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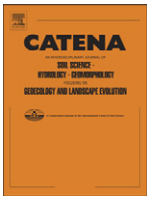

Using cesium-137 to quantify sediment source contribution and uncertainty in a small watershed

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ABSTRACT

Knowledge of sediment provenance is critical for precision conservation planning and calibration of soil erosion models. The objectives are to evaluate the ability of ^{137}Cs to apportion sediment source contributions, quantify uncertainty of the estimates, and estimate desirable sample numbers. We collected 50 surface soil samples from overland in a watershed (15.6 km^2), 28 subsoil samples from gully bank, and 43 sediment samples in channels. The ^{137}Cs activity was measured by γ spectrometry. Proportion means were calculated by solving a linear mixing model. Uncertainty was estimated by the first-order approximation and Monte Carlo (MC) simulation. The ^{137}Cs tracer was conservative in the fraction of <63 μm. The ^{137}Cs activities differed between sources in the whole watershed at $P = 0.05$. The mean proportions of the <63 μm fraction (relative error) predicted for the watershed using the mixing model were 0.42 (35%) for the overland source and 0.58 (25%) for the gully source, with a 95% confidence interval of ± 0.145 for both. Overland source contributed most uncertainty to the proportion estimates, followed by sediment, with minimum from gully. The MC simulation predicted the same mean proportions, but with relative errors being <3% for both. Compared with the first-order approximation, MC underestimated uncertainty of the means due to the large sample number used in simulation. In general, 30 to 50 samples are needed for each source and sediment to generate reliable estimates using ^{137}Cs. Estimated source proportions of the fine sediment may be converted to proportional contributions of total soil erosion by adjusting by particle sizes and sediment delivery ratio. No adjustment or weighting should be made directly to the mixing model unless conservativeness of the tracer is violated.

1. Introduction

Knowledge of sediment provenance is critical for developing precision conservation plans to effectively control soil erosion and sediment delivery in a watershed. For example, if gully bank erosion is a dominant sediment source, any conservation measure implemented in overland areas will not reduce sediment discharge in stream. In addition, contributions of sediment source types (e.g., gully versus overland) are needed for accurately calibrating hydrological and erosion models at a watershed scale because many process-based models simulate overland erosion and gully erosion separately. Because many sediment source types cannot be directly measured in a watershed setting, various sediment source fingerprints including soil physical, chemical, mineralogical, and biological properties as well as radioactive and stable isotopes have been widely used as tracers to apportion sediment sources. Among all fingerprint properties, fallout radionuclides including ^{137}Cs, excess ^{210}Pb, and ^{7}Be have been widely used as tracers in the past decades. Those fallout radionuclides have been used to estimate soil erosion redistribution (e.g., Ritchie and McHenry, 1990; Walling and He, 1999; Zapata, 2010; Mabit et al., 2013; Zhang, 2014; Zhang et al., 2015a), establish sediment chronology (e.g., Ritchie and McHenry, 1990; Appleby, 2008; Zhang et al., 2015b), and fingerprint sediment sources.

The fallout radionuclides have been shown to provide good fingerprints for identifying sediment source type (Walling, 2005). Surface soils are exposed to fallout and are tagged with the radionuclides, while gully bank or subsurface soils are often received little or no fallout and shown little or zero radioactivity. The contrasting radioactivity between surface and subsurface soils makes the fallout radionuclides the ideal tracers for discriminating overland erosion from gully erosion. Moreover, due to the nature of tillage mixing, the fallout radionuclide activity on a cultivated soil surface is much lower than that on a

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Abbreviations: CI, confidence interval; CV, coefficient of variance; MC, Monte Carlo; RE, relative error; SDR, sediment delivery ratio; SE, standard error.

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uncultivated soil surface such as those in the rangelands and forestlands (Walling, 2005). Such a large difference in activity enables the fallout radionuclides to further discriminate between cultivated and uncultivated soils.

The fallout radionuclides, primarily ^{137}Cs, have been successfully used to fingerprint sediment source type (e.g., Walling and Woodward, 1992; Olley et al., 1993; Wallbrink et al., 1996, 1998; Collins et al., 1997; Brigham et al., 2001). A key feature for an ideal sediment tracer is its conservativeness during sediment generation and transport. Cesium-137 is strongly adsorbed to or fixed by fine soil particles and organic matters, which are generally transported as suspended loads or wash loads in streams. To circumvent the enrichment of suspended sediment in fines, only fine fractions such as <63 μm have been collected from sources and sediment and analyzed for ^{137}Cs activity in the literature in order to preserve the conservativeness assumption (Wallbrink et al., 1998; Walling, 2005; Koiter et al., 2013). Motha et al. (2002) examined the conservativeness of nine tracers (six major elements, mineral magnetic properties, and radionuclides of ^{137}Cs and excess ^{210}Pb) during sediment generation under simulated rainfall, and found that only the two radionuclides behaved conservatively in the size fractions of <63 μm. This finding largely explains why the fallout radionuclides have been successfully used for fingerprinting fine sediment sources in many studies in the literature.

Due to large activity differences between surface soils and subsurface soils or gully bank materials, ^{137}Cs has been successfully used alone in a simple mixing model to apportion sediment contributions between these two sources (Olley et al., 1993; Wallbrink et al., 1996; Nagle and Ritchie, 1999, 2004; Zhang et al., 1997; Nagle et al., 2007; Ritchie et al., 2009). Cesium-137 has also been used along with the radionuclides of ^{7}Be and excess ^{210}Pb to apportion sediment source contributions between surface soil and gully erosion (Walling and Woodward, 1992; Wallbrink et al., 1999; Brigham et al., 2001; Smith and Dragovich, 2008; Hancock et al., 2014), between cultivated and uncultivated lands including forestlands, rangelands, and no-till areas (Wallbrink et al., 1998; Matisoff et al., 2002; Porto et al., 2005), and in forested watersheds (Mizugaki et al., 2008; Wilkinson et al., 2009; Smith et al., 2011, 2012; Owens et al., 2012). Given the ability of ^{137}Cs to distinguish surface from subsurface soils, it has been extensively used in conjunction with other fingerprint properties such as geochemicals (Collins et al., 1997, 2001; Walling et al., 2001; Collins and Walling, 2002; Carter et al., 2003; Gruszowski et al., 2003; Krause et al., 2003; Koiter et al., 2013), organic matter and stable isotopes of ^{13}C and ^{15}N (Mukundan et al., 2010, 2011; Samani et al., 2011; Rhoton et al., 2008), mineral magnetic property (Russell et al., 2001; Motha et al., 2002, 2004) to apportion sediment sources based on land uses including forest, pasture, croplands, roads, tire tracks, gully, and channel bank.

The use of the mean tracer concentrations to represent the true values in sediment sources and sediment mixture in the mixing model is an important source of uncertainty in the source contributions estimated by the mixing model (Lamba et al., 2014), and tends to disguise the uncertainty of the proportional estimates. Most sediment source fingerprinting studies only report the mean proportional contributions without explicitly stating the uncertainty associated with the means. This practice often leads to false certainty on estimated proportions. In recent years, more attention has been paid to uncertainty assessment (Walling, 2013). Monte Carlo simulation has been increasingly used to simulate probability distributions of estimated source proportions by randomly sampling probability distributions of each input tracer in each source and sediment mixture (Motha et al., 2003; Nagle et al., 2007; Collins et al., 2010, 2012; McKinley et al., 2013; Stone et al., 2014; Laceby and Olley, 2014), by bootstrap resampling (Clarke, 2014), and by randomly removing one sample from each source category before each new model run (Gellis and Noe, 2013; Lamba et al., 2014). Bayesian uncertainty framework has also been used to simulate probability distributions of estimated source proportions (Small et al., 2002; Koiter et al., 2013; Stewart et al., 2014). On the other hand, the Gaussian first order approximation, which is based on the theory of error propagation, has seldom been used in approximating uncertainty in sediment source ascription studies in the literature.

The objectives of this study are to (1) evaluate the ability of using radionuclide ^{137}Cs alone to apportion sediment sources between overland and gully erosion in a mixed land use watershed in central Oklahoma, (2) quantify uncertainty of the mean source contributions, estimated using the Gaussian approximation approach and Monte Carlo simulation, and further compare them, (3) estimate sample numbers required for statistically sound estimation under the study conditions to serve as guidelines for future sampling design, and (4) assess spatial variations of the estimated source contributions within the watershed.

2. Materials and methods

2.1. Watershed description

The drainage area of Bull Creek watershed is approximately 15.6 km^2. Croplands occupy about 50% of the watershed area, and are predominantly located on the gently sloped uplands (Fig. 1). Winter wheat (*Triticum aestivum*, L.) is the main crop, with limited sorghum (*Sorghum bicolor*) and millet (several different genus species combinations). Almost all croplands are terraced, and no-till and minimal tillage are implemented in most parts of the croplands. About 45% of the area is woodlands and rangelands, which are found on the rugged terrain along and near the creeks. The woodlands and rangelands are moderately grazed by cattle (*Bos taurus*). Developed areas including road and open water occupy about 5% of the drainage area.

2.2. Sample collection and processing

Surface soil samples were stratified by soil and land use types. A SSURGO soil map (Soil Survey Geographic database, 2008) was superimposed with a NLCD land cover map (National Land Cover Dataset, 2001) to delineate sampling units. Eleven major soil types were created by grouping slope categories, and two major land uses of croplands and rangelands were selected. Totally 50 surface soil samples with 28 from croplands and 22 from rangelands were taken in the watershed (Figs. 1 and 2). Each surface sample was taken from each sampling unit (a combination of a major soil and land use type). Each sample was a composite of 30 subsamples taken randomly from the sampling unit. Each subsample was taken at the soil surface with a steel ring (6.2 cm i.d. and 2.5 cm high). Twenty-eight subsoil or gully bank samples were taken from the vertical gully bank with a trowel after scraping the surface (Fig. 2). The sampling position was at least 30 cm from the ground surface to avoid the top soils, and one sample was taken on each site. Sediment samples were collected at 12 locations using sediment traps (Fig. 1). The trap is made of an aluminum pipe (14 cm i.d., and 60 cm long) with plastic funnels on each end with a 3-cm opening upstream and a 2-cm opening downstream (Fig. 1). Sediment samples were collected from 2011 to 2014. All the samples were oven dried, and ground and sieved though a 2-mm sieve. Particle size composition of clay, silt, and sand was determined for all samples with a hydrometer at 40 s and 2 h following the onset of sedimentation (Bouyoucos, 1962).

2.3. Radiometric measurement of ^{137}Cs

Radionuclide ^{137}Cs, an anthropogenic radionuclide, was generated by atmospheric nuclear bomb testing with a major fallout period between 1958 and 1964. The ^{137}Cs activity was measured by gamma spectrometry at 661.62 keV using a high-purity germanium (HPGe) coaxial detector (50% efficiency and a resolution of full width at half maximum (FWHM) of 2.2 keV at 1.3 MeV) coupled to a multi-channel analyzer.