Using environmental radionuclides as tracers in sediment budget investigations

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INTRODUCTION

The use of fallout or environmental radionuclides to trace and quantify the mobilisation, transfer and deposition of sediment in catchments can be traced back to the 1960s and to work such as that reported by Rogowski & Tamura (1965). In this early work, attention focussed on the mobility and fate of radioactive fallout from weapons testing from a health or radioecological perspective. Such work demonstrated that radionuclides such as caesium-137 (^{137}Cs) were rapidly and firmly fixed by the surface soil and that their subsequent mobilisation and transfer were effectively controlled by the erosion and transport of soil and sediment particles. The potential to exploit this feature of radionuclide behaviour to trace and quantify sediment movement was soon recognised by workers such as Ritchie and McHenry and their co-workers (e.g. Ritchie et al., 1970, 1973) who successfully used ^{137}Cs to estimate rates of soil loss from agricultural land and sedimentation rates in lakes, reservoirs and wetlands. Since that early work, ^{137}Cs has been successfully used in soil erosion studies in many areas of the world. The approach has also been extended to include other radionuclides, such as beryllium-7 (^{7}Be) and unsupported or excess lead-210 (^{210}Pb$_{ex}$) and to embrace various sediment budget components including, for example, floodplain sedimentation. Such work has increasingly demonstrated the potential for using fallout radionuclides as an essentially unique tool in sediment budget investigations.

THE BASIS

The fundamental basis for using environmental radionuclides in sediment budget investigations is that over a relatively small area their fallout input, which will be largely associated with rainfall, can be assumed to be effectively uniform. In most environments, this fallout is rapidly and firmly fixed by the surface soil and subsequent redistribution of the fallout input will therefore reflect the redistribution of soil and sediment particles in the landscape. Measurements of the distribution of the radionuclide within the landscape and its activity levels in sediment moving through the landscape can thus be used to derive estimates of rates of sediment redistribution and to trace the source of mobilised sediment. In this way, it is possible to obtain spatially distributed information on soil and sediment redistribution within the landscape and to use a common or integrated approach to investigate several components of the catchment sediment budget.

To date, most studies involving environmental radionuclides have focused on the use of ^{137}Cs, but increasing attention has been given to the use of alternative radionuclides, and more particularly ^{210}Pb$_{ex}$ and ^{7}Be. Each of these fallout radionuclides is characterised by a different origin, fallout record and half-life and thus offers different opportunities for use as a tracer. Caesium-137 is an artificial radionuclide, originating primarily from nuclear weapons testing, and thus its fallout has varied markedly through time. Fallout was first detected in the early 1950s, reached a peak in the early 1960s prior to the Nuclear Test Ban Treaty, and rapidly declined to very low levels by the mid 1970s. In some areas of Europe and adjacent regions, significant further short-term inputs were received in 1986 as a result of the Chernobyl accident. This time variable input represents an important characteristic in many applications of ^{137}Cs

measurements, for example in establishing the chronology of sediment deposits and thereby estimating sedimentation rates. Caesium-137 fallout is also characterised by significant spatial variability at the global scale, reflecting control by both location in relation to the nuclear weapons tests and global stratospheric circulation patterns and the magnitude of annual precipitation. Fallout inventories in some equatorial areas and throughout much of the southern hemisphere are almost an order of magnitude less than in the northern hemisphere, where most of the early work in developing the potential of ^{137}Cs measurements was undertaken. These reduced inventories introduce some constraints on the potential for using ^{137}Cs measurements in different areas of the world. With a half-life of 30.2 years, ^{137}Cs is relatively long-lived, although in the absence of further significant fallout, existing inventories will necessarily decline through time, making measurements increasingly difficult or unreliable in areas with low inventories.

In contrast to ^{137}Cs, $^{210}Pb_{ex}$ is of natural geogenic origin, being a natural product of the ^{238}U decay series, with a half-life of 22.3 years. It is derived from the decay of gaseous ^{222}Rn, the daughter of ^{226}Ra. Radium-226 occurs naturally in soils and rocks and will generate ^{210}Pb that will be in equilibrium with its parent. Upward diffusion of a small quantity of the ^{222}Rn from the soil introduces ^{210}Pb into the atmosphere and its subsequent fallout provides an input of this radionuclide to the soil surface that is not in equilibrium with its parent ^{226}Ra. This fallout component is commonly referred to as 'unsupported' or 'excess' ^{210}Pb, since it cannot be accounted for (or supported by) decay of the in-situ parent. Again in contrast to ^{137}Cs, the fallout input of $^{210}Pb_{ex}$ is essentially constant through time, and, by virtue of this contrasting behaviour, $^{210}Pb_{ex}$ frequently offers a useful complement to ^{137}Cs in many applications. Much less is known about the global pattern of $^{210}Pb_{ex}$ fallout, but the magnitude of fallout inputs are known to reflect both the magnitude of the annual precipitation and location on the continental land masses in relation to the prevailing winds. In some locations where ^{137}Cs activities are low due to low fallout receipt, $^{210}Pb_{ex}$ activities may be significantly higher and this radionuclide could therefore provide an alternative to ^{137}Cs in areas with low ^{137}Cs inventories.

Beryllium-7 is also of natural origin, but unlike $^{210}Pb_{ex}$, its origin is cosmogenic and it is produced in the upper atmosphere by cosmic ray spallation of nitrogen and oxygen. In contrast to ^{137}Cs and $^{210}Pb_{ex}$, the half-life of ^{7}Be is very short (53.3 days) and it therefore offers considerable potential for investigating soil and sediment redistribution over much shorter time periods (i.e. days rather than decades).

SOME EXAMPLES

In order to demonstrate the potential for using environmental radionuclides in sediment budget investigations, several examples can be usefully introduced. These relate to, firstly, quantifying rates and patterns of erosion and soil redistribution on catchment slopes and estimating sediment delivery ratios, secondly, quantifying rates and patterns of overbank deposition on river floodplains and estimating the associated conveyances losses, thirdly, establishing the relative contribution of different sources to the suspended sediment output from a catchment, and, finally, the synthesis of such results to construct a catchment sediment budget.

Quantifying rates and patterns of erosion and soil redistribution on catchment slopes

The results of using ^{137}Cs and ^{7}Be measurements to quantify both short- (i.e. several events) and longer-term (i.e. ~ 45 years) rates of soil erosion and redistribution within a 6.7 ha cultivated field at Higher Waltham Farm near Crediton, Devon, UK (cf. Walling et al., 1999), provide a useful example of the potential to document both rates and patterns of erosion within small areas. By focusing attention on an individual field, it is possible to assess the gross and net erosion from the area and to thereby estimate the sediment delivery ratio. The ^{7}Be measurements

were used to investigate the erosional response of the field under relatively extreme conditions, when a period of heavy rainfall coincided with the field being bare and compacted after the maize harvest. The resulting estimate of net soil loss for this short period (2.5 kg m^{-2}) was substantially greater than the longer-term mean value based on ^{137}Cs measurements (0.48 kg m^{-2} $year^{-1}$), although the associated sediment delivery ratios were very similar at 0.80 and 0.83 respectively.

Results such as those illustrated above require a dense network of sampling points and equivalent information is unlikely to be available for even a relatively small catchment. It is therefore necessary to devise sampling strategies capable of generating representative data that can be extrapolated to an entire catchment. A simple approach was used by Walling et al. (in press) to derive estimates of soil redistribution rates on the slopes of a 63 km^2 catchment in southern Zambia. In this case, representative transects were used to characterise sediment redistribution on the slopes under three contrasting land use types, namely, commercial cultivation, communal cultivation and bush grazing. Caesium-137 inventories are low in this region of Southerm Africa, and measurements of both $^{210}Pb_{ex}$ and ^{137}Cs were used to establish these sediment budgets, both as a means of validating the two approaches and to obtain additional information on medium-term changes in erosion rates. Since $^{210}Pb_{ex}$ fallout can be considered to be essentially constant through time, whereas that of ^{137}Cs occurred primarily between the late 1950s and the mid 1970s, comparisons of the results provided by the two radionuclides can provide information on changes in erosion rates through time.

Quantifying rates and patterns of overbank deposition on river floodplains and estimating conveyance losses

Measurements of the ^{137}Cs or $^{210}Pb_{ex}$ content of a sediment core collected from a river floodplain afford an effective and reliable means of estimating the sedimentation rate at that point (cf. He & Walling, 1996; Walling & He, 1997). Collection of a substantial number of cores from a floodplain reach in turn provides a means of investigating the spatial pattern of deposition and the influence of such factors as distance from the channel, depth of inundation and the local microtopography (cf. Walling & He, 1998). Since the estimates of sedimentation rate derived from ^{137}Cs measurements relate to the past ca. 40 years, whereas those based on $^{210}Pb_{ex}$ measurements relate to a longer period (e.g. ca. 100 years), it is also possible to investigate changes in sedimentation rates over the recent past (cf. Walling & He, 1999). Use of ^{137}Cs and $^{210}Pb_{ex}$ measurements to document rates and pattern of overbank sedimentation within a short reach of the floodplain of the River Severn at Buildwas, UK showed a close relationship between the microtopography and the sedimentation rate. The estimated mean sedimentation rate for this site over the past 40 years was 0.28 g cm^{-2} $year^{-1}$, whereas the equivalent rate for the past 100 years was 0.33 g cm^{-2} $year^{-1}$. This suggests that the sedimentation rate at this site has decreased towards the present. Collection of cores from representative sites on the floodplains of 21 rivers in the UK has permitted a more wide-ranging comparison of estimates of recent sedimentation rates provided by ^{137}Cs measurements with the longer-term values provided by ^{210}Pb measurements. In this case floodplain sedimentation rates estimated from the ^{137}Cs measurements ranged between 0.04 and 1.22 g cm^{-2} $year^{-1}$, whereas those based on $^{210}Pb_{ex}$ measurements ranged between 0.04 and 1.42 g cm^{-2} $year^{-1}$. By comparing the two estimates for each site, it was found that sedimentation rates have remained relatively constant over the past 100 years, although evidence of decreasing sedimentation rates was found at 11 sites, increasing rates at four sites and stable rates at six sites.

Although information on the detailed pattern of overbank deposition rates will be of value in many sediment budget investigations, in some case a more general estimate of the magnitude of the conveyance loss associated with overbank deposition within the floodplain system may be required. In this case there will be a need to extrapolate the findings from individual reaches or cross sections to the entire main channel system. This approach was adopted by the author and his co-workers on the Rivers Ouse and Wharfe in Yorkshire, UK (cf.

Walling et al., 1998). More than 250 sediment cores were collected from 26 representative transects located along the main channel systems of the Yorkshire Ouse and River Wharfe for ^{137}Cs analysis. The estimates of average sedimentation rate for the individual transects were extrapolated to the reaches between adjacent transects and the mean annual deposition flux or conveyance loss associated with overbank deposition on the floodplain bordering the main channel system was calculated. By comparing these losses with the mean annual sediment loads of the rivers, it was possible to establish the relative importance of floodplain storage in the sediment budget of the main channel system. In the case of the River Ouse, floodplain deposition was seen to account for ca. 40% of the total suspended sediment load delivered to the main channel system over the past 40 years. The equivalent value for the River Wharfe was ca. 49%.

Tracing suspended sediment sources

Growing recognition of the many environmental problems associated with fine sediment in river systems has focused attention on the implementation of sediment management strategies. Information on sediment source is of critical importance in developing such management strategies and in identifying and targeting key source areas where control measures should be applied. Sediment source fingerprinting procedures have been shown to possess very considerable potential for providing such information (cf. Walling & Woodward, 1995). The successful application of sediment source fingerprinting procedures relies heavily on the availability of fingerprint properties, which are able to discriminate potential sources. Environmental radionuclides offer considerable potential for discriminating sediment derived from different source types, for example cultivated land, grazing land and channel banks and gullies (e.g. Walling & Woodward, 1992). Because of their fallout source, ^{137}Cs, ^{210}Pb$_{ex}$ and ^{7}Be accumulate at the soil surface and their concentrations in mobilised sediment provide an effective means of discriminating between surface and subsurface (e.g. channel bank) sources. Furthermore, because concentrations will be lower at the surface of cultivated land (by virtue of mixing into the plough layer), they also afford a means of discriminating sediment mobilised from cultivated and pasture land. In most current applications of sediment source fingerprinting procedures, composite fingerprints comprising a range of fingerprint properties are used, but environmental radionuclides are frequently included in such fingerprints as a key, if not sole, source indicator. The use of ^{137}Cs to discriminate sediment sources is usefully illustrated by the work of Zhang et al. (1997) in the 3.86 km^2 catchment of the Zhaojia Gully in the rolling loess plateau region of Shaanxi Province, China. In that study, ^{137}Cs concentrations were used to discriminate sediment derived from the rolling plateau and the gully areas and to establish the relative importance of these two sources to the sediment output from the catchment. The results emphasised the importance of the gully areas as the main sediment source and the need to target these areas in any attempt to reduce downstream sediment yields through the implementation of soil conservation and sediment control measures.

Establishing catchment sediment budgets

By providing information on erosion rates and sediment delivery from the slopes of a catchment, sediment source areas and transmission losses associated with overbank deposition on the floodplains bordering the main channel system, results such as those presented above can be integrated with measurements of sediment yield at the catchment outlet to establish a catchment sediment budget. Thus, for example, Walling et al. (2002) were able to construct sediment budgets for two small catchments in Central England and Walling et al. (2001) established a sediment budget for the 63 km^2 Kaleya catchment in southern Zambia. In the latter case, separate sub-budgets were constructed for the slopes under communal cultivation, bush grazing and commercial cultivation and these, along with erosion of channel banks and gullies,

accounted for the inputs to the channel system. Conveyance losses of sediment passing through the main channel system were associated with reservoir deposition and overbank sedimentation on the floodplains and the overall sediment delivery ratio from the catchment was estimated to be 9%.

PERSPECTIVE

Traditional monitoring techniques employed for assembling information on catchment sediment budgets possess many limitations in terms of spatial and temporal sampling. Environmental radionuclides offer the potential to overcome many of these limitations. Some key advantages include the following:

1. Provision of spatially-distributed data on sediment redistribution, which are compatible with recent advances in physically-based distributed modelling.
2. Generation of estimates of longer-term average rates avoids the problems of representativeness frequently associated with event-based or short-term data.
3. No major disturbance of the landscape is involved.
4. Estimates of sediment redistribution rates can be obtained on the basis of a single site visit and the need for costly instrumentation and monitoring is avoided .
5. Estimates of sediment redistribution rates based on contemporary sampling are retrospective and data are available immediately rather than after a long period of monitoring.
6. Source fingerprinting data provide spatially integrated results for the entire catchment.

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