



Sediment Fingerprinting to Delineate Sources of Sediment in the Agricultural and Forested Smith Creek Watershed, Virginia, USA

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Research Impact Statement: Sediment fingerprinting helps identify and apportion sediment sources, including sediment derived from top soil and eroding streambanks, which are often overlooked sources of sediment to streams.

ABSTRACT: The sediment fingerprinting approach was used to apportion fine-grained sediment to cropland, pasture, forests, and streambanks in the agricultural and forested Smith Creek, watershed, Virginia. Smith Creek is a showcase study area in the Chesapeake Bay watershed, where management actions to reduce nutrients and sediment are being monitored. Analyses of suspended sediment at the downstream and upstream sampling sites indicated streambanks were the major source of sediment (76% downstream and 70% upstream). Current management strategies proposed to reduce sediment loadings for Smith Creek do not target streambanks as a source of sediment, whereas the results of this study indicate that management strategies to reduce sediment loads in Smith Creek may be effective if directed toward managing streambank erosion. The results of this study also highlight the utility of sediment fingerprinting as a management tool to identify sediment sources.

(KEYWORDS: sediment fingerprinting; bank erosion; Chesapeake Bay; sediment TMDL.)

INTRODUCTION

Worldwide, sediment is an important pollutant degrading aquatic habitat and impacting infrastructure, such as reservoirs (Strayer and Dudgeon 2010; Liu et al. 2017). In the United States (U.S.), sediment is one of the leading causes of stream impairment (USEPA 2017). Fine sediment can reduce light penetration and suppress primary production in algae and macrophytes (Yamada and Nakamura 2002; Izagirre et al. 2009; Jones et al. 2012). Deposited sediment can bury channel substrate and degrade habitat for macroinvertebrates (Jones et al. 2012) and fish (Sear et al. 2016). In addition, fine sediment provides a transport vector for bound nutrients, heavy metals,

and other contaminants (Owens et al. 2001; Gerbersdorf et al. 2011).

Sediment is a major contributor to ecological degradation in Chesapeake Bay (Gellis and Brakebill 2013). Smith Creek, along with two other streams in the Chesapeake Bay watershed, was selected by the U.S. Department of Agriculture as a “showcase” study area, meaning that if successfully restored, it would become a model for restoration efforts in the Chesapeake Bay watershed (Eppe 2010; Jenner 2010; USDA-NRCS 2017). Biological monitoring conducted by the Virginia Department of Environmental Quality indicated Smith Creek was violating the state’s general standard for aquatic life use where the stream should support the propagation and growth of a balanced indigenous population of aquatic life

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(VADEQ 2009). The primary stressor on the aquatic community was identified as sediment. In 2004, a Total Maximum Daily Load (TMDL) was developed for Smith Creek to reduce sediment loadings (VADEQ 2009). The successful mitigation of sediment-related impairments requires knowledge of the contribution of sediment from its various sources.

Attempting to relate sediment to its sources is a difficult task. Approaches for estimating sediment sources include sediment budget studies (Gellis and Walling 2011; Gellis et al. 2016), geographic information system (GIS) and photogrammetric analysis (Fernandez et al. 2003; Curtis et al. 2005; Roering et al. 2013; Hackney and Clayton 2015; Gellis et al. 2016), models (Aksoy and Cavvas 2005; USEPA 2008), and the use of geochemical tracers or fingerprints (Walling 2005; Gellis and Walling 2011; Mukundan et al. 2012; Collins et al. 2017). Each of these approaches has its advantages and disadvantages. Sediment budget approaches often rely on field measurements, which can provide useful data on erosion and deposition rates, but are labor intensive, and can be spatially limited. Photogrammetry/GIS analysis and model analysis, although less intensive in terms of labor and time, may produce a wide range of results that need to be validated with data collected from the watershed of interest. Ground-based and airborne lidar, as well as structure-from-motion photogrammetry with handheld cameras and unmanned aerial systems (drones), are being increasingly used to describe channel morphology and topographic change (Faux et al. 2009; Caroti et al. 2013; Roering et al. 2013; Caroti et al. 2015). The resultant scans, point clouds, and digital elevation maps can be overlain chronologically to quantify the erosion and deposition of various channel sources. However, these techniques often require field validation and the representation of morphological elements requires a high point density with large data processing demands. In addition, resolution and scale may restrict the applicability of these techniques to quantify topsoil erosion and ultimately do not quantify the delivery of these sediment sources out of the watershed.

Sediment fingerprinting is an approach that has been increasingly utilized to assist managers in identifying sources of sediment in a watershed (Collins, Walling, et al. 2010; Mukundan et al. 2012; Miller et al. 2015; Collins et al. 2017). The sediment fingerprinting approach entails the identification of specific sediment sources through the establishment of a minimal set of physical and (or) chemical properties that uniquely define each source in the watershed. In general, sediment fingerprinting results can provide information on the relative contribution of upland (soil erosion from various land use and land cover types) vs. channel contributions (streambanks and channel

beds) (Gellis and Walling 2011; Gellis et al. 2016). Differentiating between these two broad categories (upland and channel sources) is important because sediment-reduction management strategies differ by source and require very different approaches — reducing agricultural sources may involve soil conservation and tilling practices, whereas reducing channel sources of sediment may involve stream restoration, bank stabilization, and (or) grade control to arrest downcutting. The objective of this study was to identify the relative contributions of sediment from cropland, pasture, forest, and streambanks in the Smith Creek watershed using the sediment fingerprinting approach.

MATERIALS AND METHODS

Study Area

Smith Creek drains the Valley and Ridge Province in the Chesapeake Bay watershed, with land use in the area draining to the downstream station (in 2011) consisting of forest, 48%; pasture, 41%; developed, 8%; and cropland, 3% (Figure 1) (Homer et al. 2015). The area draining to the downstream station is underlain by dolostone and limestone (66%) and sandstone and shale (34%) (Dicken et al. 2005). Elevations range from 270 m at the lower reaches to 890 m in the Massanutten Mountains on the eastern side of the watershed. In the area draining to the upstream station, land use in 2011 was crop 2%; pasture 30%; forest 66%; and other 2% (Homer et al. 2015), and bedrock is 58% dolostone/limestone and 42% sandstone/shale (Dicken et al. 2005). Average annual daily discharge recorded at the U.S. Geological Survey (USGS) gage, 1961–2016, was 2.14 m³/s (USGS 2016). Precipitation in the watershed measured at the Dale Enterprise rain station near Harrisonburg, Virginia, averaged 915 mm/yr with temperatures ranging from a July mean of 23°C to a January mean of 0.44°C (University of North Carolina 2012). Most portions of Smith Creek are meandering, pool-riffle systems on gravel to sand beds with occasional bedrock outcrops. Varying thicknesses of sediment in channel storage (on the channel bed) were observed at select reaches, most noticeably in pools. Fine sediment was also present within the interstitial coarse substrate.

The Sediment Fingerprinting Approach

The sediment fingerprinting approach provides a direct method for quantifying watershed sources of

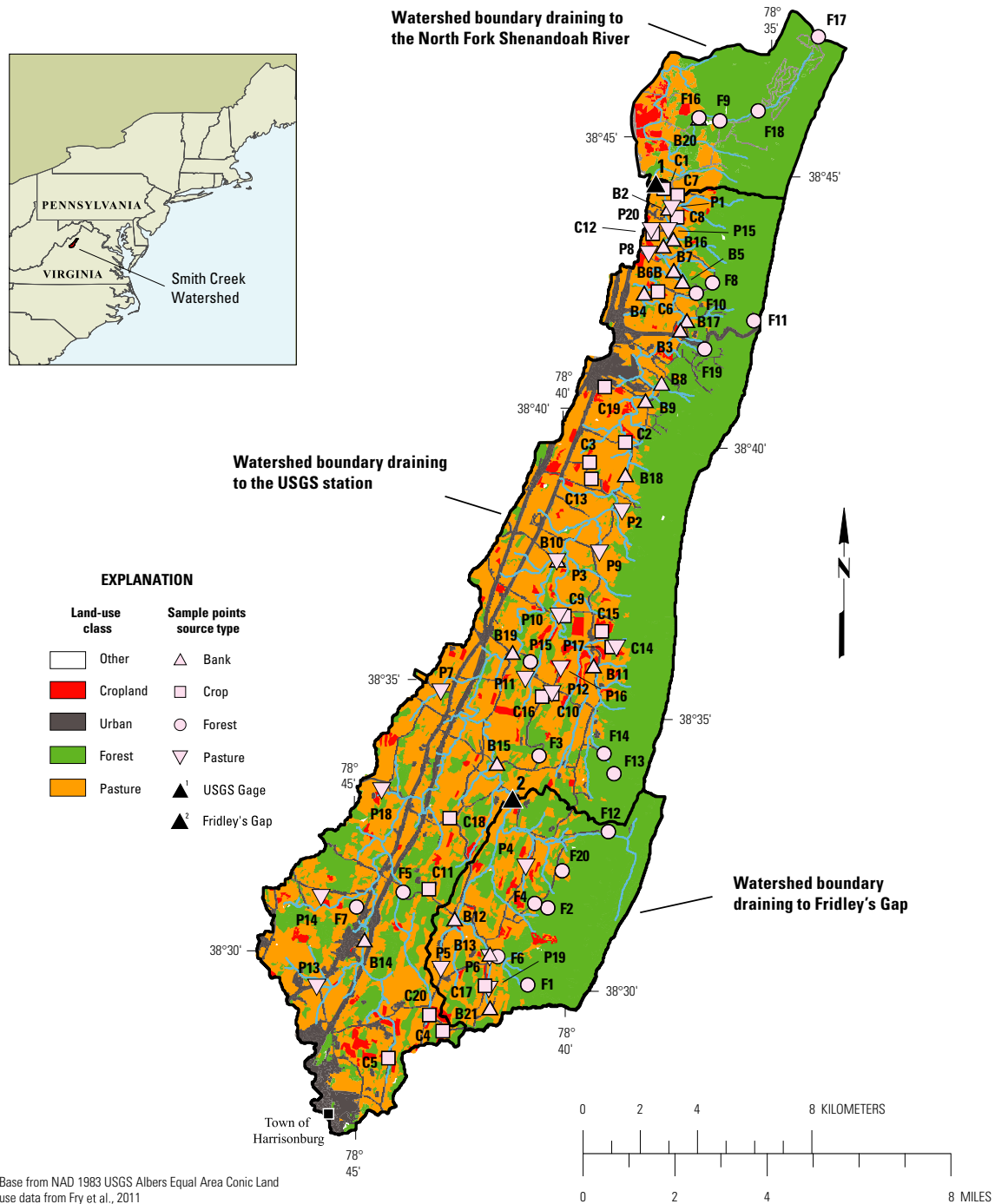


FIGURE 1. Source sample locations land use in 2011 of Smith Creek, Virginia Watershed (letter and number indicate sample ID; with letter denoting source type). USGS, United States Geological Survey.

fine-grained suspended sediment (Collins et al. 1997; Motha et al. 2003; Gellis et al. 2009; Walling 2013). The sediment fingerprinting procedure used here establishes a minimal set of physical and (or) chemical properties (tracers) based on samples collected in upland or channel locations identified as potential sources of sediment. Suspended-sediment target samples collected under different flow conditions exhibits

a composite, or fingerprint, of these properties that allows them to be traced back to their respective sources.

From 2012 through 2015, sediment fingerprinting was performed for two portions in the watershed: (1) downstream at the USGS gage at Smith Creek near New Market, Virginia (246 km²; USGS station ID 01632900), herein referred to as the “downstream

station” and (2) upstream at Fridley’s Gap located at a point midway in the watershed (44.4 km²), herein referred to as the “upstream station” (Figure 1). The sediment fingerprinting approach has been used in several watersheds of varying scales and land uses draining the Chesapeake Bay watershed (Table 1). Previous sediment fingerprinting results for these watersheds indicate that sediment sources vary spatially and temporally, partly as a result of land use changes over time, geology, storm factors, and sediment storage (Gellis et al. 2009; Gellis et al. 2015). Using this knowledge, we designed our sampling to account for variability in land use and geology in order to determine the sediment contribution from each source.

Target Samples. At both the downstream and upstream sampling stations, target samples of suspended sediment were collected during storm events in Smith Creek using a passive sampler (Phillips et al. 2000) (Figures 2 and 3). At both sampling stations, two 120 cm length, 10 cm diameter PVC tubes were each mounted on channel struts that were hammered into the channel bed. At the time of installation at the downstream station (January 20, 2012), water entered each PVC tube at a river stage of 1.0 m (discharge ~7.4 m³/s). The channel struts were often damaged at the downstream station during large runoff events and on March 27, 2013, each PVC tube was mounted on a tree on each side of the stream, approximately 50 m from each other, where flow entered the samplers at a river stage of 1.2 m (discharge ~11.0 m³/s). Sediment was retrieved after an event and composited from each tube into one sample. At the upstream station, two passive samplers were installed on May 8, 2012. The upstream station is not gaged and the discharge that starts to fill each tube was not determined. During the study period, 19 storm events were sampled at the downstream station (Table 2a) and 18 samples were collected at the upstream station (Table 2b). Other studies that have used the Phillips et al. (2000) passive sampler design include: Gellis et al. (2017) for streams in the Midwest U.S.; Pulley and Rowntree (2016) in South Africa; and Collins, Zhang, et al. (2010) in the United Kingdom.

Source Samples. Sediment source samples to apportion sediment at the downstream station were collected from cropland ($n = 20$), pasture ($n = 20$), forest ($n = 20$), and streambanks ($n = 22$) (Figure 1). A subset of source samples from the Fridley Gap watershed was used to apportion sediment at the upstream station: pasture ($n = 16$), cropland ($n = 11$), forest ($n = 8$), and streambanks ($n = 8$) (Figure 1). Samples were not collected from the 8% of the watershed identified as developed, as most of this land use

TABLE 1. Summary of sediment fingerprinting studies conducted in the Chesapeake Bay watershed.

Stream name	Drainage area, km ²	Land use, %				Sediment source apportionment (%)						Reference	
		Agriculture ¹	Forest	Urban upland ²	Urban	Agriculture ¹	Forest	Urban upland ²	Street residue	Construction sites	Drainage ditches		Streambanks
Pocomoke River, Maryland–Delaware	156.7	52	46	NA	1	46	13	—	—	—	34	7	1
Mattawoman Creek, Maryland	134.5	18	60	NA	19	17	29	—	23	—	—	31	1
Little Conestoga Creek, Pennsylvania	109.5	45	4	NA	49	77	—	—	0	—	—	23	1
Anacostia River, Maryland–District of Columbia	189.0	0	0	NA	100	—	—	30	13	—	—	58	2
Mill Stream Branch, Maryland	31.6	74	22	NA	2	—	—	—	—	—	—	100	3,4
Linganore Creek, Maryland	147.0	62	27	NA	8	45	3	—	—	—	—	52	5
Difficult Run, Virginia	14.2	NA	NA	46	54	—	—	1	10	—	—	89	6

Note: References: 1: Gellis et al. (2009); 2: Devereux et al. (2010); 3: Banks et al. (2010); 4: Massoudieh et al. (2012); 5: Gellis et al. (2015); 6: Cashman et al. (2018).

¹Agriculture includes pasture, hay, and cropland.

²Urban upland includes areas defined as parks, lawns, and forest.



FIGURE 2. Passive samplers deployed at Smith Creek, Virginia to collect suspended sediment (left photo, sampler on right bank [March 24, 2015]; right photo, sampler on left bank [March 28, 2012]).

type is impervious. Samples for source analysis from cropland, pasture, and forest 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 three transects running parallel to slope, each ~100 m in length and spaced ~30 m. At each transect, a sample was collected every 10 m and all samples were mixed into a single sample in the field. Sediment samples from streambanks were obtained by scraping the entire exposed streambank to a depth of ~1 cm with a plastic hand shovel. Three to five bank profiles spaced 10 m apart along the stream reach were sampled and composited into one sample.

Sample Preparation and Laboratory Analysis. Source and target samples were transported to the USGS Baltimore, Maryland laboratory on ice where they were wet-sieved with de-ionized water through a 63- μm polyester sieve to remove the sand, and dried again at 60°C. The silt and clay portions of the samples were sent for elemental and stable isotopic analysis (Table 3). Ideally ~2 g of fine sediment was necessary for all analyses. Elemental analysis was conducted at the U.S. Environmental Protection Agency (USEPA) facilities in Fort Meade, Maryland, which reported elemental concentrations for 18 tracers using inductively coupled plasma-optical emission spectrometry (ICP-OES) and ICP combined with mass spectrometry (ICP-MS) (Table 3; USGS Data Release Source, <https://doi.org/10.5066/f7rn36q1>). The elemental analysis followed standard USEPA laboratory protocols (<https://www.epa.gov/homeland-security-research/epa-method-2008-determination-trace-elements-water-and-wastes>; <https://www.epa.gov/homeland-security-research/method-2007-determination-metals-and-trace-elements-water-and-wastes>). Supplementary data can be found at USGS Data Release <https://doi.org/10.5066/f7rn36q1>. Grain size analysis of the <63- μm fraction was conducted at the USGS Baltimore, Maryland laboratory using a Laser In Situ Scattering

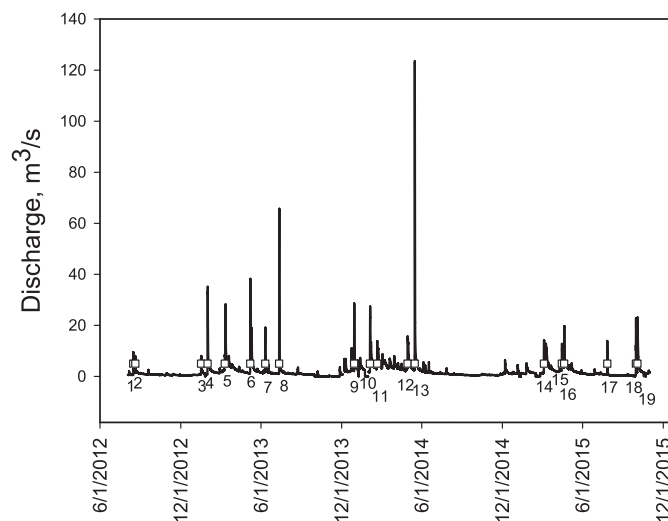


FIGURE 3. Flow hydrograph (15 min) for period of study showing sampled events (1 through 19) at the downstream station (USGS gage, Smith Creek near New Market, Virginia; station ID 01632900).

Transmissometer (LISST-100X); (<http://www.sequoiasci.com/wp-content/uploads/2013/07/manual-5.pdf>). Median values (D_{50}) of the fine-grained sediment (<63- μm) are reported in the LISST software (USGS Data Release; <https://doi.org/10.5066/f7rn36q1>).

Sediment Apportionment. The statistical approach to apportion sediment used a two-step process—(1) “a priori” test, where samples were first tested for elemental differences due to sampling in different geologic terrains and (2) multivariate sediment source apportionment based on land use differences.

“A priori” Testing for Geologic Differences

Geology can influence sediment chemistry (Pouyat et al. 2007; Woodruff et al. 2009; Tan 2011) and other fingerprinting studies have apportioned sediment by geology (Collins et al. 1998; D’Haen et al. 2013;

TABLE 2. Summary of sampled events, 2012–2015 at (a) the downstream station (USGS gage) and (b) the upstream station (Fridley's Gap), Smith Creek, Virginia.

Event #	Sample period	Event #	Sample period	Event #	Sample period
(a)					
1	8/15/2012 0800 AM–8/15/2012 0200 PM	8	7/12/2013 0715 AM–7/13/2013 0100 AM	14	3/5/2015 0345 AM–3/10/2015 0145 AM
2	8/20/2012 0130 AM–8/20/2012 0415 AM	9	12/29/2013 1030 AM–12/30/2013 1100 AM	15	4/14/2015 1115 PM–4/15/2015 0945 AM
3	1/16/2013 0645 AM–1/16/2013 1000 PM	10	2/3/2014 0815 AM–2/4/2014 1115 AM	16	4/20/2015 0415 AM–4/21/2015 0130 AM
4	1/30/2013 0845 PM–2/1/2013 0145 PM		2/5/2014 0545 AM–2/6/2014 0230 AM	17	7/27/2015 0115 AM–7/27/2015 0315 AM
5	3/10/2013 0845 PM–3/14/2013 0845 AM	11	2/19/2014 0700 PM–2/20/2014 0345 AM	18	9/29/2015 0930 PM–9/30/2015 0915 AM
	3/19/2013 0600 PM–3/20/2013 1200 PM	12	4/29/2014 0645 PM–5/1/2014 0645 PM	19	10/3/2015 0745 AM–10/4/2015 0845 PM
6	5/7/2013 0800 PM–5/9/2013 1115 AM	13	5/16/2014 1245 AM–5/17/2014 0915 PM		
	5/10/2013 1100 PM–5/11/2013 1045 AM				
7	6/10/2013 0745 PM–6/11/2013 0115 AM				
(b)					
Event #	Collection date	Collection time	Event #	Collection date	Collection time
1	8/8/2012	0200 PM	10	1/1/2014	1155 AM
2	8/16/2012	0615 PM	11	2/12/2014	0515 PM
3	8/21/2012	0350 PM	12	3/1/2014	1100 AM
4	1/23/2013	0630 PM	13	5/8/2014	1200 PM
5	2/9/2013	1200 PM	14	3/18/2015	0200 PM
6	3/16/2013	0230 PM	15	4/17/2015	1200 PM
7	5/9/2013	0300 PM	16	4/29/2015	0321 PM
8	7/16/2013	1050 AM	17	10/1/2015	1145 AM
9	12/27/2013	0300 PM	18	10/7/2015	0245 PM

Miller et al. 2015). Because Smith Creek is underlain by two main geologic units (dolostone and limestone, and sandstone and shale), we first needed to determine if the geologic signature affected the tracer signal from each source. We used the nonparametric Mann–Whitney rank sum test to determine if the median tracer properties from each land use showed a statistical difference ($p < 0.05$) when grouped by geology. If more than 50% of the tracers for a given land use showed a significant geologic signal, the land use types were separated by geology in the final analysis. If samples were not significantly different based on the Mann–Whitney rank sum test results, a decision was made to combine samples from different geologic areas with similar land use. Because only four pasture samples were collected in areas underlain by shale, the Mann–Whitney rank sum test was not used on pasture samples.

Multivariate Sediment Source Apportionment

The Sediment Source Assessment Tool (Sed_SAT version 1.0) was used to execute the statistical steps in the sediment fingerprinting approach (Gorman Sanisaca et al. 2017; available at https://my.usgs.gov/bitbucket/projects/SED/repos/sed_sat/browse). Sed_SAT is written in the statistical language R (R Core Team 2016) using a Microsoft Access[®] interface that assists

TABLE 3. Tracers used in sediment fingerprinting analysis.

Tracer	Units	Tracer	Units
Aluminum*	µg/g	Manganese*	µg/g
Arsenic*	µg/g	Nickel*	µg/g
Barium*	µg/g	Potassium*	µg/g
Beryllium*	µg/g	Strontium*	µg/g
Cadmium*	µg/g	Uranium*	µg/g
Chromium*	µg/g	Vanadium*	µg/g
Cobalt*	µg/g	Zinc [†]	µg/g
Copper*	µg/g	δ ¹³ C [†]	‰
Iron*	µg/g	δ ¹⁵ N [†]	‰
Lead*	µg/g	Total organic C [†]	%
Magnesium*	µg/g	N [†]	%

Notes: USEPA, U.S. Environmental Protection Agency; ICP-MS, inductively coupled plasma-mass spectrometry; ICP-OES, ICP-optical emission spectrometry.

*Analyzed at USEPA Laboratory facility using ICP-MS and ICP-OES.

[†]Analyzed at Isotope Tracer Technologies, Inc.¹

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

the user to step through all statistical steps to apportion sediment (Gorman Sanisaca et al. 2017). Each target sample was run through Sed_SAT individually, using the default settings, to determine the sediment contribution from each land use per event.

Prior to running any analysis in Sed_SAT, the datasets must be complete with estimated values for

nondetects. Substituting a value, such as one-half the reporting limit for a nondetect introduces error (Helsel 2012). Nondetects for this study were imputed using single imputation in the *zCompositions* R-package (Palarea-Albaladejo and Martin-Fernandez 2014). Following imputation, outliers in the source dataset were identified and evaluated. Samples with any tracer that is greater (or less than) three times the standard deviation plus or minus the mean of that source group are removed. The outlier test requires that the univariate tracer concentration data are normally distributed where each source group is tested for normality using a Shapiro–Wilk test ($p > 0.05$). If the data in its original, untransformed form are not normally distributed, each tracer in each source group is transformed using the “ladder of powers” (log base10, square root, square, cube root, inverse, and inverse square root) transforms (Helsel and Hirsch 2002). If normality is still not achieved, the outlier test is performed on the untransformed data.

A prerequisite of sediment fingerprinting is that the tracer concentration values in the target dataset must be conservative and not change during transport from the source to the sampling point (Gellis and Walling 2011). Grain size and organic content can have a strong influence on tracer concentrations (Horowitz and Elrick 1987; Miller et al. 2015). Because grain size and organic differences may exist between the source samples and target (suspended-sediment) samples, it is important to make the source and target samples comparable (Gellis and Walling 2011). In many sediment fingerprinting studies, a grain size and organic correction needs to be applied to the source samples to make them comparable to the target samples (Collins, Walling, et al. 2010; Gellis and Noe 2013).

Size and organic corrections were performed by individual sources for a given tracer. Grain size and organic corrections to the source data are performed when a significant regression occurs between grain size or total organic carbon (TOC) and the tracer’s concentration (or activity). Default settings in *Sed_SAT* define a significant relation as having: (1) normally distributed residuals (Shapiro–Wilk p -value > 0.05), (2) the slope of regression line is significant (p -value < 0.05), and (3) the $R^2 > 0.50$. Regressions of grain size (and TOC) and tracer concentrations are first tested for a significant relation with untransformed data. If a significant relation is not found, grain size is transformed using the “ladder of powers” transforms defined for the outlier test. If a significant relation is still not found, the tracer concentration is transformed using the same transformations and all possible combinations of transformed and untransformed grain size and tracer concentration data are tested for a significant relation. If after this step a significant relation is not achieved, the tracer concentrations are not corrected for grain size (and TOC). When

a significant relation occurs between grain size (or TOC) and tracer concentration, each source tracer concentration is compared to the target sample and corrected according to rules established in Gellis et al. (2016).

The bracket test is a test for the conservativeness of the tracer. For each target sample, each tracer must be bracketed by the source samples’ tracer concentrations ($<10\%$ of the minimum and $>10\%$ of maximum tracer concentration). Any tracer that did not satisfy this condition was considered to be nonconservative and was removed from further consideration. The bracket test was performed on tracers after the grain size and organic correction factors were applied.

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, a forward stepwise discriminant function analysis (DFA) was used to select an optimal group of tracers after size and organic corrections were applied. DFA looks for the linear combination of tracer coefficients that best separates or discriminates the source groups (Fisher 1936). The default significance is 0.01. Stepwise DFA is performed utilizing the *greedy.wilks()* function, which starts tracer selection with the tracer that yields the greatest separation between the groups, and adds tracers using the Wilk’s lambda criterion until there are no longer significant tracers (Mardia et al. 1979). The closer the Wilk’s lambda statistic is to 0, the more significant a tracer’s contribution to the linear discriminant function. The default significance level is 0.01.

The final step in the statistical analysis is determining the percent contribution of each sediment source using an “unmixing model” modified from Collins, Walling, et al. (2010).

$$RE = \left(\sum_{i=1}^n \left\{ \left(C_i - \left(\sum_{s=1}^m P_s S_{si} \right) \right) / C_i \right\}^2 W_i \right) \quad (1)$$

And

$$\sum_{s=1}^m P_s = 1, \quad (2)$$

where RE = relative error term; C_i = concentration of tracer property (i) in each target sample; P_s = the optimized percentage contribution from source category (s); S_{si} = mean concentration of tracer property (i) in source category (s) after size and organic corrections are applied; W_i = tracer discriminatory weighting; n = number of fingerprint properties comprising the optimum composite fingerprint; m = number of sediment source categories.

The unmixing model optimizes for the lowest relative error value using all possible source percentage combinations. The tracer discriminatory weighting value, W_i , is a weighting used to reflect tracer discriminatory power (Equation 1) (Collins, Walling, et al. 2010).

$$W_i = \frac{P_i}{P_{\text{opt}}}, \quad (3)$$

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

Target samples from both the upstream and downstream stations were input individually through Sed_SAT to apportion the sediment to cropland, pasture, forest, or streambanks. Source percentages are presented for: (1) each sample, (2) averaged for the entire study period, and (3) weighted by the sediment load for each storm event (only applicable to the downstream station).

Analysis of Uncertainty in the Sediment Fingerprinting Approach

In Sed_SAT, the ability of the final set of tracers selected to apportion sediment is evaluated by: (1) the confusion matrix, (2) the source verification test (SVT), and (3) a Monte Carlo analysis (Gorman Sanisaca et al. 2017). The confusion matrix is produced in stepwise DFA and describes the percent of source samples correctly predicted for each group vs. the actual number of source samples in each group (Kohavi and Provost 1998).

The SVT is designed to determine how well the final set of tracers discriminates the sources if the source samples are treated as target samples. The corrected source samples are entered as target samples into Sed_SAT. This test is designed to inform the user qualitatively how well the final set of tracers can correctly apportion sources. If source samples are not accurately identified (i.e., <50% of the correct source) but are characterized as others sources, it may indicate that the sources have similar chemical signatures and a decision can be made to combine source types into a general category (e.g., cropland and pasture into agriculture) (Gellis et al. 2015). In addition, if a sample is consistently misclassified (e.g., <50%) for all target samples, the user may decide to remove this sample and start the process again.

A Monte Carlo simulation was used to quantify the uncertainty in the sediment fingerprinting results

produced by the unmixing model (Collins and Walling 2007; Gellis et al. 2016). In Sed_SAT, the Monte Carlo simulation randomly removes one sample from each of the source groups and the unmixing model is run without these samples (Gorman Sanisaca et al. 2017). The Monte Carlo simulation is run 1,000 times per target sample. For each target sample, summary statistics of the Monte Carlo simulation are output by Sed_SAT. The difference between the final unmixing model results and the average of the 1,000 Monte Carlo results, and the minimum and maximum source percentage results, as well as boxplots, are used to assess the sensitivity of the final apportionment to removal of individual source samples.

Weighting Results by Sediment Load

Water year suspended-sediment loads have been computed and published by the USGS for the downstream station for 2011–2013 (Hyer et al. 2016). Sediment data were not available for the upstream station. Hourly suspended-sediment loads obtained for the downstream station were summed for the time period of each sampled event. The sediment fingerprinting source results for each sample were weighted by the suspended-sediment load for each sample relative to the total load for all samples and summed to get a weighted average for the period of study, as follows:

$$\text{Storm}_{\text{wt}(n)} = \frac{\text{SSstorm mass}_n}{\sum_{i=1}^n \text{SSstorm mass}_i}, \quad (4)$$

where $\text{Storm}_{\text{wt}(n)}$ is the weight given to the sediment load transported for each sampled event (n); SSstorm mass_n is the suspended-sediment load (Mg) computed for each target sample period n ; SSstorm mass_i is the summed sediment load transported for all samples i (from 1 to n).

$$S_v = \sum_{i=1}^n [\text{SA}_{vi} \times \text{Storm}_{\text{wt}(n)}], \quad (5)$$

where S_v = storm-weighted source apportionment at the downstream station, in percent for each source

TABLE 4. Summary of Mann–Whitney rank sum test showing the 22 tracers with a significant dependence on geology. (Samples were separated by limestone/dolomite and shale/sandstones.)

Land use type	Tracers that showed a difference by geology	Percent of samples differentiated by geology
Streambanks	{no tracers}	0
Crop	Al, Ba, Be, Fe, Mg, Ni, K	32
Forest	$\delta^{13}\text{C}$	4

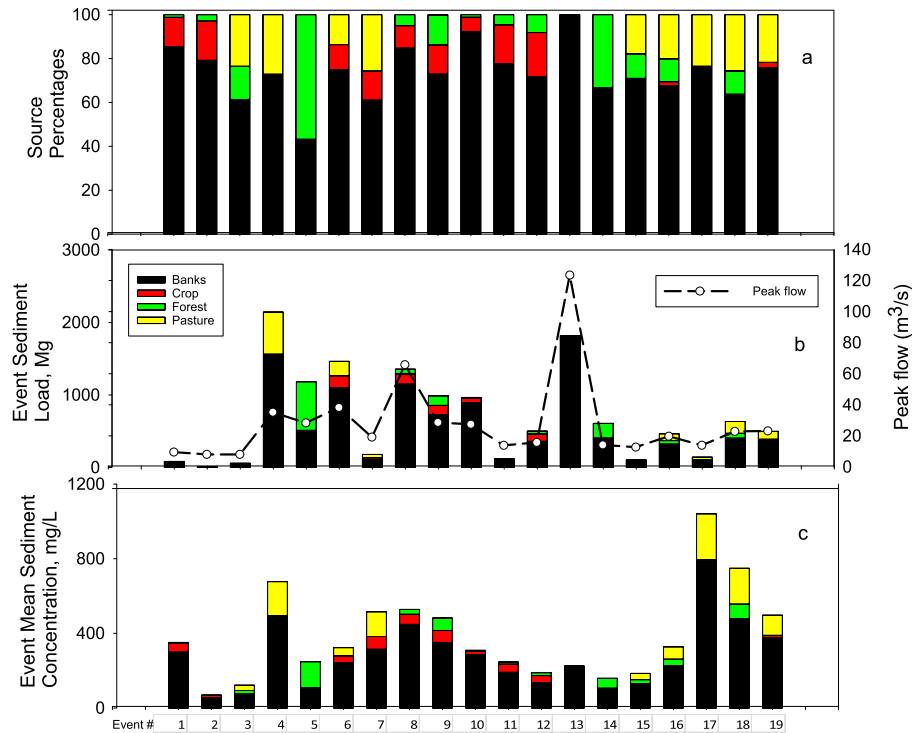


FIGURE 4. Sediment fingerprinting results for the 19 events sampled at the USGS gage, the downstream sampling site on Smith Creek, Virginia, from 2012 to 2015 shown by: (a) percentage, (b) event sediment load plotted with peak flow, and (c) event mean sediment concentration.

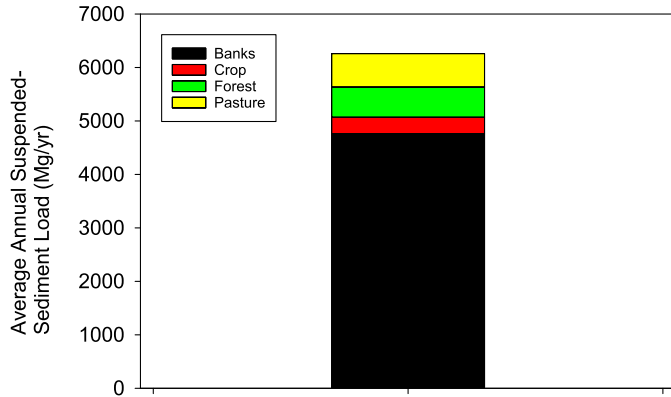


FIGURE 5. Annual allocation of sediment sources based on weighted-sediment fingerprinting results at the USGS gage applied to Smith Creek's annual suspended-sediment load, 2011–2013 (6,260 Mg/yr) (Hyer et al. 2016).

(v); (v = cropland, pasture, forest, and streambanks); SA_{vi} = sediment source apportionment from the sediment fingerprinting results (in percent) (Equation 1) for source (v) and sample i ; n = number of sampled events (i) = 19 at the downstream station.

Weighting the sediment fingerprinting results by the sediment load for each sample incorporates the importance of high loading events (Gellis and Walling 2011). It should be pointed out that the suspended-

sediment load computations include particle sizes in the sand range ($>63\ \mu\text{m}$), whereas sediment fingerprinting results are only for the $<63\text{-}\mu\text{m}$ fraction. Analysis of 145 suspended-sediment samples from Smith Creek collected between April 2010 and August 2017 (USGS 2016) indicate that on average $73 \pm 24\%$ is finer than $63\ \mu\text{m}$. Frequent suspended-sediment grain size analysis for each storm would be required to compute a fine sediment load, which was not possible with the current data. Therefore, there is some error in determining fine sediment apportionment by weighting with suspended-sediment loads that contain sand.

RESULTS AND DISCUSSION

Geologic Differences in Tracer Concentrations

The Mann–Whitney rank sum test results for elemental and stable isotope tracer biases in areas underlain by limestone/dolostone vs. sandstone/shale showed no significance for streambanks, seven tracers showed significant differences in croplands, and one tracer for forest (Table 4). Based on the low percentage ($<50\%$) of tracers showing a significant

TABLE 5. Sediment fingerprinting results for Smith Creek, Virginia.

Event #	Collection period	Cropland %	Pasture %	Forest %	Streambanks %	Relative error (Equation 1)
(a) Results at the USGS gage						
1	8/15/2012 0800 AM–8/15/2012 0200 PM	14	0	1	85	1.4919
2	8/20/2012 0130 AM–8/20/2012 0415 AM	18	0	3	79	0.9558
3	1/16/2013 0645 AM–1/16/2013 1000 PM	0	23	15	61	0.8156
4	1/30/2013 0845 PM–2/1/2013 0145 PM	0	27	0	73	0.1068
5	3/10/2013 0845 PM–3/14/2013 0845 AM	0	0	57	43	0.1961
6	3/19/2013 0600 PM–3/20/2013 1200 PM					
	5/7/2013 0800 PM–5/9/2013 1115 AM	12	14	0	75	0.0543
	5/10/2013 1100 PM–5/11/2013 1045 AM					
7	6/10/2013 0745 PM–6/11/2013 0115 AM	13	26	0	61	0.7138
8	7/12/2013 0715 AM–7/13/2013 0100 AM	10	0	5	85	0.1214
9	12/29/2013 1030 AM–12/30/2013 1100 AM	13	0	14	73	0.3642
10	2/3/2014 0815 AM–2/4/2014 1115 AM	6	0	1	92	0.0115
	2/5/2014 0545 AM–2/6/2014 0230 AM					
11	2/19/2014 0700 PM–2/20/2014 0345 AM	18	0	5	78	0.2162
12	4/29/2014 0645 PM–5/1/2014 0645 PM	20	0	8	72	0.1541
13	5/16/2014 1245 AM–5/17/2014 0915 PM	0	0	0	100	0.1728
14	3/5/2015 0345 AM–3/10/2015 0145 AM	0	0	33	67	1.0042
15	4/14/2015 1115 PM–4/15/2015 0945 AM	0	18	11	71	0.0367
16	4/20/2015 0415 AM–4/21/2015 0130 AM	2	20	10	68	0.0087
17	7/27/2015 0115 AM–7/27/2015 0315 AM	0	24	0	76	0.1454
18	9/29/2015 0930 PM–9/30/2015 0915 AM	0	26	11	64	0.6678
19	10/3/2015 0745 AM–10/4/2015 0845 PM	3	22	0	76	0.2935
Average		7	10	9	74	
Weighted by sediment load		5	10	9	76	
Event #	Sample date	Cropland %	Pasture %	Forest %	Streambanks %	Relative error
(b) Sediment fingerprinting results for Fridley's Gap, Virginia						
1	8/8/2012	0	19	11	70	0.2033
2	8/16/2012	0	0	50	50	0.1006
3	8/21/2012	0	2	30	68	0.1147
4	1/23/2013	0	0	24	76	0.9173
5	2/9/2013	0	0	0	100	0.3355
6	3/16/2013	0	0	34	66	0.1098
7	5/9/2013	0	1	18	81	0.2451
8	7/16/2013	0	8	16	76	0.0604
9	12/27/2013	0	12	5	82	0.3818
10	1/1/2014	0	56	0	44	0.4848
11	2/12/2014	0	1	0	98	0.0683
12	3/1/2014	0	0	21	79	1.5293
13	5/8/2014	0	49	0	51	0.4600
14	3/18/2015	0	0	0	100	0.1782
15	4/17/2015	1	0	27	72	0.0921
16	4/29/2015	0	60	0	40	0.0501
17	10/1/2015	0	100	0	0	0.8705
18	10/7/2015	0	0	1	99	0.1143
Average		0	17	13	70	

difference in medians, we decided not to apportion the source samples by geology.

Sediment Fingerprinting Source Apportionment and Uncertainty

Sediment fingerprinting results are presented for the downstream station (Figures 4 and 5; Table 5a) and the upstream station (Figure 6; Table 5b).

Results from Samples Collected at the Downstream Station (USGS Gage). Averaging the sediment fingerprinting results for the 19 events at the downstream station show that streambanks contributed 74% of the apportioned sediment, pasture 10%, forest 9%, and cropland 7% (Figure 4a; Table 5a). Weighting the results by sediment loads (Tables 5a and 6) showed similar results: streambanks 76%, pasture 10%, forest 9%, and cropland 5% (Table 5a). Sediment fingerprinting results displayed

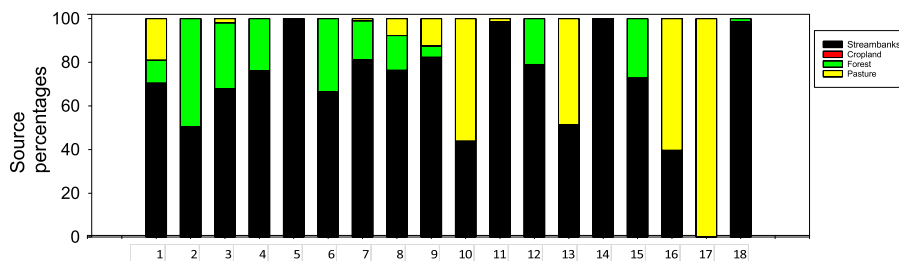


FIGURE 6. Sediment fingerprinting results for the 18 events sampled at Fridley's Gap, the upstream sampling site in Smith Creek, Virginia, from 2012 to 2015.

TABLE 6. Summary of sediment and peak flow characteristics for each sampled event at the downstream station.

Event #	Event dates and times	Suspended-sediment load, Mg	Average	Peak flow, m ³ /s	Sediment weighting factor
			suspended-sediment concentration, mg/L		
1	8/15/2012 0800 AM–8/15/2012 0200 PM	81.9	351	9.5	0.006
2	8/20/2012 0130 AM–8/20/2012 0415 AM	10.8	69.0	8.0	0.001
3	1/16/2013 0645 AM–1/16/2013 1000 PM	58.2	122	8.0	0.004
4	1/30/2013 0845 PM–2/1/2013 0145 PM	2145	677	35.1	0.161
5	3/10/2013 0845 PM–3/14/2013 0845 AM	1182	248	28.3	0.089
6	3/19/2013 0600 PM–3/20/2013 1200 PM	1464	323	38.2	0.110
	5/7/2013 0800 PM–5/9/2013 1115 AM				
7	5/10/2013 1100 PM–5/11/2013 1045 AM	179	515	19.1	0.013
	6/10/2013 0745 PM–6/11/2013 0115 AM				
8	7/12/2013 0715 AM–7/13/2013 0100 AM	1356	528	65.7	0.102
9	12/29/2013 1030 AM–12/30/2013 1100 AM	992	482	28.6	0.074
10	2/3/2014 0815 AM–2/4/2014 1115 AM	965	309	27.4	0.072
11	2/5/2014 0545 AM–2/6/2014 0230 AM	121	246	13.8	0.009
	2/19/2014 0700 PM–2/20/2014 0345 AM				
12	4/29/2014 0645 PM–5/1/2014 0645 PM	501	189	15.7	0.038
13	5/16/2014 1245 AM–5/17/2014 0915 PM	1821	225	124	0.137
14	3/5/2015 0345 AM–3/10/2015 0145 AM	608	159	14.1	0.046
15	4/14/2015 1115 PM–4/15/2015 0945 AM	104	185	12.7	0.008
16	4/20/2015 0415 AM–4/21/2015 0130 AM	464	327	19.7	0.035
17	7/27/2015 0115 AM–7/27/2015 0315 AM	142	1040	13.9	0.011
18	9/29/2015 0930 PM–9/30/2015 0915 AM	633	749	22.8	0.048
19	10/3/2015 0745 AM–10/4/2015 0845 PM	497	497	23.1	0.037

by sediment concentration and sediment loads showed that streambanks were the largest source of sediment during the highest sediment concentrations and loading events (Figure 4b, 4c). Five of the six highest mean sediment concentrations (Table 6; events 17, 18, 4, 7, and 19) had >20% contribution from pasture (Figure 4c; Table 5a). Using the weighted-sediment apportionment results (Table 5a) and applying it to the average water year suspended-sediment load for Smith Creek (2011–2013; 6,260 Mg/yr) (Hyer et al. 2016) shows the following mass contributions by source: streambanks = 4,758 Mg/yr; pasture = 626 Mg/yr; forest = 563 Mg/yr; and cropland = 313 Mg/yr (Figure 5).

DFA results indicated that the number of significant tracers varied with the target samples (Figure 7a) at the downstream station (Figure 7a; Table S1) where $\delta^{13}\text{C}$, $\delta^{15}\text{N}$, N, Ba, Mg, and Cu were significant in at

least 15 of the 19 target samples. It is important to point out that a different set of tracers can be significant in discriminating the sources for any given target sample (Figure 7). Since the size and organic content of each target sample effects the final corrected concentration of source samples, depending on the grain size and organic content of the target sample, and the bracket test removing tracers, different tracers may be significant. The confusion matrix indicated that the final set of tracers for the 19 target samples were on average able to correctly discriminate banks (94%), crop (94%), forest (86%), and pasture (82%) (Table 7). Pasture samples showed the greatest range in source discrimination, from 63% to 100% (Table 7).

The SVT results averaged by source group for the 19 sampled events at the downstream station indicated that $78 \pm 9\%$ cropland, $43 \pm 12\%$ pasture, $84 \pm 5\%$

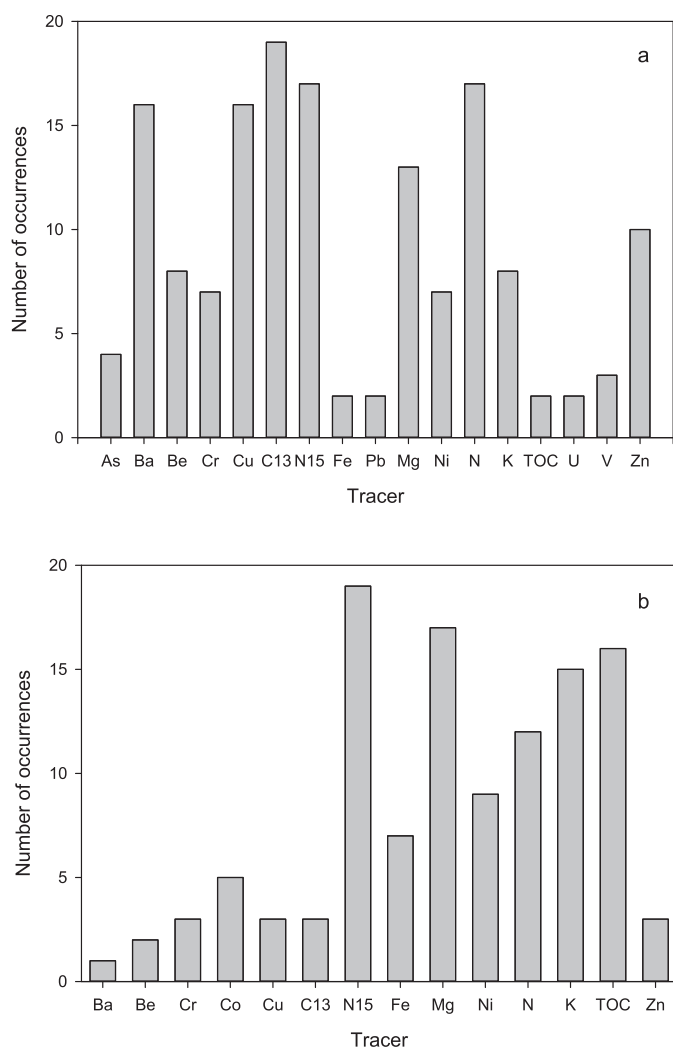


FIGURE 7. Tracers found to be significant in discriminating sediment sources (cropland, pasture, forest, and streambanks) for: (a) target samples collected at the downstream station (USGS gage Smith Creek, Virginia), and (b) target samples collected at the upstream station (Fridley's Gap, Smith Creek, Virginia).

forest, and $77 \pm 6\%$ of streambanks were correctly classified (Figure 8a) (Table S2). Results of the SVT indicated that some of the individual source samples were misclassified (Figure 9a–9d; Table 8). Individual pasture samples, which showed the lowest percentage of correctly classified (43%), were misclassified as cropland (25%), forest (27%), and streambanks (5%) (Figure 9d). Forest had the lowest percentage of misclassified samples (16%) (Figure 9c).

The average of the Monte Carlo simulation results (1,000 iterations per target sample), for the sampled events at the downstream station (Table S3), were within 1% of the mixing model results for each source group (Figure S1a). The minimum and maximum differences in the Monte Carlo results to the sediment fingerprinting results for each source group and for all events, ranged as follows: banks 2%–16%; crop

0%–9%; forest 0%–11%; and pasture 0%–16% (Table S3).

Results from Samples Collected at the Upstream Station (Fridley's Gap). Sediment fingerprinting results for the 18 events sampled at the upstream station showed streambanks as the dominant sediment source (Figure 6), contributing on average 70% of the sediment, with pasture 17%, forest 13%, and cropland 0% (Table 5b). DFA results indicated that the number of significant tracers varied with the target samples at the upstream station (Figure 7b; Table S4) where $\delta^{15}\text{N}$, Mg, TOC, and K were significant in at least 15 of the sampled events. The confusion matrix indicated that the final set of tracers for the 18 events were on average able to correctly discriminate streambanks (97%), crop (96%), forest (97%), and pasture (84%) (Table 9).

The SVT results averaged by each source group for the 18 events sampled at the upstream station indicated that $95 \pm 3\%$ cropland, $50 \pm 6\%$ pasture, $72 \pm 10\%$ forest, and $74 \pm 11\%$ of streambanks were correctly classified (Figure 8b) (Table S5). Results of the SVT indicated that some of the individual source samples were misclassified (Figure 10a–10d; Table 10). Pasture samples, which showed the lowest percentage of correctly classified (50%), were misclassified as cropland (28%), forest (13%), and streambanks (15%) (Figure 10d). Cropland had the highest percentage of correctly classified samples (95%) (Figure 10b).

The average of the Monte Carlo simulation results (1,000 iterations per target sample), for the sampled events at the upstream station (Table S6), were within 8% of the mixing model results for each source group (Figure S1b). The minimum and maximum differences in the Monte Carlo results to the sediment fingerprinting results for each source group and for all events, ranged as follows: crop 0%–13%; pasture 0%–41%; forest 0%–13%; and streambanks 0%–41% (Table S6).

Robustness of the Sediment Fingerprinting Model

Error analysis of the sediment fingerprinting results are shown using the confusion matrix (Tables 7 and 9), the SVT (Figures 8 and 9; Tables 8 and 10; Tables S2 and S5), and the Monte Carlo results (Tables S3 and S6; Figure S1). The confusion matrix indicated that on average >80% of the source samples at both sampling stations were correctly classified (Tables 7 and 9). Streambanks, on average, at the downstream and upstream stations, showed the highest percentage of correctly classified (>90%), with pasture showing the lowest percentage of correctly classified (>82%) (Tables 7 and 9).

TABLE 7. Confusion matrix results for samples collected in the watershed draining to the downstream station.

Event	Sample date	CROP	PASTURE	FOREST	STREAMBANK
1	8/17/2012	95	95	90	95
2	8/21/2012	95	95	90	95
3	1/23/2013	95	100	90	95
4	2/9/2013	91	68	85	91
5	3/27/2013	95	84	80	95
6	5/13/2013	95	84	80	95
7	6/12/2013	91	84	85	91
8	7/16/2013	91	79	90	91
9	1/1/2014	91	79	85	91
10	2/12/2014	91	68	85	91
11	3/1/2014	95	89	90	95
12	5/8/2014	95	100	95	95
13	5/21/2014	91	84	90	91
14	3/18/2015	95	84	85	95
15	4/17/2015	95	68	85	95
16	4/29/2015	91	63	85	91
17	8/3/2015	95	74	85	95
18	10/1/2015	95	79	80	95
19	10/8/2015	95	79	80	95
Average		94	82	86	94

Note: The confusion matrix describes the percent of source samples correctly predicted for each group vs. the actual number of source samples in each group (Kohavi and Provost 1998).

At both sampling locations, the SVT results indicated, on average that the final set of tracers were able to correctly apportion >70% of cropland, forest, and streambank source samples with pasture samples, on average showing the lowest percentage of correct apportionment ($\leq 50\%$) (Tables 8 and 10). At both sampling stations, the majority of misclassified pasture samples were classified as upland sources (cropland and forest) (Figures 9 and 10; Tables 8, 10, S2, and S5).

Pasture samples that were misclassified may indicate similar chemistry in soils between land uses (Miller et al. 2015). In Scotland, Stutter et al. (2009) used major and trace elements to distinguish sediment sources and also showed a lack of discrimination between pasture and cropland. Changing land use on the same field over time may also cause soil chemistry to be similar. In Linganore Creek, Maryland, the pasture and cropland fields were often rotated over time and samples for sediment fingerprinting analysis were combined into one land use—agriculture (Gellis et al. 2015). In Smith Creek, most of the agricultural land was in forest before European settlement of the area (late 1700s) (Wayland 1912; Shenandoah County 2017), and confusion between pasture and forest may indicate that some elemental characteristics of the soils have not changed since this period.

At the upstream station, 34% of streambank samples were misclassified (Figure 10a). Streambanks are constructed from sediment originating from upstream sources, and therefore may retain chemical

properties of other watershed sources (Singh and Rajamani 2001; Bølviken et al. 2004; Bogen and Ottesen 2008) resulting in this misclassification.

The average of the Monte Carlo results for all target samples are within 1% and 8% of the mixing model results for each sample, at the downstream and upstream stations, respectively (Tables S3 and S6), indicating that the removal of a random sample does not affect apportionment results. The maximum difference between Monte Carlo results and the apportionment results ranged from 16% and 41% at the downstream and upstream stations, respectively (Tables S3 and S6). The difference of 41% at the upstream station occurred in the October 1, 2015 sample, where 14 of the 1,000 iterations (1.4%) showed differences ranging from 33% to 41% from the mixing model. The remaining 986 iterations showed differences of <13%; indicating that only a small percentage of the Monte Carlo results showed this large difference.

In summary, error analysis of the sediment fingerprinting results using the confusion matrix, SVT, and Monte Carlo analysis indicates that the final set of tracers used to fingerprint sediment sources at the downstream and upstream stations in Smith Creek were able to effectively discriminate sediment sources but some overlap in apportionment results occurred between pasture and the other upland sources (cropland and forest). Therefore, samples apportioned to pasture may contain some percentage of cropland and forest, and a smaller percentage of streambanks (Figures 9 and 10).

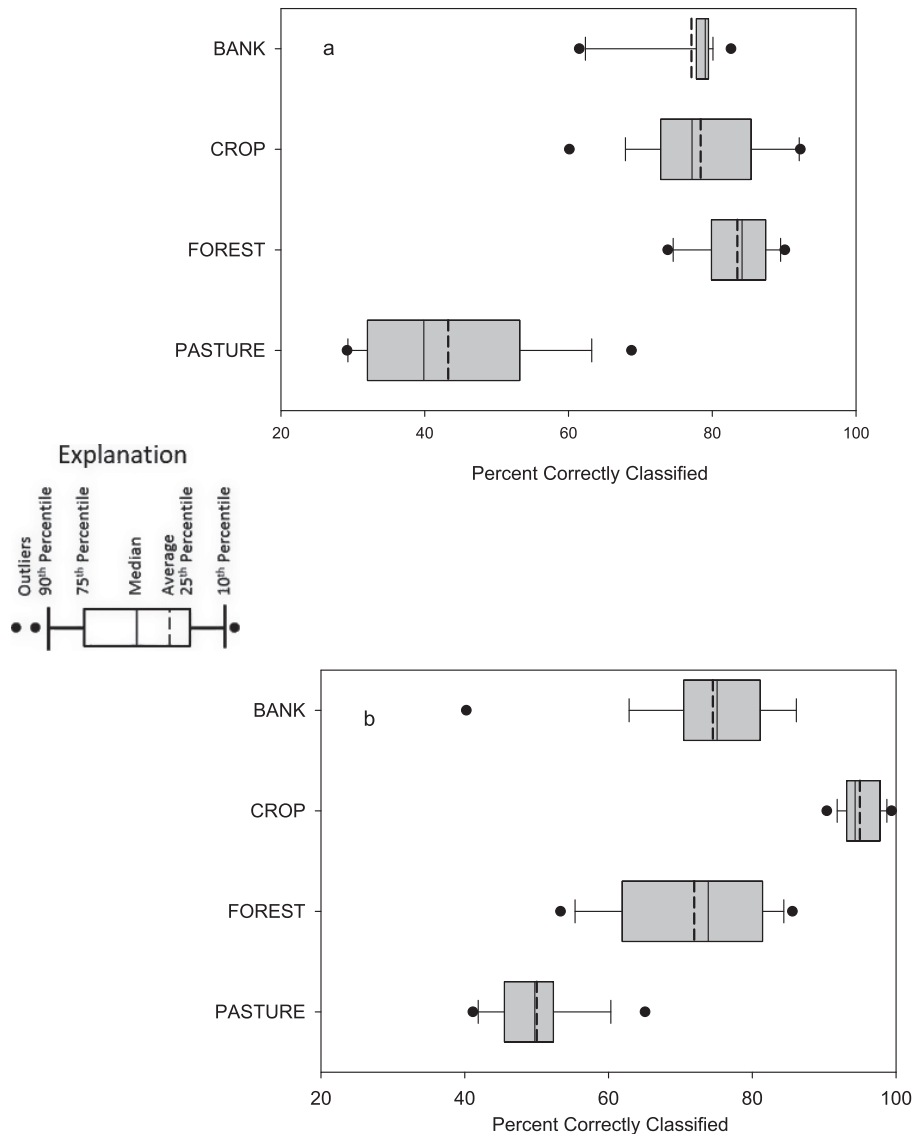


FIGURE 8. The source verification test (SVT) results showing the apportionment results averaged by source group for the: (a) 19 target samples collected at the downstream station (USGS gage) (Table S2) and (b) 18 target samples collected at the upstream station (Fridley's Gap) (Table S5). The SVT is used in Sed_SAT to determine how well the final set of tracers classifies the source samples where the source samples are treated as target samples.

Discussion of Sediment Fingerprinting Apportionment

Cropland. Cropland contributed 5% and <1% of the sediment at the downstream and upstream stations, respectively (Table 5a, 5b). The small contribution of cropland at both stations may reflect the low percentage of cropland in the watershed, 3% and 2% in the contributing area to the downstream and upstream stations, respectively. Erosion on cropland is caused by wind, water gully erosion, and tillage practices (Sojka et al. 1984; Kertis and Iivari 2006). Erosion from cropland is recognized in the Smith Creek Sediment TMDL and measures proposed to reduce erosion include grass buffers, contour plowing,

and the conversion of cropland to permanent cover, forest, or pasture (VADEQ 2009).

Pasture. Pasture contributed 10% and 17% of the sediment at the downstream and upstream stations, respectively (Table 5a, 5b). Although the area of pasture draining to the upstream station (30%) is smaller than at the downstream station (41%), the higher percentage of sediment apportioned to pasture at the upstream station may reflect the lower streambank contributions and (or) more efficient delivery from pasture lands to the upstream station. Erosion on pasture land can be related to stocking density, loss of vegetation, trampling, and lowered infiltration rates, leading to

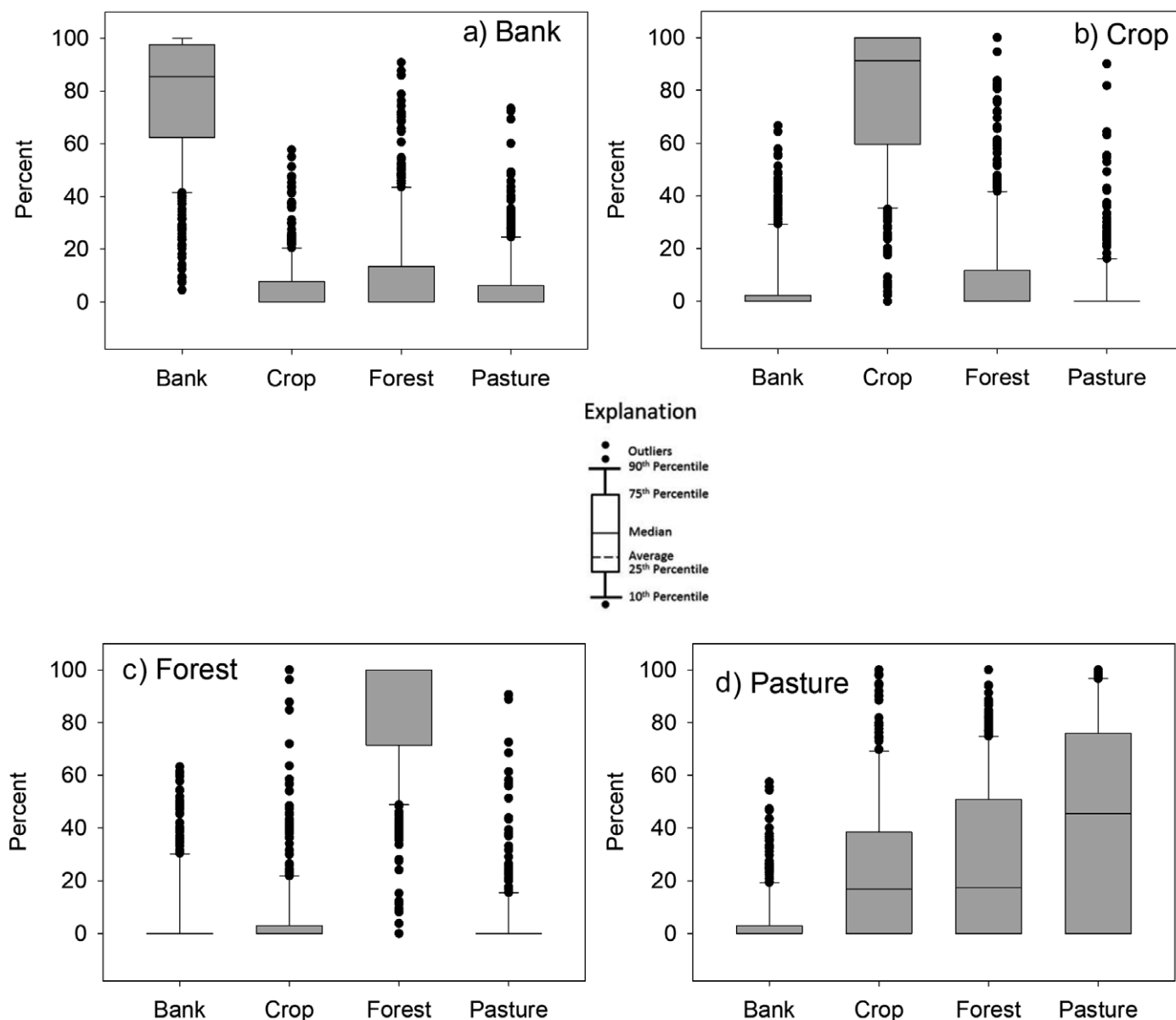


FIGURE 9. SVT results for individual source samples at the downstream station (USGS gage), (a) bank ($n = 418$), (b) crop ($n = 361$), (c) forest ($n = 380$), and (d) pasture ($n = 361$), indicating the apportioned sources for each sample.

increased surface runoff (Alderfer and Robinson 1947; Evans 1998). In the Smith Creek TMDL, strategies to reduce sediment include pasture management, reforestation of erodible pasture, and livestock exclusion from the stream through fencing (VADEQ 2009).

Forest. Forest contributed 9% and 13% of the sediment at the downstream and upstream stations, respectively (Table 5a, 5b). The percentage of sediment apportioned to forest at both sampling sites is less than the area of forest draining to each station and may indicate lower rates of erosion on forested slopes relative to other land uses, such as cropland

(Dunne and Leopold 1978). In the agricultural Lingapore Creek, Maryland, rates of erosion on forested lands using Cesium-137 (1.45 Mg/ha/yr) were an order of magnitude less than agriculture rates of erosion (pasture and cropland combined; 19.0 Mg/ha/yr) (Gellis et al. 2015). In other watersheds in the Chesapeake Bay, sediment fingerprinting results also showed forest as a source of sediment albeit lower than agriculture and streambanks (Table 1). Reasons for forest as a source of sediment in these watersheds were attributed to timber harvesting activities, land use conversion from forest to other land uses, and erosion on steep slopes occupied by forest (Gellis et al. 2009; Gellis

TABLE 8. Results of SVT showing what the sample source types were classified as at the downstream station.

Event	Source type	Classified as (%)				Event	Source type	Classified as (%)			
		CROP	PASTURE	FOREST	BANK			CROP	PASTURE	FOREST	BANK
#1	BANK	5	3	13	78	#10	BANK	6	6	9	79
#1	CROP	77	3	15	4	#10	CROP	75	10	8	8
#1	FOREST	3	3	88	6	#10	FOREST	7	4	84	5
#1	PASTURE	15	53	28	4	#10	PASTURE	29	37	29	4
#2	BANK	5	4	13	78	#11	BANK	4	4	12	80
#2	CROP	78	2	14	5	#11	CROP	75	7	12	6
#2	FOREST	3	3	87	6	#11	FOREST	3	5	87	5
#2	PASTURE	17	49	30	4	#11	PASTURE	20	46	32	2
#3	BANK	7	2	12	79	#12	BANK	5	3	12	79
#3	CROP	92	2	5	1	#12	CROP	73	6	11	11
#3	FOREST	4	4	86	6	#12	FOREST	2	4	86	8
#3	PASTURE	11	69	18	3	#12	PASTURE	11	63	22	4
#4	BANK	5	10	6	79	#13	BANK	7	8	7	78
#4	CROP	85	2	7	5	#13	CROP	92	2	2	3
#4	FOREST	11	4	78	7	#13	FOREST	14	5	74	7
#4	PASTURE	35	31	29	6	#13	PASTURE	30	39	26	5
#5	BANK	8	7	24	62	#14	BANK	7	5	26	62
#5	CROP	78	6	6	10	#14	CROP	60	4	26	10
#5	FOREST	7	7	80	6	#14	FOREST	2	1	90	7
#5	PASTURE	29	47	16	7	#14	PASTURE	11	56	26	7
#6	BANK	6	6	9	79	#15	BANK	6	7	7	79
#6	CROP	72	7	14	8	#15	CROP	87	1	8	5
#6	FOREST	5	4	86	5	#15	FOREST	11	4	78	7
#6	PASTURE	23	41	31	5	#15	PASTURE	40	29	25	6
#7	BANK	5	7	12	77	#16	BANK	5	10	6	79
#7	CROP	68	0	23	9	#16	CROP	76	8	7	8
#7	FOREST	2	4	88	6	#16	FOREST	6	5	83	6
#7	PASTURE	16	29	49	6	#16	PASTURE	24	39	32	5
#8	BANK	6	5	12	77	#17	BANK	5	7	8	80
#8	CROP	68	9	14	9	#17	CROP	85	2	8	5
#8	FOREST	4	1	90	6	#17	FOREST	9	4	80	6
#8	PASTURE	14	57	25	3	#17	PASTURE	40	30	24	6
#9	BANK	7	8	6	78	#18	BANK	3	6	8	83
#9	CROP	92	3	2	3	#18	CROP	80	6	10	4
#9	FOREST	14	5	75	7	#18	FOREST	8	5	81	5
#9	PASTURE	32	40	25	4	#18	PASTURE	43	35	18	3
						#19	BANK	5	7	10	79
						#19	CROP	76	6	12	6
						#19	FOREST	5	5	84	6
						#19	PASTURE	32	32	30	6

Note: The SVT is used in the Sediment SAT (Sed_SAT) to determine how well the final set of tracers discriminates the sources where the source samples are treated as target samples.

et al. 2015). In upland watersheds of the Patuxent River, Maryland, high measurements of sediment yield ($119 \text{ Mg/km}^2/\text{yr}$) were observed in first-order forested watersheds and thought to be due to erosion by concentrated overland flow and first-order channel enlargement (Smith and Wilcock 2015).

Erosion of forest lands in Smith Creek may be related to current and past land use practices of timber harvesting. The eastern flanks of the Smith Creek watershed are covered by forests of the George Washington and Jefferson National Forests (GWNF). The GWNF has a long history of timber harvesting

going back to the 1700s (USDA, Forest Service 2017). Between 1900 and 1933, over 63% of the forest was cut for lumber (USDA, Forest Service 2017). Lumber demands during World War II continued to put pressure on timber harvesting with a peak in forest cuts in 1941 (Satterthwaite 1993). By the mid-1990s, timber harvests averaged about 4,000 acres annually (USDA, Forest Service 2017). Timber harvesting and associated activities, such as dirt roads, are known to contribute a large amount of sediment (Beschta 1978; Patric et al. 1984; Megahan et al. 1995; Macdonald et al. 2003). A portion of the sediment generated from

TABLE 9. Confusion matrix results for samples collected in the area draining to the upstream station.

Event	CROP	PASTURE	FOREST	STREAMBANK
1	91	87	100	100
2	100	80	100	100
3	91	87	100	100
4	100	100	100	100
5	91	73	100	100
6	91	73	100	100
7	100	80	100	100
8	91	73	100	100
9	100	93	88	100
10	100	100	100	88
11	100	73	88	100
12	100	100	100	88
13	100	93	100	75
14	82	80	88	100
15	100	73	100	100
16	100	87	100	100
17	100	87	88	100
18	100	73	100	100
Average	96	84	97	97

Note: The confusion matrix describes the percent of source samples correctly predicted for each group vs. the actual number of source samples in each group (Kohavi and Provost 1998).

timber harvesting also goes into hillslope and channel storage where it is periodically re-eroded and delivered downstream (Croke et al. 1999; Anderson and Lockaby 2011). In Smith Creek, other evidence that suggests forests contribute sediment are deposits of fine-grained sediment observed in several ponds located at the base of the forested slopes. These are ponds (surface area ~250 m²) that capture the drainage of small streams draining the forested slopes. Examination of the sediment deposits by spudding with a 30 cm diameter by 2.5 m PVC pipe, indicated fine-grained sediment with depths >1 m. Future work may include determining the volume of sediment and the history of these ponds.

In the sediment TMDL for Smith Creek, forested lands are thought to represent natural conditions where no reductions in sediment from forested lands were proposed (VADEQ 2009). Increases in forest cover for riparian buffer protection are a suggested measure to improve water quality conditions (VADEQ 2009).

Streambanks. Streambanks contributed 76% and 70% of the sediment at the downstream and upstream stations, respectively, indicating they were the major source of sediment in Smith Creek but varied temporally (Figures 4 and 6; Table 5a, 5b). Streambank erosion can be related to several processes: (1) erosion that occurs during high flows related to high shear stresses on the bank, (2) mass-wasting related to bank properties, angle, and excess pore-water pressure, and (3) freeze-thaw activity (Wolman 1959; Wynn 2006).

At the downstream station, sediment source percentages showed a weak ($R^2 = 0.27$) but significant correlation to peak flow indicating that higher rates of bank erosion may occur during higher flows (Figure 11). Event 13, which had the highest peak flow (124 m³/s) and the second highest sediment loading (1,821 Mg), showed a 100% sediment apportionment from streambanks (Figure 4a).

Determining streambank erosion from mass-wasting requires collection and understanding of the geotechnical properties of the streambanks and conditions during a runoff event (Simon and Collison 2001; Wynn 2006) which were beyond the scope of this study. Freeze and thaw or frost action leading to shrinking and swelling of streambank sediment results in loosening material and forming detritus at the base of the streambanks (Wolman 1959; Gatto 1995; Couper and Maddock 2001). Subsequent flows with adequate shear stresses erode and transport this material, which can elevate sediment loads (Day et al. 2013; Henshaw et al. 2013). Winter months with greater freeze-thaw days may show greater apportionment of sediment from streambanks. Wolman (1959) reported that 85% of the observed streambank erosion at Watts Branch, Maryland, occurred during the winter months. At both sampling stations in Smith Creek, event apportionment results were categorized by season (spring, summer/fall, winter) (Tables S7a) and tested for differences in medians using a Mann-Whitney rank sum test ($p > 0.05$). Results did not show any significant difference in median values of sediment contributions by three seasons (winter, spring, summer/fall) (Tables S7b).

To determine if freeze-thaw activity is an important factor in the contributions of sediment from streambanks, the total number of freeze-thaw days (defined as the number of days the minimum and maximum temperature dropped above and below 0°C) was obtained from NOAA (2017; USGS Data Release, <https://doi.org/10.5066/f7rn36q1>) and regressed against sediment apportionment results for streambanks at the downstream station (Figure S2; Tables S8). Results at the downstream station did not show a significant relation between freeze-thaw days and streambank contributions (Figure S2). Based on this seasonal analysis, it does not appear that winter months or the total number of freeze-thaw days have a relation with streambank apportionment.

Another cause of streambank erosion is cattle trampling (Kauffman et al. 1983; Evans 1998; Zeckoski et al. 2007). In Smith Creek, the State of Virginia and the U.S. Department of Agriculture, Natural Resources Conservation Service are actively involved in a cooperative program of stream fencing with land owners to control cattle entry and crossing of streams (Schaeffer et al. 2017). By limiting cattle

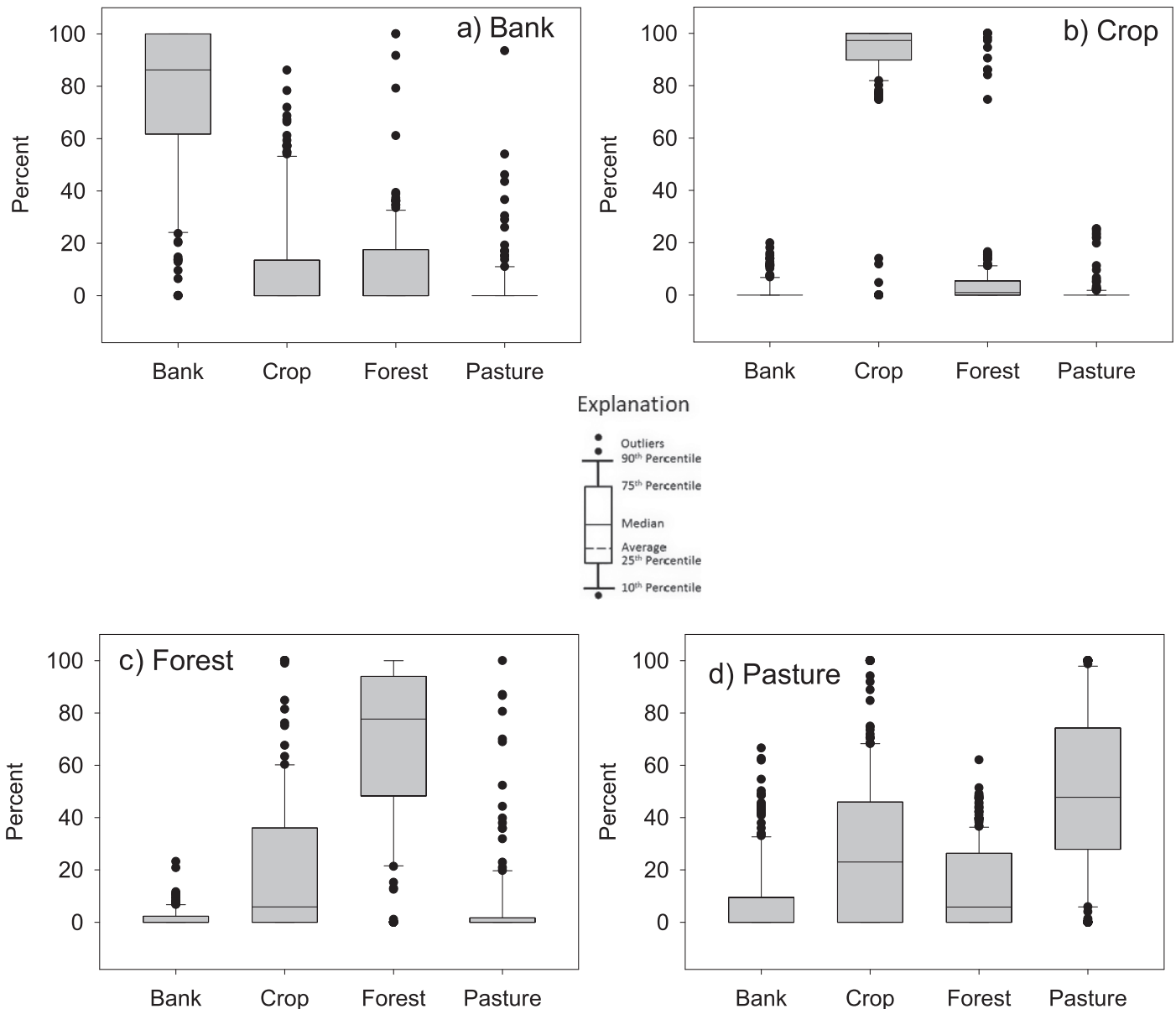


FIGURE 10. SVT results for individual source samples at the upstream station (Fridley's Gap), (a) bank ($n = 152$), (b) crop ($n = 209$), (c) forest ($n = 152$), and (d) pasture ($n = 285$), indicating the apportioned sources for each sample.

access to the river, improvements may occur in sediment, nutrients, and pathogens (Zeckoski et al. 2007; Miller et al. 2010). In the Lower Little Bow River in Alberta, Canada, total suspended-solid loads were significantly reduced (41%) by cattle exclusion (Miller et al. 2010). Improvements were due to better vegetation properties (Miller et al. 2010). In 2009, it was estimated that 278 km of fencing would be needed in Smith Creek to exclude 95% of the livestock from the stream to meet bacterial reductions (VADEQ 2009). The fencing of cattle from the stream would also have the added benefit of reducing streambank erosion. By 2016, 35 km of Smith Creek and its tributaries had

livestock fencing, which is 13% of the fencing goal (USDA-NRCS 2017).

Temporal Characteristics of Apportioned Sediment

The sediment fingerprinting approach quantifies the sources of sediment delivered to the sampling sites. It is likely that much of the delivered sediment could be from channel storage and not directly eroded and delivered to the sampling site in that storm event (Gellis and Noe 2013). In the agricultural Linganore Creek watershed, Maryland, a lack

TABLE 10. Results of SVT showing what the sample source types were classified as at the upstream station.

Event	Source type	CROP	PASTURE	FOREST	BANK	Event	Source type	CROP	PASTURE	FOREST	BANK
#1	BANK	10	10	17	63	#10	BANK	9	2	18	71
#1	CROP	98	0	2	0	#10	CROP	99	1	0	0
#1	FOREST	24	4	68	3	#10	FOREST	36	5	53	6
#1	PASTURE	18	48	17	17	#10	PASTURE	29	53	6	11
#2	BANK	16	0	9	75	#11	BANK	22	1	6	71
#2	CROP	98	0	1	1	#11	CROP	94	1	4	1
#2	FOREST	12	1	84	2	#11	FOREST	16	4	79	1
#2	PASTURE	31	55	9	5	#11	PASTURE	26	52	14	7
#3	BANK	19	3	8	71	#12	BANK	12	2	7	78
#3	CROP	95	0	5	0	#12	CROP	92	2	3	3
#3	FOREST	33	4	62	1	#12	FOREST	21	3	74	2
#3	PASTURE	30	46	17	7	#12	PASTURE	28	50	16	6
#4	BANK	10	4	5	81	#13	BANK	11	1	8	80
#4	CROP	94	0	4	1	#13	CROP	94	2	2	1
#4	FOREST	22	2	75	1	#13	FOREST	22	4	73	1
#4	PASTURE	31	45	16	8	#13	PASTURE	27	52	15	6
#5	BANK	14	0	16	70	#14	BANK	7	1	5	86
#5	CROP	99	0	0	0	#14	CROP	92	2	3	3
#5	FOREST	19	2	77	2	#14	FOREST	12	4	82	1
#5	PASTURE	25	65	5	5	#14	PASTURE	27	51	14	8
#6	BANK	11	2	8	79	#15	BANK	9	24	26	40
#6	CROP	92	2	3	3	#15	CROP	97	0	3	0
#6	FOREST	21	3	75	2	#15	FOREST	22	4	73	0
#6	PASTURE	28	49	16	7	#15	PASTURE	24	51	14	10
#7	BANK	9	4	5	83	#16	BANK	10	3	6	80
#7	CROP	94	0	4	2	#16	CROP	93	2	2	2
#7	FOREST	13	3	84	0	#16	FOREST	36	4	56	5
#7	PASTURE	32	42	16	10	#16	PASTURE	30	45	16	9
#8	BANK	10	2	7	80	#17	BANK	14	1	15	70
#8	CROP	93	2	3	2	#17	CROP	95	1	4	0
#8	FOREST	37	3	57	3	#17	FOREST	24	5	70	2
#8	PASTURE	29	48	15	8	#17	PASTURE	23	60	10	7
#9	BANK	17	0	9	74	#18	BANK	7	1	6	86
#9	CROP	98	0	2	0	#18	CROP	90	2	4	4
#9	FOREST	34	3	61	2	#18	FOREST	8	5	86	2
#9	PASTURE	33	41	15	11	#18	PASTURE	28	50	14	8

of correlation between upland source apportionment and flow conditions suggested that a portion of sediment may be derived from channel storage and not delivered directly to the stream from upland source (s) during the sampled event (Gellis and Noe 2013). Sediment delivery ratios determined for all sources in Linganore Creek: agricultural areas of cropland and pasture (4%), forests (8%), and streambanks (44%) (Gellis et al. 2015), indicated that the majority of sediment is in colluvial and channel storage before it is delivered out of the watershed. Based on the lack of correlation between peak flow and source results in this study (Table S9), similar sediment delivery processes may occur in Smith Creek. Age determination of suspended sediment could discern if the sediment is either recent (rapidly delivered from the source areas) or older (from channel storage). Several studies have used radionuclides ($^{210}\text{Pb}_{\text{ex}}$, ^7Be , ^{137}Cs) to date fine-grained sediment

(Wallbrink et al. 1998; Matisoff et al. 2005; Le Cloarec et al. 2007; Evrard et al. 2010; Belmont et al. 2014; Gellis et al. 2017).

Importance of Discriminating Upland vs. Streambank Sources

Other sediment fingerprinting studies in Chesapeake Bay have shown streambanks to be an important source of sediment. For example, five of the eight studies listed in Table 1 indicate that streambanks are among the largest sources of sediment in each watershed; in some watersheds, streambanks are 100% of the apportionment (i.e., Mill Stream Branch, Maryland). However, many of the models used by management agencies in the Chesapeake Bay watershed rely on land use/land cover for sediment sources and do not include streambanks as a

source of sediment, e.g., the USEPA Chesapeake Bay Watershed, Hydrologic Simulation Program—Fortran model (Shenk and Linker 2013), and the USGS Spatially Referenced Regressions On Watershed attributes model (Brakebill et al. 2010).

The sediment fingerprinting approach apportions sediment between upland sources (forest, pasture, crop) and streambanks which is important because sediment-reduction management strategies differ based on contributing sources. Reduction of sediment from agricultural areas may include livestock management and crop management (NHDAMF 2017), whereas reduction of sediment from streambanks may include livestock fencing, vegetative plantings, or engineered structures (Davis et al. 1994; Iowa Department of Natural Resources 2006).

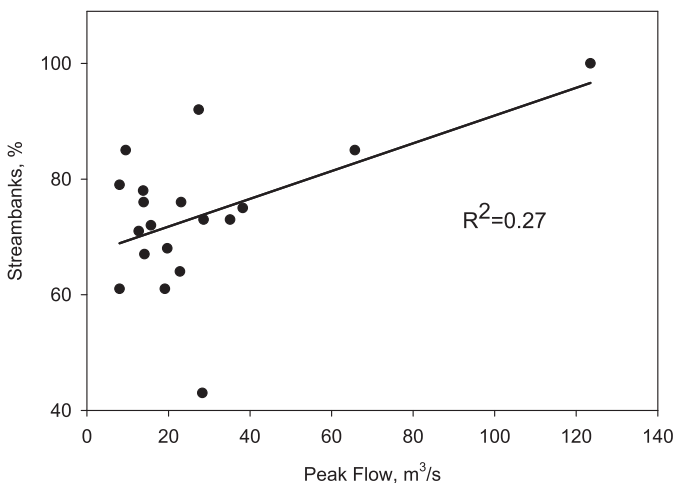


FIGURE 11. Relation of peak flow and the percentage at the downstream station of apportioned sediment from streambank sources. Regression is significant with the slope of regression line $p < 0.05$ and the residuals are normally distributed (Shapiro–Wilk p -value > 0.05).

Sediment Fingerprinting Results in the Sediment TMDL Process

Examination of several sediment TMDL reports produced by jurisdictions throughout the U.S. indicated a reliance on models, GIS analysis, and best judgment to identify sediment sources in the TMDL framework (Keyes and Radcliffe 2002; Mukundan et al. 2012). Several researchers have discussed the utility of using sediment fingerprinting results as a management tool in the Sediment TMDL process (Mukundan et al. 2012; Voli et al. 2013; Gellis et al. 2015; Gellis et al. 2016). A manual produced by the USEPA highlights sediment fingerprinting and sediment budget approaches to identify the significant sources of fine-grained sediment in the Sediment TMDL process (Gellis et al. 2016). For impaired streams draining the Neuse River, North Carolina, Voli et al. (2013) used geochemical analysis in the sediment fingerprinting approach to identify bank erosion, followed by construction sites and timber harvesting as important sediment sources.

This study demonstrates how sediment fingerprinting results can be used to guide management actions to reduce sediment loads in the Sediment TMDL process. Sediment fingerprinting results can be presented as percentage of source type and if sediment is collected, as a percentage of loads. A key finding from this study using the sediment fingerprinting approach is the importance of streambanks as a source of sediment which in the sediment load reduction strategy (TMDL) for Smith Creek, was not previously identified as a source of sediment (VADEQ 2009). In addition, many of the models available to determine sediment sources do not include streambanks as a source.

The Sediment TMDL for Smith Creek provides an opportunity to compare the sediment fingerprinting results from this study to proposed actions (VADEQ 2009). The TMDL implementation plan estimated the annual sediment load for Smith Creek as 13,648 Mg/yr, which is 218% higher than the loads reported by the

TABLE 11. Comparison of sediment Total Maximum Daily Load (TMDL) allocations (VADEQ 2009) to USGS suspended-sediment loads (Hyer et al. 2016) apportioned by the sediment fingerprinting results (Table 5a).

Land use	TMDL assigned load (Mg/yr) (VADEQ 2009)	TMDL, 22% suggested reductions (VADEQ 2009) (Mg/yr) ¹	USGS sediment load apportioned by the sediment fingerprinting results (Mg/yr)
Forest	135	0	563
Pasture	10,973	2,414	626
Crop	2,423	533	313
Urban ²	116	26	NA
Streambanks	NA	NA	4,758
Total	13,648	2,973	6,260 ³

¹The 22% reduction does not include forest.

²Includes urban, transitional, and MS4 in the VADEQ (Virginia Department of Environmental Quality) (2009) report.

³Hyer et al. (2016).

USGS (6,260 Mg/yr; water years 2011–2013) (Hyer et al. 2016) (Table 11). Differences in the reported loads are due to the different time periods examined — 2009 for the implementation plan and 2011–2013 for the USGS data — as well as the different methods used to calculate loads. The USGS computed suspended-sediment loads using the relation between discharge, turbidity, and suspended-sediment concentrations (Hyer et al. 2016). The Sediment TMDL allocations are based on the Generalized Watershed Loading Functions model, which incorporates the Universal Soil Loss Equation to generate sediment (Haith et al. 1992). The TMDL implementation plan proposes a 22% reduction in sediment loads for all land uses except forest (0%) (Table 11). The total of the proposed reductions is 2,973 Mg/yr, which is almost half (47%) of the USGS annual sediment load. It is also important to highlight that reductions in sediment from streambanks, which is 4,758 Mg/yr apportioned by sediment fingerprinting (Figure 5; Table 11), are not identified as a sediment source in the TMDL implementation plan (VADEQ 2009). Results of this study suggest that reductions in sediment loads may be effective if directed toward managing streambank erosion. Urban sediment, which is estimated as 116 Mg/yr in the TMDL implementation plan (VADEQ 2009), is not included in the sediment fingerprinting results (Table 11). Future sediment fingerprinting studies may include urban areas in their source assessment.

SUPPORTING INFORMATION

Additional supporting information may be found online under the Supporting Information tab for this article: Additional figures of output from the Sediment Source Assessment Tool (Sed_SAT) and regression analysis.

DATA AVAILABILITY

The USGS Data Release may be accessed at <https://doi.org/10.5066/f7rn36q1>. The data release is the laboratory analysis results of the source and target samples, and temperature data.

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