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Geomorphological applications of environmental radionuclides

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Background

Environmental radionuclides as tracers of geomorphic processes

Geomorphologists have shown increasing interest in environmental radionuclides since pioneering studies by Ritchie and McHenry in the USA and Campbell, Longmore and Loughran in Australia. Environmental radionuclides have attracted this interest because they provide geomorphologists with the means to trace sediment movement within the landscape. They, therefore, facilitate investigation of subjects at the core of geomorphology, namely the rates and patterns of landscape change. Most attention has been focussed on the artificial radionuclide caesium-137 (^{137}Cs) but more recently potential applications of the natural radionuclides lead-210 (^{210}Pb) and beryllium-7 (^{7}Be) have been investigated (Walling *et al.*, 1995; Wallbrink & Murray, 1996a, 1996b). The origin, characteristics and applications of these radionuclides are summarised in Table 1. These radionuclides are of value as sediment tracers because of three important characteristics:

1. a strong affinity for sediment;
2. a global distribution;
3. the possibility of measurement at low concentration.

The first characteristic has been well attested, for mineral soils, in laboratory conditions (e.g. Singh & Gilkes, 1990) and is consistent with field observations (e.g. Walling & Quine, 1992), and the remaining characteristics have strong empirical support.

Table 1. Environmental radionuclides used in geomorphological contemporary process research. (Applications: SE = soil erosion; LS = lake sedimentation; FS = floodplain sedimentation; SF = sediment fingerprinting).

	Source	Half-life	Fallout	Applications
Caesium-137	nuclear weapons tests	30 years	1950s-1970s	SE; LS; FS; SF
Caesium-134	Chernobyl accident	2 years	1986	FS; SF
Lead-210	uranium series	22 years	continuous	SE; LS; FS; SF
Beryllium-7	cosmogenic	53 days	continuous	SF

It is important to recognise, however, that the use of radionuclides in geomorphological research is based on three assumptions. These have been identified elsewhere (e.g. Walling & Quine, 1992) and their discussion lies beyond the scope of this paper. The assumptions are, therefore, listed below with reference to some of the relevant discursive literature:

1. locally spatially uniform initial fallout distribution (Sutherland, 1991; Owens & Walling, 1996; Harper & Gilkes, 1994; Quine *et al.*, 1996);
2. lateral redistribution is associated with sediment redistribution (Rogowski & Tamura, 1965, 1970; Walling & Quine, 1992);
3. quantitative sediment redistribution rates may be derived from radionuclide redistribution data (Walling & Quine, 1990; Quine, 1995; Quine *et al.*, 1996, 1997).

In applications of radionuclides, in particular ^{137}Cs, to the investigations of mineral sediment dynamics it can be suggested that these assumptions are acceptable and associated with errors which are comparable or lower than those associated with alternative assessment strategies.

Quantification of net landscape change

It should be clear that if the assumptions outlined above are accepted, radionuclides which display the characteristics described in the previous section provide a means to answer fundamental geomorphological questions concerning the distribution of erosion and deposition in the landscape and the rates of these processes. Both ^{137}Cs and ^{210}Pb have been used to quantify net landscape change on hillslopes and floodplains. The logic is similar in each case and the approaches have been discussed in detail elsewhere (cf. Walling & Quine, 1991; Walling *et al.*, 1992, 1995). In essence the approach takes advantage of the uniform spatial distribution of inventories (radionuclide abundance per unit area) which would be expected in the absence of sediment redistribution, such that deviation from a uniform spatial distribution may be attributed to the effects of erosion and deposition. The procedure typically adopted involves four stages. (1) The total amount of radionuclide fallout received at the site – the reference inventory (RI) – is established either by sampling uneroded areas or by applying a relationship between fallout receipt and a variable for which accurate data are readily available (e.g. mean annual rainfall, cf. Basher & Matthews, 1993). (2) The spatial distribution of radionuclide inventories across the study site is established by measuring the radionuclide content of soil profile samples of known surface area collected from the study site. Each sample yields a point inventory – PI. (3) A qualitative measure of soil/sediment redistribution is obtained by calculation of the percentage residual (PP) at each sampling location, as follows: PP = ((PI-RI)*100)/RI. Negative percentage residuals are taken to indicate erosion and positive residuals to indicate deposition. (4) Quantitative measures of soil/sediment redistribution are obtained by establishing the quantitative relationships between soil and radionuclide loss and soil and radionuclide gain. For hillslope studies this is best achieved by simulating the changing radionuclide content of a soil profile, or series of profiles, when subject to erosion or deposition (cf. Quine, 1995; Quine *et al.*, 1996, 1997). For floodplain studies it is necessary to estimate the radionuclide content of deposited sediment. Where this is unknown it may be estimated using information, derived from depth distributions of radionuclides, concerning deposition rates and excess radionuclide capture (cf. Walling *et al.*, 1992; Walling and He, 1992). In a few exceptional cases following the Chernobyl accident it was possible to estimate floodplain sedimentation using the spatial distributions of Chernobyl-derived ^{134}Cs and the measured content of the same radionuclide in suspended sediment (Walling *et al.*, 1992).

Further Developments

Further developments of radionuclide-based approaches have allowed significant progress to be made in two areas which may be described as 'applied geomorphology' and 'confrontation of geomorphological theory and observations'.

Applied geomorphology

The identification and quantification of soil erosion and sediment storage have obvious value to those concerned with the development of land management strategies for the minimisation of land degradation and water resource pollution. Nevertheless, such strategies often require data concerning larger areas than those which may be routinely examined using the strategies outlined in the previous section. There has, therefore, been a need to develop approaches which allow access to data which are relevant at these spatial scales. In this development attention has focused on suspended sediment for two reasons. Firstly, for low order drainage basins, the suspended sediment load may be seen as an integrated sample of sediment from the eroding areas of the catchment, although it is important to recognise the role of delivery processes in influencing the representation of eroding areas in suspended sediment and in controlling the magnitude of sediment transport to the fluvial system. Secondly, the suspended sediment carried during storm events typically represents the major source of sediment-associated contaminants to the river system. Therefore, identification of the source of suspended sediments would provide valuable information concerning both the distribution of erosion and the sources of sediment-associated pollutants. Furthermore, tracing suspended sediment transport through the fluvial system can provide valuable insights into both sediment and potential contaminant storage. As the following examples illustrate, environmental radionuclides have proved valuable in the identification of both the source and the locations of storage of fluvial suspended sediment.

Suspended sediment sources

Environmental radionuclides have been used to identify the sources of suspended sediment by taking advantage of differences in the depth distributions of radionuclides associated with different land-use practices. These differences in depth distributions result in distinctive radionuclide contents of sediment, sometimes termed 'sediment fingerprints', derived from areas under different land-uses. Some success has been achieved using measurements of a single radionuclide, but this approach is limited to situations with only two potential sources and minimal potential for transformations due to sediment delivery processes. The use of multiple radionuclide signatures has provided a much more powerful tool (Walling & Woodward, 1992; Olley *et al.*, 1996). Nevertheless, as the following example of sediment fingerprinting in the River Leira, Norway demonstrates, effective use of the approach relies on geomorphological knowledge of the system and consideration of sources of variation in radionuclide signatures.

High suspended sediment loads carried by the River Leira were considered to be a major source of pollution to ecologically important lacustrine environments and initial field observations identified two potential sources of sediment, namely channel banks and arable fields. In 1990 an attempt was made to assess the importance of these sources using environmental radionuclides. Surface samples (0-5 cm depth) were collected from the potential source areas and, because few suspended sediment samples were available, samples of suspended sediment surrogates were collected from channel and floodplain contexts. Two radiocaesium signatures were measured: caesium-134, derived from the Chernobyl accident in 1986; and caesium-137, derived from both the Chernobyl accident and nuclear weapons testing. Distinction between the sediment sources was based on the total ^{137}Cs content and the ^{134}Cs: ^{137}Cs ratio. A ratio close to 0.6 (typically found in Chernobyl fallout) would indicate that most of the radiocaesium was derived from the Chernobyl accident, suggesting a surface origin, while lower values would indicate a significant 'bomb' component, suggesting erosion or mixing since 1986. The ^{137}Cs activities would be expected to decrease with increasing erosion, or with dilution by mixing, and increase with enrichment by size-selective processes. Figure 1 shows the signatures of the probable sources – eroded banks and arable fields – and the suspended sediment and surrogates. The signatures coincide with the conceptual model with a sub-surface origin indicated for the eroded banks and a mixed soil for the arable fields. Furthermore, the suspended sediment and surrogates show the conservative nature of the ratio and the particle-size related variation in the ^{137}Cs activity of the suspended sediment and surrogates.

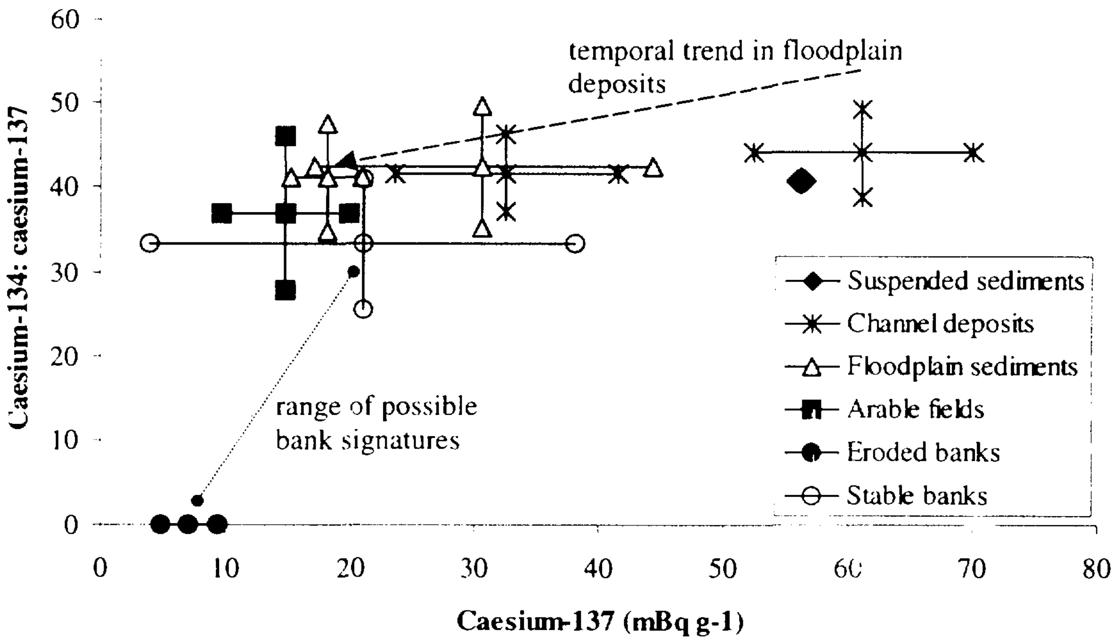

Figure 1. Sediment fingerprints of potential sediment sources and suspended sediments from the River Leira, Norway.

Examination of the eroded bank, arable field, and sediment fingerprints in Figure 1 suggests that the dominant source of sediment was the arable fields. However, before accepting this analysis, the possibility of temporal variation in the signatures of banks, fields and sediments must be considered. With respect to banks, the signature of eroded banks in Figure 1 indicates the characteristics of sediment that would be derived if the same areas were subject to further erosion. However, if bank erosion occurred as a series of shallow failures at different locations rather than successive erosion of the same scar, then the signature of sediment derived from bank erosion would be represented by a bank that appeared stable at the time of sampling. As Figure 1 shows, the signature of stable banks is close to that of arable fields and the suspended sediment surrogates. More certain characterisation of the signature of sediment derived from channel banks would only be possible with additional evidence concerning the mechanisms and spatial pattern of bank erosion, however, it is likely that the signature of sediment derived from banks would lie along the annotated dotted line in Figure 1. On this basis, channel banks may have been a more important source of sediment than indicated in the initial analysis. Although the signature of arable fields is expected to show less temporal variation than the bank signature, both higher ^{134}Cs: ^{137}Cs ratios and higher ^{137}Cs contents would be expected in sediment eroded from the fields during the period between the deposition of Chernobyl-derived radiocaesium and its mixing by cultivation through the plough layer. Evidence for a rapid decline in ratios and ^{137}Cs contents in suspended sediment would, therefore, be indicative of an important arable field component. Direct evidence for temporal variation in the signatures of suspended sediment is lacking, however, an indication of the temporal trend was obtained by examining the variation with depth in the signatures of floodplain sediments and this trend is shown as an annotated dashed line in Figure 1. The direction of this trend is certain but tight temporal control is lacking. Nevertheless, the trend is more consistent with a dominant arable source than a channel bank source. In conclusion, consideration of temporal variation in signatures may make interpretation more problematic and lead to a more qualified conclusion with identification of additional data requirements, however, this is preferable to erroneous interpretation of the fingerprint data.

Suspended sediment storage

From both geomorphological and environmental management perspectives, identification of the fate of suspended sediment can be as valuable as location of the source. This is reflected in growing interest in determining sediment budgets from drainage basins. The Chernobyl accident in 1986 provided a unique opportunity to derive a sediment budget for the River Severn (Walling & Quine, 1993). The spatial distribution of Chernobyl-derived fallout ^{134}Cs over the Severn catchment was characterised by low levels of fallout (< 250 Bq m^{-2}) in the areas of well-developed floodplain in the middle and lower reaches, and much higher levels (1000-3000 Bq m^{-2}) over the headwaters. Therefore, in the period following the Chernobyl accident, erosion of ^{134}Cs from headwater areas would be expected to lead to labelling of the suspended sediment transported through the river system with ^{134}Cs. Therefore, a ^{134}Cs budget with a tightly controlled temporal framework, could be established by monitoring the ^{134}Cs flux from the basin and deriving estimates of conveyances losses to intermediate storage through measurement of excess ^{134}Cs in floodplain and channel sediments. The methods used and data obtained are discussed in detail elsewhere (Walling & Quine, 1993) and Table 2 provides only a summary of the approach and results. The derivation of the sediment budget from the ^{134}Cs budget rests on the assumption that the ^{134}Cs mirrors the behaviour of fine sediment passing through the fluvial system. Despite this assumption and the caveats discussed by Walling and Quine (1993), this example demonstrates the potential for deriving hitherto unattainable insights into sediment dynamics at the catchment scale using environmental radionuclide data.

Table 2. Summary of the derivation of a sediment budget for the River Severn, UK, upstream of Upton on Severn, for a 3 year period from April 1989. A full discussion is presented by Walling and Quine (1993).

	Sediment output	**Floodplain storage**	**Channel storage**
^{134}Cs data	temporal variation in ^{134}Cs content of suspended sediment at Upton on Severn	excess ^{134}Cs at 27 sites	excess ^{134}Cs at 10 sites
Other data	annual suspended sediment load at Upton on Severn	active floodplain extent at 2 km intervals	channel width at 2 km intervals
Derived data	total ^{134}Cs throughput at Upton on Severn: April 1986- April 1989:	total ^{134}Cs storage in floodplain sediments upstream of Upton on Severn	total ^{134}Cs storage in channel sediments upstream of Upton on Severn
^{134}Cs budget	3.4×10^{10} Bq	1.1×10^{10} Bq	8.6×10^{8} Bq
Suspended sediment budget (%)	74.6	23.6	1.8

Confrontation of geomorphological theory and observations

The second area of development focuses on the attempts to integrate or compare data derived from radionuclide studies, in these cases ^{137}Cs-based investigations, with other geomorphological observations and 'theory' (the term is used loosely to represent an accepted view). This is discussed in the context of two examples. The first examines the processes of erosion on agricultural land in Europe, while the second addresses the sources of sediment in the 'rolling hills' area of the Loess Plateau, China.

Soil erosion on agricultural land in Europe

On the basis of field observations, many geomorphologists have identified water erosion as the dominant geomorphic process on agricultural land (cf. Boardman & Bell, 1992). However, patterns and rates of soil redistribution derived using ^{137}Cs do not conform to expectations based on water erosion. Firstly, observed and simulated patterns of water erosion indicate that rates increase with upslope contributing area and maximum rates are often identified in areas of flow convergence in slope and valley floor concavities. In contrast, ^{137}Cs-derived soil redistribution patterns typically indicate that maximum soil loss occurs on upper, convex slope elements and that concave elements are associated with net soil gain (cf. Quine & Walling, 1993a). Secondly, most gross rates of soil erosion derived using ^{137}Cs significantly exceed comparable observed rill erosion rates. For example, the same field at Dalicott Farm in Shropshire was monitored for rill erosion over a 9 year period (Evans, 1992) and sampled for ^{137}Cs (Quine & Walling, 1991). The rill-based water erosion rate of 0.3 t ha^{-1} yr^{-1} (Evans, 1992) was an order of magnitude lower than the ^{137}Cs-derived gross erosion rate of 10.2 t ha^{-1} yr^{-1} (Quine & Walling, 1991). Such deviations have led some to suggest that the ^{137}Cs-derived rates are unreliable, however, at Dalicott Farm, independent data for net landscape change were identified which indicated that both the patterns and the rates of soil redistribution derived using ^{137}Cs were reliable (Quine & Walling, 1993b). Much of this apparent contradiction can be accounted for if tillage erosion is taken into account (Quine *et al.*, 1996). Experimental studies of tillage translocation have demonstrated that it is a diffusive process (characterised by soil loss from concavities and gain in convexities) of sufficient magnitude to be responsible for significant within-field soil redistribution (Lindstrom *et al.*, 1990, 1992; Govers *et al.*, 1994). These authors have suggested that mean annual tillage translocation and tillage erosion associated with the use of an implement in opposing directions in alternate years may be described as follows:

$$Qt = kS \quad (1)$$

$$Rt = k.dS/dx \quad (2)$$

where, Qt is the soil flux due to tillage (kg m^{-1}); S is the slope tangent; Rt is the soil redistribution rate due to tillage (kg m^{-2}); x is horizontal distance (m); k is the tillage constant (kg m^{-1}), with values ranging from 230-360 for a mouldboard plough (Lindstrom *et al.*, 1990, 1992; Govers *et al.*, 1994). In view of this evidence for the importance of tillage erosion, an approach was developed to account for tillage erosion in the derivation of erosion rates from ^{137}Cs data (Quine *et al.*, 1994, 1996, 1997; Govers *et al.*, 1996). An outline of the approach is shown in Figure 2.

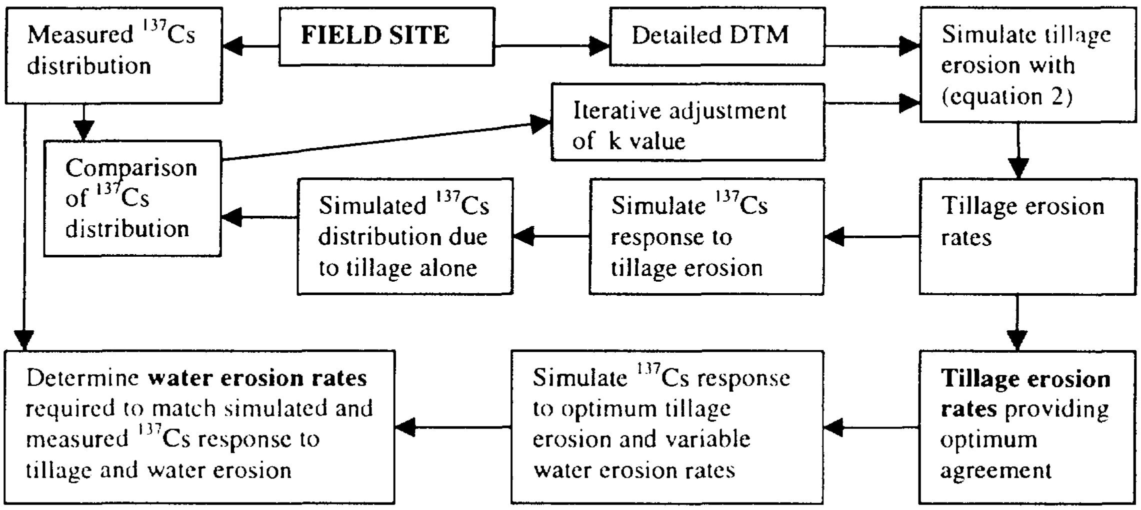

Figure 2. Approach to derivation of tillage and water erosion rates from ^{137}Cs data (Quine *et al.*, 1994, 1996, 1997).

Because the approach outline in Figure 2 provides separate estimates of water erosion and tillage erosion, the results may be evaluated by comparison of the ^{137}Cs-derived water erosion rates with field observations of water erosion. Table 3 provides a summary of two examples of such comparisons. At Dalicott Farm, UK, even when tillage erosion has been taken into account, both the ^{137}Cs data and the topographic data (derived from differences in height between fields adjacent to a boundary, see Quine *et al.* , 1996) indicate rates of water erosion (1.1-1.5 and 0.25-0.5 kg m^{-2} $year^{-1}$) which are very significantly higher than the rill-based estimate for the field (0.1 kg m^{-2} $year^{-1}$) obtained by Evans (1992). This may be due to the fact that the former rates correspond to a small part of the field, characterised by the highest slope angle and length, whereas the rill-based estimates represent erosion over the whole field area. However, it is also possible that the field observations underestimate erosion at these low levels. The maximum rates of denudation and accretion derived using ^{137}Cs for the Dalicott slope equate to 0.4-0.5 cm $year^{-1}$ and 0.3-0.4 cm $year^{-1}$, respectively. Surface impacts of this magnitude would not be readily identifiable by field observation even if the erosion was concentrated over a small part of the area.

Table 3. Comparison of water erosion rates derived from field survey and ^{137}Cs (Quine *et al.*, 1994, 1996). The Dalicott field observations were made by Evans (1992).

	Huldenberg, Belgium (m^3 $year^{-1}$)		Dalicott, UK (kg m^{-2} $year^{-1}$)
	Rills	Ephemeral Gully	
Field observation	1.5 – 4	5-10	0.1 (field, 9 years)
^{137}Cs-derived	3.3	5.1	1.0 - 1.5 (transect, 38 years)
Topographic data			0.25 - 0.5 (transect, >500 years)

At Huldenberg in Belgium, the close quantitative agreement in the integrated rates was mirrored by close agreement between the field observations and the ^{137}Cs-derived spatial distribution of water erosion rates, with both distributions showing maximum erosion in the valley floor thalweg and slope concavities (Quine *et al.*, 1994, 1997). The magnitude of tillage erosion at the site is demonstrated by the high 'k' value of 550 kg m^{-1} $year^{-1}$ which was derived. This reflects the annual cycle of mouldboard ploughing, discing and harrowing. The significance of tillage erosion is demonstrated in the pattern of landscape evolution at the site (obtained by examining the net effect of both water and tillage erosion) which, despite the incision of the concave slope elements by water erosion, was dominated by infilling of the concavities. Furthermore, tillage erosion was found to be the dominant process over >70% of the field area (Quine *et al.*, 1997). In conclusion, confrontation of accepted views concerning erosion on agricultural fields with data derived from ^{137}Cs has contributed to a recognition of the importance of tillage erosion, and a re-evaluation of the pattern of landscape evolution.

Sediment sources in the Loess Plateau, China.

The second example of confronting geomorphological theory and observations addresses the sources of sediment in the 'rolling hills' area of the Loess Plateau, China. Data derived from erosion plots has been widely used to derive empirical relationships between erosion rates on cultivated land and topographic attributes of fields. On the basis of these relationships, it has been suggested that sediment from the cultivated fields accounts for *ca* 50% of the very high sediment yields (*ca* 10 000 –25 000 t km^{-2} $year^{-1}$) of the rivers draining the area, the remainder being derived from gully slopes. In order to evaluate this suggestion, a ^{137}Cs-based investigation of soil erosion rates was undertaken in an area of cultivated fields near Ansai, in the rolling hills area of the Loess Plateau in Shaanxi Province (Quine *et al.*, 1997; Zhang *et al.*, 1998). Comparison of the ^{137}Cs-derived rates and estimates based on the empirical relationships suggested that the latter led to a significant over-estimate of erosion. Furthermore, detailed comparison suggested that the over-estimation was particularly acute when predictions were made for slopes with lengths in excess of the longest erosion plot (50 m) used in the derivation of the empirical relationship (Zhang *et al.*, 1998). This is significant because the majority of the slopes in the area have lengths in excess of 50 m. The ^{137}Cs-derived data suggest that erosion rates from the cultivated land are closer to 8000 – 9000 t km^{-2} year^{-1} than the 10000 – 16000 t km^{-2} year^{-1} suggested by the empirical relationships and, therefore, that the contribution of the cultivated fields to the total sediment yield of the rivers draining the area would be 20-40%. Nevertheless, these estimates must be treated with caution because the ^{137}Cs data were derived from a small number of fields. In order to further investigate the contribution of cultivated fields to the sediment production, a second radionuclide-based investigation was conducted. In this case, the origin of sediments deposited in the Zhaojia dam were estimated using ^{137}Cs data (Zhang *et al.* 1997). The two principal sources were identified as mass movements from the gully slopes (characterised by a ^{137}Cs content of 0 mBq g^{-1}) and cultivated fields. A stratified sampling strategy was employed to characterise the sediment derived from the gently sloping and steep cultivated fields. Areal estimates were used in conjunction with ^{137}Cs-based erosion rate estimates to derive weighted average ^{137}Cs contents of the sediment eroded from the fields. These estimates were then adjusted for radioactive decay (Table 4). The dam had a complex, but well-documented history and it was, therefore, possible to identify sediments associated with 2 specific floods in 1993 and also a series of floods between 1973 and 1977 (Table 4). A simple mixing model was used to derive the contribution of the cultivated fields and gullies to the total sediment yield:

$$Cd = Cp * fp + Cg * fg \qquad (3)$$

where, Cd, Cp and Cg represent the ^{137}Cs contents of flood deposits in the dam, sediment derived from the plateau fields and the gully slopes, respectively (mBq g^{-1}); fp and fg represent the fractions by mass of the sediment in the dam derived from the plateau and gully. The values for Cd, Cp and fp for the period from 1973-1977 and for the two 1993 floods are summarised in Table 4. These data show the same pattern as in the previous study, namely a significantly reduced estimate of the contribution of the cultivated slopes to sediment yield (17-27%), represented by the dam sediments. There is, therefore, a limited but growing body of internally consistent evidence from radionuclide studies to suggest that a re-evaluation is needed concerning the major sediment sources in the rolling hills area of the Loess Plateau. Although the body of available radionuclide data represent only a small area, the same criticism can be levelled at the datasets used in the development of the empirical relationships on which current estimates of sediment sources are based.

Table 4. The ^{137}Cs content of sediment derived from cultivated fields (Cp) and deposited in dams (Cd), and the estimated contributions of cultivated fields (fp) to sediment yield (fp) in Zhaojia gully, Shaanxi Province, China (Zhang *et al.*, 1997).

	1973	1974	1975	1976	1977	21.7.93 – 1.8.93	21.8.93
Cp (mBq g^{-1})	11.5	10.5	9.9	9.7	9.4	3.9	3.9
Cd (mBq g^{-1})	2.2	1.8	2.3	1.7	2.2	0.7	1.1
fp*100 (%)	19	17	23	17	24	18	27

Overview

The two examples illustrate the insights which may be gained when radionuclide studies are used to confront or evaluate accepted geomorphological 'theory'. These have implications beyond academic interest. Recognition of the soil redistribution potential of tillage erosion has important implications for two reasons. Firstly, estimates of decline in soil productivity based on water erosion rates will greatly underestimate the potential decline which may occur as a result of tillage and water erosion, if soil depth is an important consideration. Secondly, water erosion may be promoted by tillage erosion because of the transfer by tillage erosion of erodible material from the slopes into concavities where it is more susceptible to erosion by water. Evidence concerning sediment sources in the Loess Plateau suggests that, where off-site impacts are of significant concern, a shift in emphasis concerning erosion and sediment control strategies may be necessary.

Conclusions

Geomorphological applications of environmental radionuclides provide unique access to detailed quantitative data concerning landscape change over a range of timescales. When combined or compared with other geomorphological data, valuable insights may be obtained and, in some cases, it may be necessary to re-evaluate accepted views. New applications continue to be developed such as the use of the tracing properties of radionuclides in tillage experiments (Lobb *et al.*, 1995; Quine *et al.*, 1998). Nevertheless, there remain areas of development of radionuclide approaches which require attention. In particular, a better understanding of the relationships between radionuclide transport and sediment transport are needed. Ongoing experimental work should contribute further to this. Applied geomorphological use of the approach will be greatly enhanced by the development of extrapolation procedures. In this area integration with erosion models offers considerable potential (de Roo and Walling, 1993; Gillieson *et al.*, 1996) which requires further development (Quine, 1998).

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