



Combining sediment fingerprinting with age-dating sediment using fallout radionuclides for an agricultural stream, Walnut Creek, Iowa, USA

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Abstract

Purpose The main purpose of this study was to demonstrate the utility of the sediment fingerprinting approach to apportion surface-derived sediment, and then age date that portion using short-lived fallout radionuclides. In systems where a large mass of mobile sediment is in channel storage, age dating provides an understanding of the transfer of sediment through the watershed and the time scales over which management actions to reduce sediment loadings may be effective.

Materials and methods In the agricultural Walnut Creek watershed, Iowa, the sediment-fingerprinting approach with elemental analysis was used to apportion the sources of fine-grained sediment (croplands, prairie, unpaved roads, and channel banks). Fallout radionuclides (^7Be , $^{210}\text{Pb}_{\text{ex}}$) were used to age the portion of suspended sediment that was derived from agricultural topsoil. Age dating was performed at two different scales: $^{210}\text{Pb}_{\text{ex}}$ which can date sediment to ~85 years and ^7Be to ~1 year.

Results and discussion Sediment fingerprinting results indicated that the majority of suspended sediment is derived from cropland (62%) with streambanks contributing 36%, and prairie, pasture, and unpaved roads each contributing $\leq 1\%$. The topsoil-derived portion of sediment (primarily agriculture) dated using $^{210}\text{Pb}_{\text{ex}}$ has ages ranging from 1 to 58 years, and using ^7Be , a component of much younger sediment that yields ages ranging from 44 to 205 days. The occurrence of ^7Be indicates that some portion of the sediment is young, on the order of months, whereas the dating based on $^{210}\text{Pb}_{\text{ex}}$ indicates that some of the surface-derived sediment has been in channel storage for decades. Published studies in Walnut Creek indicate that a large component of sediment is stored in the channel bed.

Conclusions We conclude that the $^{210}\text{Pb}_{\text{ex}}$ -based ages are a reasonable estimate for the mean age of the surface-derived fraction and that ^7Be activities are evidence that there is a smaller fraction of very young sediment in the stream. We propose a geomorphic model where agricultural soil is delivered to the channel and conveyed to the watershed outlet at three time scales: a geologic-millennial time scale, decades, and a young time scale (< 1 year).

Keywords Age dating sediment · Agriculture · Radionuclides · Sediment fingerprinting

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1 Introduction

Worldwide, fine-grained sediment (<63 μm) is a major pollutant impacting aquatic habitat and infrastructure through: burial of substrate (Wood and Armitage 1997; Izagirre et al. 2009), light attenuation (Wilber and Clarke 2001; Langland and Cronin 2003), clogging of fish gills (Cavanagh et al. 2014), altering oxygen demand and effect on egg gestation (Collins et al. 2014), transporting pollutants (Warren et al. 2003; Owens et al. 2005; Yi et al. 2008), sedimentation in reservoirs (Liu et al. 2017), and clogging of municipal water filtration systems (McHale and Siemion 2014). Identifying the sources of this sediment and quantifying its contribution to the total load of streams is information useful not only to management agencies but also to our understanding of processes occurring in the critical zone, the near surface layer that covers all land on Earth. Sediment fingerprinting offers an effective tool for management agencies to identify sediment inputs. Management agencies are also interested in the volume (or mass) of sediment in channel storage and the age of this sediment, as this can inform managers of the timescales when sediment controls may show an effect. Both the sources of sediment and its residence time are important critical zone processes. Sediment fingerprinting provides information on the contributions from topsoil erosion as well as the delivery of this sediment to the fluvial system, which are critical zone processes that help us understand agricultural sustainability and aquatic ecosystem stressors.

Attempting to relate sediments to their upstream sources is challenging. However, there have been advances using geochemical and mineralogical data, including radionuclides and stable isotopes, to “fingerprint” sediment sources (Collins et al. 1997, 2010, 2017; Papanicolaou et al. 2003; Walling 2013; Evrard et al. 2016; Gellis et al. 2016; Manjoro et al. 2017). The sediment fingerprinting approach is based on characterizing each of the potential sediment sources within a watershed by a composite fingerprint, defined by a number of physical or geochemical properties of the source materials and comparing the fingerprint of sampled suspended or bed sediment (target sediment) with the fingerprints of the potential sources. By using a statistical model, it is possible to estimate the relative contributions from different source types (Collins et al. 2010; Haddadchi et al. 2013; Gellis et al. 2016). Results from sediment fingerprinting studies have been used to guide management actions to reduce erosion and sediment input worldwide (Caitcheon et al. 2006; Evans et al. 2006; Gellis and Walling 2011; Mukundan et al. 2012; Gellis et al. 2016).

Distinction between sediment transit times and residence times can be confusing (Wallbrink et al. 1998; Smith et al. 2014; Moody 2017). As sediment travels from its origin to a point of interest in the watershed, it can be deposited in storage on upland surfaces (i.e., colluvial slopes) (Smith et al. 2014),

in channel storage (i.e., the active channel bed, bars) (Estrany et al. 2011; Harper et al. 2017), or on floodplains (Walling et al. 1998; Fryirs and Brierley 2001) for periods of time that range from days or weeks to millennia (Lancaster and Casebeer 2007; Pizzuto et al. 2014; Hoffmann 2015). The sediment transit time is the time it takes for sediment to travel from a starting point in the watershed to an endpoint where sediment leaves the area of interest (Matisoff et al. 2005). Residence time is the mean time sediment particles spend in a storage reservoir. Determining the transit time and residence time of sediment depends on the framework or scale of interest. An upland transit time would examine the average time it takes for a sediment particle to leave an upland source and enter the channel (Slattery et al. 2002). At the watershed scale, a sediment transit time could include the time sediment spends in all storage reservoirs, i.e., on hillslopes and on the channel bed (Bonniwell et al. 1999; Hoffmann 2015). Residence time for sediment is often computed as the mass or volume of sediment in a storage reservoir divided by the average sediment transport rate (Kelsey et al. 1987; Malmom et al. 2002). The residence time of sediment can refer to the time spent in a discrete depositional reservoir, i.e., bed or floodplain (Skalak and Pizzuto 2010; Fryirs 2013), or the entire watershed (Dominik et al. 1987; Evrard et al. 2010). At the watershed scale, sediment residence time would include time spent in both storage and transport through its journey down the fluvial system (Pizzuto et al. 2014). For this paper, the transit time is defined as the time from when sediment is delivered to the channel to when it arrives at a (downstream) sample location. The transit time includes the time sediment is in transport and channel storage but does not include the time spent in floodplain or upland storage.

The U.S. Geological Survey did an intensive study of water quality and ecology in 100 wadeable midwestern US streams in 2013 (Van Metre et al. 2018). In 99 of those streams, Gellis et al. (2017) used ^7Be to age fine-grained suspended sediment and bed material to < 1 year and $^{210}\text{Pb}_{\text{ex}}$ to apportion sediment sources between land surface and channel (streambanks and sediment in channel storage). Results indicated that channel sources (streambanks and sediment in channel storage) dominate; 79% of the sites had > 50% channel-derived sediment. The age of the surface-derived fraction of bed sediment ranged from 0 to 174 days with a median of 82 days and suspended-sediment sample ages ranged from 0 to 84 days with a median of 41 days. Gellis et al. (2017) noted that their approach could not determine whether the channel-derived sediment originated from streambanks or from older topsoil-derived sediment that had been in channel storage for several decades or longer. One of the 99 streams was Walnut Creek in central Iowa and is the subject of this study. Our objective was to combine the sediment fingerprinting approach, which apportions sediment into channel banks and surface-derived sediment (pasture, prairie, cropland, and unpaved roads) based on

elemental analyses, with fallout radionuclides to determine ages of the surface-derived portion of this sediment.

1.1 Sediment source determination

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. Sediment sources are typically separated into channel sources (streambanks, channel bed, and bars) and land-surface sources (classified by land use, land cover, or by geology and soil units) (Collins and Walling 2002; Collins et al. 2017). Properties that have been used to identify sediment sources include elemental analysis (Collins et al. 2012a; Gellis and Noe 2013), radionuclides (Belmont et al. 2014; Evrard et al. 2016), stable isotopes (Fox 2009; Lacey et al. 2015), compound-specific stable isotope (CSSI) analysis (Zhang et al. 2017), and soil enzyme activity (Nosrati et al. 2011).

Target sediment samples collected at a defined watershed outlet represent a composite of the source properties (fingerprints). Target sediment can be suspended sediment, channel bed, and floodplain deposits (Walling 2005; Martínez-Carreras et al. 2010; Collins et al. 2012b; Haddadchi et al. 2013; Miller et al. 2015; Gellis et al. 2016). Target samples can also include lake and reservoir cores, which enable interpretations of historical changes in sediment source from natural and anthropogenic forcings (Foster and Walling 1994; Foster et al. 2008; Belmont et al. 2011; D'Haen et al. 2013; Manjoro et al. 2017).

The apportionment of sediment sources contributing to the target sediment is determined using various statistical procedures (Collins et al. 2010; Haddadchi et al. 2013; Miller et al. 2015). Generally, two statistical approaches to apportion target sediment to its sources have appeared in the literature: a tracer reduction approach and a Bayesian approach. The tracer reduction approach uses a multi-step procedure, beginning with tests to remove potential tracers that cannot differentiate between sources using tests such as a Mann-Whitney or Kruskal-Wallis, then using stepwise discriminate function analysis (DFA) to identify an optimum composite fingerprint (Collins et al. 1997, 1998; Martínez-Carreras et al. 2010; Gellis and Noe 2013). The Bayesian approach involves probabilistic treatment of the data which allows for understanding uncertainties in the spatial and temporal variability in source and riverine sediment geochemistry (Fox and Papanicolaou 2008; Stewart et al. 2014; Cooper and Krueger 2017). Error analysis to test the sediment fingerprinting results has included a relative error comparison or goodness of fit (GOF) model (Collins et al. 1998; Manjoro et al. 2017) where the sample fingerprint properties of the target samples are compared with the corresponding values predicted by the model. Motha et al. (2003) and Collins and Walling (2007a) used a Monte Carlo

design where samples of the tracer properties are randomly drawn from their corresponding normal distributions and run through the mixing model to test variability in source apportionment results.

1.2 Age determination

Short-lived fallout radionuclides have been used to examine sediment residence times in watersheds and ages of channel sediment (Dominik et al. 1987; Wallbrink et al. 2002; Evrard et al. 2010; Mabit et al. 2014). Wallbrink et al. (2002), using $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs , estimated residence times of sediment in the channel for two river systems in Australia ranging from 0 to 21 years and 0 to 9 years. Dominik et al. (1987), using ^7Be , $^{210}\text{Pb}_{\text{ex}}$, and ^{137}Cs , proposed a two-box age model of residence times for the alpine Rhône River, with a geologic box where a component of topsoil particles travel slowly (800 to 1400 years) and a rapid box where high surface erosion rates for a small fraction of the sediment moves particles at short residence times between 1 and 220 days. Evrard et al. (2010) examined sediment residence times in a tropical watershed in central Mexico using ^7Be , $^{210}\text{Pb}_{\text{ex}}$, and ^{137}Cs and concluded that residence times were similar to the two-box model of Dominik et al. (1987); a geologic box where soil is transported to the watershed outlet at time scales of 5000 to 23,000 years and a rapid box, where once in the channel, sediment travels to the monitoring point from 50 to 200 days.

2 Materials and methods

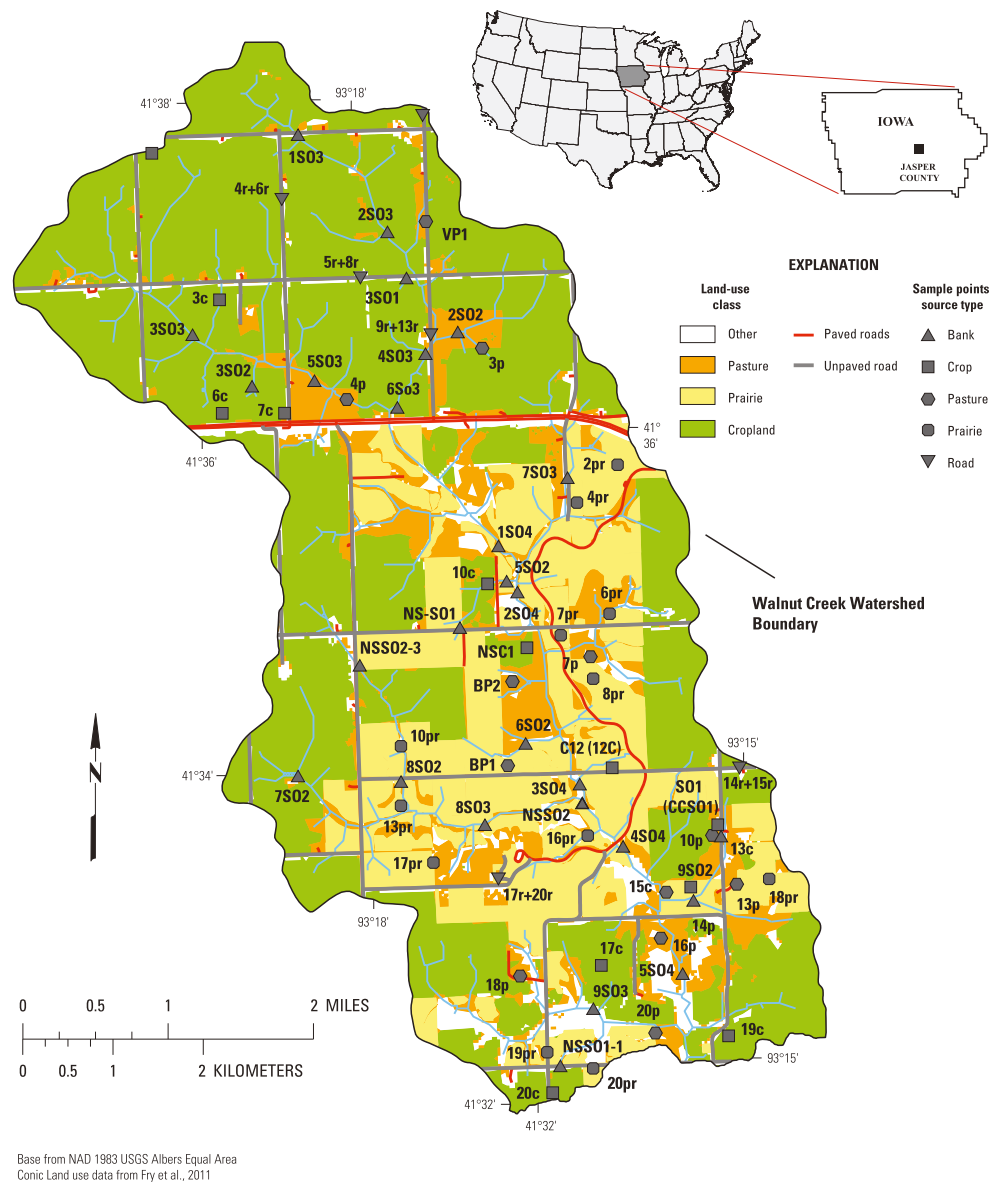
2.1 Study area

The 52.6-km² Walnut Creek watershed as defined here is a 4th-order system draining south-central Iowa in Jasper County (Fig. 1). The climate is humid continental with an average annual precipitation of 750 mm (Palmer et al. 2014). Soils in the watershed are primarily silty clay loams, silt loams, or clay loams formed in loess and till (Schilling et al. 2011). Upland areas in Walnut Creek are loess mantled pre-Illinoian till with Holocene alluvial fills (Schilling et al. 2011). Legacy or post-Euro-American-settlement sediment (nineteenth century) is common in the valley fills where channel banks are primarily composed of silt and clay (Schilling et al. 2011).

Walnut Creek is an agricultural basin, with 65% of the watershed in cropland, 19% in prairie, 8% in pasture, and 8% in developed land and other (Homer et al. 2015). The length of unpaved roads in the watershed obtained from 2000 and 2013 coverages (U.S. Census Bureau 2001; U.S. Department of Agriculture 2014) was 53 km or an unpaved road density of $\sim 1 \text{ km}^{-2}$.

Walnut Creek is one of the 99 streams selected as part of the U.S. Geological Survey's (USGS) Midwest Stream Quality

Fig. 1 Walnut Creek, Iowa watershed study area showing location of sediment source samples and land use (Fry et al. 2011)



Assessment (MSQA), a study conducted by the USGS National Water-Quality Assessment (NAWQA) (Van Metre et al. 2012, 2018). The overall goals of the MSQA were to characterize water-quality stressors—contaminants, nutrients, sediment, streamflow, and habitat and ecological conditions in streams across the Midwest Region and to determine the relative effects of these stressors on aquatic organisms.

2.2 Flow, turbidity, and suspended sediment

Instantaneous discharge (10 min intervals), instantaneous suspended sediment (selected times), and turbidity (30 min intervals) were obtained from the U.S. Department of Agriculture, Walnut Creek near Vandalia, Iowa streamgage (discontinued USGS Station ID 05487550; herein called the streamgage) for the MSQA study period (23 May 2013 to 8

August 2013). Discharge, stage, and record computation followed USGS protocols and operating procedures (Kennedy 1983) which can be found in Schilling (2000). Suspended sediment was collected at least once a week during low flows using a manual dip sample, and at higher flows using isokinetic samplers with equal width increments (EWI) (Edwards and Glysson 1999). Event samples were collected using an automatic sampler to collect flow-based event composite samples comprising 100 mL subsamples of stream water taken on a discharge-volume interval of one subsample for every 18,406 m³ of flow. The 100-mL subsamples were composited in 3.8 L bottles; multiple bottles were available for large events. When the event was finished, the bottles were put in a churn splitter for mixing and a subsample was taken for suspended sediment. EWI sample results were used to determine correction coefficients to remove the bias from the

grab and automatic samples. The method bias coefficient (EWI:sample) was calculated to be 0.50 for the automatic sampler suspended-sediment concentration (SSC) and 0.93 for the dip sample SSC. Suspended sediment was analyzed for concentrations at the USDA laboratory in Ames, Iowa following procedures and protocols in ASTM (2002).

Turbidity was recorded at the streamgage every 30 min using a Hydrolab DS5X sonde following standards outlined in Wagner et al. (2006). A regression model of turbidity and suspended sediment concentration (SSC) developed by USDA was used to estimate SSC during periods of missing record. A mean daily SSC was determined for low flow periods and events. On low flow days where one or more samples of SSC were obtained, samples were averaged to obtain a mean daily SSC. For periods of low flow where a SSC sample was not obtained, 10 min intervals of SSC were estimated using the time series management system (Water Information Systems by KISTERS (WISKI), Kisters North America, Citrus Heights, California, USA; www.kisters.net). For high flow days and events, SSC was estimated from the automatic sampler-collected composite sample. If the automatic sampler malfunctioned, the relation of SSC and turbidity and 30 min turbidity values were converted to SSC and averaged for the day.

Suspended-sediment daily loads were computed using the mean daily SSC and mean daily discharge, as follows:

$$SS_{\text{daily}} = \text{Flow}_{\text{daily}} \times \text{SSC}_{\text{daily}} \times 0.0864 \quad (1)$$

where SS_{daily} is the daily suspended-sediment load (Mg day^{-1}), $\text{Flow}_{\text{daily}}$ is the mean daily flow ($\text{m}^3 \text{s}^{-1}$), $\text{SSC}_{\text{daily}}$ is the mean daily SSC (mg L^{-1}), and 0.0864 is a conversion coefficient based on the unit of measurement of water discharge that assumes a specific weight of 2.65 for sediment (Gray and Simões 2008).

Suspended sediment was collected for tracer analyses using passive samplers (Phillips et al. 2000) that were deployed over 14 weeks and sampled periodically (Table 1). Two pairs of PVC tubes mounted on steel struts were placed in the center of the channel with the upper tube placed on top of the lower tube. This placement of the tubes allowed flows over a range in stage to be sampled (Fig. S1, Electronic supplementary material). Sediment was retrieved from the passive samplers biweekly or after storm events and both tubes were composited into one sample; referred to herein as target samples.

The total suspended-sediment load was computed by summing the daily load for the period of each target sample and for the period of study (Table 1). Because suspended-sediment load is computed as a daily and not an hourly load, the sediment load transported for each sample period is determined from the day the sampler was deployed to the day prior to the next retrieval. The weighted sediment load is determined by dividing the sediment load for each sample by the total load for the period of study and applying this weighting to the

apportionment results for each sample (Walling et al. 1999; Gellis et al. 2015). Target sample 8 retrieved on 1 August 2013 did not have enough mass ($< 1 \text{ g}$) for radionuclide analysis; therefore, the period from 24 July 2013 to 1 August 2013 was not used to compute a total sediment load for this timeframe.

2.3 Sampling and sediment fingerprinting

The sediment-fingerprinting approach was used to determine the sources of fine-grained ($< 63 \mu\text{m}$) sediment in the Walnut Creek watershed draining to the streamgage. Nine target samples were used for source analysis between 23 May 2013 to 8 August 2013 (Table 1; sample 8 was not used). The period was chosen because it coincided with the USGS National Water Quality Assessment sampling for streams in the Midwest Region of the USA that included Walnut Creek (Van Metre et al. 2012).

Sediment-source samples were collected from pasture ($n = 12$), cropland ($n = 13$), prairie ($n = 12$), channel banks ($n = 26$), and unpaved roads ($n = 6$) (Fig. 1). A similar number of source samples were collected for sediment fingerprinting studies in agricultural watersheds of Chesapeake Bay (Gellis et al. 2009, 2015). Source sample locations were selected by rasterizing each of the source areas using a GIS and assigning a unique value to each cell for each source. A random number generator was used to select individual grid cells within each land use for sampling. The randomized design avoided bias in site selection and provided a random sample that is representative of land use in Walnut Creek.

Cropland, prairie, and pasture are present in Walnut Creek as large fields (10s–100s ha). In the randomly selected cropland, prairie, and pasture sites, portions of the entire field were selected for sampling that were representative of the entire field, and topsoil was collected across three to five transects each parallel to slope, $\sim 30 \text{ m}$ apart and $\sim 100 \text{ m}$ long. At each transect, topsoil was collected every 10 m from the top $\sim 1.0 \text{ cm}$ of the soil surface with a plastic hand shovel. All samples from a given site were mixed into a single sample in the field.

Each unpaved road sample was collected across three transects spaced 10 m apart that traversed the road using a hand brush and plastic dustpan and composited into one sample. Eroding streambanks were sampled by scraping the entire vertical face of the exposed streambank to a depth of $\sim 1 \text{ cm}$ 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. The channel bed is assumed to be a temporary storage reservoir of sediment originating from a variety of upstream sources (Gellis et al. 2015) and was not sampled as a source.

After collection, source samples were put on ice and transported to the Iowa State University Soil and Plant Analysis Laboratory, Ames, Iowa. Target samples were shipped

Table 1 Summary of suspended-sediment sample dates and times, including mean daily flow and sediment loads for Walnut Creek, Iowa (Gellis et al. 2018)

Sample type	Target sample No.	Sample period and times	Collection duration (h)	Meandaily flow(m ³ s ⁻¹)	Peak flow (m ³ s ⁻¹) during collection period	Date and time of peak flow	Total suspended-sediment load (Mg)	Mean suspended-sediment concentration (mg L ⁻¹)
Suspended sediment	1	23 May 2013 at 08:45–6 June 2013 at 11:00	338.5	2.71	4.14	5 June 2013 at 04:30	4868	838
Suspended sediment	2	6 June 2013 at 11:00–13 June 2013 at 09:15	166.5	0.92	1.24	9 June 2013 at 21:20	101	132
Suspended sediment	3	13 June 2013 at 09:15–20 June 2013 at 11:00	169.8	1.14	7.04	15 June 2013 - at 03:00	385	312
Suspended sediment	4	20 June 2013 at 11:00–27 June 2013 at 11:00	168.2	0.82	4.45	24 June 2013 - at 17:50	69.9	108
Suspended sediment	5	27 June 2013 at 11:00–11 July 2013 at 08:45	333.8	0.40	All baseflow no events	All baseflow no events	30.8	52
Suspended sediment	6	11 July 2013 at 08:45–18 July 2013 at 9:50	169.3	0.18	All baseflow no events	All baseflow no events	4.8	35
Suspended sediment	7	18 July 2013 at 9:50–24 July 2013 at 12:45	147.0	0.12	All baseflow no events	All baseflow no events	1.4	20
Suspended sediment	8	24 July 2013 at 12:45–1 August 2013 at 09:20	188.8	0.07	All baseflow no events	All baseflow no events	2.3	38
Suspended sediment	9	1 August 2013 at 09:20–8 August 2013 at 1000	168.8	0.04	All baseflow no events	All baseflow no events	1.8	62

Because of low mass (<1 g), target sample 8 was not used in this analysis

on ice to the USGS MD-DC-Water Science Center in Baltimore, Maryland. Source and target samples were wet sieved with de-ionized water through a 63-µm polyester sieve to remove the sand and dried at 60 °C. The silt and clay portion (< 63 µm) of the source and target samples were split for three analyses: (1) elemental analysis using inductively coupled plasma optical emission spectrometry (ICP-OES) and inductively coupled plasma mass spectrometry (ICP-MS); (2) grain size, and (3) radionuclide analysis (⁷Be and ²¹⁰Pb_{ex}) on target samples and for selected source samples (²¹⁰Pb_{ex}).

Elemental analyses on the source and target samples were conducted at the USGS Central Mineral and Environmental Resources Science Center in Denver, Colorado, which reported 38 elemental concentrations for the source tracers and target samples used in sediment fingerprinting (Table 2; Gellis et al. 2018). Samples were dried and ground then digested at low temperature using a mixture of hydrofluoric and hydrochloric-nitric-perchloric acids, with analysis by ICP-MS (Briggs and Meier 2002). ICP-MS and ICP-OES data are acceptable if recovery of each element in reference materials analyzed with samples is within ± 15% of reference materials at five times the lower limit of determination (LOD) and if the calculated relative standard deviation (RSD) of replicates is within 15% (<https://minerals.usgs.gov/science/analytical-chemistry/method17.html>). Ten samples from the study were split and analyzed in replicate using the same

ICP-MS and ICP-OES elemental methods as used here (Gellis et al. 2018). Samples with non-detects were not included in determining a RPD. The median relative percent difference (RPD) for the 38 elements used in sediment fingerprinting had a median RPD ranging from 0.0% (Cs) to 30.7% (Sb), and the median of the medians was 4.2%, indicating relatively good precision.

Table 2 Tracers used in sediment fingerprinting

Aluminum	Gallium	Scandium
Antimony	Iron	Silver
Arsenic	Lanthanum	Sodium
Barium	Lead	Strontium
Beryllium	Lithium	Thallium
Bismuth	Magnesium	Thorium
Cadmium	Manganese	Titanium
Cesium	Molybdenum	Total organic carbon (TOC)
Calcium	Nickel	Uranium
Cerium	Niobium	Vanadium
Chromium	Phosphorus	Yttrium
Cobalt	Potassium	Zinc
Copper	Rubidium	

Particle size determination was performed at the USGS MD-DE-DC Water Science Center in Baltimore, Maryland using a Laser In Situ Scattering Transmissometer (LISST-100X). The LISST-100X uses laser diffraction to measure the size of sediment particles (instrument specifications can be found at <http://www.sequoiasci.com/library/standards/>; accessed 3 January 2017). Prior to analysis, fine sediment (< 63 μm) undergoes the following preparation procedure: an aliquot (0.0210–0.0300 g) of each sample is transferred into a 125-mL Erlenmeyer flask. A glass pipet is used to add 10 mL of a 50-g L^{-1} solution of hexametaphosphate (NaPO_4)₆ to each sample to aid in deflocculation. Each sample is placed in an ultrasonic bath for 5 min to disperse the particles and then put on a shaking table for a minimum of 16 h. The results are expressed as the median particle diameter (D_{50}) of the sediment by the LISST software (Gellis et al. 2018).

2.4 Sediment-source apportionment using the sediment-fingerprinting approach

Several analytical and statistical steps were used to determine which tracers are most effective in defining sediment sources using the sediment-fingerprinting approach (Gellis et al. 2015, 2016). The sediment source assessment tool (Sed_SAT) was used to assist the user through all necessary statistical steps in sediment fingerprinting (Gorman Sanisaca et al. 2017). Sed_SAT is written in the statistical language R (R Core Team 2016) using Microsoft Access© (Gorman Sanisaca et al. 2017; available at <https://my.usgs.gov/bitbucket/projects/SED>). A full description of the decision tree is available within Gellis et al. (2016) and Gorman Sanisaca et al. (2017). The statistical defaults in Sed_SAT were used to apportion sediment in Walnut Creek. Sed_SAT follows a five-step procedure to apportion sediment: (1) option to remove outliers, (2) option to perform grain size and organic content corrections to the source data, (3) a bracket test for the conservativeness of the tracer, (4) stepwise discriminant function analysis (DFA), and (5) percent contribution from each source using an “unmixing model” (modified from Collins et al. 2010) as follows:

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

and

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

where RE is the relative minimum error term; C_i is the concentration of tracer property after size and organic correction factors are applied (i) to the suspended sediment sample; P_s is the the relative contribution from source category (s); S_{si} is the mean concentration of tracer property

(i) 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

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

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

The unmixing model minimizes the relative error term in Eq. (2) through an optimization procedure using the `optim()` function in the programming language R.

In Walnut Creek, each target sample was apportioned to five sources: streambanks, cropland, pasture, prairie, and unpaved roads. Source percentages are presented three ways: (1) as an average for the entire study period, (2) by weighting the sediment load transported for each sample’s time period relative to the total sediment load of all samples, and (3) by the sediment concentration for each sampling period. Weighting the sediment-fingerprinting results by the sediment load is as follows:

$$\text{Collection}_{\text{wt}(n)} = \frac{\text{SSmass}_n}{\sum_{i=1}^n \text{SSmass}_i} \quad (5)$$

$\text{Collection}_{\text{wt}(n)}$ is the weight given to the sediment load transported for each sample’s collection period n ; SSmass_n is the summed daily sediment load for each sample’s collection period; n is determined using the computation method in Eq. (1); and SSmass_i is the summed sediment loads transported for each sample i (from 1 to n). Note that this total does not include the period 24 July 2013 at 12:45–1 August 2013 at 09:20, where the mass of sediment sampled was too low to analyze. The final storm-weighted source percentages are determined as follows:

$$S_v = \sum_{(i=1)}^n [\text{SA}_{vi} \times \text{Collection}_{\text{wt}(n)}] \quad (6)$$

S_v is the storm-weighted source apportionment (in percent) for source (v) (v includes channel banks, cropland, prairie, pasture, or unpaved roads); SA_{vi} is the sediment source apportionment from the sediment-fingerprinting results (in percent) for source (v) and sample i ; and n is the number of samples (i) collected during sampling period = 8.

Apportionment of sediment sources is also presented relative to the suspended-sediment concentration (mg L^{-1}) for

each sample by taking the mean daily suspended-sediment concentration for each sampling period (Gellis et al. 2018) and multiplying it by the apportionment results for each sample.

2.5 Uncertainty in the sediment-fingerprinting approach

Uncertainty tests in the sediment-fingerprinting results determined for each target sample include: (1) the confusion matrix, (2) Monte Carlo uncertainty analysis, (3) the source verification test (SVT), and (4) a Monte Carlo leave-one-out cross validation. The confusion matrix is an output of stepwise DFA that shows the number of source samples predicted for each group versus the actual number of source samples in each group (Provost and Kohavi 1998). It is a measure of how well the tracers can discriminate the sources.

A Monte Carlo uncertainty analysis was used to quantify the uncertainties associated with the optimized sediment source contributions predicted by the unmixing model (Eq. (2)) (Collins and Walling 2007b; Martínez-Carreras et al. 2010). To perform an uncertainty analysis some of the statistical properties of initial source data were used to generate random deviates. The sample median and robust-scale estimator (Q_n) were proposed to generate corresponding normal distribution following Collins et al. (2012b) and Zhang et al. (2017). The present method is an alternative to the more common method of incorporating mean and standard deviation as shape and scale estimators. The notation of random deviate is:

$$X \sim N(M, Q_n) \quad (7)$$

where X is a random deviate of the sediment property of normal distribution with shape estimator—median (M) and a robust-scale estimator (Q_n).

Incorporating both a random number generator and an inverse cumulative distribution function (quantile function) with obtained property statistics generated $n = 1000$ samples for each Monte Carlo iteration.

The SVT is designed to see how well the final set of tracers discriminates the sources. Each source sample is entered as a target sample into the unmixing model to determine how well it can correctly identify each source sample. Because soil properties in some land uses can often be similar (i.e., pasture and cropland), the SVT can be used to decide whether to combine source samples into one category (i.e., pasture and cropland into agriculture) (Gellis et al. 2015). In addition, if a sample is misclassified (e.g., < 50% of that source), the user has the option to remove this sample and start the process again.

The Monte Carlo leave-one-out cross validation was used to quantify the sensitivity of the sediment-fingerprinting results to the removal of samples (Gellis et al. 2016). In

Sed_SAT, the Monte Carlo leave-one-out cross validation randomly removes one sample from each of the source groups, which affects the group means, and the unmixing model is run without these samples (Gorman Sanisaca et al. 2017). The Monte Carlo simulation is run 1000 times on each target. For each target sample, the final unmixing model results are compared with the difference from two outputs in the Monte Carlo leave-one-out cross validation output; (1) the average of the Monte Carlo results and (2) the minimum and maximum source percentage results in the Monte Carlo results.

The robustness of the source ascription results for both Monte Carlo tests were assessed using a “goodness of fit” (GOF) (Collins and Walling 2007a, b; Manjoro et al. 2017) where the sample fingerprint properties of the target samples are compared with the corresponding values predicted by the model, as follows:

$$\text{GOF} = 1 - \sqrt{\left(\frac{\sum_{i=1}^n \left\{ \left(C_i - \left(\sum_{s=1}^m P_s S_{si} \right) / C_i \right)^2 W_i \right\}}{n} \right)} \quad (8)$$

where all variables are defined in Eq. (2).

The GOF test is run for each iteration of both the Monte Carlo uncertainty analysis and the Monte Carlo leave-one-out cross validation. GOF values > 0.85 are considered acceptable values (Sherriff et al. 2015).

2.6 Sediment age determinations

The age of the topsoil-derived portion (pasture, prairie, or cropland) of each target sample was determined using the radionuclides ^7Be and ^{210}Pb for two age classes: ^7Be up to ~1 year, and ^{210}Pb up to ~85 years. ^7Be (half-life of 53.3 days) is a naturally occurring radionuclide produced in the upper atmosphere by cosmic ray spallation of nitrogen and oxygen. It attaches to airborne particulates and reaches the earth surface mostly through precipitation, where ^7Be -bearing particles sorb to fine sediment (Baskaran et al. 1993). ^7Be is typically found in the top centimeter of soil (Baskaran et al. 1993) and is negligible in streambanks.

Lead-210 (half-life 22.3 years) is a naturally occurring radionuclide of lead in the ^{238}U decay series. ^{238}U decays to ^{226}Ra , which in turn decays to the noble gas ^{222}Rn . Evasion of some of the ^{222}Rn from continental land masses to the atmosphere occurs where it ultimately decays to ^{210}Pb that attaches to airborne particulates and aerosols. Atmospheric fallout of this ^{210}Pb enriches surface soils in ^{210}Pb above the level “supported” by decay of ^{226}Ra in geologic materials; the unsupported fraction is commonly referred to as excess ^{210}Pb ($^{210}\text{Pb}_{\text{ex}}$). $^{210}\text{Pb}_{\text{ex}}$ is primarily delivered to the earth surface in the form of atmospheric aerosols during rainfall events that sorb strongly to fine sediment (Baskaran et al. 1993). The $^{210}\text{Pb}_{\text{ex}}$ decays at a known rate until only the supported ^{210}Pb remains, providing a date marker for when surface soils

and sediments were last exposed to direct fallout, if the initial activity of $^{210}\text{Pb}_{\text{ex}}$ is known. The activity of $^{210}\text{Pb}_{\text{ex}}$ has been shown to be higher in topsoil relative to streambanks (Matisoff et al. 2005; Hancock et al. 2014) and is dependent on fallout rate and the amount of post-fallout disturbance of the soil (e.g., erosion losses, mixing by plowing). Although unpaved road material does obtain some ^7Be and $^{210}\text{Pb}_{\text{ex}}$, because it is brought in from nearby quarries and applied periodically, we did not perform age dating on this surface source.

Radionuclide analysis for ^7Be , and $^{210}\text{Pb}_{\text{ex}}$, were conducted at the USGS Sediment Radioisotope Laboratory in Menlo Park, California, using high-resolution germanium detector gamma spectrometers following methods described in Fuller et al. (1999) and Van Metre et al. (2004). Measured activities of ^7Be were corrected for radioactive decay from the date of sample collection to the date of analysis. Excess $^{210}\text{Pb}_{\text{ex}}$ is the difference between the measured total ^{210}Pb and ^{226}Ra , which is determined from the short-lived intermediate gamma-emitting isotopes ^{214}Pb and ^{214}Bi . Method detection limits (MDLs) for ^7Be and $^{210}\text{Pb}_{\text{ex}}$ were 6.7 mBq g^{-1} . Because of differences in times between sample collection and analysis, the sample-specific detection limit for ^7Be was determined by correcting the MDL for decay, which varies among samples (11.7 to 31.7 mBq g^{-1}).

The reported 1-sigma uncertainty in the measured radionuclide activity (s1) was calculated from the random counting error of samples and background standard spectra at the 1 standard deviation level. Uncertainty in measured activity was typically within $\pm 10\%$ of the measured activity for total ^{210}Pb and ^{226}Ra and $\pm 18\%$ for ^7Be . Uncertainty in $^{210}\text{Pb}_{\text{ex}}$ was propagated from the uncertainties in total ^{210}Pb and ^{226}Ra activity and averaged $\pm 20\%$.

To obtain $^{210}\text{Pb}_{\text{ex}}$ radionuclide activities of sediment source types in Walnut Creek (streambanks, pasture, prairie, and

cropland), the samples used in sediment fingerprinting were composited into fewer samples and sent for analysis (Gellis et al. 2018). The age dating approach used here relies on the activity of $^{210}\text{Pb}_{\text{ex}}$ in the target sample that originated from topsoil (pasture, prairie, and cropland). Radionuclide activities are present in lower amounts in streambanks and unpaved roads and were subtracted from activities on the target samples as shown in the following equations. Because of the time constraints in analyzing ^7Be , source samples were not analyzed for ^7Be . The activity of ^7Be in topsoil uses the estimate found in surface soils obtained for the Midwestern USA (Gellis et al. 2017 and references therein).

^7Be is first corrected to the percent of surface-derived sediment as follows:

$${}^7\text{Be}_{(\text{corr})} = \frac{{}^7\text{Be}_{(\text{target})}}{(\text{surface}\%/100)} \quad (9)$$

where ${}^7\text{Be}_{(\text{corr})}$ is the estimated surface ^7Be activity (mBq g^{-1}); ${}^7\text{Be}_{(\text{target})}$ is the measured ^7Be activity in the target sample (suspended sediment) (mBq g^{-1}); and surface% is the percentage of surface-derived sediment from the sediment fingerprinting results (pasture + crop + prairie + unpaved roads).

The age of the target sediment up to ~ 1 year is determined as:

$${}^7\text{Be}_{(\text{age})} = \frac{\text{Ln}\left(\frac{{}^7\text{Be}_{(\text{corr})}}{{}^7\text{Be}_{(95)}}\right)}{-\lambda {}^7\text{Be}} \quad (10)$$

where ${}^7\text{Be}_{(\text{age})}$ is the age of topsoil-derived sediment (days); ${}^7\text{Be}_{(\text{corr})}$ is the estimated surface ^7Be activity (mBq g^{-1}) (Eqs. 8 and 9); ${}^7\text{Be}_{(95)}$ is the estimated surface material ^7Be activity for Midwestern US soils (542 mBq g^{-1}) (Gellis et al. 2017); and $\lambda_{7\text{Be}}$ is the decay constant for $^7\text{Be} = 0.01305 \text{ day}^{-1}$.

The estimated topsoil activity of $^{210}\text{Pb}_{\text{ex}}$ is determined as:

$${}^{210}\text{Pb}_{\text{ex}(\text{corr})} = \frac{\left[{}^{210}\text{Pb}_{(\text{target})} - \left({}^{210}\text{Pb}_{(\text{roads})} \times \frac{\text{source}\%(\text{roads})}{100} + {}^{210}\text{Pb}_{(\text{banks})} \times \frac{\text{source}\%(\text{banks})}{100} \right) \right]}{(\text{surf_topsoil}\%/100)} \quad (11)$$

where ${}^{210}\text{Pb}_{\text{ex}(\text{corr})}$ is the estimated surface $^{210}\text{Pb}_{\text{ex}}$ activity (mBq g^{-1}); ${}^{210}\text{Pb}_{\text{ex}(\text{target})}$ is the measured ^{210}Pb activity in the target sample (suspended sediment) (mBq g^{-1}); ${}^{210}\text{Pb}_{(\text{roads})}$ is the mean ^{210}Pb activity in unpaved roads (Gellis et al. 2018); source%(roads) is the sediment-fingerprinting results in percent for unpaved roads; ${}^{210}\text{Pb}_{(\text{banks})}$ is the mean ^{210}Pb activity in streambanks (Gellis et al. 2018); source%(banks) is the sediment-fingerprinting results in percent for streambank; and surf_topsoil% is from Eqs. 8 and 9.

The age of target sediment using $^{210}\text{Pb}_{\text{ex}}$ up to ~ 85 years, is as follows:

$${}^{210}\text{Pb}_{\text{ex}(\text{age})} = \frac{\text{Ln}\left(\frac{{}^{210}\text{Pb}_{\text{ex}(\text{corr})}}{{}^{210}\text{Pb}_{\text{ex}(\text{surf})}}\right)}{-\lambda {}^{210}\text{Pb}_{\text{ex}}} \quad (12)$$

where ${}^{210}\text{Pb}_{\text{ex}(\text{age})}$ is the age of topsoil-derived sediment (days); ${}^{210}\text{Pb}_{\text{ex}(\text{corr})}$ is the estimated surface $^{210}\text{Pb}_{\text{ex}}$ activity (mBq g^{-1}) (Eq. 12); ${}^{210}\text{Pb}_{\text{ex}(\text{surf})}$ is the weighted surface

activity of $^{210}\text{Pb}_{\text{ex}}$ (75.5 mBq g^{-1}) from samples in Walnut Creek (Gellis et al. 2018); and $\lambda_{210_{\text{ex}}}$ is the decay constant for $^{210}\text{Pb}_{\text{ex}} = 8.50999 \times 10^{-5} \text{ day}^{-1}$.

For $^{210}\text{Pb}_{\text{ex}}$ and ^7Be , age as defined here is the transit time between when sediment enters the channel from a surface source and when the target sample is collected. Sediment that does not contain measurable quantities of $^{210}\text{Pb}_{\text{ex}}$ is considered to be older than ~ 85 years. This was determined by entering the MDL for $^{210}\text{Pb}_{\text{ex}}$ into Eq. (12).

3 Results

3.1 Flow and suspended sediment

The first half of the sampling period (23 May 2013 to 1 July 2013) was a period of higher flows (Fig. 2). This period of sampling accounted for 57% of the total flow and 89% of the total suspended-sediment load, where SSCs ranged from 6.4 to 3204 mg L^{-1} (Gellis et al. 2018). The second half of the sampling period corresponds to summertime baseflow with SSCs ranging from 13 to 149 mg L^{-1} (Gellis et al. 2018).

3.2 Sediment-fingerprinting results and uncertainty

The unmixing model showed that sources varied between suspended-sediment samples and that cropland was the largest source of sediment on average (48%) and weighted by sediment loads (62%) (Fig. 3; Table 3). Based on weighting by sediment loads, channel banks accounted for most of the remainder of the sediment (36%), with prairie, pasture, and unpaved roads each contributing $\leq 1\%$ (Table 3). For the two sampling periods with the highest mean sediment concentration (target samples 1 and 3, means of 838 and 312 mg L^{-1} , respectively), cropland was the largest source (61 and 71%), followed by streambanks (37 and 27%) (Fig. 3b; Table 3).

The median grain size (D_{50}) of the $< 63 \mu\text{m}$ fraction of target samples was finer than most source samples and the TOC in target samples was higher than most of the source samples (Fig. 4).

Nineteen of the thirty-eight tracers (elements) had at least one source group corrected for grain size and twenty-one tracers had at least one source group corrected for organic content (Table S1A, B, Electronic supplementary material). Only one tracer, uranium, was corrected for both size and organic corrections for the same source group (unpaved roads) (Table S1A, B, Electronic supplementary material). Smith and Blake (2014) suggest that applying both a size and organic correction to a tracer can result in over correction. Because only 1 of the 38 tracers, uranium, was corrected for both size and organics, over correcting the data was not a concern.

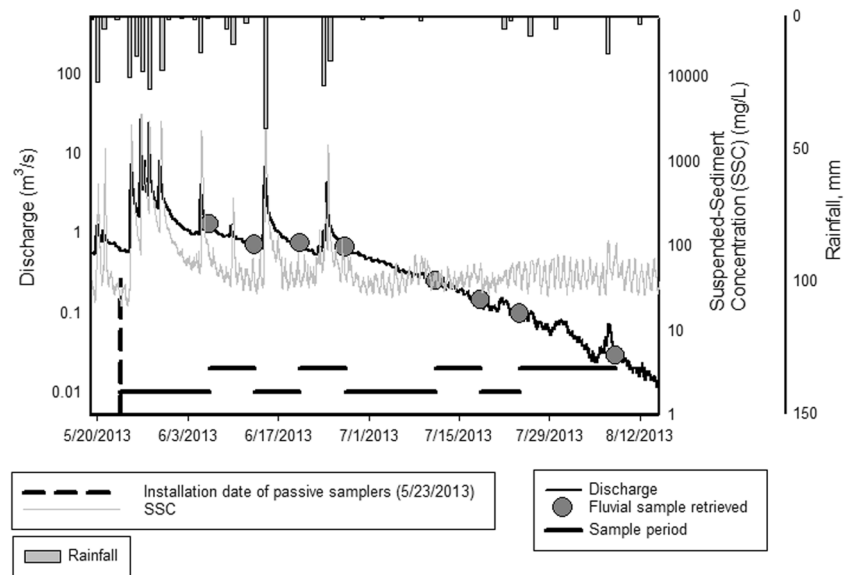
Results of stepwise DFA indicated that five of the tracers (Ca, Mo, Na, Sc, and TOC) were significant for all target samples with other elements being significant for one or more target samples (Fig. 5; Table S2, Electronic supplementary material). It is important to point out that a different set of tracers can be significant in discriminating the sources for any given target sample (Table S2, Electronic supplementary material). Because the size and organic content of each target sample affects the final corrected concentration of source samples, depending on the grain size and organic content of the target sample, different tracers may be significant.

Averaging all results in the confusion matrix shows that streambanks and unpaved roads have 100% of the samples classified correctly, with most samples classified correctly for pasture (87%), crop (99%), and prairie (93%) (Fig. 6). The SVT results are used to determine how well the final tracers selected in stepwise DFA can identify the source samples (Table 4; Table S3, Electronic supplementary material). Results of the SVT indicate that all sources were classified as $> 73\%$ of their source type; with prairie (73%), pasture (77%), crop (78%), and banks and unpaved roads (97%) (Table 4). Misclassified cropland samples were classified as banks (8%), pasture (4%), prairie (9%), and unpaved roads (1%) (Table 4). The majority of misclassified pasture samples were classified as prairie, and the majority of misclassified prairie samples were classified as pasture (Table 4).

Results of the Monte Carlo uncertainty analysis indicate that the cumulative distribution functions produced mean apportionment results that ranged from 0 to 8% of the original unmixing model results (Table 3). The Monte Carlo uncertainty analysis showed that confidence intervals (5 and 95 percentiles) were less than 10% except for three source apportionments which showed uncertainty of 11, 14, and 16% (Table 3; Fig. S2, Electronic supplementary material). The GOF test for all samples were > 0.90 indicating that the model provided reliable estimates of the relative contributions from the individual source types (Table 3).

The Monte Carlo leave-one-out cross validation produces a range in source group means which affects the unmixing model results. The average of the 1000 Monte Carlo iterations for each target sample were within 0 to 8% of the unmixing models results (Fig. S3, Electronic supplementary material). Six of the eight target samples showed $< 16\%$ difference from the unmixing model results to the minimum and maximum of any Monte Carlo iteration with four of the eight target samples showing a $< 9\%$ difference. Target sample 4 had 19% of the 1000 Monte Carlo iterations with a maximum difference to the unmixing model of $> 10\%$ and all occurring for pasture and prairie samples. Target sample 6 had 59% of the 1000 Monte Carlo iterations with a maximum difference to the unmixing model of $> 10\%$ and all occurring for pasture and prairie samples. The GOF test showed results > 0.90 for each sample (Table 3), indicating that the model was not sensitive to the removal of samples.

Fig. 2 Discharge, turbidity, rainfall, and suspended sediment sample collection shown for the period (19 May 2013 to 15 August 2013) (Gellis et al. 2018)



3.3 Ages of fluvial sediment

Lead-210 excess and ^7Be were detected in all target samples (Gellis et al. 2018). ^7Be activity on the surface-derived fraction of target samples indicated that ages ranged from 44 to 205 days, averaging 131 days (Table 5). $^{210}\text{Pb}_{\text{ex}}$ indicated ages from 1 to 58 years (average = 37 years) (Table 5). The age differences from the two radionuclides indicate that all of the sediment samples are comprised of sediments of a range of ages.

4 Discussion

4.1 Sediment source apportionment

The robustness of the sediment-fingerprinting results can be assessed by examining the confusion matrix (Fig. 6), Monte Carlo uncertainty analysis (Table 3; Fig. S2, Electronic supplementary material), SVT (Table 4; Table S3, Electronic supplementary material), and Monte Carlo leave-one-out cross-validation results (Fig. S3, Electronic supplementary

Fig. 3 Results of sediment fingerprinting apportionment. Results are shown by **a** source percentages and **b** by sample period mean sediment concentration

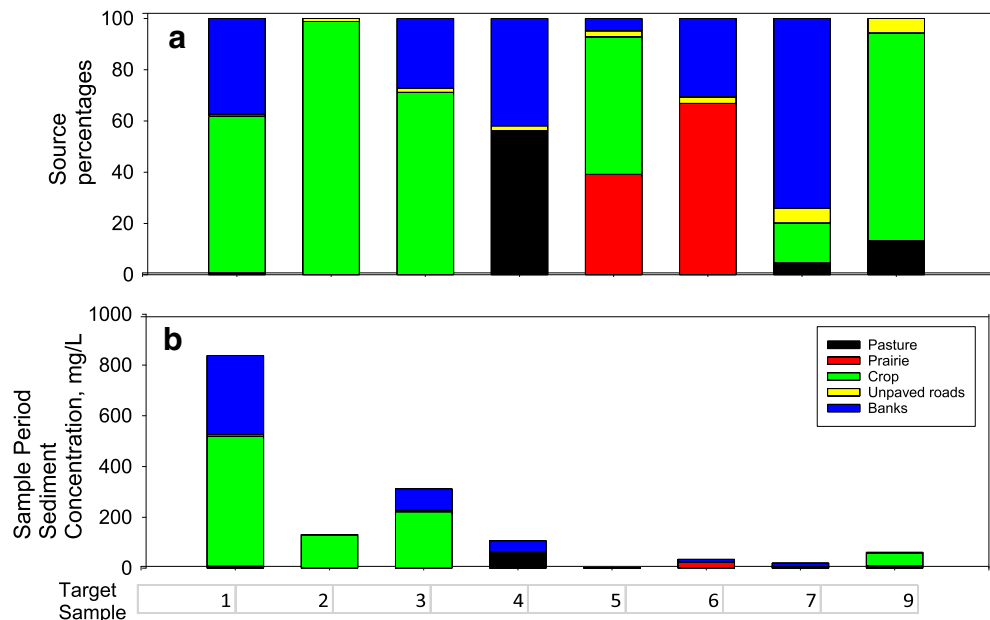
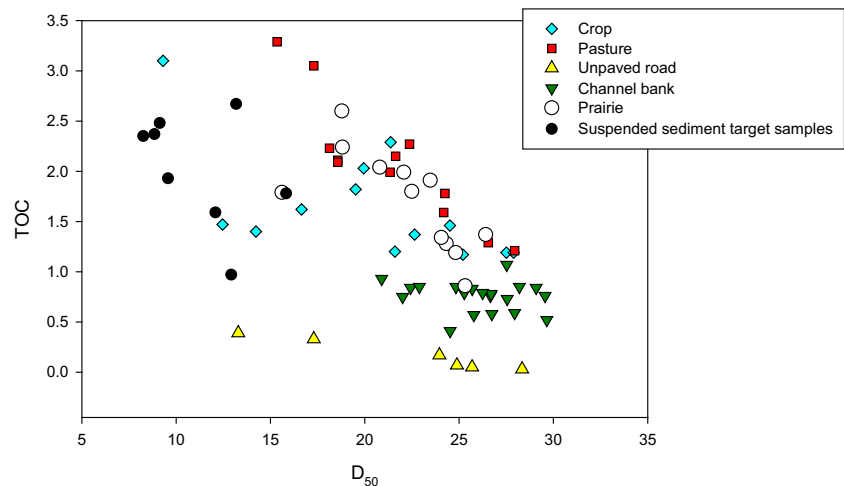


Table 3 Sediment fingerprinting results for Walnut Creek, Iowa

Sample period	Target sample No. (Table 1)	Pasture %	Prairie %	Crop %	Road %	Bank %	Relative minimum error (Eq. 2)	Sediment loading weighting factor (Eq. 5)	Monte Carlo uncertainty GOF test	Monte Carlo leave-1-out cross-validation GOF test
23 May 2013 at 08:45–11:00	1	1 (2 ± 4)	0 (0 ± 0)	61 (60 ± 7)	1 (1 ± 0)	37 (37 ± 3)	1.0982	0.89	0.93	0.91
6 June 2013 at 11:00–13:00	2	0 (0 ± 0)	0 (0 ± 0)	99 (99 ± 0)	1 (1 ± 0)	0 (0 ± 0)	0.3994	0.02	0.94	0.95
13 June 2013 at 09:15–11:00	3	0 (6 ± 4)	0 (0 ± 0)	71 (63 ± 6)	2 (2 ± 0)	27 (29 ± 2)	0.5480	0.07	0.93	0.93
20 June 2013 at 11:00–11:00	4	56 (58 ± 1)	0 (0 ± 0)	0 (0 ± 0)	2 (2 ± 0)	42 (40 ± 1)	1.0593	0.01	0.90	0.91
27 June 2013 at 11:00–11:00	5	0 (0 ± 0)	39 (34 ± 8)	54 (58 ± 11)	2 (3 ± 0)	5 (5 ± 4)	0.8801	0.01	0.91	0.91
11 July 2013 at 08:45–9:50	6	0 (1 ± 14)	67 (64 ± 16)	0 (0 ± 0)	3 (3 ± 0)	31 (33 ± 2)	0.8259	0.001	0.91	0.91
18 July 2013 at 9:50–12:45	7	5 (12 ± 5)	0 (0 ± 0)	16 (7 ± 9)	6 (6 ± 0)	74 (76 ± 4)	0.7814	0.0003	0.94	0.94
August 2013 at 09:20–1000	9	13 (17 ± 4)	0 (0 ± 0)	81 (78 ± 4)	6 (6 ± 0)	0 (0 ± 0)	1.0937	0.0003	0.92	0.92
Average		9	13	48	3	27				
Weighted by sediment loads		1	0	62	1	36				

Values in parentheses for source apportionment results were produced from the Monte Carlo uncertainty analysis with 95 confidence interval

Fig. 4 Median grain size (D_{50}) and total organic carbon (TOC) of source and target samples, Walnut Creek, Iowa (Gellis et al. 2018)



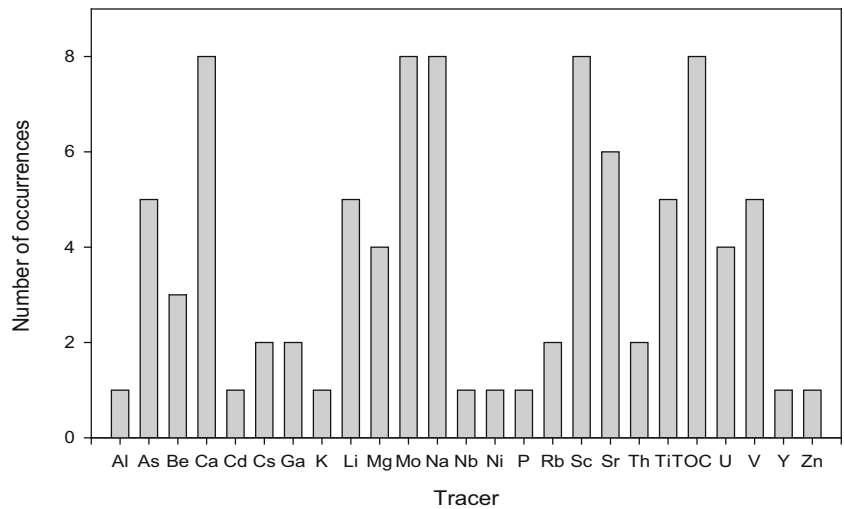
material). The confusion matrix results indicate that the tracers used in Walnut Creek are able to correctly classify the sources (Fig. 6). Monte Carlo uncertainty analysis and the GOF results showed that most apportionment results were > 0.90 , which are considered acceptable values (Sherriff et al. 2015) (Table 3). Results of the SVT indicated that banks and unpaved roads on average had $\sim 99\%$ correct classification, whereas the top-soil sources (pasture, cropland, and prairie) are often classified as other sources (Table 4). The removal of a random sample in the Monte Carlo leave-one-out cross-validation analysis, which changes the source group means, showed little difference to the unmixing model results ($< 8\%$) except for target samples 4 and 6 which showed $> 10\%$ difference to the unmixing model results. The differences all occurred for pasture and prairie samples. Six of the eight target samples showed a maximum difference of the minimum or maximum Monte Carlo leave-one-out cross-validation result to the unmixing model of up to 16% for any iteration.

Land use has a strong effect on soil chemistry due to differences in how the soil is managed, for example, differences in erosion, compaction, biologic activity, and addition of fertilizers and nutrients (Aguilar et al. 1988; Yanai et al. 2012; Veenstra and Lee Burras 2015; Yesilonis et al. 2016). As land use is converted, soil chemistry changes are variable over time where selected elements change while others remain constant (Veenstra and Lee Burras 2015). Misclassification of the top-soil source groups in the SVT (i.e., pasture and crop) may indicate similar chemistry in soils between topsoil land use types (Miller et al. 2015) or changing land use on the same field over time (Gellis et al. 2015). Palazón et al. (2015) indicated that an overlap in DFA results between agriculture and forest was due to land use conversion of agriculture reverting to forest after land abandonment. In Linganore Creek, Maryland, an agricultural watershed in Maryland, pasture and cropland fields were often rotated over time and source samples were combined for sediment fingerprinting analysis (Gellis et al. 2015).

In Walnut Creek, the intermixing of apportionment results between crop, pasture, and prairie can also be explained by its land use history. The Neal Smith National Wildlife Refuge, located in the Walnut Creek watershed, is part of a large-scale effort by the U.S. Fish and Wildlife Service (USFWS) to restore agricultural areas in the watershed to native prairie. Beginning in 1992, an effort to convert agricultural lands to native prairie was started (Schilling et al. 2002) and by 2005, approximately 23.5% of the Walnut Creek watershed had been converted from agricultural land to native prairie (Schilling et al. 2006). Pasture and prairie show a higher percentage of interclassification (confusion matrix and SVT) (Table 4) that may be both due to prairie lands being former pasture fields and/or that both land types are dominated by grasses. Although cropland in Walnut Creek was also converted to prairie, the lower percentage of misclassified prairie samples as cropland may be due to plowing and tilling operations on cropland which continually mix the soil, whereas prairie is no longer tilled. In summary, the uncertainty results from Sed_SAT suggest that for each target sample, the final set of tracers are able to significantly discriminate between topsoil types, streambanks, and roads in Walnut Creek but that an overlap in apportionment results occurred between pasture and prairie that may be due to their land use history.

Stepwise discriminant function analysis indicated that up to 25 tracers were significant in identifying the sediment sources with 5 tracers significant for all target samples (Fig. 5). Koiter et al. (2013) proposed that an environmental basis for selection of tracers used in sediment fingerprinting, such as a relation to geology and land use, should be established. Although a rigorous evaluation of this basis for all of the tracers used here is beyond the scope of this study, logical differences are evident among the five tracers that are significant for each target sample (Ca, Mo, Na, Sc, and TOC) that may be related to pedogenesis and anthropogenic influences (Fig. S4, Electronic supplementary material). Unpaved roads have the highest Ca concentration of any land use (Fig. S4a, Electronic

Fig. 5 Summary of the tracers shown to be significant in discriminating the five sediment sources (banks, unpaved roads, prairie, pasture, and cropland) for all eight target samples in Walnut Creek, Iowa



supplementary material). Secondary (unpaved) roads in Iowa are surfaced with either stream gravel or a crushed limestone aggregate obtained from local sources (Bergeson et al. 1990). The high Ca concentrations observed in the Walnut Creek watershed for unpaved roads is due to the high Ca content found in limestone. Because unpaved road material is

obtained from mines and quarries, this material is lower in concentration of the other tracers (Mo, Na, Sc, TOC), which are related to soil properties.

Molybdenum, TOC, and Sc are highest in topsoil sources (cropland, pasture, prairie) (Fig. S4b, d, e in the Electronic supplementary material). Total organic carbon is correlated

Fig. 6 Summary of the confusion matrix results indicating the percent of source samples classified correctly by the final set of tracers

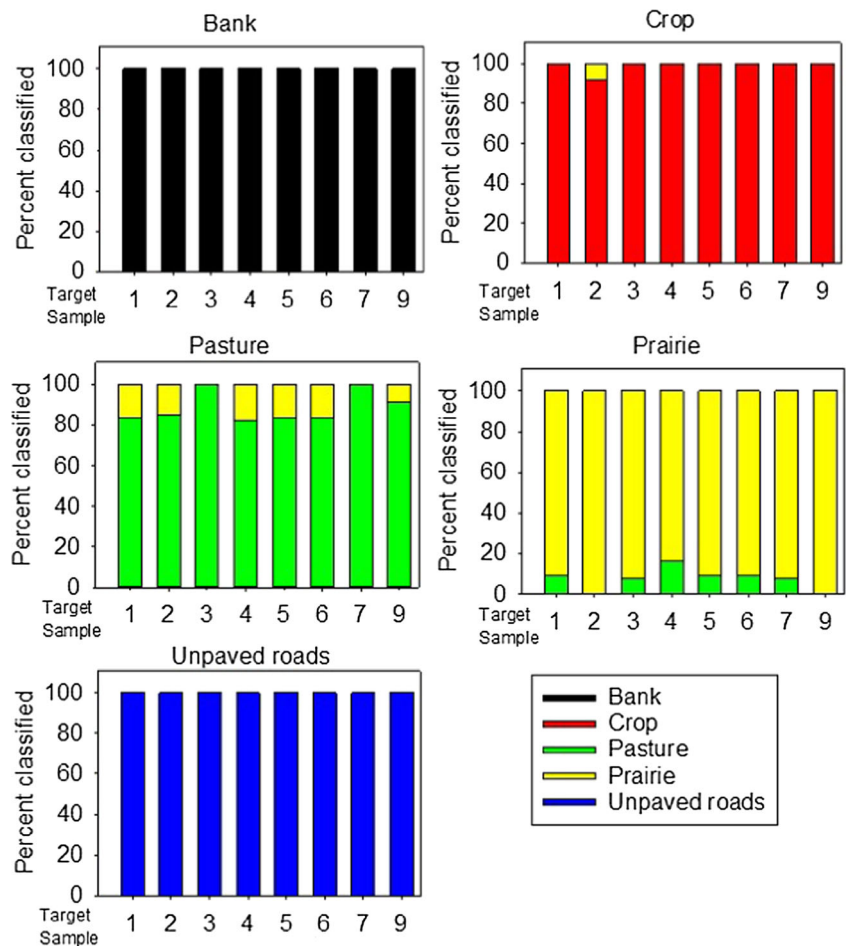


Table 4 Results of source-verification test (SVT)

Target sample No.	Source type	Classified as (%)				
		Bank	Crop	Pasture	Prairie	Unpaved road
1	Bank	97	1	1	0	0
2	Bank	96	3	2	0	0
3	Bank	97	1	1	1	0
4	Bank	98	1	1	1	0
5	Bank	97	1	1	0	0
6	Bank	97	1	1	0	0
7	Bank	97	1	1	1	0
9	Bank	97	2	1	0	0
Average	Bank	97	1	1	0	0
1	Pasture	1	3	78	17	0
2	Pasture	3	5	81	10	0
3	Pasture	1	7	64	28	0
4	Pasture	0	7	63	29	0
5	Pasture	1	5	87	7	0
6	Pasture	1	5	83	11	0
7	Pasture	1	2	74	22	0
9	Pasture	4	4	87	5	0
Average	Pasture	2	5	77	16	0
1	Unpaved road	2	0	0	0	98
2	Unpaved road	5	0	0	0	95
3	Unpaved road	4	0	0	0	96
4	Unpaved road	2	0	0	0	98
5	Unpaved road	2	0	0	0	98
6	Unpaved road	2	0	0	0	98
7	Unpaved road	4	0	0	0	95
9	Unpaved road	4	0	0	0	96
Average	Unpaved road	3	0	0	0	97
1	Crop	13	73	4	6	1
2	Crop	12	70	1	16	0
3	Crop	5	78	5	12	0
4	Crop	4	84	5	8	0
5	Crop	7	77	1	15	0
6	Crop	6	77	5	3	4
7	Crop	9	84	7	0	0
9	Crop	8	82	1	9	0
Average	Crop	8	78	4	9	1
1	Prairie	3	5	18	74	0
2	Prairie	5	6	15	73	0
3	Prairie	3	5	16	77	0
4	Prairie	2	4	17	77	0
5	Prairie	3	6	17	73	0
6	Prairie	2	7	20	70	0
7	Prairie	8	3	23	67	0
9	Prairie	6	4	18	72	0
Average	Prairie	4	5	18	73	0

Table 5 Surface-derived portion of the target sediment for the percent that is in the age of 1963, and ages using $^{210}\text{Pb}_{\text{ex}}$ and ^7Be

Target sample No.	Age of surface-derived sediment	
	^7Be (days)	$^{210}\text{Pb}_{\text{ex}}$ (years)
1	141	22
2	155	43
3	105	49
4	82	23
5	191	47
6	205	58
7	44	57
9	127	1
Average	131	37

to organic matter which is commonly high in the topsoil of these land uses. The low TOC in banks reflects the decrease in organic matter commonly observed in deeper soils. Scandium is a rare earth element common in soils as a result of weathering of parent material in soil formation (Horowitz 1975; Vermeire et al. 2016). The higher concentrations of Sc observed in topsoil classes (crop, pasture, and prairie) versus streambanks and unpaved roads (Fig. S4d, Electronic supplementary material) may reflect differences due to pedogenesis. The higher concentrations of Mo in cropland, pasture, and prairie (Fig. S4b, Electronic supplementary material) might reflect the deposition of Mo from atmospheric pollution in the twentieth century. Chappaz et al. (2008, 2012) observed higher Mo concentrations over background levels in lake sediments in Canada, which were attributed to atmospheric inputs of Mo from coal combustion and ore processing plants. High levels of Mo in soil from industrial pollution in the twentieth century have also been described in Austria by (Neunhuserer et al. 2001) and in the Midwestern USA (Elrashidi et al. 2016). Streambanks are composed of older sediment and may not be subject to atmospheric Mo deposition and thus have lower levels. The slightly higher Na concentrations in banks compared with surface soils (Fig. S4d, Electronic supplementary material) might result from the leaching of Na from surface soils, as it is a strongly hydrated monovalent cation and is relative mobile in soils (Laird et al. 2010). It also is possible that leaching of soluble constituents from surface soils is enhanced by agricultural practices. We recognize that additional research would be needed to understand the differences in Mo, Sc, and Na among these land use types.

4.2 Age of suspended sediment

Suspended sediment is a mixture of ages. The age of suspended sediment calculated from $^{210}\text{Pb}_{\text{ex}}$ activities indicates that a portion of the target sediment is of decadal ages

averaging 37 years (Table 5). The age of suspended-sediment samples using ^7Be activity (Gellis et al. 2018) ranged from 44 to 205 days (averaging 131 days) (Table 5), indicating that a portion of fine-grained sediment is moving rapidly through Walnut Creek.

Clearly, two very different ages for the same sediment sample cannot both be accurate. The occurrence of ^7Be indicates that some portion of the sediment is young, on the order of months, whereas the dating based on $^{210}\text{Pb}_{\text{ex}}$ indicates that some of the surface-derived sediment has been in channel storage for decades (Table 5). Unlike dating sediment in lakes and reservoirs with $^{210}\text{Pb}_{\text{ex}}$, which assumes that each depositional layer is isolated from new sediment, the scour and fill of sediment in Walnut Creek leads to a mixture of ages. There are two processes that are reducing the activity of ^7Be in surface-derived sediment below that of surface soil: radioactive decay and dilution by older sediment with no ^7Be activity. The ages calculated using ^7Be (mean of 131 days) assume only decay and no mixing. However, we know that much of the surface-derived sediment is much older based on $^{210}\text{Pb}_{\text{ex}}$ age estimates. This implies that some smaller portion of sediment must have higher ^7Be activities and be even younger than the ^7Be age estimates, but there is no way at present of separating out the effects of decay and dilution on ^7Be activities. Future research in age dating suspended sediment may provide some insight into this.

Based on $^{210}\text{Pb}_{\text{ex}}$, the mean age of the same surface-derived sediment fraction averaged 37 years, indicating that much of this surface-derived sediment remains in channel storage for decades. Unlike ^7Be , the $^{210}\text{Pb}_{\text{ex}}$ age might be a reasonable estimate of the mean age of the whole surface-derived fraction because most or all of that fraction probably entered the channel within the ~85-year time period dateable with ^{210}Pb . We therefore conclude that the $^{210}\text{Pb}_{\text{ex}}$ -based ages are a reasonable estimate for the mean age of the surface-derived fraction and that the ^7Be activities are evidence that there is a smaller fraction of very young sediment being transported in the stream. Other studies have also reported that the age or transit time of sediment falls into several age groups (Table 6).

4.3 Comparison of streambank and agriculture as sediment sources in Walnut Creek

The results of this study indicate that streambank sediment is approximately one-third (36%) of the apportioned suspended sediment (Table 3), which is at the lower end of estimates from other studies in Walnut Creek (Table 7). In a program to monitor water quality, sediment, and channel changes as they relate to management activities at the Neal Smith National Wildlife Refuge in Walnut Creek, streambank contributions of sediment were estimated for three different time periods: (1) 1996 to 1998, (2) 1996 to 2005, and (3) 2004 to 2010 (Table 7). For each time period, the percent contribution from

Table 6 Summary of studies age-dating suspended sediment

Study area	Age dating using	Transit and residence of sediment	Reference
Murrumbidgee River, Australia	$^{210}\text{Pb}_{\text{ex}}$	10 + – 5 years	Wallbrink et al. (1998)
Old Woman Creek, Ohio; Weeks Bay, Alabama; and South Slough, Oregon	^7Be , $^{210}\text{Pb}_{\text{ex}}$	46–79 days	Matisoff et al. (2005)
Seine River, France	^7Be , $^{210}\text{Pb}_{\text{ex}}$, ^{137}Cs	< 365 days; 4800–30,000 years	Le Cloarec et al. (2007)
Cointzio River, Mexico	^7Be , $^{210}\text{Pb}_{\text{ex}}$, ^{137}Cs	50–200 days; 5000–23,300 years	Evrard et al. (2010)
Chesapeake Bay, USA	Mass balance approach	100–1000 years	Pizzuto et al. (2014)
Pleasant Valley Watershed, Wisconsin	^7Be , $^{210}\text{Pb}_{\text{ex}}$	123 ± 12 to 322 ± 114 days	Lamba et al. (2014)
Midwest, USA	^7Be	68% of sampled streams < 100 days	Gellis et al. (2017)
Louroux catchment, France	^7Be , $^{210}\text{Pb}_{\text{ex}}$	0 to 215 days	Le Gall et al. (2017)

streambanks changed: period 1 banks contributed 51%, period 2 contributions ranged from 38.6 to 64.4%, and period 3 contributions ranged from 0 to 53% (Table 7).

Although the literature results in Table 7 show some periods with a higher percentage of sediment from streambanks than the current study, the literature also indicates that streambank contributions vary temporally with flow variability, where on some occasions, the contributions from streambank erosion were considered to be zero (Palmer et al. 2014). Therefore, the results shown here, where streambanks have a contribution ranging from 0 to 71% (weighted average of 36%), are still within the range of values reported in the literature for these longer-term studies.

Estimates of agricultural erosion are limited for Walnut Creek and only include modeled results using the Revised Universal Soil Loss Equation (RUSLE) (Schilling et al. 2011). For 2005, 20,490 Mg of sediment was estimated to have eroded from upland areas, a value almost three times higher than the average annual sediment load from 1996 to 2005 (7606 Mg year⁻¹) (Table 7). Depending on the sediment delivery ratio of sediment from upland areas, upland sediment based on RUSLE could be a

large source of sediment equal to or greater than streambank erosion. Our results confirm that upland sediment (cropland) is an important source of sediment in Walnut Creek.

4.4 Sediment in channel storage

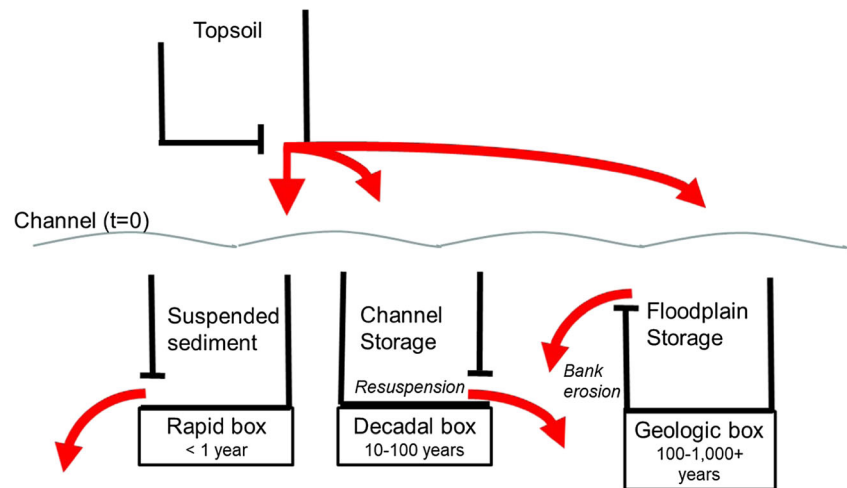
The volume of sediment in channel storage can be an important component of the total sediment budget (Lambert and Walling 1988; Marttila and Kløve 2014; Piqué et al. 2014; Wilson et al. 2004), particularly in streams with accumulations of large woody debris and beaver dams (Fisher et al. 2010; Wohl and Scott 2017). In Pleasant Valley, a small 19-km² agricultural lowland stream in Wisconsin, fine grained soft sediment stored along the channel bed is estimated to be equivalent to 8 years' worth of annual loading exported from the watershed (Faith Fitzpatrick, USGS, written communication 9 December 2017). Stream surveys conducted in 1998 by Schilling and Wolter (2000) along the thalweg of Walnut Creek for a length of 10,123 m, showed an average thickness of 12.5 cm (dominantly silt). Much of the fine-grained sediment was stored behind logs and debris jams where in one

Table 7 Literature values for rainfall, sediment loads, percent streambank contributions, and upland erosion for Walnut Creek, Iowa

Period	Rainfall average (mm)	Average annual sediment load (Mg)	Range of sediment load contribution from streambanks (%)	Upland erosion (Revised Universal Soil Loss Equation (RUSLE)) (Mg year ⁻¹)	Reference
1996–1998	845	12,732	51		Schilling and Wolter (2000)
1996–2005	751	7606	38.6–64.4%	20,490 ^a	Schilling et al. (2011)
2004–2010	984	15,958	0 to 53		Palmer et al. (2014)

^a Estimated for 2005

Fig. 7 Depiction of transit time of sediment in Walnut Creek, Iowa as a three-box model: rapid, decadal, and geologic



portion of the stream, bed sediment accumulations reached > 0.3 to >0.6 m (Schilling and Wolter 2000). Based on average flow measurements for 1997, Schilling and Wolter (2000) estimated that it would take 8.8 years to remove the sediment in channel storage if no new sediment were introduced.

4.5 Sediment age model

The transit time of sediment can be viewed from two perspectives: (1) the time it takes a particle to move from its source (i.e., topsoil) to the nearest channel or from its source to a point in the channel (i.e., outlet) or (2) the time it takes a surface-derived particle to move from when it enters the channel system to the outlet (or sampling point). Here, we examine the second perspective and assume that the age of a fine-grained sediment particle begins when it enters the channel (Fig. 7), an assumption that is made for only the topsoil-derived portion of sediment. Because streambanks do not acquire substantial activities of ^7Be and $^{210}\text{Pb}_{\text{ex}}$, this sediment cannot be dated. Instead, we assume that when fine-grained streambank sediment is eroded, it travels in the channel in a similar fashion to the topsoil-derived sediment.

Several studies have used radionuclides to date surface-derived fine-grained sediment at decadal scales ($^{210}\text{Pb}_{\text{ex}}$, ^{137}Cs) and days using ^7Be (up to 1 year). In Old Woman Creek, Ohio, the age of suspended sediment using ^7Be ranged from 46 to 79 days (Matisoff et al. 2005) (Table 6). Using $^{210}\text{Pb}_{\text{ex}}$, ^{137}Cs , and ^7Be , Evrard et al. (2010) in the Coitzio River, Mexico, and Le Cloarec et al. (2007) for the Seine River, France, both depicted a two-box model to explain sediment age times. A geologic box representing watershed storage ranging in age in Mexico from 5000 to 23,300 years and in France from 4800 to 30,000 years, and a rapid box in Mexico from 50 to 200 days and in France, to less than 1 year (Dominik et al. 1987; Evrard et al. 2010). In the agricultural Walnut Creek watershed, Iowa, we propose that sediment which is eroded from agricultural areas and streambanks can

reside in three different storage age boxes: (1) a rapid box < 1 year, (2) a decadal box (10–100 years), and (3) a geologic box (100–> 1000 years) (Fig. 7).

Based on ^7Be results, a rapid box is depicted for Walnut Creek where a portion of the fine-grained sediment moves through the system in less than 1 year (Fig. 7). Based on $^{210}\text{Pb}_{\text{ex}}$ results, a decadal storage box is depicted for Walnut Creek (Fig. 7), where some of the sediment from agricultural areas remains in storage for decadal time periods with limited transport during storm events. A decadal transit time of sediment also was observed for Australian rivers (0–21 years) (Wallbrink et al. 2002). Most of the sediment that travels in the decadal and rapid storage box in Walnut Creek originates from agricultural topsoil.

Although an analysis of the ages of floodplain sediment was not undertaken in Walnut Creek, for the geologic box, based on other studies, sediment that is deposited on floodplains is likely to remain there over mean time scales of a hundred to a thousand or more years (Lancaster and Casebeer 2007; Phillips et al. 2007; Pizzuto et al. 2014). Sediment deposited on the floodplain in Walnut Creek is depicted as the geologic box in Fig. 7, where it is assumed to remain in storage for a period of 100 years or longer.

4.6 Data limitations and uncertainty

The age model in this paper for ^7Be (Eq. 10) relies on the surface activity of ^7Be and, as discussed in Sect. 4.2, it is applied to the whole surface-derived sediment fraction without accounting for mixing with sediment too old to date with ^7Be . It was not within the scope of this study to directly measure fallout activities. The value, 542 mBq g^{-1} , was estimated by Gellis et al. (2017) for Midwestern US streams and is 35% less than the average value of ^7Be surface soil activity in the literature (mean = 838 mBq g^{-1} , Fig. S5, Electronic supplementary material); however, the number of studies is limited ($n = 6$) and the variability is large (standard deviation, ± 616)

(Fig. S5, Electronic supplementary material). We recognize that the ^7Be surface soil activity in Walnut Creek may be temporally and spatially variable. If the mean soil surface ^7Be activity is higher or lower than our estimate, the resulting age estimates of suspended sediment will get older and younger, respectively.

Beryllium-7 and $^{210}\text{Pb}_{\text{ex}}$ activity are used as indicators of the age of the surface-material component of suspended sediment. Most of the ^7Be activity in surface soils is found in the top centimeter (Wallbrink et al. 1998; Huh and Su 2004). In our age model for ^7Be , it is assumed that the top most surface (~1 cm) is being eroded and deeper erosion of topsoil is not occurring. Deeper surface erosion would tend to lower the ^7Be activity in the transported particles but not necessarily lower $^{210}\text{Pb}_{\text{ex}}$. If deeper surface erosion did occur, then our results are indicating older ages.

We recognize that some of the ^7Be and $^{210}\text{Pb}_{\text{ex}}$ falls directly on the wetted areas of the stream channels, which may cause an increase in activity on sediment already in suspension and channel storage. Hancock et al. (2014) examined the contribution of surface soils using fallout radionuclides and estimated that the direct contribution of ^7Be to the Bowen River (9400 km²) and Logan River (3700 km²) in eastern Australia could cause a 12 and 10% increase, respectively, in the measured values. Hancock et al. (2014) further commented that this estimate is high considering that most of the rainfall fell early in the event prior to rising stage and presumably before the input of sediment to the system. Wallbrink et al. (2002) examined ratios of $^{210}\text{Pb}_{\text{ex}}$ to ^{137}Cs in source and target samples and based on the higher ratios in target samples determined that enrichment of $^{210}\text{Pb}_{\text{ex}}$ on suspended sediment was occurring through direct rainfall on the channel. A similar analysis was run for Walnut Creek and results do not show enrichment of $^{210}\text{Pb}_{\text{ex}}$ in target samples relative to source samples and we therefore conclude that there is not a significant contribution of $^{210}\text{Pb}_{\text{ex}}$ on the target sediment from rainfall (Table S4, Electronic supplementary material).

Because ^7Be and $^{210}\text{Pb}_{\text{ex}}$ were not measured in rainfall in this study, correction factors cannot be applied to the direct input of ^7Be to the wetted area of the channel. If rainfall inputs of ^7Be directly to the stream increase suspended-sediment activities, this increase would result in a younger apparent age. For example, if 10% of the ^7Be activity in the target sample is attributed to dosing from direct rainfall on the stream channel, and a correction was made to the ^7Be activity, the suspended sediment would increase in the age of 8 days, a relatively small amount.

Higher radionuclide activity can be correlated to finer grain sizes (He and Walling 1996). Although a size correction factor was applied to the elemental concentrations in the sediment-fingerprinting method, because of the small amount of source samples that were analyzed for ^7Be and $^{210}\text{Pb}_{\text{ex}}$ (Gellis et al. 2018), we are unable to apply the same regression approach to these samples. This study did analyze the finer portion of

sediment (< 63 μm) where most of the radionuclide activity is found.

Although the focus of this paper was to test the efficacy of combining the sediment fingerprinting approach with age dating, limitations related to the short duration of the study period are recognized. Sediment sources and ages may have seasonal variations that are not captured in this relatively short period of sampling. To determine how the sampling period compared with longer-term hydrologic conditions, historical monthly rainfall (NOAA) data were obtained for the Prairie City, Iowa raingage (2002–2017) (<https://www.ncdc.noaa.gov/>; accessed 31 March 2018) (Table S5, Electronic supplementary material), located ~3 km from the USGS station on Walnut Creek. Monthly rainfall summaries show that the rainfall totals for June and July 2013 (the sampling months for this study) were similar to the 2002–2017 June median and 64% less than the July median. To determine if antecedent conditions prior to sampling were different than the long-term average, monthly totals for the period January to May were summed for each year and indicated that January to May 2013 was 78% higher than the median long-term value. In summary, compared with historic records, antecedent conditions prior to sampling at Walnut Creek in 2013 were wetter and during the sampling period conditions were drier. We do not know, however, how these differences affect sediment sources and ages.

5 Conclusions

Understanding the contributions from agricultural topsoil and its residence time are important critical zone processes that can assist managers in developing actions to reduce sediment and improve the health of aquatic habitats. This study shows how the sediment fingerprinting approach combined with fallout radionuclides can apportion fine-grained sediment to its sources and date the topsoil-derived portion. Previous studies on age dating sediment using fallout radionuclides were unable to distinguish older channel sediment from sediment that was diluted by streambanks. Here, we use elemental analysis to fingerprint the sources of sediment and short-lived fallout radionuclides to date the surface-derived portion of sediment into two age groups: ^7Be to < 1 year and $^{210}\text{Pb}_{\text{ex}}$ to decadal ages. Results from Walnut Creek indicate that the majority (63%) of the sediment is surface-derived agricultural topsoil and has a mean age of 37 years (using $^{210}\text{Pb}_{\text{ex}}$) and includes a component of much younger sediment that results in a mean age of 131 days (using ^7Be). For Walnut Creek, we propose a model of sediment ages, where sediment which is eroded from agricultural areas and streambanks can reside in three different storage age boxes: (1) a rapid box (< 1 year), (2) a decadal box (10–100 years), and (3) a geologic box (100–> 1000 years).

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