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Composite suspended sediment particles in river systems: their incidence, dynamics and physical characteristics

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Abstract:

Most of the existing data on the effective particle size characteristics of fluvial suspended sediment derive from instantaneous sampling methods that may not be representative of the overall suspended sediment loads. This presents difficulties when there is a need to incorporate effective particle size data into numerical models of floodplain sedimentation and sediment-associated contaminant transfer. We have used a field-based water elutriation apparatus (WEA) to assemble a large (36 flood) database on the time-integrated nature of the effective and absolute particle size characteristics of suspended sediment in four subcatchments of the River Exe basin of southwest England. These catchments encompass a wide range of terrains and fluvial environments that are broadly representative of much of the UK and temperate, low relief northwest Europe. The WEA provides important data on the physical characteristics of composite particles that are not attainable using other methods. This dataset has allowed, for the first time, detailed interbasin comparisons of the time-integrated particle size characteristics of suspended sediment and reliable estimates of the contribution of five effective size classes to the mean annual suspended sediment load of the study catchments. The suspended sediment load of each river is dominated by composite rather than primary particles, with, for example, almost 60% (by mass) of the sediment load of the River Exe at Thorverton transported as composite particles >16 μm in size. All the effective size classes contain significant clay components. A key outcome of this study is the recognition that each catchment has a distinctive time-integrated effective particle size signature. In addition, the time-integrated effective particle size characteristics of the suspended loads in each of the catchments display much greater spatial variability than the equivalent absolute particle size distributions. This indicates that the processes producing composite particles vary significantly between these catchments, and this has important implications for our understanding of the dynamics of suspended sediment properties. Copyright © 2007 John Wiley & Sons, Ltd.

KEY WORDS suspended sediment; composite particles; aggregation; flocculation; particle size; water elutriation; time-integrated sampling

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INTRODUCTION

It is now widely accepted that the suspended sediment loads of many rivers are dominated by composite particles (Droppo, 2001; Walling and Woodward, 2000). Most researchers in this field now routinely distinguish between the effective particle size distribution of fluvial suspended sediment, which includes composite particles, and the absolute particle size distribution, which represents the primary mineral particles (Ongley *et al*., 1981; Walling and Moorehead, 1987; Walling and Woodward, 1993). Composite particles can be at least two orders of magnitude larger than the majority of their constituent mineral grains (Woodward and Walling, 1992; Droppo and Ongley, 1992; Walling and Woodward, 2000), resulting in greatly enhanced settling velocities for the finest portions of the sediment load. This has important implications for how we conceptualize the hydrodynamic behaviour of suspended sediment and for the calibration of sediment transport and channel and floodplain sedimentation models (Ongley *et al*., 1992; Nicholas and Walling, 1996; Droppo *et al*., 1998; Bungartz and Wanner, 2004). Furthermore, because suspended sediment geochemistry is closely related to particle size (Horowitz and Elrick, 1987; Horowitz, 1991), information on the nature and significance of composite particles is essential to provide more realistic assessments of the flux and fate of sediment-associated contaminants in fluvial systems. Phillips and Walling (1999) have shown, for example, that the deposition of composite sediment particles in the hyporheic zone of gravel bed streams is an important process.

From a sediment monitoring perspective, it is also clear that there are considerable theoretical and practical difficulties associated with measuring the *in situ* or effective particle size characteristics of fluvial suspended sediment and the global dataset is still small—with a complete absence of effective particle size data for many environments upstream of the estuarine zone. It is becoming increasingly clear that the ubiquity of composite particles has important implications for sediment and water quality research, since the chemically active clay fraction is typically combined with a variety of organic materials within much larger composite particles (Droppo *et al*., 1998; Droppo, 2001). Most of the research in this field

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has focused on the development of new field techniques for measuring the effective particle size characteristics of fluvial suspended sediment and most of these approaches involve instantaneous sampling. To date, however, there has been little published information on interbasin variability in the effective particle size characteristics of fluvial suspended sediment. The nature of the relationships between catchment characteristics, the physical characteristics of composite particles and the spatial variability in the processes promoting their formation have not been addressed. The clay content of composite particles is an important physical characteristic that is difficult to quantify using traditional methods. Similarly, it is far from straightforward to quantify the relative importance of discrete and composite particles in suspended sediment loads. A key aim of this paper is to demonstrate how use of a field-based water elutriation apparatus to collect time-integrated samples can provide new insights into the nature and dynamics of composite particles that are not attainable using other methods.

In this paper, following Woodward and Walling (1992) and Walling and Woodward (2000), we use the term composite particle to refer to any suspended particulate that comprises two or more particles, irrespective of its mode of formation, and an example from the Hampshire Avon (1478 km^2) in southern England is shown in Figure 1. We refer to composite particles that form by flocculation in the river channel as flocs and those that originate from hillslopes and channel margin materials before being entrained into the channel flow, as aggregates. This distinction is not always straightforward as aggregates and flocs may break up during transport to produce discrete particles and smaller composite forms–and they may also flocculate due to a range of physical, chemical and biological processes. The detailed mechanisms involved are beyond the scope of this paper and we use composite particle(s) as a non-genetic term that includes both source material-derived aggregates and the products of in-stream flocculation (Walling and Woodward, 2000; Droppo, 2001; Woodward *et al.*, 2002).

In order to improve our understanding of the occurrence and nature of composite particles in suspended sediment transport and the spatial and temporal variability in the effective particle size characteristics, the authors have developed and evaluated several sampling techniques for this purpose that can be deployed during flood events (Table I) (Woodward and Walling, 1992, 1999; Walling and Woodward, 1993, 2000; Woodward *et al.*, 2002). These methods have been tested in reconnaissance studies in the Warwickshire Avon, the Hampshire Avon and the Dorset Stour and in catchments in the River Exe basin (Figure 2) and coupled to a more detailed and long-term sampling programme at four key sites in the River Exe basin (Table I and Figure 3).

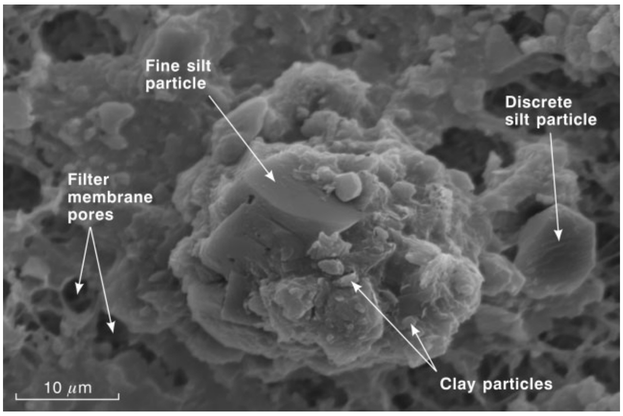

Figure 1. SEM image of a composite suspended sediment particle from the Hampshire Avon at Fordingbridge. See text for the definition of composite particle used in this paper. The location of this site is shown on Figure 3. This composite particle is approximately 25 μm in diameter (medium silt). Its constituent particle matrix includes a range of grain sizes with fine silt particles and clays and very fine submicron particles. Smaller composite particles and discrete mineral grains are present on the filter paper substrate

A RECONNAISSANCE SURVEY IN MIDLAND AND LOWLAND BRITAIN

Walling and Woodward (2000) presented effective particle size data for six sites in lowland river catchments across Midland, Southern and Southwest England (Figure 2 and Table I). This survey incorporated a wide range of catchment land uses, rock types and soils that are representative of many river catchments in Britain. Reconnaissance samples from each river indicated that the suspended sediment loads were dominated by composite particles and each of these instantaneous samples revealed marked contrasts between the effective and absolute particle size characteristics (Figure 2). These observations have been supported by scanning electron microscopy (SEM) investigations of suspended sediment samples collected from each sampling station (e.g. Figure 1). The particle size data shown in Figure 2 also show evidence of marked spatial variability in both the effective and absolute particle size distributions. The Warwickshire Avon at Stratford (1273 km^2), for example, displays distinctive peaked distributions for both curves and has the finest absolute size distribution with a high proportion of clay-sized material. In contrast, the Hampshire Avon at Fordingbridge has the coarsest absolute distribution with a significant medium and coarse silt fraction and a prominent size mode around 25 μm. Further downstream in the Warwickshire Avon at Evesham (2210 km^2), the effective particle size distribution contrasts markedly with the upstream data from Stratford (Figure 2) and the much coarser distribution at Evesham may reflect the operation of in-stream flocculation processes or inputs of coarser, aggregated sediments. It is important to appreciate that these particle size data were derived from instantaneous suspended sediment samples and a systematic survey of the spatial and temporal dynamics of the effective and absolute particle size characteristics of suspended sediment in British rivers has

Table I. Sample site details for the catchments in this study where measurements of the effective particle size characteristics of fluvial suspended sediment have taken place. Ordnance Survey Grid References and gauging station elevations above sea level are given for each site. The maximum elevation in the upstream catchment is also shown

River basin	Sampling station[a]	Drainage area (km^2)	Mean flow (m^3 s^{-1})	Maximum elevation (m)	Methods used[b]
Hampshire Avon	Fordingbridge SU158144 (25·6 m)	1478	15·18	294	① ② ③
Dorset Stour	Throop SY113958 (4·4 m)	1073	13·21	277	① ② ③
Warwickshire Avon	Stratford SP205549 (34 m)	1273	No data	320	① ② ③
Warwickshire Avon	Evesham SP040438 (19·5 m)	2210	15·35	320	① ② ③
River Exe	Thorverton SS936016 (25·9 m)	601	15·88	519	① ② ③ ④ ⑤
River Culm	Rewe SX946992 (23 m)	273	3·7[c]	293	① ② ③ ④ ⑤
River Dart	Bickleigh SS935076 (48 m)	46	<2·0[d]	270	① ② ③ ④ ⑤
Jackmoor Brook	Pynes Cottage SX902988 (27 m)	9·8	<0·5[d]	235	① ② ③ ④ ⑤

[a] Samples were collected at or close to the gauging station sites listed.
[b] ① Resuspension of composite particles from bulk (20 l) samples (Phillips and Walling, 1995b); ② samples collected for analysis by SEM (Woodward and Walling, 1992); ③ bulk samples collected for the measurement of absolute particle size; ④ rapid transport of bottle samples for effective size measurement using a Mastersizer (Walling and Woodward, 2000); ⑤ use of a water elutriation apparatus (Walling and Woodward, 1993).
[c] Mean flow data from Woodmill gauging station upstream (226 km^2).
[d] Estimated.

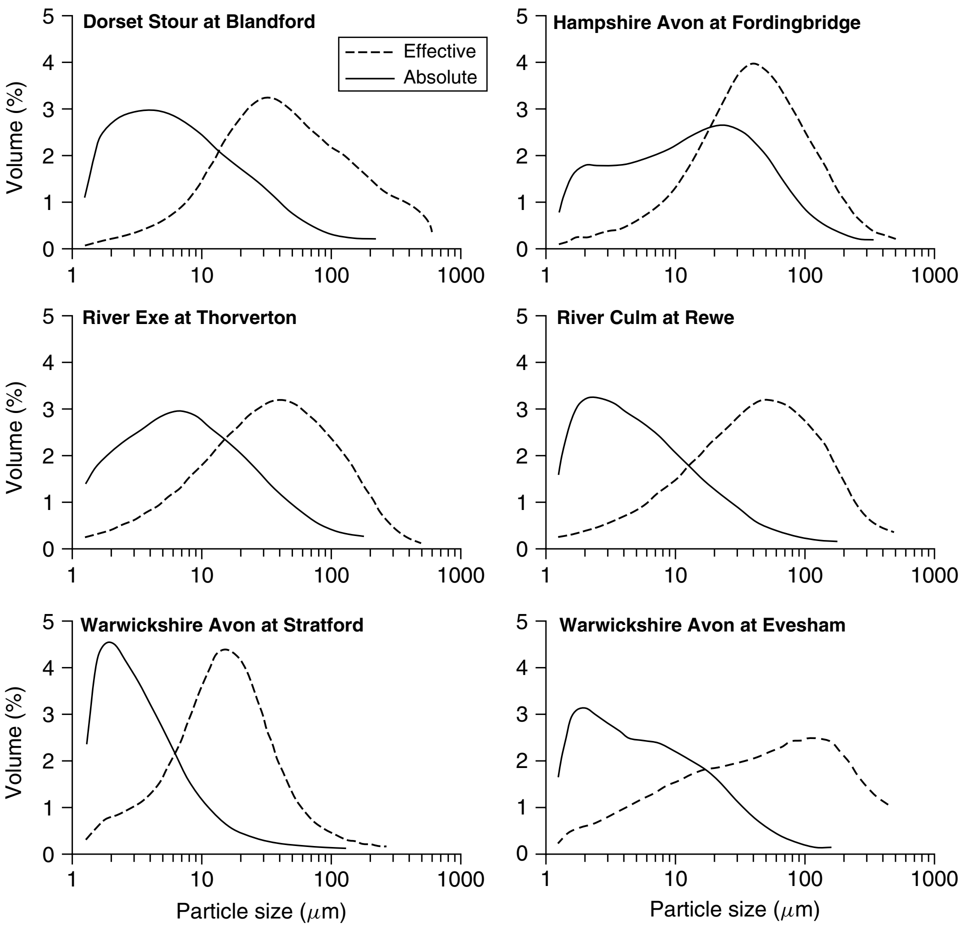

Figure 2. Effective and absolute particle size distributions from six sampling stations on river systems across lowland Midland and Southern England (after Walling and Woodward, 2000). Note that these data are derived from instantaneous samples collected during flood events

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not been carried out. Similarly, little is known about the time-integrated nature of the effective and absolute particle size characteristics of suspended sediment, not only in UK rivers, but in fluvial systems more generally, and it can be argued that these data are the most relevant for sediment transport and contaminant flux modelling.

THE WATER ELUTRIATION SAMPLING PROJECT

One of the approaches developed by the authors involves a custom-built field-based water elutriation apparatus (WEA), which, uniquely, allows continuous sorting by hydraulic separation of a time-integrated suspended sediment sample into five effective size classes during the course of a flood event. This apparatus draws sediment-laden floodwaters directly from the river channel and it can be operated for several hours during the course of a storm discharge event to ensure that a representative sample of the sediment wave is collected and elutriated within the sedimentation chambers. The WEA was first deployed for this purpose in the River Exe at Thorverton gauging station (601 km^2) over 10 years ago and an evaluation of the approach with data from six flood events was reported by Walling and Woodward (1993). After initial testing at the Thorverton station, the sampling programme was expanded to incorporate a total of four sampling sites, and four water elutriation systems were built and installed to allow simultaneous sampling of runoff events across the middle and lower sections of the Exe basin (Figure 3). This strategy was designed to investigate the significance of contrasts in catchment characteristics on the nature of the time-integrated particle size characteristics (for both the effective and absolute distributions) of fluvial suspended sediment and to identify the most appropriate sampling methods to generate reliable and relevant data

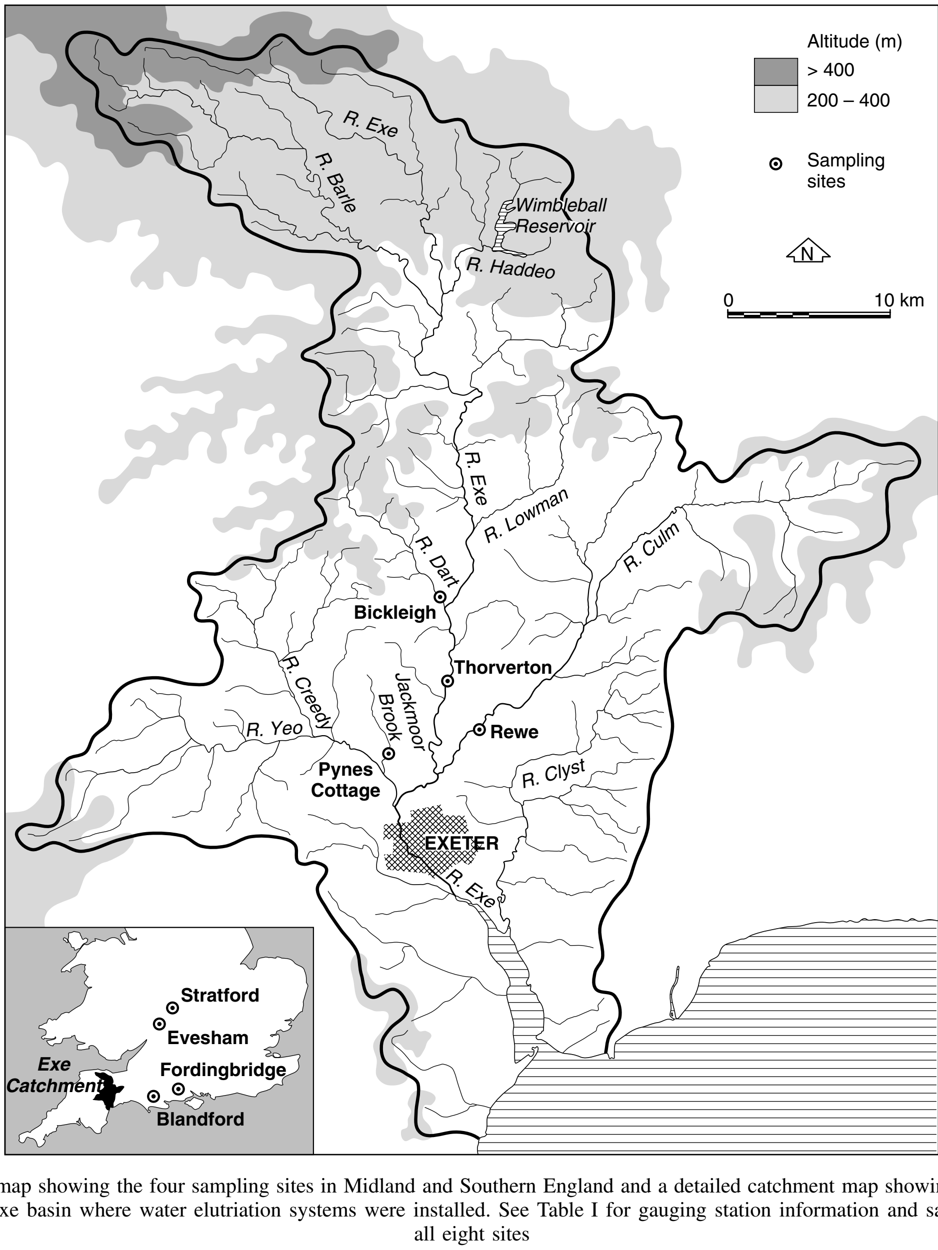

Figure 3. Location map showing the four sampling sites in Midland and Southern England and a detailed catchment map showing the four sampling sites in the River Exe basin where water elutriation systems were installed. See Table I for gauging station information and sampling methods for all eight sites

for model calibration (see Nicholas and Walling, 1996). A key objective was to improve our understanding of the nature of composite particles, especially their clay content, and to evaluate any contrasts in the effective particle size properties between the catchments under investigation. In view of the considerable body of background data available for these four catchments, they provided excellent contexts in which to explore these issues and, together, they incorporate a wide range of hillslope and fluvial environments that are representative of terrains in many river basins across the UK and in temperate northwest Europe. This paper presents, for the first time, the full time-integrated particle size dataset for 36 flood events from the four Exe basin catchments. It reports both effective and absolute time integrated particle size data for each flood event and discusses the spatial and temporal dynamics of these properties. The relative importance of composite and discrete particles in suspended sediment flux is discussed and the effective size data are used to estimate the role of composite particles, subdivided into five size classes, in the mean annual flux of suspended sediment in each catchment.

THE STUDY SITES IN THE RIVER EXE BASIN

Four catchments in the River Exe basin (1500 km^2) were selected for this investigation. The monitoring sites are shown in Figure 3 and background data for each catchment are given in Table I. Continuous long-term records of discharge and suspended sediment concentration and estimates of mean annual sediment yield were available for each site (Walling and Webb, 1987). Mean annual suspended sediment yield values range from 20 t km^{-2} yr^{-1} for the River Culm at Rewe (273 km^2) to 58 t km^{-2} yr^{-1} for the River Dart at Bickleigh (46 km^2). A marked north–south precipitation gradient is evident in the Exe basin with values ranging from >1800 mm on Exmoor above 400 m to *c*. 750 mm in the lower basin near Exeter just upstream of the estuary. The four study catchments show marked contrasts in scale (>600 km^2 to <10 km^2), topography, lithology, soil type, land use, channel and floodplain geomorphology and runoff hydrology. These contrasts in catchment characteristics combine to produce differences in both the annual suspended sediment fluxes and in the relative contributions from the major suspended sediment source types, such as channel bank erosion and surface erosion from arable land and pasture land (Collins *et al*., 1988; Walling and Woodward, 1992, 1995; Walling *et al*., 1993). Similarly, previous work in these catchments has shown that spatial variations in the absolute particle size characteristics of the suspended load can be related to these catchment variables and the nature of the sediment delivery system (Walling and Moorehead, 1987). For example, previous work has shown that suspended sediment collected at Pynes Cottage and Rewe has a finer median absolute grain size, less sand and higher clay content than that sampled at Thorverton and Bickleigh (Figure 3). Maximum storm period suspended sediment concentrations also show significant spatial variability across the Exe basin, with values at Pynes Cottage and Bickleigh frequently exceeding 1500 mg l^{-1} but rarely exceeding 800 and 400 mg l^{-1}, respectively, in the reaches at Thorverton and Rewe (Phillips and Walling, 1999).

FIELD SAMPLING OF SUSPENDED SEDIMENT USING THE WATER ELUTRIATION APPARATUS

The custom-built water elutriation apparatus used in this study is shown in Figure 4. The design and field deployment of this equipment and the principles of hydraulic sorting of suspended sediment using an elutriation system have been reported fully by Walling and Woodward (1993) and only an outline is given here. The WEA consists of four glass sedimentation chambers, a peristaltic

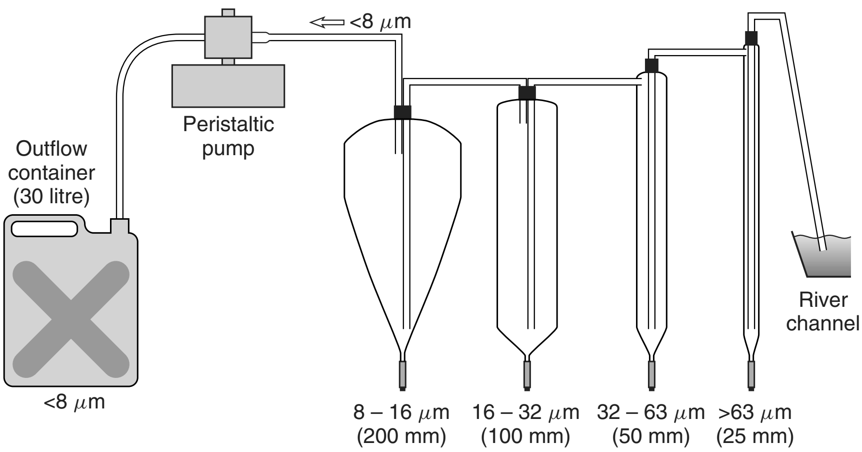

Figure 4. Schematic diagram of the field-based water elutriation apparatus used for sampling flood events in this study. The internal diameters of the cylindrical portion of each sedimentation chamber are shown in parentheses. The dimensions of the apparatus shown take advantage of the fact that doubling the diameter of a sedimentation chamber (i.e. from 25 to 50 mm and from 100 to 200 mm) causes its cross-sectional area to be increased by a factor of four. To obtain the desired particle sizes, the velocity of the flow in successive chambers must decrease by a proportion determined by Stokes' Law: $V = kr^2$, where V is the velocity of a settling particle, k is the proportionality constant and r is the radius of the particle (Follmer and Beavers, 1973, p. 546)

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pump with variable speed control, and a 30-l plastic outflow container. The sedimentation chambers contain a long inlet glass tube and short outlet tube and the chambers are linked with flexible plastic tubing (Figure 4). The cylindrical sections of the chambers have internal diameters of 25, 50, 100 and 200 mm, respectively. The sample inflow is a length of plastic tube connected to the first (25 mm) sedimentation chamber. The peristaltic pump maintains a constant flow rate throughout the sampling period and is positioned in-line after the 200 mm sedimentation chamber. The outflow from the pump is retained in the 30-l container (Figure 4). In the laboratory, the use of water elutriators is commonly recommended for the size range 10–200 μm for particles with densities greater than 2·0 g cm^{-3} and this is compatible with the composite sediment particles transported in the Exe basin and the effective size classes we have selected (see below). The effective size classes were defined using Stokes' Law assuming a particle density of 2·65 g cm^{-3}. The composite suspended sediment particles in each effective size class will display a range of shapes and densities, so these groupings should be seen as 'hydraulically-equivalent' classes. Thus, while the value of 2·65 g cm^{-3} may overestimate the density of those particles with a significant organic content, these classes will directly reflect their 'hydraulic size' in the river channel (see Walling and Woodward, 1993). Prior to each sampling run, the WEA was filled with untreated clear river water that had been collected from the channel at the respective sampling sites. Earlier demonstrations of water elutriation systems in the laboratory analysis of soils and sediments are provided by Beavers and Jones (1966) and Follmer and Beavers (1973).

Field sampling of suspended sediment

At each of the four sampling sites in the Exe basin shown in Figure 3, a water elutriation system was installed close to the river channel, either in the gauging station hut itself or in purpose-built housing. The sample intake tube has an internal diameter of 4 mm and was fixed either to a permanent stake in the stream bed or to a stake fixed normal to the channel bank. In all cases, the inlet tube was positioned to sample floodwaters above the zone of bed load transport and below the mean annual flood stage. Turbidity monitors were available at each site and the elutriation apparatus was operated during floods when suspended sediment concentrations exceeded 100 mg l^{-1}. In total, 36 flood discharge events were sampled between October 1991 and January 1993. All of these floods took place between October and April so it is not possible to examine any summer/winter contrasts from these data. Eight floods were sampled in the Jackmoor Brook and River Culm catchments and 10 in the Exe and Dart catchments, respectively. Many of these events were sampled concurrently at two or more sites during basin-wide storm discharge events. River water was drawn directly into the WEA for up to 8 hours during individual sampling runs. As flood events in these catchments are typically characterized by clockwise hysteresis, with the peak in sediment concentration preceding the peak in discharge, it was often possible to obtain a representative sample of a given sediment wave in the first few hours of a flood discharge event. With less flashy responses and typically lower suspended sediment concentrations, WEA sampling runs at the Exe and Culm monitoring stations (Figure 3) were typically rather longer (between 4 and 8 hours). The flow rate through the field apparatus was adjusted for each run according to ambient river water temperature to retain particles with equivalent spherical diameters (assuming a density of 2·65 g cm^{-3}) of >63 μm, 63–32 μm, 32–16 μm, and 16–8 μm in the four sedimentation chambers (Figure 4). The <8 μm size class was retained in the outflow container (Figure 4). The results obtained therefore represent a *time-integrated* measure of the effective particle size distribution of the transported sediment. This approach offers significant advantages over those sampling techniques that generate essentially instantaneous estimates of the effective particle size distribution, although the latter can be useful if several samples are collected during the course of a flood event. The length of the sampling period for the WEA varied according to the magnitude and duration of the runoff event and the associated suspended sediment concentrations.

Walling and Woodward (1993) reported the results of a number of tests that demonstrated that the sediment concentrations in the river water entering the apparatus are representative of those within the river channel. This is largely due to the fact that clay- and silt-sized particles (often referred to as the cohesive sediment fraction) dominate the absolute grain size distribution of the suspended sediment load of all four catchments and the sand fraction rarely exceeds 5% of the total loads (Table II). The sample intake tube on the WEA has the same internal diameter (4 mm) as the horizontal within-channel suspended sediment trap sampler described by Phillips *et al.* (2000). Field tests have shown that the particle size characteristics of the sediment collected by that sampler were also representative of the ambient suspended sediment (Phillips *et al.*, 2000).

Walling and Woodward (1993) have argued that composite particle breakage due to shear effects within the inlet tube is not a significant process. The SEM analysis of composite particles in these rivers has shown that they commonly display a compact arrangement of constituent particles, with many similarities to soil aggregates (Woodward and Walling, 1992; Walling and Woodward, 2000), and these forms appear to be resistant to breakage during the water elutriation process. Turbulence does occur in the lower portion of the sedimentation chambers at the base of each inlet tube. However, since this was no greater than that occurring naturally in the river channel, significant break-up of composite particles was considered unlikely and the composite particles drawn into the apparatus are believed to be representative of those in the river (Walling and Woodward, 1993). Laminar flow exists

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Table II. The magnitude of the >63 μm fraction of the suspended sediment load reported by Walling and Moorehead (1987) and the water elutriation apparatus (WEA) discussed in this paper at the four sampling sites. The Walling and Moorehead figures are mean values based on the analysis of a large number of bulk samples using wet sieving. The effective size data (>63 μm) from the WEA are also shown. Mean values (%) >63 μm are given in each case

River	Sampling station	Walling and Moorehead Absolute	WEA Absolute	WEA Effective	WEA (n)
Jackmoor Brook	Pynes Cottage	4·0	4·52	11·31	8
River Dart	Bickleigh	4·2	4·17	11·14	10
River Culm	Rewe	1·2	1·91	2·11	8
River Exe	Thorverton	5·0	5·68	11·37	10

within the central and upper portions of each chamber (Beavers and Jones, 1966; Follmer and Beavers, 1973).

Laboratory procedures

At the end of each sampling run the suspended sediment samples ($n = 180$) were emptied from each sedimentation chamber into plastic bottles and, with the outflow container, were returned to the laboratory. The sediment from each effective size class was recovered by centrifugation prior to freezing and freeze-drying. The amount of sediment associated with each of the five effective size classes was then determined by weighing. These mass data represent the effective particle size distribution for each flood event. All the sediment samples were then treated to remove the organic fraction and chemically dispersed following standard procedures. The absolute particle size characteristics (including the clay content) of the mineral sediment comprising each effective class were determined using a Malvern Mastersizer laser particle size analyser. The absolute particle size data for each class were then weighted according to the measured mass of those classes so as to generate the time-integrated absolute particle size distribution for each of the 36 flood events (Walling and Woodward, 1993).

RESULTS AND DISCUSSION

Effective particle size

The sediment mass data for the effective classes provide a valuable representation of the effective particle size distribution of the suspended sediment sampled during each run and all the available data for the four catchments are shown in Figure 5. These plots show that the form of the time-integrated effective size distribution is broadly consistent for each catchment over time, although it is important to appreciate that the dataset does not include any events between May and September. Marked intercatchment (spatial) variability is, however, evident. The 10 sample runs from the River Exe at Thorverton show that the effective size classes >8 μm commonly account for about 75% of the sediment load and that the >32 μm fraction accounts for over 30% of the sediment load. This contrasts markedly with the River Culm at Rewe, where the <8 μm effective size class displays the highest temporal variability for all the samples, but the proportion of the load associated with this class is always greater than the equivalent class sampled at Thorverton and Bickleigh. On average, the two effective size classes >32 μm account for less than 10% of the sediment load of the River Culm at Rewe. The dominance of the <8 μm effective size class, in particular at Rewe, may partly reflect the presence of a significant number of low-density composite particles in this river system. The nature of the relationship between hydraulic equivalence, settling velocity and composite particle density requires further work in a range of catchment types (Droppo, 2001).

The effective size distributions for the Dart and Exe are broadly comparable in form, with a comparatively even distribution of material across the five effective size classes when compared with the Jackmoor Brook and River Culm. The similarity between the Dart and the Exe datasets is partly explained by the routing of sediment pulses from the high yielding central part of the Exe basin to the main channel reaches at Thorverton approximately 5 km downstream of the Dart–Exe confluence (Figure 3). In contrast, Jackmoor Brook drains a much smaller catchment (9·8 km^2) that is dominated by arable agriculture and drains into the River Creedy a few kilometres to the northwest of Exeter (Figure 3). It is interesting to note that the >63 μm effective size class at Jackmoor Brook accounts for a slightly larger proportion of the suspended load (mean = 11·3%) than the 32–63 μm class (mean = 10·2%). This contrasts with the other three catchments where the >63 μm class forms a significantly smaller proportion of the total load. The >63 μm effective class comprises a relatively minor part of the sediment load of the River Culm at Rewe, with a mean value of 2·1%. On average, the median particle size for the effective distributions lies within the following classes: River Culm (<8 μm), Jackmoor Brook (8–16 μm), River Dart (16–32 μm) and River Exe (16–32 μm).

As mentioned above, because most of the sampled flood events (34 out of 36) took place between late October and March, it was not possible to explore seasonal contrasts in the effective particle size characteristics of the suspended sediment transported by these rivers. It is important to appreciate, however, that the long-term records (>30 years) of suspended sediment transport for these catchments show that most of the annual suspended sediment flux takes place during a small number of flood events between October and April (Walling and Webb,

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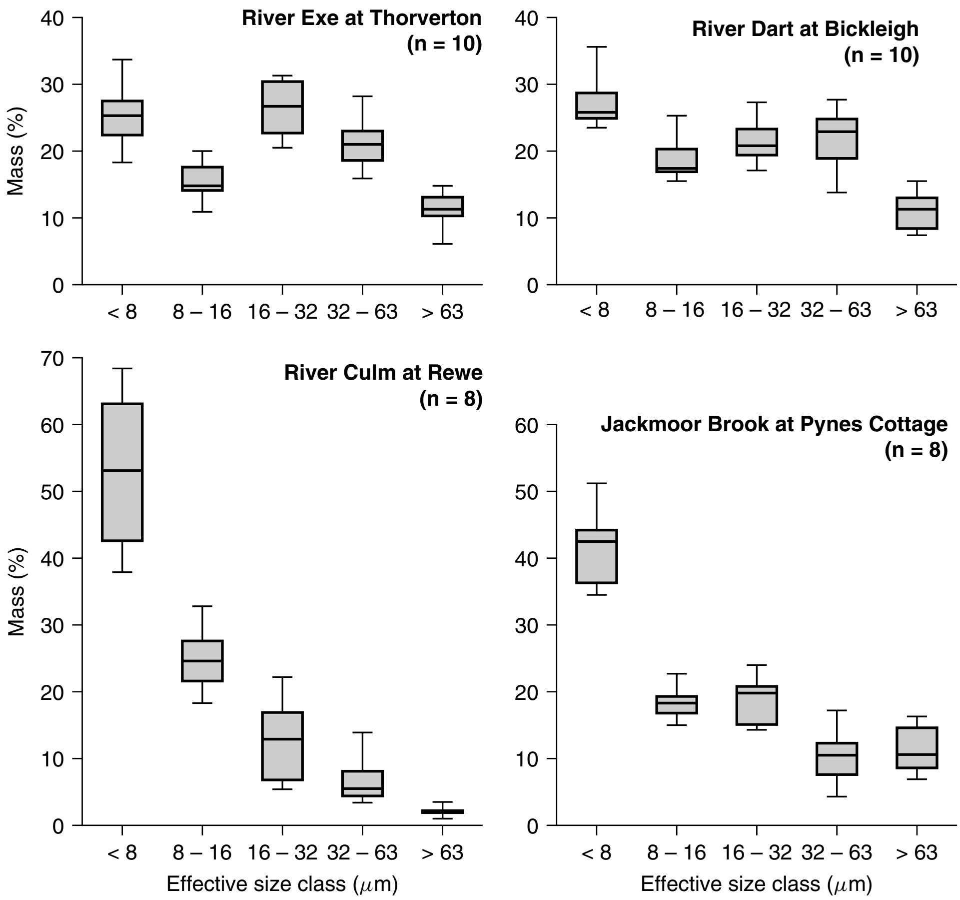

Figure 5. Box and whisker plots based on the effective particle size data derived from the water elutriation apparatus for all 36 flood events sampled in the Exe basin study catchments between October 1991 and January 1993. These plots show the sample ranges, the lower and upper quartiles and the median. Note the interbasin contrasts in the time-integrated effective particle size characteristics

1987; Walling *et al*., 1992). As the interquartile ranges are generally low (with the notable exception of the <8 μm class at Rewe), the data presented in Figure 5 for two flood seasons indicate that catchment-specific signatures that are representative of the long-term effective particle size distribution of the sediment load can be identified using time-integrated sampling. These effective size signatures may be a key attribute of the Exe basin catchments presented here and of comparable fluvial systems across the UK.

Absolute particle size

The effective size data are summarized in Figure 6, along with the equivalent absolute particle size distributions for every WEA run in each catchment. As mentioned above, the latter have been produced from the absolute particle size data for each of the effective class fractions and weighted according to the mass of those fractions (Walling and Woodward, 1993). These data are in good agreement with existing information on the absolute particle size characteristics of suspended sediment from these catchments (Walling and Moorehead, 1987). Some differences are to be expected in view of the operationally defined nature of particle size measurement (see Horowitz, 1991), and the fact that previous absolute size data from the Exe basin were mainly derived from samples obtained using instantaneous sampling methods using different laboratory methods such as the SediGraph (e.g. Walling and Kane, 1984). It is clear that the absolute distributions contrast markedly with the effective size distributions (Figure 6). The former are dominated by fine silt and clay material and in all cases the <8 μm fraction is the dominant class. The WEA dataset shows that the sediment loads of the Jackmoor Brook and River Culm display the finest overall absolute distributions with mean values of 70% and 68·4% respectively, for the <8 μm fraction. This is in good agreement with absolute particle size data for the study catchments obtained using the Par-Tec 200 Laser backscatter probe. Phillips and Walling (2005) report median particle size values of 4·6 μm and 5·0 μm, respectively, for the Jackmoor Brook and River Culm catchments. These catchments are underlain mainly by Permian rocks, while the River Exe at Thorverton (that drains Devonian rocks) and the River Dart at Bickleigh (that drains Carboniferous strata) show slightly coarser median size values of 6·5μm and 6·1μm, respectively (Phillips and Walling, 2005).

The sediment load of the River Culm at Rewe contains the smallest proportion of primary sand particles (mean = 1·9%) which is in agreement with previous estimates (1·2%) based on particle size determinations using the SediGraph and wet sieving reported in

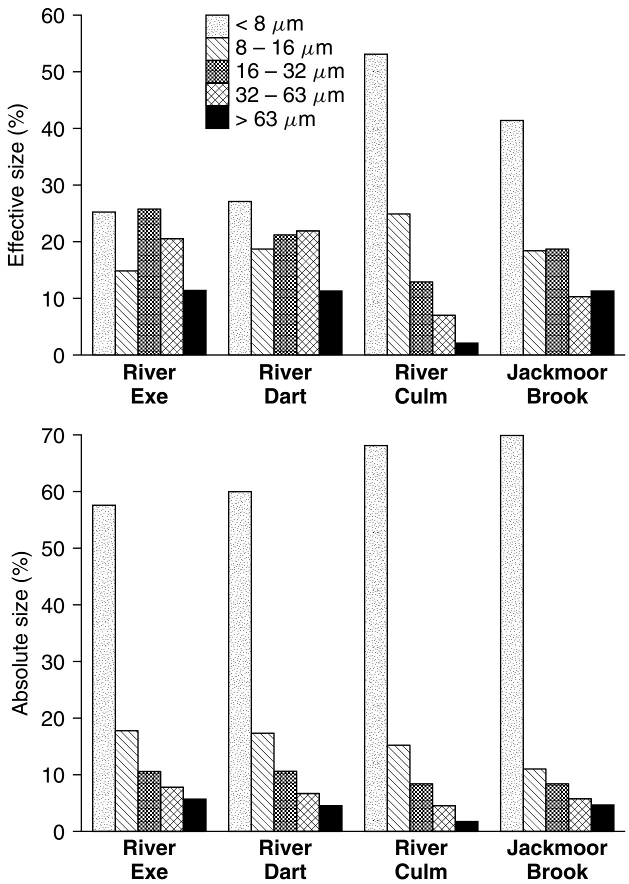

Figure 6. Mean values for the effective and absolute particle size classes for all the flood events sampled by the water elutriation apparatus in the four study catchments in the Exe basin

Walling and Moorehead (1987). It is significant that the sand (>63 μm) content data generated from the time-integrated absolute distributions from each sampling station are in excellent accord with existing mean data derived from wet sieving of a large number of bulk suspended sediment samples collected over a range of flow conditions and flood seasons (e.g. Walling and Kane, 1984; Walling and Moorehead, 1987) (Table II). The close agreement between these two independent assessments of the magnitude of the sand fraction adds further weight to our view that the WEA collects representative samples of suspended sediment in a reliable and consistent manner across a range of channel sizes and flow conditions.

A key feature of the absolute distributions derived from the WEA shown in Figure 6 is the marked reduction in spatial variability in comparison to the equivalent effective particle size distributions derived from the same apparatus. In each of the 36 events, the median (D_{50}) grain size of the absolute size distribution lies in the <8 μm fraction. These results agree well with recent (1994–1998) absolute particle size data obtained from 13 sampling sites on river systems within the Humber (*c.* 24 000 km^2) and Tweed (*c.* 4390 km^2) rivers; two large British catchments that drain to the North Sea. The mean D_{50} values range from 4·1 μm (River Don) to 9·2 μm (River Nidd) and the mean value for the total silt and clay fraction (<63 μm) ranges from 92·9% (River Trent) to 99·6% (River Don) (Walling *et al.*, 2000). Thus, the bulk of the sediment load in these river systems is finer than 63 μm, with a sand component typically <5%, and the range of values recorded is in good agreement with the absolute particle size data for catchments in the Exe basin. Walling *et al.* (2000) have also argued that the absence of a well-defined relationship between absolute particle size and discharge for the rivers within the Tweed and Humber catchments suggests that the form of the absolute particle size distribution is controlled more by sediment supply factors than transport capacity or flow hydraulics.

It is clear that the WEA data for the four Exe basin catchments show a marked increase in intercatchment variability for the effective particle size characteristics of the suspended loads relative to the absolute particle size characteristics. This pattern has emerged from the analysis of 36 flood events - the largest dataset yet compiled on the physical characteristics of suspended sediment based on time-integrated sampling for individual flood events. This pattern suggests that the process dynamics promoting the occurrence of composite particles, whether these mainly reflect inputs of source-derived (soil) aggregates (Meyer *et al.*, 1980) and/or the production of in-stream flocs, vary in their intensity *between* these catchments. These processes are complex (see Droppo, 2001) and catchment sediment sources and soil types, as well as sediment delivery dynamics (including slope-channel coupling and hydrological pathways) and a range of biological and flow-related processes, are likely to be important. Further work is required to ascertain the key catchment parameters governing the operation of these processes in British rivers and in freshwater fluvial systems more generally.

To further examine spatial (intercatchment) variability in the effective and absolute particle size characteristics in the Exe basin it is helpful to consider the relative importance of large (>32 μm), medium (8–32 μm) and small (<8 μm) composite and discrete particles, respectively, in suspended sediment delivery. These data are derived from the water elutriation datasets and have been plotted in Figure 7. The composite particle data occupy three distinct zones of the ternary matrix, with the Exe and Dart basins plotting to the left of centre, the Culm samples at lower centre and the Jackmoor Brook samples lying between these two extremes. A key contrast is evident between the two largest catchments, since the River Culm produces a much smaller proportion of large composite particles than the River Exe at Thorverton, with mean values of 6·4% and 32·4%, respectively. Again, the Dart and the Exe have similar distributions because of sediment routing from the latter to the former station and the contribution to the main channel of the River Exe from other tributaries dominated by Devonian rocks with similar terrain to the Dart basin. The SEM analysis reported in Woodward and Walling (1992) has demonstrated the presence of composite particles >200 μm in size in samples collected from the River Exe at Thorverton. The

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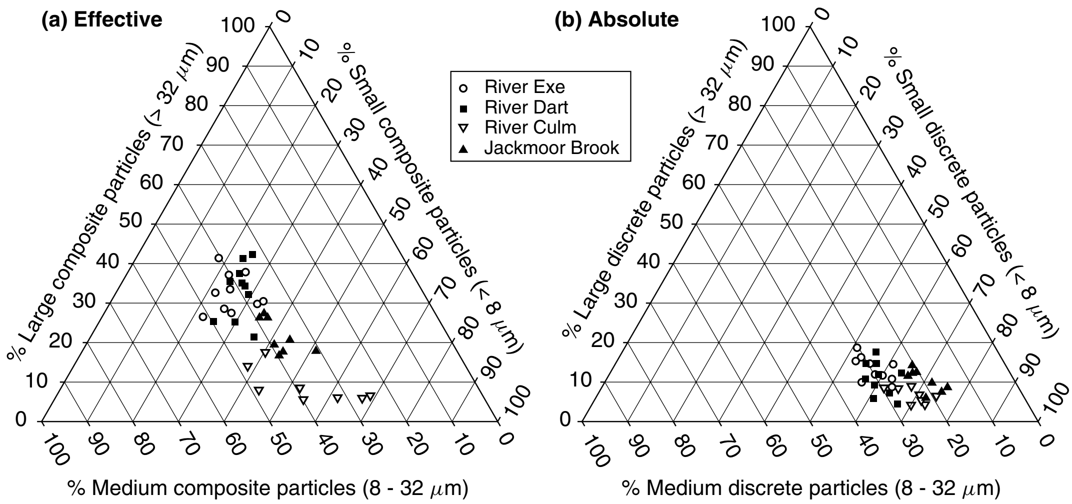

Figure 7. Ternary diagrams showing the proportion of large, medium and small particles in the effective and absolute particle size distributions for each of the study catchments

much lower proportion evident in samples from the River Culm at Rewe may be due to the much lower suspended sediment concentrations during flood events commonly found at this site, which limits opportunities for flocculation. It may also reflect differences in source material properties, but this issue can only be resolved by further research.

The Jackmoor Brook drains a small subcatchment of the Exe basin characterized by intensive mixed arable farming and typically displays the highest suspended sediment concentrations during flood events (up to *c.* 3500 mg l^{-1}). It is likely that the cohesive, clay-rich soils derived from the Permian rocks underlying this small basin deliver a significant proportion of aggregates to the fluvial system (see Meyer *et al.*, 1980; Phillips and Walling, 2005). Thus, in this catchment, within-channel flocculation of source-derived aggregates into larger composite forms is likely to be an important process. In contrast, there is much less spatial variation and less interstorm variation in the absolute particle size data (discrete particles). It is clear that the Jackmoor Brook and the River Culm transport sediment loads that are much finer grained than the Exe and Dart, with a significant number of flood events exhibiting values in excess of 70% for small (<8 μm) discrete particles and all the Culm samples contain fewer than 10% of large discrete particles (>32 μm) by mass (Figure 7). Some examples of composite particles from the rivers Exe and Culm are shown in Figure 8.

The relative importance of discrete and composite particles

The data provided by the WEA do not allow direct assessment of the proportion of particles within an effective size class existing as either discrete primary particles or as composite particles. The SEM and optical microscope investigations have shown that discrete particles are present in the suspended sediment loads of freshwater rivers (see Woodward and Walling, 1992; Droppo and Ongley, 1994; Walling and Woodward, 2000; Woodward *et al.*, 2002) and such particles are likely to be more abundant as the proportion of coarse silt and sand in the overall load increases (Figure 8). In practice, however, it is difficult to quantify the proportion of an effective particle size distribution that is composed of discrete particles. The proportion can be estimated from approaches based on particle counting using undisturbed samples mounted on customized microscope slides (e.g. Droppo and Ongley, 1992), but these methods are time consuming and the database is still small. It is possible, however, to estimate these proportions indirectly using the absolute size data derived from the WEA presented above and this is a further advantage of this approach. By comparing the absolute size distribution of the sediment associated with a particular effective size class with the lower size limit of that effective class, the amount of sediment finer than the lower limit of the effective class provides a minimum estimate of the proportion of sediment incorporated into composite particles (Walling and Woodward, 1993). This exercise has been carried out for flood events in each catchment and an example for each is shown in Table III. The proportion of discrete particles that is finer than the lower size limit of these effective size classes represents material previously incorporated into composite particles. As indicated above, the values given in Table III are likely to be underestimates, since many of the sediment particles coarser than the lower size limit of these effective classes will also be incorporated into composite particles. These results clearly demonstrate that the suspended sediment load of each catchment is dominated by composite particles (Table III) and this is likely to be the case in all freshwater rivers.

The clay (<2 μm) content of effective size classes

As shown above, a key advantage of the WEA lies in its ability to allow measurement of the absolute size characteristics of a specified portion of the effective particle size distribution. This allows the physical properties of a particular grade of composite particles to be investigated and the WEA is the only sampling approach that

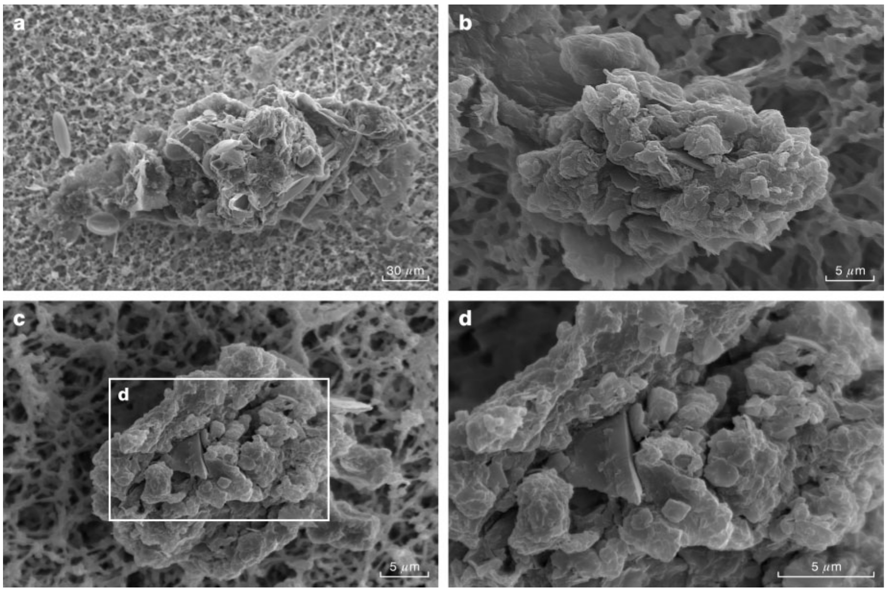

Figure 8. Scanning electron microscopy images of composite suspended sediment particles from the River Exe at Thorverton and the River Culm at Rewe. (a) Large composite particle from the River Culm. The dense core of this composite particle is bounded by a much looser outer matrix that includes medium silt-sized particles with diatoms and organic matter incorporated into the constituent particle matrix. (b) Medium composite particle from the River Culm composed of platy fine silt and clay-sized particles. The constituent particle matrix is much denser than the example shown in (a) from the same river. (c) A large composite particle from the River Exe at Thorverton showing a range of constituent particle grain sizes. (d) Detail from (c) showing an angular fine silt particle embedded within a finer constituent particle matrix of fine silts and clays. Note the discrete clay particles attached to the surface of larger constituent particles

Table III. The amount of dispersed mineral particles (%) in each effective size class finer than the lower size limit of that effective size class. These data provide an estimate of the proportion of inorganic sediment incorporated into composite particles within the four coarsest effective size classes. A typical example from each catchment is given. It is not possible to obtain an equivalent estimate for the <8 μm effective class

River	Date of event	% <8 μm in 8–16 μm class	% <16 μm in 16–32 μm class	% <32 μm in 32–63 μm class	% <63 μm in >63 μm class
Jackmoor Brook	30/11/92	65	79	79	77
River Dart	28/04/92	62	83	89	89
River Culm	29/11/92	74	58	66	81
River Exe	31/10/91	68	85	83	80·5

allows such insights. The exponential increase in surface area with decreasing particle size means that the clay content of each effective size class is of considerable interest since it may exert an important control on the size and stability of composite particles, and it will exert a strong influence on the geochemistry and contaminant loading of each effective size class (see Horowitz and Elrick, 1987; Umlauf and Bierl, 1987). The mean clay content of each effective size class is shown in Figure 9. All of the effective size classes contain a substantial proportion of clay-sized particles, with the highest values for the four classes <63 μm recorded for the Jackmoor Brook samples. In general, the composite particles in the suspended sediment load transported by the Jackmoor Brook contain significantly more clay than the other three rivers (Figure 9). The <8 μm effective size fraction for the Jackmoor Brook averages 52·8% clay, while the clay components in each of the effective size classes in the other three catchments are broadly comparable, particularly for the 16–32 μm and 32–63 μm classes (Figure 9).

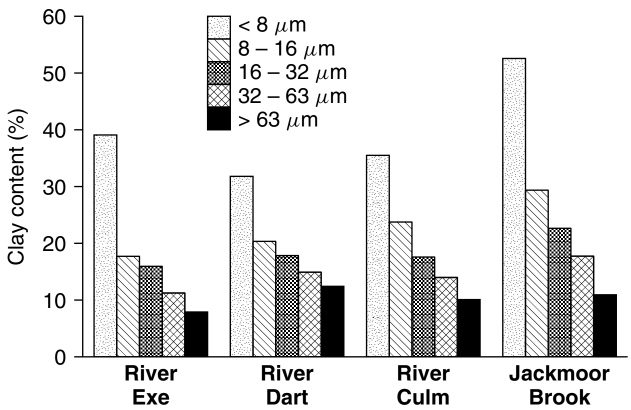

Figure 9. The mean clay (<2 μm) content of each effective size class for the 36 flood events shown in Figure 5

It is likely that the clay fraction (and its interaction with various kinds of organic matter) plays an important role in the formation and stability of composite sediment particles. The SEM images shown in Figure 8 show that clay-sized particles form an important component of these composite particles. They are present in a variety of

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shapes and sizes and the finest clay particles with their charged surfaces and very high surface areas are likely to be important binding agents in both aggregates and flocs. Previous work has documented the clay mineralogy of the suspended sediment load in these catchments using XRD (Walling and Kane, 1984). Use of an EDAX system in tandem with SEM to determine the dominant clay minerals in composite particles from different catchments is a potentially promising avenue of future research. This approach might allow an assessment of aggregation and flocculation potential based on catchment characteristics such as lithology and soil type.

The mean clay content of the <8 μm effective size class is actually higher for the suspended sediment load of the River Exe (39·1%) than for the River Culm (35·4%), although the <8 μm effective class for the River Culm accounts for 53·1% of the effective size distribution in comparison to 25·6% for the River Exe. For each of the 36 flood events the proportion of the total amount of clay transported within each effective size class has been calculated. These results indicate that, in the case of the River Dart and the River Exe, the effective size classes covering the range >8 μm commonly account for the transport of more than 50 and often >60% of the total load of clay-sized particles. The equivalent value for the Jackmoor Brook and River Culm, which both transport much finer overall effective suspended sediment loads, is around 30–35% (Table IV). These data have important implications for contaminant and nutrient transport, because a significant portion of the chemically active clay fraction is transported within medium (8–32 μm) and large (>32 μm) composite particles, whose hydrodynamic behaviour and fall velocity are markedly different to most of their constituent particles.

This paper has concentrated on the physical characteristics of composite particles, especially their clay content. However, further work is needed to improve our understanding of the role of organic matter and aquatic microfauna in composite particle dynamics in a range of environments. Composite particles typically contain inorganic and organic materials, with the latter present in a wide variety of forms, including microfauna such as diatoms (which are clearly identifiable in the SEM images shown in Figures 1 and 8), a microbial component, and extracellular polymeric substances (Droppo, 2001). It has been argued that the extracellular polymeric material (or fibrils) can provide a binding framework for the floc or aggregate and will influence composite particle density and stability as well as providing an important high surface area medium for the adsorption of nutrients and contaminants from the water column (Droppo, 2001). Hydraulically sorted samples generated by the WEA could provide a useful basis for further investigation of the nature and role of organic materials and microfauna within composite particles of different sizes and densities, and this could lead to new insights into the relative importance of source-derived aggregates and flocculated particles in fluvial systems.

THE ROLE OF COMPOSITE PARTICLES IN SUSPENDED SEDIMENT FLUXES

In contrast to other methods that have been used to monitor the particle size characteristics of fluvial suspended sediment, the water elutriation apparatus provides a *time-integrated* sample and this may represent several hours of suspended sediment transport. It is therefore possible to sample most, if not all, of the sediment wave associated with a given flow event in catchments of this size. As suspended sediment transport in the study catchments is highly episodic, and significant fluxes are essentially limited to flood events (Walling *et al*., 1992), it can be argued that the mean effective size data derived from the water elutriation apparatus provide the best available estimates of the role of composite particles in the longterm suspended sediment fluxes from these catchments (Figure 6).

The recognition of catchment-specific time integrated effective particle size signatures in the Exe basin means that these data are especially suited for sediment flux calculations and budgeting studies of sediment-associated transport. Thirty-six flood events incorporating a representative range of magnitudes were sampled between October 1991 and January 1993 and these data have been used to estimate the contribution of different effective size classes to the overall suspended sediment load for each catchment. The mean annual suspended sediment yields of these catchments have been estimated using continuous monitoring with optical turbidity meters (Walling and Webb, 1987) and these data are shown in Table V. The importance of each effective size class in the annual flux of suspended sediment in each catchment is also shown in Table V. For the River Exe at Thorverton, over 5500 t of suspended sediment is transported within the effective size classes >32 μm, while the <8 μm class (25·6%) accounts for over 4000 t. Much of the annual suspended sediment load at Rewe (River Culm) is transported in the effective classes <16 μm (>4200 t), while the effective classes >16 μm are dominant in the River

Table IV. Mean values for the contribution (%) to total clay transport of each effective size class

River	< 8 μm	8–16 μm	16–32 μm	32–63 μm	> 63 μm
River Exe ($n = 10$)	48·7	13·9	20·8	11·9	4·6
River Dart ($n = 10$)	41·2	19·0	17·5	15·5	6·9
River Culm ($n = 8$)	66·2	21·0	8·5	3·6	0·8
Jackmoor Brook ($n = 8$)	62·9	15·2	12·4	5·5	4·1

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Table V. The contribution of each effective size class (μm) to the mean annual suspended sediment load of the four study catchments. All values in tonnes per year

River	Suspended sediment load	< 8 µm	8–16 µm	16–32 µm	32–63 µm	> 63 µm
Jackmoor Brook	294	121·6	54·1	54·8	30·2	33·3
River Dart	2668	722·5	498·9	564·5	584·8	297·2
River Culm	5460	2901·4	1361·7	702·2	379·5	115·2
River Exe	16828	4304·6	2608·3	4456·1	3545·7	1913·3

Dart at Bickleigh. Figure 6 highlights the fact that discrete particles, <8 µm in size, dominate the absolute particle size characteristics of all four rivers in the Exe basin. However, it is important to appreciate that, due to the presence of composite particles, the proportion of the suspended sediment load transported within composite particles >8 µm in size is 58% for the Jackmoor Brook (170 t yr^{-1}), 72% for the River Dart (1920 t yr^{-1}), 74% for the River Exe (12 451 t yr^{-1}) and 45% for the River Culm (2457 t yr^{-1}). In combination with information on sediment quality, these data could provide the basis for further investigation of sediment-associated contaminant or nutrient fluxes from these catchments.

CONCLUSIONS

The field-based WEA developed by the authors generates consistent and reliable data on both the time-integrated effective and absolute particle size characteristics of fluvial suspended sediment. It offers insights into suspended sediment properties that are not attainable using other methods. Data from four subcatchments in the Exe basin reveal distinctive spatial patterns in the effective particle size characteristics of suspended sediment, while the absolute particle size properties typically show much less spatial variability. The latter are comparable with absolute size data collected with traditional sampling methods from a range of large UK rivers. The data provided by the WEA for the rivers in the Exe basin indicate that it may be possible to identify characteristic effective particle size signatures for river basins that can be used for the calibration of sediment transport and deposition models (e.g. Nicholas and Walling, 1986) and nutrient and contaminant budget studies. Investigations of interstorm and intrastorm variability in the effective particle size characteristics of suspended sediment in the Exe basin have been reported by Phillips and Walling (2005). The WEA is not suitable for such comparisons, as the time-integrated sampling averages out any intrastorm changes for a given sampling period.

The WEA is unique in that it performs passive hydraulic separation of floodwaters so that the physical, chemical and biological properties of composite particles of different sizes and densities can be studied. This paper has focused on some of the physical properties and it has been shown that clay grade material forms an important component of each effective size class in the sediment load of the Exe basin catchments. Clay content may be an important control on composite particle form and stability and sediment-associated contaminant loading. Much of the clay-grade material is transported within composite particles >8 µm in size. These findings have important implications for the development of channel and floodplain sedimentation models. Empirical and theoretical studies of contaminant and nutrient budgets in river systems also need to incorporate data on the effective particle size characteristics of the suspended sediment load. Sediment monitoring programmes need to provide realistic and representative effective particle size data for a range of fluvial environments that can be used in the development of numerical models.

The results presented in this paper provide a detailed and systematic evaluation of the time-integrated nature of the particle size characteristics of fluvial suspended sediment, and the findings have led to important advances in our understanding of the significance and nature of composite particles. Several important avenues for further research have also been highlighted. The marked spatial contrasts in these effective particle size signatures contrast with the lower variability evident in the absolute particle size datasets. This observation suggests that there are marked spatial variations in composite particle formation related to catchment characteristics and the processes involved require further research.

It is important to appreciate that we need to obtain reliable and representative data for both the absolute and effective particle size characteristics of fluvial suspended sediment if we are to advance our understanding of river system sediment delivery dynamics from source to sink. The former will commonly relate to the nature of catchment source materials as well as grain-size selectivity and delivery processes during transport (see Walling *et al.*, 2000). The effective size distribution will provide a more meaningful representation of the hydrodynamic behaviour of the suspended sediment load during transport. The distinction between the two distributions is not always clear cut, as both are dynamic and will change during the course of a flood event as sediment sources become exhausted, for example, and flow hydraulics and shear stresses change. Further work is required on the relationship between catchment characteristics, flow conditions and the relative importance of aggregates and flocs in fluvial systems.

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