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3.5.2. Tracing Fine-Sediment Using Artificial Radionuclides

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ABSTRACT: The environmental characteristics of many artificial gamma-emitting radionuclides provide a number of distinct advantages over other forms of fine-sediment tracer that are readily exploitable when undertaking erosion investigations. This makes them particularly effective for accurately measuring the movement of small quantities of fine-sediment, especially in low-energy environments where soil movement is commensurately slow, or for measuring erosion rates over short (i.e. event-based) timescales. The major advantages and disadvantages associated with their use as tracers are outlined and briefly discussed. Three examples of tracing applications using caesium-134 (^{134}Cs) and cobalt-60 (^{60}Co) are summarised in order to highlight their versatility and demonstrate the diverse ways in which they can be used in field- and laboratory-based investigations.

KEYWORDS: artificial radionuclide; sediment-tracer; fine-sediment; caesium-134; cobalt-60.

Introduction

Fine-sediment (< 2 mm dia.) acts as a vector for the transfer of nutrients and contaminants and its removal can ultimately lead to a loss of agricultural productivity (Walling and Quine, 1992), as well as pollute terrestrial and aquatic ecosystems (Walling et al., 2003; Walling, 2004). Against this background, numerous approaches have been developed for quantifying soil movement through catchments. One approach is through the use of artificially applied sediment tracers. A tracer of this type can be defined as any identifiable substance which, by examining its behaviour over a given time-frame, may provide information on the behaviour of the host sediment (Sauzay, 1973; Evans, 1983). To be suitable, a tracer must meet certain requirements. The most fundamental include the ability to faithfully replicate the behaviour of the medium being traced, yet remain distinguishable from its surroundings in order to facilitate its re-identification and measurement (Sauzay, 1973; Evans, 1983; Guiresse & Revel, 1995; McCubbin & Leonard, 1995; Foster & Lees, 2000). Many different materials and substances have previously been used as sediment tracers. Their success in providing representative data, however, relies on an assumption that the tracer will behave in the same manner as the host sediment. In order for this assumption to be met, the physical characteristics of the tracer (i.e. approximate aggregate size, bulk density, etc.) must match the host material as closely as possible (Evans, 1983; Parsons et al., 1993; Ventura et al., 2002). Achieving this has proven to be extremely difficult (*cf.* Parsons et al., 1993; Riebe, 1995; Ventura et al., 2002; Zhang et al., 2003; Polyakov and Nearing, 2004), owing to the physical characteristics of fine-sediment and its tendency to be dispersed, often over relatively long distances and sometimes in multiple directions (Fullen, 1982). The use of artificial radionuclides offers a convenient way in which this obstacle can be overcome.

Radionuclides as fine-sediment tracers

Each radionuclide (or radioisotope) emits high-energy ionising radiation at a unique wavelength. This characteristic 'signature' can be used to identify an individual radionuclide and the radiation emission represents a powerful tool that can be used to trace physical and chemical pathways (Jones and Atkins, 2000). Gamma rays represent the most energetic form of ionising radiation because their high energy and extremely small mass allow them to pass through most materials. For those reasons, radionuclides that undergo radioactive decay by gamma radiation arguably represent one of the most effective fine-sediment tracers.

In the context of this technical report, an artificial radionuclide is defined as being of anthropogenic origin, but excludes those deposited by globally-distributed fallout, such as caesium-137 (^{137}Cs). In this respect, artificial radionuclides do not occur naturally in the wider environment (with the exception of licensed or accidental releases in local or regional areas). Typical methods for introducing an artificial radionuclide into a given environment when tracing fine-sediment are by: 1.) directly labelling surface-sediment in situ over a predefined study area; 2.) introducing quantities of 'pre-labelled' sediment onto or over a chosen study site; or 3.) conducting tracing experiments under a confined (i.e. laboratory) environment.

Health and safety

Due to inherent dangers associated with radioactive material, particularly unsealed sources, health and safety precautions must be followed in order to minimise the exposure-time to ionising radiation. A comprehensive guide to radiation protection is provided in Connor et al. (2007). In summary, the main risks associated with over-exposure can be alleviated by: 1.) shielding the radioactive source within an enclosed (i.e. lead-lined) container until required for use; 2.) minimising the time spent in proximity to a radioactive source; 3.) maintaining as much distance from the radioactive source as is practicable; 4.) wearing personal safety equipment and adhering to the Control of Substances Hazardous to Health (COSHH) protocol (HSE, 2012); and 5.) partitioning and diluting to a lower activity concentration (e.g. 100-200 Bq ml^{-1}) a sufficient quantity of radionuclide material from the stock supply to meet the needs of a given investigation.

Measuring radionuclides

The most common method for accurately measuring low-level radioactivity is by gamma spectrometry. This is a relatively simple procedure that involves minimal sample preparation and is non-destructive, which allows repeat measurements to be undertaken on individual samples. Field-based areal activity measurements can be performed *in-situ* using a field gamma spectrometer, shown in Figure 1. Although no sample preparation is required, the crucial factor for obtaining reliable results is to ensure that the radiometric protocol adopted at the beginning of an investigation remains consistent throughout the monitoring campaign (*cf.* He et al., 2002; Greenwood et al., 2008).

Figure 1. Using an in-situ gamma spectrometer to measure areal activity.

In contrast, estimating the mass activity of individual sediment samples is typically undertaken using laboratory-based, or static, gamma spectrometers (Figure 2). The key stages of sample preparation for this form of analysis involve drying, gently disaggregating and screening the sediment through a 2 mm dia. sieve. The material is then weighed usually to an accuracy of 0.1 g (Pennock and Appleby, 2002). For reasons of precision, samples presented to a detector should be of similar mass (i.e. within ± *ca.* 0.1 g) and radiometric assays should always be performed in containers of identical dimensions so that the sample-geometry, and hence the distance from the radioactive source to the detector-head, remain constant.

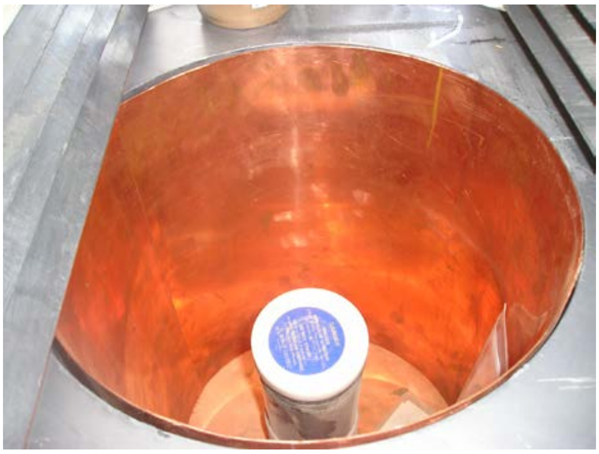

Figure 2. The interior of a static gamma spectrometer. The weighed sediment sample is placed, in a container, on the detector-head (fitted with a white end-cap).

Failure to comply with this protocol can profoundly influence the precision of individual measurements and ultimately produce erroneous results. A fuller explanation of the importance of sample presentation, and of other factors affecting analytical precision in gamma spectrometry is provided in Wallbrink et al. (2002). Where the mass, and hence the geometry, of samples varies, the ability of the detector(s) to 'capture' and record gamma-photons can also vary. In order to compensate and account for this effect, the geometry-efficiency of the detector must be re-calculated in order that sufficient analytical precision between samples of different mass is obtained (Wallbrink et al., 2002). This can be achieved by preparing a quantity (i.e. a set) of 'in-house' geometry-calibration standards. Each standard should increase in mass at regular (e.g. 5 g) intervals and the range of each set should be sufficient to correspond to the lightest and heaviest sediment samples obtained from a particular investigation. Geometry-calibration standards should, ideally, be labelled with the same radionuclide used in the investigation and at a similar mass activity (i.e. Becquerel per gram (Bq g^{-1})) as the soil(s) under investigation. Each standard should be analysed a minimum of, for instance, three times over a suitable range of counting times (i.e. 1, 2 and 3 hours) to derive an accurate result. All results should be decay-corrected to a common date of analysis in order to account for the short half-life and the radioactive decay process. The mean activity value derived from each standard is then used as basis for re-calculating the geometry-efficiency of the detector at each mass. The appropriate detector efficiency value should be selected from the particular calibration standard whose mass most closely corresponds to the mass of the sample under analysis.

Benefits and advantages

In order to be of maximum value as a sediment tracer, an artificial radionuclide must possess certain environmental characteristics. Of particular importance is the need for a radionuclide to express a rapid and strong affinity for soil particles, in order to limit the effect of leaching, and have a relatively short half-life, in order to minimise environmental contamination (Fullen, 1982). Gamma-emitting radionuclides offer five distinct advantages that are readily exploitable when undertaking erosion investigations. The first relates to the relative ease with which energetic gamma-photons can be detected with a high degree of analytical sensitivity (Lang, 2008). The second relates to the highly penetrative nature of gamma-photons; coupled with the fact that only a relatively few are needed to identify the radionuclide and estimate the relative activity (Fullen, 1982). The third and potentially most powerful advantage relates to the extremely small mass of individual gamma-photons (Courtois, 1973; Sauzay, 1973; McCubbin and Leonard, 1995). In this respect, sediment labelled with a suitable artificial radionuclide for the purpose of tracing its movement arguably represents one of the most effective ways of precisely matching the tracer characteristics to the sediment being traced. Exploiting this attribute provides a (theoretical) means of obtaining an absolute indication of the movement of the eroded sediment without having unduly perturbed or adversely influenced its behaviour (Wooldridge, 1965; Fullen, 1982; Showler et al., 1988; McCubbin and Leonard, 1995). Once the correct environmental permits relating to the release of unsealed sources of radioactive material have been obtained (refer to EA, 2012), a fourth advantage is that the initial dose-rate of an artificial radionuclide released over a given area can be tailored, within reason, to suit the objectives of an investigation. For example, if an objective is to measure erosion rates over very short (i.e. event-based) timescales, or in low-energy environments where soil movement is likely to be commensurately small, a high dose rate will allow radiometric assays to be performed relatively quickly and accurately, even on small samples, and also provide the best opportunity of obtaining results within an acceptable error range (i.e. usually ± 5-10%) (Sutherland, 1994; Wallbrink et al., 2002; Parsons and Foster, 2011). Lastly, an initially high dose rate applied directly to the soil-surface (at predetermined locations) within a given study-site can provide a means of lengthening the duration of an investigation so that erosion rates can continue to be documented over one or more seasons (see Wooldridge, 1965; Fullen, 1982; Syversen et al., 2001).

Taking account of the characteristics outlined above, artificial radionuclides offer a significant number of advantages over many other forms of tracer. With careful experimental design, these can be exploited and used to develop novel tracing-techniques that can be applied in a range of erosion scenarios where other tracers or existing tracing-techniques would be ineffective or lack the necessary level of sensitivity needed to provide reliable data.

Disadvantages and limitations

Arguably, one of the most vexing problems when using artificial radionuclide relates to a dearth of established techniques for directly labelling the surface-soil uniformly (Syversen et al., 2001). This limitation has two major implications. The first is that field-based investigations are typically limited to studying soil movement over areas equivalent to a few m^2 (Wooldridge, 1965; Fullen, 1982; Quine et al., 1999; Syversen et al., 2001). This constraint can be overcome, to a certain extent, by applying the radionuclide to small areas, or ‘plots’, within a larger study-site. This approach overcomes one of the major spatial constraints associated with using artificial radionuclides, since it represents a convenient and effective way of obtaining contiguous erosion data from plots strategically sited at key areas, or on particular landform-features within a study-site (e.g. Greenwood et al., 2008).

The second limitation associated with the non-uniform application of radionuclide material means that the pre-event areal activity at any location within the labelled area is unknown and must, as a vital prerequisite, be accurately measured before the next erosion event. Importantly, all post-event measurements must thereafter be performed at precisely the same locations as the pre-event measurements, in order to permit changes in the areal activity, and hence, in the movement or redistribution of the labelled soil, to be accurately quantified.

Arguably, one of the most profound disadvantages with using artificial radionuclides arises from the stigma associated with radioactivity, and the fact that field-based investigations inevitably involve deliberately releasing radioactive material into an environment. This problem can be mitigated against, to a certain extent by, firstly, adhering to the precept that any exposure to ionising radiation should be kept as low as reasonably possible (Connor et al., 2007), and secondly, by selecting an artificial radionuclide with an appropriate (i.e. short) half-life. Artificial radionuclides with a suitably contrasting half-life range that have been successfully used in sediment-tracing investigations include iron-59 (^{59}Fe; $t_{0.5}$ = 45 d) (Wooldridge, 1965; Fullen, 1982), caesium-134 (^{134}Cs; $t_{0.5}$ = 2.06 yrs.) (Quine et al., 1999; Syversen et al., 2001; Greenwood et al., 2008), and cobalt-60 (^{60}Co; $t_{0.5}$ = 5.27 yrs) (Toth and Alderfer, 1960; Greenwood et al., 2008).

Any researcher considering using artificial radionuclides in sediment tracing investigations must also ensure that their research institute has the necessary radioisotope permits and adequate facilities to purchase, securely store and safely dispose of radioactive waste. The researcher will also need access to comprehensive analytical facilities, or be prepared to pay for analytical time on radiometric facilities elsewhere, and be trained to handle and partition the unsealed source material to an appropriate concentration. Lastly, a vital requirement when conducting field-based investigations using artificial radionuclides is the need to identify landowners willing to allow radioactive material to be deliberately introduced onto their land.

Practical applications

Examples of three field / laboratory-based tracing techniques using the artificial radionuclides, ^{134}Cs and ^{60}Co, are now briefly described, in order to demonstrate the novel or diverse ways in which they can be applied.

Tracing eroded earthworm casts on pasture

The presence of earthworm casts on soil-surfaces has led many workers to speculate that the dispersed sediment can potentially contribute to soil erosion (Le Bayon and Binet, 1999, 2001; Le Bayon et al., 2002). Despite efforts to confirm or refute those claims, a notable degree of uncertainty still exists and is attributed, as described earlier, to the difficulty of accurately tracing fine-sediment. Capitalising on this continuing uncertainty, a technique was successfully developed for labelling earthworm casts with either ^{134}Cs or ^{60}Co, for the purpose of tracing the dispersed sediment. Labelling was achieved by immersing the intact air-dried casts into a solution of water, mixed with a known concentration of either ^{134}Cs or ^{60}Co. After labelling, the casts were deployed across a small prepared area of pasture in a semi-moist condition, which was considered to be most representative of the stability, and hence, the potential erodibility, of casts of a wide range of ages (Binet and Le Bayon, 1999). In order to provide information on relative travel distance of the dispersed sediment, a batch of ^{134}Cs-labelled casts was evenly distributed across the upslope half of the plot, at a distance of > 0.3 m from the channel outlet, and a batch of ^{60}Co-labelled casts was evenly distributed across the downslope half of the plot, at a distance of < 0.3 m from the channel outlet (Figure 3). All casts were subjected to natural weather events over a two-month period. Runoff samples were collected and the eroded sediment was separated from the liquid, processed following the method described earlier and radiometrically assayed. A mass balance was used to partition and quantify the relative proportions of labelled sediment from the unlabelled surface soil. The response of both tracers indicate that, in proportional terms, 14.1% and 4.5% of the original mass of ^{60}Co and ^{134}Cs-labelled casts were transported downslope by surface runoff over distances < 0.3 m and > 0.3 m, respectively. The results provide essentially unique information that confirms that earthworm casts can contribute to soil erosion, certainly over relatively short distances, on vegetated hillslopes.

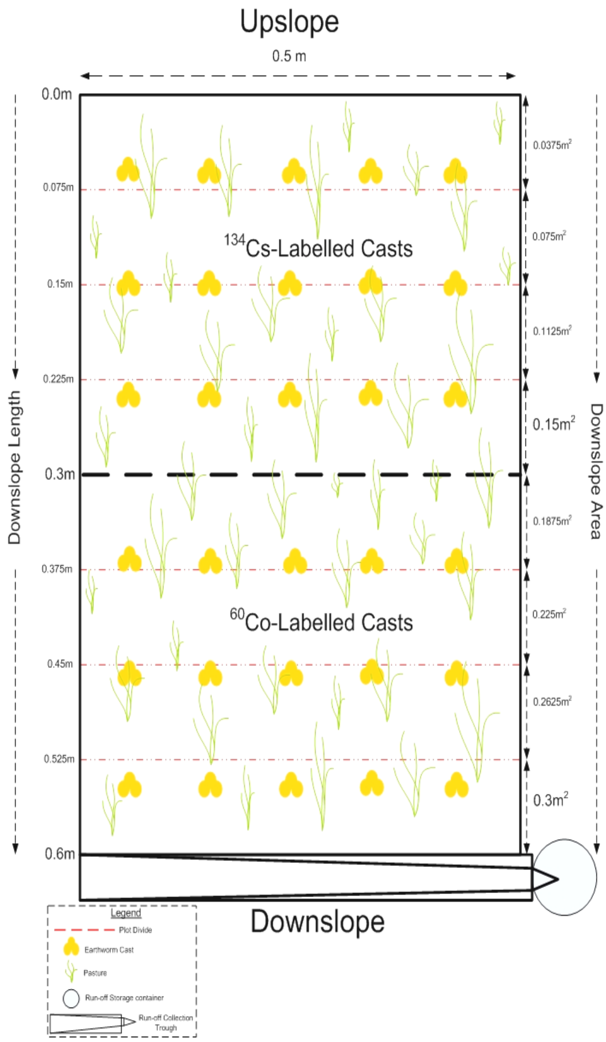

Figure 3. The design-layout and relative positions of each batch of labelled casts within the erosion plot (not to scale).

Measuring soil redistribution on livestock-poached pasture

Recent developments in fingerprinting and sediment-source tracing techniques have demonstrated that the contribution of fine-sediment from pasture is considerably greater than originally assumed (Collins et al., 1997; Russell et al., 2001). Stimulated by those findings, numerous studies seeking to identify potential sediment-sources (Brazier et al., 2007; Bilotta et al., 2008) have singled-out the effect of poaching by livestock (Warren et al., 1986a, 1986b; Pietola et al., 2005; Haygarth et al., 2006; Bilotta et al., 2007, 2008). Poaching occurs where livestock congregate, such as on the banks of watercourses, around feed-troughs and in gate-entrances (Figure 4). This increases the susceptibility of the soil to erosion, particularly by rainsplash and surface runoff, due to its degraded structure (Kauffman and Krueger, 1984; Warren et al., 1986b).

Figure 4. An example of a heavily poached gateway.

Little work has been done to quantify the contribution of sediment from poached areas, however, owing to a paucity of tracing-techniques capable of measuring its movement at the required spatial and temporal scales (Granger et al., 2007). A field-based technique was thus developed to quantify rates of soil redistribution on small areas of poached pasture over a series of rainfall events. The approach was adapted from an established technique for estimating catchment-scale erosion rates in undisturbed soils using the fallout radionuclide, ^{137}Cs (Walling and Quine, 1990, 1992; Walling et al., 2002). The approach involved directly labelling small plots of poached soil with a known activity of either ^{134}Cs or ^{60}Co. Predetermined measuring points were established across each plot and the areal activity at each point was estimated before and after periods of rainfall. By documenting spatial variations in the areal activity before and after periods of rainfall, quantitative soil redistribution values could be estimated at each of the measuring points within each plot using an established mathematical accounting procedure (Loughran, 1989; Walling and Quine, 1990, 1992; Walling et al. 2002).

By the end of the 65 d monitoring period, all plots recorded a mean net reduction in the overall soil-depth ranging between -6.8 to -15.2 mm. In quantitative terms, these values represent an equivalent mean soil loss of between 0.134 to 0.302 kg d^{-1}. Overall, these findings provide evidence that areas of livestock-poached pasture can represent a substantial source of eroded sediment.

Determining the grain-size composition of inter-rill and rill-eroded sediment

Many attempts have been made to determine whether contrasting hydraulic conditions associated with inter-rill and rill erosion are reflected in the grain-size selectivity and hence, in the particle-size composition of eroded material (e.g. Young and Onstad, 1978; Alberts et al., 1980; Farenhorst and Bryan, 1995; Fox and Bryan, 1999; Chaplot and Le Bissonais, 2000; Basic et al., 2002; Leguédois and Le Bissonais, 2004; Yang et al., 2006). Attributing the erosion of sediment to a particular process is extremely difficult, however (Yang et al., 2006), and where attempts have been made, the results have often been contradictory. Against this uncertainty, a stratified tracing technique, using both radionuclides simultaneously, was developed to identify changes in sediment-source during the transition from inter-rill to rill erosion. The approach was adapted from techniques developed by Wallbrink and Murray (1993), Wallbrink et al. (1999), Whiting et al. (2001) and Yang et al. (2006), all of whom exploited the contrasting depth-distribution of varying fallout radionuclides to determine the depth-origin of eroded sediment and predict the mobilisation process responsible. Conducting a similar investigation using artificial radionuclides, however, afforded a level of convenience that permitted testing eight different agricultural soils under laboratory-conditions relatively rapidly, and under a standard suite of eroding conditions. The approach involved filling a small erosion plot with a soil, pre-labelled with a known activity of ^{134}Cs, to a depth of ca. 0.1 m. This was covered with an additional 0.01 m layer of the same soil, pre-labelled with a known activity of ^{60}Co. The principle behind the 'stratified' configuration, seen in Figure 5, capitalises on erosion as a time-variant process (Yang et a., 2006), during which, the early stages are typically dominated by inter-rill erosion which preferentially removes fine material almost exclusively from the surface due to its low transport capacity (Alberts et al., 1980; Abrahams et al., 1998; Parsons et al., 1998; Leguédois and Le Bissonais, 2004; Ahmadi et al., 2006).

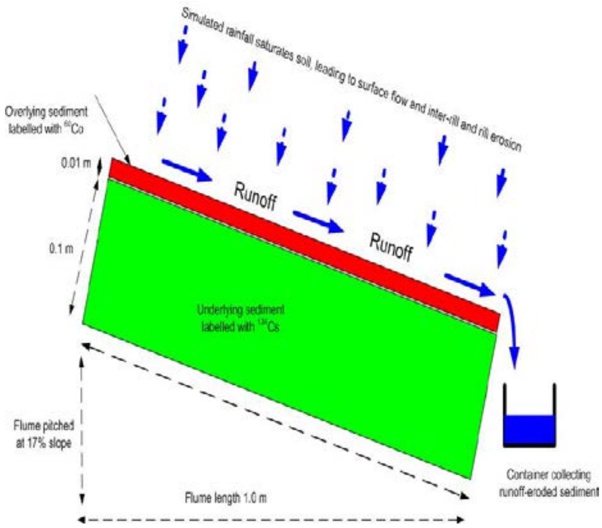

Figure 5. The stratified configuration of the two layers of labelled soil in the erosion plot.

As the rainfall event proceeds in time, surface-flow becomes concentrated, rill erosion becomes the predominant mobilisation process (Figure 6) and the increased transport capacity erodes coarser material when compared with inter-rill eroded sediment. In order to test this hypothesis, simulated rainfall was applied to each soil at a sufficient intensity and duration to initiate inter-rill erosion leading to rill erosion. Runoff was collected continuously over equal time-intervals and the sediment was separated from the liquid, processed following the method described earlier and radiometrically assayed. By examining changes in the radiometric signals of individual samples, it was possible to identify the transition from inter-rill to rill erosion and hence, the change from surface to sub-surface sediment-source. The radiometric data provided a basis by which a pair of samples were selected from each soil that were considered to be most characteristic of each mobilisation process.

The particle size composition of each pair of samples from each soil was determined and the grain-size was measured at corresponding 10 percentile intervals and statistically tested to determine whether the size-difference was significant. The results indicate that rill-eroded sediment was significantly coarser than inter-rill eroded material for the majority of soils investigated.

Figure 6. A partially developed rill-system.

Conclusion

Artificial gamma-emitting radionuclides posses a number of unique characteristics that are readily exploitable when undertaking sediment tracing investigations. With careful experimental design, these can be utilised to develop novel or diverse tracing techniques to investigate certain erosion scenarios where other tracers or existing tracing-techniques would be ineffective or lack the sensitivity needed to provide accurate data at the required spatial and temporal scales. This conclusion is supported by the findings from the three investigations described.

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