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A Review of the Radionuclide, Cobalt-60, as a Fine-Sediment Tracer

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Abstract

The chapter firstly outlines the global crisis concerning accelerated soil erosion, the implications that this could have on longer-term food security and ways that soil loss has been quantified originally through catchment-scale monitoring but with a recent shift toward the use of sediment tracers. It then briefly reviews some of the most commonly used tracers in sedimentation studies, before focusing on artificial, gamma-emitting radionuclides, and in particular, on Cobalt-60 (Co-60). Some historical background information on previous uses of Co-60 are then provided, and the suite of key environmental characteristics that, from the perspective of studies in hydrology and geomorphology, make Co-60 a potentially attractive candidate for fine-sediment tracing. The chapter then outlines and reviews three ways in which Co-60 has been experimentally applied in varying erosion and sedimentation scenarios where most of the more commonly used tracers would be unsuitable or would lack the level of sensitivity needed to return meaningful data. It then outlines some of the notable drawbacks associated with using Co-60, before highlighting refinements and prospects for future work. The chapter finally concludes by evaluating the versatility and efficacy of Co-60 as a fine-sediment tracer.

Keywords: soil erosion, land degradation, Cobalt-60, fine-sediment tracer, sediment transfer mechanism, river floodplain, remobilisation, earthworm casts, livestock-poaching, sediment-source

1. Introduction

1.1. The importance of sediment studies

The mobilisation, transportation and deposition of fine sediment (defined here as particles and aggregates <2 mm dia.) by rainfall and surface run-off are natural processes but are often accelerated by certain human activities that include *inter alia*, deforestation, modern

agricultural practises and/or adopting inappropriate land uses [1, 2]. The recognition that soil and sediment both represent a vital resource has led to concerns that accelerated soil erosion, along with the concomitant degradation of agricultural land, poses significant threats to sustainable food production to long-term global food security [3, 4] and to the quality of the environment *per se* [5]. Although inherently difficult to quantitatively express the degree to which land is, or has been, degraded at the global scale, varying authors have offered numerous statistics in an attempt to judge the severity of the problem. In the mid-1980s, for instance, Brown [6] concluded that global soil depletion annually exceeded the amount that could be replenished under natural conditions by around 23 billion tonnes each year. In the 1990s, Oldeman et al. [91] debated that two billion hectares (ha), representing 22.5% of all productive land on Earth, was more degraded than it was in the mid-1940s. More recently, Imeson [7] reported that global agricultural cropland productivity is diminishing by an average of 5% each year, principally by water-driven soil erosion but undoubtedly exacerbated by modern agricultural practises, whereas others such as Cherlet and Ivits [8] concluded that approximately 24 billion tonnes of fertile soil is lost to erosion and degradation each year, which, they speculate, could lead to a 12% reduction in global food production in the next 25 years. Against an expanding world population reliant on finite land on which to produce food and offer continuing food security [9], it is now acknowledged that the accelerated loss of a non-renewable resource such as soil is unsustainable which not only leads to land degradation and reduced food production *per se*, but also can lead to many environmental problems seemingly unconnected to agriculture and food production [10].

Reasons for many environmental problems can be attributed to fine to fine sediment which acts as a vector for the transfer and off-site conveyance of nutrients, which, when in concentrated form, can readily act as a pollutant. The loss of nutrient-rich fine sediment from agricultural land not only leads to a gradual reduction in the fertility and yield of crops from that land, but also the transfer of fine sediment carried away in agricultural run-off inevitably contributes to reduced water quality in receiving watercourses and to the widespread degradation and eutrophication of aquatic habitats downstream of its entry point [11]. Indeed, excessive concentrations of fine sediment, carried in suspension from field to watercourse, also have the ability to physically disrupt the internal functioning of aquatic environments and can act as a diffuse source of contamination in both terrestrial and aquatic ecosystems alike [12].

In response to the threat that excessive loss of soil poses to the environment, hill slope and catchment processes have come under increasing scrutiny in their role as sediment sources and sinks. Against the need to improve water quality in fluvial and aquatic environments, legislation such as the European Union (EU) Water Framework Directive (WFD) [13] has sought to promote a holistic approach to river basin management, with the net effect of reducing pollution and the amount of sediment entering into watercourses, in order to preserve and enhance their biodiversity and amenity status and sustain their future well-being for the benefit of all stakeholders [13]. Motivated by this legislation, the formulation of catchment-scale management plans has highlighted an urgent need for effective sediment control strategies to manage and mitigate the deleterious effects of fine sediment in the wider environment but particularly to reduce agricultural run-off from leaving the field (**Figure 1**) and ultimately entering aquatic environments. The effectiveness of such measures, however, ultimately

depends on the research community being able to identify and thereafter quantify the spatial and temporal flux of fine sediment within catchments [14].

Prognoses for estimating sediment movement within catchments have typically relied on monitoring techniques, with the focus on quantifying sediment yields at catchment outlets [15]. Although this approach has provided an initial framework around which overall annual sediment budgets have been constructed, recognition that not all mobilised fine sediments actually reaches the river but, instead, may be temporarily stored on depositional areas that have directed increasing attention to the internal functioning of river catchments themselves [11]. Researchers have thus sought to gain a more detailed understanding of the myriad sediment conveyance mechanisms that link upstream mobilisation of fine sediment to the downstream sediment yield and to determine the relative efficiency of such mechanisms in conveying sediment from the hill slope to the river [16]. However, identifying and elucidating how important those links are remains one of the most complex and least understood components of a catchment system. Importantly, it has also highlighted limitations in the ability of many existing monitoring techniques to provide sufficiently detailed information to enable more meaningful sediment budgets that integrate sediment sources and sinks, and the conveyance pathways interposed between them to be constructed. Those limitations have necessarily prompted a shift from purely monitoring the movement of fine-sediment toward tracing its movement, but the latter approach was very much viewed as mutually complementing the former approach rather than replacing it [17, 18].



Figure 1. Agricultural run-off leaving a recently cultivated field is often laden with fine-sediment and sediment-bound nutrients, such as phosphorous and nitrogen.

2. Sediment tracers

A tracer can be defined as any physical object or chemical substance capable of being introduced into a physical, chemical, or biological system, in order that it may be subsequently tracked over a given timeframe and provide information that can be inferred by its redistribution or dispersal [19, 20]. However, the suitability of any tracer depends on its ability to meet certain requirements. From the perspective of work presented and reviewed here, arguably the most fundamental characteristics are:

- i. ease of identification;
- ii. the ability to mimic the behaviour of the media being traced as closely as possible, yet remain distinguishable from its surroundings; and
- iii. an ability to remain as conservative in the environment as possible [19–21].

A wide variety of materials and substances meet those requirements, and the choice of tracer will ultimately depend on a number of things. Arguably, those that are most fundamental are the cost and availability of the tracers themselves, the area under investigation, and hence, the area over which the tracers can be applied and recovered, but also the textural characteristics of the sediment being traced and whether it is fine, medium, or coarse grained. Since most sediment-related environmental problems relate to the fine-sediment fraction, a very brief review follows that accordingly focuses on some of the most commonly used types of tracers that have been used to study the movement of fine sediment.

2.1. Passive and active tracers: a brief review

For clarity, sediment tracers can be divided into two sub-groups: those that are ‘passive’ and those that are ‘active’. Definitions of passive and active tracers can vary, depending on the research discipline making the distinction, the nature of the research being undertaken, and the frequency and stage during an investigation at which measurements of the movement or dispersal of the tracer/sediment can be undertaken (i.e., during or after an event). In light of this uncertainty, the following text provides a working yet extended definition of passive and active tracers, in the context of the work presented and reviewed in this chapter.

2.1.1. *Passive tracers*

Passive tracers are defined as those that have no additional energy source external to the combined force moving both sediment and tracer together. In essence, therefore, passive tracers are inert, and their redetection cannot be undertaken via an internal function of the tracer itself. Passive tracers come in many forms, but ideally their physical size and density should match, as closely as possible, the texture of the soil being investigated. Examples of some of the more commonly used passive tracers in soil erosion studies include metal cubes (e.g. [22]), plastic beads (e.g. [23]) and coloured gravel (e.g. [24]). As such, passive tracers are often commonly referred to as physical point tracers, and importantly, their inert nature means that methods of recovery and translocation distance typically involve relocation by

physical-based approaches. For fine, particulate-sized material, this may involve systematically removing small quantities of surface material at different locations along a line of erosion and then recovering the tracer material from the mobilised/deposited sediment. Rates of tracer displacement, and hence rates of mobilisation, can then be inferred based on changes in the concentration of the tracer relative to its original concentration prior to the erosion event. For larger physical point tracers, their relocation may involve more elementary approaches, such as field-walking, to observe and then record the location of individual tracers relative to their original position before the erosion event. By necessity, however, all recovery methods are typically performed after one or a series of consecutive erosion events. As the recovery of passive tracers is exploratory, it is invariably also invasive for accessing and trampling the site, and disturbing and removing at least some of the host sediment. Consequently, this typically restricts the use of passive tracers to measuring erosion over single, yet high energy, events, or over events that occur in quick succession. For this reason, passive tracers have proven to be ideally suited for measuring rates of soil translocation during tillage operations on hill slopes (e.g. [25]).

Substances known as rare earth elements (REEs) have proved particularly useful for studying erosion over relatively short timescales, particularly in areas where high erosion rates reduce the effectiveness of other techniques (e.g. [26, 27]). Numerous characteristics have contributed to their popularity, and these include their availability, their conservative behaviour, and their ability to bind to individual clay particles but also to coalesce and thereafter mimic the behaviour of aggregates. Low background concentrations in most environments also permit high analytical sensitivity [28]. Other positive attributes include their inert nature, benign characteristics, and the ability to detect different REEs simultaneously, usually by inductively coupled plasma-mass spectrometry (ICP-MS). Disadvantages with this type of material, however, include the need for detailed sampling campaigns covering the full extent of an area under investigation to provide a sufficient level of spatial resolution. As this represents a prerequisite, to ensure representative erosion estimates are obtained from across the entire study area and not just erosion 'hot-spots', this usually limits the area over which REEs can feasibly be applied to just a few 10 s of m². In addition to this, larger areas are difficult to access to physically take samples for analysis without altering or manipulating the very soil surface on which subsequent data may be based.

2.1.2. *Active tracers*

In contrast to passive tracers, active tracers are defined as any material or substance whose redetection and subsequent measurement are facilitated by an internal function of the tracer itself [19]. Such internal functions often have the ability to operate both independently and irrespective of environmental conditions [29]. This permits modes of identification and measurement to be conducted using a variety of different techniques, many of which are non-invasive and/or non-destructive and can thus be performed repeatedly, either in-situ and/or remotely. Those attributes permit measurements to be undertaken at different stages during the operational phase of an experiment; an attribute that affords researchers the opportunity to iteratively monitor the movement or dispersal of the tracer over a wider range of timescales. In most instances, therefore, the main characteristics evidenced by active tracers are

often viewed positively, as they confer a major advantage over passive tracers, by allowing repeat measurements to be performed over one or a sequence of erosion events occurring in quick succession or even over slightly longer (i.e., seasonal) timescales [30, 31].

Some examples of active tracers previously used in erosion experiments include magnetic powder (i.e., magnetite) sorbed onto small polystyrene beads and incorporated into a host soil at known concentrations. Changes in the magnetic susceptibility of the soil relative to the original value thereafter provide a means of determining the quantity of mobilised soil (e.g. [32]). Similarly, crushed magnetite with textural characteristics similar to a host soil has been introduced at a point-source location at known concentrations within a bounded erosion plot. Samples of run-off-derived sediment were recovered from the plot outlet after a series of erosion events and re-measured. An elevated magnetic signal relative to the background level from the host soil allows researchers to not only quantify the travel distance of the labelled material but also infer how much of the magnetised material has been mobilised (e.g. [33]).

In addition to the above, ceramic prills have been labelled with dysprosium oxide (Dy_2O_3), which, after exposure to rainfall, have been recovered and subjected to neutron activation analysis (NAA), in order to make the dysprosium oxide (temporarily) radioactive, thereby allowing soil redistribution rates to be determined (e.g. [34]). Alternatively, a more complex labelling approach has been undertaken, whereby Europium (Eu_2O_3) was incorporated into molten glass, which was then cooled and grounded to different sizes to represent soil aggregates. Prior to cooling, lead was also introduced into the mixture, which facilitated adjusting the bulk density of the tracer to the host soil [35]. Similar to the previous approach, the mixture was then subjected to NAA; transforming the Europium into the gamma-emitting radionuclide, Eu-125 m ($^{125\text{m}}\text{Eu}$), whereupon the labelled material was used in a variety of erosion investigations.

2.1.3. *Fallout radionuclides as sediment tracers*

A number of globally distributed (i.e. fallout) radionuclides, namely lead-210 (^{210}Pb), beryllium-7 (^7Be), and caesium-137 (^{137}Cs), naturally decay by γ -radiation and express a very high affinity for fine sediment. Consequently, all three sorb to individual clay and silt particles, and aggregates, rapidly and very strongly on contact. This means that in undisturbed soils, most of the fallout inventory remains within the upper part of the soil profile. Both attributes render all three radionuclides useful for studying the redistribution of surface sediment. Over the last ca. five decades, therefore, researchers have increasingly sought to harness those characteristics through developing and refining ways in which those particular fallout radionuclides can be exploited and used to investigate soil redistribution processes at the field to catchment scale (e.g. [36, 92]). Continual refinements to the original technique have steadily provided researchers with an increasingly deeper understanding of sediment dynamics operating both within and between river catchments. Reasons for many of these process-based advances have largely been attributed to the use of tracers that undergo nuclear decay. This produces a signature along parts of the electromagnetic spectrum (EMS) that is not only unique and provides means of identifying each radionuclide, but the rate of decay is specific to each radionuclide and operates independently of fundamental environmental conditions; namely temperature,

pressure, light, and moisture, which can unduly alter or influence the behaviour of other tracers, while investigations are still in progress [29]. As such, the rate at which certain radionuclides undergo nuclear decay has duly allowed a temporal perspective to be established for many environmental processes [37]. Indeed, timeframes have now been attributed to a wide range of sedimentation-related mechanisms once considered too slow and hence immeasurable or simply too innocuous to merit closer attention [11, 38–42]. In the case of the anthropogenic ^{137}Cs ($t_{0.5} = 30.2$ years) and the lithogenic ^{210}Pb ($t_{0.5} = 22$ years), those two medium-term fallout radionuclides have permitted field and catchment-scale information on sediment redistribution to be estimated retrospectively over timescales of ca. 50 and 100 years, respectively (e.g. [43, 44]). This has provided researchers with means of quantifying a number of fundamental catchment processes that include estimating the volume of sediment deposited across river floodplains during flood events, quantifying the residence time of deposited sediment on river floodplains and seeking evidence of catchment responses to longer term land-use changes [44]. However, while those factors represent major advances in our understanding of the way in which catchments function, estimates of erosion and deposition using fallout ^{137}Cs and ^{210}Pb must, by necessity, be presented as yearly averages over the aforementioned time periods only. Consequently, those approaches are unable to quantify soil redistribution over short or event-based timescales, or where erosion may have occurred due to sudden changes in land-use conditions [45]. In situations where event-based redistribution data are required at the field-scale and larger, recent developments using the short-lived cosmogenic fallout radionuclide, ^7Be ($t_{0.5} = 53$ days), have proven successful (e.g. [46, 47]); to the point where it is now established as an additional technique that compliments the use of the two medium-term tracers described above. However, a major disadvantage associated with fallout ^7Be is that it can be readily sequestered by surface vegetation [48]. Depending on the type and density of the vegetation at a given study area, its presence can adversely influence the amount of inventory that actually reaches the soil surface. Since this can initiate changes in the depth distribution of ^7Be in the soil profile, while the investigation is still in progress [49], small-scale variations in inventory values may produce erroneous results. Consequently, studies using ^7Be have tended to be focused on areas where soils have recently been cultivated and the surface is still free of vegetation, or areas where vegetation cover is sparse or crop residue is low [48].

2.1.4. Artificial gamma-emitting radionuclides as sediment tracers

The use of artificial gamma-emitting radionuclides as sediment tracers, or to clarify, radionuclides that have been deliberately introduced into or across a small predefined study area or used to 'label' small quantities of fine sediment with low-level radiation have the potential to overcome many of the limitations associated with some of the more commonly used tracers outlined and reviewed above. They are not, however, designed to replace existing tracers but instead should be viewed as an additional tool in the proverbial 'toolbox' of tracers and tracing techniques that researchers can draw upon during instances where detailed information about sediment redistribution is required at the small scale and at high temporal resolution.

There are a number of notable advantages associated with certain artificial radionuclides as fine-sediment tracers; particularly, species that undergo nuclear decay by emitting gamma radiation. Among the most noteworthy advantages included in their limited distribution in

the wider environment, the level of convenience afforded by their use in soil erosion investigations and, above all, the conservative behaviour, and hence, the ability of certain artificial radionuclides to sorb to individual soil particles and aggregates rapidly and strongly. This attribute enables them to faithfully mimic the behaviour of fine sediment during an erosion event without physically perturbing or adversely influencing its behaviour while in transit. 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 [38]. This particular advantage can be attributed to the 'minimal mass theory' proposed by authors such as Sauzay [20], Courtois (1973) and echoed latterly by McCubbin and Leonard [50]. The general principle has previously been alluded to in an earlier section, but to explain in more detail, it argues that the behaviour of any object or substance used as a tracer must be as compatible as possible with the medium being traced in order to derive accurate and representative information. Because individual gamma photons carry very little mass but emit a powerful and unique signature that is distinct to each radionuclide, any labelled sediment that is subsequently redistributed will carry vestiges of that same signature, albeit not as strongly as the original signature before dispersal. By measuring and comparing the strength of the signature (also referred to as an 'inventory') before and after an erosion event, the change not only indicates that labelled material had been dispersed but can also be used to quantify the mass of sediment dispersal [30, 31, 38, 50, 51]. The process of nuclear decay is particularly useful to tracer scientists undertaking studies of certain sedimentation processes. This is because one of the most commonly occurring, ubiquitous and hence important erosion mechanisms is particularly subtle, since it occurs on shallow slopes under low-energy conditions. The process, which is known as 'sheetwash' or 'inter-rill erosion', preferentially entrains small particles that have previously been detached by raindrops as they impact against the soil surface. It then carries the liberated sediment under the influence of gravity downslope in a thin layer of water (often <3 mm deep) [52]. Sheetwash commonly occurs on all low-gradient slopes and is the precursor to the more visually obvious process known as rill erosion, where topography serves to concentrate the overland flow, whereupon it becomes much faster flowing and has sufficient hydraulic power to incise its way down through the soil profile [38], indiscriminately detaching and transporting fine-sediment of all sizes (**Figure 2**). In areas where deposition of sheetwash-derived sediment takes place, gamma-emitting radionuclides can be particularly useful for measuring certain sedimentation processes. This is because only a few gamma photons are required to identify the species of radionuclide. Arguably, therefore, one of the biggest attributes associated with radionuclides that emit γ -radiation is the relative ease with which gamma photons can be detected, measured, and used to speciate individual radionuclides. Due to the fact that gamma photons are ejected at very high energy but have negligible mass, they are highly penetrative and easily travel through most substances, including thin layers of deposited sediment. Consequently, re-identification of small quantities of pre-labelled sediment is possible, even in depositional areas, where subsequent burial by unlabelled material may have occurred [39, 40]. Because of the above attributes, soil that has been pre-labelled, even with relatively low levels of a suitable radionuclide, will theoretically not only behave in a manner that is identical to unlabelled soil, but also its redistribution or burial can be detected and accurately measured with a level of sensitivity that far exceeds the majority of other tracers.



Figure 2. When un-concentrated overland flow becomes concentrated, it forms a network of small channels called rills which gradually incise into the soil profile and transports the eroded material downslope.

Despite numerous demonstrations of the versatility and effectiveness of artificial radionuclides to provide reliable information on fine-sediment redistribution over short timescales, however (e.g. [30, 31, 38, 40, 53–56]), their use in erosion investigations has attracted surprisingly little attention over the last ca. 50 years. Reasons for the slow uptake remain unclear but may be attributed to their controversial nature and the commensurately stringent suite of laws that govern the purchase and subsequent release of open-source radionuclides in the environment (e.g. [57]), difficulties associated with uniformly labelling soils and sediments prior to undertaking in- or ex-situ erosion experiments and the limited areal extent over which they can realistically be applied (*cf.* [54]). With the last constraint in mind, research using artificial radionuclides has tended to focus on small-scale investigations, where the area of application may be less than one square metre [58] and certainly no larger than a few hundred square metres [54]. Despite this apparent limitation, numerous workers have circumvented this scale constraint by utilising a point-source tracing approach, whereby the radionuclide may be applied to a network of small areas across, for instance, a field-scale study area (e.g. [38, 40]). Such an approach has proven to be both convenient and very effective at iteratively documenting soil redistribution data over one or a more consecutive rainfall or flood events, particularly if care is taken to select representative locations within the larger study area.

2.1.5. *The anthropogenic radionuclide, Cobalt-60*

Cobalt-60 is an anthropogenic, gamma-emitting radionuclide with a relatively short half-life ($t_{0.5}$) of approximately 5.26 years [59]. It is classified as an activation product that is inadvertently manufactured in nuclear reactors by neutron activation of the naturally occurring isotope, Cobalt-59 (^{59}Co). Cobalt-59 is a stable metal element that is relatively ubiquitous in

the environment and occurs in various minerals and metal ores and typically remains as an impurity within most finished metal products. Metals containing ^{59}Co are frequently, and inadvertently, used in the construction of nuclear reactors. Upon exposure to the intense radiation field within the reactor, ^{59}Co absorbs a free neutron into its nucleus, transforming it into the unstable radioisotope, ^{60}Co . Once transformed, ^{60}Co begins to emit ionising radiation in the form of beta particles (β^-) and gamma radiation (γ -radiation). After 5.26 years has elapsed (i.e. one half-life), radioactive decay transforms 50% of the original ^{60}Co nuclei into its stable progeny, nickel-60 (^{60}Ni) [60]. Cobalt-60 emits γ -radiation at photo-peaks of approximately 1173 and 1332 kilo electron volts (keV). The energy field, which is defined as the proportion of ^{60}Co atoms produced per 100 disintegrations, is ca. 100% at each photo peak. It is therefore theoretically possible to obtain accurate results from samples emitting activities as low as 1.0 Becquerel (Bq) kg^{-1} of (dry) sediment, assuming long (≥ 1 day) analytical counting times on a laboratory-grade gamma spectrometer.

With regards to its presence in the environment, small quantities of ^{60}Co are discharged under licence as effluent into marine environments from nuclear reprocessing plants such as La Hague, in northern France, and Sellafield in Cumbria, UK [61–63]. Discharges of ^{60}Co can be detected in close proximity to its release point in samples of seawater [61, 64], in surface sediments or from shallow sediment cores [62, 63]. Some researchers have exploited these licenced releases and used the signature of ^{60}Co as a tracer or geochemical marker to elucidate varying environmental processes [61, 62, 64, 65]. Further to those licenced releases noted above, areas in close proximity to disused underwater nuclear test sites also represent additional, yet highly localised, sources of ^{60}Co [63].

Owing to its purely anthropogenic origin, its relatively short half-life and limited discharge sources, ^{60}Co does not, therefore, naturally occur in the wider environment. This not only simplifies things when undertaking sediment tracing investigations, but it also represents a notable advantage for a number of other reasons. Principally, an absence of background contamination removes the possibility of obtaining spurious data due to spatially variable background levels (*cf.* [66]). This permits both tracer inputs and outputs to be very precisely quantified [20]. This also facilitates the likelihood that subtle changes in radionuclide inventories after an erosion event could signify very subtle movement of labelled sediment, instead of being potentially attributable to spatial variations in background concentrations. Further to the above, an absence of background contamination also permits the use of lower initial dose rates (i.e. the amount of inventory sorbed to a given mass of sediment, or sorbed over a given area) when undertaking tracing investigations [53]. Importantly, all the above factors collectively contribute to potentially increasing the analytical sensitivity, to improving data accuracy, and increasing the longevity of the monitoring campaign.

A desktop review of ^{60}Co reveals that a substantial amount of literature has been compiled which documents the characteristics, behaviour, and mobility of ^{60}Co under varying environmental scenarios (e.g. [59, 63, 67–69]). Most relevant to work presented in this chapter are the rate and strength of retention (sorption) to fine sediment, the solubility and migration of ^{60}Co , and its rate of biological uptake (bio-uptake) and accumulation, and thus its potential toxicity to organisms.

With respect to the first two parameters, the solid/liquid distribution coefficient (K_d) values have been used to describe the mobility, sorption, and desorption of ^{60}Co in response to a range of environmental and physical controls. These range from particle size characteristics of the material being labelled and traced, the organic matter (OM) content, pH, Eh, temperature, moisture and time [59, 68, 70]. K_d values for ^{60}Co , obtained from wide ranging soil-types, indicate that sorption is typically high to very high [59, 69]. Numerous researchers have reported that pH represents a major controlling factor that can frequently dominate the rate at which sorption occurs, especially in acidic (i.e. $\leq \text{pH } 6$) environments [59, 68]. However, the influence exerted by pH diminishes very rapidly from ca. pH neutral and upwards, and by $\sim \text{pH } 8$, sorption to sediment is reportedly $\sim 100\%$ [59, 71, 72]. Under environmental conditions typically encountered in agricultural environments, therefore, sorption of ^{60}Co to soil is both rapid and considered to be largely irreversible [59, 69, 71, 73]. The environmental characteristics of ^{60}Co have resulted in it being used as a tracer in a wide variety of investigations over approximately the last six decades. To demonstrate this, some investigations from the available literature have been compiled and are listed in **Table 1** in the order of publication.

Although the list is neither exhaustive nor comprehensive, it provides some indication of the diverse applications in which ^{60}Co has previously been used and thus highlights its potential versatility as a tracer of many environmental processes. Owing to the relatively limited number of ^{60}Co sources and pathways for its entry into the environment, however, very little literature exists regarding its biological uptake, accumulation and potential toxicity to organisms. Studies that have made reference to those particular characteristics do not discuss its toxicity potential in great detail (e.g. [51, 55, 56, 69, 74, 75]) but infer that its likely rate of transfer and accumulation from soils into and through the food chain and its injurious potential to organisms at low concentrations are minimal. This inference is corroborated, albeit by logic alone, via the wide variety of ecological and medical tracing applications where ^{60}Co has been introduced onto, or into, numerous organisms (including humans) during tracing investigations, some of which are listed in **Table 1**.

With regard to the use of ^{60}Co in sediment tracing studies, Toth and Alderfer [55] document two of the earliest known applications, with the first publications providing details of the development of a procedure for uniformly tagging (labelling) water stable aggregates (WSAs) by immersion into solutions of ^{60}Co and water. The success of the labelling technique culminated in the same authors conducting a year-long investigation into the formation and breakdown of the ^{60}Co -labelled WSAs [56]. The reported success of both the labelling technique and the tracing application provided an initial indication of the potential value of ^{60}Co as a candidate for sediment tracing work of a similar nature. However, the apparent dearth of sediment-related tracing investigations since those published in 1959 implies that the versatility and possible value of ^{60}Co may have been ignored or inadvertently overlooked by subsequent researchers.

The following section now briefly describes procedures for sample processing, as well as, arguably, the most accurate and cost-effective means of measuring low level radioactivity in soils and sediments, namely by gamma spectrometry.

Data source	Publication year	Nature of investigation
Arnason et al. (in Ref. [51])	1950	Determine the movement of wireworms (<i>Ctenicera</i> spp.) in soil
Rings and Layne (in Ref. [51])	1953	Determine dispersal of beetles (<i>Conotrachelus</i> spp.) in woodland
Sullivan (in Ref. [51])	1953	Determine dispersal and habitat range of beetles (<i>Pissodes</i> spp.) in woodland
Babers et al...	1954	Determine dispersal of Boll Weevils (<i>Anthonomus</i> spp.)
Green and Spinks (in Ref. [51])	1955	Determine the movement of wireworms (<i>Agroites</i> spp.) in soil
Green et al. (in Ref. [51])	1957	Determine the movement of moth larvae (<i>Rhyacionia</i> spp.) in soil
Green et al. (in Ref. [51])	1957	Determine the dispersal of mature moths (<i>Rhyacionia</i> spp.)
Toth and Alderfer	1959a	Establish a procedure for labelling water stable aggregates by immersion
Toth and Alderfer	1959b	Monitor the formation and breakdown of labelled water stable aggregates
Traniello et al. (in Ref. [51])	1985	Determine territorial feeding patterns of termites (<i>Reticulitermes</i> spp.)
Rosengaus et al. (in Ref. [51])	1986	Determine of termites (<i>Reticulitermes</i> spp.)
Thorén et al. [94]	1991	Determine location of cancerous brain tumours in humans
Cundy and Croudace	1996	Wastewater effluent as a geochemical marker for estimating sedimentation rates
Alam et al. [90]	2001	Determine rates of nutrient uptake in agro-crops
Capowiez et al.	2001	Determine 3-dimensional trajectories of burrowing earthworms
Sattar et al.	2002	Determine feeding habits of termites (<i>Heterotermes</i> , <i>Microtermes</i> and <i>Reticulitermes</i> spp.)
Thompson et al.	2002	Wastewater effluent as a geochemical marker for estimating sedimentation rates
Greenwood et al.	2008	Used as a fine-sediment tracer
Schreiner et al. [93]	2009	Tomotherapy cancer treatment in humans
Greenwood	2012	Used as a fine-sediment tracer
Greenwood et al.	2013	Used as a fine-sediment tracer
Greenwood et al.	Accepted	Used as a fine-sediment tracer

Table 1. Selected studies, published from 1950 to the present day, that have used the artificial radionuclide, ^{60}Co , as a tracer of biological or environmental processes.

2.2. Sample processing & methods and units of measurement

As previously mentioned, each gamma-emitting radionuclide produces one or more photo peaks, or signatures, at known locations along the EMS, which allows each radionuclide species to be readily identified and their prevalence to be quantitatively determined. There are several methods of determining low-level radioactivity from soil and sediment samples. One of the most accurate and cost-effective methods is by gamma spectrometry using detectors fitted with a high-purity germanium crystal (HPGe) [76]. Sample analysis is a relatively simple procedure that involves minimal prior sample preparation. It is also non-destructive, which allows repeat measurements to be undertaken on individual samples. For laboratory-based analysis, the key stages involve drying, gently disaggregating and screening each sample through a 2-mm diameter sieve. Sieved material should be weighed to a predetermined quantity (i.e. ca. 100 g) and to an accuracy of one decimal place [77]. For reasons of precision, samples presented to a detector should be of similar mass (i.e. within ± 0.1 g), and radiometric assays should be performed in containers of identical proportions, so that the sample geometry, and hence, the distance from the sample to the germanium crystal, remains constant. Failure to comply with this rule can profoundly influence the precision of measurements, leading to erroneous results [76]. The sample is then placed in very close proximity to the germanium crystal, which is housed in the detector head (**Figure 3A**). Gamma-photons ejected

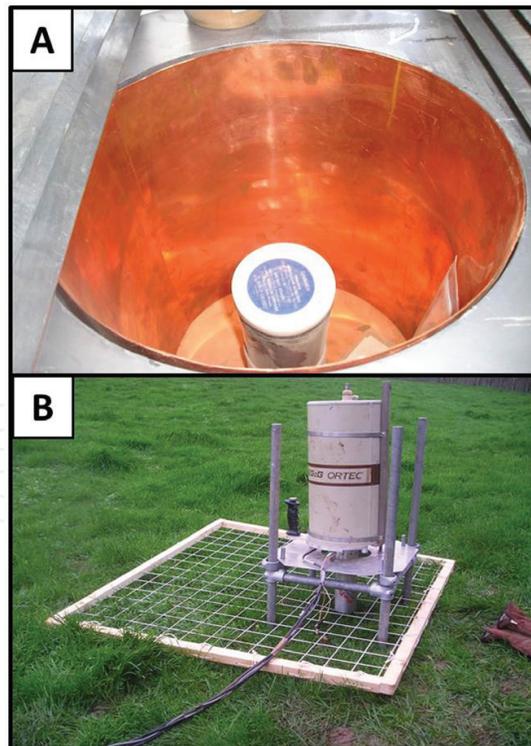


Figure 3. (A) The inside of a laboratory-grade gamma-spectrometer showing the top of the germanium crystal onto which a sediment sample is placed for analysis, and the lead and copper surround, with sliding cover lids to reduce interference from incoming background gamma radiation. (B) An in-situ gamma spectrometer is placed with the detector head facing downwards and close to the soil surface. Establishing and maintaining a consistent measurement protocol throughout a monitoring campaign is necessary to ensure that results are reliable.

from the sample interact with the electrically charged germanium crystal, which in-turn emits signals or pulses that correspond to the energy of the incoming gamma photons. These are routed through a preamplifier to an amplifier and then to a multi-channel analyser (MCA), whereupon the signals are displayed as a spectrum on a computer screen. Since the energy of the emitted photons is unique to each radionuclide, those that are present in the sample can be identified and their respective activity quantified. Field-based areal activity measurements can be performed in-situ using a field-based gamma spectrometer, shown in **Figure 3B**. Although no sample preparation is required when taking in-situ measurements, the crucial factor to obtain reliable results is to ensure that the radiometric protocol adopted at the beginning of the investigation remains consistent throughout the monitoring campaign (*cf.* [16, 40, 45]). More information about in-situ measurement protocols is given in the following section and can also be found in Greenwood [16] and Greenwood et al. [40].

The most convenient and appropriate measurement unit, which is also recognised by the International System of Units (SI), for assessing low-level radioactivity emitted by any radionuclide in soils and sediments is the Becquerel (Bq). One Bq is equal to one decay per second (dps). Because this unit is small, it is often convenient to use additional prefixes, such as the kBq (i.e. 10^3 Bq) or MBq (10^5 Bq). It is also often necessary to use an additional prefix, for instance, where the concentration of fallout radionuclides is being determined from small soil samples at typical environmental activity levels, which enable the activity concentration to be measured and expressed in milli-Becquerels (mBq) that are equal to 10^{-3} Bq.

3. Practical applications

The following sections now describes and reviews examples of three field-/laboratory-based prototype tracing techniques using ^{60}Co in order to demonstrate the novel and diverse ways in which it has recently been used as a fine-sediment tracer. The underlying motivation for each investigation was to acquire essentially unique information that would otherwise be unobtainable if cruder or less-sensitive types of tracer were used. As indicated earlier, the attribute of sensitivity has proven to be particularly beneficial during attempts to study, for instance, the movement, redistribution or remobilisation of fine sediment in environments where the energy regime may be low, and hence, redistribution rates are likely to be commensurately small, or where mobilisation occurs over very short (i.e. event based) timeframes.

3.1. Example 1: assessing flood-derived sediment remobilisation on river floodplains

3.1.1. *Experimental aim*

The aim of this investigation was to explore the potential for documenting the extent to which fine sediment, recently deposited on a river floodplain during an overbank flood, might be subject to remobilisation and redistribution. Further details regarding the approach and the findings can be found in Greenwood et al. [39, 40]. Motivation for the investigation arose due to the realisation that river floodplains not only represent important stores of fine sediment

but also act as stores of sediment-associated nutrients and contaminants [78]. Consequently, increasing awareness of their environmental importance has resulted in attempts to quantify the capacity of river floodplains to store fine sediment and to determine the residence time of the deposited material [79]. While those factors have been satisfactorily quantified to a certain extent using the aforementioned medium-term fallout radionuclides, ^{137}Cs and ^{210}Pb , measuring sediment accretion rates over shorter timescales and assessing the extent to which recently deposited sediment may be remobilised during subsequent flood events have, by contrast, received limited attention. Reasons for the oversight have traditionally been attributed to difficulties in tracing the movement of small quantities of fine sediment over short time periods. Scope therefore existed to design an experiment for the purpose of testing ^{60}Co on this hitherto unrecognised and unstudied sedimentation scenario by undertaking the following approach.

3.1.2. Method

The approach involved pre-labelling small (i.e. ca. 200 g) quantities of fine sediment with 80 Bq of ^{60}Co activity mixed in 250 ml of water (each forming an aliquot). Each aliquot was deposited back onto the floodplain during a simulated flood. A total of 15 aliquots were deposited at representative locations to form a series of 'active plots' (hereafter referred to as AP1–15)), within the flood-zone, so that labelled sediment on each plot would be subjected to the potential remobilising effect of the overbank floodwaters, when the floodplain was next inundated. A further three plots were installed above the flood zone and used as controls (i.e. hereafter referred to as control plots (CP1–3)), to account for the loss or removal of labelled sediment, and hence, radioactive inventory, by processes other than by floodwaters. A simulated flood was created using a 300-mm diameter bottomless container which was embedded into the floodplain surface, so that a watertight seal was obtained. In order to recreate overbank flood conditions, one aliquot of labelled sediment was added to a further 25 l of water, vigorously shaken and then poured into the container. The water was allowed to infiltrate into the ground, thereby depositing the ^{60}Co -labelled sediment onto the floodplain surface (**Figure 4**). This approach was repeated for all 18 plots, with each simulated flood depositing a layer of ^{60}Co -labelled sediment to a depth of approximately 1.4 mm, which was representative of conditions in local rivers during typical flood events. The radiometric inventory associated with each plot was recorded before and after consecutive flood events using an in-situ gamma spectrometer. Data from the active plots over the three flood events were then correlated against the magnitude of each event, as indicated using peak river discharge, as well as two key spatial controls, namely the depth to which each active plot was inundated and their respective distances from the main river channel.

3.1.3. Key results and discussion

Before and after inventory values were corrected for radioactive decay and the proportion of the radiometric inventory lost due to processes other than by remobilisation by floodwaters, as indicated by the average of the three control plots, was removed from all 15 active plots. Minimum and maximum error bars were calculated at the 90% confidence level in order to



Figure 4. A bottomless container was embedded into the soil surface of a floodplain and 200 g of ^{60}Co -labelled sediment, mixed in 25 l of water, were poured into the container and allowed to infiltrate, thereby depositing the labelled sediment onto the soil surface under simulated flood conditions.

show the uncertainty associated with before and after measurements for each plot. At this particular juncture in the research, it was considered more meaningful, should similar work be conducted in the future on river floodplains elsewhere, if variations in initial inventories between plots were normalised and converted to an equivalent mass of sediment (expressed in g m^{-2}). Consequently, all plots started with an initial depositional mass of sediment equivalent to 1386 g m^{-2} . Where error bars between before and after measurements for each active plot do not overlap, the change in inventory is considered to be significant. This is interpreted as clear evidence that the labelled sediment was remobilised by overbank floodwaters. **Figure 5A–C** shows which plots recorded significant remobilisation over three consecutive flood events (i.e. termed the extent of remobilisation). They also indicate how much labelled sediment remained at each active plot after each flood event (i.e. termed the magnitude of remobilisation). For brevity, both the extent and mean magnitude, based on the mean of all 15 active plots, are summarised in **Tables 2** and **3**, respectively. Looking at data shown in **Table 2**, a clear inverse trend can be seen, which shows numbers of active plots that recorded significant remobilisation diminished with increasing time after initial deposition. Indeed, significant remobilisation ceased completely during the third inundation event at 119 days after initial deposition. There was neither correlation between the magnitude of each flood and the magnitude of remobilisation nor any correlation between magnitude of remobilisation and depth. A moderate yet significant direct correlation was recorded between magnitude of remobilisation and distance from river channel during the first flood event only. The direct relationship indicates that remobilisation intensified with increasing distance from the river. This finding, although

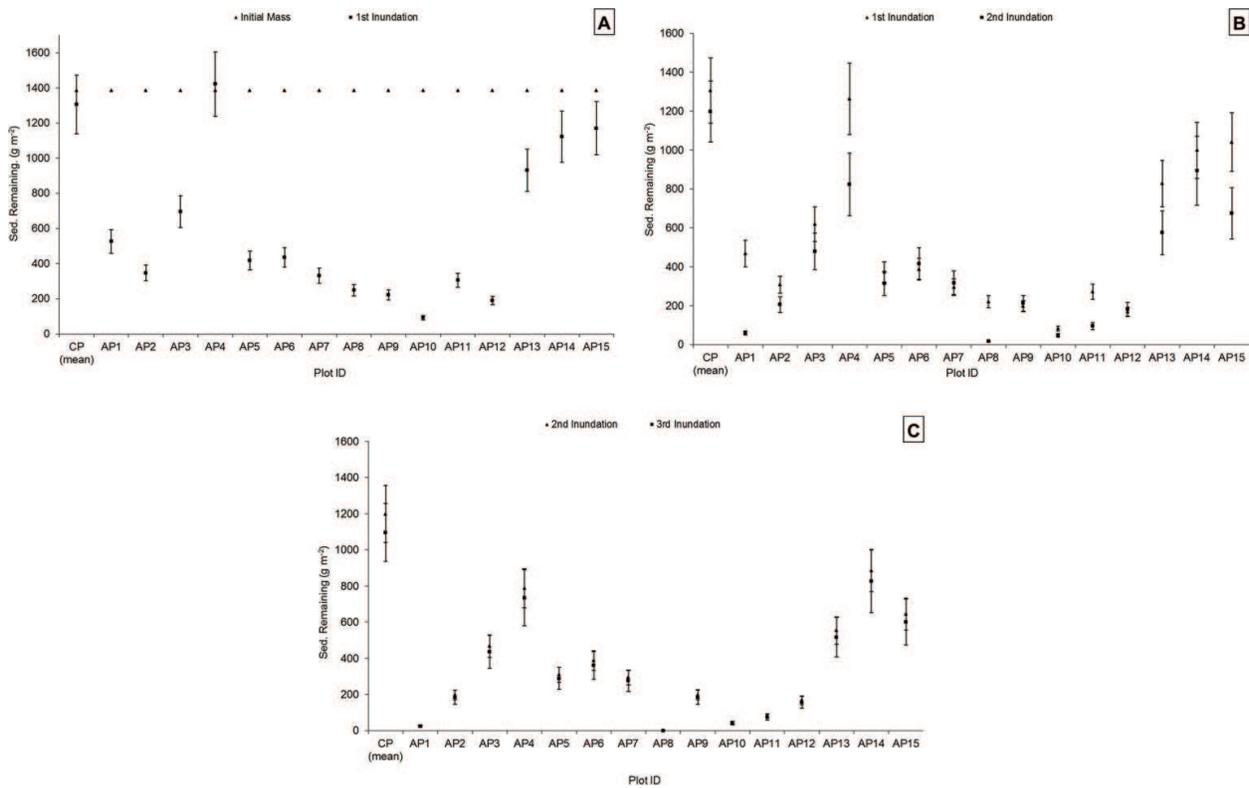


Figure 5. (A) Results of remobilisation after the first flood event. Error bars indicate the uncertainty associated with each measurement. Where error bars associated with before and after inundation values do not overlap, significant remobilisation of the ⁶⁰Co-labelled material has occurred at that location. Copyright © 2013 John Wiley & Sons, Ltd. (B) Remobilisation after the second flood event, although significant, was notably less than for the first flood event. Copyright © 2013 John Wiley & Sons, Ltd. (C) No significant remobilisation was recorded during the third flood event. Copyright © 2013 John Wiley & Sons, Ltd.

somewhat paradoxical, is attributed to the geometry and certain topographic features within this particular study site, both of which serve to temporarily divert but channel the fast-flowing floodwaters away from the river at the mid-point of the floodplain.

Interestingly, if data from **Table 2** are cross-referenced with data seen in **Table 3**, a number of key processes are revealed from which certain conclusions may be drawn. Firstly, flood-derived sediment appears to be extremely susceptible to remobilisation by subsequent floods during the first few weeks after deposition. Secondly, a significant amount of the same sediment

	Nos. of active plots	Inundation event		
		1st	2nd	3rd
% of active plots recording significant remobilisation	15	93 (14)	53 (8)	0
Time since initial deposition (days)	n/a	16	76	119

Nos active plots in parenthesis.

Table 2. The extent of remobilisation over three consecutive flood events, as indicated by ⁶⁰Co, and time since initial deposition for the River Taw floodplain.

Initial sed. mass (g m ⁻²)	Mean remobilisation mass (g m ⁻²)			Mean sed. mass remaining (g m ⁻²)
	Event 1	Event 2	Event 3	
1386	884.63 (63.8)	165.06 (11.9)	24.27 (1.8)	312.04 (22.5)

Equivalent % values in parenthesis.

Table 3. The mean magnitude of remobilisation over three consecutive flood events, as indicated using ⁶⁰Co, and the mean amount of labelled sediment remaining after the third flood event for the River Taw floodplain.

also appears to remain vulnerable to remobilisation for up to 2 months after initial deposition. Thirdly, despite the average magnitude of remobilisation being insignificant at 1.8% during the third event (**Table 3**), an average of 22.5% of the initial inventory (equivalent to 312.04 g m⁻²) remained after the third event. This is interpreted as evidence that the remaining portion of the flood-deposited (⁶⁰Co-labelled) sediment gradually stabilised. This stabilising effect is attributed to the material gradually becoming incorporated into the main soil profile and entering into temporary storage. This not only effectively made it unavailable for further remobilisation but also storage occurred almost 4 months after the sediment was initially deposited.

3.1.4. Uncertainties

A number of key uncertainties need to be acknowledged. Arguably, the most profound of which is that ⁶⁰Co is largely untried and untested as a fine-sediment tracer, and thus, the resultant data remain uncorroborated. Three independent approaches were undertaken in an attempt to rectify this uncertainty. The first approach involves presenting data from a parallel study using a different artificial radionuclide, namely Cs-134 (¹³⁴Cs; $t_{0.5} = 2.06$ years). Sediment labelled with ¹³⁴Cs was duly deployed in exactly the same manner described above but on another floodplain in a different river system. Although the flow regime of the two rivers and the floodplain characteristics differ markedly, equivalent remobilisation values and general timelines from data recorded using ¹³⁴Cs over three flood events in **Tables 4** and **5** are striking in their similarity with data from **Tables 2** and **3** using ⁶⁰Co. The second approach involved performing independent sorption tests on surface soil taken from each floodplain in order to determine whether the response of both radionuclides to provide information on remobilisation dynamics was comparable. Textural characteristics on surface soils taken from each floodplain (**Table 6**) indicated that <63 µm fraction (i.e. silt and clay) was generally similar and represented ca. 80–85% of each sample, as were other key indicators, including clay fractions

	Nos. of active plots	Inundation event		
		1st	2nd	3rd
% of active plots recording significant remobilisation	15	87 (13)	47 (7)	0
Time since initial deposition (days)	n/a	50	74	104

Nos active plots in parenthesis.

Table 4. The extent of remobilisation over three consecutive flood events, as indicated using ¹³⁴Cs, and time since initial deposition for the River Culm floodplain.

Initial Sed. Mass (g m ⁻²)	Mean remobilisation mass (g m ⁻²)			Mean sed. mass remaining (g m ⁻²)
	Event 1	Event 2	Event 3	
1386	687.17 (49.6)	173.03 (12.5)	37.94 (2.7)	487.82 (35.2)

Equivalent % values in parenthesis.

Table 5. The mean magnitude of remobilisation over three consecutive flood events, as indicated using ¹³⁴Cs, and the mean amount of labelled sediment remaining after the third flood event for the River Culm floodplain.

(<2 µm), median particle size (D₅₀) and SSA (**Table 6**). Sorption tests were undertaken on 12 soil samples from both floodplains according to Sparks [72], six samples of which were labelled with ⁶⁰Co and six labelled with ¹³⁴Cs, at the same activity concentration. Samples were then allowed to rest for 24 h, and then the solid and liquid fractions of each aliquot were assayed together. After decanting off the supernatant (liquid) fraction, both fractions were then re-assayed separately. After averaging individual values for each group, results indicated for both radionuclides that the solid (i.e. sediment) fraction retained 99.8% of the applied inventory. The third method of corroboration involved undertaking a ‘paired-plot’ approach, with the aim of comparing the response of both radionuclides under similar floodwater-flow conditions. Two active plots were established 0.15 m apart (measured from perimeter to perimeter) on a uniform area of one of the floodplains, one installed using ¹³⁴Cs-labelled sediment and the second installed using ⁶⁰Co-labelled sediment. An assumption was then made that floodwater-flow conditions across flat topography with sparse vegetation cover would be sufficiently similar over short distances [80, 81] to allow results from each set of results to be compared. Four sets of paired plots were established (hereafter referred to as PP1-PP4), all within a 6 m radius of each other, and all plots were duly inundated during the same flood event, to a depth of ca. 0.15 m. The results, which are shown in **Table 7**, reveal that ⁶⁰Co consistently records a slightly lower magnitude of remobilisation in comparison to data generated using ¹³⁴Cs. Differences range from a negligible 0.5% (PP4; ⁶⁰Co < ¹³⁴Cs) to a notably larger 12.5% (PP2; ⁶⁰Co < ¹³⁴Cs), with a mean difference of 6.5% (⁶⁰Co < ¹³⁴Cs; n = 4). One possible reason for the contrasting values recorded by PP2 may be due to spatially variable flow conditions at that location. However, if this value is treated as an outlier and excluded, the average difference between the two radionuclides falls to just 4.5% (⁶⁰Co < ¹³⁴Cs; n = 3).

	River Taw floodplain soil	River Culm floodplain soil
>63 µm (%)	14.6	20.8
<63 µm (%)	85.4	79.2
<2 µm (%)	6.8	4.7
Median D ₅₀ (µm)	22	28
Specific surface area (SSA) (m ⁻² g ⁻¹)	1.31	1.40
Bulk density (g cm ⁻³)	1.66	1.70

Table 6. Key characteristics associated with surface soil from the River Taw and River Culm floodplains.

	¹³⁴ Cs		⁶⁰ Co		% Difference
	Mass remaining (g m ⁻²)	% Remaining	Mass remaining (g m ⁻²)	% Remaining	
PP1	1191.93	86	1264.51	91.2	5.2
PP2	902.36	65.1	1075.92	77.6	12.5
PP3	1032.28	74.5	1140.39	82.3	7.8
PP4	1001.44	72.3	1009.64	72.8	0.5
Average	1032.00	74.5	1122.62	81.0	6.5

Table 7. Results from a paired-plot approach to determine differences in ⁶⁰Co and ¹³⁴Cs response under similar remobilisation conditions during one flood event.

3.2. Example 2: dispersed earthworm casts as a sediment-source

3.2.1. Experimental aim

The aim of this investigation was, again, to exploit the high sorption characteristics of ⁶⁰Co by testing its tracing capabilities on a biologically driven sediment detachment mechanism that would be particularly challenging for most other forms of tracer. The underlying rationale for this work was to confirm or refute the implication that fine sediment from disintegrating earthworm casts contributes to agricultural run-off. Previous attempts to quantify this process have met with only limited success [82–84]; however, due to the challenging nature of the erosion mechanism under investigation and of inherent difficulties of tracing objects that start as tangible 3-dimensional structures, but when weathered and disintegrated, lose their physicality by reverting back to fine sediment. Consequently, a prototype technique for labelling intact earthworm casts was designed in order to attempt to document the mobility of dispersed earthworm casts on vegetated hill slopes. More detailed background information on this particular tracing approach can be accessed in Greenwood [16]. Motivation for attempting a tracing investigation of this nature revolves around the fact that earthworms are prevalent in all edaphic environments, except deserts and permafrost regions. Anecic, or deep-burrowing, species are responsible for the vertical movement of soil due to a number of fundamental lifecycle traits namely burrowing and feeding. Although both activities are generally acknowledged to beneficially modify the soil and improve soil structure through mixing and incorporating surface organic matter into the upper soil horizon, a by-product of those activities is the production of earthworm casts on the soil surface (**Figure 6**). Researchers have thus speculated that the presence of surface casts not only represents an on-going soil detachment mechanism but also inadvertently renders casts vulnerable to weathering by impacting raindrops and leaves the resultant fine sediment susceptible to mobilisation by surface run-off. It is possible, therefore, that earthworm casts could represent a substantial sediment source. Given the ubiquity of anecic earthworms on vegetated hill slopes in temperate environments, along with their constant burrowing and feeding, it is likely that notable quantities of fine sediment and sediment-bound nutrients may remain in a constant state of flux, particularly on areas, where earthworms are particularly active and surface casts are abundant.



Figure 6. The on-going production of casts makes the sediment vulnerable to weathering and possible downslope mobilisation, even on relatively gentle slopes.

3.2.2. Method

The initial method for labelling intact earthworm casts with ^{60}Co was based on a technique originally reported by Toth and Alderfer [55] for labelling WSAs by immersion into a known activity of ^{60}Co mixed with water. Small groups of casts were air dried, weighed, and then immersed 'en masse' into a shallow tray containing a known solution of ^{60}Co mixed with a further 2 l of water. After immersion for a predetermined period of time (usually around 90 seconds), casts were removed, allowed to drip for ca. 30 s and then re-weighed (wet). The increase in mass (g) was multiplied by the original activity concentration (Bq ml^{-1}) of the radionuclide solution to derive an initial inventory for each group of casts. Labelled casts were then deployed on a 0.15 m^2 area of prepared grassland located at the hill slope divide of a ca. 10% slope (5.7°). Vegetation within the plot was trimmed, and each side of the plot was bounded using an aluminium sheet embedded into the soil surface to prevent run-on. A metal channel was embedded at the downslope end of the plot, flush with the soil surface, to guide any run-off and run-off-eroded sediment into an awaiting container located in a nearby reception pit (**Figure 7**). Any unlabelled casts within the prepared plot were removed, and 15 ^{60}Co -labelled casts, representing 190.7 g of sediment and with a group inventory of 224.3 Bq (equivalent to 1.18 Bq g^{-1}), were evenly spread across the upper part of the erosion plot. Casts were exposed to natural weather events over the following 76 days. Monitoring generally took place on a weekly basis, but the frequency of site visits was ultimately driven by the occurrence of rainfall events. During the operational phase, 186.3 mm of rainfall was recorded which generated 16 run-off samples. Sediment was separated from the run-off and



Figure 7. A number of ^{60}Co -labelled earthworm casts were spread across a bounded area of pasture and exposed to natural weather events for 76 days.

radiometrically assayed on a laboratory-grade gamma spectrometer, and a mass balance approach was used to partition the relative proportions of run-off-derived ^{60}Co -labelled sediment from any unlabelled sediment.

3.2.3. Key results and discussion

Data presented in **Table 8** show that of the 190.7 g of sediment originally deployed across the erosion plot in the form of 15 ^{60}Co -labelled earthworm casts, 26.82 g of the labelled sediment (equivalent to 14.1% of the original mass) was removed from the plot by surface run-off. In descriptive terms, the largest sample mass was 9.29 g and was recorded on day 5, the lowest sample mass was 0.14 g and was recorded on day 61 and the average mass based on all 16 samples was 1.68 g. The average mass for the first eight samples recovered from day 5–30 was 2.51 g, whereas the average mass for the latter eight samples recovered between days 32–76 was 0.85 g. **Figure 8** illustrates temporal variations in sediment recovery. Higher sediment recovery rates during the early stage of the investigation probably reflect higher sediment

	⁶⁰ Co (travel distance 0.1–0.3 m)	Unlabelled
Total initial sed. mass (g)	190.7	n/a
Total run-off removed (g)	26.82	59.51
% Removed	14.1	n/a
% Retained	85.9	n/a

Table 8. The proportion of ⁶⁰Co-labelled earthworm-derived sediment removed by surface run-off and the proportion retained.

availability. In contrast, lower recovery rates during the latter half of the experiment could either indicate the gradual exhaustion of the available (labelled) sediment supply, or it could indicate stabilisation and incorporation of the remaining labelled material back into the soil profile. Despite the gradual reduction in labelled sediment recovery, data seen in **Figure 8** clearly highlight that sediment from dispersed earthworm casts can act as a notable sediment source for at least a few months after initial production. Interestingly, unlabelled sediment, presumably generated after deployment of the labelled casts, was sporadically recovered on days 23 and 27 and then generally continuously recovered from day 38 onwards. If the unlabelled material comes from fresh earthworm casts, then the initial absence may reflect the fact that all unlabelled casts were removed from the plot when the investigation commenced. Similarly, the sporadic appearance of unlabelled sediment on days 23 and 27, followed by a generally sustained presence from day 38 onwards, may reflect a steady increase in unlabelled cast production by the resident earthworm population within the vicinity of the erosion plot.

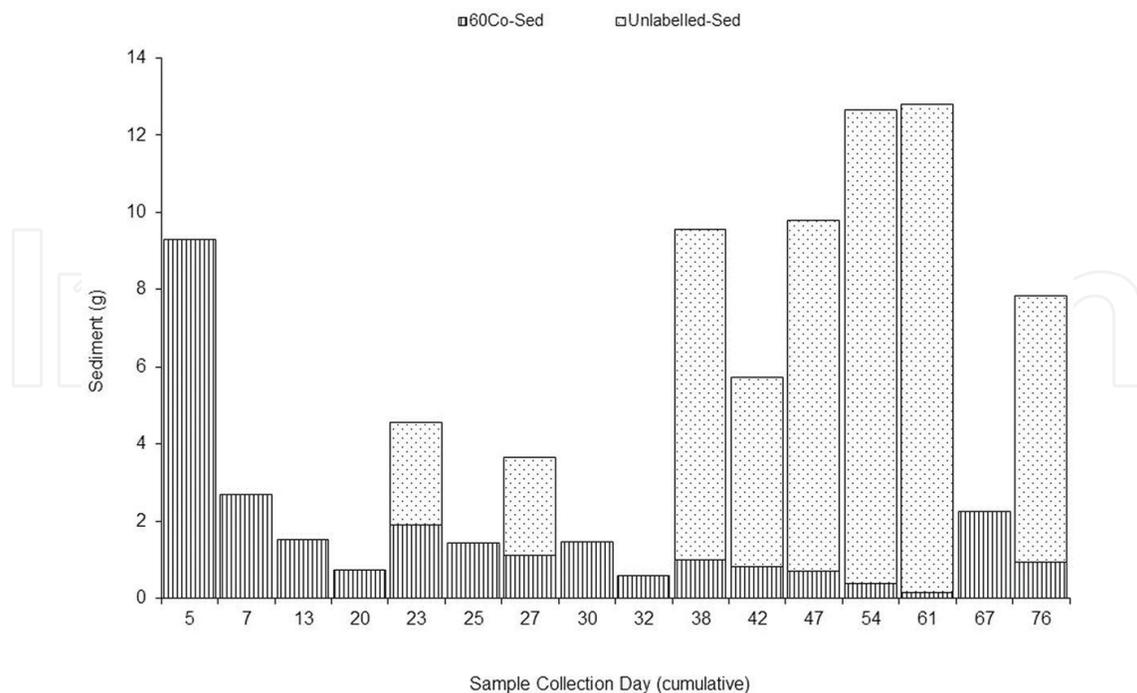


Figure 8. Results of an in-situ erosion experiment showing temporal variations in the amount of both ⁶⁰Co-labelled and unlabelled sediment recovered in surface run-off over the 76 day operational period.

3.2.4. Uncertainties

While the results presented above provide 'proof of concept' and support the idea that the tracing approach is generally sound and certainly worth developing, there are a number of issues and uncertainties that need to be addressed. One of the most pressing is the way in which initial inventories were calculated. Since a failure to determine an accurate initial inventory will generate erroneous data without an appropriate correction factor, initial refinements should focus on ways in which an initial group inventory can be established with more certainty. The original assumption predicted that dried, intact earthworm casts will sorb an amount of radionuclide that is proportional to the original concentration. Subsequent sorption tests were undertaken to test this hypothesis on small groups of earthworm casts using ^{60}Co , as well as ^{134}Cs , again, in order to assess and compare their respective sorption behaviours. A total of six aliquots were prepared, three containing ^{60}Co concentrations equivalent to 0.25, 0.5, and 1.0 Bq ml⁻¹, each in 400 ml of water, and three containing ^{134}Cs at similar concentrations and volumes. Earthworm casts were oven dried and then sorted into 18 groups, with each group consisting of five similar-sized casts, with an average weight of 39.8 g. The labelling procedure firstly involved measuring the radionuclide activity of a 100-ml sub-sample of radionuclide solution from each aliquot. After measuring, each sub-sample was then merged back into its respective aliquot, and the first group of casts was immersed into an aliquot for 4 min (hereafter referred to as Dip 1), removed and allowed to drain for ca. 30 s. A 100 ml sub-sample of the remaining liquid was, again, taken from each aliquot and analysed to determine the change in activity. This approach was repeated twice more (i.e. Dip 2 and Dip 3) until three groups of casts had been immersed into each aliquot. This refined approach follows the premise that the change (i.e. reduction) in inventory concentration after immersion of a group of casts must solely be due to the casts themselves. This technique is believed to represent a more accurate way of establishing a precise inventory for each group. Preliminary results from the refined sorption tests are shown in **Figures 9** and **10** for ^{60}Co and ^{134}Cs , respectively, and suggest that the original assumption may not necessarily be correct for contrasting reasons. By way of explanation, a group of casts recording zero along the Y-axis would signify sorption at an equivalent concentration as the original solution (i.e. no net loss or gain). Conversely, positive values along the Y-axis signify that a group of casts sorbed more than the available inventory concentration, whereas negative values signify that casts sorbed less than the available inventory concentration. For ^{60}Co , three groups of casts did indeed sorb more than the original concentration, whereas six groups sorbed notably less than the available inventory. For ^{134}Cs , one group sorbed slightly less than the available inventory concentration, whereas eight groups sorbed notably more than the available concentration. While these findings suggest sediment recovery data presented in **Figure 10** may be underestimated, by-products of the recent sorption tests are 18 groups of casts, each with a precise inventory. Consequently, scope exists to utilise those groups of casts experimentally by undertaking a series of replicate erosion experiments to further explore the potential of this particular tracing technique.

3.3. Example 3: livestock-poached pasture as a sediment source

3.3.1. Experimental aim

In the previous two examples presented, labelling was undertaken by exposing small quantities of fine-sediment to quantities of open-source ^{60}Co ; whereupon the resultant labelled material

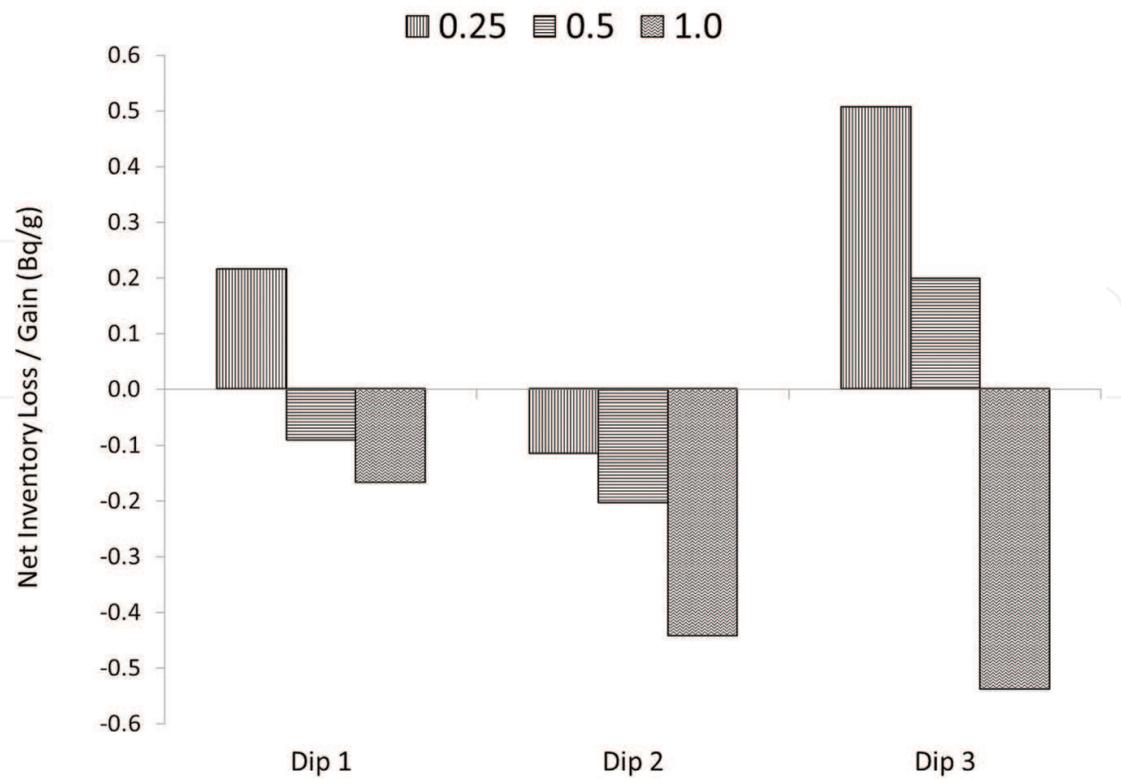


Figure 9. Results of sorption tests on small groups of intact earthworm casts by immersion into known activities of ^{60}Co , each mixed in 400 ml of water.

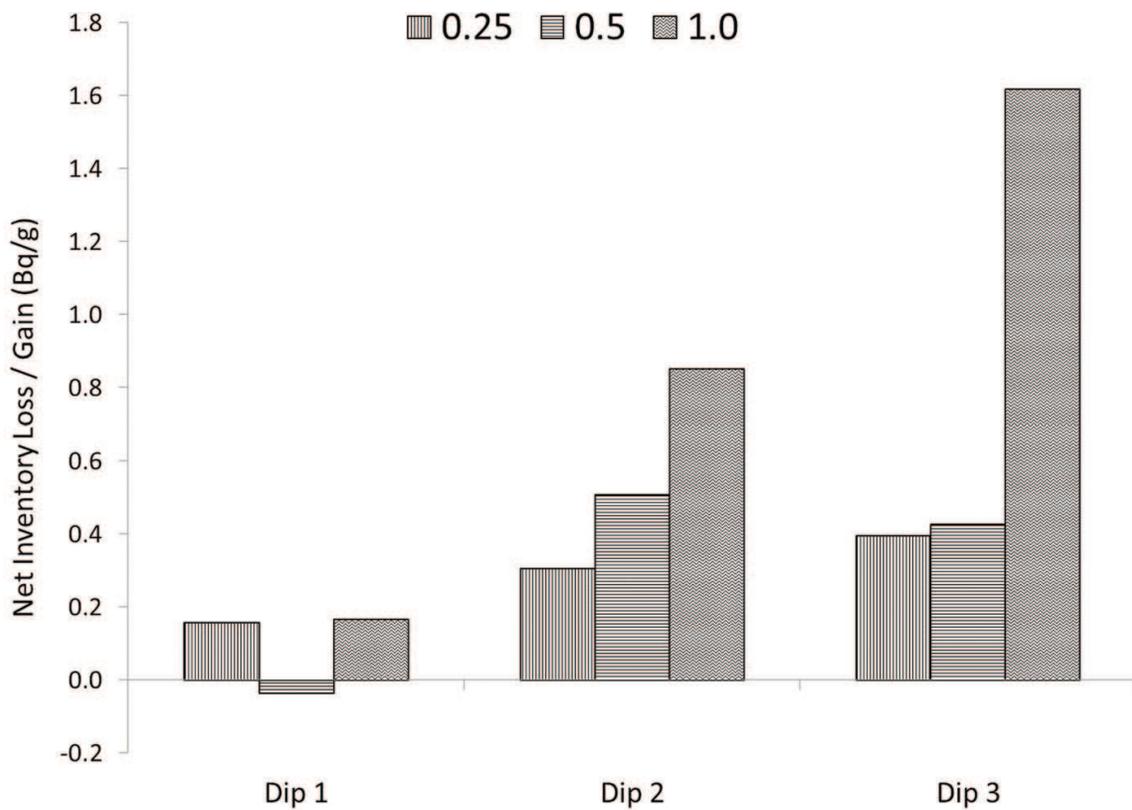


Figure 10. Results of sorption tests on small groups of intact earthworm casts by immersion into known activities of ^{134}Cs , each mixed in 400 ml of water.

was redeployed, albeit in different settings and under different erosion scenarios. In this final example, labelling was undertaken in-situ across six 0.2×0.2 m areas of grassland that had been heavily trampled and de-vegetated by cattle over the winter period, which left the soil with a slurry-like consistency, in a process known as livestock poaching (**Figure 11**). Again, more detailed background information on this particular tracing approach can be accessed in Greenwood [16] and in Greenwood et al. [58]. In brief, poaching is a common trait associated with domestic herd animals that frequently occur within confined areas of intensively managed grasslands, when the herd repeatedly congregates in certain areas. This can include field gateways, around feed-mangers and water-troughs, or along the banks of unfenced water-courses. If left unchecked, the repeated churning action of their hooves tramples and mixes the soil with any surface water. Vegetation cover is quickly depleted, and the soil can take on a slurry-like consistency with poor structure and limited cohesion. In this state, it is particularly vulnerable to redistribution by, for instance, impacting raindrops, and/or by surface run-off. Against this background, advances and recent developments in catchment-scale sediment fingerprinting techniques [17, 85] have demonstrated that intensively managed grasslands may contribute more fine sediment and sediment-associated nutrients and contaminants than originally presumed. Given legislation such as the WFD that aims to force EU member states to improve water quality in rivers and streams, a considerable number of studies have sought to identify potential sediment sources within intensively managed grasslands [10, 86, 87]. Resulting from those studies, poaching by livestock is been cited as a likely source of eroded sediment [38]. However, little work has been undertaken to quantify the process, again, due



Figure 11. Areas in fields where livestock repeatedly congregate, such as gate entrances, often become heavily poached; causing the surface soil to lose its cohesion and ability to resist erosion. In intensively managed grassland systems, areas such as this may act as sediment-sources.

to the dearth of available techniques capable of accurately documenting the movement of very small quantities of fine sediment at the required spatial and temporal scales [88]. Motivated by an urgent need to quantify this process and challenged by the complexities of the erosion mechanism and the very short timescales over which it can potentially operate, an in-situ method of labelling poached areas with ^{60}Co , for the purpose of documenting the extent to which sediment might be subsequently redistributed, was devised and executed.

3.3.2. Method

Small areas (0.04 m^2) of livestock-poached soil were each directly labelled with 1040 Bq of ^{60}Co activity mixed in 250 ml of water, using a correspondingly sized drip-type rainfall simulator, which was able to distribute an even supply of the radionuclide material over each area under controlled conditions. Nine areas were labelled within a 2.6 ha. field using this approach; three areas were established within the poached gateway seen in **Figure 11** on slopes ranging from 4.5 to 8% ($2.6\text{--}4.6^\circ$), three were established along the bank of a poached watercourse on slopes ranging from 6 to 21.3% ($3.4\text{--}12^\circ$), and three were established on a nearby flat, fully vegetated and un-poached area for use as controls. After labelling, each area was precisely marked and then allowed to rest for 24 h to allow the material to fully infiltrate and sorb to the soil before an initial surface inventory was established. Again, exploiting the high sorption capacity of ^{60}Co to fine sediment, a pilot study was undertaken 1 week before installation of the nine areas, which indicated that ca. 90% of the applied inventory remained within the upper 25–30 mm of an undisturbed soil profile. Initial surface inventories were established at 16 predetermined measurement points across each area using the in-situ gamma spectrometer previously seen in **Figure 3B**. An analytical protocol was devised to ensure that consistency was maintained throughout the measurement procedure, which coupled with undertaking measurements at the same 16 predetermined points, ensured the same ca. 86% of each area was measured every time. After installation, the nine areas were thus operational for a total of 65 days over the winter period, 2007. During this time, all nine areas were re-measured on three separate occasions after one or a series of natural rainfall events. Changes in surface inventories were determined relative to previous measurements, and these were used as a basis for inferring sediment redistribution across each poached area. On terminating the experiment, areas used as controls were shallow cored, sectioned in 3 mm intervals and assayed on a laboratory-grade gamma spectrometer to establish the vertical distribution of ^{60}Co within the soil profile. This information was then incorporated into a calibration procedure and used to convert changes in radiometric values to equivalent soil redistribution values.

3.3.3. Key results and discussion

Due to this investigation still being at a preliminary stage, data are presented in **Table 9** from one poached area only over the three re-measurement campaigns in order to provide proof-of-concept and to tentatively demonstrate the viability of the tracing technique. Radiometric values are decay and control plot corrected, and resultant erosion values are presented in $\text{kg m}^{-2} \text{ d}^{-1}$, with \pm uncertainty values calculated at the 95% confidence level. In instances where the surface inventory was less than the previous value, this is interpreted as evidence of

Re-measurement 1									
	Column 1		Column 2		Column 3		Column 4		Mean (kg m⁻² d⁻¹)
	Redist.	± Error							
	(kg m⁻² d⁻¹)		(kg m⁻² d⁻¹)		(kg m⁻² d⁻¹)		(kg m⁻² d⁻¹)		
Row 1	-0.88	0.15	-2.20	0.29	dep	n/a	-1.27	0.24	
Row 2	-0.14	0.03	-1.31	0.22	-1.10	0.14	dep	n/a	
Row 3	-0.36	0.06	-0.46	0.08	dep	n/a	-1.03	0.17	
Row 4	-2.14	0.41	-2.51	0.44	-1.68	0.30	dep	n/a	
Mean (kg m ⁻² d ⁻¹)	-1.26								
Re-measurement 2									
	Column 1		Column 2		Column 3		Column 4		Mean (kg m⁻² d⁻¹)
	Redist.	± Error							
	(kg m⁻² d⁻¹)		(kg m⁻² d⁻¹)		(kg m⁻² d⁻¹)		(kg m⁻² d⁻¹)		
Row 1	-1.15	0.27	-1.08	0.24	-1.15	0.20	-0.93	0.19	
Row 2	-1.33	0.24	-0.46	0.07	-1.38	0.21	dep	n/a	
Row 3	-0.95	0.28	-0.54	0.14	-1.17	0.33	-1.54	0.46	
Row 4	-1.23	0.26	-0.79	0.15	-2.25	0.61	dep	n/a	
Mean (kg m ⁻² d ⁻¹)	-1.14								
Re-measurement 3									
	Column 1		Column 2		Column 3		Column 4		Mean (kg m⁻² d⁻¹)
	Redist.	± Error							
	(kg m⁻² d⁻¹)		(kg m⁻² d⁻¹)		(kg m⁻² d⁻¹)		(kg m⁻² d⁻¹)		
Row 1	-1.27	0.34	-0.83	0.24	-0.69	0.18	-0.58	0.12	
Row 2	-1.65	0.29	-0.61	0.11	-1.01	0.24	dep	n/a	
Row 3	-1.62	0.42	-1.49	0.36	-0.99	0.28	-0.99	0.27	
Row 4	-1.14	0.22	-1.00	0.18	-1.75	0.41	dep	n/a	
Mean (kg m ⁻² d ⁻¹)	-1.11								

Table 9. Soil redistribution values measured over three separate re-measurement campaigns spanning 65 days.

the removal of labelled sediment at that particular location. In contrast, where the surface inventory was greater than the previous value, this is interpreted as evidence of the deposition of labelled sediment at that location. Due to the complexities of calculating deposition, however, no data are currently presented for those points. Despite their absence, however, evidence of deposition was only recorded at four points during the first re-measurement

and at two points each during the second and third re-measurements. Consequently, the predominant redistribution mechanism over the three re-measurement campaigns, certainly at this particular poached area, was clearly erosion; signifying a net loss of material from within the confines of the monitored area during the 65 days the experiment was operational. Average erosion values over the three re-measurement campaigns were 1.26, 1.14 and 1.11 kg m⁻² d⁻¹, respectively. The stepwise reduction in net soil loss over the three re-measurement campaigns is thought to reflect the poached soil gradually drying, regaining its structure and offering increasing resistance against redistribution processes as weather conditions improved. In addition to this, the gradual regrowth and recovery of grass probably exerted an increasingly stabilising influence on the soil over time. These very tentative yet encouraging findings, albeit from one location only, support the original hypothesis that livestock-poached areas can act as a sediment source in intensively-managed grassland systems. It is worth emphasising, however, that as poaching more often occurs as isolated islands surrounded by a sea of dense grassland, it is highly likely that any eroded material would be trapped by the surrounding grass and probably quickly reincorporated back into the soil matrix. However, on poached areas lying immediately adjacent, or close to watercourses, it is possible that the eroded material could migrate into the watercourse and contribute to reducing water quality downstream of its entry point.

3.3.4. *Uncertainties*

Soil redistribution depths were determined using one of many available calibration procedures, each of which converts inventory values to equivalent soil redistribution values in a slightly different way. Depending on which calibration procedure is chosen, therefore, redistribution rates may vary, even when based on the same radiometric data. As ⁶⁰Co is still largely untried and untested as a fine-sediment tracer, and the tracing approach is not only novel but also is still in its infancy, a logical way of off-setting at least some of the uncertainty associated with the resultant redistribution data would be to process the same radiometric data using two different calibration models. Such an approach would permit data sets to be compared and provide an indication of the degree of uncertainty involved [89].

4. Problems with using Cobalt-60

Despite the numerous benefits and advantages that have been highlighted in previous sections regarding open-source ⁶⁰Co as a fine-sediment tracer, during the course of undertaking these, and other, sediment tracing investigations using radioactive ⁶⁰Co, a number of problems were encountered. One of the most prominent was initially locating a supplier of open-source radioactive material and thereafter obtaining/ensuring that the necessary licences and permits were in-place to enable the radioactive material to be purchased, stored, used and, where applicable, disposed of. Also important was the availability of trained personnel qualified to partition and dispense stock material at activities suitable to undertake experimental work. Also paramount when attempting to develop novel tracing techniques is to have ready access to highly specialised, and expensive, in-situ and laboratory-based gamma spectrometers so

that analyses can be performed under a range of environments, sometimes more than once. Also problematic was finding sympathetic landowners or farm managers that were willing to allow work to be conducted on their land, largely due to the stigma associated with radioactivity. This particular issue admirably illustrates the contentiousness associated with radiation *per se*, despite it being very low level and especially with the deliberate, and arguably contentious, release of such material into the environment, particularly for the purpose of studying what many members of the general public would probably view as a trivial subject with limited real-world benefits.

5. Key refinements and prospects for future work

In terms of key refinements and prospects for future work, with regard to Example 1, rather than using intuition and experience to select sites on which to install active plots, more representative spatial coverage could be obtained by adopting a more focused approach where, for instance, plots are installed using an equidistant grid system, with spacings that are appropriate to the size of the floodplain under scrutiny. In addition, a progressive reduction in the analytical precision of most active plots was noted over the second, but particularly over the third, inundation event. This was attributed to diminishing radiometric signals emitted by the finite supply of labelled sediment as supply gradually became exhausted, and/or high uncertainties due to the necessarily short counting time (600 s) devoted to each analysis. Both effects could be mitigated in several ways. One way is to increase the amount of labelled sediment and associated inventory deposited at each plot. Alternatively, a simpler, somewhat less contentious and more obvious way is to extend the counting time per assay. While this would initially appear to be a good alternative, since flooding occurs almost exclusively in the autumn/winter period, a time when natural daylight hours are around 6–7 h only, this would necessarily extend the total analytical time from 3 days to probably in excess of 4 days per floodplain site (i.e. >8 days in total). Undertaking analyses over timescales such as this and taking into account the time needed for floodwaters to recede, there exists a good chance that multiple floods could occur in quick succession before further analyses could be performed, thereby resulting in a loss of information on the extent and magnitude of remobilisation over single flood events.

With regard to Example 2, in light of the refined labelling approach and presumed increased precision, just a flavour of the possible tests that could be undertaken in future might include using simulated rainfall and erosion plots lined with turf to simulate permanent pasture. Such an approach could offer a convenient and precise way of looking at key controlling factors, such as slope and vegetation density, and the degree to which they influence rates of sediment recovery and the travel distance of the eroded material. Further work could also be undertaken on simulated cultivated soil; again, using artificial rainfall and erosion plots; but in this instance, to not only determine travel distance of the dispersed sediment from earthworm casts on unconsolidated soils, but perhaps also study the rate of removal from crusted soils. While the use of both artificial rain and erosion plots is a convenient way of undertaking experiments, neither approach can substitute for the effect that time has on weathering and

erosion, however. Consequently, it could also be worth deploying groups of casts in situ on natural pasture and use natural rainfall as the principle weathering agent, in a similar manner to that described earlier and monitor over several months or until the radiometric signal associated with run-off-recovered sediment falls to below detection levels. As a further addendum to this basic idea, it might also be worthwhile installing in-situ plots, bounded and spaced relatively close to each other, but each with a progressively longer distance from the area of deployment to the channel outlet in order to explore the relationship that space and time has on the travel distance of eroded casts.

With regard to Example 3, in light of the tentative yet encouraging results presented earlier, a number of changes could be made that could reduce the uncertainty. Firstly, rather than installing areas based on observations and intuition, a denser network of systematically placed plots across the full extent of each poached area would generate more representative spatial information. As for the same reasons above, analytical counting times were deliberately kept short due to lack of daylight hours when the experiment was done. While extending them would reduce the uncertainty associated with individual redistribution estimates, it would also mean that total analytical times would be commensurately longer. Consequently, a trade-off would need to be made between longer count times, resulting in increased accuracy and completing the analyses in a realistic period of time. One additional refinement to the existing technique that would be adopted in future would be to label poached areas and ascertain initial surface inventories, but re-measure once only and then abandon, in favour of installing new sites on different poached areas elsewhere. Reasons for this seemingly drastic approach relate to the sensitivity of the mathematical conversion procedure, and the fact that its ability to convert changes in surface activities to equivalent soil redistribution depends on the distribution of the radionuclide declining exponentially with depth. While this relationship remained valid immediately after labelling, the progressive redistribution of labelled surface material, along with the likely ingress and mixing of unlabelled sediment from elsewhere, would probably cause the exponential relationship to steadily break down. Although the timescale over which this would occur are unknown, the end result would be increasingly erroneous data.

6. Concluding remarks

The chapter has described and reviewed three prototype sediment tracing approaches using the artificial gamma-emitting radionuclide ^{60}Co , each of which was designed to measure the redistribution, remobilisation or dispersal of fine-sediment under a different scenario. The first example looked at the fate of flood-deposited sediment on river floodplains and found that the material was very susceptible to remobilisation during the first flood event immediately after deposition. However, it also found that a significant proportion was still mobile for up to 2 months after deposition, after which, whatever remained was incorporated into the bulk soil profile and stored. The second example presented in the chapter sought to determine whether dispersed earthworm casts act as a sediment source on vegetated hill slopes. The study found that sediment from dispersed casts did indeed act as a sediment source, and

importantly, it also remained mobile for at least a few months after initial production. The final example sought to determine the extent to which areas of livestock-poached pasture were subjected to redistribution processes. The study found that such areas did indeed experience sediment redistribution and that erosion, or a net loss of fine-sediment from poached areas, was the predominant process. The above findings, although preliminary, are not only sufficiently encouraging to warrant further refining each approach using ^{60}Co but also collectively represent essentially unique information that contributes to an improved understanding of some of the widespread yet largely under-studied sediment mobilisation and transfer mechanisms that commonly operate within catchments. While some limitations and issues were duly noted with ^{60}Co , most of those could be readily circumvented with sufficient advanced planning. Indeed, it is therefore difficult to imagine how information gained from the three experimental examples presented above could be obtained using the more common tracers reviewed earlier in the chapter. Based on these findings, the versatility of ^{60}Co as a fine-sediment tracer has been confirmed, and its efficacy to provide accurate erosion data in challenging settings has undoubtedly expanded the overall scope and diversity of sedimentation scenarios that can currently be investigated. With additional work to explore and further develop the capabilities of ^{60}Co , it might eventually be added to the existing suite of tracers and tracing techniques currently available to researchers studying sedimentation processes.

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References

- [1] Barrow CJ. Environmental Management: Principles and Practise. London: Routledge; 2000

- [2] Zapata F. The use of environmental radionuclides as tracers in soil erosion and sedimentation investigations: Recent advances and future developments. *Soil & Tillage Research*. 2003;**69**:3-13
- [3] Walling DE. The changing sediment loads of the world's rivers. In: *Sediment Dynamics in Changing Environments*. International Association of Hydrological Sciences (IAHS), Publication No. 325. 2008. p. 323-338
- [4] Walling DE, Quine TA. The use of caesium-137 measurements in soil erosion surveys. In: *Erosion and Sediment Transport Monitoring Programmes in River Basins*. International Association of Hydrological Sciences (IAHS), Publication No. 210. 1992. p. 143-152
- [5] El-Swaify SA, Flach KW. Foreword. In: Lal R, editor. *Soil Erosion Research Methods*. Iowa: USA: Soil and Water Conservation Society; 1988
- [6] Brown LR. Conserving soils. In: Brown LR, editor. *State of the World*. Vol. 53. New York: Norton; 1984, 1984. p. 53-75
- [7] Imeson A. *Desertification, Land Degradation and Sustainability: Paradigms, Processes, Principles and Policies*. UK: Wiley-Blackwell Publishing; 2012
- [8] Cherlet M, Ivits E. Compilation of a new atlas of desertification and contribution to a global assessment. In: de Boever M, Khlosi M, Delbecque N, de Pue J, Ryken N, Verdoodt A, Cornelis W, Gabriels D, editors. *Desertification and Land Degradation: Processes and Mitigation*. Ghent University, Belgium: UNESCO Chair of Eremology; 2013
- [9] Gibbs HK, Salmon JM. Mapping the world's degraded lands. *Applied Geography*. 2015;**57**:12-21
- [10] Evans R, Collins AL, Zhang Y, Foster IDL, Boardnam J, Sint H, Lee MRF. A comparison of conventional and ¹³⁷Cs-based estimates of soil erosion rates on arable and grassland across lowland England and Wales. *Earth-Science Reviews*. 2017;**173**:49-64
- [11] Walling DE, Collins AL. The catchment sediment budget as a management tool. *Environmental Science & Policy*. 2008;**11**:136-143
- [12] Pulley S, Foster I, Antunes P. The application of sediment fingerprinting to floodplain and lake sediment cores: Assumptions and uncertainties evaluated through case studies in the Nene Basin, UK. *Journal of Soils and Sediments*. 2015;**15**(10):2132-2154
- [13] Europa 2017. The EU Water Framework Directive <http://ec.europa.eu/environment/water/water-framework/info/timetableen.htm> [Accessed 25/07/17]
- [14] Pulley S, Van der Waal B, Collins AL, Foster IDL, Rowntree K. Are source groups always appropriate when sediment fingerprinting? The direct comparison of source and sediment samples as a methodological step. *River Research and Applications*. 2017:1-11
- [15] Walling DE. Tracing versus monitoring: New challenges and opportunities in erosion and sediment delivery research, Chpt. 2. In: Owens PN, Collins AJ, editors. *Soil Erosion and Sediment Redistribution in River Catchments: Measuring, Monitoring and Management*. Oxfordshire, UK: CABI Publishing; 2006

- [16] Greenwood P. Applications of active tracer techniques in soil erosion and catchment sediment source investigations [unpublished PhD thesis]. University of Exeter, UK: Department of Geography; 2010 https://www.researchgate.net/publication/303494922_Applications_of_Active_Tracer_Techniques_in_Soil_Erosion_and_Catchment_Sediment_Source_Investigations
- [17] Collins AL, Pulley S, Foster IDL, Gellis A, Porto P, Horowitz AJ. Sediment source fingerprinting as an aid to catchment management: A review of the current state of knowledge and a methodological decision-tree for end-users. *Journal of Environmental Management*. 2016;**194**:86-108
- [18] Nosrati K. Ascribing soil erosion of hillslope components to river sediment yield. *Journal of Environmental Management*. 2017;**194**:63-72
- [19] Evans GV. Tracer techniques in hydrology. *The International Journal of Applied Radiation and Isotopes*. 1983;**34**:451-475
- [20] Sauzay G. Tracer techniques in sediment transport: Report of the panel. In: *Tracer Techniques in Sediment Transport*. Technical Report Series No. 145. Vienna, Austria: International Atomic Energy Agency (IAEA); 1973. p. 3-8
- [21] Foster IDL. *Tracers in Geomorphology*. UK: John Wiley & Sons Ltd.; 2000
- [22] Heckrath G, Halekoh U, Djurhuus J, Govers G. The effect of tillage direction on soil redistribution by mouldboard ploughing on complex slopes. *Soil & Tillage Research*. 2006;**88**:225-241
- [23] Spokas K, Forcella F, Archer D, Reicosky D. SeedChaser: Vertical soil tillage distribution model. *Computers and Electronics in Agriculture*. 2007;**57**:62-73
- [24] Tiessen KHD, Mehuys GR, Lobb DA, Rees HW. Tillage erosion within potato production systems in Atlantic Canada I. Measurement of tillage translocation by implements used in seedbed preparation. *Soil & Tillage Research*. 2007;**95**:308-319
- [25] Lobb DA, Kachanoski RG. Quantification of tillage translocation and tillage erosion. *Canadian Journal of Soil Science*. 1994;**74**:353
- [26] Liu G, Hai X, Liu P, Zhang J. Using rare earth elements to monitor sediment sources from a miniature model of a small watershed in the three gorges area of China. *Catena*. 2016a;**142**:114-122
- [27] Liu G, Hai X, Liu P, Zhang Q, Zhang J. An improved method for tracing soil erosion using rare earth elements. *Journal of Soils and Sediments*. 2016b;**16**(5):1670-1679
- [28] Zhang XC, Friedrich JM, Nearing MA, Norton LD. Potential use of rare earth oxides as tracers for soil erosion and aggregation studies. *Soil Science Society of America Journal*. 2001;**65**:1508-1515
- [29] Lang A. Recent advances in dating and source tracing of fluvial deposits. In: *Sediment Dynamics in Changing Environments*. International Association of Hydrological Sciences (IAHS), Publication No. 325. 2008. p. 3-12

- [30] Fullen MA. Laboratory and field studies in the use of the isotope ^{59}Fe for tracing soil particle movement. *Earth Surface Processes and Landforms*. 1982;**7**:285-293
- [31] Wooldridge DD. Tracing soil particle movement with Fe-59. *Soil Science Society of America Journal*. 1965;**29**:469-472
- [32] Ventura E, Nearing M, Norton LD. Developing a magnetic tracer to study soil erosion. *Catena*. 2001;**43**:277-291
- [33] Parsons AJ, Wainwright J, Abrahams AD. Tracing sediment movement in interrill overland flow on a semi-arid grassland hillslope using magnetic susceptibility. *Earth Surface Processes and Landforms*. 1993;**18**:721-732
- [34] Plante AF, Duke MJM, McGill WB. A tracer detectable by neutron activation for soil aggregation and translocation studies. *Soil Science Society of America Journal*. 1999;**63**:1284-1290
- [35] Riebe B. Monitoring the translocation of soil particles using a neutron activated tracer. In: Hartge KH, Stewart BA, editors. *Soil Structure: Its Development and Function*. Florida, USA: CRC Press Inc.; 1995. p. 277-294
- [36] Walling DE. Quantifying the fine sediment budgets of river basins. *National Hydrology Seminar*. 2004:9-20
- [37] Campbell BL, Loughran RJ, Elliott GL. A method for determining sediment budgets using caesium-137. In: *Sediment Budgets*. International Association of Hydrological Sciences (IAHS), Publication No. 174. 1988. p. 171-179
- [38] Greenwood P. Tracing Fine-Sediment Using Artificial Radionuclides. *British Society for Geomorphology Publication*; 2012 http://britishgeomorphology.org.uk/sites/default/files/geom_tech_chapters/3.5.2_SedimentTracing.pdf
- [39] Greenwood P, Walling DE, Quine TA. Assessing the remobilization of recently deposited sediment from river floodplains during single overbank flood events, using caesium-134 and cobalt-60 as tracers. *International Association of Hydrological Sciences Publication, Number 325*. 2008:13-23
- [40] Greenwood P, Walling DE, Quine TA. Using caesium-134 and cobalt-60 as tracers to assess the remobilization of recently-deposited overbank-derived sediment on river floodplains during subsequent inundation events. *Earth Surface Processes and Landforms*. 2013;**39**(2):228-244
- [41] Wallbrink PJ, Olley JM, Hancock G. Estimating residence times of fine sediment in river channels using fallout ^{210}Pb . In: *The Structure, Function and Management Implications of Fluvial Sedimentary Systems*. International Association of Hydrological Sciences (IAHS), Publication No. 276. 2002a. p. 425-432
- [42] Walling DE, Owens PN. The role of flood plain sedimentation in catchment sediment and contaminant budgets. In: *The Structure, Function and Management Implications of Fluvial Sedimentary Systems*. International Association of Hydrological Sciences (IAHS), Publication No. 276. 2002. p. 407-416

- [43] He Q, Walling DE. Interpreting particle size effects in the adsorption of ^{137}Cs and unsupported ^{210}Pb by mineral soils and sediments. *Journal of Environmental Radioactivity*. 1996;**30**:117-137
- [44] Walling DE, He Q. Rates of overbank sedimentation on the floodplains of several British rivers during the past 110 years. In: *Variability in Stream Erosion and Sediment Transport*. International Association of Hydrological Sciences (IAHS) Publication, No. 224. 1994. p. 77-86
- [45] He Q, Walling DE, Wallbrink PJ. Alternative methods and radionuclides for use in soil-erosion and sedimentation investigations. Chpt. 9. In: Zapata F, editor. (2002) *Handbook for the Assessment of Soil Erosion and Sedimentation Using Environmental Radionuclides*. London: Kluwer Academic Publishers; 2002. p. 185-215
- [46] Blake WH, Walling DE, He Q. Fallout beryllium-7 as a tracer in soil erosion investigations. *Applied Radiation and Isotopes*. 1999;**51**:599-605
- [47] Blake WH, Walling DE, He Q. Using cosmogenic Beryllium-7 as a tracer in sediment budget investigations. *Geografiska Annaler A*. 2002;**84**:89-102
- [48] Mabit L, Benmansour M, Walling DE. Comparative advantages and limitations of the fallout radionuclides ^{137}Cs , $^{210}\text{Pb}_{\text{ex}}$ and ^7Be for assessing soil erosion and sedimentation. *Journal of Environmental Radioactivity*. 2008;**99**:1799-1807
- [49] Walling DE, Schuller P, Zhang Y, Iroumé A. Extending the timescale for using beryllium-7 measurements to document soil redistribution by erosion. *Water Resources Research*. 2009;**45**:W02418
- [50] McCubbin D, Leonard KS. Use of radiotracers for studies of metal sorption behaviour. *Science of the Total Environment*. 1995;**173/174**:259-266
- [51] Showler AT, Knaus RM, Reagan TE. The versatility of radiotracer methods for studying insect ethology and ecology. *Florida Entomologist*. 1988;**71**(4):554-580
- [52] Kinnell PIA. The effect of flow depth on sediment transport induced by raindrops impacting shallow flows. *American Society of Agricultural Engineers*. 1991;**34**(1):161-168
- [53] Quine TA, Govers G, Poesen J, Walling DE, van Wesemael B, Martinez-Fernandez J. Fine-earth translocation by tillage in stony soils in the Guadalentin, south-east Spain: An investigation using caesium-134. *Soil & Tillage Research*. 1999;**51**:279-301
- [54] Syversen N, Øygarden L, Salbu B. Cesium-134 as a tracer to study particle transport processes within a small catchment with a buffer zone. *Journal of Environmental Quality*. 2001;**30**:1771-1783
- [55] Toth SJ, Alderfer RB. A procedure for tagging water-stable aggregates with Co-60. *Soil Science*. 1959a;**89**:36-37
- [56] Toth SJ, Alderfer RB. Formation and breakdown of Co-60-tagged water-stable aggregates in a Norton Silt Loam Soil. *Soil Science*. 1959b;**90**:232-238

- [57] Ridley N. The Radioactive Substances (Substances of Low Activity) Exemption Order 1986. Statutory Instruments, No 1002. Atomic Energy and Radioactivity Substances. 1986. http://www.ionactive.co.uk/pdfs/Substances_Low_Activity_Exemption_Order_1119018891.pdf [Accessed 25/07/17]
- [58] Greenwood P, Walling DE, Quine TA. Accepted. Documenting sediment redistribution on livestock-poached pasture using the artificial radionuclides caesium-134 and cobalt-60 as tracers. Land degradation and development. Special Issue: The impact of grazing on land degradation: A common problem with many driving forces
- [59] Khan SA. Sorption of the long-lived radionuclides cesium-134, strontium-85 and cobalt-60 on bentonite. *Journal of Radioanalytical and Nuclear Chemistry*. 2003;**258**:3-6
- [60] Environmental Protection Agency (EPA). Radiation Protection: Cobalt. USA: Environmental Protection Agency; 2002 www.epa.gov/rpdweb00/radionuclides/cobalt.html [Accessed 25/07/17]
- [61] Bailly du Bois P. Mapping of water masses in the North Sea using radioactive tracers. *Endeavour*. 1996;**20**:2-7
- [62] Cundy AB, Croudace IW. Sediment accretion and recent sea-level rise in the Solent, Southern England: Inferences from radiometric and geochemical studies. *Estuarine, Coastal and Shelf Science*. 1996;**43**:449-467
- [63] Matishov GG, Matishov DG, Namjatov AA, Carroll JL, Dahle S. Anthropogenic radionuclides in Kola and Motovsky Bays of the Barents Sea, Russia. *Journal of Environmental Radioactivity*. 1999;**43**:77-88
- [64] Bailly du Bois P, Guéguéniat P. Quantitative assessment of dissolved radiotracers in the English Channel: Sources, average impact of la Hague reprocessing plant and conservative behaviour (1983, 1986, 1988, 1994). *Continental Shelf Research*. 1999;**19**:1977-2002
- [65] Thompson J, Dyer FM, Croudace IM. Records of radionuclide deposition in two salt marshes in the United Kingdom with contrasting redox and accumulation conditions. *Geochimica et Cosmochimica Acta*. 2002;**66**(6):1011-1023
- [66] Sutherland RA. Spatial variability of ^{137}Cs and the influence of sampling on estimates of sediment redistribution. *Catena*. 1994;**21**:57-71
- [67] Caron F, Mankarios G. Pre-assessment of the speciation of ^{60}Co , ^{125}Sb , ^{137}Cs and ^{241}Am in a contaminated aquifer. *Journal of Environmental Radioactivity*. 2004;**77**:29-46
- [68] Payne TE, Itakura T, Comarmond MJ, Harrison JJ. Environmental mobility of cobalt – Influence of solid phase characteristics and groundwater chemistry. *Applied Radiation and Isotopes*. 2009;**67**:1269-1276
- [69] Shinonaga T, Schimmack W, Gerzabek MH. Vertical migration of ^{60}Co , ^{137}Cs and ^{226}Ra in agricultural soils as observed in lysimeters under crop rotation. *Journal of Environmental Radioactivity*. 2005;**79**:93-106

- [70] Environmental Protection Agency (EPA). Understanding Variation in Partition Coefficient K_d Values – Attachment C: Radiological Properties for SSL Development. USA: Office of Air & Radiation, Environmental Protection Agency; 1999
- [71] Chen L, Lu S. Sorption and desorption of radiocobalt on montmorillonite—Effects of pH, ionic strength and fulvic acid. *Applied Radiation and Isotopes*. 2008;**66**:288-294
- [72] Sparks DL. *Environmental Soil Chemistry*. 2nd ed. Amsterdam, The Netherlands: Academic Press, Elsevier Publishers Ltd.; 2003
- [73] Shor JT, Dial C. Volatilization of cobalt, technetium, and uranium isotopes from soils through salt amendment and calcination. *Journal of Environmental Radioactivity*. 2000;**48**:35-48
- [74] Capowiez Y, Renault P, Belzunces L. Three-dimensional trajectories of ^{60}Co -labelled earthworms in artificial cores of soil. *European Journal of Soil Science*. 2001;**52**:365-375
- [75] Sattar A, Traniello JFA, Salihah Z, Farid A. Determining different parameters of Co-60 bait as a radiotracer for three species of subterranean termites. *The Nucleus*. 2002;**39**:89-93
- [76] Wallbrink PJ, Walling DE, He Q. Radionuclide measurement using HPGe gamma spectrometry. Cp. 5. In: Zapata F, editor. (2002) *Handbook for the Assessment of Soil Erosion and Sedimentation Using Environmental Radionuclides*. London: Kluwer Academic Publishers; 2002b. p. 67-96
- [77] Pennock DJ, Appleby PG. Site selection and sampling design, Chpt. 2. In: Zapata F, editor. *Handbook for the Assessment of Soil Erosion and Sedimentation Using Environmental Radionuclides*. London, UK: Kluwer Academic Publishers; 2002. p. 15-40
- [78] Owens PN, Walling DE, Leeks GJL. Deposition and storage of fine-grained sediment within the main channel system of the River Tweed, Scotland. *Earth Surface Processes and Landforms*. 1999;**24**:1061-1076
- [79] Walling DE, Owens PN, Carter J, Leeks GJL, Lewis S, Meharg AA, Wright J. Storage of sediment-associated nutrients and contaminants in river channel and floodplain systems. *Applied Geochemistry*. 2003;**18**:195-220
- [80] Nicholas AP, Walling DE, Sweet RJ, Fang X. New strategies for upscaling high-resolution flow and overbank sedimentation models to quantify floodplain sediment storage at the catchment scale. *Journal of Hydrology*. 2006;**329**:577-594
- [81] Simm DJ, Walling DE. Lateral variability of overbank sedimentation on a Devon floodplain. *Hydrological Sciences*. 1998;**43**:715-732
- [82] Le Bayon RC, Binet F. Rainfall effects on erosion of earthworm casts and phosphorous transfers by water runoff. *Biological Fertility of Soils*. 1999;**30**:7-13
- [83] Le Bayon RC, Binet F. Earthworm surface casts affect soil erosion by run-off and phosphorous transfer in a temperate maize crop. *Pedobiologia*. 2001;**45**:430-442

- [84] Le Bayon RC, Moreau S, Gascuel-Oudoux C, Binet F. Annual variations in earthworm surface-casting activity and soil transport by water run-off under a temperate maize agroecosystem. *Geoderma*. 2002;**106**:121-135
- [85] Collins AL, Walling DE, Leeks GJL. Source type ascription for fluvial suspended sediment based on a quantitative composite fingerprinting technique. *Catena*. 1997;**29**:1-27
- [86] Bilotta GS, Brazier RE, Haygarth PM, MacLeod CJA, Butler P, Granger S, Krueger T, Freer J, Quinton J. Rethinking the contribution of drained and undrained grasslands to sediment-related water quality problems. *Journal of Environmental Quality*. 2008;**37**:906-914
- [87] Orr RJ, Murray PJ, Eyles CJ, Blackwell MSA, Cardenas LM, Collins AJ, Dungait JAJ, Goulding KWT, Griffith BA, Gurr SJ, Harris P, Hawkins JMB, Misselbrook TH, Rawlings C, Shepard A, Sint H, Takahashi T, Tozer KN, Whitmore AP, Wu L, Lee MRF. The North Wyke Farm Platform: Effect of temperate grassland farming systems on soil moisture contents, runoff and associated water quality dynamics. *European Journal of Soil Science*. 2016;**67**:374-385
- [88] Granger SJ, Bol R, Butler PJ, Haygarth PM, Naden P, Old G, Owens PN, Smith BPG. Processes affecting transfer of sediment and colloids, with associated phosphorous, from intensively farmed grasslands: Tracing and organic matter. *Hydrological Processes*. 2007;**21**:417-422
- [89] Walling DE, He Q, Appleby PG. Conversion models for use in soil-erosion, soil-redistribution and sedimentation investigations. Chpt. 7. In: *Handbook for the Assessment of Soil Erosion and Sedimentation Using Environmental Radionuclides*, F Zapata. London, UK: Kluwer Academic Publishers; 2002. p. 111-164
- [90] Alam SM, Ansari R, Khan A. Application of radioisotopes and radiation in the field of agriculture: Review. *Journal of Biological Sciences*. 2001;**1**(3):82-86
- [91] Oldeman LR. *Soil Degradation: A Threat to Food Security?* Report 98/01. International Soil Reference and Information Centre: Wageningen; 1998
- [92] Ritchie JC, McHenry JR. Application of radioactive fallout cesium-137 for measuring soil erosion and sediment accumulation rates and patterns: A review. *Journal of Environmental Quality*. 1990;**19**:215-233
- [93] Schreiner LJ, Joshi CP, Darko J, Kerr A, Salomons G, Dhanesar S. The role of cobalt-60 in modern radiation therapy: Dose, delivery and image guidance. *Journal of Medical Physics*. 2009;**34**(3):133-136
- [94] Thorén M, Rahn T, Guo WY, Werner S. Stereotactic radiosurgery with the cobalt-60 gamma unit in the treatment of growth hormone-producing pituitary tumors. *Neurosurgery*. 1991;**29**:663-668

