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Sediment source analysis in the Linganore Creek watershed, Maryland, USA, using the sediment fingerprinting approach: 2008 to 2010

Allen C. Gellis · Gregory B. Noe

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Abstract
Purpose Fine-grained sediment is an important pollutant in streams and estuaries, including the Chesapeake Bay in the USA. The objective of this study was to determine the sources of fine-grained sediment using the sediment fingerprinting approach in the Linganore Creek watershed, a tributary to the Chesapeake Bay.
Materials and methods The sediment fingerprinting approach was used in the agricultural and forested, 147-km^2 Linganore Creek watershed, Maryland from 1 August 2008 to 31 December 2010 to determine the relative percentage contribution from different potential sources of fine-grained sediment. Fine-grained suspended sediment samples (<63 μm) were collected during storm events in Linganore Creek using an automatic sampler and manual isokinetic samplers. Source samples were collected from 40 stream bank sites, 24 agricultural (cropland and pasture) sites, and 19 forested sites. Suspended sediment and source samples were analyzed for elements and stable isotopes.
Results and discussion Results of sediment fingerprinting for 194 samples collected in 36 separate storm events indicate that stream banks contributed 53% of the annual fine-grained suspended sediment load, agriculture contributed 44%, and forests contributed 3%. Peak flows and sediment loads of the storms correlate to stream bank erosion. The highest peak flows occurred in the winter and, along with freeze–thaw activity, contributed to winter months showing the highest rate of stream bank erosion. Peak flow was negatively correlated to sediment sources from agricultural lands which had the greatest contribution in non-winter months. Caution should be observed when trying to interpret the relation between sediment sources and individual storms using the sediment fingerprinting approach. Because the sediment fingerprinting results from individual storms may not include the temporal aspects of the sourced sediment, sediment that is in storage from previous events, remobilized and sampled during the current event, will reflect previous storm characteristics. Stream bank sediment is delivered directly to the channel during an event, whereas the delivery of upland sediment to the stream is lower due to storage on hillslopes and/or in channels, sediment from stream banks are more likely to be related to the characteristics of the sampled storm event.
Conclusions Stream banks and agricultural lands are both important sources of fine-grained sediment in the Linganore Creek watershed. Peak flows and sediment loads for the 36 storms show a significant relation to sediment sources from stream bank erosion. Attempting to link upland sediment sources to flow and seasonal characteristics is difficult since much of the upland sediment eroded in an event goes into storage. By averaging sediment sources over several storms, it may be possible to determine not only the sediment sources that are directly contributed during the current event but also sediment from previous events that was in storage and remobilized.

Keywords Bank erosion · Chesapeake Bay · Sediment budget · Sediment fingerprinting

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A. C. Gellis (✉)
US Geological Survey, 5522 Research Park Drive,
Baltimore, MD 21228, USA
e-mail: agellis@usgs.gov

G. B. Noe
US Geological Survey, 430 National Center,
Reston, VA 20192, USA

1 Introduction

Worldwide, sediment is recognized as a water quality problem affecting aquatic ecosystem health (Ongley 1996; Taylor and Owens 2009; Walling 2009). In the Chesapeake Bay and its tributaries in the USA, fine-grained sediment is a major contributor to ecological degradation (Brakebill et al. 2010; Langland et al. 2012). Sediment has an adverse effect on the health of streams in the bay watershed and on submerged aquatic vegetation and living resources in the estuary; it results in degraded water quality, loss of habitat, and population declines in biological communities.

The successful mitigation of sediment-related problems often requires knowledge of sediment sources. Linganore Creek is listed on Maryland's streams for sediment impairment (Maryland Department of the Environment 2008). The study area is upstream of Lake Linganore, a reservoir that is used for water supply and recreation. Lake Linganore is also experiencing a sedimentation problem (Sekellick and Banks 2010). Therefore, identifying the major sources of fine-grained sediment in the upper portions of Linganore Creek can assist in developing strategies to reduce sediment delivered to Lake Linganore and improve aquatic conditions, reservoir storage capacity, and reduce water treatment costs.

In characterizing sediment sources, an important first step is proportioning the sediment that is derived from uplands (i.e., agriculture) versus channel (i.e., stream banks) sources. This first step is important because management strategies designed to reduce sediment from these sources would have very different approaches. In Linganore Creek, upland sources are related to land cover and land use practices (i.e., agriculture (cropland and pasture) and forests). Erosion on upland surfaces can occur though sheetwash, rilling, gullying, and mass movements. Periods of heavy rain that lead to saturation excess overland flow or infiltration excess overland flow can erode and mobilize upland sediment. Gellis et al. (2009) showed that for two of four events in the Pocomoke River, Maryland and Delaware, a tributary of the Chesapeake Bay, upland sediment (agriculture and forest) was an important source when the sampled storms were likely to have produced overland flow which is necessary to erode surface sediment.

Channel sources are typically the channel bed and stream banks. The channel bed can be a source of sediment in incising channels. However, the beds of the main stem and tributaries in many watersheds are not considered as sediment sources, since in the absence of significant channel incision, sediment mobilized from the channel bed is likely to reflect temporary storage of sediment originating from upstream sources and is, therefore, not treated as a separate source (Gellis et al. 2009).

Stream banks can be an important sediment source and erode by three mechanisms: (1) freeze–thaw processes (Wolman 1959; Wynn 2006); (2) fluvial erosion (Julian and Torres 2006; Wynn 2006); and (3) mass wasting (Darby et al. 2007; Wynn 2006). Freeze–thaw action of the bank surfaces causes the soil to expand and loosen. The material that is loosened is readily available for transport by a range of flows that inundate the bank surface (Lawler 1986; Wolman 1959). Fluvial erosion is the detachment, entrainment, and removal of particles or aggregates from the stream bank by the hydraulic forces of water. Hydraulic forces are related to the shear stress that the flow exerts on the bank. Sediment grain size, cohesiveness of grains, and vegetation are also important in whether stream banks are erodible (Wynn 2006). Hooke (1979) and Julian and Torres (2006) performed statistical analysis on the factors controlling bank erosion and determined that peak flow best predicts bank erosion. Mass wasting is the failure of all or part of the stream banks as a result of geotechnical instabilities. Mass failures can occur from fluvial erosion undercutting the toe of the stream banks and creating unstable conditions leading to bank failure (Simon et al. 2000). Bank failures and mass wasting are common during the recessional period of stormflow when seepage forces overcome the resistance of a grain's cohesion and the banks may fail (Fox et al. 2007; Simon et al. 2000).

In the Chesapeake Bay watershed, both upland and channel sources have previously been identified using the sediment fingerprinting approach in several small watersheds. In the agricultural watersheds (Little Conestoga Creek and the Pocomoke River), the primary sediment sources identified were stream banks and croplands (Gellis et al. 2009). In the agricultural Mill Stream Branch watershed, stream banks were 100% of the sediment sources for five suspended sediment samples (Banks et al. 2010). In the mixed land use (urban, agriculture, and forest) Mattawoman Creek watershed, significant sources of sediment include cropland, construction sites, and forests (Gellis et al. 2009). Stream banks, street residue, and upland areas were the primary sources in the urban Anacostia River watershed (Devereux et al. 2010). Results of sediment fingerprinting for watersheds in the Chesapeake Bay indicate that sediment sources vary among the watersheds, partly as a result of differences in past and present land use and geology.

Linganore Creek is also part of a network of small watersheds in the Chesapeake Bay where the US Geological Survey (USGS) and its partners are studying the erosion, transport, storage, and delivery of fine-grained sediment (Gellis and Brakebill 2013). In the Linganore Creek watershed, the USGS partnered with Frederick County to develop this study in which the objective was to determine the sources of fine-grained sediment using the sediment fingerprinting and sediment budget approaches. This paper describes the results of the sediment fingerprinting portion of the study.

2 Study area

The study was conducted in the upper portions of Linganore Creek, a tributary to the Monocacy River that drains parts of

Frederick and Carroll Counties, Maryland (Fig. 1). Located in the Piedmont Physiographic Province, the area is characterized by rolling hills and moderately to deeply incised, well-drained valleys with altitudes ranging from near sea level to more than 275 m above sea level (DiLisio 1983). The Piedmont Physiographic Province also has the highest sediment yields of any physiographic province in the Chesapeake Bay region (Gellis et al. 2009). Soils in the Linganore Creek watershed are primarily loams, silt loams, and gravelly loams of the Mt. Airy, Glenelg, and Blocktown Series developed on weathered phyllite and schist (Kraft 2002). Slope percentages for the Linganore Creek watershed derived from 30 m cells (n = 163,022) created from the 2004 National Elevation Dataset indicate that the 25th, 50th, and 75th percentiles of slopes are 4.9, 7.6, and 10.7%, respectively.

Average precipitation in the basin is 1,063 mm year^{-1} (US Department of Commerce et al. 2009). Temperatures range from a July mean of 26 °C to a December mean of 0.7 °C (Sekellick and Banks 2010; US Department of Commerce et al. 2011). Land use in 2006 in the 147-km^2 watershed was 27% forest, 26% hay and pasture (herein referred to as pasture), 36% cropland, 8% developed, and 3% other (Fry et al. 2011). The agriculture acreage is comprised primarily of corn, wheat, barley, soybean, and pasture/hay (US Department of Agriculture (USDA) and National Agricultural Statistics 2009). Cultivated crops are planted from April to May and harvested during August and September. Both till and no-till operations are used. Pasture land is used for livestock grazing and dairy. Forests are primarily secondary growth forests which have developed after land clearing in the late eighteenth century and early nineteenth century for agriculture (Sprague et al. 2006). During the study period, forests were not managed for commercial uses.

3 Materials and methods

3.1 The sediment fingerprinting approach

Source analysis of fine-grained sediment using the sediment fingerprinting approach was conducted in the watershed draining to USGS streamflow gauging station 01642438 (Fig. 1; Linganore Creek near Libertytown, MD, with a drainage area of 147 km^2). The sediment fingerprinting approach provides a direct method for quantifying watershed sources of fine-grained suspended sediment (Collins et al. 1997; Gellis

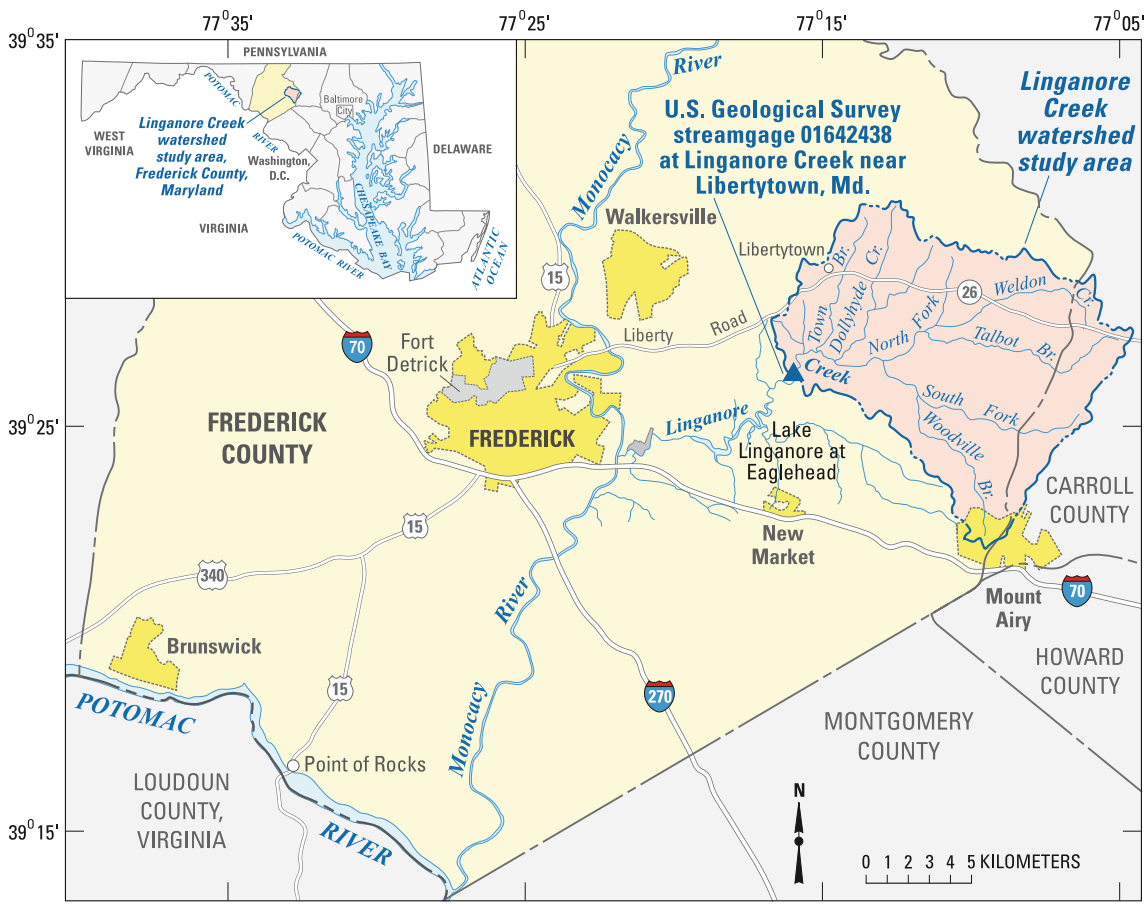

Fig. 1 Location of study area, Linganore Creek watershed, Frederick County, MD, USA

et al. 2009; Motha et al. 2003; Mukundan et al. 2012). This approach entails the identification of specific sources through the establishment of a minimal set of physical and/or chemical properties, such as tracers that uniquely define each source in the watershed. Suspended sediment collected under different flow conditions exhibits a composite or fingerprint of these properties that allows them to be traced back to their respective sources. Tracers that have been successfully used as fingerprints include mineralogy (Motha et al. 2003), radionuclides (Collins et al. 1997; Nagle et al. 2007; Walling and Woodward 1992), trace elements (Devereux et al. 2010), magnetic properties (Slattery et al. 2000), and stable isotope ratios ($^{13}C/^{12}C$ and $^{15}N/^{14}N$) (Papanicolaou et al. 2003). Sources of sediment include, but are not limited to, upland land use and land cover (such as agriculture and forest) and the channel corridor (stream banks). By comparing the fingerprints of the suspended sediment samples to the fingerprints of the source samples and by using a statistical unmixing model, the sources of the suspended sediment can be apportioned.

3.2 Collection and analysis of fluvial and source samples

Fine-grained suspended sediment samples were collected at the USGS streamflow gauging station during storm events in Linganore Creek watershed from 1 August 2008 to 31 December 2010 by an automatic sampler that was programmed to activate when water levels in the stream exceeded a set gauge height. When possible, suspended sediment was also collected during storm events using manual isokinetic samplers (US DH-81 and US D-74). During the study period, 194 suspended sediment samples over 36 separate storm events were sampled for fine-grained source analysis (Electronic Supplementary Material Table 1).

Sediment source samples were collected from upland source areas and stream banks (40 from stream banks, 24 from agricultural areas, and 19 from forested sites) (Fig. 2). Site selection for forest and agricultural upland samples were based on obtaining a spatially representative dataset and cooperative landowner permission. Stream bank sites selection was based on a randomized design for each stream order using

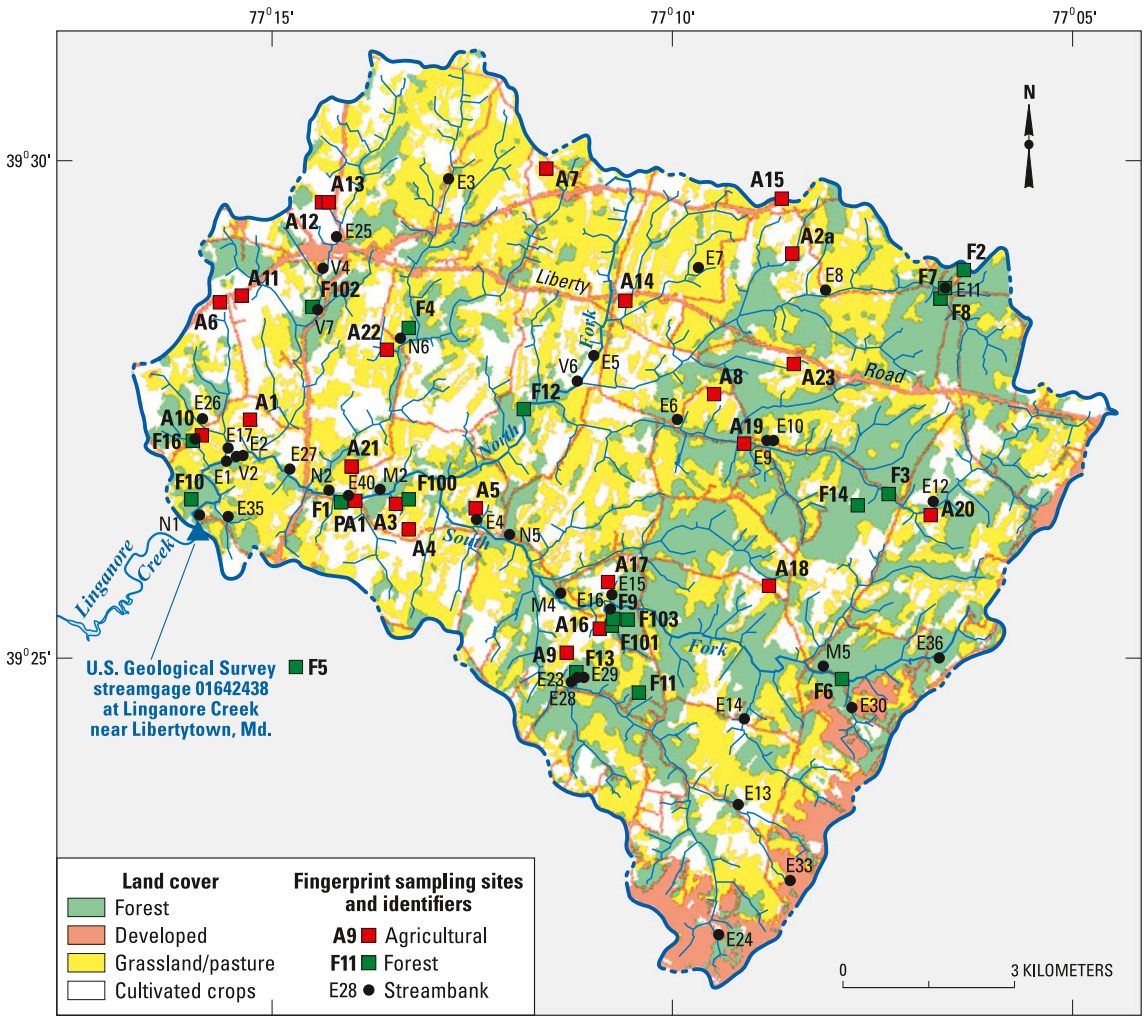

Fig. 2 Location of sampling sites for sediment fingerprinting analysis, Linganore Creek watershed

a geographic information system and a field reconnaissance of the watershed. As part of the sediment budget portion of the project, resurveys of the channel in Linganore Creek between 2008 and 2010 did not show that the channel was incising, and therefore, the beds of the channels were not considered as a sediment source. After discussions with local landowners, it was determined that cropland and pasture fields transition over time, and sites that are now in permanent pasture were cropland within the last two decades; therefore, cropland and pasture were combined into one sediment source category—agriculture.

Soil samples for source analysis from agriculture and forested areas were collected from the top ~1.0 cm of the soil surface with a plastic hand shovel. To account for variability in the tracer properties at agriculture and forested sites, sediment was collected across transects (100 by 30 m) and composited into one sample for each site. To obtain a representative sample of the stream banks, the surface of the exposed stream banks (~1 cm) were sampled vertically from the bottom to the top of the bank face. Three to five transects spaced 10 m apart along the stream reach were sampled and composited into one sample.

3.3 Laboratory analyses

Fluvial samples were centrifuged (1–4 l) at the USGS Atlanta Sediment Laboratory facility. Because we were interested in determining the sources of fine-grained sediment and most of the tracer's activity is found on the fine material, sand was removed from the samples by wet sieving them through a 63-μm polyester sieve. Because of mass constraints in the laboratory analysis of sediment, selected storm samples taken across the hydrograph were combined. Source samples collected from agriculture, forest, and stream banks were taken to the USGS laboratory at Baltimore, MD, dried at 60 °C, disaggregated with a mortar and pestle, wet-sieved with deionized water through a 63-μm polyester sieve to remove the sand, and dried again at 60 °C.

The silt and clay portion (<63 μm) of fluvial, upland, and stream bank samples were sent to the USGS-National Research Program (NRP) laboratory in Reston, VA for elemental and stable isotope analyses (Table 1). At the USGS-NRP laboratory, the samples were analyzed for 19 elements (see Table 1). All samples were analyzed using inductively coupled plasma combined with optical emission spectrometry (ICP-OES) and inductively coupled plasma combined with mass spectrometry (ICP-MS) after microwave-assisted multi-acid digestion (a mixture of nitric and hydrofluoric acids). Standard reference soil (NIST2709, San Joaquin Soil) and blanks were digested in each round of microwave digestion. The average blank value was subtracted from all sediment and reference samples for each element. Only elements with measured concentrations within 10% of the published value for NIST2709 are reported. Specific details regarding this method can be found in the paper of Noe and Hupp (2005).

Table 1 List of elements used as tracers to identify the sources of fine-grained sediment

Tracer analyzed from ICP-OES and ICP-MS analyses			
Boron (B) (μg g^{-1})	Copper (Cu) (μg g^{-1})	Lead (Pb) (μg g^{-1})	Aluminum (Al) (mg g^{-1})
Vanadium (V) (μg g^{-1})	Cadmium (Cd) (μg g^{-1})	Lithium (Li) (μg g^{-1})	Iron (Fe) (mg g^{-1})
Manganese (Mn) (μg g^{-1})	Molybdenum (Mo) (μg g^{-1})	Calcium (Ca) (mg g^{-1})	Phosphorus (P) (mg g^{-1})
Cobalt (Co) (μg g^{-1})	Antimony (Sb) (μg g^{-1})	Magnesium (Mg) (mg g^{-1})	Titanium (Ti) (mg g^{-1})
Nickel (Ni) (μg g^{-1})	Arsenic (As) (μg g^{-1})	Potassium (K) (mg g^{-1})	
Tracer analyzed from stable isotope analysis			
δ^{13}C (‰)	TOC (%)	Nitrogen (N) (%)	δ^{15}N (‰)

$\delta^{13}C$ stable isotopic total carbon-13, *TOC* total organic carbon, *$\delta^{15}N$* stable isotopic nitrogen-15

Samples were analyzed for the ratios of the stable isotopes of carbon and nitrogen ($^{13}C/^{12}C$ and $^{15}N/^{14}N$) and total carbon and nitrogen (TC and TN) at the Reston Stable Isotope Laboratory (RSIL) of the USGS, Reston, VA (Table 1). Samples were first processed to remove inorganic carbon using HCl vapor digestion (Hedges and Stern 1984), thus sample total organic carbon (TOC) was measured. Methods for analysis are described in the RSIL web site http://www.isotopes.usgs.gov (accessed September 2007). The relative carbon isotopic results for the sample, δ^{13}C, are reported in per mill relative to Vienna Pee Dee Belemnite and normalized on a scale such that the relative carbon isotope ratios of L-SVEC Li_2CO_3 (lithium carbonate reference material prepared by H. Svec, Iowa State University) and NBS 19 $CaCO_3$ (National Bureau of Standards Reference Material 19 for calcium carbonate) are −46.6 and +1.95‰, respectively (Coplen et al. 2006). Nitrogen isotope ratios, δ^{15}N, also reported in per mill, are expressed relative to N_2 in air using several internationally distributed isotopic reference materials, as discussed by Révész and Qi (2006). The 2σ uncertainty for both δ^{13}C and δ^{15}N analyses is ±0.50‰. The total number of tracers analyzed from the USGS laboratory was 23.

The size distribution of the <63 μm sample was determined at the USGS-NRP laboratory in Reston, VA, using a laser in situ scattering transmissometer (LISST-100X with mixing chamber) (Wolf et al. 2011). A mass of 20 mg was used for size analysis. Median values (D_{50}) of the sediment that are <63 μm were obtained from the LISST output data.

3.4 Statistical analysis

Several analytical and statistical steps were used to determine which tracers were most significant in defining sediment sources (α =0.05; Fig. 3).

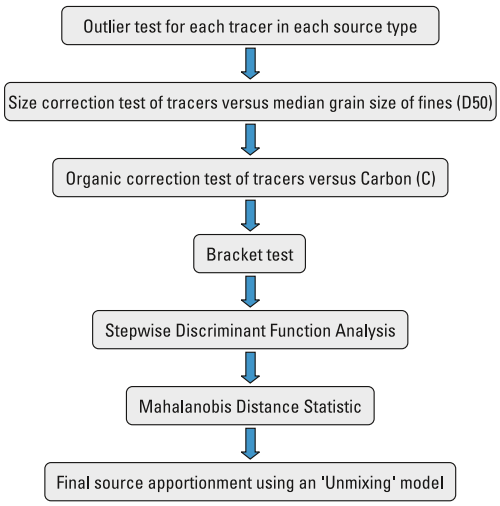

Fig. 3 Outline of statistical operations used in determining sediment sources using the sediment fingerprinting approach

3.4.1 Outlier test

The first step in the statistical procedure was to remove outliers. In each source group, each tracer was tested to determine if it had a normal distribution using the Shapiro–Wilk test (Ho=samples are random and come from a normal distribution). All variables that were not normally distributed were tested again for normality after transformation using log, power, square root, cube root, inverse, and inverse square root functions (Helsel and Hirsch 1997). The best transformation for normality was selected, and the tracers were transformed. The average and standard deviation within each source group for each transformed tracer was determined. If the tracer value for a given source sample exceeded three times the standard deviation of the average value, this sample was considered an outlier and the entire sample was removed for all 23 tracers (Wainer 1976). Outliers were determined for transformed tracers in each source type and indicated that two stream bank samples were outliers (Electronic Supplementary Material Tables 2, 3, and 4) and were removed.

3.4.2 Correcting source tracers for sediment size and organic content variability

The property of a sediment tracer not only depends on source material but on grain size and organic content (Collins et al. 2010; Horowitz 1991). The finer grain sizes have potentially more area to sorb constituents and have a higher tracer concentration. Conversely, iron oxides that develop on coarser sediment in the silt range may also contain more sites for constituents to sorb onto and can have higher concentrations. Organic matter on sediment can also become sites for sorption of tracers or include tracer elements in organic molecules.

To assure that tracers from sediment sources are comparable to tracers in fluvial sediment, based on grain size and organic content, a regression analysis was performed to determine if a tracer's property in each source group was significantly related to grain size and/or organic content. This test was developed with assistance from Professor Desmond Walling, University of Exeter, UK (D.E. Walling, 2012, personal communication) and represents a new approach for correcting for grain size and organic content differences between fluvial and source samples. For each source group, linear regression was used to determine if the relation of D_{50} or TOC to a given tracer's concentration was significant. An example of how the correction was applied is shown in Fig. 4. The statistical program Statistical Analysis System (SAS) was used in the regression analysis (SAS Institute 1985)[1].

Guidelines used to determine if the relation of D_{50} or TOC to a given tracer's concentration is significant included determining that the slope of the regression line is significant. If the slope of the regression was found to be significant, the residuals of the regression equation were tested to see if they are independent and followed a normal distribution. The Shapiro–Wilk test (Ho=samples are random and come from a normal distribution) along with plots of residuals versus predicted values and histograms of the residuals were used to determine if the regression model was reasonable (Helsel and Hirsch 1997).

The steps used to determine the best regression model were as follows: (1) determine if the relation of untransformed D_{50} and TOC versus each source group's tracer concentration is significant; (2) if untransformed D_{50} and TOC did not show a significant relation to the select tracers, then D_{50} and TOC were transformed using a log, power, square root, cube root, inverse, and inverse square root function; the transformed D_{50} and TOC were then regressed against each source group's tracer concentration to find the best regression model; (3) if after this step, select tracers did not show a significant statistical relation to D_{50} or TOC, then a source group's tracer was transformed using a log, power, square root, cube root, inverse, and inverse square root. The transformed tracers in each source group were regressed against all possible combinations of transformed D_{50} and TOC (including untransformed) and the optimum regression model was selected. If no relation was found with D_{50} or TOC, a correction factor was not applied. The tracers, δ^{13}C, δ^{15}N, and %N, are affected by the relative proportions of different kinds of organic matter in the sample, not the total organic matter content (C. Kendall, USGS, 2013, personal communication). The tracers, δ^{13}C, δ^{15}N, and N, were not corrected by TOC.

If the regression model of a source group's tracer versus D_{50} or TOC was determined to be significant, then a correction factor was applied to the tracer as follows:

[1] "Any use of trade, firm, or product names is for descriptive purposes only and does not imply endorsement by the US Government."

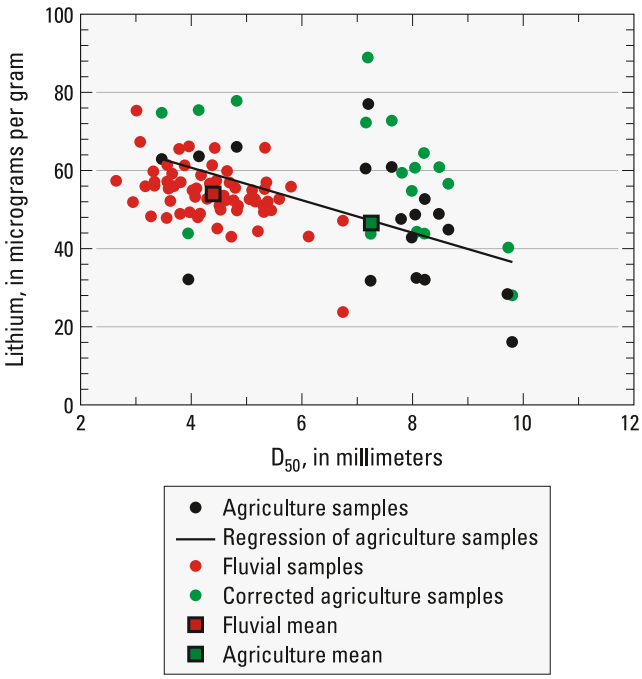

Fig. 4 Example of how the size correction factor is applied to a source group. In this example, the D_{50} from agriculture samples was regressed against the tracer lithium (Li). The line of best fit of agriculture D_{50} and agriculture Li is negative. The concentration of Li in the agriculture samples decreases as D_{50} increases. The average of D_{50} of fluvial samples is finer than the average D_{50} of agriculture samples. To correct for the differences in size, Li should be increased

$$C_n = \{Ti_i - ([(CORR_n - CORR_z) \times m]\}^{\wedge} \quad (1)$$

where C_n is the untransformed tracer after particle size or organic correction, Ti_i is the original value of tracer (i) (normalized) in source group n, $CORR_n$ is the D_{50} or TOC of the source samples (if necessary transformed for normality), $CORR_z$ is the mean D_{50} or mean TOC of fluvial samples (if necessary transformed by the same transformation for normality as the source D_{50} or TOC samples), m is the slope of the regression line of the tracers (if necessary transformed for normality) in source group n, versus D_{50} or TOC (if necessary transformed for normality) in source group n, and ^ denotes that if the tracer is normalized by a transform, the final corrected tracer is untransformed. When tracers were transformed and then corrected using Eq. 1, a bias correction factor was applied to the untransformed tracer values (Stuart and Ord 1991).

3.4.3 Bracket test

A requirement of sediment fingerprinting is that the fluvial tracers must be conservative and not change during transport from the source to the sampling point. Consequently, the next step in the statistical analysis was determining that, for a given tracer, the fluvial samples were within the range of the equivalent values obtained for the potential sources (Gellis and Walling 2011) (Fig. 3). The bracket test is an important prerequisite before further statistical analyses are performed. Any tracers that did not satisfy this constraint within measurement error (10% of each fluvial sample's tracer value) were considered to be nonconservative and were removed from further consideration. The bracketing test was performed on tracers after the particle size and organic correction factors were applied.

3.4.4 Stepwise discriminant function analysis

Collins et al. (1997) and Collins and Walling (2002) have suggested that a composite of several tracers provides a greater ability to discriminate between sources than a single tracer. To create the optimal group of tracers that differentiate the source groups, a stepwise discriminant function analysis (DFA) was used to select tracers after size and organic corrections were applied (Fig. 3). This procedure assumes normality among the variables being analyzed; thus, all variables used in the DFA were tested for normality using the Shapiro–Wilk test (Ho=samples are random and come from a normal distribution). All variables that were not normally distributed were tested again for normality after transformation using a log, power, square root, cube root, inverse, and inverse square root function (Helsel and Hirsch 1997).

The best transformation for normality was selected and stepwise DFA was performed on the data. Stepwise DFA incrementally identifies which tracers significantly contribute to correctly differentiating the sediment sources and rejects variables that do not contribute based on the minimization of the computed value of the variable Wilks' λ (Collins et al. 1997). A λ close to 1.0 indicates that the means of all tracers chosen are equal and cannot be distinguished among groups. A λ close to 0 occurs when any two groups are well separated (within-group variability is small compared to overall variability). Thus, the model selects a combination of tracers that provide optimal separation—no better separation can be achieved using fewer or more tracers. The SAS statistical program was used in stepwise DFA (SAS Institute 1985).

Another requirement of sediment fingerprinting is that the final set of tracers determined from stepwise DFA can correctly differentiate each source type. The Mahalanobis distance is a measure of the distance between groups and takes into account the covariance among the variables in calculating distances (De Maesschalck et al. 2000). The Mahalanobis distance has been used in other sediment source studies (Karlin 1980; Minella et al. 2008; Poulenard et al. 2009) and was used in this analysis to verify that the set of transformed tracers determined from stepwise DFA can correctly identify between each source type (Fig. 3).

3.4.5 Identify source percentages

The final step in the statistical analysis was determining the percentage contribution of each significant source of sediment using an "unmixing model" modified from Collins et al. (2010) (Eqs. 2 and 3; Fig. 3), as follows:

$$\sum_{i=1}^{n}\left\{\left[C_i-\left(\sum_{s=1}^{m}P_sS_{si}\right)\right]/C_i\right\}^2 W_i \quad (2)$$

and

$$\sum_{s=1}^{n}P_s=1 \quad (3)$$

where C_i is the concentration of tracer property (i) in the suspended sediment collected during storm events; Ps is the optimized percentage contribution from source category (s); S_{si} is the mean concentration of tracer property (i) after size and TOC correction factors are applied in source category (s); W_i is the tracer discriminatory weighting; n is the number of fingerprint properties comprising the optimum composite fingerprint; and m is the number of sediment source categories.

Collins et al. (2010) applied the particle size and organic correction factors directly in the unmixing model. In this modified version (Eq. 2), the set of tracer values that are determined from the stepwise DFA are used in the unmixing model with the particle size and organic correction factors applied. The unmixing model uses the untransformed data. The unmixing model was written in the programming language Python.

The unmixing model iteratively tests for the lowest error value using all possible source percentage combinations. A P_s step of 0.01 is used in the source computations. The tracer discriminatory weighting value, W_i, is a weighting used to reflect tracer discriminatory power (Eq. 2) (Collins et al. 2010). This weighting is based on the relative discriminatory power of each individual tracer provided by the results of the stepwise DFA. This weighting ensures that tracers with greater discriminatory power are optimized in the unmixing model solutions. The weighting for each tracer that passed the stepwise DFA test was determined as follows:

$$W_i=\frac{P_i}{P_{opt}} \quad (4)$$

where W_i is the tracer discriminatory weighting for tracer i, P_i is the percentage of source type samples classified correctly using tracer i, and P_{opt} is the tracer that has the lowest percentage of sample classified correctly. The percentage of source type samples classified correctly is a standard output from the DFA statistical results. Thus, a W_i value of 1.0 has low power of discriminating samples.

Sediment apportioned to stream banks, agriculture, and forest was determined for each sample, for the sampled storm event, and for the entire study period. Source percentages are presented for each storm and for the entire study period.

The source percentages are weighted first by the sediment load of each storm event (storm-weighted) and then by the total sediment load transported by all events (total load weighted) in the sediment fingerprinting analysis. (1) The source percentages determined for each storm event are weighted by the sediment load transported for the time interval ascribed to each sample and divided by the total sediment load of that storm. (2) The source percentages determined for the entire study period are weighted by the sediment load transported by each storm and divided by the total sediment load for all events that were analyzed for source analysis. A similar approach to weighting the individual sediment fingerprinting source results by the total sediment load was used by Walling et al. (1999). Suspended sediment loads used in the weighting of source percentages were computed using the USGS program Graphical Constituent Loading Analysis System (Koltun et al. 2006).

3.5 Uncertainty in the sediment fingerprinting approach

A Monte Carlo approach was used to quantify the uncertainty in the sediment fingerprinting results produced by the unmixing

Table 2 Results of test to determine if regression of the median grain size of material <63 μm (D_{50}) versus tracer activity in source samples is significant

Summary of regression analysis used for size correction factor

Sediment source type	Tracer	Transformation for normality for D_{50}	Transformation for normality of tracer	Slope of line of best fit	p value for slope of regression line	R^2	p value of normality of residuals	Correction factor
Agriculture	Li	No transformation	No transformation	−4.14	0.04	0.24	0.93	−11.9
Stream banks	B	No transformation	Inverse	−0.0004	0.03	0.21	0.32	−0.0018
Stream banks	Co	Inverse	No transformation	−48.1	0.04	0.19	0.79	−0.0039
Stream banks	P	Inverse	Cube root	−0.86	0.04	0.18	0.64	−0.0008
Stream banks	Sb	Squared	No transformation	0.0016	0.05	0.17	0.77	0.093
Forest	Ni	Inverse	Squared	916	0.05	0.50	0.74	125

Results are shown for normalized data. Slope of the regression line for the normalized variables is shown for significance $p<0.05$

Table 3 Results of test to determine if regression of carbon versus tracer activity in source samples is significant

Summary of regression analysis used for organic-correction factor

Sediment source type	Tracer	Transformation for normality of carbon	Transformation for normality of tracer	Slope of line of best fit	p value for slope of regression line	R^2	p value of normality of residuals	Correction factor
Agriculture	Co	No transformation	No transformation	−3.02	0.04	0.17	0.68	−2.32
Agriculture	Fe	No transformation	No transformation	−6.48	0.03	0.19	0.37	−4.98
Agriculture	Li	No transformation	No transformation	4.91	0.04	0.18	0.82	3.77
Agriculture	Mg	Inverse	Inverse square root	−0.58	0.03	0.19	0.91	0.033
Agriculture	Pb	No transformation	No transformation	4.79	0.02	0.22	0.48	3.68
Agriculture	Ti	No transformation	No transformation	−0.85	0.04	0.17	0.30	−0.651
Agriculture	V	No transformation	No transformation	−16.8	0.03	0.20	0.46	−12.9
Banks	P	No transformation	Log	0.070	0.02	0.14	0.96	−0.098
Banks	Pb	No transformation	No transformation	4.62	0.04	0.12	0.22	−6.50
Banks	Sb	No transformation	No transformation	0.144	0	0.27	0.28	−0.202
Banks	Ti	No transformation	No transformation	−0.55	0.04	0.11	0.44	0.773
Banks	V	No transformation	Inverse	0.0008	0	0.21	0.64	−0.0011
Forest	B	No transformation	Inverse	−0.0012	0.04	0.22	0.57	−0.0054
Forest	Fe	Log	No transformation	−44.2	0.05	0.21	0.77	−16.7
Forest	Mg	No transformation	Log	−0.075	0.05	0.21	0.61	−0.331
Forest	Ti	Log	Log	−0.23	0.01	0.31	0.44	0.314

Slope of the regression line for the normalized variables is shown for significance $p<0.05$. Results are shown for normalized data. Carbon in stream bank samples was log transformed

model (Collins and Walling 2007). A Monte Carlo simulation was written in the programming language Python. The Monte Carlo simulation randomly removes one sample from each of the three source type groups and the unmixing model is run without these samples. The Monte Carlo simulation is run 1,000 times on each fluvial sample. For each of the 1,000 iterations, the average and the minimum and maximum sediment source percentage for each source are determined. For each of the fluvial samples, the difference between the final unmixing model sediment source percentage results and the average of the 1,000 Monte Carlo sediment source percentage results and the minimum and maximum source percentage results produced by the Monte Carlo simulation are used to assess the robustness of the final set of source samples and tracers.

4 Sediment fingerprinting: results and discussion

Total suspended sediment transported during the study period (1 August 2008 through 31 December 2010) at Linganore Creek was 15,840 or 6,374 Mg year^{-1} of which 5,454 Mg year^{-1} was fine-grained sediment. The annual sediment yield was 43.4 Mg km^{-2} year^{-1} and is within the range of sediment yields reported for the Piedmont Physiographic Province of the Chesapeake Bay (Gellis et al. 2005). The suspended sediment load for the 36 storm events, which included sediment >63 μm, totaled 12,070 Mg which is 76% of the total suspended sediment load transported at the Linganore Creek near Libertytown, MD.

Plots of grain size (D_{50}) versus tracer concentration indicated a significant relation for Li in agricultural samples; B, Co, P, and Sb in stream bank samples; and Ni in forest samples. A correction factor was applied to these tracers (Table 2). Plots of TOC versus tracer concentration indicated a significant relation for Co, Fe, Li, Mg, Pb, Ti, and V in agricultural samples; P, Pb, Sb, Ti, and V in stream banks samples; and B, Fe, Mg, and Ti in forest samples. A correction factor was applied to these tracers (Table 3).

Results of the bracketing test indicated that the fluvial samples were not bracketed by 10 tracers, and these were removed (Table 4). Thirteen tracers were used for stepwise DFA (Al, C, δ^{13}C, Cu, Fe, Li, Mg, Mn, N, Ni, P, Pb, and V). Stepwise DFA was applied to the transformed set of tracers for all source types and showed that six tracers were significant (C, N, δ^{13}C, V, Cu, and Li) (Tables 4 and 5). Results of the Mahalanobis distance test indicated that this final set of six tracers was able to discriminate between all three source types (stream banks, agriculture, and forest) with a $p<0.05$. The DFA with six tracers successfully classified 93% of the source samples to their correct source category.

Tracer discriminatory weightings were applied to the six tracers (Table 4) and the unmixing model was run for each fluvial sample (Table 6; Electronic Supplementary Material Table 5). Results of the unmixing model, which are weighted

Table 4 Statistical tests (bracket test, normality tests, and stepwise DFA) performed on tracers for all source types after a size and organic correction were applied

Tracer	Bracket test result	Test for normality before stepwise DFA	Stepwise DFA[a]	Percent source type samples classified correctly by tracer	Cumulative source type samples classified correctly by tracer	Tracer discriminatory weighting (W_i; Eq. 2)
Al	PASS	cubrt				
As	FAIL					
B	FAIL					
C	PASS	log	Step 1 (0.0001)	84	84	1.8
C13	PASS	sqrt	Step 3 (0.0001)	63	91	1.5
Ca	FAIL					
Cd	FAIL					
Co	FAIL					
Cu	PASS	invsqrt	Step 5 (0.0001)	47	91	1.0
Fe	PASS	log				
K	FAIL					
Li	PASS	no trans	Step 6 (0.0001)	51	93	1.1
Mg	PASS	log				
Mn	PASS	cubrt				
Mo	FAIL					
N	PASS	log	Step 2 (0.0001)	76	88	1.8
N15	FAIL					
Ni	PASS	log				
P	PASS	no trans				
Pb	PASS	cubrt				
Sb	FAIL					
Ti	FAIL					
V	PASS	invsqrt	Step 4 (0.0001)	53	91	1.1

Transformed tracers were used in the stepwise DFA

no trans no transformation needed for normality, *log* log transformation, *inv* inverse transformation, *cubrt* cube root transformation

[a] Results shown for stepwise DFA include the significant steps in forward stepwise selection and p values<0.05

by the sediment load of each sample and the sediment load of each event, show stream banks as the major source of sediment (53%), followed by agriculture (44%) and forest (3%) (Fig. 5; Table 6).

4.1 Relation of sediment sources to flow and season

Peak flow (or peak discharge) of an event is an indication of the strength of a storm event and is a function of the amount and intensity of precipitation and runoff generated to the stream (Rankl 2004). In streams studied in Puerto Rico (Gellis 2013), peak flow was significant in explaining storm-generated suspended sediment loads and concentrations. In Wyoming, USA, peak flow was significantly correlated to storm event sediment loads (Rankl 2004). In Linganore Creek, peak flow for each event (Electronic Supplementary Material Table 5) shows a significant relation to the event suspended sediment load (Fig. 6a). Plots of storm-weighted contributions

Table 5 Average and standard deviation for final six tracers used in sediment fingerprinting analysis

		δ^{13}C (‰)	C (%)	N (%)	V (μg g^{-1})	Cu (μg g^{-1})	Li (μg g^{-1})
Stream banks	Average	−25.67	1.50	0.17	132	46.3	48.7
	Standard deviation	0.71	0.76	0.07	24	16.6	9.8
Agriculture	Average	−25.44	3.67	0.38	133	44.5	46.6
	Standard deviation	1.54	1.24	0.11	47	18.0	14.5
Forest	Average	−27.59	7.33	0.51	98.4	38.0	40.4
	Standard deviation	0.44	2.66	0.16	24.2	12.1	13.9

Table 6 Unmixing model results for 36 storm events in Linganore Creek

		Source apportionment for each storm event determined by weighted the suspended sediment load of each sample by the suspended sediment load for that storm event and summed (storm-weighted)					Source apportionment for each storm event determined by weighted the suspended sediment load of each sample by the suspended sediment load for that storm event and summed (storm-weighted)		
Event no.	Date: sample times (month/day/year)	Stream banks	Agriculture	Forest	Event no.	Date: sample times (month/day/year)	Stream banks	Agriculture	Forest
1	9/6/2008:1236 to 9/7/2008:1233	50	50	0	19	8/28/2009:0449 to 8/28/2009:2343	36	56	8
2	9/13/2008:0045 to 9/13/2008:0415	6	94	0	20	10/17/2009:1132 to 10/18/2009:0433	49	51	0
3	9/28/2008:0030 to 9/28/2008:0603	30	70	0	21	10/24/2009:1626 to 10/24/2009:2327	37	52	11
4	9/30/2008:2115 to 10/1/2008:0248	4	96	0	22	10/27/2009:0848 to 10/28/2009:2153	54	42	3
5	10/25/2008:1900 to 10/26/2008:0933	14	77	9	23	11/24/2009:0600 to 11/24/2009:1231	22	40	38
6	11/15/2008:1529 to 11/16/2008:0733	0	73	27	24	12/3/2009:0330 to 12/3/2009:1231	11	70	19
7	11/30/2008:2345 to 12/01/2008:1540	39	33	27	25	12/9/2009:0330 to 12/10/2009:0708	56	41	3
8	12/11/2008:1515 to 12/13/2008:0422	42	58	0	26	12/13/2009:1700 to 12/13/2009:2200	34	62	4
9	12/19/2008:1400 to 12/20/2008:0433	24	73	3	27	12/26/2009:0030 to 12/27/2009:0211	77	18	5
10	1/7/2009:0845 to 1/8/2009:0343	50	50	0	28	1/17/2010:1430 to 1/18/2010:0131	40	37	24
11	4/3/2009:1200 to 4/3/2009:1800	16	84	0	29	1/25/2010:0730 to 1/26/2010:0040	71	26	4
12	4/20/2009:0945 to 4/23/2009:0442	57	43	0	30	2/6/2010:1245 to 2/6/2010:1745	65	0	35

Table 6 (continued)

Event no.	Date: sample times (month/day/year)	Source apportionment for each storm event determined by weighted the suspended sediment load of each sample by the suspended sediment load for that storm event and summed (storm-weighted)			Event no.	Date: sample times (month/day/year)	Source apportionment for each storm event determined by weighted the suspended sediment load of each sample by the suspended sediment load for that storm event and summed (storm-weighted)		
		Stream banks	Agriculture	Forest			Stream banks	Agriculture	Forest
13	5/3/2009:1730 to 5/5/2009:0820	41	57	2	31	3/12/2010:0900 to 3/12/2010:1213	43	57	0
14	5/28/2009:2000 to 5/28/2009:0703	2	98	0	32	3/13/2010:0900 to 3/14/2010:1101	65	34	1
15	6/4/2009:0200 to 6/4/2009:0400	26	74	0	33	3/29/2010:0215 to 3/29/2010:0715	19	81	0
16	6/18/2009:0915 to 6/19/2009:1319	15	84	0	34	4/25/2010:2115 to 4/26/2010:0816	36	59	4
17	7/22/2009:0115 to 7/22/2009:0415	23	77	0	35	8/12/2010:0645 to 8/14/2010:0340	51	47	2
18	7/23/2009:2015 to 7/24/2009:1100	19	80	1	36	9/30/2010:0800 to 10/1/2010:1220	31	68	0
	Source apportionment for all storm events determined by weighting the suspended sediment load of each storm event by the suspended sediment loads for all events and summed (total load weighted)	53	44	3					

Storm event results are averages of individual samples collected on the storm hydrograph (Electronic Supplementary Material Table 5)

from stream banks versus peak flow for the 36 storm events indicate that, as peak flow increases, stream bank contributions increase (Fig. 6c). The increase in contribution from stream banks with higher peak flow indicates that stream banks may have been eroded during these events (discussed below). Contributions from storm-weighted source percentages from agriculture versus peak flow for the 36 storm events show that, as peak flow increases, the source contributions from agriculture decrease (Fig. 6b). The decrease in agricultural sediment with increasing peak flows may reflect the greater contributions from stream banks relative to agricultural sources during periods of high flow (discussed below). Storm-weighted source percentages from forested areas did not show a relation to peak flow (Fig. 6d).

Plots of storm-generated suspended sediment loads versus storm-weighted source percentages show similar results to peak flow (Fig. 7). As storm-generated suspended sediment loads increase, contributions from stream banks increase and contributions from agriculture decrease (Fig. 7). Forest contributions do not show a strong correlation to storm-generated suspended sediment loads (Fig. 7).

The time of year may also be a factor in determining whether specific sediment sources may be important. Bare ground in agricultural areas is common before the growing season and after harvesting (Gellis et al. 2009). Wolman (1959) and Gatto (1995) have shown that, in winter months (December to February), freeze–thaw action is an important process in the erosion of stream banks. Plots of storm-weighted sediment percentages are shown by month and averaged by season: winter (December to February), spring (March to May), summer (June to August), and fall (September to November) (Fig. 8a–c). The highest contributions from

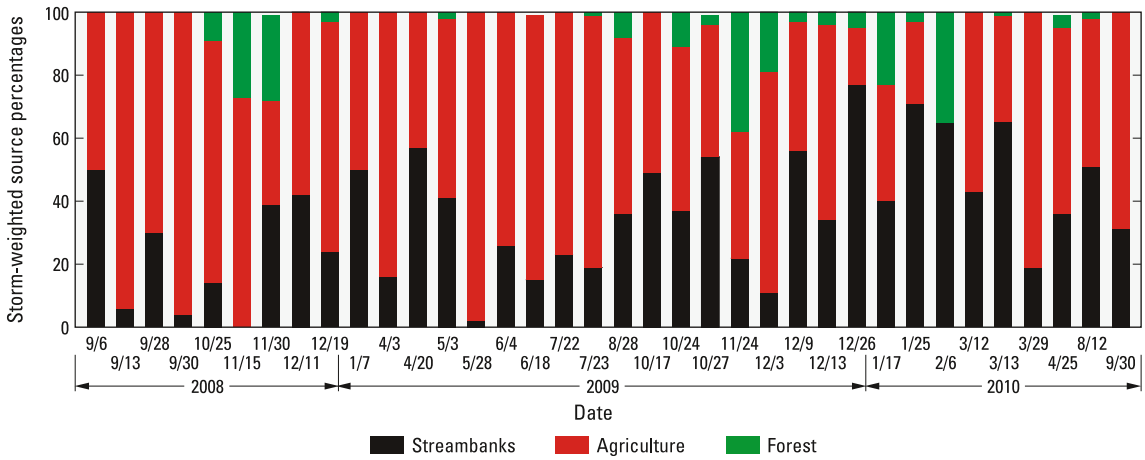

Fig. 5 Results of sediment fingerprinting analysis for 36 storm events in Linganore Creek, 1 August 2008 to 31 December 2010. Each storm's source percentages are storm-weighted where the sediment load of each sample in an event is weighted by the total sediment load of that event. Dates are presented as month/day/year

stream banks occurred in winter months (47%) (Fig. 8a). A *t* test performed on seasonal differences of sediment derived from stream banks indicates that winter months are significantly higher than non-winter months (Fig. 8a).

Peak flow plotted against time of year show the highest peak flows in the winter months, with three of the highest peak flows occurring in December (Fig. 8d). The largest peak flow of the sampled events (55.5 $m^3 s^{-1}$) occurred on 26 December 2009 and contributed the highest storm-weighted percentage from stream banks for any event (average=77%). This event had rainfall totals of 88 mm, with a recurrence interval of 1 to 2 years (Electronic Supplementary Material Table 5). A *t* test performed on seasonal differences of peak flows indicates that peak flows are significantly higher in winter months than non-winter months (Fig. 8d).

In the three primary mechanisms of bank erosion—freeze–thaw processes, fluvial erosion, and mass wasting—peak flow plays an important role in eroding stream banks. Greater masses of sediment produced by these three mechanisms are removed as peak flow increases. Higher peak flows increase shear stress and erode stream banks and can be important when bank failures occur during streamflow recession and excess porewater pressure, following a high stage.

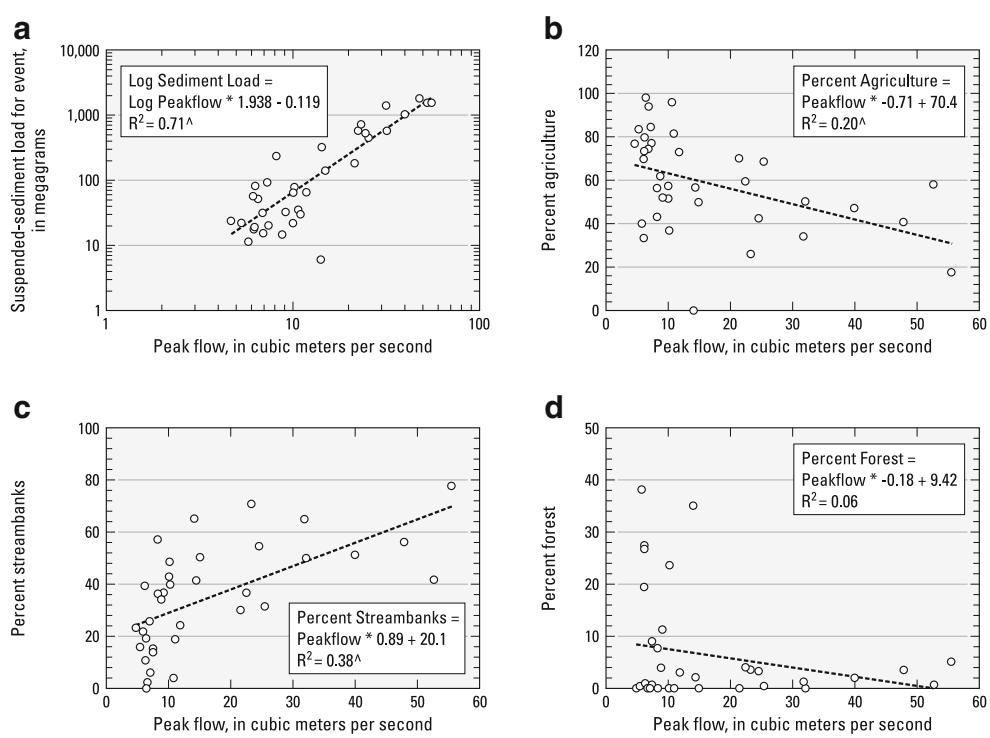

Fig. 6 **a** Relation of peak flow to suspended sediment load for sampled events. Relation of peak flow to storm-weighted source percentages for **b** agriculture, **c** stream banks, and **d** forest. ^$p<0.05$, the slope of the regression line is significant and the residuals are normally distributed

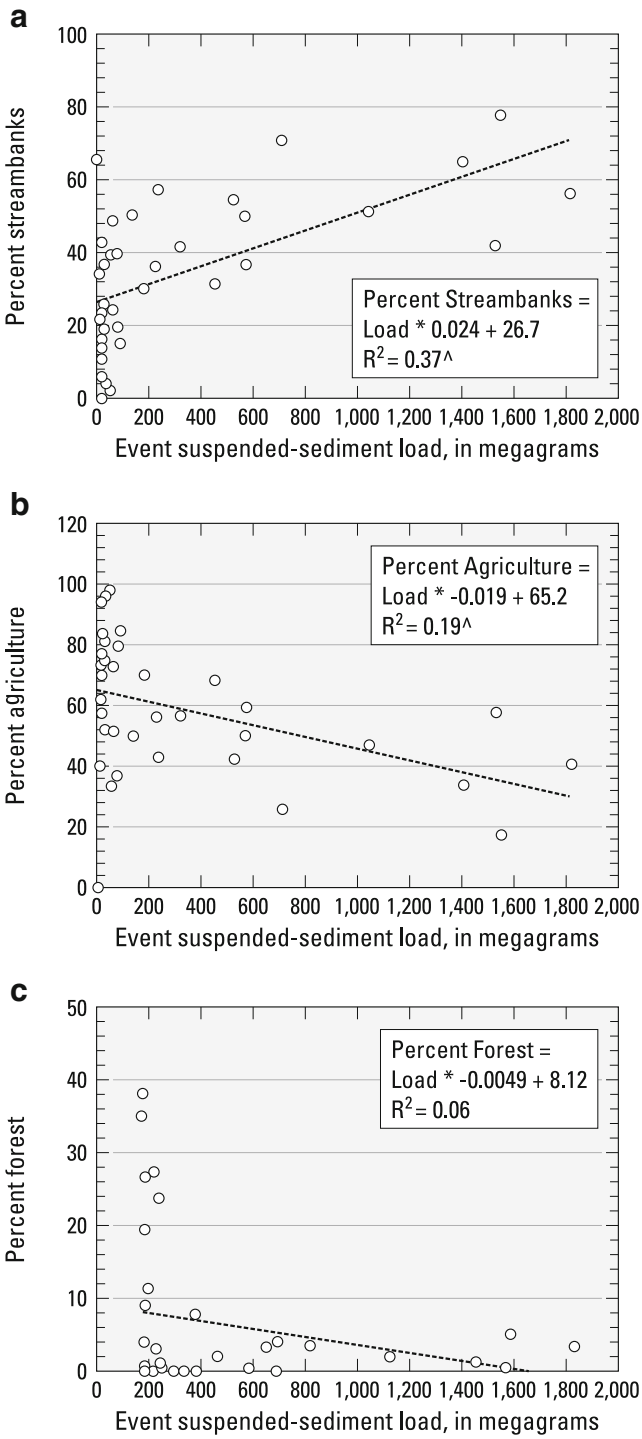

Fig. 7 Relation of suspended sediment load to storm-weighted source percentages for **a** stream banks, **b** agriculture, and **c** forest. ^$p<0.05$, the slope of the regression line is significant and the residuals are normally distributed

In Linganore Creek, late fall through early spring commonly have days when temperatures fall below and above freezing. Plots of source areas with season indicate that the highest storm-weighted percentage of bank erosion occurs in the winter, during December through February (average=47%) (Fig. 8a). Examination of climatic data for Frederick, MD indicates that all storms sampled in the winter had at least 1 day of freeze–thaw in the 5 days preceding the sampled event. Therefore, freeze–thaw days may be a contributing factor to stream bank erosion in Linganore Creek.

The lowest storm-weighted contributions of sediment from agriculture occurred in winter (43%) (Fig. 8b). A *t* test performed on seasonal differences of sediment derived from agriculture indicates that winter months are significantly lower than non-winter months (Fig. 8b). In Linganore Creek, the lower contributions from agriculture in the winter may reflect the higher contributions from bank erosion. The highest storm-weighted contributions from agriculture (>90%) occurred during three storms (event 14, 28 May 2009, 94%; event 4, 30 September to 1 October 2008, 96%; and event 2, 13 September 2008, 98%) (Electronic Supplementary Material Table 5). Events 2, 4, and 14 were moderate events where peak flows ranked 27, 17, and 29, respectively, and rainfall totals ranked 20, 22, and 28, respectively, where a rank of 1 has the highest value (Electronic Supplementary Material Table 5). Spring, summer, and fall conditions on agricultural lands range from bare ground associated with tillage in the spring to full vegetative cover in the summer to bare ground following harvest in the fall. Higher contributions from agriculture in spring and fall could reflect conditions when fields have less vegetation. In agricultural watersheds in Belgium, summer contributions of sediment were low due to the increased vegetative cover (Steegen et al. 1998). Vegetative cover consisted of land in cultivation (wheat, sugar beets, and corn), pasture, and woodlands. High-intensity thunderstorms occurring in the summer also have the ability to erode farm fields and transport agricultural sediment. In the Netherlands, soil loss in the summer was twice as high as winter soil loss and was caused by high-intensity rainfall events (Kwaad 1991). Intense summer thunderstorms were observed in Linganore Creek during the study period, but rainfall intensity was not measured.

The highest contribution from forests occurred in fall to winter months (10%) (Fig. 8c). Significant differences in the weighted sediment source contributions from forest occurred between winter–fall and spring–summer (see Fig. 8c). The five highest storm-weighted sediment contributions from forest (>20%) occurred during November, January, and February (events 23, 30, 7, 6, and 28; Electronic Supplementary Material Table 5). These events had moderate peak flows (ranked 34, 14, 33, 31, and 18, respectively) and moderate rainfall totals (ranked 25, missing data, 30, 10, and 24, respectively) (Electronic Supplementary Material Table 5). November, January, and February are months when leaf-off conditions typically occur in these deciduous forests, and it is possible that bare ground was eroded during these storms.

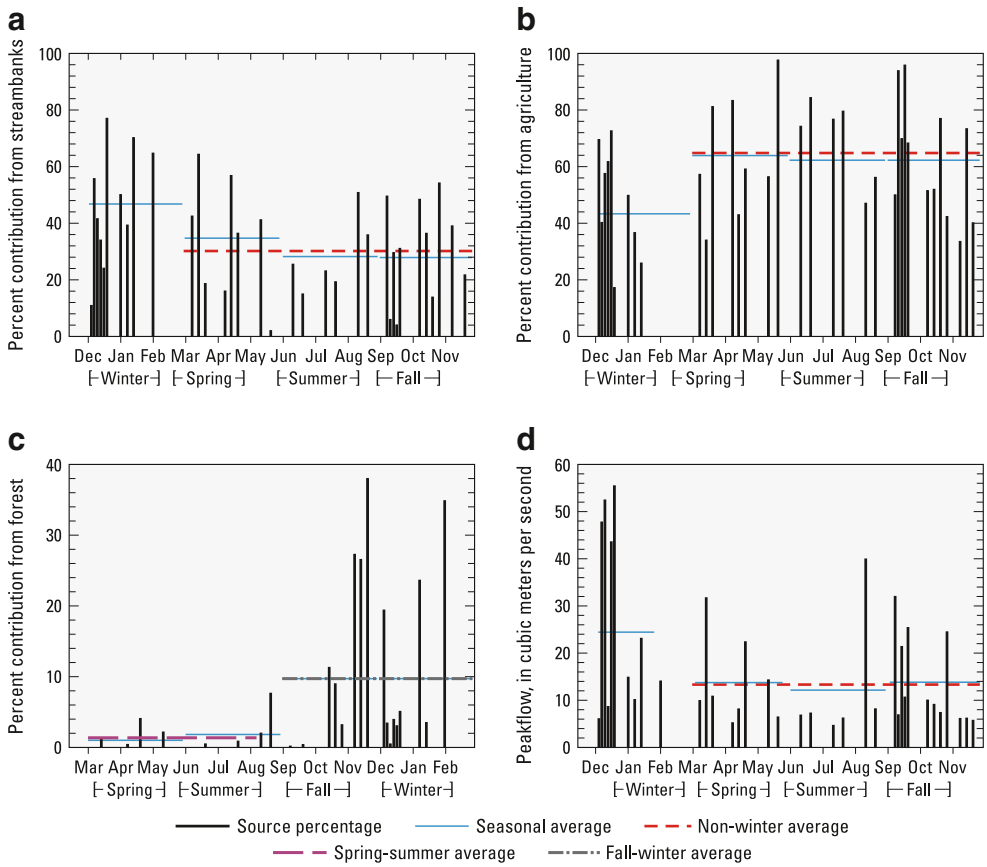

Fig. 8 Storm-weighted source percentages shown by months and season for **a** stream banks (results of a *t* test ($p=0.023$; data normally distributed) on the data indicated a statistical difference between winter and non-winter months); **b** agriculture (results of *t* test ($p=0.009$; data normally distributed) on the data indicated a statistical difference between winter and non-winter months); and **c** forest (results of a Mann–Whitney rank sum test ($p=0.031$; data non-normally distributed) on the data indicated a statistical difference between spring–summer and fall–winter months). **d** Peak flow by month and season (results of a Mann–Whitney rank sum test ($p=0.046$; data non-normally distributed) on the data indicated a statistical difference between winter and non-winter months)

4.2 Limitations and uncertainty

It should be noted that the sediment fingerprinting method quantifies the relative source percentage of sediment that are delivered to a point of interest in the watershed. Although the sediment fingerprinting approach provides sediment source percentages for a given storm, it should not be inferred that the sampled sediment was eroded, transported, and delivered in that event. While it is possible that some of the sampled samples could have been eroded, transported, and delivered in the sampled event, it is also likely that a portion of the sampled sediment may have been eroded in a previous storm(s) and been in storage for a period of time until it was remobilized and sampled in the select event (Gellis 2013). Therefore, the sediment fingerprinting method does not determine what the temporal aspects of the sourced sediment may have been. In addition, since sediment that is in storage may become remobilized over subsequent events, the source of the stored sediment and its erosion, transport, and deposition may be related to previous land management actions and storm events.

Because we do not know the history of when the sediment was eroded and where the sediment may have resided in storage (i.e., slopes, floodplains, channels), caution should be observed when trying to interpret how sediment sources relate to the flow of a given sample. A large percentage of sediment eroded from upland surfaces, such as agricultural lands, is not delivered directly to the channel but goes into storage. The sediment delivery ratio (SDR) is the amount of sediment delivered to a point in the channel from a given source(s) divided by the gross erosion of the source(s). The SDR for cropland and pasture in the York River watershed, Virginia (a tributary of the Chesapeake Bay) was 0.31 (Herman et al. 2003) and the SDR for cropland in the Little Conestoga Creek watershed, Pennsylvania (also a tributary to the Chesapeake Bay) was 0.20 (Gellis et al. 2009). A value of SDR<1 indicates that some of the eroded sediment went into storage. Therefore, it is likely that some of the agricultural sediment that is exported during storm events in Linganore Creek is upland sediment (e.g., agricultural) that was in storage.

Stream bank sediment is eroded directly into the channel during high flows and, therefore, has a high potential to be delivered to the sampling station during that event. In Luxembourg, stream banks had an SDR of 1.0, indicating that all of the sediment eroded from stream banks was delivered out of the watershed (Duijsings 1986). However, Skalak and Pizzuto (2010) describe the potential of sediment derived from stream banks in the South River, Virginia (a tributary to the Chesapeake Bay) to remain in channel storage behind large

woody debris up to decades, which would indicate an SDR< 1.0. Large amounts of sediment from all sources, both upland and stream banks, can also be trapped by floodplains for long time periods prior to sampling as suspended sediment at a gauge (Schenk et al. 2013). Compared to upland-derived sediment (agriculture and forest), stream bank-derived sediment has a greater potential to be eroded and delivered during an event due to fewer potential storage locations in transit, and it is likely that the sediment fingerprinting results for sediment derived from stream banks will show a significant relation to flow and seasonal conditions.

Sediment fingerprinting results will reflect sediment that was eroded and transported during the event as well as contributions from sediment that was in storage. The remobilized stored sediment will have conservative tracers that reflect its source origin. By examining sediment sources over several storms or seasons, it may be possible to determine the sediment from previous events that was in storage and remobilized.

Results of the Monte Carlo analysis indicates that all 194 suspended sediment samples had average source percentages within 2% of the final sediment fingerprinting results (Electronic Supplementary Material Table 5). For stream banks, the range of all Monte Carlo results is 8% higher and lower than the Monte Carlo average. For agriculture, the range of all Monte Carlo results is 13% higher and 12% lower than the Monte Carlo average. For forest, the range of all Monte Carlo results is 5% lower and 7% higher than the Monte Carlo average.

Since the majority of samples have sediment source differences between the average Monte Carlo results and the final set of fingerprinting results that are 2% or less, the unmixing model results shown for the final set of source samples are considered robust. The minimum and maximum differences around the average Monte Carlo source apportionment have close to 50% of the samples within 5% of the final fingerprinting results and the majority of samples are <10% of the final fingerprinting results. This small spread of the Monte Carlo results between minimum and maximum values confirms that the final set of tracers give reliable results.

5 Summary and conclusions

Fine-grained sediment is an important pollutant in the Chesapeake Bay. The USGS is engaged in studies to understand sediment processes in small watersheds of which the results from this study are an example. Linganore Creek, a tributary to Chesapeake Bay, is listed by the State of Maryland as being impaired by sediment. The upper portions of the watershed drain into Lake Linganore which is also threatened by sedimentation. Understanding the sources of sediment in the Linganore Creek watershed may assist management agencies in developing plans to reduce the input of sediment to the stream and receiving waters.

The sources of fine-grained sediment were examined for the 147-km^2 Linganore Creek watershed, Maryland between 1 August 2008 and 31 December 2010. During the period of study, 194 suspended sediment samples were collected over 36 separate storm events and used to determine the sources of sediment using the sediment fingerprinting approach. Sediment source percentages were weighted against sediment loads of the individual samples and then weighted against the total sediment transported by the 36 storm events. Results indicate that stream banks were the major source of sediment (53%), followed by agriculture (44%) and forested lands (3%). Uncertainty analysis indicated that the averages of the Monte Carlo source simulations are within 2% of the sediment fingerprinting results. The minimum and maximum values of source percentages produced from the Monte Carlo simulation are within 5 to 10% of the average value. Therefore, the final set of source samples and tracers used in this study was considered a robust dataset.

Peak flows and sediment loads for the 36 storm events show a significant positive relation to contributions from stream banks. Seasonality is also a factor in contributing sediment from stream banks. Winter months (December to February) had the greater contributions of sediment from stream banks. This may be due to the greater freeze–thaw activity that occurs in winter months and caused the banks to erode. Higher peak flows that occur in winter months would remove the eroded sediment. High peak flows may also increase bank erosion through increased shear stress and higher porewater pressures on the falling limb of large events.

Peak flows and suspended sediment loads were also significantly negatively correlated to the contribution of sediment from agriculture. Contributions from forests did not show any relation to flow. Contributions from agriculture were highest in non-winter months and those from forests were highest in the fall and winter. The higher contributions from agriculture and forests could be related to leaf-off conditions in early spring and fall.

Results of this study have implications to land management agencies interested in reducing sediment in Linganore Creek. The sediment fingerprinting results indicate that the two main sources of fine-grained sediment delivered out of the watershed were stream banks (53%) and agriculture (44%) and suggest that, to reduce sediment in Linganore Creek, both sources need to be addressed. A companion study that is focused on sediment budgets (erosion and deposition) will also be used to target which stream banks in the watershed are eroding. Combining the sediment fingerprinting results with the sediment budget results will also identify areas in the watershed where sediment storage has occurred. For example, rates of sediment trapping (in megagrams per square kilometer

per year) by floodplains of Linganore Creek are very large relative to watershed sediment yield (>100), even after accounting for stream bank erosion, indicating that floodplains are hot spots of sediment removal (Schenk et al. 2013).

This paper cautions researchers to be aware of over-interpreting sediment source results using the sediment fingerprinting approach. Attempting to link upland sediment sources to flow characteristics is difficult since much of the upland sediment that is eroded in an event may not be delivered to the sampling point in that event but may go into storage. Storage areas include channel and upland areas. Sediment from stream banks is delivered directly into the channel, is less likely to be stored prior to delivery to a stream suspended sediment sampling station, and has a better relation to flow characteristics. By analyzing sediment sources over several storms, it may be possible to determine not only the sediment sources that are directly contributed from the current event but also sediment from previous events that was in storage and remobilized. Since seasonal analysis incorporates many storms, the seasonal interpretations of sediment sources may be valid. Because the sediment sampled for the 36 storm events for this study occurred over several years and over different flow and seasonal conditions, it likely provides a robust long-term estimation of sediment sources in the watershed.

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