Use of radiometric fingerprints to derive information on suspended sediment sources

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ABSTRACT There is an increasing need for information on suspended sediment sources which is difficult to meet using traditional monitoring techniques. The fingerprinting technique offers an alternative approach, but it is important to select appropriate fingerprinting properties. Fallout radionuclide concentrations appear to offer considerable potential as fingerprinting properties, since they are essentially independent of lithology and soil type and can clearly distinguish surficial and channel sources. Examples of the use of ^{137}Cs, unsupported ^{210}Pb and ^{7}Be concentrations to decipher suspended sediment sources in two small drainage basins in Devon, UK are presented.

INTRODUCTION

Recent concern for the role of suspended sediment in non-point pollution from land use activities and in the transport of nutrients and contaminants has highlighted the need for sediment monitoring programmes to provide information on sediment sources as well as concentrations and loads. This need has also been further promoted by the desire to establish sediment budgets for drainage basins, by attempts to develop distributed sediment yield models and by attempts to develop more meaningful geomorphological interpretations of sediment yield data in terms of landscape evolution. Although in some circumstances it is important to ascertain the precise *spatial location* of sediment sources within a drainage basin, more often the requirement is for information on the *type* of source involved and, for example, whether the sediment has originated primarily from erosion of cultivated land, pasture or forest, or from channel or gully erosion, irrespective of the precise spatial location of the source.

Detailed information concerning the types and relative importance of sediment sources within a drainage basin is, however, difficult to assemble for all but very small basins using traditional monitoring techniques (cf. Peart & Walling, 1986). The "fingerprinting" technique offers an alternative approach to determining sediment source which would appear to offer very considerable potential (cf. Wall & Wilding, 1976; Oldfield *et al.,* 1979; Peart & Walling, 1986,1988). In essence, this method involves firstly, the selection of a physical or chemical property which clearly differentiates potential source materials and, secondly, comparison of measurements of the same property obtained from suspended sediment with the equivalent values for the potential sources. In the case of a simple distinction between surficial and channel sources, the property could be one that differentiated topsoil from the underlying parent material and bed rock. The essential simplicity of the fingerprinting technique is, however, complicated by a number of potential problems, including enrichment of the sediment relative to the source material and transformation of sediment properties within the fluvial system (cf. Peart, 1990). Furthermore, it is important to identify physical or chemical properties which vary over a substantial range and are therefore able to distinguish different sources in an unequivocal manner. Peart & Walling (1988) advocated the use of several alternative diagnostic properties, rather than a single indicator, in order to establish the consistency of the results obtained.

The quest for diagnostic properties capable of distinguishing a range of source types has embraced a large number of sediment properties, including sediment colour (Grimshaw & Lewin, 1980), clay mineralogy (Wall & Wilding, 1976), sediment chemistry (Peart & Walling, 1986), radionuclide concentrations (eg. Peart & Walling, 1986; Burch *et al.*, 1988; Wallbrink & Murray, 1990; Froehlich & Walling, 1991) and mineral magnetic parameters (eg. Oldfield *et al.*, 1979; Oldfield & Clark, 1990; Foster *et al.*, 1990), with varying degrees of success. For many applications, particularly in heterogeneous drainage basins with a variety of soil and rock types, the "ideal" property should be independent of lithology and soil type and for this reason fallout radionuclides, and more especially ^{137}Cs have attracted particular attention (cf. Walling & Bradley, 1990).

THE USE OF FALLOUT RADIONUCLIDES AS INDICATORS OF SEDIMENT SOURCE

Fallout radionuclides would appear to afford particularly valuable indicators of sediment source, because they can be seen as labelling the surficial materials of a drainage basin in a manner that will be consistent over large areas and effectively independent of soil and rock type. By virtue of their arrival at the land surface as fallout, they will commonly be rapidly adsorbed by surficial materials. Where the soil is undisturbed, the radionuclides will accumulate at or near the surface and concentrations will be relatively high. Where the soil is cultivated, the radionuclides will be mixed within the plough layer and surface concentrations will be lower. It should therefore be possible to distinguish sediment eroded from cultivated and undisturbed land. Near vertical surfaces such as river banks and steep gully walls will receive relatively little fallout and will therefore be characterized by low radionuclide concentrations. Furthermore, where erosion penetrates through the upper horizons of the soil, material mobilised from the lower horizons will again be characterized by low radionuclide concentrations, and surfaces that are eroding rapidly will have little opportunity to accumulate radionuclides and will also be associated with low radionuclide concentrations. Contrasts in the fallout history and decay rates of individual radionuclides may introduce further differences in their distribution which can be exploited to provide essentially independent evidence of sediment source. Three fallout radionuclides with different fallout histories and decay rates which appear to offer particular potential as fingerprint properties are ^{137}Cs, unsupported ^{210}Pb and ^{7}Be. All three are gamma emmitters and simultaneous non-destructive measurements of their activity can be undertaken relatively easily using modern high resolution gamma spectrometry equipment.

Caesium-137 is an artificial radionuclide (half-life 30.1 years) which is present in the environment largely as a result of global fallout associated with the testing of thermonuclear weapons during the period 1954-1968. The Chernobyl accident caused significant additions of this radionuclide during 1986 over large areas of Europe. Unsupported ^{210}Pb and ^{7}Be are natural radionuclides which also reach the soil surface via atmospheric fallout, although in this case the annual fallout is essentially constant through time rather than having occurred during a particular period. Lead-210 is a product of the ^{238}U decay series with a half-life of 22.26 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 which will be in equilibrium with its parent. Diffusion of a small proportion of the ^{222}Rn from the soil introduces ^{210}Pb into the atmosphere and its subsequent fallout provides an input of this radionuclide to surface soils and sediments which is not in equilibrium with its parent ^{226}Ra. This component is termed "unsupported" ^{210}Pb, since it cannot be accounted for (or supported) by decay of the in-situ parent. The amount of unsupported or atmospherically-derived ^{210}Pb in a sediment sample can be calculated by measuring both ^{210}Pb and ^{226}Ra and subtracting the supported or in situ component. Beryllium-7 is a cosmogenic radionuclide produced in the upper atmosphere by cosmic ray spallation of nitrogen and oxygen, which again arrives at the land surface as fallout. In this case, the radionuclide is short-lived (half-life 53.3 days) and its behaviour in terrestrial and aquatic environments has been less well documented. However, several studies have suggested that it may be useful for investigating recent sediment movement (cf. Larsen & Cutshall, 1981; Burch *et al.*, 1988; Wallbrink & Murray, 1990).

Oldfield & Clark (1990) have recently indicated that most existing attempts to use radionuclides to decipher suspended sediment sources within a drainage basin have used only

^{137}Cs. While a number of studies have also exploited unsupported ^{210}Pb as a tracer (eg. Wasson *et al.*, 1987), few investigations have used two or more radionuclides as fingerprints. As noted above, the use of several fingerprinting properties has been advocated by Peart & Walling (1986) as a means of ensuring the consistency and reliability of the results obtained. This paper describes a study undertaken by the authors in two small tributary basins of the River Exe in Devon, UK, which has attempted to examine the fingerprinting potential of the three radionuclides listed above. These can be expected to provide essentially independent evidence of sediment source by virtue of contrasts in both their fallout histories and their decay rates. The study had two basic aims; firstly, to investigate the distribution and behaviour of the three selected fallout radionuclides within the two drainage basins as a means of confirming their utility for distinguishing individual types of sediment source, and, secondly, to use the fingerprinting approach to identify the dominant sediment sources within the two basins.

THE STUDY BASINS

The location, relief and drainage networks of the two study basins are shown in Fig. 1. The drainage basin of the River Dart (46 km^2) is underlain by sandstones and shales of Upper Carb-

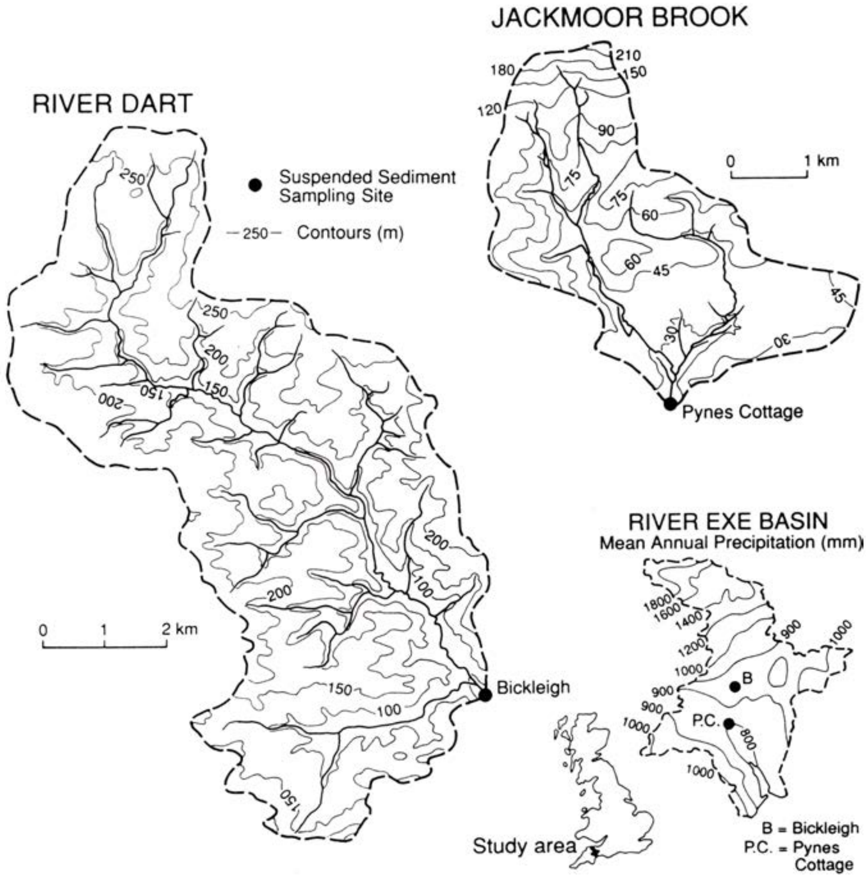

FIG. 1 The study basins.

oniferous age. The basin has a deeply dissected, incised terrain and is characterized by steep slopes (>11°) and tributary streams with steep gradients. The land use is dominated by permanent pasture, although mixed woodland is found on many of the steeper slopes. The mean annual precipitation received by the basin is ca. 1100mm. The mean annual suspended sediment yield is estimated to be about 75 t km^{-2} $year^{-1}$. The Jackmoor Brook basin is located 10km to the south and drains an area underlain by Permian sandstones and argillaceous breccias and conglomerates. The relief of this basin is more subdued and the slopes are commonly < 4°. The land use is dominated by mixed arable farming, although there are some small areas of mixed deciduous woodland and permanent pasture. The mean annual precipitation is about 800 mm. Within the local area this drainage basin is noteworthy for the relatively high suspended sediment concentrations (up to 3500 mg l^{-1}) which occur during storm events. Available records indicate that the mean annual suspended sediment yield is ca. 60 t km^{-2} $year^{-1}$. In both basins, neither the channel network nor the agricultural land with its pattern of hedge bank boundaries, provide obvious evidence of major sediment sources and in this situation the fingerprinting technique offers an attractive approach to sediment source identification. Furthermore, contrasts between the basins in terms of land use, relief and underlying geology could be expected to give rise to differences in the relative importance of the major sediment source types.

FIELD AND LABORATORY METHODS

In order to ascertain the distribution of the three radionuclides within the soils of the study basins, profiles were sampled at undisturbed and cultivated sites in both basins using the scraper plate technique described by Campbell *et al.* (1988). This provides bulk down-profile samples at 1 or 2cm increments and these samples (<2mm fraction) were used for ^{137}Cs and unsupported ^{210}Pb measurements. Short (15cm diameter) cores were also collected to depths of 10 cm and sub-divided into 3 and 5 mm increments for ^{7}Be determinations. Characterization of potential source materials was based on the collection of 40 samples from each basin which were representative of surface (0-2 cm) material from cultivated, uncultivated and woodland sites and of eroding channel banks. In order to permit direct comparison of these materials with suspended sediment, the <63μm fraction was separated by sieving and dried and disaggregated prior to gamma spectrometry.

Bulk water samples were collected at the outlets of both study basins during storm runoff events, using a submersible pump to fill several 25-litre polythene containers. The sediment was recovered from these bulk samples by continuous flow centrifugation and subsequently screened through a 2mm mesh sieve and freeze dried prior to gamma spectrometry. In all cases, these samples consisted almost exclusively of <63μm particles.

Measurements of the ^{137}Cs, unsupported ^{210}Pb and ^{7}Be content of both suspended sediments and source materials were undertaken directly by gamma spectrometry using a high purity germanium detector housed in a copper lined lead shield. Unsupported ^{210}Pb and ^{7}Be measurements followed the procedures described by Joshi (1987) and Larsen & Cutshall (1981).

THE VERTICAL DISTRIBUTION OF ^{137}Cs, UNSUPPORTED ^{210}Pb and ^{7}Be IN THE SOILS OF THE STUDY BASINS

Figure 2 illustrates the vertical distribution of the three radionuclides in soils representative of both cultivated and uncultivated sites in the study catchments. The forms of the ^{137}Cs profiles conform with existing evidence from other investigations in the UK and elsewhere (cf. Walling & Quine, 1990, 1992) and confirm that a large proportion of the inventory of this radionuclide is held within the upper 10 cm of undisturbed soils, whereas in cultivated soils the radiocaesium is mixed within the plough layer to depths of ca. 20-25 cm. As a result, material from the surface of undisturbed soils will be characterized by ^{137}Cs concentrations that are several times higher than those from cultivated soils.

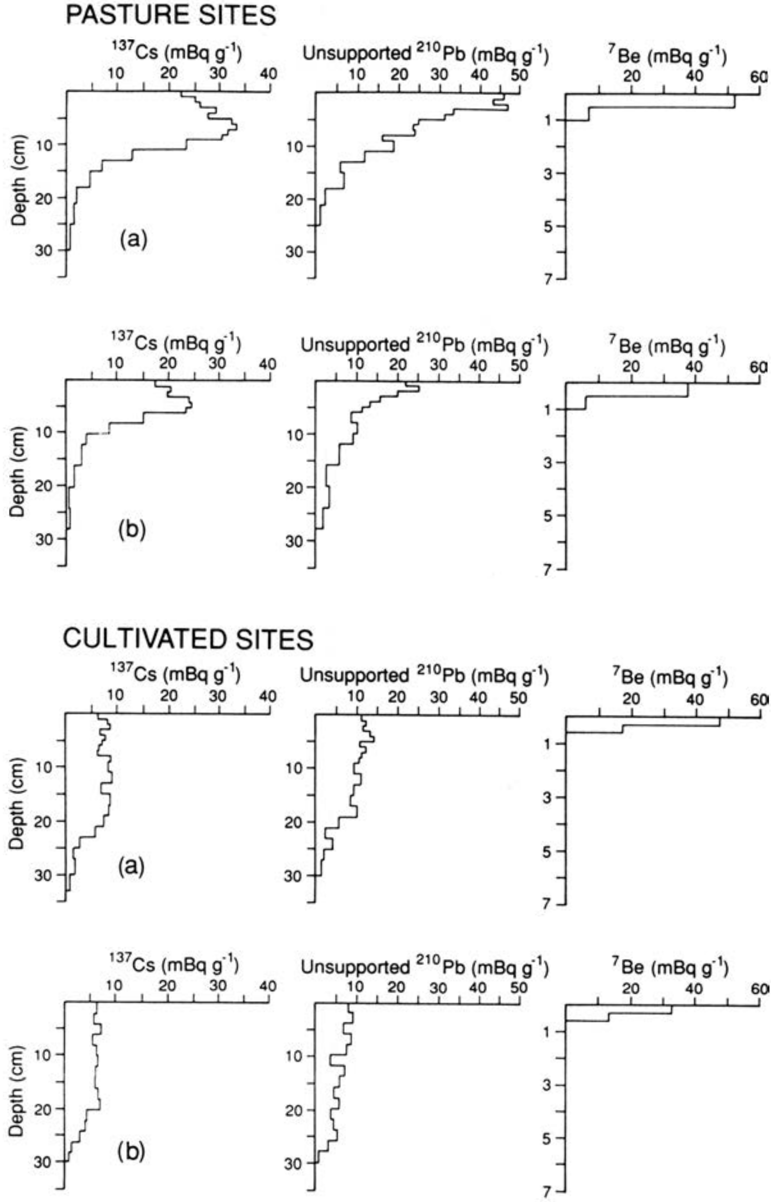

FIG. 2 The vertical distribution of ^{137}Cs, unsupported ^{210}Pb, and ^{7}Be in soil profiles from representative pasture and cultivated sites in the (a) Dart and (b) Jackmoor Brook basins.

Much less information is currently available regarding the vertical distribution of unsupported ^{210}Pb and ^{7}Be in UK soils, but the profiles illustrated in Fig. 2 conform to expectations based on physical reasoning. The unsupported ^{210}Pb profiles at undisturbed sites show maximum values at the soil surface (0-2 cm) and a progressive decrease with depth, with the ^{210}Pb : ^{226}Ra ratio reaching unity at approximately 25 cm in both profiles. The minor contrast with the ^{137}Cs profiles for undisturbed soils, where there is slight reduction of radiocaesium concentrations at the surface, may be related to the different fallout histories of the two radionuclides. In the case of ^{210}Pb the annual fallout input to the surface has been essentially constant through time, whereas in the case of ^{137}Cs surface inputs have been negligible over the past 20 years. Where mixing occurs as a result of cultivation, the shapes of the unsupported ^{210}Pb profiles from both basins closely match those of ^{137}Cs. As with ^{137}Cs, there is a clear contrast between the unsupported ^{210}Pb concentrations in material from the surface of undisturbed and cultivated soils, with the former exceeding the latter by 3-4 times. By virtue of its short half-life, ^{7}Be is largely concentrated in the top 5 mm of both pasture and cultivated soils, where it reflects fallout inputs during the preceding months. Any ^{7}Be moving to greater depths over longer periods will be undetectable due to decay. It is possible that the surface layer containing the majority of the ^{7}Be is in fact even thinner than that depicted in Fig. 2, since the cores used to collect the samples were sectioned at either 3 or 5 mm increments. Because the flux of ^{7}Be to the earth's surface is closely related to the pattern of rainfall input, it will exhibit appreciable temporal variability. As a result of its short half-life, the ^{7}Be concentrations in surface material will therefore vary through time, unlike those of ^{137}Cs and unsupported ^{210}Pb which will remain effectively constant in the medium-term. It is therefore more difficult to use concentrations of this radionuclide as a fingerprint property, but it would, nevertheless, appear to offer considerable potential for fingerprinting surface-derived material and for investigating the dynamics of sediment generation during the course of a storm event. For example, if rilling is an important process, the rapid incision of rills during a storm event could be expected to generate an increasing proportion of sediment from which ^{7}Be is absent. In comparing the radionuclide concentrations associated with cores from the two study basins, the higher activities associated with materials from the Dart basin can be largely attributed to the higher annual precipitation received by this basin (Fig. 1).

FINGERPRINTING SOURCE MATERIALS

The soil profiles depicted in Fig. 2 indicate that it should be possible to make a clear distinction between cultivated and uncultivated surface materials and channel bank materials from depths >10 cm below the surface in terms of their ^{137}Cs and unsupported ^{210}Pb concentrations.

The ^{137}Cs and unsupported ^{210}Pb activity of the <63μm component of four categories of potential source material, namely, cultivated fields, areas of permanent pasture, woodland areas and channel banks, have been measured in order to test this assumption. The results of these measurements are presented in Fig. 3. Figure 3 indicates that the four potential source materials

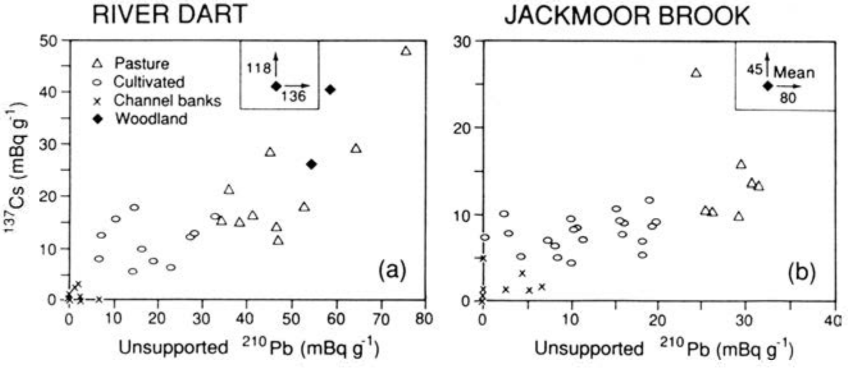

FIG. 3 A comparison of four potential source materials in terms of their ^{137}Cs and unsupported ^{210}Pb activities.

can be clearly distinguished in terms of their ^{137}Cs and unsupported ^{210}Pb concentrations. It is notable that the ^{137}Cs content of material from some permanent pasture sites is similar to that from cultivated sites, yet the unsupported ^{210}Pb values are much higher. In practice it is very often difficult to establish with certainty whether or not a pasture field has been ploughed and reseeded within the last 30 years or so. It indeed seems likely that some of the pasture sites plotted on Fig. 3 represent fields that were ploughed 10 or 20 years ago, thereby mixing the ^{137}Cs profile and reducing surface concentrations of radiocaesium, but have since received substantial amounts of unsupported ^{210}Pb fallout which have restored the surface concentrations of that radionuclide to levels similar to those associated with permanent pasture. Measurements of these two radionuclides could therefore be used to provide information on the land use history of particular fields. The woodland sites in the Jackmoor Brook basin exhibit ^{137}Cs and unsupported ^{210}Pb concentrations that are higher than those for permanent pasture. A similar distinction appears to exist in the Dart basin, but it is less clear. These elevated levels probably reflect the very high organic content of the surface layer of woodland sites.

COMPARING THE FINGERPRINTS OF POTENTIAL SOURCE MATERIALS WITH THOSE OF SUSPENDED SEDIMENT

The ^{137}Cs and unsupported ^{210}Pb activities associated with potential source materials are compared with those associated with suspended sediment in Fig. 4(a). For convenience of plotting, values for surface material from forest sites have not been included for the Jackmoor Brook, since these are very much higher than those associated with suspended sediment and

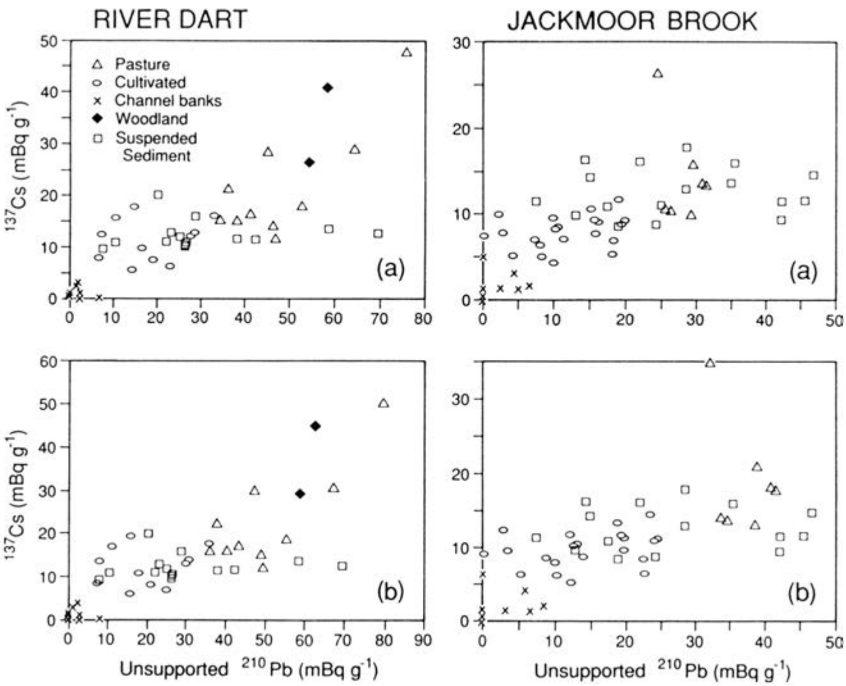

FIG.4 A comparison of the ^{137}Cs and unsupported ^{210}Pb activities of suspended sediment with those of representative samples of potential source materials. a) is based on the raw data, whereas in b) the values for the source materials have been corrected for enrichment effects (i.e. increased) to make them directly comparable with the suspended sediment values.

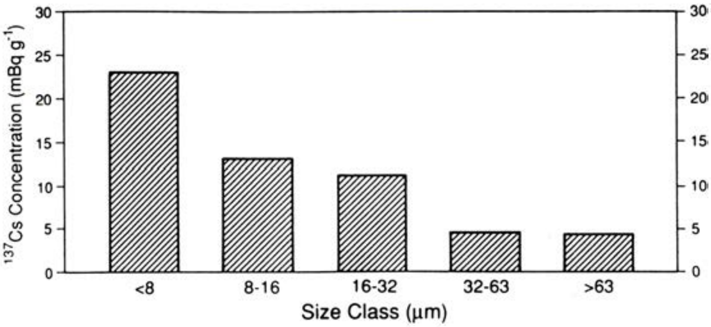

FIG. 5 Caesium-137 activity of individual size fractions separated from a representative sample of surface material from a cultivated field in the Jackmoor Brook basin.

effectively exclude it from being a potential sediment source. Furthermore, the relatively small area involved also intuitively precludes it from being an important source. Inspection of the plots for the River Dart and the Jackmoor Brook presented in Fig. 4(a) indicates that in several cases the suspended sediment is associated with values of ^{137}Cs and unsupported ^{210}Pb concentration that exceed those in the potential source materials. This reflects the influence of enrichment, wherein the suspended sediment samples are enriched in fines relative to the source material. It is well known that radionuclides are preferentially associated with fines and it is therefore necessary to take account of this enrichment effect when comparing fingerprints of sediment and source materials.

Figure 5 provides further evidence of the preferential association of ^{137}Cs with the fine fraction of soils and sediment. The data presented relate to a sample of surface material from a cultivated field in the Jackmoor Brook basin which was separated into five size classes using a combination of air elutriation (<63µm) (cf. Horowitz & Elrick, 1986) and wet sieving (>63µm). The specific ^{137}Cs activity for the five size classes is shown in Fig. 5. This information shows that the <8µm component is associated with a substantially higher ^{137}Cs activity than the coarser size fractions, although the medium silt classes are also important. By assuming that unsupported ^{210}Pb demonstrates similar grain size selectivity to ^{137}Cs and that source materials from both basins respond in a similar manner, information on the particle size composition of the source materials and the suspended sediment have been used in conjunction with the fractionation data presented in Fig. 5 to correct the ^{137}Cs and unsupported ^{210}Pb values for source materials to make them directly comparable with those for the suspended sediment (Fig. 4(b)). The correction factors applied to the Jackmoor Brook data are considerably higher than those applied to the source materials from the River Dart because of the finer nature of suspended sediment relative to potential source materials in the former case. The corrected data are also summarised in Table 1.

The evidence provided in Fig. 4(b)and Table 1 suggests that in the case of the River Dart the suspended sediment exhibits a range of ^{137}Cs activity which is directly comparable with that from cultivated fields. However, the range of unsupported ^{210}Pb activity associated with suspended sediment overlaps that for surface materials from permanent pasture, suggesting that pasture may represent a significant secondary source. Channel banks would appear not to be a major sediment source in this basin since the activities associated with this material are very considerably lower than those for suspended sediment. It is possible, however, that the mixing of sediment derived from channel banks with topsoil material eroded from uncultivated fields could produce intermediate radionuclide values for suspended sediment, comparable to those of cultivated fields. However, the ^{7}Be measurements indicate that suspended sediment always contains appreciable ^{7}Be activity (Table 1) and must therefore be derived primarily from surface sources. While such mixing is clearly a potentially significant process, the ^{7}Be values indicate that channel sources are not of sufficient magnitude to effect such a marked dilution.

TABLE 1 A comparison of the mean radiometric fingerprints of suspended sediment with those of potential source materials corrected for contrasts in grain size composition (mBq g^{-1}).

	RIVER DART				JACKMOOR BROOK			
Material	Unsupp ^{210}Pb	^{137}Cs	^{7}Be	n	Unsupp ^{210}Pb	^{137}Cs	^{7}Be	n
Woodland	90.8	68.5		3	112.4	63.5		2
Pasture	50.6	22.7		10	37.3	18.6		7
Cultivated	20.0	12.3		11	14.4	14.4		21
Banks	1.1	0.7		16	2.4	1.8		10
Suspended Sed.	30.4	12.3	64.6	14	27.2	12.5	87.4	17

In the case of the Jackmoor Brook, the suspended sediment fingerprints are similarly close to those for cultivated soils in the case of ^{137}Cs and overlap into the range of permanent pasture in the case of unsupported ^{210}Pb. Bank material can again be discounted as a significant source and it can be suggested that the dominant source in this basin is cultivated fields with pasture representing a secondary source

INTER- AND INTRA-EVENT VARIATIONS IN SEDIMENT SOURCE

The data presented in Fig. 4 provide only generalised information on the likely importance of the several potential sediment sources, since suspended sediment data exhibit a range of fingerprint values, indicating that the precise importance of the individual sediment sources varies from event to event and even within events. The radiometric fingerprint data can, however, also be used to investigate inter- and intra-event variations in sediment sources and several examples of this potential are provided in Fig. 6.

Figure 6(a) plots the values of ^{7}Be and unsupported ^{210}Pb activity associated with suspended sediment samples collected during a series of storm events in March 1991. In each case the samples were collected around the time of the hydrograph peak and their radiometric fingerprints suggest that the dominant sediment sources varied significantly between these three events. The lowest ^{7}Be and unsupported ^{210}Pb activities are associated with the event of March 8, which was the largest runoff event and may therefore have effected deeper rilling and possibly some channel scouring. The comparatively low values associated with this event may be partly explained by antecedent catchment conditions, since the highest activities in this sequence of storm events are associated with the smallest event four days earlier (4.3.91) when topsoil sources appear to have been dominant. Figure 6(b) provides information on the variation of ^{137}Cs, ^{7}Be and unsupported ^{210}Pb activity during a storm event in the River Dart basin. The levels of ^{137}Cs and unsupported ^{210}Pb and also ^{7}Be associated with all the samples suggest that surface material, and more particularly cultivated soils, represent the dominant source. However, the progressive decline in ^{7}Be and unsupported ^{210}Pb activity during the sampled period indicates a shift in sediment source during the course of the storm event. This trend could be the result of an increasing contribution by sediments with comparatively low radionuclide concentrations at the time of peak discharge. The greater relative reduction in ^{7}Be and unsupported ^{210}Pb activity relative to that of ^{137}Cs may also point to the dominance of sources where recent fallout inputs of these two radionuclides have accumulated at the surface, whilst ^{137}Cs activity remains more uniformly distributed throughout the profile. These could represent fields cultivated during the previous autumn. As erosion proceeds, the surface layer bearing higher levels of ^{7}Be and unsupported ^{210}Pb would be removed, exposing material with lower activities to erosion.

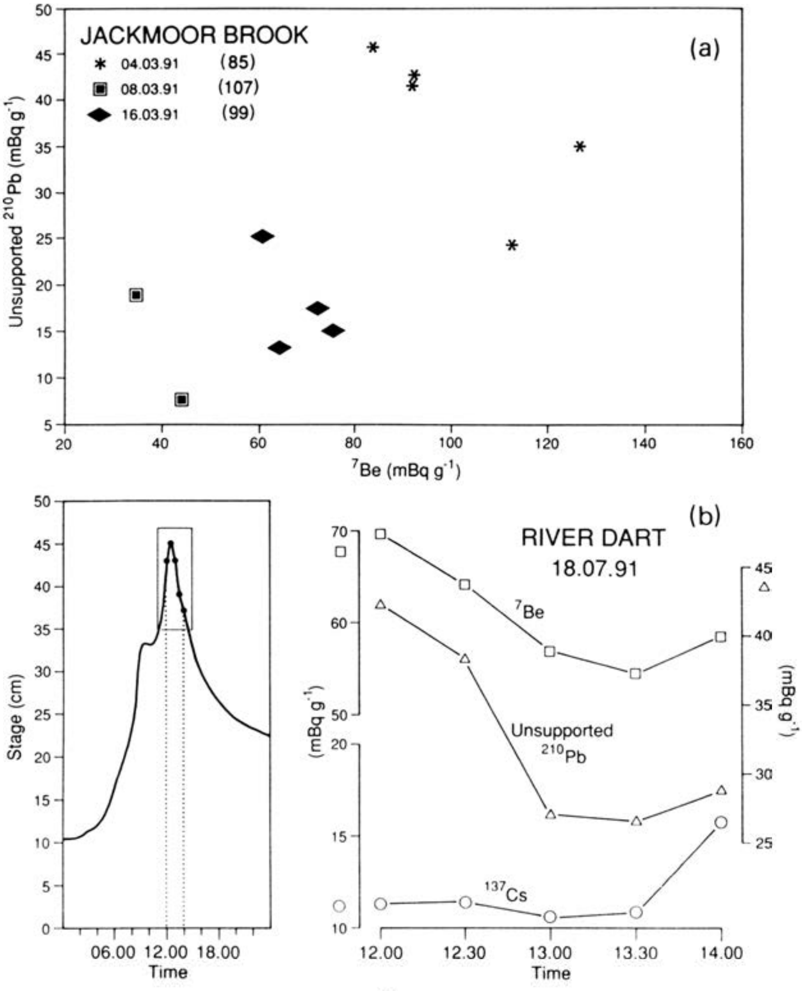

FIG. 6 a) Unsupported ^{210}Pb and ^{7}Be concentrations for suspended sediment samples collected during a series of storm events in March 1991 in the Jackmoor Brook basin (Stage peaks in cm). b) Changes in the radionuclide concentration of suspended sediment during a single storm event in the River Dart basin on 18.7.91.

PERSPECTIVE

The results presented above demonstrate that fallout radionuclides can provide valuable fingerprints for identifying suspended sediment sources. These fingerprints are effectively independent of the underlying geology and soil type and can therefore be used in heterogeneous basins where other potential fingerprinting properties may have limited application. However, following Peart & Walling (1986), the need to use a range of fingerprinting properties must be emphasised. Radiometric indicators should ideally be used in conjunction with other sediment properties which provide further clarification of the sources involved and may thus help to exclude possibilities not definitively resolved by the former. There is also a need to develop multivariate techniques capable of processing the evidence afforded by a range of fingerprint properties.

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