



Tracing suspended sediment sources in catchments and river systems

D.E. Walling*

Department of Geography, University of Exeter, Amory Building, Rennes Drive, EXETER, EX4 4RJ, UK

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Abstract

Recent years have seen a growing awareness of the wider environmental significance of the suspended sediment loads transported by rivers and streams. This includes the importance of fine sediment in the transport of nutrients and contaminants, including phosphorus (P). Sediment source exerts a key control on the physical and geochemical properties of suspended sediment, including its P content, and will also influence the potential for implementing effective sediment and diffuse source pollution control strategies. Information on suspended sediment source, defined in terms of both source type and spatial origin, is therefore increasingly needed. Such information is difficult to obtain using traditional monitoring techniques, but source tracing or fingerprinting techniques afford a valuable and effective alternative approach to establish the relative importance of potential sediment sources. This contribution reviews the development of source fingerprinting techniques, presents several examples of their application in UK catchments and discusses the need for future development of the approach and the potential for extending its application.

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1. Introduction

1.1. The context

Recent years have seen a growing awareness of the wider environmental significance of the suspended sediment loads transported by rivers and streams. This includes the importance of fine sediment in the

transport of nutrients and contaminants, such as phosphorus (P), pesticides, PCBs, heavy metals and pathogens through fluvial systems (e.g., Shear and Watson, 1977; UNESCO, 1983; Allan, 1986; Warren et al., 2003). Table 1 serves to emphasise the important role of fine sediment in catchment P exports, by showing that sediment-associated transport can account for a major proportion of the P load in UK rivers. Recognition of the wide-ranging environmental significance of fine sediment has generated a need for improved information on the amounts of sediment involved (i.e., loads and

* Corresponding author. Tel.: +44 1392263345; fax: +44 1392263342.

E-mail address: d.e.walling@exeter.ac.uk.

Table 1
Phosphorus (P) export from selected UK catchments and the proportion transported in particulate form (based on Withers et al., 1998)

River	Catchment area (km ²)	Total P export (kg ha ⁻¹ year ⁻¹)	Particulate (%)
Avon (Warwickshire)	2674	2.10	26
Severn	6850	1.62	43
Exe	601	1.64	68
Dart	46	1.87	75
Ouse	3315	2.07	55
Swale	381	0.84	33
Calder	899	6.40	34
Don	1320	0.93	67
Dee	2100	0.26	69
Ythan	689	0.73	79

concentrations) and on the changes in those amounts through time, consequent upon longer-term changes in land use and other facets of environmental change. However, it is also important to obtain information on the main sources of the transported sediment, since sediment source can exert a key control on both the physical and geochemical properties of fine sediment, including its P content, which in turn exerts a fundamental control over the magnitude of sediment-associated nutrient and contaminant fluxes.

The suspended sediment load transported by a river or stream will commonly represent a mixture of sediment derived from different locations and from different source types within the contributing catchment. Thus, for example, a relatively small area of the catchment, underlain by a particular rock type or supporting a particular land use, could contribute most of the suspended sediment load at the catchment outlet. Equally, in some catchments, sheet and rill erosion could dominate the sediment supply, whereas in others, channel erosion or gully erosion could represent the primary source. Information on sediment source is of fundamental importance in understanding the suspended sediment dynamics and the sediment budget of a catchment (e.g., Dietrich and Dunne, 1978; Trimble, 1983; Walling, 1988; Walling et al., 2001a, 2002).

Information on sediment source also represents a key requirement from the management perspective, since identification of sediment sources is a key precursor to the design of effective sediment manage-

ment and control strategies. Whereas soil conservation programmes are primarily concerned with controlling on-site soil loss from agricultural land, sediment control programmes are more concerned with downstream problems and must consider a wider range of potential sources. Resources could be effectively wasted if, for example, control measures focussed on reducing surface erosion, when most of the sediment transported through a river system was contributed by channel and gully erosion. As indicated above, sediment source can exert a fundamental control on the nutrient and contaminant content of fine sediment, since the source of the sediment is likely to influence its physical and chemical properties and its contaminant loading and any management strategy aimed at controlling sediment-associated nutrient and contaminant fluxes would again need to take account of sediment source. In the case of P, for example, it may be important to consider the P content of the various sources and its bioavailability, so that the primary sources of P linked to specific impacts can be targeted. Thus, although the dominant sediment source within a catchment might be channel bank erosion, the P content of this sediment is likely to be substantially lower and less bioavailable than that mobilised from the surface of agricultural land. From a P management perspective, it could therefore prove more effective to control sediment inputs from agricultural land. The precise type of information on sediment source required will depend on the purpose in hand and the nature of the sediment-related problem. However, information on both the source type (e.g., sheet and rill erosion of areas under different land use, gully erosion, channel erosion or mass movements) and the spatial location of the sources (e.g., which tributary or part/parts of the basin) will frequently be required to address the above issues.

1.2. Quantifying suspended sediment sources

Although the need for information on suspended sediment source is clear, it has proved less easy to assemble such data (see Collins and Walling, 2004). Traditional methods for assessing the relative importance of individual source types employ an indirect approach and involve visual observations or measurements of erosional activity, which are in turn used to infer the relative importance of different potential

sources. Thus, aerial photography could provide evidence of the incidence of channel and gully erosion and perhaps sheet and rill erosion (e.g., Eriksson et al., 2003), erosion pins could be employed to record the rate of surface lowering or retreat of features such as eroding river banks or gully walls (e.g., Haigh, 1977; Lawler et al., 1999; Stott, 1999) and erosion plots could be used to document rates of soil loss from surface sources (e.g., Soons, 1968; Loughran, 1990). However, this indirect approach faces many problems. Firstly, it will commonly require some a priori assumptions as to the likely sources, and in some environments, these may not be clear. Secondly, the use of erosion pins and erosion plots to provide information on the relative magnitude of erosion rates associated with different potential sources is difficult in anything but small drainage basins, due to the spatial variability of erosion and thus spatial sampling problems (c.f. Peart and Walling, 1988). Thirdly, and perhaps most importantly, the approach only provides information on sediment mobilisation and is unable to take account of the efficiency of sediment delivery to the stream system, for which the information on the source of transported sediment is ultimately required. As an alternative approach, some workers have attempted to infer sediment source contributions using models and prediction procedures. Thus, for example, the Universal Soil Loss Equation (USLE) could be used to estimate sheet and rill erosion from a small catchment and the difference between this estimate and the sediment yield could be attributed, at least in theory, to channel and gully erosion.

Attempts to obtain information on the spatial source of the sediment transported by a river commonly involves less uncertainty, since, theoretically, it would be possible to monitor the sediment load at a large number of points within a river network and therefore evaluate the relative importance of different tributaries or different parts of the catchment as sediment sources. However, this approach is commonly precluded by both practical and cost constraints. Furthermore, storage of sediment within the channel system could introduce problems in terms of relating the downstream flux to the fluxes from individual tributaries.

Faced with the many problems and constraints associated with the use of traditional approaches to obtaining information on sediment source and the growing need for such information, in the 1970s, a

number of workers attempted to exploit the potential of an alternative direct approach to quantifying suspended sediment sources, based on source tracing or ‘fingerprinting’ (e.g., Klages and Hsieh, 1975; Wall and Wilding, 1976; Walling et al., 1979). 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 equivalent values for potential sources, in order to identify the likely source of that sediment. Early work successfully used geochemical (e.g., Wall and Wilding, 1976), mineralogical (e.g., Klages and Hsieh, 1975) and mineral magnetic (e.g., Walling et al., 1979; Oldfield et al., 1985) properties for source fingerprinting. However, the scope of these studies was generally limited in terms of providing, firstly, only a qualitative indication of the likely importance of particular sources and, secondly, only broad discrimination between a small number of potential sources, typically surface and subsurface/channel bank materials.

2. The development of sediment source tracing or fingerprinting procedures

Subsequent development of the source tracing or fingerprinting approach directed attention to a number of important methodological aspects, with a view on refining the approach and improving the reliability of the results obtained. The first area of development involved the search for fingerprint properties that were capable of clearly discriminating several potential sources. Geochemical, mineralogical and mineral magnetic properties of soils and sediments continued to be used, but sediment colour (Grimshaw and Lewin, 1980), plant pollen (Brown, 1985), isotopic signatures (e.g., Douglas et al., 1995, 2003) and particularly the activity of fallout radionuclides, including caesium-137 (^{137}Cs), excess lead-210 (^{210}Pb) and beryllium-7 (^7Be) (e.g., Peart and Walling, 1986; Walling and Woodward, 1992; Olley et al., 1993; He and Owens, 1995; Wallbrink et al., 1996, 1998, 1999) also attracted attention. Fallout radionuclides were shown to be particularly valuable for distinguishing surface and subsurface materials, since concentrations are

commonly relatively high in the former and low or non-existent in the latter. Furthermore, they frequently also afford a means of distinguishing the surface horizons of cultivated and uncultivated soils, since ploughing and tilling of a soil mixes the radionuclide into the plough layer and produces lower surface concentrations than found in undisturbed soils, where the majority of the radionuclide inventory remains near the surface. In addition, fallout radionuclide concentrations are effectively independent of soil type and underlying geology and thus well-suited to use in heterogeneous catchments. It might be argued that the inclusion of fallout radionuclides in sediment fingerprinting exercises greatly increased the validity and scope of such work and provided the impetus necessary for the approach to be more widely adopted. In considering potential fingerprint properties, emphasis was initially frequently placed on the search for a single diagnostic property that would be capable of clearly discriminating a number of potential sources. However, the quest for such a property was soon recognised to be an elusive goal and attention switched to the identification and use of several properties, which together would provide a composite fingerprint capable of discriminating unequivocally between several potential sources (e.g., Walling et al., 1993).

Most early source fingerprinting studies were essentially qualitative in their results, showing, for example, which sources were likely to be most important and identifying temporal changes in the relative importance of these sources, both during storm events or between different seasons. The incorporation of quantitative mixing models, in association with composite fingerprints, into source fingerprinting studies in the late 1980s and early 1990s marked a second and major methodological advance, that made it possible to obtain quantitative estimates of the relative contributions from different sources (cf. Yu and Oldfield, 1989; Walling et al., 1993; Shankar et al., 1994; Walling and Woodward, 1995; Walden et al., 1997). By carefully selecting the composite fingerprint used and including a substantial number of fingerprint properties with contrasting origins, environmental behaviour and controls, it was possible to discriminate between several potential sources and to quantify their relative contributions to the sediment load of a stream.

The increased precision available to source fingerprinting studies through the use of quantitative mixing models also directed attention to the need to rigorously test the ability of individual fingerprint properties to discriminate between potential sources and to identify the optimum combination of sediment properties to include in a composite fingerprint. The resulting refinement of the technique is seen as representing a third key development. Cluster analysis coupled with analysis of variance (Walling and Woodward, 1995) and discriminant function analysis (Collins et al., 1996, 1997a,b) were successfully employed to identify the optimum combination of tracer properties to include in a composite fingerprint and the application of these procedures was frequently preceded by the use of statistical tests, such as the Kruskal–Wallis test, to confirm the ability of individual fingerprint properties to discriminate between potential sources and to thus be considered for inclusion in the final composite fingerprint (Collins et al., 1996, 1997a,b).

Other important developments and refinements in the source fingerprinting technique have included the need to take account of differences in grain size composition and organic matter content between source material and suspended sediment samples, recognition of the problems associated with the use of ratio properties in multi-component mixing models, the need to consider the precision of the laboratory analyses employed for individual sediment properties when optimising the mixing model and incorporation of uncertainty analysis into the computation procedures. Corrections for differences in particle size composition and organic matter content between suspended sediment and potential source materials are clearly an important consideration in obtaining reliable results from any fingerprinting study, since it is well known that the concentrations of many soil and sediment properties are influenced by the grain size and organic matter content of the sample (Horowitz, 1991). Thus, for example, the sediment mobilised from a particular source could have different properties from those of the source material, if the mobilisation process was size selective and the mobilised sediment was, for example, enriched in fines. Procedures used to take account of these differences have varied in complexity. Simple approaches have, for example, involved analysing

only the <0.063-mm fraction of the samples, thereby restricting attention to the dominant size class of the suspended sediment (see Dyer, 1998; Wallbrink et al., 1998) and correcting concentrations by using the ratio of the specific surface areas of the suspended sediment and source material samples (see Collins et al., 1997a,b, 1998; Gruszowski et al., 2003). More complex approaches have included incorporation of more detailed information on the precise relationship between grain size composition and the concentration of individual properties into the correction procedure (e.g., He and Walling, 1996; Russell et al., 2001) or derivation of adjusted source material concentrations, by using information on the grain size composition of the suspended sediment and the property concentrations associated with particular size fractions of the source material, to estimate the property concentration associated with source material with the same grain size composition as the suspended sediment (e.g., Walling and Woodward, 1992; Slattery et al., 1995; Motha et al., 2003). Less attention has been given to the need to undertake similar corrections to take account of differences in the organic matter content of source material and suspended sediment, partly because enrichment in organic matter is closely linked to enrichment in fines. However, several workers have attempted to address this consideration. Collins et al. (1997a,b, 1998) used a simple ratio between the organic matter content of source material and sediment, whereas Motha et al. (2003) measured the property concentrations associated with the organic fraction and adjusted source material concentrations to reflect a similar organic matter content to that in suspended sediment.

In the search for fingerprint properties able to readily distinguish between a range of sources, attention was frequently directed to property ratios. Such ratios also offered potential to overcome contrasts in grain size composition between source material and sediment samples, since they are likely to be insensitive to such contrasts. However, it is now recognised that property ratios should not generally be used in simple linear mixing models, since they are not linearly additive. In view of the need to use such mixing models to obtain quantitative estimates of source contributions, their value is therefore less clear. The varying precision of laboratory analyses of individual sediment properties has also been taken

into account by some workers when optimising a mixing model, since greater reliance should arguably be placed on those fingerprint properties which have the greatest precision (cf. Collins et al., 1997a,b, 1998). The application of optimisation procedures to the mixing model used to establish the proportions of the total sediment load derived from a set of potential sources may also introduce problems of equifinality, in that several different parameter combinations could possibly produce the same goodness of fit. Recognition of this problem and the uncertainties introduced by the natural variability of source material properties has prompted the application of Bayesian statistics, Monte Carlo routines and other uncertainty analysis to the model fitting procedure (e.g., Rowan et al., 2000; Motha et al., 2003; Small et al., 2002; Douglas et al., 2003).

Source fingerprinting procedures have now been successfully used in many studies, to obtain detailed and reliable information on suspended sediment sources, and their application is becoming increasingly accepted as affording a valuable and essentially unique basis for assembling information on catchment suspended sediment sources. The approach has been applied in both small and large river basins (e.g., Slattery et al., 1995; Russell et al., 2001; Collins et al., 1996; Wallbrink et al., 1996; Douglas et al., 2003) and for establishing the contribution of individual sources defined in terms of both source type and spatial location (e.g., Collins et al., 1997a). Furthermore, its use has also been extended to include sediment sources in urban catchments (Carter et al., 2003), employing recent floodplain sediments as an alternative to suspended sediment samples in determining the primary sediment sources within a catchment (e.g., Bottrill et al., 1999) and to analysis of lake sediment and floodplain cores to elucidate recent changes in sediment sources within river basin cores (e.g., Foster and Walling, 1994; Owens et al., 1999; Owens and Walling, 2002; Foster et al., 2003).

Within the UK, the fingerprinting approach has been increasingly used to assemble much needed information on catchment suspended sediment sources, particularly by the author and his co-workers. Several examples of the studies undertaken, the methods employed and the results obtained are presented below, in order to demonstrate the utility of the approach.

3. UK case studies

3.1. Sediment sources in the Upper Torridge catchment

The Upper Torridge catchment in Devon, UK, drains an area of 258 km² above the Environment Agency flow gauging station at Rockhay Bridge. With its moderate relief (maximum altitude 220 m), relatively high annual precipitation and runoff (ca. 1250 mm and 900 mm, respectively) and heavy soils, the land use of the catchment is dominated by pasture (ca. 80%) with arable land and woodland accounting for ca. 16% and 4%, respectively. Livestock grazing (cattle and sheep) constitutes the main farming activity. The River Torridge is a well-known salmon river, but in recent years the salmon stocks have shown a marked decline. The siltation of salmon spawning redds has been identified as a cause of declining salmon numbers and this has in turn directed attention to the relatively high suspended sediment yields of catchments in this area of Devon. The fingerprinting approach was used to investigate suspended sediment sources in the Upper Torridge catchment during 1997 and 1998 as part of a wider investigation of spawning gravel siltation within the catchment (see Nicholls, 2001). The results of the source fingerprinting exercise provide an interesting perspective on both the fine sediment dynamics of the catchment and the influence of land use on the suspended sediment output from the catchment, as well as the potential for developing a sediment control strategy to reduce suspended sediment fluxes.

The source tracing investigation focussed on source types, rather than spatially differentiated sources, and four potential sources were identified. These comprised channel banks, and the surface of areas under cultivation, pasture and woodland. In order to derive composite fingerprints for these four potential sources, representative samples of river bank sediment and surface soil from the three land use types were collected from different areas of the catchment. In all, 170 samples were collected, comprising 50 samples each from areas under cultivation and pasture, 40 samples from channel banks and 30 samples from woodland areas. These samples were air-dried, disaggregated and passed through a 0.063-mm sieve prior to laboratory analysis.

Bulk suspended sediment samples were collected from the flow gauging station at Rockhay Bridge during a representative selection of high flow events, in order to provide information on the properties of the suspended sediment load of the river, that could be compared with those of the potential sources. The samples (ca. 100 l) were collected using a pump sampler and stored in 20-l plastic containers, which were transported to the laboratory. There, the sediment was recovered by continuous flow centrifugation, freeze dried and sieved to <0.063 mm prior to laboratory analysis.

Selection of fingerprint properties for use in the investigation was based on previous experience of source discrimination, as well as being constrained by available analytical facilities and the time available for analytical work. The 19 properties finally selected (see Table 2) comprised a range of heavy metals, trace metals and base cations, organic carbon, nitrogen (N), two fallout radionuclides (¹³⁷Cs and excess ²¹⁰Pb) and one geogenic radionuclide (radium-226 (²²⁶Ra)). The grain size distribution of both the source material and suspended sediment samples was determined using a

Table 2

Results of the Kruskal–Wallis *H*-test applied to the measurements of fingerprint properties made on the potential source materials collected from the Upper Torridge catchment (based on Nicholls, 2001)

Potential fingerprint property	<i>H</i> value
Nitrogen	60.84*
Carbon	61.46*
Aluminium	0.61
Calcium	40.85*
Chromium	47.91*
Cobalt	24.81*
Copper	8.40*
Iron	44.66*
Lead	14.78*
Magnesium	5.12
Manganese	20.17*
Nickel	24.69*
Potassium	52.16*
Strontium	30.51*
Sodium	10.37*
Zinc	10.43*
Caesium-137	50.15*
Radium-226	55.51*
Excess lead-210	48.44*
Critical Value	7.82

* Difference significant at *P*=0.05.

Coulter LS130 laser granulometer, after removal of the organic matter and chemical and ultrasonic dispersion. In addition to measuring the grain size distribution of each sample, this equipment also provided an estimate of its specific surface area, assuming spherical particles.

A two-stage statistical procedure was used to identify the optimum set of source material properties for use as composite fingerprints (cf. Collins et al., 1997a,b). First the non-parametric Kruskal–Wallis *H*-test was used to test which properties exhibited significant differences between the individual source types. Secondly, stepwise multivariate discriminant function analysis was applied to the list of properties selected in the first stage, in order to identify the set of properties or composite fingerprint that afforded optimum discrimination between source groups. The results of this statistical analysis are presented in Tables 2 and 3. Table 2 indicates that, with the exception of aluminium (Al) and magnesium (Mg), all the fingerprint properties initially selected evidenced statistically significant differences between the four source types. Table 3 indicates that the final composite fingerprint comprising seven properties was capable of allocating 100% of the source samples to the correct source type and therefore provided a powerful means of discriminating between the potential sources. Each source type was characterised by the mean concentration for each of the seven fingerprint properties, on the assumption that sediment contributed by a particular source type within the catchment could be expected to be characterised by a concentration close to that of the mean for the representative samples collected from that source type, since the sediment

would be mobilised from many different locations within the catchment.

A multivariate mixing model based on that employed by Collins et al. (1996) and Walling et al. (1999a) was subsequently used to estimate the relative contribution of the potential sediment sources to a particular suspended sediment sample. In this method, the proportions *P* contributed by the *m* individual sources *s* are established by minimising the sum of the squares of the residuals (*R*_{es}) for the *n* tracer properties involved, where:

$$R_{es} = \sum_{i=1}^n \left(\frac{C_{ssi} - \left(\sum_{s=1}^m C_{si}P_s \right)}{C_{ssi}} \right)^2 \quad (1)$$

and *C*_{ssi} is the concentration of tracer property *i* in the suspended sediment sample, *C*_{si} is the mean concentration of tracer property *i* in source group *s* and *P*_{*s*} is the relative proportion from source group *s*.

To ensure that equal weight is given to the individual fingerprint properties included in the linear equations within the mixing model and thus contributing to the overall sum of squares of the residuals, all property concentrations were scaled to the range 0–1. The effects of contrasts in grain size composition between these different materials were partly addressed by restricting analysis to the <0.063-mm fraction, but further correction was introduced by using the ratio of the specific surface area of the suspended sediment to that of the mean for the individual source types. No corrections were made for differences in organic matter content between source materials and suspended sediment, because the relationship between organic matter content and element concentration is complex and difficult to generalise.

The goodness of fit provided by the optimised mixing model was assessed by comparing the actual fingerprint property concentrations for the suspended sediment samples with the corresponding values predicted by the mixing model, based on the estimates of the magnitude of the contributions from each of the sources. The mean (average for all properties within each composite fingerprint) relative errors for the mixing model calculations were typically around 10%, confirming that the relative contributions from the individual source types generated by the mixing model were meaningful.

Table 3
The results of the stepwise discriminant function analysis used to identify the optimum composite fingerprint for distinguishing the four source types (based on Nicholls, 2001)

Fingerprint property added	Wilks' lambda	Cumulative percent of samples classified correctly
Nitrogen	0.19443	69.74
Potassium	0.06426	89.47
Caesium-137	0.02351	98.41
Carbon	0.01355	98.41
Chromium	0.00864	95.24
Iron	0.00631	95.24
Radium-226	0.00499	100.00

Fig. 1 summarises the source ascription results obtained for the 16 bulk suspended samples collected from the River Torridge at Rockhay Bridge during the study period. The substantial variability in the contributions from the four sources, between the individual samples, is a key feature of the results. There is, however, a reasonable degree of consistency in the magnitude of the contribution from bank erosion, which typically ranges between 20% and 40%, with a mean of 26%. The variation between the samples undoubtedly reflects both intra- and inter-event and seasonal variability in the importance of the bank erosion contribution and the timing of the sample relative to the routing of sediment from different parts of the catchment. In the case of inter-event variability in the importance of bank erosion, it is important to recognise that sediment supply from bank erosion will reflect both natural controls (e.g., antecedent conditions and flow magnitude) as well as the more adventitious effects of livestock in trampling and degrading the river banks. Furthermore, it must be recognised that changes in the relative magnitude of the contribution from any one source could also

reflect changes in the magnitude of the contributions from the other sources rather than a real change in the absolute contribution from a particular source. Fig. 1 indicates that the contribution from woodland areas is also consistently very low, reflecting both the relatively small proportion of the catchment occupied by woodland and the lack of surface runoff and sediment generation commonly associated with areas of woodland and forest. Greater variations between the samples are evident for the contributions from cultivated and pasture areas. The contribution from pasture sources ranges from 2% to 85% for the individual samples, although the contribution from this source dominates the majority of samples. The importance of pasture areas as a sediment source partly reflects the high proportion of the catchment occupied by this land use, but can also be linked to the high stocking densities and poaching of the heavy soils, particularly during the wetter months of the year. The contrasts in the relative importance of the contributions from pasture and cultivated areas to the individual samples again reflect seasonal and inter-event variations (e.g., event magnitude) in sediment

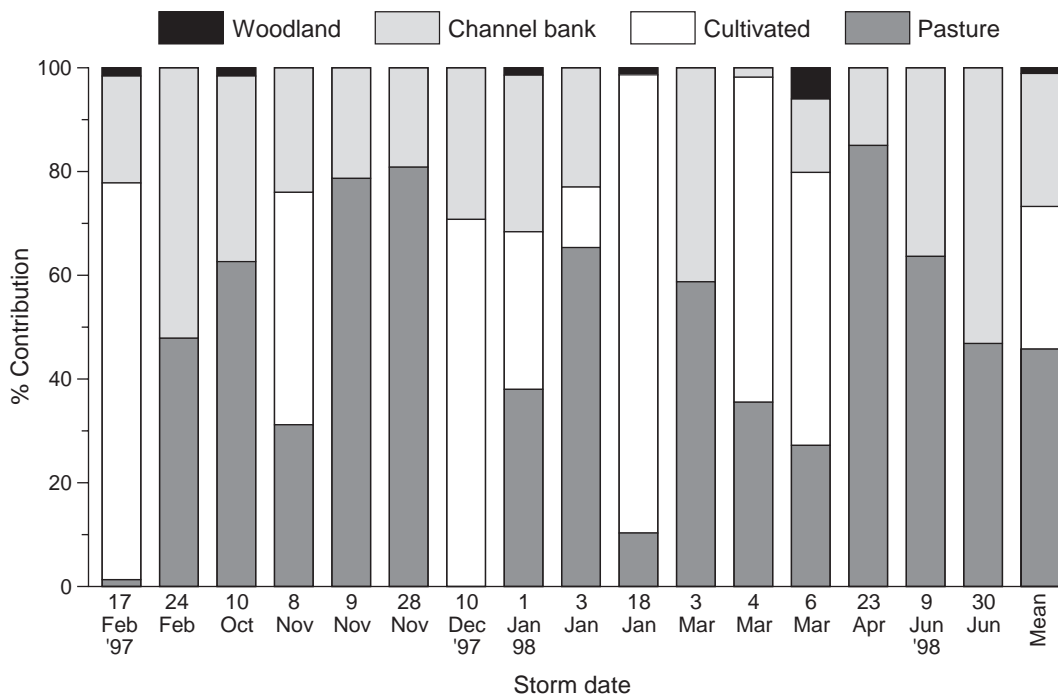


Fig. 1. Source contributions to bulk suspended sediment samples collected from the River Torridge at Rockhay Bridge, Devon, UK (based on Nicholls, 2001).

mobilisation from the two land use types and, probably more importantly, the timing of the sample collection during an event and its relation to the routing of runoff and sediment from different parts of the catchment through the catchment outlet, where the suspended sediment samples were collected.

More samples would ideally be required to generate an estimate of the precise overall contribution of the four potential sources to the annual or study period suspended sediment load of the Upper Torridge catchment, but a ‘best-estimate’ can be provided by taking account of the different magnitudes of the sampled events and calculating a load-weighted mean contribution, based on the suspended sediment load at the time of sampling (derived as the product of the instantaneous values of discharge and suspended sediment concentration) viz.

$$P_{sw} = \sum_{x=1}^n P_{sx} \left(\frac{L_x}{L_t} \right) \quad (2)$$

where P_{sw} is the load-weighted relative contribution from source type s , L_x (kg s^{-1}) is the instantaneous suspended sediment load for suspended sediment sample x , L_t (kg s^{-1}) is the sum of the instantaneous loads (L_x) associated with the n sediment samples collected from the sampling site and P_{sx} is the relative contribution from source type s for sediment sample x . This load-weighted approach (cf. Walling et al., 1999b) provides a more realistic estimate of the proportion of the total suspended sediment load passing the sampling site contributed by individual sources, than a simple average of the percentage contribution values associated with individual suspended sediment samples, some of which may represent periods with relatively low suspended sediment loads that contribute only a small proportion of the longer-term suspended sediment yield.

The results of the calculations of the load-weighted mean contributions from the four sources presented in Table 4 indicate that surface erosion from pasture areas represents the dominant sediment source in the Upper Torridge catchment, but that both surface erosion from cultivated areas and channel erosion also represent important sources. The importance of channel erosion, which includes ditches and ephemeral channels incised into the subsoil/regolith, reflects a number of factors, including the high number of actively eroding banks on the main river, the

Table 4

Load-weighted mean contributions of each potential source type to the suspended sediment samples collected from the River Torridge at Rockhay Bridge (based on Nicholls, 2001)

Source type	Contribution (%)
Woodland topsoil	2
Pasture topsoil	47
Cultivated topsoil	28
Channel banks	23

trampling and degradation of the banks by livestock using the river for water, the high drainage density of the catchment under wet conditions, the ditches associated with agricultural drainage systems and the flashy discharge response of the catchment. Taking account of the estimate of the annual suspended sediment yield of the catchment based on the records assembled for the study period, i.e., $89 \text{ t km}^{-2} \text{ year}^{-1}$, the specific suspended sediment yield attributable solely to bank erosion is ca. $20 \text{ t km}^{-2} \text{ year}^{-1}$. This is equivalent to ca. 50% of the typical suspended sediment yield of a UK catchment (cf. Walling and Webb, 1987), a fact that further emphasises the importance of bank erosion as a sediment source in the study catchment.

In assessing further the magnitude and significance of the contributions from bank erosion and from pasture and cultivated areas, it is again important to take account of the proportions of the catchment area supplying these contributions and thus the equivalent suspended sediment yields from these areas. Based on the proportions of the catchment occupied by pasture and arable areas (i.e., 80% and 16%, respectively), the specific suspended sediment yields from these two areas may be estimated to be ca. $52 \text{ t km}^{-2} \text{ year}^{-1}$ from pasture areas and ca. $155 \text{ t km}^{-2} \text{ year}^{-1}$ from arable areas. The contrast between arable and pasture areas is consistent with existing understanding of the relative rates of soil loss from the two land use types, with pasture areas evidencing much lower rates of soil loss, due to the greater vegetation cover density. However, both values must be seen as relatively high when compared with existing information on the specific suspended sediment yields of UK catchments, and taking into account that the total sediment yield from a catchment is likely to include an appreciable additional contribution from channel erosion sources. These relatively high sediment yields are, however, consistent with existing knowledge of sediment

mobilisation within the Upper Torridge catchment. Increased rates of sediment mobilisation from pasture areas can be readily linked to the high stocking densities on the heavy soils, which are frequently waterlogged in winter and on which poaching is a common occurrence. Equally, the increased rates of sediment mobilisation from arable areas reflect, at least in part, the importance of fodder maize as an arable crop. Rates of soil loss from maize fields are frequently high, since the crop is generally harvested in mid-autumn, when the soils have wetted up and are readily compacted by the heavy harvesting machinery, and the fields, which are frequently left bare until the following spring, are a major source of surface runoff and soil loss (cf. Walling et al., 1999a). Furthermore, the heavy soils of the Upper Torridge catchment are characterised by high drainage densities and relatively high rates of surface runoff under wet conditions in winter, when most sediment mobilisation occurs. These two factors combine to increase slope-channel connectivity and associated sediment delivery ratios for arable fields, which are often bare or with a low cover density in winter (cf. McHugh et al., 2002; Walling and Zhang, 2004).

3.2. The role of field drains as a sediment source and transfer pathway

In the study of sediment sources in the Upper Torridge catchment introduced above, emphasis was placed on a broad distinction between surface erosion under different land use classes and channel or subsurface erosion. Although it was known that underdrainage systems existed in many areas of the catchment, no explicit attempt was made to identify sediment that had passed through the tile drain systems. It was assumed that this was either of surface or subsurface origin and it was therefore subsumed within those source types. In some investigations, however, there may be a requirement to assess the relative importance of field drains as either a sediment source or pathway. Such information could be assembled by monitoring the outlets of drains within a catchment and comparing the sediment flux from the drains with that at the catchment outlet. However, this is likely to prove a costly and labour intensive process and in many catchments both the location of, and the areas contributing to, the drains

may be poorly documented. Source fingerprinting offers an alternative approach, since if sediment issuing from the drains can be clearly distinguished, in terms of its properties, from sediment derived from other sources, it is possible to assess the contribution of sediment discharged from drains to the sediment load at the catchment outlet. A study undertaken in a small catchment in Herefordshire, reported by Russell et al. (2001), which used this approach, provides a useful example of its potential.

The 1.5-km² Rosemaund catchment (see Fig. 2), a headwater tributary of the River Lugg, comprises part of the ADAS Rosemaund Research Centre and has provided the focus for intensive investigations of P and sediment fluxes (cf. Hodgkinson and Withers, 1996; Johnes and Hodgkinson, 1998). The catchment is characterised by gentle slopes (<5°) and is underlain at a depth of ca. 1–3 m by impermeable mudstones. The soils are primarily light silty clay loams of the Bromyard series (argillic brown earths), with some small areas of Middleton (stagnogleyic argillic brown earths) and Compton series (pelo-alluvial gley soils) at the base of slopes, in hollows and in the stream corridor (cf. Fig. 2c). Further details of the soil hydrology are provided by Williams et al. (1996). The mean annual precipitation for the locality is ca. 600 mm. Most of the catchment is given over to arable cultivation and hopyards, although there are several fields occupied by longer-term temporary pasture (cf. Fig. 2b). The catchment is extensively underdrained (92% of the catchment area) and there are at least 22 known outfalls in the catchment. Stream monitoring and sampling stations have been installed at the outlets of the main catchment (Belmont) and an upstream subcatchment (Jubilee, 0.31 km²). The outfalls of three subsurface tile drains are also monitored at Foxbridge, Longlands and Moorfield (see Fig. 2b). Available data indicate that the mean annual suspended sediment yield at the Belmont sampling station is ca. 82.0 t km⁻² year⁻¹ (cf. Walling et al., 2002).

In order to characterise potential suspended sediment sources in the catchment, 100 samples of potential source material were collected from the catchment in 1997. These comprised samples of channel bank material and of surface material (0–2 cm depth). The latter were collected to be representative of material likely to be mobilised by surface erosion processes and of the range of land use and soil

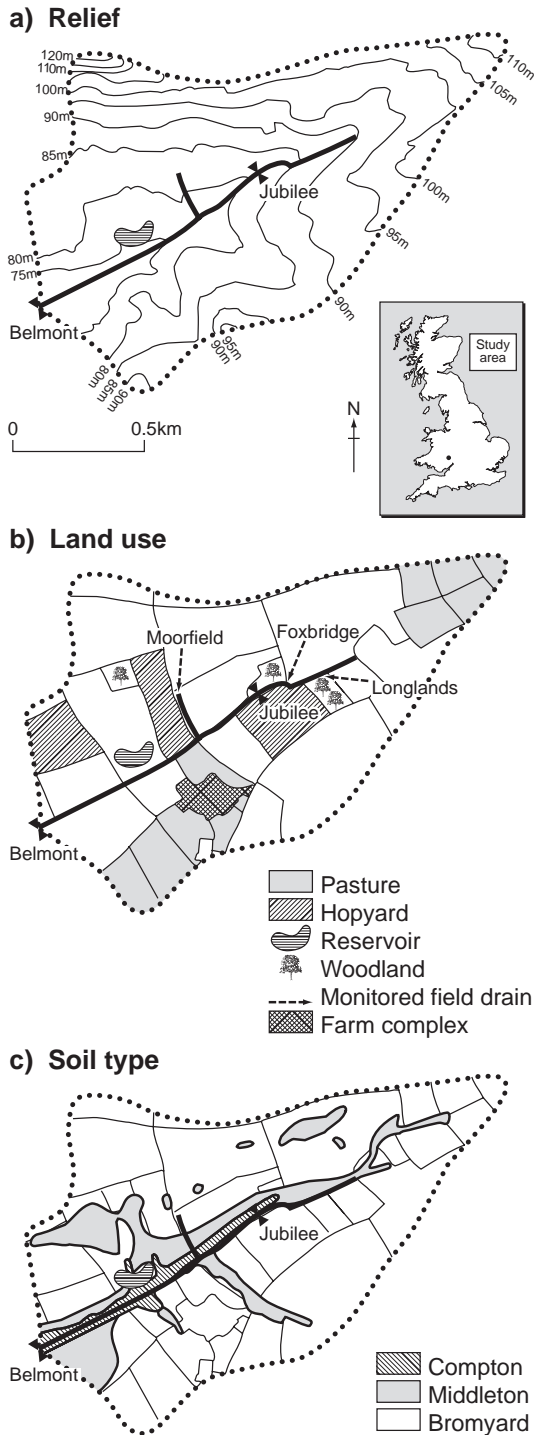


Fig. 2. The Rosemaund catchment showing the location, the topography and flow gauging station sites (a), the land use and monitored drains (b) and the distribution of soil types (c).

types. The source samples were oven dried at 40° and disaggregated prior to analysis. Bulk suspended sediment samples (50–200 l) were also collected from the monitored drains during storm events during the period 1997–1999, in order to characterise sediment discharged from the drain systems within the catchment. Bulk suspended sediment samples (50–200 l) for characterising the suspended sediment output from the catchment were collected from the Jubilee (74 samples) and Belmont (57 samples) monitoring stations over a range of discharge conditions, again during the period 1997–1999. The sediment was recovered from both the stream and drain samples by continuous flow centrifugation and freeze dried prior to analysis.

The source material and drainflow sediment samples were analysed for a range of potential geochemical and radiometric fingerprint properties, similar to those employed in the Upper Torridge investigation described above. However, these also included free-oxide forms of iron (Fe), aluminium (Al) and manganese (Mn) (Bascomb, 1986). In addition, several mineral magnetic properties including low frequency and frequency dependent susceptibility (χ_{lf} and χ_{fd}), anhysteretic remanent magnetisation (ARM) and two values of isothermal remanent magnetism (SIRM and IRM at -0.1 T) were also measured (see Russell et al., 2001). In total, 32 properties were considered. A similar two-stage selection procedure to that employed in the Upper Torridge study was used to confirm the ability of these fingerprint properties to discriminate between both the bank material and surface samples and the samples of drainflow sediment. This procedure was applied to both the Belmont and the Jubilee catchments. Two approaches were used to classify further the surface samples for these discrimination tests. The first was based on land use (i.e., pasture, arable and hopyards) and the other on soil type (i.e., Bromyard, Middleton and Compton series). Two sets of source types were therefore considered. The first comprised channel banks, field drains and surface soils under pasture, arable and hopyards, whereas the second comprised channel banks, field drains and surface soils from areas with soils of the Bromyard, Middleton and Compton series. The results of the stepwise multiple discriminant function analysis, which are summarised in Table 5, demonstrate that it is possible to identify composite fingerprints that are able to discriminate

Table 5

The composite fingerprints selected using stepwise discriminant function analysis for the two source type sets for the Belmont and Jubilee catchments (based on Russell et al., 2001)

Catchment	Source set	Parameters*	Discrimination (%)
Belmont	Land use	Al _p , Fe, Mg, Mn, ¹³⁷ Cs, K, χ_{if} , ARM, SIRM	87.4
	Soil type	Al _p , SIRM, ARM, ¹³⁷ Cs, χ_{if} , Pb, Mg, K, Fe, Mn	88.3
Jubilee	Land use	¹³⁷ Cs, As, N, ARM, SIRM, Pb, χ_{if} , C	89.4
	Soil type	K, Mg, As, Mn, ¹³⁷ Cs, χ_{if} , ARM, SIRM	91.1

SIRM—saturated isothermal remanent magnetisation.

* List in order of inclusion into the composite fingerprint.

successfully between the five potential sources in each source type set. These composite fingerprints comprised between eight and 10 properties.

Once the composite fingerprints had been established, measurements of the properties involved were undertaken on the suspended sediment samples collected from the Belmont and Jubilee catchments. The multivariate mixing model (Eq. (1)) was then used to estimate the relative contributions from the five sources comprising the two source sets. In view of the significant differences in particle size composition between source materials and the drainflow and stream suspended sediment samples, a detailed grain size correction procedure, based on the relationship between fingerprint property concentrations and specific surface area was applied to the data (see Russell et al., 2001). The results of the source ascription exercise, expressed as load-weighted mean contributions from the various potential sources, presented in Table 6, evidence a strong degree of consistency both between the results provided by the two source sets and by the two catchments and indicate that the drains contribute about 50% of the total sediment output from the two catchments. This value is in turn consistent with available evidence of the importance of the drain contribution, based on measurements of the sediment output from the drain systems and comparisons with the catchment sediment yields. The major contribution of the drains to the overall sediment yield from the two catchments has important implications for the development of sediment management strategies in lowland catchments (cf. Chapman et al., 2001), since it would seem that land drainage is likely to increase the overall sediment flux from a catchment by increasing the connectivity between the catchment surface and the channel network. Existing evidence suggests that

much of the sediment discharged from the drain systems in the Rosemaund catchment originates from the catchment surface and passes from the surface to the drains through desiccation cracks and other macropores. Further scope clearly exists to assess the relative importance of different soil horizons in contributing to the sediment flux from a drain system, by using the fingerprinting approach to discriminate between sediment derived from different soil horizons (cf. Hardy et al., 2000).

Table 6

Load-weighted mean contributions of the potential sediment sources to the suspended sediment samples collected from the Belmont and Jubilee monitoring stations, estimated using the mixing model applied to the two source type sets (based on Russell et al., 2001)

Catchment and	Source set	Source	Contribution (%)
Belmont	Land use	Pasture	3.9
		Arable	17.5
		Hopyard	12.2
		Channel banks	11.1
		Field drains	55.3
Belmont	Soil type	Bromyard	12.9
		Middleton	11.8
		Compton	8.9
		Channel banks	11.9
		Field drains	54.5
Jubilee	Land use	Pasture	3.1
		Arable	30.1
		Hopyard	7.0
		Channel banks	12.0
		Field drains	47.8
Jubilee	Soil type	Bromyard	3.7
		Middleton	30.5
		Channel banks	11.1
		Field drains	54.7

3.3. Spatial source contributions within the catchment of the Yorkshire Ouse

The case studies of the Upper River Torridge and the Rosemaund catchments provided above focus on source types. Such information is likely to prove of greatest value when establishing a catchment sediment budget or developing sediment control or management strategies. However, the source fingerprinting approach has also been used in the UK to assess the contributions to the total suspended sediment flux

from different zones of a catchment or different subcatchments (see Walling and Woodward, 1995; Collins et al., 1997a; Walling et al., 1999b; Owens et al., 2000; Russell et al., 2001). A good example of this application is provided by the study reported by Walling et al. (1999b), which used a similar approach to that described above to assess the relative contributions from the three main geological/topographic zones within the 3315-km² catchment of the Yorkshire Ouse (see Fig. 3). The three main geological zones, namely those underlain by Carboniferous, Permian

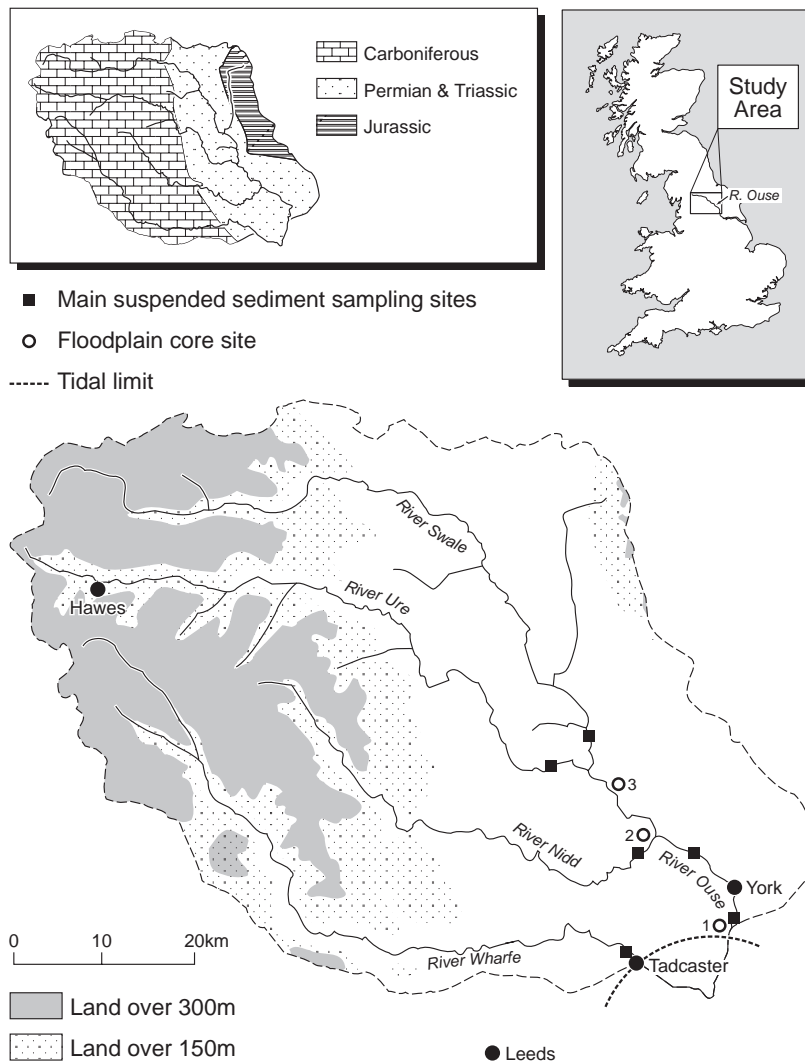


Fig. 3. The catchment of the River Ouse, Yorkshire, UK, showing the location of the suspended sediment sampling and floodplain coring sites and the main geological subdivisions.

and Triassic, and Jurassic strata, broadly correspond to the three main topographic zones in the catchment, namely the uplands of the Pennines (Carboniferous), the Vale of York (Permian and Triassic) and the uplands of the North York Moors (Jurassic). Although in places the underlying solid geology is covered by glacial drift deposits, most of these deposits are located in the Vale of York and they have been subsumed into the general geological zonation.

As with the previous studies, representative samples of the main potential source types were collected from within the catchment. In this case, three broad sources were identified (i.e., channel bank material and surface material from areas of cultivated and non-cultivated land). The ca. 160 source material samples were then classified according to the underlying geology of the areas from which they had been collected and attention focussed on establishing composite fingerprints capable of discriminating the three main geological zones. Again, a range of geochemical and radionuclide measurements were considered as potential fingerprint properties, and three mineral magnetic parameters namely the low frequency (χ_{lf}) and frequency dependent (χ_{fd}) magnetic susceptibility and the saturated isothermal remanent magnetisation (SIRM) were also included. The Kruskal–Wallis test and stepwise discriminant function analysis were employed to select the optimum composite fingerprint, which in this case included eight properties, namely, nitrogen (N), Mn, Mg, potassium (K), strontium (Sr), nickel (Ni), χ_{lf} and χ_{fd} . Bulk suspended sediment samples, representative of a range of high flow conditions, were collected during the period November 1994 to February 1997 from downstream sampling sites on both the main Ouse (30 samples) and from its major tributaries, namely, the Swale (19 samples), Ure (14 samples), Nidd (14 samples) and Wharfe (7 samples) at the sites identified in Fig. 3. The source contributions to these individual samples were determined by optimising the mixing model presented as Eq. (1), and the load-weighted mean contributions from each geological source are listed in Table 7, along with the proportions of the individual geological zones comprising each catchment.

The results presented in Table 7 indicate that the relative contributions from the individual geological zones within each of the catchments are broadly

Table 7

Load-weighted mean contributions of each geological source group to the suspended sediment samples collected from the main downstream sampling stations on the River Ouse and its tributaries (based on Walling et al., 1999b)

River	Geological source group contribution (%)		
	Carboniferous	Permian and Triassic	Jurassic
Swale	28.6 (39)	36.6 (41)	34.8 (20)
Ure	55.2 (79)	44.8 (21)	0 (0)
Nidd	75.9 (67)	24.1 (33)	0 (0)
Ouse	23.8 (50)	41.4 (40)	34.8 (10)
Wharfe	90.6 (90)	9.4 (10)	0 (0)

Values in parentheses represent the approximate percentage of the catchment underlain by each geological group.

consistent with the proportions of the catchments occupied by these zones and therefore that there are no major contrasts between the zones in terms of sediment yield. However, the results from the Swale, Ure and Ouse indicate that the contributions from their upper catchments, which lie within the Pennine Hills and which are underlain by Carboniferous strata, are significantly less than might be expected from their spatial extent, suggesting that the sediment yield from at least part of the Carboniferous zone is relatively low. Equally, the contributions from the areas of the Jurassic/North York Moors zone, lying within the catchments of the Swale and Ouse, the only two catchments extending into this zone, are significantly higher than might be expected from their spatial extent, and indicate that the sediment yields from this zone are relatively high.

3.4. Using floodplain cores to reconstruct past changes in sediment source contributions within the catchment of the Yorkshire Ouse

Since the overbank deposits of fine sediment found on river floodplains can provide a record of the properties of fine sediment transported by the river in the recent past (e.g., Walling et al., 2000; Owens et al., 1999; Owens and Walling, 2002, 2003), it is possible to use sediment cores retrieved from floodplains in the lower reaches of a catchment to provide information on longer-term (i.e., 50–100 years) changes in fine sediment sources within the upstream catchment. If a chronology can be established for a core, the timing of the changes in source contribution can be defined. It is, however, important to recognise that the fine

sediment associated with overbank floodplain deposits is unlikely to be directly equivalent to the suspended sediment load, since the deposited sediment is likely to be coarser than the suspended sediment. This approach to investigating longer-term trends in sediment sources is applicable to both source types and spatial sources and has been successfully applied in the Yorkshire Ouse and Tweed catchments (Owens et al., 1999, 2000; Walling et al., 2003b).

As part of an investigation of longer-term trends in the relative importance of the main source types, undertaken in the Yorkshire Ouse catchment (cf. Owens et al., 1999), three cores were collected from the floodplain bordering the River Ouse, downstream of its confluence with the River Nidd (see Fig. 3) and these cores were sectioned into 2-cm depth increments. Measurements of ^{137}Cs and excess ^{210}Pb activities in the cores were used to provide chronologies for the cores. To investigate downcore changes in source type, it was necessary to select fingerprint properties for which downcore variations would reflect changes in suspended sediment source, rather than other factors such as the age of the sediment. For this reason, it was not possible to include ^{137}Cs and excess ^{210}Pb in the suite of potential fingerprint properties considered, and metals, such as Pb and Zn that were sensitive to the history of metal mining in the upstream catchment were also excluded. As a preliminary filter, the correlations between fingerprint property concentrations and depth were considered. Where strong correlations with depth existed, this suggested that the downcore variations in the fingerprint property may have been influenced by either the age of the sediment or the depth of burial and these properties were not used for fingerprinting. Once the suite of 16 potential fingerprint properties was selected, the data available from the source materials previously collected from the catchment for studies of contemporary sediment sources (see above) were used to test their ability to discriminate between the different sources and to identify the optimum composite fingerprint. In this exercise, source type was defined quite broadly in terms of two possible sources, namely, topsoil and channel bank/subsoil, since the reduced number of potential fingerprint properties reduced the level of discrimination possible. Since only two potential sources were distinguished, the Mann–Whitney test was used to assess the ability of the individual

fingerprint properties to distinguish the two source categories and to produce a preliminary selection of the 16 potential fingerprint properties. These were then input into the stepwise discriminant function analysis used to establish the optimum composite fingerprint for discriminating between the two potential sources. The result of the discriminant function analysis presented in Table 8 shows that the composite fingerprint used to establish the contributions of the two source types included seven fingerprint properties and produced a high level of discrimination between the two potential source types.

The results of using the composite fingerprints listed in Table 8 in a mixing model to establish downcore variations in sediment source type are presented in Fig. 4. Tentative dates have been appended to several depths within the vertical profiles, based on age–depth relationships derived from ^{137}Cs and excess ^{210}Pb profiles in the sediment cores. The downcore variations in the contribution of the two primary source types shown in Fig. 5 indicate that significant changes in the suspended sediment sources have occurred in the catchment of the River Ouse over the past 100 years or so. Fig. 5 provides evidence of a general increase in the contribution from topsoil sources from 1900 through to about 1960, with some evidence of a decline after this date. The increase can be readily linked to the increasing intensity of agricultural land use during the twentieth century (e.g., expansion of arable cultivation, use of heavier machinery and increased stocking densities). The apparent reversal of this trend after the 1960s is less easy to explain, but it may reflect the introduction of improved land management practices

Table 8

The results of using stepwise discriminant function analysis to identify the optimum combination of tracer properties for use as a composite fingerprint for discriminating source types within the catchment of the Yorkshire Ouse (based on Owens et al., 1999)

Tracer property	Cumulative percent of samples classified correctly
Z _{ir}	61.5
Iron	79.1
Potassium	80.9
Strontium	84.6
Z _{fd}	83.6
Manganese	84.6
Nickel	90.0

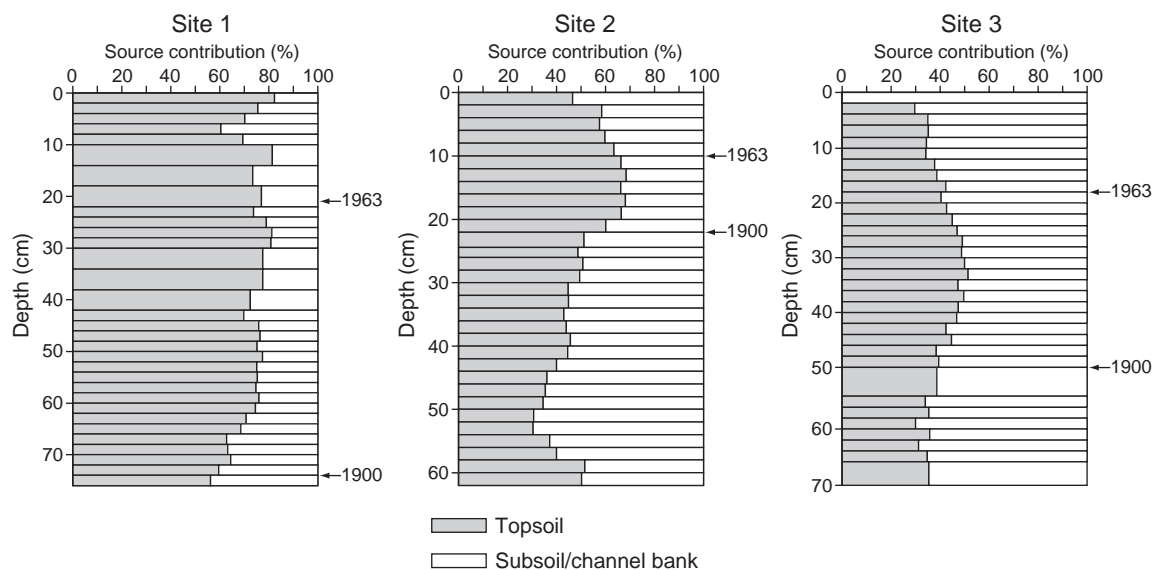


Fig. 4. Downcore changes in the relative contribution of topsoil and channel bank/subsoil sources for the three cores collected from the River Ouse floodplain (based on Owens et al., 1999).

aimed at reducing sediment mobilisation and delivery from cultivated areas.

3.5. The relative importance of individual sediment source types: the UK scene

Essentially similar procedures to those used in the study of source type contributions within the Upper Torridge catchment described above have been employed by the author and his co-workers in more than 30 catchments, both in Devon and elsewhere in the UK (see Fig. 5). In some cases, the proportions reported for the source type contributions are simple means of the values for the individual bulk samples collected from the river, rather than load-weighted means, but this is seen to be of limited importance in terms of the overall consistency and comparability of the results presented. The spatial coverage of the results and the variation in the size of the catchments investigated also reflect the opportunistic nature of the various studies. However, although far from comprehensive in terms of national coverage, the results of these studies, which are summarised in Table 9, provide a useful, perspective on suspended sediment sources within UK catchments and the countrywide variability in the relative importance of the various potential sources.

Any attempt to synthesise the results presented in Table 9 must take account of catchment size, since channel erosion could be expected to be more important in larger catchments with well-developed channel banks, whilst smaller catchments provide greater opportunity for a particular land use to be dominant and to therefore dominate the source contribution. Equally, it is important to recognise that the results are presented in terms of relative contributions and that equivalent information on the sediment yields from the catchments would be required to assess the absolute importance of a particular source. Thus, in the case of a catchment with a high contribution from channel erosion, the actual amount of sediment contributed by this source could be considerably less than for another catchment of similar size, where channel erosion accounts for a smaller proportion of the sediment load, but that load is substantially greater. The explanation for the relative magnitude of the contribution from a particular source could thus lie in the factors controlling the importance of the contributions from the other sources, rather than the source in question.

Bearing in mind the above constraints, Table 9 indicates that channel or bank erosion commonly accounts for between ca. 4 and 40% of the suspended sediment loads of UK rivers, and that the contribution is typically around 5–15% (see Fig. 6). Surface

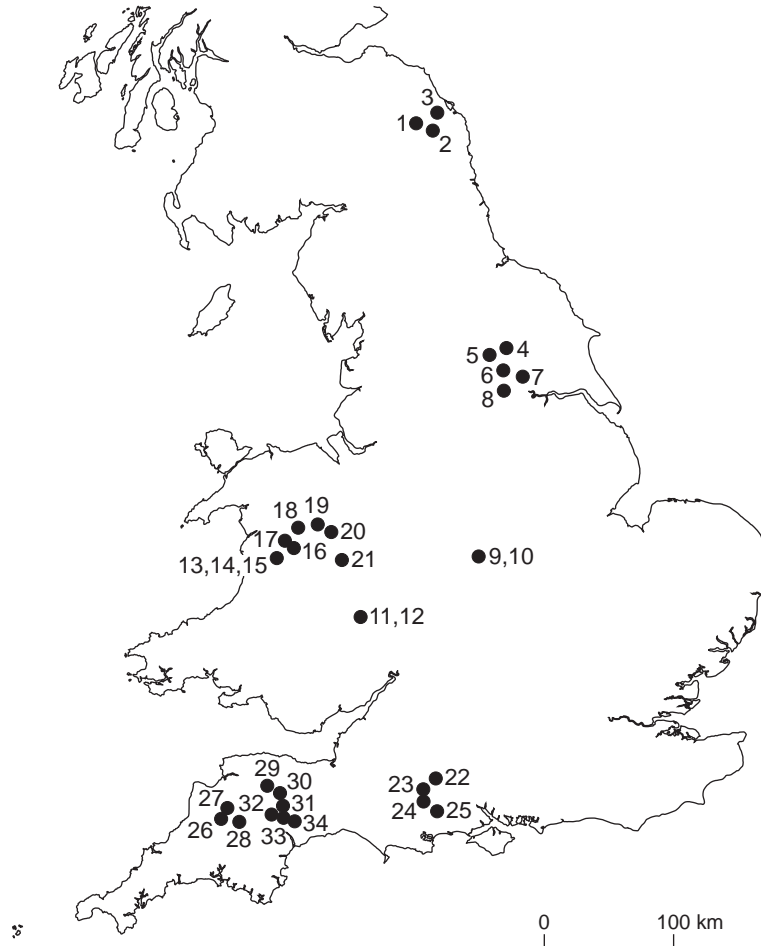


Fig. 5. The location of drainage basins in the UK for which sediment source data have been obtained by the author and his co-workers using the fingerprinting technique.

sources, taken together, are the dominant source in all catchments, accounting for 60–96% of the sediment yield, but with values of 85–95% being more typical (see Fig. 6). Turning to spatial patterns, the data presented in Table 9 suggest that bank erosion contributes a greater proportion to the total sediment load in upland catchments (e.g., the Tweed and its tributaries and the tributaries of the River Ouse in Yorkshire), where contributions of ca. 30% or more are common. Equally, surface sources appear to be more dominant (and therefore channel sources less important) in more lowland catchments, and particularly the smaller lowland catchments, where land use activities can be important in increasing sediment

mobilisation from surface sources. This contrast between upland and lowland areas has important implications for the design of effective sediment control strategies, since it is clear that reducing channel erosion is unlikely to prove an effective means of significantly reducing sediment loads in most lowland catchments, where available resources would arguably be better deployed in reducing erosion and sediment mobilisation from surface sources. Additional data regarding the importance of land drains as a sediment source are provided in Table 9. The drainflow contributions to the sediment yields of catchments 9 and 10 (31%) are less than those reported for the Rosemaund catchments (11 and 12)

Table 9
Estimates of source type contributions for a selection of UK catchments obtained using the source fingerprinting technique

Catchment No. ^a	River/catchment	Area (km ²)	Woodland	%Contribution ^b topsoil under pasture/moorland	Cultivated	Channel banks	Drains	Study
1	Ettrick Water	500	3.0	49	–	48		a
2	Teviot	1110	15	21	24	39		a
3	Tweed	4390	7	20	35	39		a
4	Swale	1350	–	42	30	28		b
5	Ure	914	0.7	45	17	37		b
6	Nidd	484	6.9	75	2.8	15		b
7	Ouse	3315	0	25	38	37		b
8	Wharfe	814	4.4	70	3.6	23		b
9	New Cliftonthorpe	0.96	–	30	33	6.0	31	c
10	Lower Smisby	2.6	–	26	37	6.2	31	c
11	Jubilee	0.31	–	3.1	37	12	48	c
12	Belmont	1.5	–	3.9	30	11	55	c
13	Upper Hore		11	63	–	26		d
14	Hafren		78	28	–	4		d
15	Upper Severn	8.7	22	68	–	12		d
16	Upper Severn	580	48	29	–	23		d
17	Rhiw	140	2.0	89	2.0	7		d
18	Vyrnwy	778	2.0	83	4.0	11		d
19	Perry	181	2.0	71	22	5.0		d
20	Tern	852	1.0	40	53	5.0		d
21	Severn	4325	2.0	65	25	8.0		d
22	Upper Avon	99	1.8	12	78	8.2		e
23	Wylfe	445	1.7	14	73	11		e
24	Nadder	221	1.3	16	69	14		e
25	Lower Avon	1477	1.4	16	64	19		e
26	Waldon	78	4	48	27	21		f
27	Upper Torridge	115	2	48	29	21		f
28	Torridge	258	2	47	28	23		f
29	Barle	128	6.0	85	1.0	8.0		d
30	Bathern	64	1.0	87	3.0	9.0		d
31	Lowman	54	2.0	54	40	4.0		d
32	Dart	46	3.0	82	11	5.0		d
33	Exe	601	3.0	72	20	5.0		d
34	Culm	276	–	30	60	10		g
34	Culm	276	–	35	53	12		h

^a See Fig. 5.

^b In several cases, contribution values were abstracted from histogram plots and represent approximate values, a—Owens et al. (2000); b—Walling et al. (1999a,b); c—Russell et al. (2001); d—Collins et al. (1997a,b); e—Heywood (2003); f—Nicholls (2001); g—Walling and Woodward (1995); h—He and Owens (1995).

considered above, but, nevertheless further emphasise the importance of drain systems as a suspended sediment source and delivery pathway.

4. The prospect

The case studies and results presented above clearly demonstrate the potential for using source

tracing or fingerprinting techniques to obtain information on suspended sediment sources within river basins, both in the UK and more generally. However, there remains a need to refine and further develop several aspects of the approach and scope clearly exists to extend the application of the source fingerprinting approach to embrace other aspects of the catchment sediment system. Both aspects are considered below.

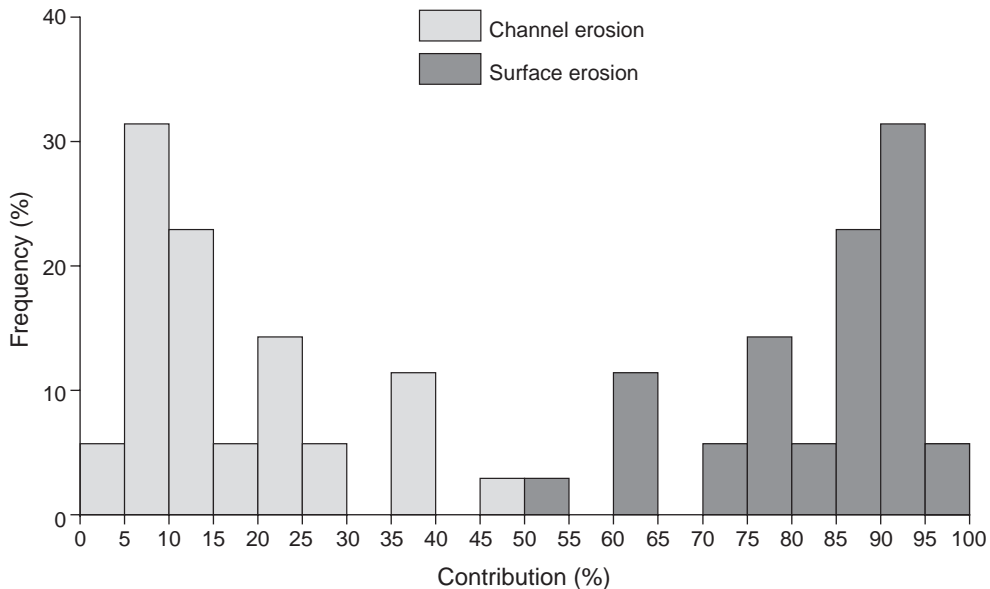


Fig. 6. Frequency distributions for the contribution of channel/subsurface and surface sources to suspended sediment samples collected from the catchments identified in Fig. 5 and Table 9 (in the case of catchments 9–12, drain contributions have been treated as a surface source).

4.1. Further refinement of the approach

Five key areas can be identified as being particularly deserving of further development and refinement. These can be summarised as follows:

- (1) identification of further fingerprint properties that will permit increased discrimination between potential sources and provision of guidelines for preselecting fingerprint properties;
- (2) development of improved procedures for taking account of contrasts in particle size composition and organic matter content between sediment and source material samples;
- (3) further investigation of the conservative behaviour of fingerprint properties;
- (4) more explicit incorporation of uncertainty into existing methodologies;
- (5) development of improved sampling strategies for obtaining suspended sediment samples.

Each of these areas will be briefly reviewed.

The successful use of composite fingerprints, involving a substantial number of fingerprint properties, has reduced the need to seek new properties capable of discriminating more clearly between

potential sources. However, scope clearly exists to increase the number of potential sources that can be included in a mixing model, if fingerprint properties capable of distinguishing those sources can be identified. Equally, scope still undoubtedly remains to improve the level of discrimination afforded by a composite fingerprint (cf. Tables 5 and 8) by including additional fingerprint properties influenced by different environmental controls. In the latter case, rare earths, stable isotopes and specific fractions of more commonly used fingerprint properties, such as heavy metals, could prove useful. In the former case, agrochemicals associated with specific crops and land use practices could provide a basis for discriminating sediment mobilised from areas under different crops or land use might prove useful. In this context, further attention could usefully be given to stable carbon and N isotopes which offer the potential to distinguish different crop residues (e.g., Papanicolaou et al., 2003). In addition, it should be recognised that existing procedures, which are frequently based on the analysis of a wide range of potential fingerprint properties and subsequent selection of a subset of these properties to provide the composite fingerprint, can prove highly demanding in terms of analytical resources. There is clearly a need to provide guidance

on the initial selection of fingerprint properties, in order to reduce the number of properties analysed. Such selection should clearly take account of the local conditions (e.g., underlying geology and soil types etc.) as well as the purpose of the fingerprinting exercise (e.g., source type or spatial source discrimination). Collins and Walling (2002) have begun this process, but further work is required to provide a basis for recommending sets of fingerprint properties for particular applications.

In most of the examples cited above, consideration was given to the need to take account of contrasts in grain size composition between the suspended sediment samples and the potential source materials, in order to make their fingerprint property concentrations directly comparable. However, there is an important need to refine the procedures employed for this purpose. It is known that differences in grain size composition will be of varying importance for different fingerprint properties and improved procedures should clearly take this into account. The procedure employed by Russell et al. (2001) in their study of drainflow contributions to the sediment output from the Rosemaund catchment was based on property-specific relationships between concentration and specific surface area and this approach merits further development. Equally, recent work reported by Motha et al. (2003) has taken a different approach involving fractionation of the source material sample to determine the fingerprint property concentrations associated with individual size fractions, which in turn made it possible to estimate the property concentration that would be associated with a sample of a known grain size composition. This approach could prove quite time consuming but, nevertheless, offers considerable potential as a means of dealing with the grain size problem. Motha et al. (2003) also introduced a further grain size correction, by adjusting the fingerprint property concentrations associated with individual source material samples used to characterise a particular source type to a standard grain size distribution, before calculating the mean concentration for the source type to be used in the mixing model. As indicated above, there have been few attempts to incorporate corrections for organic matter content into source fingerprinting procedures and several workers have argued that the grain size

correction will partly take account of differences in organic matter content, since the organic content of soils and sediments commonly increases as grain size decreases. However, there is again a need to explore this aspect more fully. The approach used by Motha et al. (2003) would appear to offer one way forward, although it is again demanding in terms of laboratory time. In this approach, the organic fraction was extracted from samples and analysed separately for the fingerprint properties. The resulting information provides a basis for directly estimating the effect of increasing or reducing the organic matter content of a sample on the fingerprint property concentration.

The source fingerprinting approach relies heavily on the assumption of conservative behaviour of the fingerprint properties during sediment mobilisation and transport. This assumption is usually addressed by selecting fingerprint properties that are known to be conservative. However, further work is undoubtedly required to explore this problem further and to verify empirically the assumption of conservative behaviour for a range of fingerprint properties. The study reported by Motha et al. (2002), which involved use of a rainfall simulator in the field, to simulate the mobilisation of sediment from the land surface and permitted direct comparisons between the properties of the mobilised sediment and the in situ source material, provides one potential approach for addressing this issue. Closely related to such considerations of conservative behaviour is the need to ensure that the assumption of linear additivity implicit in the use of mixing models is valid. Whilst this is likely to be the case for element concentrations and related geochemical measures, doubts have been raised regarding its validity for some mineral magnetic properties (cf. Lees, 1997).

The source ascription results provided in this paper, as well as those reported in many other investigations, are presented as absolute values. This could generate a false sense of precision, and it is important to recognise the many sources of uncertainty involved in generating such values. More particularly, the mixing model results are based on minimising the sum of squares of the relative deviations and thus represent a 'best estimate' with associated problems of equifinality. As indicated above a number of approaches involving Bayesian

statistics and Monte Carlo procedures have been employed to provide more explicit recognition of these uncertainties. Equally, it is important to recognise the problem of characterising an individual source type by a single fingerprint concentration and the many uncertainties necessarily associated with such a value again need to be more explicitly recognised and incorporated into the final results (cf. Foster and Lees, 2000). Furthermore, even though calculation of load-weighted mean contributions for particular source types undoubtedly increases the reliability of such values in terms of providing a value which is representative of the longer-term (e.g., annual) suspended sediment flux, there remains a need to explore more fully the number of bulk suspended samples required to characterise the suspended sediment flux from a catchment. This number will clearly depend on catchment size and could be greatly reduced if load- or time-weighted composite samples can be collected (see below).

The final problem area relates to the collection of suspended sediment samples for use in source fingerprinting exercises. Most existing fingerprinting studies have focussed on characterising the contribution of a set of potential sources to the longer-term sediment yield from a catchment and have therefore been based on the collection of a representative suite of essentially instantaneous samples from the catchment outlet. However, it must be recognised that the source composition of the sediment transported from a basin is likely to vary continuously through time, particularly in larger basins. During a storm, for example, such variations will reflect the routing of sediment from different parts of the catchment and temporal variations in the efficacy of the various sediment mobilisation processes operating within the catchment. Equally, when considering longer timescales, such as an annual cycle, source contributions could be expected to vary in response to changing surface conditions (e.g., land use) and changing hydrological response within the catchment. In order to provide reliable estimates of the source contributions, it is clearly important that a substantial number of samples, spanning a wide range of hydrological conditions, event magnitudes and timing within the event, should be collected in order to provide a representative result. In the catchment-based studies described in this paper, the load-weighted mean contribution has

been calculated. If a sampling device or installation, capable of collecting a load-weighted composite sample, was available, use of this sample in the fingerprinting exercise would provide a reliable estimate of the source contributions for the period covered by the sample. However, such samples are rarely available.

A further problem associated with the sediment sampling procedures commonly used in suspended sediment source tracing investigations is the need to obtain a sufficient dry mass of sediment to permit analysis of a wide range of fingerprinting properties. Determination of ^{137}Cs and related radionuclide concentrations by gamma spectrometry, will, for example, require several grammes, if not tens of grammes, of sediment. Depending on the ambient suspended sediment concentrations in the river at the time of sampling, obtaining this amount of sediment is likely to involve recovery of the sediment from a large volume sample (e.g., 50 l or more). Use of a pump linked directly to a continuous flow centrifuge could avoid the need to collect and transport these large volume samples, but in most cases, the transport of large samples to the laboratory will prove necessary and could represent an important operational constraint on the sampling programme. The suspended sediment trap sampler recently described by Phillips et al. (2000) and Russell et al. (2000) offers a means of overcoming this problem, in that the trap sampler is installed in the stream or river and a small proportion of the flow passes through the sampler body, where the sediment settles out and is trapped. When the sampler is emptied, the sediment is recovered from the relatively small volume of water contained within the sampler body. This avoids the need to collect large volumes of water. Furthermore, since the device collects a time-integrated composite sample, it affords a means of overcoming the problems of temporal representativeness highlighted above. The volume of water passing through the trap will increase as flow velocity increases during higher flows, but since the cross-sectional area of the intake remains constant, the sampler does not collect a true load-weighted composite sample. Nevertheless, the time-integrated sample should afford an effective means of characterising the sediment load during the period of sample collection. To date, the device has been primarily used in smaller catchments, but its principle is equally applicable to

larger streams and rivers, providing the sampler can be mounted in the flow and retrieved for emptying, when required.

4.2. *Extending the application of the approach*

As indicated above, introduction of further fingerprint properties into suspended sediment source tracing investigations could increase the potential for increasing the number of sources discriminated. These could include, for example, fields occupied by a specific land use or both topsoil and subsoil under particular land use types (cf. Gruszowski et al., 2003). Similarly, where a catchment contains urban areas, it should prove possible to include particulate material washed from urban surfaces (e.g., roads, car parks and roofs) as a discrete source (cf. Carter et al., 2003; Charlesworth et al., 2000). Equally, the successful use of source fingerprinting to distinguish sediment issuing from field drains reported above could be extended to include other transport pathways within a catchment. For example, Gruszowski et al. (2003) were able to discriminate sediment either derived from or transported via roads, in a study undertaken within a small (15 km²) rural catchment in Warwickshire. Scope also undoubtedly exists to distinguish sediment derived directly from primary source areas within a catchment and that remobilised from temporary storage, for example within the channel system (cf. Blake et al., 2002).

In the examples presented in this contribution, emphasis has been placed on obtaining estimates of the overall contribution of a particular source to the longer-term (i.e., annual or seasonal) sediment flux from a catchment. However, the temporal variability of source contributions has been highlighted (see Fig. 1) and scope clearly exists to investigate both intra- and inter-storm variations in source contributions in more detail. Such work could provide valuable insights into the temporal variability of sediment mobilisation processes during individual storm events (cf. Russell et al., 2001; Slattery et al., 2000), as well as changes in source contributions according to event magnitude. In the latter case, contrasts in the dominant sediment sources between high magnitude low frequency events and more 'normal' events and the existence of threshold conditions for increased mobilisation of sediment from certain sources could

usefully be investigated. Equally, more attention could be directed to seasonal contrasts in source contributions. In all cases, such investigations should couple source fingerprinting with detailed monitoring of suspended sediment fluxes, so that both the absolute and relative magnitude of contributions from particular sources can be established.

This paper has focussed almost exclusively on provision of information on the source of the suspended sediment flux from a catchment, although overbank floodplain deposits were used as a surrogate for suspended sediment, when considering past changes in sediment source. Considerable potential exists to extend such reconstruction to sediment deposits in lakes and reservoirs, which effectively provide a record of past sediment properties and therefore changes in sediment source (cf. Dearing and Foster, 1986; Dearing et al., 1990; Foster and Walling, 1994; Foster et al., 2003). The same basic approach can, however, also be applied to contemporary fine sediment in other situations. For example, Walling et al. (2003a) report a fingerprinting investigation aimed at clarifying the source of the fine matrix sediment accumulating in salmonid spawning gravels in rivers in different areas of Britain. It cannot be assumed that the source of such fine bed sediment is the same as that of suspended sediment more generally, since it is likely that a substantial proportion of the fine sediment infiltrating the gravel framework does so during low flows, when the fine sediment transported by the flow may have a different source to the main suspended sediment load. In this case, only a broad distinction between surface- and channel bank-derived sediment was possible, but this demonstrated contrasting sediment sources in different areas of the country, which in turn emphasised the need for different approaches to sediment management to control gravel siltation in these different areas.

As emphasised in the introduction to this contribution, sediment source can exercise an important control over the physical and geochemical properties of fine sediment. This control will also extend to the P content. P content will reflect a number of controls (cf. Walling et al., 2001b) but can be expected to vary significantly according to whether the sediment was mobilised from channel/subsurface sources or the catchment surface. Equally, sediment mobilised from

different soil types or areas under different land use can also be expected to vary in P content. Furthermore, the physical and geochemical properties of suspended sediment will control the uptake of dissolved P discharged from sewage treatments works and other sources and subsequent exchange between the particulate and dissolved phases, as the sediment is transported through the river system. This contribution has focused on establishing the source of the fine sediment transported by rivers. There is clearly a need for further work to explore in more detail the implications of sediment source for particulate P fluxes in catchments and river basins.

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