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Fingerprinting the sources of suspended sediment delivery to a large municipal drinking water reservoir: Falls Lake, Neuse River, North Carolina, USA

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Abstract
Purpose We employ a geochemical-fingerprinting approach to estimate the source of suspended sediments collected from tributaries entering Falls Lake, a 50-km^2 drinking water reservoir on the Neuse River, North Carolina, USA. Many of the major tributaries to the lake are on North Carolina's 303(d) list for impaired streams, and in 2008, the lake was added to that list because of high values of turbidity, likely sourced from tributary streams.
Materials and methods Suspended sediments were collected from four streams with a time-integrated sampler during high-flow events. In addition, composite sediment samples representing potential sources were collected from stream banks, forests, pastures, construction sites, dirt and paved roads, and road cuts within tributary basins. Radiocarbon dating and magnetic susceptibility measurements were used to determine the origin of stream bank alluvial deposits. Sediment samples were analyzed for the concentrations of 55 elements and two radionuclides in order to identify tracers capable of distinguishing between potential sediment sources. The relative sediment source contributions were determined by applying a Monte Carlo simulation that parameterized the geochemical tracer data in a mixing model.
Results and discussion Radiocarbon and magnetic susceptibility measurements confirmed the presence of "legacy" sediment in the Ellerbe and New Light Creek valley bottoms. Mixing model results demonstrate that stream bank erosion is the largest contributor to the suspended sediment load in New Light Creek (62%), Ellerbe Creek (58%), and Little Lick Creek (33%), and is the second largest contributor in Lick Creek (27%) behind construction sites (43%).
Conclusions We find that stream bank erosion is the largest nonpoint source contributor to the suspended sediment load in three of the four catchments and is therefore a significant source of turbidity in Falls Lake. The presence of legacy sediment appears to coincide with increased contributions from stream bank erosion in Ellerbe and New Light creeks. Active construction sites and timber harvesting were also significant sources of suspended sediment. Water quality mitigation efforts need to consider nonpoint-source contributions from stream bank erosion of valley bottom sediments aggraded after European settlement.

Keywords Atlantic Piedmont · Nonpoint-source pollutant · Total maximum daily load · Total suspended sediments · Radiocarbon · Magnetic susceptibility

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1 Introduction

1.1 Excess suspended sediments in surface waters

Suspended sediments in surface waters are the most common nonpoint-source pollutant in streams and reservoirs within the USA (US Environmental Protection Agency 2012). These sediments degrade waterways by limiting light penetration through the water column during high-flow events, leading to stressed aquatic biota (Henley et al. 2000), the burial of aquatic communities after high-flow events (Lisle 1989; Wood and Armitage 1997), and transport of nutrients and other contaminants adsorbed to the suspended particles (Delfino 1977; Lau et al. 1989). High suspended sediment loads have also been responsible for reductions in the operational capacity of municipal water supply facilities relying on surface water sources (Morris and Fan 1997). Falls Lake is a 50-km^2 reservoir on North Carolina's Neuse River that is the primary source of drinking water for nearly 500,000 people (Fig. 1). The lake and associated water treatment plant are impaired by high levels of turbidity (North Carolina Department of the Environment and Natural Resources 2012). The focus of this paper is upon quantifying the primary sources of suspended sediment transported into Falls Lake in order to help guide future surface water sediment reduction efforts for turbidity-impaired streams.

The 1972 Clean Water Act requires that states develop a list of impaired waters under their jurisdiction and calculate Total Maximum Daily Loads (TMDLs), or the maximum amount of a pollutant that a body of water can receive and still meet water quality standards, for all impaired waters whose impairment is caused by a particular pollutant. TMDLs are effective at assigning waste load allocations to point-source pollutants; while in comparison, they are often difficult to construct for nonpoint-source pollutants due to their diffusive nature (Chen et al. 1999). One technique useful in the source-area discrimination of nonpoint-source pollutants is sediment fingerprinting using geochemical analyses that may include radionuclides (e.g., Peart and Walling 1986; Mukundan et al. 2012; Mckinley et al. 2013).

1.2 Agricultural history and its impact on modern-day water quality

Water quality issues attributed to soil erosion throughout the Atlantic Piedmont physiographic province are a relatively recent occurrence. Early European records describing southern Piedmont streams anecdotally suggest that soil erosion rates were minimal compared to the levels they reached in the following centuries (Trimble 1974, and references therein). Early settlers wrote in detail about how clear these streams were during both normal and high flow conditions (e.g., Bartram 1791). It was not until the widespread exploitation of Piedmont soils for cash crops (e.g., tobacco, cotton, and

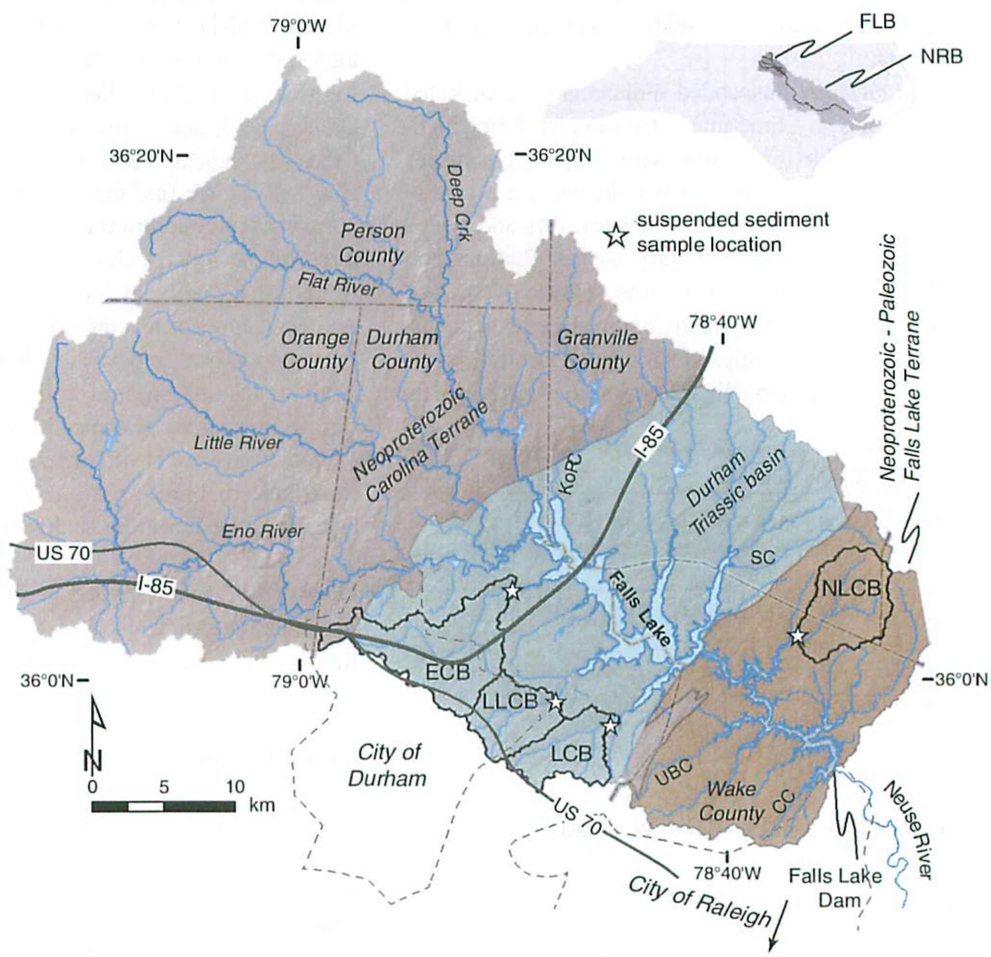

Fig. 1 Shaded relief map of the Falls Lake basin (*FLB*) that comprises the upper Neuse River basin (*NRB*; see *inset map* of North Carolina, USA). The upper half of Falls Lake is underlain by sedimentary and igneous rocks of the Triassic Durham basin, which are inset into Neoproterozoic metamorphic rocks of the Carolina and Raleigh terranes. Stream suspended sediment samples (*stars*) were collected from four Falls Lake tributary basins (Ellerbe Creek—*ECB*, Little Lick Creek—*LLCB*, Lick Creek—*LCB*, and New Light Creek—*NLCB*). The following additional tributary streams are identified: Knap of Reeds (*KoRC*), Smith (*SC*), Upper Barton (*UBC*), and Cedar (*CC*) creeks

corn) that erosive land use changed the morphology of valley bottoms, contributing to the impairment of surface waters observed today.

During the European-American colonial era, land was abundant enough that when the soil productivity of one field was exhausted it was abandoned and new land was cleared and cultivated in its place (e.g., Cathey 1951). Poor soil-conservation methods continued into the 1930s, leading to an average vertical soil loss of 15 cm for piedmont uplands across North Carolina (Trimble 1975). Most of the soil eroded from these uplands during colonial and post-colonial times is still in storage in Piedmont valley bottoms (e.g., Meade 1982; Wegmann et al. 2012). For example, Jackson et al. (2005) calculated that it would take between 6 and 10 millennia for a Georgia Piedmont stream to remove the valley bottom sediment aggraded during the cotton-farming era (AD 1820 to 1930) at current export rates.

1.3 Post-European valley bottom sediment accumulation

In the late 1600s, early settlers started building dams across streams in eastern North America to support milling operations, and, by 1840, more than 65,000 mills and associated dams existed along these streams (Walter and Merritts 2008). These dams impounded large volumes of sediment transported from upstream, turning many streams into aggradational sediment sinks (Wohl and Merritts 2007). The subsequent natural or purposeful breaching of mill–dams leads to a rapid drop in local base level followed by vertical stream incision and headcutting, which is then followed by slow lateral channel migration and widening (Doyle et al. 2002; Merritts et al. 2011). Though a substantial amount of sediment is transported downstream shortly after dam removal, the majority of the impounded sediment aggraded on relict floodplains is removed over time during subsequent high stream discharge events. Large volumes of valley bottom aggraded legacy sediment is documented for North Carolina Piedmont streams, even in the absence of mill–dam construction, and thus, it is hypothesized to be a nearly ubiquitous contributing source to the Total Suspended Sediment (TSS) concentration of modern streams (Phillips 1992; Wegmann et al. 2012). To understand how these erodible valley bottom sediments, and stream bank sediments specifically, are contributing to the turbidity of Falls Lake, a sediment fingerprinting study was conducted within four of the lake's sub-basins. Our hypothesis predicts that the erosion of post-European settlement sediments, now exposed along stream banks, is a significant, if not primary, source of current suspended sediment in the tributary basins, and by extension, into Falls Lake.

1.4 Sediment fingerprinting

Sediment fingerprinting has been developed as a method for determining the percent contribution of sediment from a particular physiographic region (e.g., Devereux et al. 2010), geologic area (e.g., Collins et al. 1998), soil type (e.g., Walling 2005), or land use type (e.g., Walling et al. 1993; Collins et al. 1997; Walling 2005; Gellis and Landwehr 2006; Devereux et al. 2010; Mukundan et al. 2010) to the TSS load of a stream. The method is performed by: (a) identifying potential sediment sources and collecting representative samples of those sources, (b) identifying unique tracers from each sediment source, (c) collecting the unique fingerprint signature of suspended sediment samples, (d) accounting for sediment and tracer fate, and (e) utilizing a mixing model to assign relative source contributions (Davis and Fox 2009). Researchers have used this technique to investigate both the cause and source of high suspended sediment loads impacting streams worldwide, and recently have applied the technique in aid of TMDL programs in the United States for streams directly and indirectly impaired by turbidity and TSS (e.g., Mukundan et al. 2010).

Researchers interested in determining the sources of fine sediment (≤63 μm; silt and clay) introduced to local reservoirs, coastal estuaries and large rivers in the mid-Atlantic region of the USA have often suspected upland soil erosion as a significant source (e.g., Gellis et al. 2009), and thus have turned to sediment fingerprinting as a means of ascribing and quantifying the relative source-area contributions. For example, Devereux et al. (2010) reported that 61% of suspended sediment collected within the northeast branch of Maryland's Anacostia River, a primarily urban drainage basin spanning both the Piedmont and Coastal Plain physiographic provinces, originated from the Piedmont and that 58% of this sediment was eroded from stream banks during flashy discharge events enhanced by the high density of impervious surfaces. Conversely, in rural Pennsylvania, Gellis et al. (2009) found that croplands were the primary source of suspended sediment within the Little Conestoga Creek drainage basin. In Georgia, Mukundan et al. (2010) determined that ~60% of the suspended sediment in the North Fork River was attributable to lateral erosion of stream banks now incised into historically aggraded (legacy) valley bottom sediments. Both current and historical land use activities have been implicated as important controls on the primary source of suspended sediment in this region. Following these examples, we predicted that stream bank erosion of historically aggraded sediments would also be a significant source in streams draining into the Falls Lake reservoir.

2 Materials and methods

2.1 Study area

The upper Neuse River was dammed in 1978 for water supply, flood control, and recreational purposes (US Army Corps of Engineers 1981). The 2,000-km^2 Falls Lake catchment drains parts of six counties (Fig. 1). The reservoir begins at the confluence of the Eno, Flat and Little Rivers northeast of the city of Durham and extends for 25 km downstream (Fig. 1). The lake's catchment encompasses three distinct geologic provinces; the Neoproterozoic-to-Paleozoic Carolina and Falls Lake Terranes, and the Durham Triassic rift basin (Fig. 1). We employ a sediment fingerprinting approach to determine the sources of suspended sediments collected from three impaired drainage basins on the south side of the lake (Ellerbe, Little Lick, and Lick Creeks), and one non-impaired north-side basin (New Light Creek; Fig. 1). The four study basins occupy 7.5% (150 km^2) of the reservoir's total catchment area. Most of the Ellerbe and western portion of the Little Lick Creek catchments are dominated by urban lands associated with the city of Durham, while the Lick Creek basin is primarily forested (Table 1). These three south-side basins are underlain almost entirely by sedimentary rocks and diabase sills of the Durham basin (North Carolina Geological Survey 1985; Electronic Supplementary Material, Table 1). The dominant upland soil of these three drainage basins is the White Store series (Electronic Supplementary Material, Table 1), which is characterized by a thick, smectitic B horizon with a high shrink–swell and erosion potential (Daniels et al. 1999). To our knowledge, there are no records of historic mill–dams located within any of the south-side basins; although stream bank stratigraphy along at least one section of Ellerbe Creek suggests otherwise (see Section 3.1). Ellerbe Creek is on the North Carolina 303(d) list for both poor biological integrity and for high levels of zinc, Little Lick Creek is listed for high levels of turbidity and low dissolved oxygen, and Lick Creek is listed for having fair biological integrity of benthic communities (North Carolina Department of the Environment and Natural Resources 2012).

New Light Creek flows southwest into Falls Lake from piedmont uplands. The basin is underlain by metasedimentary rocks (Fig. 1 and Electronic Supplementary Material, Table 1; Wylie 1984), and the dominant upland soil is the Cecil series. This soil is characterized by a thick, red (2.5 YR 4/6–5/8), and kaolinitic Bt horizon with pedogenic clay accumulation to a depth of ~2 m (Daniels et al. 1999). Surface erosion of Cecil soils is moderate-to-severe following exposure (Cawthorn 1970). Due to extensive prior erosion of the upper soil horizons the subsoil (Bt horizon) is now exposed at the surface across much of the study area (e.g., Trimble 1974; Phillips 1993). The New Light catchment is largely undeveloped (Table 1). New Light Creek is not listed on the North Carolina 303(d) list. It is the only basin in this study to contain a historic record of mill–dams (Bevers 1871).

Table 1 Land use for the four study catchments

	Land use (%)					
	Urban	Forest	Herbaceous	Pasture	Row crop	Valley bottom
Ellerbe Creek	74	16	3	2	0	4
Little Lick Creek	60	30	5	4	0	1
Lick Creek	13	60	13	7	1	6
New Light Creek	5	69	8	16	0	2

Reported values are rounded to the nearest percent

2.2 Water quality impairment in Falls Lake

The main water quality stressors within the Falls Lake basin are nutrient and sediment loading, high chlorophyll-*a* and fecal coliform levels, low dissolved oxygen, and habitat degradation (Deamer 2009). Pollutant sources such as agricultural field runoff, existing and new urban development, and point-source dischargers (e.g., sewage treatment, light industrial, and contained animal feeding operations) are typically blamed for these surface water stressors (Deamer 2009). Turbidity and chlorophyll-*a* levels within the upper section of Falls Lake, from the confluence of the Eno and Flat Rivers to the I-85 bridge (Fig. 1), often exceed the water quality standards of 25 Nephelometric Turbidity Units (NTU) and 40 μg l^{-1} chlorophyll-*a*. As a result, the reservoir was placed on North Carolina's 303(d) list for impaired and threatened waters in 2008. Chlorophyll-*a* was removed as a Falls Lake pollutant in 2012, making turbidity the existing lake pollutant (North Carolina Department of the Environment and Natural Resources 2012).

2.3 Sample collection

Land use was classified from the 2006 national land cover dataset (Fry et al. 2011) for each study basin (Table 1). We used 2010 aerial photographs to identify recent surficial erosion (i.e., erosional scars, rills, and gullies), for example from timber harvest sites, pastures, and dirt roads with close proximity to surface waters and the potential to contribute to stream suspended sediment loads. Web-based monthly construction reports were regularly checked to locate active sites within the Ellerbe, Lick, and Little Lick basins. Each trunk channel was surveyed to determine if post-European settlement legacy sediments were present in valley bottoms and exposed along stream banks.

A minimum of three sediment source samples were collected for each type between July 2011 and February 2012 (Fig. 2 and Electronic Supplementary Material, Table 2). Forest, pasture, dirt road, and construction site sediment samples were collected from the top 2 cm of the ground surface. Dirt road and construction samples were collected in extensive rill-erosion areas. Construction site samples were collected only in places of recent topsoil removal where the subsurface was exposed to erosion. Steep stream bank and road cut sites

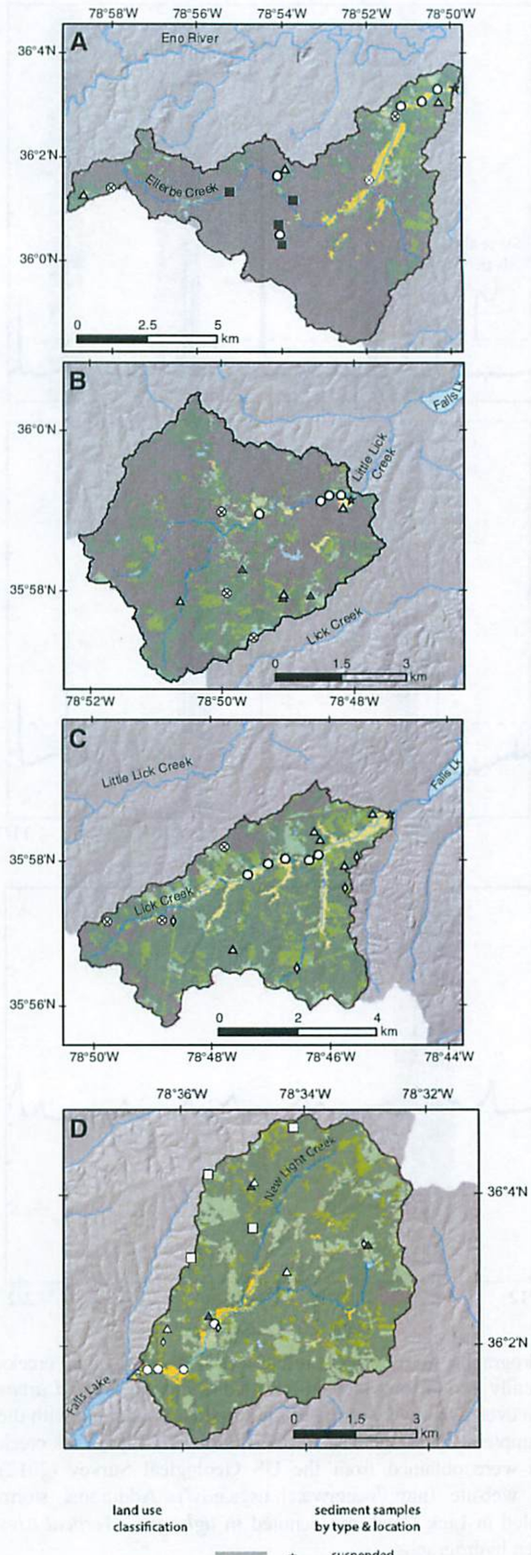

Fig. 2 Shaded relief image overlain by the 2006 National land cover assessment map for **a** Ellerbe, **b** Little Lick, **c** Lick, and **d** New Light creek basins. *Different shaped* and *shaded symbols* denote source and suspended sediment sampling locations

were cleaned of surficial materials before sampling. Paved road source samples were collected near storm drains.

Suspended sediment samples were collected near the confluence of each trunk channel with Falls Lake (Figs. 1 and 2 and Electronic Supplementary Material, Table 2), with the exception of Little Lick Creek, where samples were collected 3 km above the confluence in order to avoid flooding from a downstream impoundment (Fig. 2b). All samples were collected during high flow events (Fig. 3) with an integrated suspended sediment sampler designed specifically for fingerprinting studies modified from the design of Phillips et al. (2000; see Electronic Supplementary Material, Fig. 1). The sampler utilizes a polypropylene dewatering bag to collect and integrate the ambient suspended sediment (Voli 2012). As a result, each high discharge event can be sampled individually in its entirety, collecting a large enough sediment volume for multiple geochemical analyses. Initially, the bag retains >80-mesh (~180 μm) particles, but as the pores of the bag fill with sediment and as flocs form in the surrounding stream, the sampled grain size decreases to include silt and clay (<63 μm). Samples were retrieved 2 to 4 days after storm events in order to capture both the rising and falling limbs of the hydrograph. The PVC sampler bodies were cleaned and new dewatering bags installed between storm events. Sediment-filled dewatering bags were transported back to the laboratory where sediment was rinsed from the bags with deionized water into a clean bucket. The water in the buckets was evaporated with the aid of a 60 °C heat lamp. Dewatering bags were cleaned by a power washer after each use and returned to service in their natal stream.

Stream stage data were retrieved from USGS gauges 02086849 and 0208700550 for Ellerbe and Little Lick Creeks, respectively. We also used the Little Lick Creek gauging station as a proxy for the ungauged Lick Creek, as the two basins are similar in size and their suspended sediment collection sites were only 7 km apart. All storm events sampled in Little Lick Creek were also sampled in Lick Creek (Fig. 3). New Light Creek stage data were recorded by a Solinst (Georgetown, Ontario, Canada) levelogger pressure meter deployed next to the suspended sediment sampler between December 2011 and February 2012. Two storm events were sampled prior to the deployment of the levelogger in New Light Creek.

2.4 Stream bank geochronology

Bulk sediment and woody organic material were collected for radiocarbon dating from stream bank sections along Ellerbe, Lick, and New Light Creeks in order to determine the

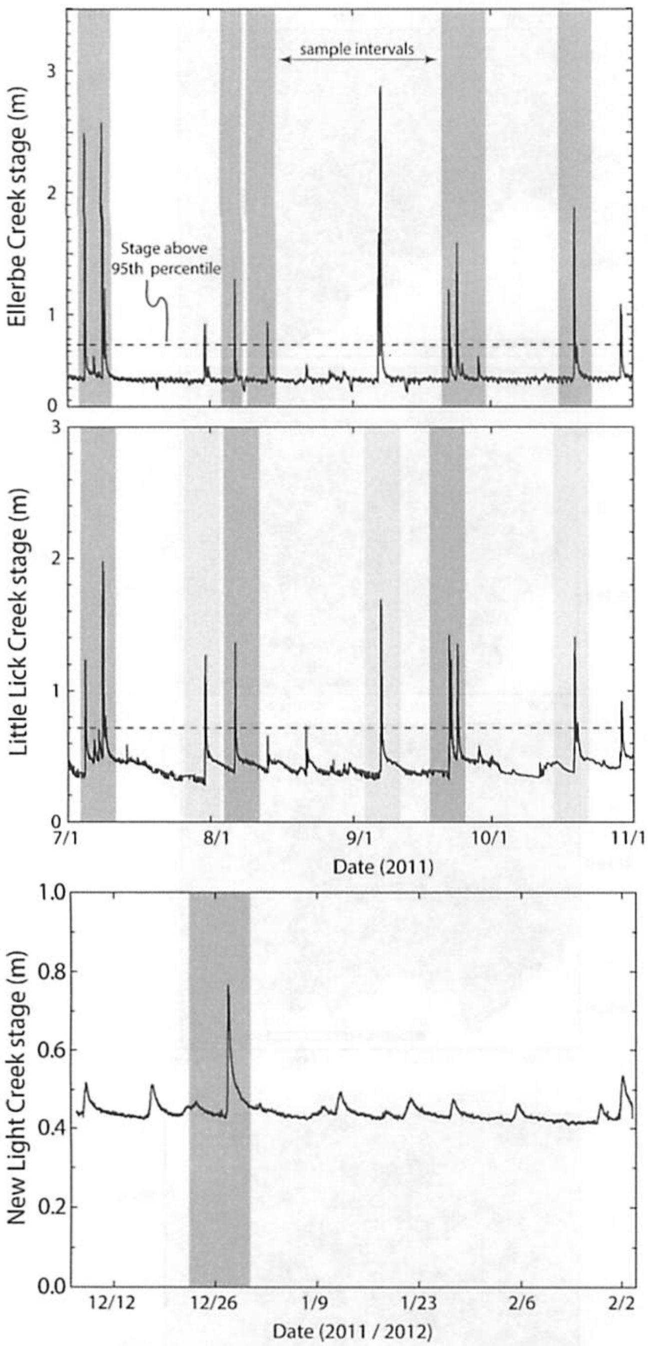

Fig. 3 Hydrographs for Ellerbe, Little Lick, and New Light creeks during the study period, provided in month/day format. *Shaded areas* outline storm events that were sampled for suspended sediment with the integrated samplers. The stage data for the Ellerbe and Little Lick creek hydrographs were obtained from the US Geological Survey (2012) WaterWatch website (http://waterwatch.usgs.gov/). Additional storm events sampled in Lick Creek are denoted in *light gray*. *Vertical axes* differ between hydrographs

depositional age of the 1–3-m-thick alluvial deposits. The stream bank was cleared back 30 cm with a shovel in order to expose fresh surfaces for sample collection. Samples were collected from the gleyed saturated soils exposed at the base of the stream banks, from the stratigraphic interval hypothesized as the horizon between pre- and post-European sediments, and from above that contact). All samples were disaggregated in sonic baths for 1 h and collected on a 100-mesh sieve (150 μm). The coarse fraction was retained, and the woody material and charcoal were selected and treated with an acid–base–acid pretreatment wash to remove soil carbon and soluble humic residue (Olsson 1986). Accelerator mass spectrometry radiocarbon dating was performed at DirectAMS (Seattle, USA). Ages are reported following standard procedures (Stuiver and Polach 1977) and converted to calibrated calendar years BC/AD with the CALIB v. 6.0 Radiocarbon Calibration online calculator (http://calib.qub.ac.uk/calib/; Stuiver et al. 2005).

2.5 Magnetic susceptibility

Stream bank bulk magnetic susceptibility values (κ) were obtained using a GF Instruments (Brno, Czech Republic) SM-20 instrument at each site where radiocarbon samples were collected. The reported κ values represent three repeat measurements (mean and standard deviation) collected every 10–30 cm along smooth 30–50 cm deep freshly exposed vertical profiles extending from the top of the stream bank to the baseflow water surface.

2.6 Geochemical analysis

Dried source and suspended sediment samples were prepared for geochemical (major, minor, and radionuclide) analyses by disaggregating and homogenizing with a ceramic mortar and pestle, followed by sieving to ≦ 63 μm, ensuring that only the source component likely to become part of the stream suspended load was analyzed. Element analyses were performed by Acme Labs (Vancouver, Canada) for 55 elements, including 14 rare earth elements (REEs) via inductively coupled plasma–mass spectrometry using the four-acid digestion (HNO_3, $HClO_4$, HF, and HCl) procedure of Kimbrough and Wakakuwa (1992) from a 0.25-g sediment split. The weight-percent of major elements were converted to oxides using stoichiometric conversion. Radionuclide (^{137}Cs and ^{40}K) analyses for New Light and Little Lick Creek basin samples were performed at the U.S. Department of Agriculture's Southwest Watershed Research Center with a gamma ray spectrometry system utilizing two n-type high-purity closed-end coaxial germanium detectors with >30% relative efficiency.

2.7 Particle size analysis

Particle size can exert an important control on trace element concentrations as they may increase as a function of available surface area (Horowitz and Elrick 1987). For this reason, we determined sample grain size (D_{50} and standard deviation) with a Beckman Coulter LS 13–320 laser particle size analyzer in order to normalize the geochemical data. Homogenized and disaggregated 1 g splits were used from at least two samples of sediment from each source type (e.g., stream bank, forest, construction site) along with the ≤63 μm fraction from two suspended sediment samples from each basin. We calculated a particle size correction factor prior to statistical analyses for each sediment source by utilizing the ratio of the source to suspended sediment D_{50} with the following equation:

$$GCcor = GCorg \times \frac{SCD_{50ij}}{SSD_{50j}} \quad (1)$$

where *GCcor* is the particle-size corrected geochemical concentration, *GCorg* is the original geochemical concentration, SCD_{50} is the median particle size of sediment source *i* collected in basin *j*, and SSD_{50} is the median particle size of the suspended sediment samples collected from basin *j*. A separate organic matter correction was not included in this study, as enrichment in organic matter is closely linked to enrichment in fine-grained sediments (Walling 2005).

2.8 Statistical analysis

We tested geochemical tracers for their ability to distinguish between sediment sources with the Kruskal–Wallis one-way analysis of variance *H* test (Kruskal and Wallis 1952), which is able to test for the independence of more than two variables without presuming either normal or non-normal distributions. Tracers proving significance ($p<0.05$) between sources were retained. Tracers passing the Kruskal–Wallis *H* test that were non-conservative (suspended sediment tracer values that were not bracketed by sediment source tracer values) were removed before performance of the mixing analysis.

Tracers passing the first stage of statistical analysis were entered into a stepwise Discriminant Function Analysis (DFA) intended to optimize the number used in the mixing model. This analysis results in the smallest combination of tracers that are capable of correctly distinguishing 100% of the sources through the minimization of Wilks' Lambda (Collins et al. 1998). The analysis was run separately for each drainage basin using IBM SPSS Statistics v. 20.0.

2.9 Mixing model

We used Monte Carlo sampling to determine the percent contribution of sediment from each source to the total suspended sediment load. In reality, only a limited number of surface sediment samples could be collected to represent all of the sources of suspended sediment in each drainage basin. This limitation results in uncertainty in the estimation of sediment source percent contribution values. The Monte Carlo approach was deemed appropriate because it incorporates the effects of this uncertainty into the mixing model by random sampling of derived parameter probability distributions (Small et al. 2002). Monte Carlo sampling and mixing model runs were completed for each drainage basin using MATLAB version 7.9.

Conservative tracers passing the Kruskal–Wallis *H* test were formulated with a multivariate normal distribution using the following equation.

$$x_k^t \sim MVN_T(\mu_k, \Sigma_k); k = 1, \ldots, K, t = 1, \ldots, T \quad (2)$$

where *k* is an index for sediment source, *t* represents each tracer data value, *K* is the total number of sediment sources, and MVN_T is a multivariate normal distribution for *T* tracers, with a specified mean μ and the ($T \times T$) dimensional covariance matrix Σ. The model was run by taking 50,000 random samples from the multivariate normal distributions from both the sediment source and suspended sediment samples for each basin. Each set of random values were used to solve the following mass balance equation.

$$Z^T = \sum_K \left(X_K^T \times P_K\right), 0 \le P_K \le 1, \sum_K P_k = 1 \quad (3)$$

where *z* and *x* are the suspended and source sediment tracer data, respectively, and *P* is the fraction of suspended sediment originating from each *k* sediment source. The non-negativity constraint resulted in a considerable amount of zeros being sampled from the multivariate normal distributions; therefore each solution that contained one or more zeros was removed from the pool of 50,000 solutions. This resulted in substantially more solutions being removed from New Light Creek than from the other basins; therefore the New Light Creek model was re-run using 100,000 random samples. The mean of the remaining pool of solutions to the mass balance equation provides the final estimate of the relative sediment contribution from each source to the suspended sediment load.

3 Results

3.1 Stream bank geochronology

Radiocarbon dating of stream bank profiles along Ellerbe, Lick, and New Light creeks suggests a difference in basin depositional history (see Electronic Supplementary Material, Table 4). Dating along Ellerbe Creek revealed that 2.5–3 m of legacy sediment has aggraded within the valley bottom since European settlement (Fig 4). The presence of numerous alternating fine and coarse-grained strata within this profile are characteristic of deposition in slack-water environments punctuated by flood events (Hunt 2011; Wegmann et al. 2012). As such, we interpret the majority of these aggraded sediments to have resulted from the damming of the stream for water-

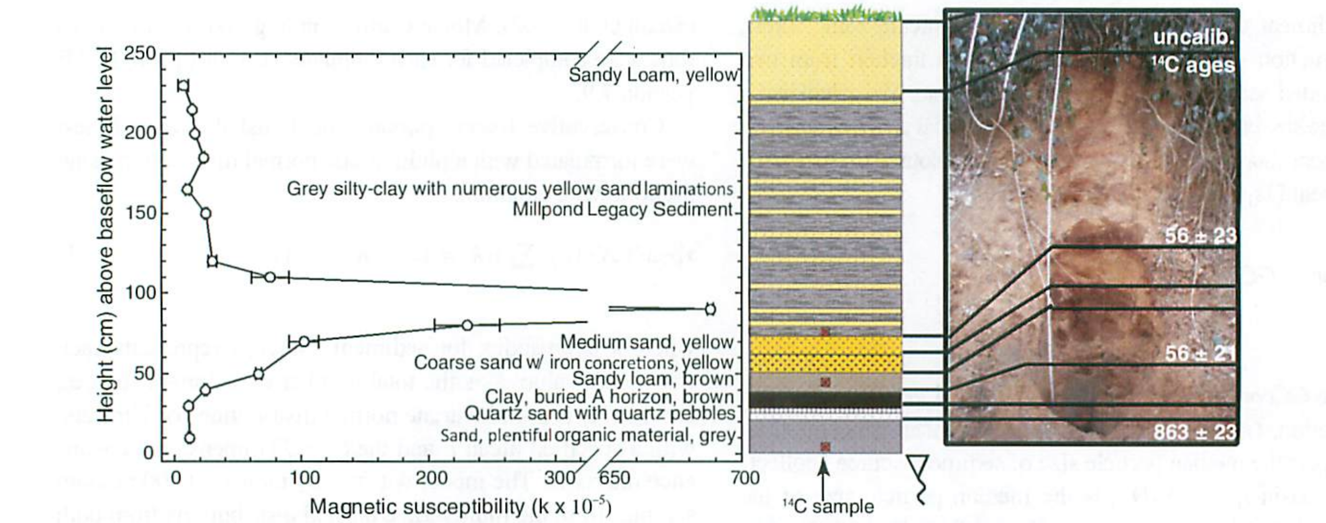

Fig. 4 Stratigraphic, radiocarbon, and magnetic susceptibility results from an Ellerbe Creek stream bank. The magnetic susceptibility plot shows a prominent spike approximately 90 cm above baseflow water level, which coincides with young radiocarbon ages. The stratigraphic section and photograph illustrate a large section of alternating fine and coarse-grained strata, likely the result of slackwater sedimentation occurring upstream of a mill–dam impoundment

power milling. Although 2–3 m of aggraded sediments (above base flow level) is also exposed along the banks of Lick Creek, ^{14}C samples date to well before European settlement (Fig. 5 and Electronic Supplementary Material, Table 4). Further, the presence of only faint laminae and strongly structured soils in the Like Creek exposures suggests a lack of post-European depositional disturbance. The New Light Creek basin is the only study catchment with a mapped record of mill–dams along its bottom lands, but charcoal collected ~80 cm below the modern day ground surface, was dated at 300 ^{14}C year BP (Fig. 6 and Electronic Supplementary Material, Table 4), prior to European settlement. Since charcoal tends to be recalcitrant, it is possible that it overestimates the true timing of sediment aggradation in this basin.

3.2 Magnetic susceptibility

Magnetic susceptibility (MS) aided in interpreting the degree of basin-wide anthropogenic disturbance recorded in stream bank sediments. Magnetic susceptibility of fine-grained alluvial deposits is useful for identifying valley bottom sediments derived from hillslope soils that experienced heating due to forest burning from both anthropogenic and natural causes (e.g., Ketterings et al. 2000). Mean MS values between ~0–20 κ were observed at the base of the stream banks along three measured exposures, indicating a low abundance of ferrimagnetic minerals, and potentially a high abundance of diamagnetic materials (e.g., Mullins 1977). Stream bank sections along Ellerbe and New Light Creeks exhibit prominent increases in MS (>100 κ) above the demonstrably pre-European settlement sediments (see Figs. 4 and 6). These large κ values, which coincide with younger radiocarbon ages, are interpreted as reflecting an increase in secondary ferrimagnetic minerals due to the heating of soils occurring from slash and burn practices employed during European land clearing, and subsequent upland erosion of the upper portions of these soils. Both indigenous peoples and European-American settlers employed slash-and-burn practices in order to quickly clear land and provide needed soil nutrients for farming along with forage for wild game and livestock (Van der Donck 1841; Otto and Anderson 1982; Stinchcomb et al. 2011). The MS values decrease close to the modern ground surface since the highest stratigraphic units are commonly recent over bank deposits of quartz-rich sand. Lick Creek did not show a prominent spike in MS, nor did it have post-European settlement radiocarbon ages (see Fig. 5), consistent with our interpretation that minimal legacy sediment aggradation occurred in this basin.

3.3 Particle size analysis

Slight differences in sample D_{50} and total grain-size distributions are observed for the sediment sources from each basin (Voli 2012; Table 2 and Fig. 7). Forest soils have the largest median grain size in three of the basins, a reflection of the sandy loam texture of the A horizon for typical study area soils (Cawthorn 1970; Kirby 1976). In the urban Ellerbe Creek basin, the coarsest source sediment is associated with paved roads. Construction sites are a consistently fine-grained source, likely due to exposure of illuvial (Bt) horizons following topsoil removal. The grain size of stream banks is intermediate to the other sources. Particle size correction factors derived from Eq. (1) as a function of sediment source from each basin varied between 0.65 and 1.67. These were applied to the geochemical data prior to statistical and mixing model analysis by multiplying the tracer and correction-factor values.

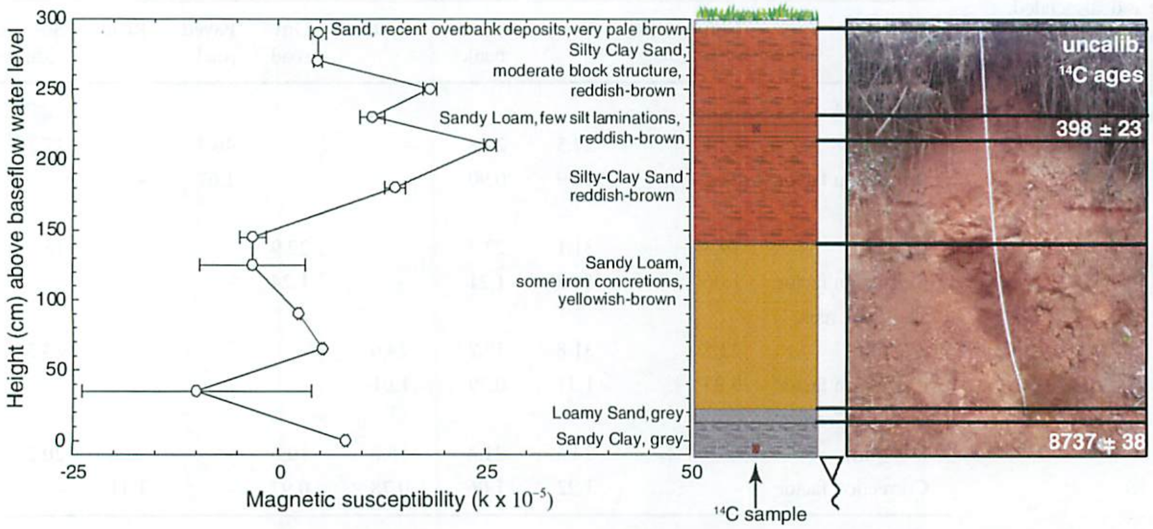

Fig. 5 Stratigraphic, radiocarbon, and magnetic susceptibility results from a Lick Creek stream bank. The magnetic susceptibility plot lacks any distinct spikes, and the radiocarbon samples date to well before European settlement. The stratigraphic section and photograph show that the approximately 300 cm of aggraded sediment within the valley bottoms of Lick Creek are strongly structured and lack any prominent laminae typical of deposition within ponded (mill–pond) environments

3.4 Geochemical results and statistical analysis

After parsing the elements through the Kruskal–Wallis H test and removal of non-conservative tracers, 40, 10, 12, and 19 elements, remained for the Ellerbe, Lick, Little Lick, and New Light basins, respectively, that were significantly different between sediment sources ($p<0.05$; see Electronic Supplementary Material, Table 3). The presence of non-conservative tracers may result from unaccounted sediment sources or from biogeochemical transformations of tracers during erosion and transport, or via inappropriate accounting for differences in tracer concentrations as a function of sampled particle size (e.g., Russell et al. 2001). Because of the relatively small number of source sample elemental determinations from each basin, the DFA often resulted in relative sediment contribution uncertainties larger than those obtained without the use of the DFA after the running of the mixing model. Thus, all conservative tracers passing the Kruskal–Wallis H test were used in the mixing model.

3.5 Mixing model

Mean source contributions to suspended sediment vary between the Falls Lake tributary basins (Fig. 8). Stream bank sediment is

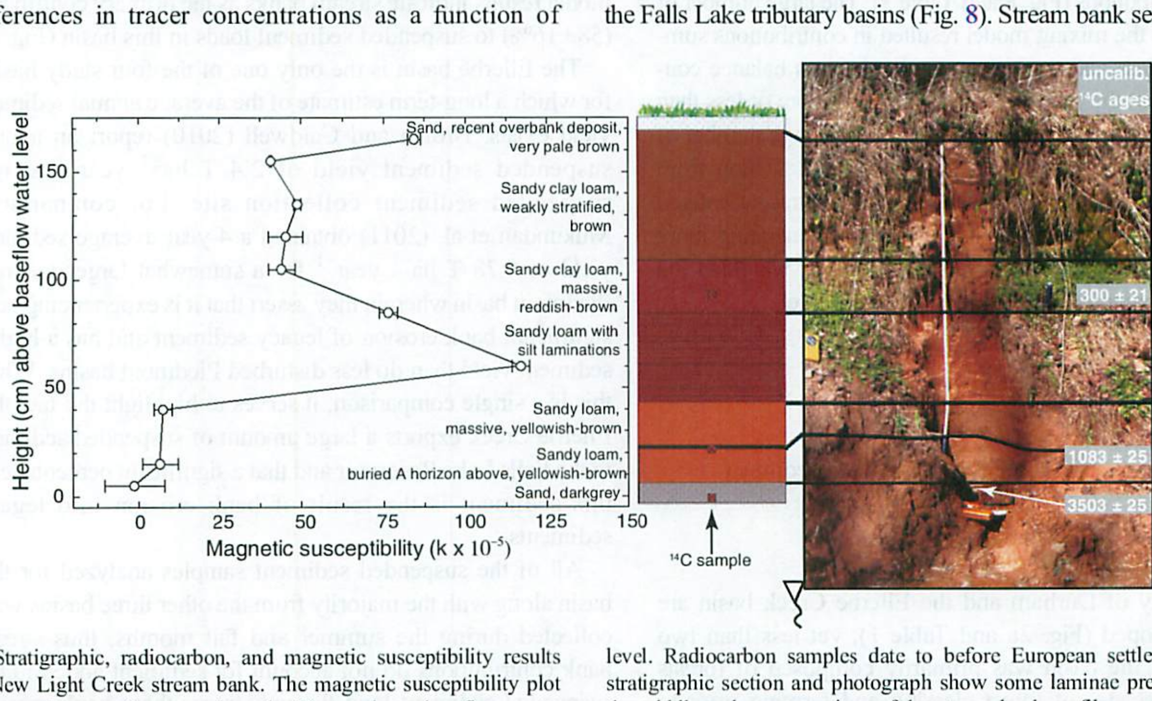

Fig. 6 Stratigraphic, radiocarbon, and magnetic susceptibility results from a New Light Creek stream bank. The magnetic susceptibility plot shows a prominent spike approximately 60 cm above baseflow water level. Radiocarbon samples date to before European settlement. The stratigraphic section and photograph show some laminae present along the middle and upper portions of the stream bank profile

Table 2 Source and suspended sediment grain size (D_{50}) and calculated correction factors used in the geochemical mixing model for each catchment

	Construction	Forest	Stream bank	Pasture	Dirt road	Paved road	Road cut	Suspended sediment
Ellerbe Creek								
D_{50} (μm)	18.1	27.5	24.8	–	–	46.3	–	27.7
Correction factor	0.65	0.99	0.90	–	–	1.67	–	–
Lick Creek								
D_{50} (μm)	19.5	31.1	22.3	–	22.9	–	–	18.5
Correction factor	1.06	1.69	1.21	–	1.24	–	–	–
Little Lick Creek								
D_{50} (μm)	22.5	31.8	19.2	24.6	–	–	–	24.3
Correction factor	0.93	1.31	0.79	1.01	–	–	–	–
New Light Creek								
D_{50} (μm)	–	24.6	21.4	15.8	19.6	–	22.4	20.2
Correction factor	–	1.22	1.06	0.78	0.97	–	1.11	–

the dominant source in New Light (62±12%), Ellerbe (58±16%), and Little Lick (33±20%) creeks, while construction sites are the dominant source in Lick Creek (43±19%). A summary of the modeled estimates of source sediment contributions and 1-sigma uncertainty ranges is given in Table 3. The small number of sediment samples collected from the basins results in large uncertainties for many of the contribution estimates. New Light Creek is the one basin that produced low uncertainties, which we attribute to the near identical signature between the stream bank and suspended sediment geochemical data used in the mixing model (see Section 4.1.4). Despite large uncertainties, most of the mixing model results are normally distributed, with the exception of the sources whose mean contributions are close to zero, as the model does not allow for negative contributions (Fig. 8 and Table 3). The large number of tracers used in the mixing model resulted in contributions summing to both less and more than 100%. A mass balance constraint to prevent the model from summing to more or less than 100% was added to the mixing model, but the large number of tracers used minimized its effectiveness. The deviation from 100% increases with an increasing number of tracers entered into the mixing model. This has the effect of generating more uncertainty in the model results than if we had weighted the mass balance parameter more heavily in the model.

4 Discussion

4.1 Catchment-specific sources of suspended sediment

4.1.1 Ellerbe Creek

Today, the city of Durham and the Ellerbe Creek basin are densely developed (Fig. 2a and Table 1); yet less than two centuries ago, the basin was primarily composed of forests and farms. Decades of forest clearing and farming prior to urbanization led to the accumulation of a significant amount of aggraded valley bottom sediment. Radiocarbon geochronology and magnetic susceptibility measurements confirm that several meters of legacy sediment currently buries the pre-European floodplain along the upper portion of the basin (Fig. 4 and Electronic Supplementary Material, Table 4). The presence of these easily-eroded sediments combined with a mid-twentieth century U.S. Army Corps of Engineers channelization project in the lower basin has left the majority of Ellerbe Creek incised from its source to its confluence with Falls Lake. High peak discharges resulting from the large amount of impervious surfaces across the basin coupled with the unstable banks of its incised channels undoubtedly leads to high rates of stream bank erosion, and is likely why the mixing model results indicate stream banks as the primary contributor (58±16%) to suspended sediment loads in this basin (Fig. 8).

The Ellerbe basin is the only one of the four study basins for which a long-term estimate of the average annual sediment yield exists. Brown and Caldwell (2010) report an annual suspended sediment yield of 2.4 T ha^{-1} year^{-1} at our suspended sediment collection site. For comparison, Mukundan et al. (2011) obtained a 4-year average sediment yield of 0.78 T ha^{-1} year^{-1} for a somewhat larger Georgia Piedmont basin wherein they assert that it is experiencing both significant bank erosion of legacy sediment and has a higher sediment yield than do less disturbed Piedmont basins. While this is a single comparison, it serves to highlight the fact that Ellerbe Creek exports a large amount of suspended sediment to the Falls Lake Reservoir and that a significant percentage of this sediment is the result of bank erosion into legacy sediments.

All of the suspended sediment samples analyzed for this basin along with the majority from the other three basins were collected during the summer and fall months, thus stream bank contributions do not account for sediment added to the suspended sediment load through freeze-thaw bank erosion

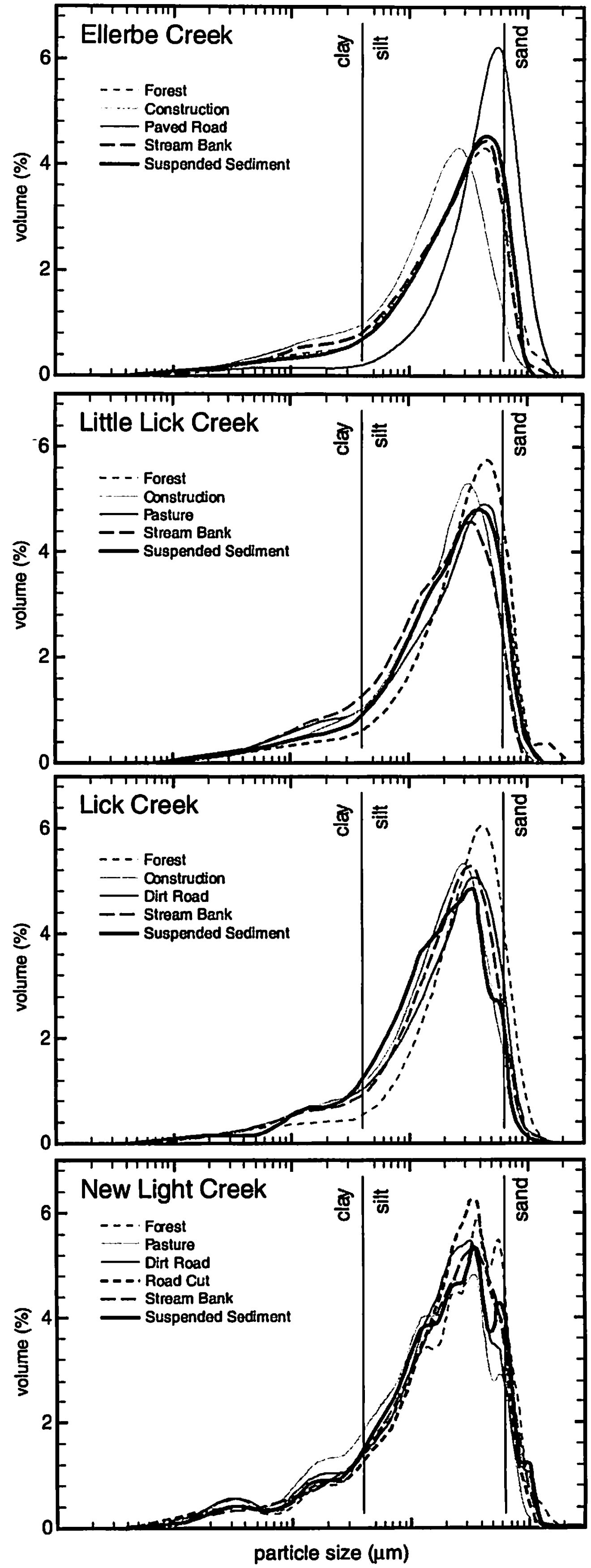

Fig. 7 Particle size distributions of source and suspended sediment samples from each basin where the particle diameter is plotted against the percent volume of the sample. Clay, silt, and sand size demarcations are indicated on each distribution plot

during the winter months, which is when most of the stream bank erosion occurred on several mid-Atlantic Piedmont streams previously monitored for rates of bank erosion (e.g., Wolman 1959; Merritts et al. 2011), implying that we might be underestimating the contribution of stream banks to the total annual suspended sediment load in Ellerbe and the other study basins.

Paved roads cover a large portion of the basin, but in the mixing model, this represent only a 13±4% contribution to the suspended load. A limited amount of fine-grained street residue is available during storm events; thus, paved roads likely contribute little sediment to the suspended load during long duration or closely spaced storm events. Although active construction sites exist within the basin, they are not a major contributor to the suspended sediment load (19±17%). Active construction sites are primarily found along the margins of the basin and the fringes of the city of Durham, which occupying the majority of the catchment was developed many decades ago. The moderate contribution from forests (26±18%) was not expected in an urbanized drainage basin; however, this may reflect the erosion of soils following commercial timber harvesting in the upper catchment (Voli 2012). Though timber harvesting can lead to a localized increase in sediment loads from Piedmont forests, pre-harvesting sediment delivery levels are typically re-established within 2–5 years, and best management practices that reduce soil disturbance and loss can minimize the negative water quality effects associated with mechanized harvesting on erodible soils (Aust and Blinn 2004).

4.1.2 Lick Creek

The Lick Creek basin is primarily forested, yet mixing model results suggest that recent development occurring in only a small portion of the basin is having a significant impact on suspended sediment loads (43±19%). During our monitoring, several active construction sites were located along the drainage divide between Lick and Little Lick creeks, where White Store soils are prevalent (Fig. 2 and Electronic Supplementary Material, Table 1). It has been observed that streams draining areas experiencing active urbanization can have suspended sediment concentrations 10–20 times higher than those draining woodlands (Wolman and Schick 1967). These differences in sediment concentrations may be even higher in basins such as Lick Creek, where active construction is taking place on highly erodible soils (Cawthorn 1970).

Radiocarbon geochronology and magnetic susceptibility results show no evidence of legacy sediments within the

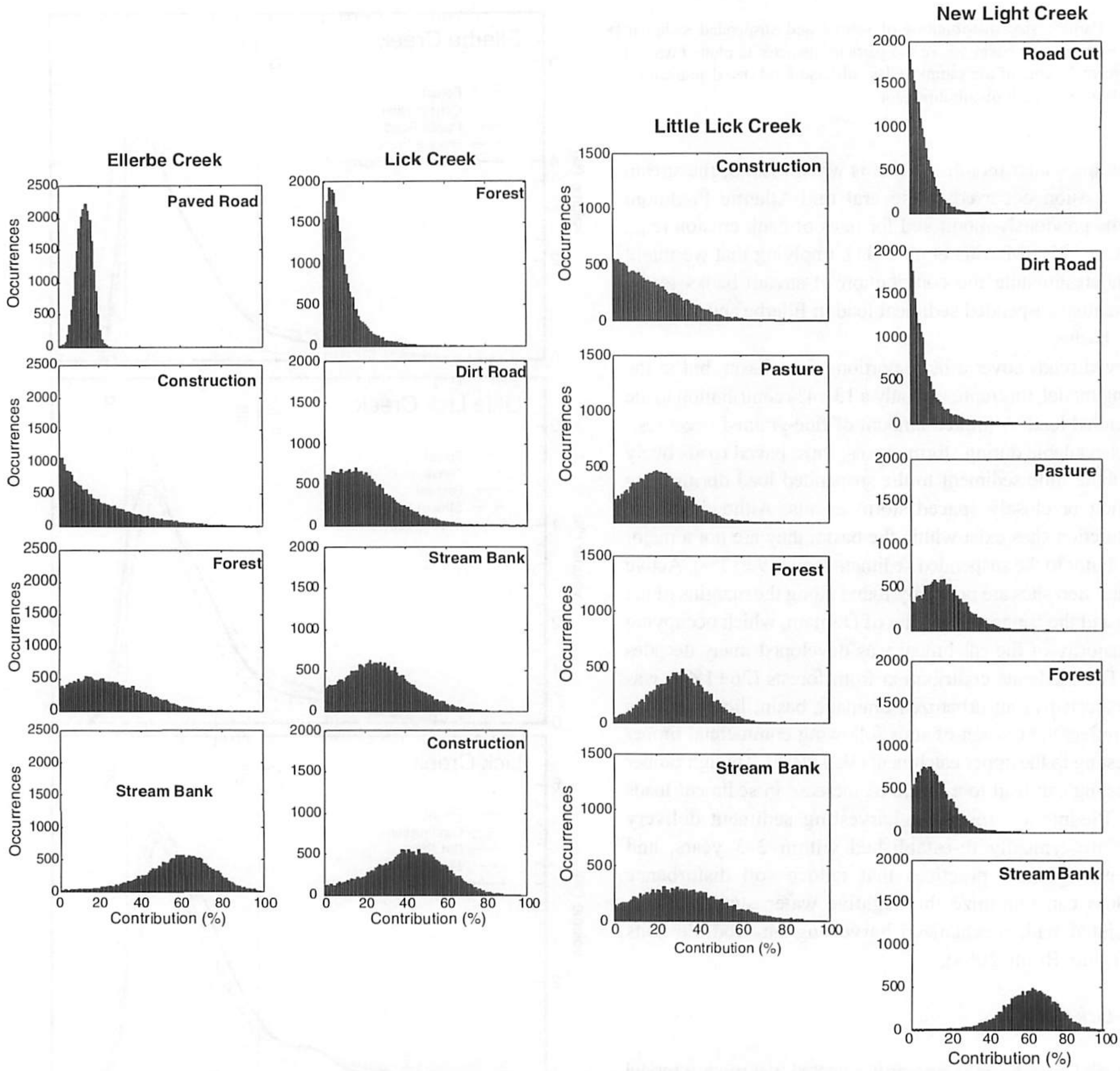

Fig. 8 Monte Carlo simulation results for each drainage basin where the percent contribution from a given source is plotted against the number of modeled occurrences that fall within each 1% bin. The number of model iterations varied between 5,000 and 10,000, depending on the number of solutions eliminated for producing a zero-percent contribution. Modeled sources are arranged in order of increasing mean percent contribution for each basin

Lick Creek valley bottom (Fig. 5). The moderate structure of the stream bank soils, not found along the other study streams, is also indicative of a soil that has not formed recently, yet the trunk channel is characterized by steep banks, which contribute the second highest amount (27±16%) to the suspended sediment load. This suggests that the stream bank morphology and erosion within the basin may be a result of recent channel incision. Small to moderate-sized basins typically display a 2–3 times increase in peak discharge following urban-to-suburban development (Booth 1990). Such hydrograph modifications result in increased shear stress exerted on the bed and banks of the channel that in turn cause greater sediment entrainment as well as channel incision and widening. This suggests that the recent development in the upper portions of the basin is causing not only an increase in fine sediment from construction sites, but may also be responsible in large part for the recent incision and channel widening of the creek, and thus enhanced erosion of stream bank sediments.

Table 3 Monte Carlo-based geochemical mixing model results reported as mean and median percentages by contributing source and their 1-σ uncertainty for each catchment

Ellerbe Creek	Construction	Paved road	Forest	Stream bank	
Mean contribution	19	13	26	58	
Median contribution	14	13	23	59	
1-σ uncertainty[a]	3 to 36	9 to 17	8 to 43	42 to 74	
Lick Creek	Construction	Dirt Road	Forest	Stream Bank	
Mean contribution	43	22	9	27	
Median contribution	44	19	7	25	
1-σ uncertainty	24 to 61	6 to 37	2 to 15	11 to 43	
Little Lick Creek	Construction	Pasture	Forest	Stream Bank	
Mean contribution	18	22	31	33	
Median contribution	16	21	31	31	
1-σ uncertainty	4 to 33	9 to 34	18 to 44	13 to 52	
New Light Creek	Road Cut	Dirt Road	Pasture	Forest	Stream Bank
Mean contribution	6	9	13	16	62
Median contribution	5	6	12	15	62
1-σ uncertainty	1 to 11	2 to 18	5 to 22	7 to 24	50 to 74

All contributions and uncertainties are rounded to the nearest 1%

[a] Based upon non-parametric quintile values that correspond to 1-sigma (68%) uncertainty

4.1.3 Little Lick Creek

Little Lick Creek is primarily an urban drainage basin situated on the outskirts of the city of Durham. The basin is similar in size, geology, and soils to Lick Creek, but did not have as many active construction sites during the fingerprinting study. The lack of active construction sites coupled with a moderate density of impervious surfaces is reflected in the mixing model results, which show stream banks (33±20%) and construction sites (18±15%) as the largest and smallest contributors, respectively (Fig. 8). Forest soils (31±13%) are the second largest mean contributor, similar to observations from the Ellerbe Creek catchment, which again may be due to commercial timber harvesting, especially when the trees are harvested across ephemeral channels (Voli 2012). Material for radiocarbon dating and magnetic susceptibility measurements were not collected along Little Lick Creek. However, the morphology and stratigraphy of the stream banks are similar to Lick Creek, and thus it is assumed that the high stream bank-derived suspended sediment contributions are from erosion and incision caused by the high peak flows associated with this urban drainage basin (Table 1).

The lack of legacy sediment within the Lick and Little Lick Creek valley bottoms can be explained by the geologic and pedologic conditions. White Store soils, derived from Triassic sedimentary rocks, are dominant in these basins (see Electronic Supplementary Material Data, Table 1). Because White Store soils have a narrow range of suitable moisture regimes and low natural fertility, they have not been used extensively for agricultural production (Kirby 1976; Daniels et al. 1999; Helms 2000). In addition, the lack of resistant lithologies and low stream gradients render the valley bottoms as unfavorable locations for the construction of water-powered mill–dams (Heron 1978). These agricultural and industrial limitations likely resulted in relatively little post-European valley bottom sediment aggradation and storage in these basins.

4.1.4 New Light Creek

The New Light Creek basin is undeveloped with current land use dominated by forest and pasture lands (Fig. 2d and Table 1). Historic aerial photographs reveal that a substantial number of today's woodlands and pastures were farms during the nineteenth and early twentieth century (Voli 2012). Historic maps also show the presence of several mill–ponds along New Light Creek (Bevers 1871). Many of the stream banks expose several meters of aggraded sediment, and the radiocarbon geochronology and magnetic susceptibility measurements suggest that almost 1 m of valley bottom aggradation has occurred during the last three centuries (Fig. 6 and Electronic Supplementary Material, Table 4).

With well-vegetated woodlands and pastures currently covering much of the basin, stream bank erosion contributed 62±12% to the suspended sediment load (Fig. 8). With only relatively small volumes of sediment captured by the sampler during storm events, a hydrograph with low peak flows during those events (Fig. 3), and New Light Creek's absence from the

North Carolina 303(d) list, suggest that this stream is not moving a substantial amount of suspended sediment during storm events. This result is expected in an undeveloped drainage basin with minimal non-vegetated surface area where half to three quarters of the suspended load is likely derived from stream bank erosion of predominantly aggraded legacy sediments.

5 Conclusions

The mitigation of nonpoint-source pollutants, such as sediment, in larger basins is rarely a straightforward procedure due to the number of sources and erosional processes contributing to their concentration in waterways. Sediment fingerprinting revealed that stream bank erosion in general, and of legacy sediments in particular, from valley bottoms of streams draining to Falls Lake is at the root of the regional sediment loading problem. Several methods have previously been employed to restore mid-Atlantic Piedmont streams suffering from high sediment loads arising from the erosion of legacy sediments. Traditional stream restoration methods promote the protection of incised stream bank reaches with large structures, such as boulder and/or rootwad revetments, or bank stabilization through grading to a stable angle followed by revegetation (Brown 2000). The bank protection approach is often expensive and fails to reduce bank erosion in streams characterized by high sediment loads, excess shear stress, and easily erodible banks, where stabilizing structures are often stranded in the middle of channels in the years to decades after implementation (e.g., Miller and Kochel 2010). Alternative approaches have focused on valley bottom removal of legacy sediments in order to restore Piedmont streams to their pre-legacy sediment condition (Merritts et al. 2011). The legacy sediment removal option would ensure a large reduction in the contribution of stream bank sediment to the suspended load, but is likely unfeasible in large and often heavily developed basins. We suggest that this leaves a combination of better stormwater management aimed at reducing peak flows, and stabilization to reduce erosion of near-vertical banks produced by both high flow events and freeze-thaw actions as the preferable method of sediment mitigation in Ellerbe Creek.

Lick and Little Lick Creek could also benefit from additional stormwater management implementations, as it appears that a substantial amount of stream bank erosion and channel incision in these basins is due to increased peak flows. New Light Creek is not on the North Carolina 303(d) list and thus does not currently need mitigation, but the amount of valley bottom legacy sediment storage demonstrates that there is potential for high suspended sediment loads here during the late winter and early spring months following gravitational failure of bank sediments aided by freeze–thaw processes.

There is also potential to reduce sediment contributions from surface sources within the studied basins. Limiting the amount of time soils are left unvegetated and enforcing erosion and sediment control measures will further mitigate construction site erosion. Timber harvesting on gentle slopes away from stream channels and re-vegetating skid trails following harvesting should reduce forest soil contributions.

Our results demonstrate that stream bank erosion resulting from anthropogenic alterations of the pre-European valley bottom is the largest contributor to the suspended sediment load in three of four Falls Lake tributary basins and a significant contributor in the fourth. Valley bottom aggradation of legacy sediments is evident along Ellerbe and New Light creeks, from both of which stream banks contributed >50% of the suspended sediment load during the monitoring period. Stream bank erosion by means of channel incision and widening due to increased stormwater runoff appears to add to stream bank contributions in the Ellerbe, Lick, and Little Lick Creek basins, but plays a less significant role where the presence of legacy sediment is diminished (Lick and Little Lick Creeks). Although we document a significant stream bank erosion component to the suspended sediment load in each of these basins, our results may underestimate the annual percent contribution from stream banks, since the bank erosion is often more prevalent during the winter months (Wegmann et al. 2012; Starek et al. 2013). Sources of sediment other than stream bank erosion that had large contributions were exposed forest soils, presumably from commercial timber harvesting activities, and sediment runoff from active construction sites. Better use and enforcement of existing best management practices and erosion and sediment control measures will likely decrease contributions from forests and construction sites, respectively. Without taking the proper measures to address and mitigate nonpoint sources of suspended sediment, particularly stream bank erosion, these streams will continue to contribute to surface water turbidity problems via the transport of high TSS loads to Falls Lake, a regionally important drinking water source, into the foreseeable future.

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