to agricultural moving south from the northernmost alluvial sampling station. The majority of the lower basin is cleared land utilized for cattle grazing and agriculture (wheat, poultry, and dairy). While it would be an overgeneralization to characterize pastures and agriculturally developed floodplains as more prone to gully development and channel erosion than headwater forests, localized but significant subsoil contributions to alluvium are observed in this 37 km² basin. This is in spite of a generally aggrading floodplain due to the influence of stream flow management, the dam’s influence on base level and channel processes, and impacts of landscape changes on soil erosion such as deforestation, road and bridge construction, and cattle ranching. Radionuclide data (Figures 6a, 6b, and 6c) are in congruence with this scenario, depicting alluvium as increasingly subsurface derived with distance downstream. The subsoil component comprises nearly all alluvium present (~91%) during flood by ~15 km and most (~74%) during bank-full stage by ~19 km downstream from the dam.

Table 3. Ra/Th Mean and Discrete Values for Sources and Alluvium, Respectively

<table>
<thead>
<tr>
<th>Sample</th>
<th>A $^{226}$Ra/$^{232}$Th</th>
<th>B $^{226}$Ra/$^{230}$Th</th>
<th>$\Delta$Ra/$^{228}$Ra</th>
<th>Interfluvial A–B, %</th>
<th>Slopes A–B, %</th>
<th>Down-Reach Distance, km</th>
</tr>
</thead>
<tbody>
<tr>
<td>Interfluvial ($\eta_i$)</td>
<td>0.85 ± 0.09</td>
<td>1.03 ± 0.11</td>
<td>1.50 ± 0.19</td>
<td>...</td>
<td>...</td>
<td>...</td>
</tr>
<tr>
<td>Subsoil ($\eta_s$)</td>
<td>0.55 ± 0.05</td>
<td>0.69 ± 0.07</td>
<td>1.56 ± 0.17</td>
<td>...</td>
<td>...</td>
<td>...</td>
</tr>
<tr>
<td>LSCB3</td>
<td>0.75 ± 0.07</td>
<td>0.91 ± 0.09</td>
<td>1.29 ± 0.11</td>
<td>67–65</td>
<td>33–35</td>
<td>7.2</td>
</tr>
<tr>
<td>LSCB5</td>
<td>0.64 ± 0.05</td>
<td>0.75 ± 0.06</td>
<td>1.75 ± 0.16</td>
<td>31–18</td>
<td>69–82</td>
<td>14.7</td>
</tr>
<tr>
<td>LSCB6</td>
<td>0.60 ± 0.07</td>
<td>0.64 ± 0.04</td>
<td>1.73 ± 0.19</td>
<td>18–0</td>
<td>82–100</td>
<td>15.1</td>
</tr>
<tr>
<td>LSCB7</td>
<td>0.83 ± 0.09</td>
<td>0.97 ± 0.10</td>
<td>1.23 ± 0.19</td>
<td>94–82</td>
<td>6–12</td>
<td>7.2</td>
</tr>
<tr>
<td>LSCB8</td>
<td>0.75 ± 0.07</td>
<td>0.85 ± 0.07</td>
<td>1.66 ± 0.22</td>
<td>65–47</td>
<td>35–53</td>
<td>15.1</td>
</tr>
<tr>
<td>LSCB9</td>
<td>0.71 ± 0.07</td>
<td>0.69 ± 0.05</td>
<td>1.57 ± 0.22</td>
<td>53–0</td>
<td>47–100</td>
<td>19.2</td>
</tr>
</tbody>
</table>

*a* This is January–February 2001 sampling.

*b* This is April 2001 sampling.

The gradient in sediment source compartment contributions to lower Loco Bayou channel alluvium observed by this technique over stream length has important management aspects. In the north, where land cover is dominated by forests, the contribution of interfluvial surface sediments is more important, regardless of hydrologic stage. Although not entirely unexpected, this condition appears to mask the impact of localized channel degradation and scour associated with the dam effluent and Moccasin Creek tributary portions of the system, where subsoil contributions to the distribution of sources in the basin and their relationship to the channel under decreasing energy conditions.

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highly concentrated erosion rather than general upland sheet-and-rill erosion is critical. This interpretation is strengthened by observations of land use that directly result in localized channel and riparian zone degradation, cattle trails and crossings, which are concentrated in the southern portion of the basin. Such a relationship has been observed by others [Magilligan and McDowell, 1997; Lyons et al., 2000; Flenniken et al., 2001], particularly in small streams like Loco Bayou [Williamson et al., 1992], and relates well to the scenario offered by the radiochemical technique.

Observations at lower Loco Bayou may provide insight into landscape sensitivity and feedback in response to development. Land use change in the Upper Mississippi Valley triggered hydrologic responses that were transmitted nearly simultaneously to all watershed scales and flood-driven hydraulic adjustments in channel and floodplain morphologies contributed to feedback effects, causing long-term lag responses [Knox, 2001]. There, agricultural land use had escalated landscape sensitivity to such a degree that modern process rates provide a very distorted representation of process rates that occurred in the geologic past prior to human disturbance.

Research on effects of climate change on fluvial systems is sparse, compared to studies relating base level change to fluvial morphology and sediment transport capacity, and most of these studies focus on timescales greater than 10^7 years [Blum and Tornqvist, 2000]. Moreover, unique responses by fluvial systems to climate change, particularly over relatively short timescales (<100 years) are not well defined. Knox [1983] has shown that changes in global atmospheric circulation do influence both flood magnitudes and vegetation cover and that these two variables then interact to mutually effect fluvial morphology and sediment dynamics. While Texas has experienced several lasting and severe droughts during the 1990s, the short duration of climatic variability here, coupled with the “overprinting” on the system in the form of land utilization and hydrologic management, in particular, do not allow for definitive interpretations of climatic influence based on the radiochemical technique presented here. Techniques such as these, when combined with mineralogical or geochemical methods, may provide a valuable tool in the examination of...

**Figure 4.** Source signatures for interfluvial surface and subsoil source compartments (Bq/kg): (a) 226Ra/232-Th, (b) 226Ra/230-Th, and (c) 230-Th/232-Th.

**Figure 5.** Alluvial signatures within mean error for 228Th/228Ra for all alluvial samples.
observed $^{228}$Ra concentrations in soils in excess of its parent $^{232}$Th by as much as 30% and used this observation to estimate residence times of alluvial channel sediment [Olley et al., 1997]. At lower Loco Bayou, $^{228}$Ra was ubiquitously deficient relative to $^{232}$Th in all samples collected (Table 2). Several contrasts between Loco Bayou and the field sites for Olley et al. [1997] exist, including bedrock geology, climate, and topography. These factors no doubt influence the general mobility of $^{228}$Ra, the initial concentrations of both $^{228}$Ra and $^{232}$Th, as a function of parent rock lithology, and a host of biological and geochemical processes, which may be the cause for the observations of Olley [1994] and Olley et al. [1997].

4. Summary and Conclusions

[29] Throughout the lower Loco Bayou basin, samples of the Weches Formation, surfaces of upland interfluves and subsoil materials on slopes were collected from actively eroding sites. Two soil cores were collected and floodplain and channel alluvial sediments were sampled. Alluvial channel samples were collected during both flood and bank-full stages and stream water was filtered during flood stage to examine radium in solution. The isotope ratios $^{230}$Th/$^{232}$Th, $^{226}$Ra/$^{232}$Th, and $^{226}$Ra/$^{230}$Th were employed to (1) determine if fractionation in the soil profile created discrete signatures for different spatial sediment source compartments, (2) allow for modeling of channel alluvium sediment sources and their change over distance or with hydrologic setting, and (3) relate results to coupling of hillslope and channel processes and land use in lower Loco Bayou.

[30] On the basis of the U/Th series radionuclide analyses of soils and sediments from lower Loco Bayou, Texas, we conclude the following.

1. The $^{226}$Ra/$^{232}$Th and $^{226}$Ra/$^{230}$Th data for interfluvial surfaces and subsoils produced discrete signatures, allowing for a simple mixing model to be applied to resolve relative contributions of each compartment to alluvial sediments. The $^{230}$Th/$^{232}$Th data graphically support this distinction.

2. While exhibiting no clear spatial or temporal trends, floodplain sample ratios are constrained by soil compartment end-members.

3. Subsoil derived alluvial sediment became dominant with increasing distance downstream during both flood and bank-full stages. Separation of alluvial sediments between flood and bank-full stages indicate a more rapid and total dominance of subsoil sources over downstream length during flood and more gradual, less complete dominance during bank-full stage. This is interpreted as indicating strong coupling between hillslope and channel process in lower Loco Bayou, directly reflecting the impacts of human agency.

4. No excess of $^{228}$Ra over its parent $^{232}$Th was observed in soils and sediments, preventing use of $^{228}$Ra/$^{232}$Th to estimate residence times of alluvium at lower Loco Bayou.

5. Agricultural development, while influencing alluvial sources and transport dynamics at lower Loco Bayou, may also be providing for a strongly coupled fluvial system, wherein landscape sensitivity to extrinsic forcing is enhanced. These relations are fundamental to our understanding of human influences on fluvial systems at any scale and warrant further investigation.

Figure 6. Loco Bayou alluvial signatures for January–February (LSCB 3, 5, and 6) and April (LSCB 7, 8, and 9) of 2001 (Bq/kg): (a) $^{226}$Ra/$^{232}$Th, (b) $^{226}$Ra/$^{230}$Th, and (c) $^{230}$Th/$^{232}$Th. Arrows denote increasing downstream distance.
6. An important contribution to watershed-scale sediment transport modeling described herein is the ability to recognize and quantify sediment sources to the river not only by source rock lithology and its characteristic isotope signatures, where present, but also the ability to do the same for sediment based on its location on the landscape, whether lithologic contrast in isotopic signatures is or is not the case.

7. The ubiquity, chemical conservancy, long half-lives, ease of modeling, and wide climatic and geographic range over which fractionation of these primordial radionuclides in soils is expected represents an important tool in understanding fluvial processes. Additional research focusing on fractionation processes as related to both natural and anthropogenic factors and testing the applicability of these methods in larger, more geologically diverse watersheds is needed.

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