



# The sources and dynamics of fine-grained sediment degrading the Freshwater Pearl Mussel (*Margaritifera margaritifera*) beds of the River Torridge, Devon, UK

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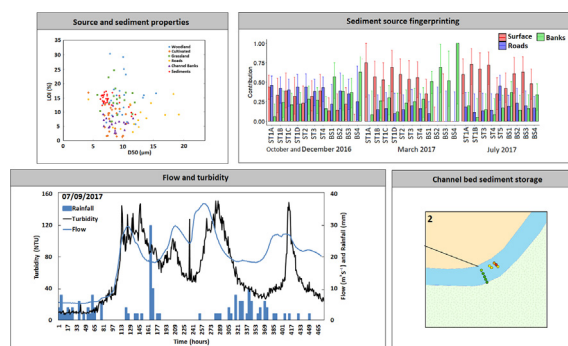
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## HIGHLIGHTS

- Fine sediment is degrading Freshwater Pearl Mussel habitats in the River Torridge.
- Roads, woodland, channel banks and agriculture were all major sediment sources.
- Bank erosion contributed more to bed sediment than suspended sediment.
- Turbidity was high in storm events and dominant sediment sources change over time.
- High turbidity peaks were elongated in duration.

## GRAPHICAL ABSTRACT



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## ABSTRACT

The Freshwater Pearl Mussel (*Margaritifera margaritifera*) is an endangered organism across its entire range. It has a complex life cycle and stringent habitat requirements and is therefore an indicator species for the general ecosystem health of host rivers. Whereas historical intensive pearl fishing contributed to population declines, excess nutrient and sediment loss associated with current land use pressures in host river catchments, including modern intensive farming practices, are now highlighted as primary contributory factors. Accordingly, this study investigated the sources and dynamics of fine-grained sediment sampled in the mussel beds of the River Torridge, SW England. Sediment source fingerprinting using a combination of colorimetric and radiometric tracers to construct different composite signatures revealed the importance of roads both as a sediment source and delivery pathway for fine-grained sediment mobilised from fields predominantly supporting lowland livestock farming. Grassland fields with evidence of soil poaching were highlighted as important sediment sources, but equally, riparian woodland was also identified as important, especially during the latter stages of consecutive runoff events when its rainfall buffering capacity was exceeded. Bed sediment storage levels (median up to 393 g m<sup>-2</sup>) were found to be low (41st percentile) compared to typical values reported by a recent strategic scale survey across England and Wales, whereas elevated turbidity peaks were shown to be long duration (days) in conjunction with consecutive days of rainfall and corresponding runoff events. Hysteresis patterns varied but were generally clockwise during the largest runoff events associated with consecutive rain days; again, suggesting mobilisation of sediment from proximal woodland sources following exceedance of rainfall buffering

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capacity. In combination, the data assembled by this study provides a basis for planning sediment control measures for protecting the Freshwater Pearl Mussel (FPM) beds from excessive fine-grained sediment inputs associated with the intensive use of primarily grazing land.

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## 1. Introduction

Globally, diffuse pollution from agriculture has been identified as a pervasive problem (Novotny, 1999). Excess losses of fine-grained sediment, as well as transfers of nutrients, pesticides and organic wastes originating from modern farming have been shown to be detrimental to water quality and aquatic biodiversity (Johnston and Dawson, 2005; Foley et al., 2005; Donald and Evans, 2006; Kemp et al., 2011; Jones et al., 2012a, 2012b, 2014), contributing to the decline of many threatened species (Richter et al., 1997; Suttle et al., 2004).

A case in point is the Freshwater Pearl Mussel (FPM; *Margaritifera margaritifera*) which is one of the most endangered of all aquatic organisms (Bogan, 1993, 1998; Williams et al., 1993; Neves et al., 1997; Strayer et al., 2004; Geist and Auerswald, 2007). The FPM was previously widespread and abundant, covering an area from the Arctic and temperate regions of Western Russia through Europe to the NE seaboard of northern America (Jungbluth et al., 1985). However, the decline of the FPM has been reported across its entire range (Baer, 1969; Jungbluth, 1971; Valovirta, 1977; Bauer, 1979, 1986, 1989; Dettmer, 1982; Wells et al., 1983; Young and Williams, 1983; Young, 1991; Young et al., 2000; JNCC, 2018). Only a few populations in Europe exhibit successful recruitment with the risk of extinction therefore being a reality (Ziuganov et al., 1994; Young et al., 2001; Geist, 2005; Geist and Auerswald, 2007). The FPM population in the Fichtelgebirge in northern Bavaria was reported to be 700,000 in the early 20th century (Meissner, 1914), but only 20,000 sixty years later (Bauer, 1979). Equally in the UK, the FPM was once widely abundant. Today, however, all populations are at risk of extinction with virtually no active recruitment (Chesney and Oliver, 1998; Skinner et al., 2003). As a result, FPM are currently protected by legislation including Schedule 5 of the Wildlife and Countryside Act (1981) in the UK and annexes II and V of the EU Habitats and Species Directive (Skinner et al., 2003). They are also listed on the IUCN Invertebrate Red List (IUCN, 1990).

Polluted host rivers have been highlighted as suffering declines in the FPM (Baer, 1970; Bauer, 1980; Dettmer, 1983; Waechtler, 1986), with factors such as eutrophication and enrichment of fine-grained sediment levels in the river substrate being highlighted as important (Phillips, 1928; Bjork, 1962; Bauer, 1988; Hastie et al., 2000).

The key habitat requirements for FPM are fast-flowing, shallow, riffles with a well-oxygenated coarse gravel or cobble substrate. For most of their >100 years lifespan, they are part buried within the substrate filter feeding organic particles suspended in the water column (Bauer, 1992). An important control on substrate habitat quality concerns hyporheic exchanges between free-flowing water and the interstitial zone (Geist and Auerswald, 2007). Insufficient exchanges encourage anoxic conditions and the resulting oxygen deficits can impact on the growth of FPM (Belanger, 1991; Geist and Auerswald, 2007). Here, increased ingress of fine-grained sediment clogs interstices and reduces exchange rates (Richards and Bacon, 1994) as well as impeding burrowing, respiration and feeding by FPM (Beasley, 1996).

Excessive fine-grained sediment represents a leading threat to successful FPM reproduction since it smothers their channel bed habitat causing direct impacts such as suffocation (Bauer, 1988; Buddensiek et al., 1993; Box and Mossa, 1999; Moorkens, 2000; Hastie et al., 2000; Hansen et al., 2016). Indirectly, excess fine-grained sediment reduces light penetration and photosynthesis, thereby reducing food resources for both host fish and FPM (Munn and Meyer, 1988; Box and Mossa, 1999). Fine-grained sediment can also act as a vector for nutrients and

contaminants which can impact both juvenile and adult FPM (Bauer, 1988; Österling et al., 2010). The highly organic sediments observed in areas with intensive modern agriculture, and especially livestock farming, will often have a high oxygen demand (Chevalier et al., 1984; Greig et al., 2005; Collins et al., 2017; Sear et al., 2017). Substrate stability is also important for FPM survival since it avoids scouring and drift to less favourable sites (Johnson and Brown, 2000; Hastie et al., 2001; Gangloff and Feminella, 2007). The importance of excess fine-grained sediment supply for the decline of the FPM is underlined by revelation that the youngest mussels found in a reach are older with higher levels of sedimentation, suggesting high juvenile mortality (Österling et al., 2010).

Fine-grained sediment delivery to rivers is estimated to have increased by 10–20% relative to the pre-agricultural landscape (Wilkinson and McElroy, 2007) and Collins and Zhang (2016) reported that median sediment delivery to UK river channels of <61 t·km<sup>-2</sup>·yr<sup>-1</sup> above background rates can be attributed to modern agriculture post World War II. It has long been recognised that the optimum way to mitigate excessive loss of fine sediment is at its key sources. This requires the identification of the major sediment sources present in a catchment or a locality used by a particular aquatic organism (Collins et al., 2011). Here, however, it is also important to consider source-specific toxicity since recent work by Sear et al. (2016) showed that in the case of Brown Trout and Atlantic Salmon, increased toxicity was attributable to a higher organic matter concentration and therefore oxygen demand.

Given the reported decline of the FPM across its range, and the concomitant ongoing need to assemble data for informing improved management of host rivers including those in the UK, this study undertook an investigation into the fine-grained sediment degrading the FPM beds within the River Torridge, Devon, UK. More specifically, the objectives were to determine temporal changes in the sources and quantities of fine-grained sediment transported in the river channel and deposited within its bed along a reach hosting FPM. The methods used in this case study are applicable for investigating similar issues at other locations where rivers host the FPM.

## 2. Study area

The River Torridge (Fig. 1; 857 km<sup>2</sup>) is located in the southwest of the UK. The catchment experiences high average annual rainfall of 1053 mm (measured at North Wyke, Okehampton). There is considerable variability in annual rainfall across the catchment, with ~1000 mm close to the river mouth, 1200 mm in its western headwaters and up to 2200 mm at its highest elevations (Nicholls, 2000). The catchment geology (Fig. 1) is composed of Carboniferous sandstones, siltstones and mudstones, apart from in a small area at high altitude in the south of the catchment underlain by granite, doleritic and basaltic rocks. This igneous area forms the northern most extent of Dartmoor. The generally poorly permeable geology leads to low groundwater water storage and flashy patterns of river flow (National Rivers Authority, 1990).

Land use is primarily (~80%) grassland for sheep and cattle grazing, but a small amount of the catchment is used for the cultivation of maize and winter barley (~15%) (Nicholls, 2000). Large areas of the catchment (5–10% in the upper Torridge) have been drained since 1952 to improve the quality of grazing land, and the areas of grazing land and stocking densities of livestock have increased (Nicholls, 2000; National Rivers Authority, 1990). Woodland is present in a partial

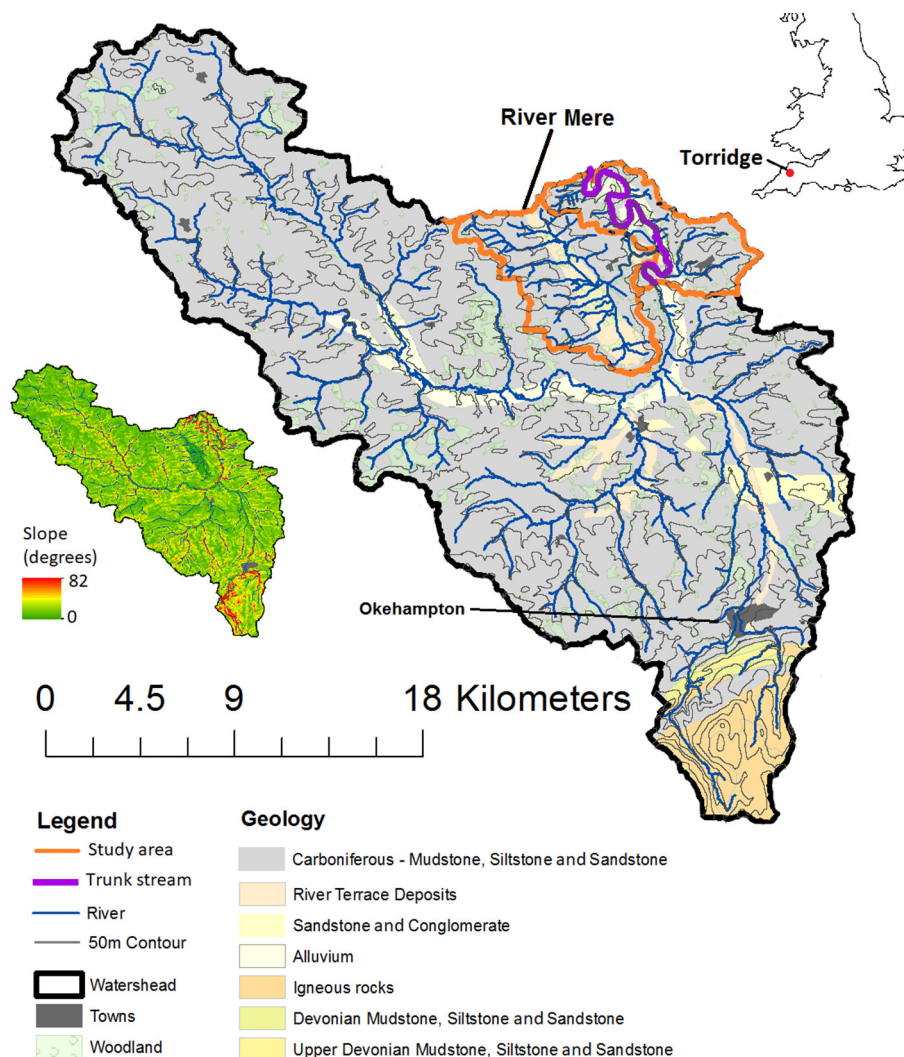


Fig. 1. The River Torridge catchment, showing the study area.

corridor adjacent to river channels where the land is steeply sloped, although it only covers a small proportion (<5%) of the total catchment area. Highly sloped woodland is present adjacent to much of the FPM priority area. Because of the steep slope of the woodland, many small channels flow through it to the trunk stream. Roads and farm tracks are present in high densities throughout the catchment and many of these contain large quantities of bare sediment deposited by surface runoff or from agricultural machinery. Many of the roads are sunken beneath the elevation of adjacent fields promoting erosion next to gates and sediment transport from fields to the roads. Channel banks are mostly shallow and composed of un-weathered rock, apart from where alluvial deposits are present. The river channel is highly confined by heavily sloped bedrock, creating a narrow v-shaped valley. Alluvial floodplain deposits are intermittently present where valley width allows. In many places poaching by cattle on the alluvial deposits can be observed; however, recent catchment management has focused on the installation of riparian fencing to exclude livestock.

The study reported in this paper focused on the River Mere tributary and the reach of the River Torridge main stem close to its confluence, as this area covers part of the FPM priority area. This spatial focus allowed for an intensive sediment source sampling campaign which would not be possible in the very large catchment as a whole, due to the limited resources available for the work. However, previous research into fine sediment sources and dynamics in the upper River Torridge has been conducted by Nicholls (2000), thereby reducing the need to study this

area upstream of the priority area in detail. The River Mere has been shown to experience high suspended sediment loads in comparison with the rest of the River Torridge, highlighting the need for its investigation, as directed by the Devon Wildlife Trust. As with the River Torridge as a whole, the River Mere tributary is primarily used for low-land grazing livestock farming and has shallow channel banks which are often composed of exposed bedrock.

FPM populations in the River Torridge are the fourth largest in England; however, like many UK populations, they have not successfully reproduced since the 1960s. There is currently an ongoing three-year project run by the Freshwater Biological Association which aims to safeguard the future of FPMs through river restoration and community engagement (Devon Wildlife Trust, 2018). Restoration work includes the identification of threats to the FPM populations and improvement works such as encouraging best farming practice and the installation of on-farm mitigation measures (see examples of such measures in Collins et al., 2016; Zhang et al., 2017).

The River Torridge has a high suspended sediment yield of  $89 \text{ t} \cdot \text{km}^2 \text{ yr}^{-1}$  (Nicholls, 2000) and contributions to the sediment yield in the upper River Torridge have previously been estimated at 2% woodland, 48% pasture, 29% cultivated land and 21% channel banks (Nicholls, 2000). This previous work, however, did not focus specifically on the key sources of fine-grained sediment impacting upon the FPM beds, and nor were roads investigated as a potential sediment source. More recent studies in the UK have identified road sediment as an

important source of the fine-grained sediment sampled in lowland rural river channels (e.g. Gruszowski et al., 2003; Collins et al., 2010a, 2010b, 2012).

This study was preceded by a period of low rainfall with few high flow events (Fig. 2). A large storm event occurred in late November 2016 causing the highest flow event during the study period. Between December 2016 and March 2017, there was a series of six high flow events and river levels remained high throughout this time. Between March 2017 and July 2017, flows returned to mostly low levels despite the largest daily rainfall event being observed in early May 2017.

### 3. Materials and methods

#### 3.1. Field sampling and monitoring

To obtain a database of sediment source samples most representative of the material being delivered to the river channel, samples were only collected from locations where there was both visible erosion and good structural connectivity with the river (Fig. 3). In the cases of roads, this was where bridges crossed the river and runoff and sediment could enter the channel. For channel banks, poached or actively eroding areas of the bank were sampled. In woodland ditches, small channels and tracks were sampled, as well as visibly disturbed soil on sloping land close to the river channel. For grassland and cultivated land, visibly disturbed or bare soil was collected to an approximate 2 cm depth from locations where it could potentially be transported by surface or drain flows into the river channel. This was often where land was trampled and poached by livestock or disturbed by vehicle tramlines close to gates and fences. The source samples were retrieved between November 2016 and February 2017.

At the lower end of the FPM priority area, fine-grained sediment deposited on the channel bed was sampled on four transects spaced evenly using the method of Lambert and Walling (1988). The mass of sediment stored within the bed at each sampling location was calculated and recorded and a composite sample of sediment from each of the four transects was retained for sediment source tracing. The bed sediment sampling captured a range of channel habitats, including sand deposits on the inside of meanders and fast flowing riffles where bedrock was exposed.

Samples of suspended sediment were obtained using time-integrating traps based upon the design of Phillips et al. (2000). Four traps were placed upstream of the FPM priority area to characterise the sediment originating from the middle and upper River Torridge

catchment (ST1; Fig. 3). Three traps (ST2–4, Fig. 3) were also positioned in sequence through the main stem reach of the River Torridge to characterise any potential changes in sediment sources. A single trap was also placed in the River Mere tributary (Fig. 3; ST5) to characterise localised sediment inputs. The sediment traps were installed in early November 2016 and were subsequently emptied in December 2016, March 2017 and July 2017 capturing the first and largest high flow in November 2016, the sequence of high flows between December 2016 to March 2017 and the period of lower flows between March and July 2017 (Fig. 2). The trap in the River Mere tributary was, however, damaged during the first two sampling periods; so, a sample was only available for July 2017. Channel bed sediment samples were retrieved in October 2016, March 2017 and July 2017.

A Seametrics 'Turbo' turbidity smart sensor was installed at Torrington approximately 2 km downstream of the channel bed sediment sampling locations to obtain a 15-min record of turbidity from between the 15th December 2016 and 31st October 2017. The sensor was installed in a section of pipe to protect it from debris and was wiped prior to each measurement. Flow data for the study period was obtained from the Environment Agency stage gauging station which was also located at Torrington. Hourly rainfall data was obtained from the Met Office monitoring station located at Rothamsted Research, North Wkye, which is approximately 5 km from the Torridge catchment.

#### 3.2. Laboratory analyses

Sediment and source samples were dried at 105 °C for 24 h and gently disaggregated using a pestle and mortar. The samples were then dry sieved through a 63 µm stainless steel mesh, before being mixed with distilled water and wet sieved to <25 µm. The <25 µm fraction was selected for analysis to reduce the potential for particle size related changes to the tracer concentrations (Horowitz, 1991; Lacey et al., 2017) to impact on the sediment source apportionment estimates. Additionally, most (~80%) of the <63 µm fraction of the source and sediment samples fell within this range. After wet sieving, the <25 µm fraction was left to settle overnight before excess water and any suspended organic particles were poured off and each sample was again oven dried at 105 °C.

Sediment colour has been used successfully as an inexpensive tracer as it has been shown to be representative of numerous sediment characteristics, which can provide discrimination between multiple sediment sources (Pulley and Rowntree, 2016). After sample preparation was completed, each individual sample was packed into a transparent

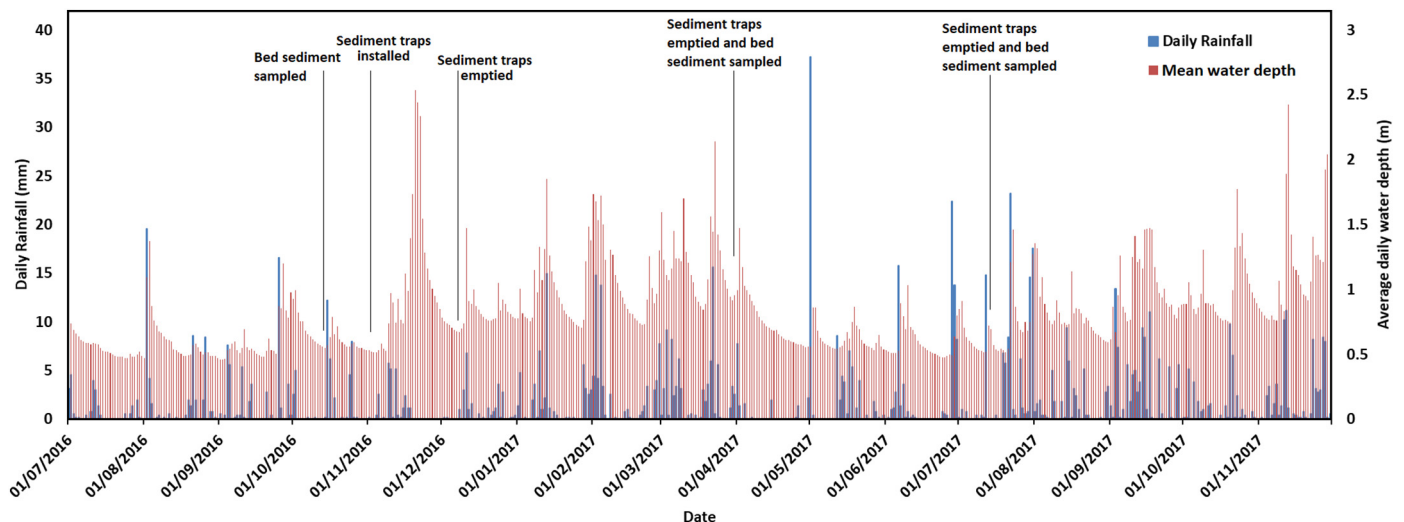


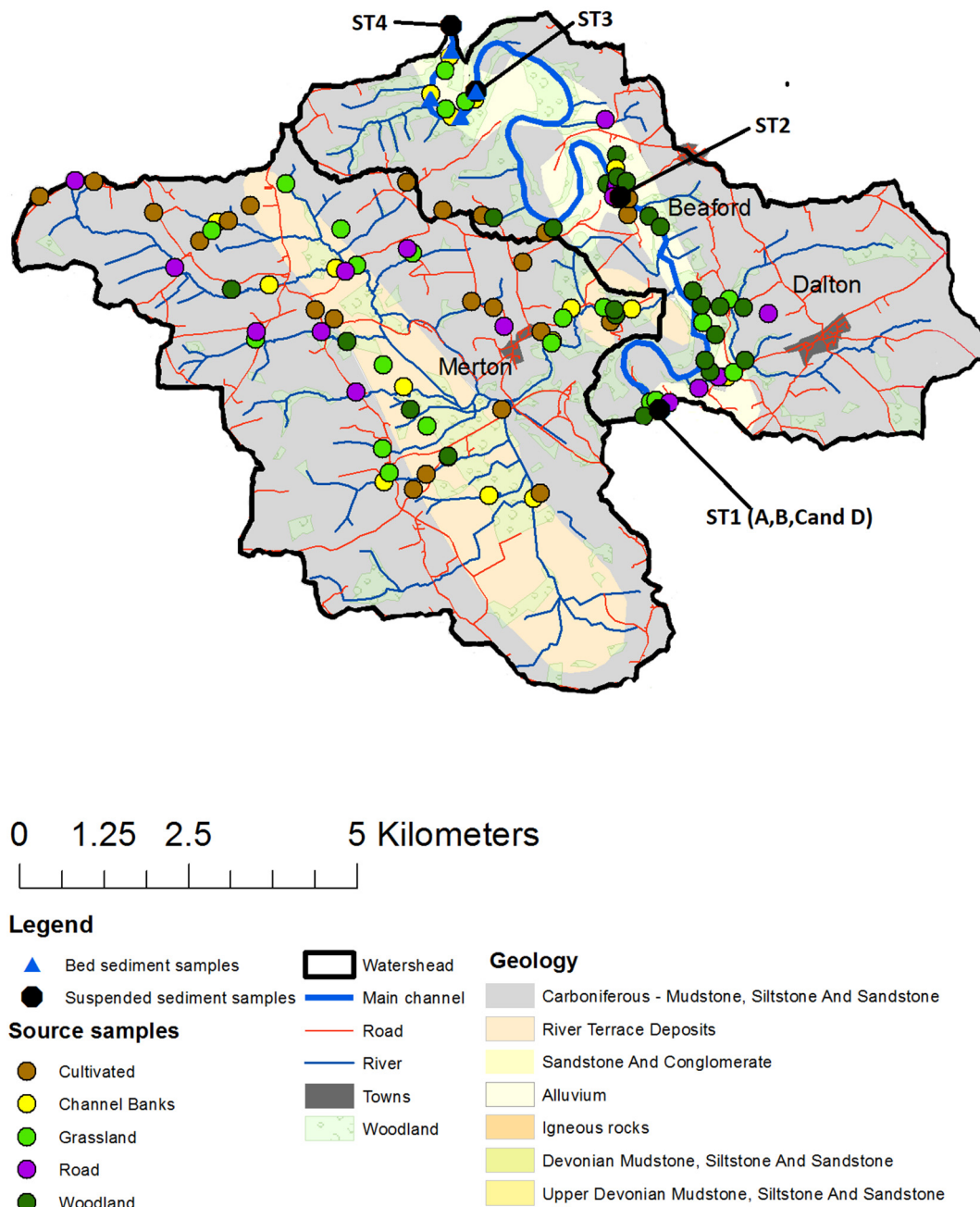
Fig. 2. Rainfall and sediment sampling dates during the study period.

polythene bag and scanned using a Ricoh MPC3504 colour scanner. The intensity of reflected red, green and blue light (0 to 255) using the RGB colour model was quantified from the scanned images using GIMP 2 open source image editing software. The Saturation Index (SI), Hue Index (HI), Colouration Index (CI) and Redness Index (RI) as well as HRGB, IRGB and SRGB were calculated on the basis of their sensitivity to specific soil components (Ray et al., 2004; Viscarra Rossel et al., 2006).

Fallout radionuclides have been shown to be strong indicators of sediment source (Walling and Woodward, 1992; Evrard et al., 2016). High activities of  $^{210}\text{Pb}_{\text{un}}$  and  $^{137}\text{Cs}$  have been found in undisturbed woodland and grassland topsoils, whilst activities are lower in cultivated land and zero in channel banks that have not been exposed to atmospheric fallout (Walling, 2004; Walling and Woodward, 1992). Subsamples of the source materials and sediment samples weighing

approximately 3 g were packed to a depth of 4 cm in PTFE tubes and sealed with paraffin wax. They were then left to equilibrate for 21 days before analysis (Pennock and Appleby, 2002). The activities of  $^{137}\text{Cs}$ ,  $^{210}\text{Pb}_{\text{un}}$ ,  $^{234}\text{Th}$ ,  $^{235}\text{U}$ ,  $^{214}\text{Pb}$ ,  $^{228}\text{Ac}$ ,  $^{212}\text{Pb}$  and  $^{40}\text{K}$  were measured using an Ortec hyper-pure germanium (HPGe) detector with two day count times (Foster et al., 2007).

Sediment organic matter content and particle size have been shown to affect its potential to cause harm to FPM (Österling et al., 2010). In addition, changes to sediment particle size and organic matter concentration during sediment erosion, transport and deposition, can introduce changes to its colour and radionuclide activities, representing a source of uncertainty associated with their use in sediment source tracing studies (Ben-Dor et al., 1998; Madruga et al., 2014). For these reasons, both the particle size distribution and organic matter concentration of the source and sediment samples were quantified.



**Fig. 3.** The priority study area for FPM including the River Mere tributary sub-catchment and reach of the main channel of the River Torridge. Locations of source and sediment sampling are shown. Refer to Fig. 1 for spatial context within the River Torridge catchment.

To measure the particle size distributions, organic matter was first removed using 30% hydrogen peroxide added to a ~0.2 g subsample of the source or sediment. The samples were left at room temperature for 24 h and then heated at 70 °C until bubbling had stopped. Immediately prior to analysis, 5 ml of 3% sodium hexametaphosphate solution was added and the samples were ultrasonically dispersed for two minutes (Blott et al., 2004; Gray et al., 2010). Sediment particle size distribution was quantified using a Malvern Instruments laser granulometer equipped with a Malvern Hydro 2000 unit. The  $D_{10}$ ,  $D_{50}$  and  $D_{90}$  of the samples were recorded. The organic matter content of the samples was determined using loss on ignition (LOI). Here, approximately 2 g of each sample were burnt in a Carbolite muffle furnace at 450 °C for 4 h.

## 4. Data processing

### 4.1. Organic matter and particle size effects on sediment tracers

To determine the effects of organic matter and particle size on the source sample tracer concentrations, a Spearman rank correlation analysis was performed between LOI,  $D_{50}$  and each tracer. Bi-plots were then produced between tracers representative of the entire dataset, and  $D_{50}$  and LOI, highlighting differences between the relationships in the different source groups.

To explore the relationship between particle size and sediment colour more thoroughly, a composite mixture of all samples in each source group was further fractionated to 25–10  $\mu\text{m}$  and <10  $\mu\text{m}$  by wet sieving and timed settling in a measuring cylinder. Due to resource limitations, this was not possible for the radionuclides. A bi-plot of red and blue was used to determine the effects of particle size on the colour of the individual source groups and to determine if changes to sediment particle size could potentially mask the sediment provenance signal represented by its colour. The  $D_{50}$  and LOI of each sediment sample was then compared to identify any differences between the bed and suspended sediment samples and between the different sampling periods. A bi-plot of LOI and  $D_{50}$  was used to compare the source groups to the sediment samples to determine if there had likely been any change to the sediment sample LOI or  $D_{50}$  during its transport to, and deposition within, the study reach hosting the FPM.

### 4.2. Sediment source fingerprinting procedure

The sediment source fingerprinting procedure used the new SIFT (Sediment Fingerprinting Tool; Pulley and Collins, 2018a) following the methods of Pulley and Collins (2018b). A brief summary of the SIFT procedure used is provided in the online supplementary material. Other tools can be used to process fingerprinting data (e.g. Gorman Sanisaca et al., 2017).

## 5. Results

### 5.1. Source and sediment organic matter content and particle size distributions

Significant ( $p < 0.05$ ) relationships were found between LOI and most tracers measured in the source samples (Supplementary Table 1). A higher organic matter concentration resulted in less light being reflected from the samples causing reduced values of most colour tracers. Fallout radionuclides ( $^{210}\text{Pb}_{\text{un}}$  and  $^{137}\text{Cs}$ ) were positively correlated with LOI, which likely reflects their abundance in undisturbed woodland and grassland topsoils. Lithogenic radionuclides were negatively correlated with LOI, possibly reflecting their dilution by organic matter. Most tracers were also significantly correlated with  $D_{50}$ , although correlation coefficients were lower than those for LOI. A coarser particle size was associated with lower values of all tracers apart from  $^{210}\text{Pb}_{\text{un}}$  and RI. It is of note that  $D_{50}$  and LOI were not significantly correlated with each other, suggesting that finer particles do not have a significantly higher LOI as may have been expected.

LOI was highest in woodland samples; this source therefore had the lowest values for red and highest  $^{137}\text{Cs}$  activities (Fig. 4a, b). Road samples were notable as they fell outside of the linear relationship between colour tracers and LOI found in the other sources with less reflected red light for a given LOI. Road samples also had a notably coarser  $D_{50}$  than the other sources and low red values, partially explaining this difference (Fig. 4c, d).

The effects of particle size on the colour of the composite mixtures of source samples in each group were generally smaller than differences between the source groups (Supplementary Fig. 1). Particle size had a

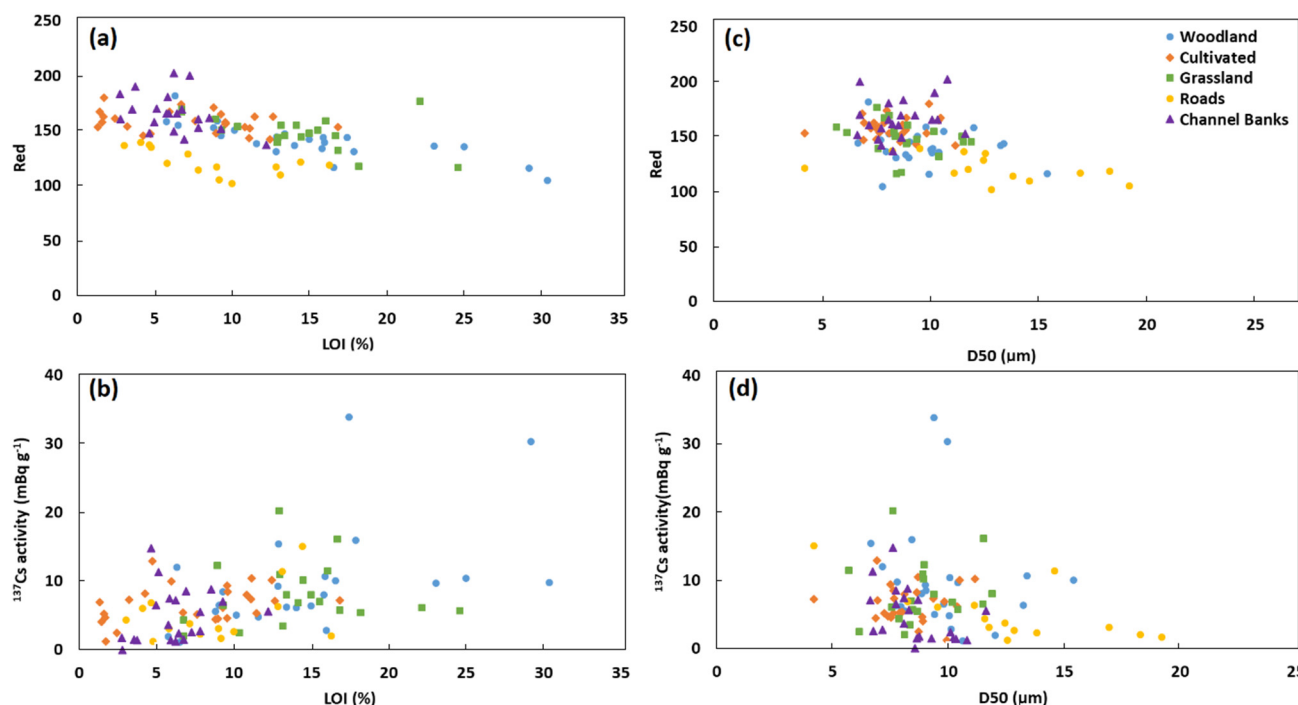


Fig. 4. The relationship between LOI,  $D_{50}$  and red and  $^{137}\text{Cs}$  activity in the sediment source samples.

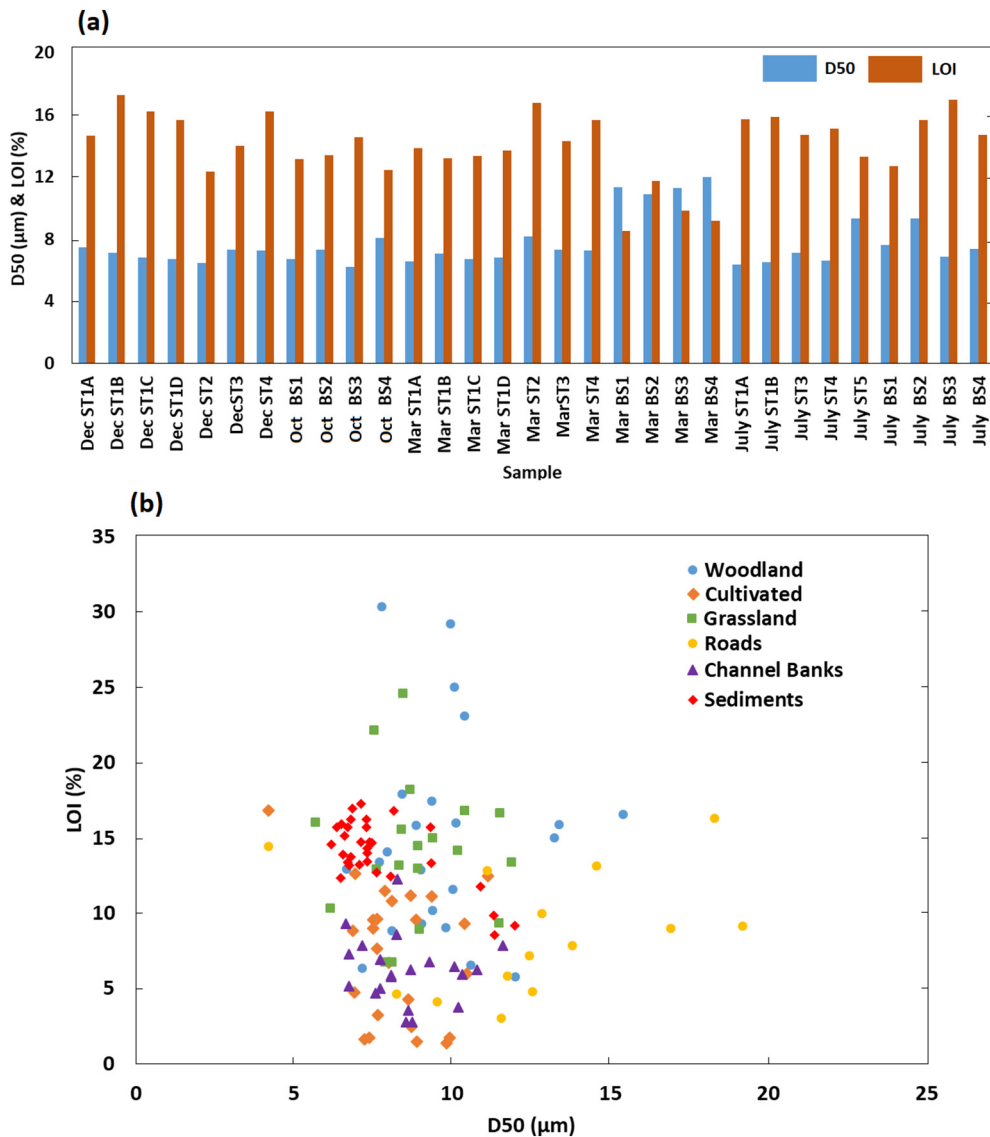


Fig. 5. The  $D_{50}$  and LOI of the suspended (ST) and channel bed (BS) sediment samples (a), and a bi-plot of  $D_{50}$  and LOI for the source and sediment samples (b).

particularly large effect on road sediments at  $<10 \mu\text{m}$ , possibly reflecting the preferential adsorption of material from vehicle wear and exhaust emissions on the greater surface area of finer particles.

The  $D_{50}$  and LOI of the sediment samples within the  $<25 \mu\text{m}$  size fraction was highly consistent, with the exception of the channel bed sediment retrieved during March 2017, which had a high  $D_{50}$  and low LOI (Fig. 5a). Both the LOI and  $D_{50}$  of the sediment samples fell within the range found in the source samples, although the  $D_{50}$  was finer and the LOI higher than the median values for the entire source sample dataset (Fig. 5b).

## 5.2. Sediment source tracing

### 5.2.1. Source group classification and misclassified samples

The initial linear discriminant analysis (LDA) indicated that there is good discrimination between all sources apart from cultivated land and grassland using the available tracers (Fig. 6). Roads and channel banks were particularly strongly discriminated. An initial trial of the sediment source fingerprinting procedure identified that un-mixing models with more than three source groups were unable to accurately apportion the composition of virtual mixtures. Because of this, two

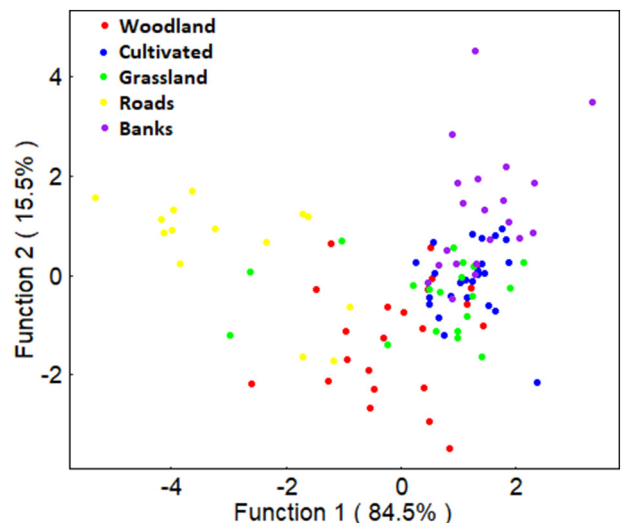


Fig. 6. Bi-plot of the two largest discriminant functions of the initial LDA.

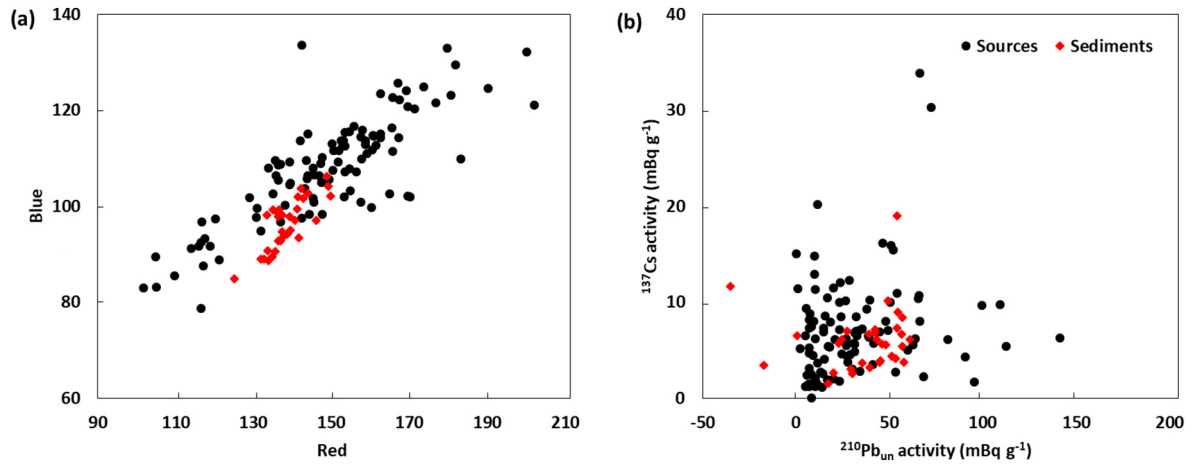


Fig. 7. Bi-plots of source (black) and sediment (red) sample tracers.

different source group classifications each consisting of three source groups were used in the fingerprinting procedure:

Classification 1: Surface sources (woodland, cultivated and grassland), Roads, and Channel banks.

Classification 2: Woodland, Roads, and Agriculture (cultivated, grassland and channel banks).

Three road samples had properties more comparable to woodland sediments which was likely due to the build-up of leaf litter and eroded soil on the road. These samples introduced significant within-source group variability to the roads source group and therefore were reclassified as woodland. One farm track sample which was retrieved from a grassland field was better classified into the road group so was also reclassified accordingly. Other potentially misclassified samples (22 for classification 1 and 14 for classification 2) were judged to be a result of natural within-source variability and therefore were not reclassified.

5.2.2. Tracer variability ratios

Variability ratios for the colour tracers were high with differences between source group medians being a mean of 1.7 times higher than mean within-source group variability (Supplementary Table 2). HRGB was the only colour tracer that failed to achieve a mean variability ratio higher than 1 in both source classifications. Mean variability ratios for lithogenic radionuclides, except for  $^{40}\text{K}$ , in Classification 1, were lower than the threshold of 1 for all tracers, indicating little difference in their activities between sources. As a result,  $^{226}\text{Ra}$ ,  $^{228}\text{Ac}$ ,  $^{234}\text{Th}$ ,  $^{235}\text{U}$  and  $^{212}\text{Pb}$  were removed from further use.

Table 1

The percentage of sediment samples falling within the specified range test thresholds of the source groups.

	Classification 1		Classification 2	
	Percent within medians $\pm$ one MAD	Percent within minimum - maximum	Percent within medians $\pm$ one MAD	Percent within minimum - maximum
Red	100	100	100	100
Green	100	100	100	100
Blue	100	100	100	100
HRGB	100	100	90	100
IRGB	100	100	100	100
SRGB	100	100	100	100
SI	100	100	77	100
HI	100	100	100	100
CI	100	100	100	100
RI	100	100	100	100
$^{210}\text{Pb}_{\text{un}}$	94	94	94	94
$^{137}\text{Cs}$	94	100	97	100
$^{40}\text{K}$	65	100	-	-

5.2.3. Tracer conservatism testing

The colour tracers of the sediment samples fell within the range of values measured for the source samples (Fig. 7a). However, values for blue light are close to the lower end of the acceptable range for many samples. Of the five samples with the lowest red and blue values, most were bed sediment samples from March 2017. The other bi-plots for colour tracers were comparable to that of red and blue. For  $^{210}\text{Pb}_{\text{un}}$  and  $^{137}\text{Cs}$ , sediment samples mostly fell within the range found in the source samples (Fig. 7b). However,  $^{210}\text{Pb}_{\text{un}}$  activities were low in three of the four March 2017 channel bed sediment samples. In the case of this bi-plot, there was not an  $r^2$  higher than the 0.8 threshold required, so the figure is only provided for reference.

All tracers fell within the median  $\pm$  one MAD range of the source groups for most sediment samples. All tracers, apart from  $^{210}\text{Pb}_{\text{un}}$  in two sediment samples fell within the minimum to maximum range found in the source samples (Table 1). There is therefore no indication of any substantial tracer non-conservatism other than in the March 2017 channel bed sediment samples.

5.2.4. Distributions of tracers in the source groups

Woodland was differentiated by its high  $^{137}\text{Cs}$  and  $^{210}\text{Pb}_{\text{un}}$  activities, and channel banks by their lower  $^{210}\text{Pb}_{\text{un}}$  and  $^{137}\text{Cs}$  activities (Supplementary Fig. 2). Approximately 50% of channel bank samples had high  $^{137}\text{Cs}$  activities, reflecting the likelihood that the channel banks are composed of recently deposited alluvium. All colour tracers separated the sources in the same way, with channel banks, agriculture and roads having the lowest or highest values and woodlands being intermediate between the two other sources.

Table 2

The results of the stepwise LDA with the percentage of source samples correctly classified into their respective groups and tracers selected as the optimum composite fingerprint.

Fingerprint	Correctly classified	Tracers
Classification 1		
Basic	80.8	Red, Green, SI, CI, RI, SRGB, $^{210}\text{Pb}_{\text{un}}$ , $^{137}\text{Cs}$ , $^{40}\text{K}$
Conservative	82.2	Red, Green, Blue, SRGB, IRGB, SI, HI, CI, RI, $^{137}\text{Cs}$
High variability	81.5	Red, Green, IRGB, SI, HI, CI, RI, $^{210}\text{Pb}_{\text{un}}$ , $^{40}\text{K}$
Classification 2		
Basic	79.5	Red, Green, SRGB, SI, CI, RI, $^{210}\text{Pb}_{\text{un}}$ , $^{137}\text{Cs}$
Conservative	80.8	Red, Green, Blue, IRGB, SRGB, SI, HI, CI, RI, $^{210}\text{Pb}_{\text{un}}$ , $^{137}\text{Cs}$
High variability	80.4	Green, Blue, SRGB, SI, HI, CI, RI, $^{210}\text{Pb}_{\text{un}}$ , $^{137}\text{Cs}$



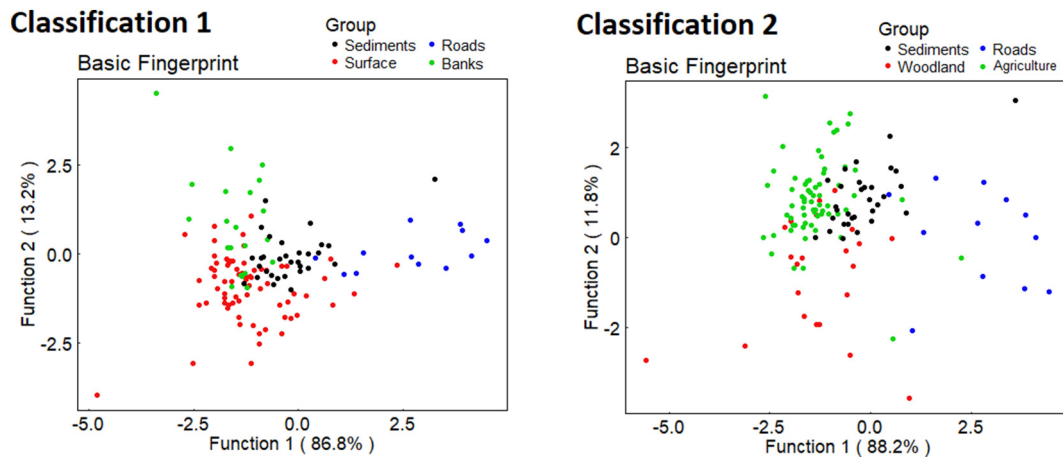


Fig. 8. Bi-plots of the two discriminant functions for the basic composite fingerprints, plots for the conservative and high variability fingerprints are provided in Supplementary Fig. 3.

### 5.2.5. Discriminant analysis

The linear discriminant analysis was able to classify correctly between 79.5% and 82.2% of source samples (Table 2). Both  $^{210}\text{Pb}_{\text{un}}$  and  $^{137}\text{Cs}$  were used in all composite fingerprints for Classification 2, but both were not used in every fingerprint for classification 1.  $^{40}\text{K}$  is only present in the Classification 1 fingerprints due to it failing the range test with Classification 2. Misclassified samples were primarily between channel banks and surface sources for Classification 1 and woodlands and agriculture for Classification 2.

Bi-plots of the two largest discriminant functions of each composite fingerprint show a good separation between the three sources, but some samples do overlap other source groups (Fig. 8; Supplementary Fig. 3). One sediment sample fell outside of the range of the source groups suggesting tracer non-conservatism. The sediment samples fell closest to surface and agricultural sources on these bi-plots, which also suggests a significant contribution from roads to some sediment samples. Contributions from channel banks and woodland were suggested to be more minor.

### 5.2.6. Virtual mixture apportionment

The un-mixing model produced similar results for the three composite fingerprints when apportioning the composition of the virtual mixtures; as such, only the results for the basic fingerprint are presented here (Fig. 9). The mean differences between the median contribution estimated by the model for each source and the actual mixture composition were 13.06% on the 0–100% contribution scale for the Basic fingerprint, 12.00% for the Conservative and 13.42% for the High variability fingerprint with Classification 1, and; 10.42% for the Basic fingerprint, 10.02% for the Conservative and 10.27% for the High variability fingerprint with Classification 2. The virtual mixtures therefore suggested that the apportionment of the dominant sources in each mixture is correct; however, the Classification 1 models underestimated contributions of surface sources by approximately 30% when contributions from this source are 100% and approximately 15% when contributions are 50%. An approximate 10–20% over and under estimated contribution from each source is also likely to occur when their actual contribution is close to 0% or 100%. The weighting of a variety of tracer combinations was trialled, but none were found to improve the accuracy of sediment source apportionment using virtual mixtures.

### 5.2.7. Sediment provenance

Almost 100% of model iterations passed the 0.35 GOF threshold for each sediment sample, apart from March 2017 sediment trap B and December 2017 sediment trap D for Classification 2, where close to 70% of iterations passed. The mean GOF of the samples passing the 0.35 threshold was close to 0.9 for all samples apart from March 2017 Bed

sediments 2, 3 and 4, where it was between 0.75 and 0.8 for five of the six composite fingerprints.

In December 2016, roads and surface sources were estimated to be dominant with a comparable contribution from woodland and agriculture (Fig. 10; Supplementary Fig. 4; Supplementary Fig. 5). Woodland was estimated to contribute a greater proportion of suspended sediment, and channel banks a greater proportion of bed sediment during this first sampling period.

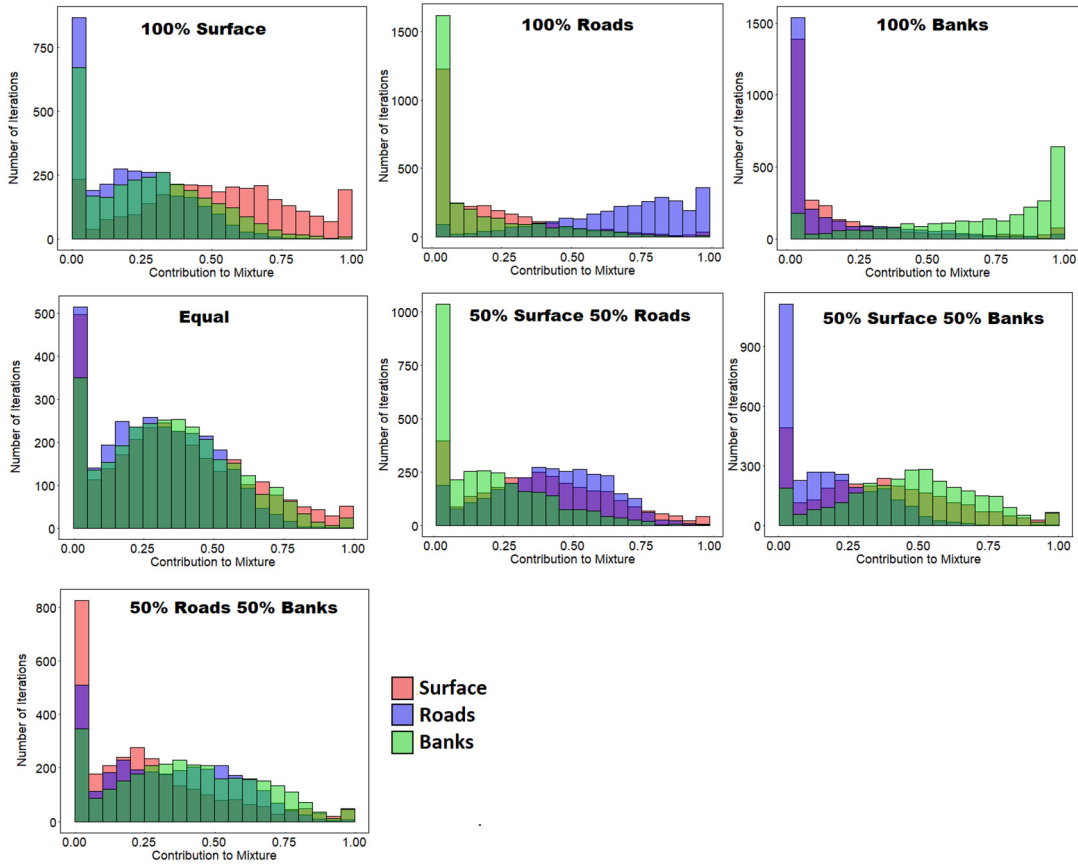
In March and July 2017, roads were a very minor sediment source and contributions from woodland and agriculture dominated. There is large range of uncertainty associated with the apportionment of woodland and agricultural sources; therefore, it is not possible to ascertain which of these is the dominant sediment source. However, the bi-plots (Fig. 8) suggest agriculture is likely to be a slightly more important source than the model outputs suggest. Channel banks were a more minor sediment source contributing ~20% of sediment. There is, however, a high estimated contribution from channel banks to the channel bed sediment samples collected in March 2017. This is likely an artefact of the coarser particle size and low organic matter content of these samples (Fig. 5) and may not reflect actual sediment provenance. There was, however, also a higher estimated contribution of channel banks to channel bed sediment during the other sampling periods suggesting channel banks are an important contributor to bed sediment along the FPM study reach. There was an approximately 25% higher contribution from roads and 25% lower contribution from woodland to the sediment originating from the River Mere tributary (ST5; Fig. 1) compared with the sediment retrieved from the River Torridge trunk stream.

### 5.3. Turbidity, rainfall and river flow

In high flow events, the durations of increased turbidity, the maximum turbidity value recorded and hysteresis trends were found to be highly variable and highly dependent upon rainfall patterns. Turbidity peaks lasted significantly longer when rainfall occurred across multiple days, and especially when multiple high flow events often took place in short succession (Supplementary Fig. 6). Antecedent conditions appeared more important than total daily rainfall. For example, the rainfall event at the start of May 2017 (Fig. 4) resulted in a low intensity and short duration turbidity peak, despite being the largest individual daily rainfall event. In contrast, most increases in turbidity continued over a number of days (a mean of approximately 6 days in each peak) and were characterised by rainfall over an extended number of days. There are multiple occurrences of many high flow events in short succession, resulting in extremely long periods of elevated turbidity, such as between the 30th January 2017 and 6th February 2017.

The maximum recorded turbidity in each runoff event was positively correlated with the highest flow, yielding an  $r^2$  of 0.62 (Supplementary

### Classification 1



### Classification 2

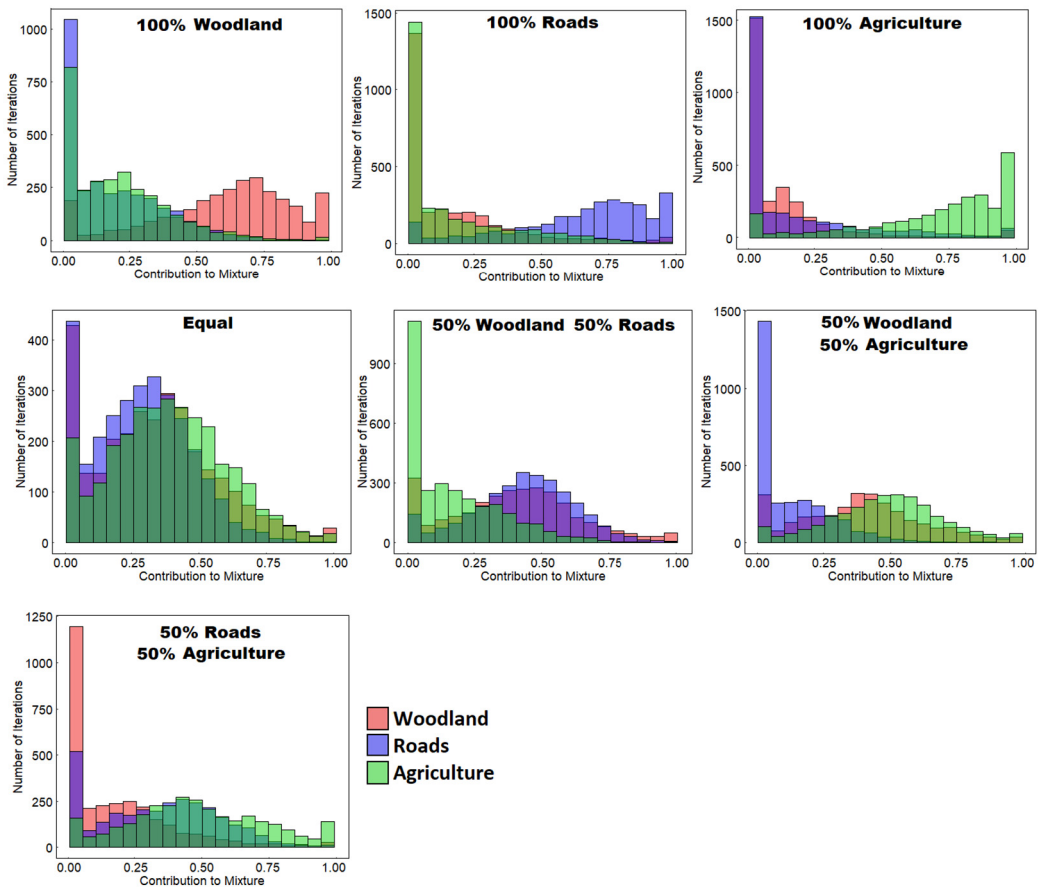


Fig. 9. Probability density functions of the virtual mixture source apportionment.

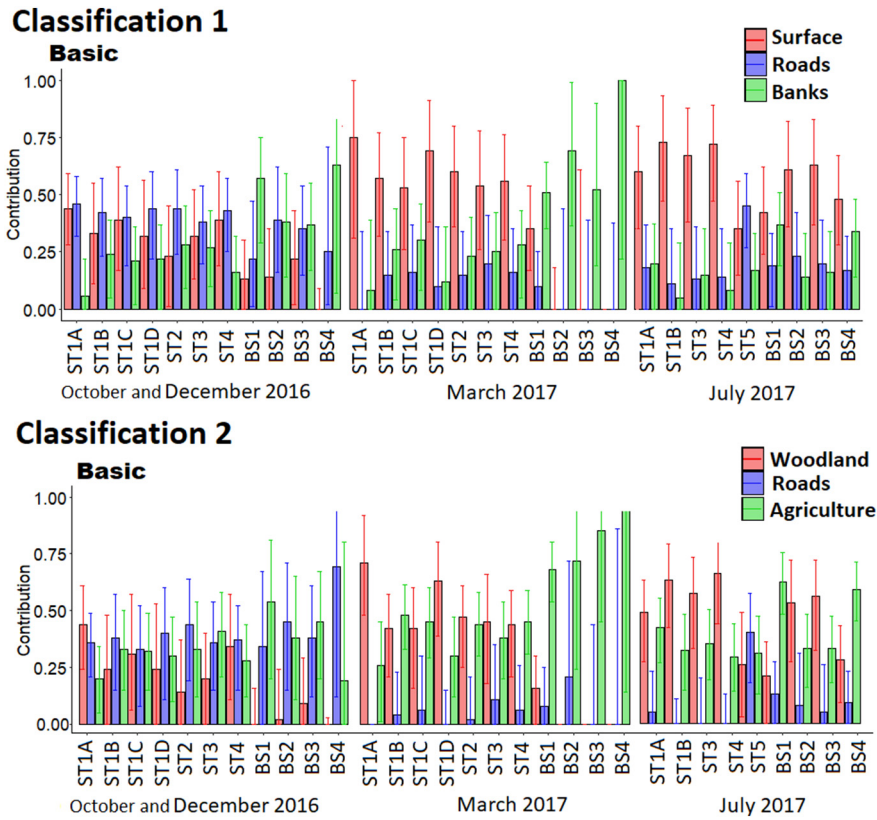


Fig. 10. The median contribution from each sediment source group and the basic composite fingerprint in classification 1 (a) and classification 2 (b) with the 25th and 75th percentile range of uncertainty.

Fig. 7). The highest magnitude events generally resulted in flow and turbidity peaks with either no hysteresis or a clockwise hysteresis, whereas smaller flows were characterised by anticlockwise hysteresis. Of the 28 high flow events recorded, five followed a strongly anti-clockwise

hysteresis pattern, nine showed an anticlockwise pattern, 12 were flat or a slight figure of eight and two were clockwise. Both observed clockwise peaks were characterised by a sharp peak and subsequent fall in turbidity early in the flood, preceding the rise in flow marking the

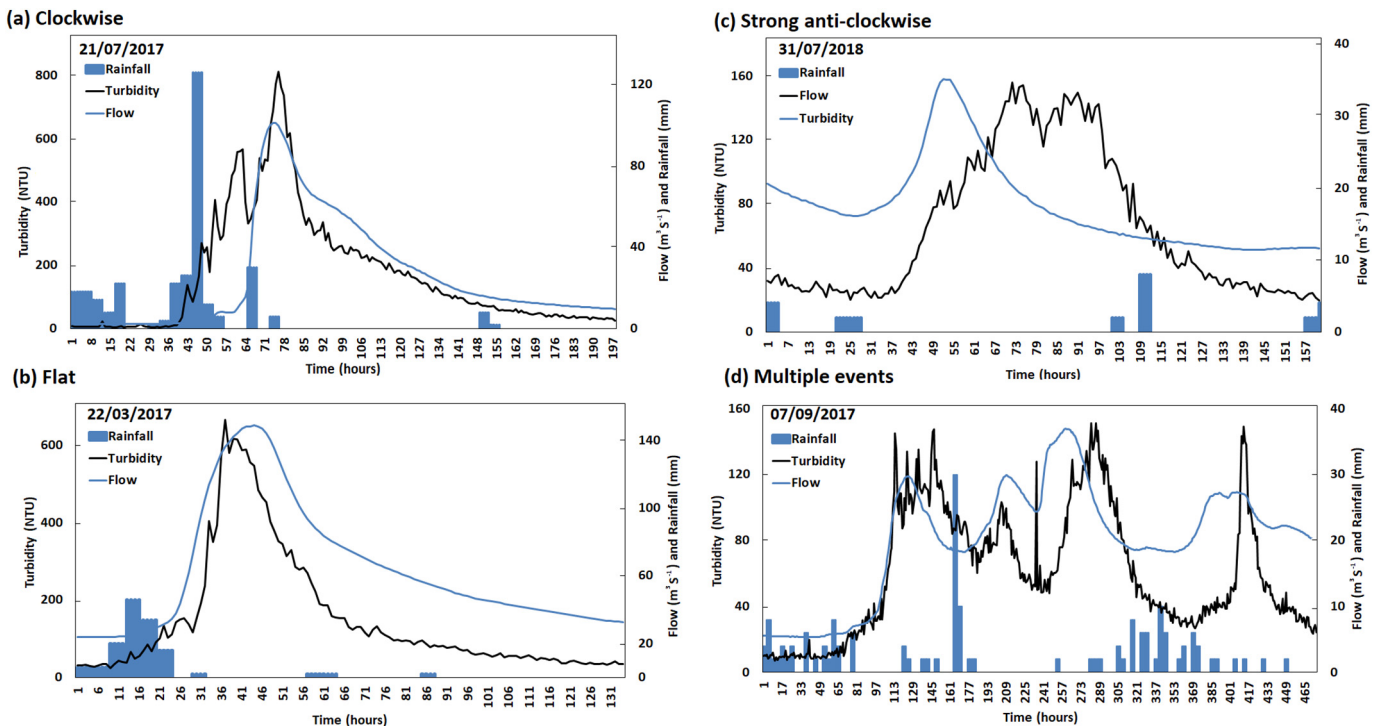


Fig. 11. Rainfall, turbidity and flow over example high flow events during the study period following different hysteresis patterns.

start of the event. Five of the 12 events with a flat hysteresis also had this initial peak prior to the main rise in turbidity (Fig. 11a). This initial peak likely represents sediment inputs from a local easily mobilised source, such as road deposited sediments. It was not possible to predict the type of hysteresis which would occur using the time since the last major flood event. However, from the 7th July 2017, four high flows occurred over 19 days with multiple rainfall events taking place during that period (Fig. 6d). Hysteresis patterns changed over the course of these events. For the first two events, turbidity rose with flow, and for the second two events turbidity rose only in the falling limb of runoff. The duration of the rise in turbidity also decreased over the sequence of storm events. There is, therefore, a trend of more easily mobilised sediment sources being depleted and sources such as woodland which may have a high rainfall buffering capacity or distal agricultural sources gaining increased importance over time.

#### 5.4. Channel bed fine-grained sediment storage

Quantities of fine-grained sediment (classified as all re-suspended sediment and not only the <25  $\mu\text{m}$  fraction) stored on the channel bed of the River Torridge were generally low, falling well below the median values for other UK rivers sampled in a recent strategic survey (Table 3; Naden et al., 2016). There was a reduction in sediment storage between October 2016 and March 2017, when a number of high flow events took place; there was, however, little change between March and July 2017 despite low rainfall. Due to the high spatial variability in bed sediment storage, differences between sampling periods were not statistically significant.

There was considerable spatial variability in fine-grained sediment storage on the channel bed. In much of site 2 and all of site 3 where there were shallow fast flowing riffles over exposed bedrock, there was almost no fine-grained sediment present (Supplementary Fig. 8). In the deeper and slower waters of sites 1 and 4, there was a greater amount of sediment stored; the largest sediment deposits were found on the inside of meanders where the bed was composed of a sandy matrix.

## 6. Discussion and conclusions

The sediment source fingerprinting exercise identified that during the first large storm event after a period of low flow, roads were a major sediment source. During periods of low flow, roads were revealed to be a much more minor source; however, it is likely that roads remain an important transport pathway for agricultural sediments to the river channel. During large storm events, visible runoff from fields to the river via roads was observed. It is unclear how long sediment must be deposited on the road surface until its properties change significantly from its original source to be recognised as the road source group. It is unlikely, however, that the short duration between the floods throughout most of the study period would have been sufficient for this property transformation.

It was not possible to differentiate between sediment originating from cultivated land and grassland, most likely reflecting the impact of land use rotation. However, due to the limited area of the catchment utilised for cultivation and its typically large distance from the river channel, it is likely that grassland sources are the most important. Intensive modern agriculture on grassland has previously been reported to contribute to soil erosion, sediment problems and wider water quality issues (Evans, 1998; Singleton et al., 2000; Drewry et al., 2008; Deeks et al., 2008; Collins et al., 2010a). It was also concluded by Nicholls (2000) that cultivated sources were a very minor source in the upper River Torridge catchment.

Woodland was suggested to be a major sediment source in the River Torridge catchment. In other UK catchments, woodland is typically a very minor sediment source when compared to agriculture (e.g. Collins et al., 1997; Walling et al., 1999) apart from during tree felling

or fire, or where farm vehicle tracks are present (Swanston and Swanson, 1976; Morris and Moses, 1987). The high contribution of sediment from woodland here is likely due to a partial wooded corridor along much of the length of the River Torridge. This corridor is on highly sloped land and small channels frequently flow through it to the river channel. Footpaths and tracks also increase connectivity between the woodland and the river channel. Prior to agricultural intensification, woodland would have covered a far larger area of the catchment than in the present day. It is therefore likely that the river has been characterised by a high sediment yield for decades, given that the deciduous woodland would have been managed to some degree. The 25% to 50% contribution from woodland to the 89  $\text{t km}^2 \text{yr}^{-1}$  sediment yield for the upper River Torridge reported by Nicholls (2000) would be a yield of 22–44  $\text{t km}^2 \text{yr}^{-1}$  from woodland which is close to the 44  $\text{t km}^2 \text{yr}^{-1}$  estimated as typical for UK catchments in the present day where intensive agriculture is the dominant land use (Walling et al., 2007). However, it should be borne in mind that the temporal characteristics of sediment delivery to the river channel from today's agricultural landscape is likely to be highly different to those for the wooded and rough grazing covered catchment present when the FPM successfully reproduced. Woodland typically has a high precipitation buffering capacity due to enhanced incorporation of organic matter into the soil resulting from litter fall, higher root densities and diameters, greater diversity of soil fauna and ultimately improved macro-porosity and structure (Beven and Germann, 1982; Chandler and Chappell, 2008) and therefore sediment mobilisation is unlikely to take place without high antecedent rainfall. The trend of sequential days of high rainfall within the River Torridge catchment provides such conditions. If only woodland was present within the catchment, a rise in river turbidity would only be expected predominantly in the later stages of high flow events after considerable precipitation had fallen. In the present-day, it is hypothesized that roads likely provide easily mobilised sediment during the initial stages of storm events, but that agricultural land is also a major sediment source and will contribute sediment after roads and before woodlands, whilst woodlands deliver sediment in the latter stages of storm events. Therefore, a change in sediment source likely takes place as precipitation falls over consecutive days causing the observed elongated peaks in turbidity. This aspect of hydro-sedimentological response is likely to be the most important factor by which agricultural fine-grained sediment reduces the chances of successful FPM reproduction.

The findings of this study have shown that fine-grained sediment movement in the lower River Torridge where the majority of FPM are present, is highly-dependent upon rainfall patterns. A high percentage of days having high rainfall totals in sequence results in elongated peaks in turbidity. This is important since it has been shown that FPM are able to tolerate suspended sediment concentrations of 30  $\text{mg l}^{-1}$  only over short time scales such as a single flood event, but that longer duration concentrations of only 10  $\text{mg l}^{-1}$  may be harmful (Valovirta, 1998). This implies that longer duration peaks in turbidity will be more harmful to the sensitive juvenile mussels than shorter duration peaks. In the upper River Torridge, Nicholls (2000) found a high sediment yield of 89  $\text{t km}^2 \text{yr}^{-1}$  with suspended sediment concentrations of below 2  $\text{mg l}^{-1}$  during low flows and up to 1200  $\text{mg l}^{-1}$  and a corresponding mean of 433  $\text{mg l}^{-1}$  during high flows. SSCs therefore substantially exceed concentrations which are likely to be tolerable to

**Table 3**

Mean and median quantities of fine-grained sediment storage ( $\text{g m}^{-2}$ ) on the bed of the River Torridge; percentiles from Naden et al. (2016).

	Oct-16	Mar-17	Jul-17
Mean	559	352	334
Standard deviation	683	499	262
Median	393	144	344
Percentile for UK rivers (of mean)	41st	32nd	31st

juvenile FPM, albeit that we must bear in mind likely dilution with flow accumulation as you move downstream in the River Torridge system.

In contrast to the long duration elevated levels of turbidity recorded, low quantities of fine-grained sediment were stored on the channel bed in the study area, with a mean value of 0.42 kg m<sup>2</sup> which was close to the lower 1/3 of values reported for UK rivers on the basis of a strategic sampling survey (Naden et al., 2016). The estimate of bed sediment storage generated by the study reported in this paper is slightly higher than the 0.14 kg m<sup>2</sup> stored in riffles in the upper River Torridge reported by Nicholls (2000). However, riffles over exposed bedrock within the study area used in this new paper had comparable or lower quantities of sediment than the upper River Torridge, indicating considerable spatial variability in this component of the sediment system (Nicholls, 2000).

Within the Torridge, the remaining FPM will typically live in the most stable habitats amongst bedrock fissures or cobbles, as opposed to riffle sections which are currently experiencing increased fine sediment accumulation. Hastie et al. (2000) found a similar trend that the inside bends of meanders are rarely inhabited by mussels due to their instability. Juvenile FPM are pedal feeders for their first few years and live burrowed amongst the gravel in the riverbed, before emerging as filter feeders. They therefore require higher flow rates, amongst stable pockets of gravel, in order to keep the gravels well oxygenated and free of sediment. Higher quantities of sediment were found deposited on the channel bed in these locations.

A high amount of temporal variability in bed storage was also found with a reduction in sediment storage after the first and largest high flow event of the study period, which was preceded by a period of low flow. It was reported by Nicholls (2000) that infiltration of sediment deep into gravels is event driven and depends on supply of material. In both the new study reported here, and Nicholls (2000), eroding channel banks were identified as a major source of sediment deposited on the channel bed; therefore, channel bank erosion during the falling limb of storms or periods of low flow is likely to be an important sediment supply process. However, due to the low quantities of fine-grained sediment stored on the channel bed and its regular flushing from the substrate during high flows, it likely that suspended sediment reflected in the turbidity recordings, represents a greater obstacle to FPM reproduction in the study river than channel bed sediment storage.

It is most likely that the long periods of elevated turbidity due to the high connectivity of different sediment sources to the river channel is a key driver resulting in the harmful effects of sediment mobilisation and delivery on FPM reproduction in the study river. Further research is needed into changes in sediment sources over the duration of individual storm events to confirm the hypothesis generated by this investigation.

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