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Are source groups always appropriate when fingerprinting sediment? The direct comparison of source and sediment samples as a methodological step

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1. Abstract

The classification of sediment source groups is often the least thoroughly considered part of a sediment fingerprinting methodology; however, the use of inappropriate source groups can be the cause of significant uncertainty. In many catchments, source groups based on land use or geology are a poor fit for their geomorphological processes and the nature of the tracers used. Against this context, this study directly compared the average percentage difference in the standardised concentrations of all tracers between a sediment sample and each individual source sample, to map the similarity between the properties of sources and sediment in three study catchments. In the River Nene, UK, the mean percentage differences between source and sediment tracer concentrations were primarily controlled by the presence of distinctive ironstone and urban sources, which had very dissimilar properties to the target sediment. However, a generally consistent trend of certain source samples having more similar properties to multiple target sediment samples than others was also found; a finding which could not be identified when using conventional source groups. In the Sywell reservoir catchment, UK, sediment originated from throughout its catchment, apart from in the case of damaged road verges, and there was little indication of any major change in sediment sources through recent time. In the Vuvu catchment, South Africa, there was a larger contribution from distal igneous sources during high flow events. The trialled method, however, provided little advantage over the standard fingerprinting approach in this case, due to the existing good fit between catchment geomorphology, the tracers used and the geological source groups. The method trialled herein can provide distinct advantages over the conventional fingerprinting approach and, whilst it should not replace it, provides a useful supplement by permitting an assessment of whether potential source groupings make best environmental sense, and providing increased resolution of sediment provenance.

Keywords: sediment fingerprinting, source classification, geomorphological processes, uncertainty

2. **Introduction**

The classification of source groups is perhaps the least thoroughly explored stage of the sediment fingerprinting approach, but in many ways is the most important. Source groups are the context in which tracer concentrations of sources and sediments are compared, forming the foundation of the sediment fingerprinting approach (Walling *et al.*, 1993; Collins *et al.*, 1997). They are also the context in which results are usually expressed to end users, e.g. as a percentage contribution from each source. When combined with sediment yield data, this allows for the evaluation of the magnitude of sediment loss from individual sources such as cultivated land, eroding farm tracks or damaged road verges (Collins and Anthony 2008; Collins *et al.*, 2010). In the context of conceptualising the catchment sediment system and delivering useful results to managers, *a priori* sediment source group classification based on land use (e.g. arable or grassland) and specific sources (e.g. channel banks or damaged road verges) is the most common source classification used, with catchment geology and / or soil types used less frequently (Collins and Walling, 2004; Haddadchi *et al.*, 2013).

The use of source groups ideally requires the selection of tracers which can robustly discriminate between them (Collins and Walling, 2002). For example, ^{137}Cs, ^{210}Pb and ^{7}Be have been shown to be strong discriminators between surface and subsurface sources

(Walling and Woodward 1992; Evrard *et al*., 2016). However, only a few of these robust tracers exist and they are rarely able to classify 100% of source samples into their respective groups when a linear discriminant analysis is used, requiring the inclusion of additional tracers in a so-called composite fingerprint (Walling *et al.*, 1993; Collins *et al.*, 1997). As a result, most tracers used in a composite fingerprint will discriminate on the basis of a 'black box' type methodology (Owens and Xu, 2011) and, individually, are often poor discriminators.

The size of the uncertainty generated within Monte Carlo based un-mixing model results is associated with a poor signal-to-noise ratio. Uncertainty has been shown to be increased by poor discrimination associated with a small contrast in tracer signatures between-source groups (signal) and large within-source group variability (noise) (Small *et al.*, 2002; Collins and Walling, 2002; Pulley *et al.*, 2015a). A variety of factors have been shown to affect tracer concentrations including: land use (Fox, 2005), geology (Wilson, 1989), soil type and drainage (Blundell *et al.*, 2009), anthropogenic pollutants (de Miguel *et al.*, 2005), management practices (McDowell *et al.*, 2016), erosion intensity (Wilkinson et al. 2015), particle size (Bihari and Dezső, 2008, Horowitz and Elrick 1987) and organic matter (Hirner *et al.*, 1990). As soils under a given land use or overlying a specific geology will vary spatially in terms of these factors, significant within-source variability will often be identified.

An increased within-source group variability is also associated with the delineation of a catchment into poorly differentiated source groups. Rotation between cropping and grassland can mean that there is sometimes poor discrimination between these two land uses due to mixing of fallout tracers through the ploughed layer. Soil type and underlying geology may not be accurately represented on maps of the catchment where deposits are small; or the underlying geology covers too small an area to significantly affect overlying soils. Sediment transport may cause eroded material from upslope to form a blanket over native soils, meaning that the underlying geology is not necessarily reflected in the source samples collected. Channel banks may be ill defined and low and share their properties more closely with surface material rather than subsurface material. Some areas of channel banks may be composed of recently deposited alluvium and others by older valley fill with very different properties, and channel bank collapse may also cause the banks to be composed of displaced surface material. Therefore, the misclassification or misfit of source samples is likely, and will act to reduce effective source discrimination.

The impact of tracer variability and associated poor source discrimination is important in three ways. First, tracer non-conservatism will have larger effects on sediment provenance results when there are small contrasts in tracer concentrations between source groups (Pulley *et al.*, 2016a). Secondly, many un-mixing models assume a normal distribution of tracer concentrations in the groups (Barthod *et al.*, 2015). Other models assume a tracer distribution using a non-parametric scaler, such as median absolute deviation or Qn (Collins *et al.*, 2010). In reality, tracer concentrations often will not follow a smooth regular distribution due to the numerous factors shown to control them within a source group. Therefore, the data input in the un-mixing model may be a poor fit to the actual distribution of tracer concentrations within a source group, increasing uncertainty associated with model results. Thirdly, uncertainties surrounding within-source group variability become even more problematic when the nature of erosion and sediment delivery is considered. Sediment inputs to a river can be highly localised. For example, different fields in the UK have been shown to erode in different years making sediment source areas highly spatially and temporally variable (Evans and Boardman, 2015; Evans *et al.*, 2016). Similarly, areas of channel bank experiencing erosion have been shown to be variable between flood events (Bull, 1996) and are likely to be concentrated in distinct 'process domains' (Abernethy and Rutherfurd, 1998; Couper and Maddock, 2001). The delivery of eroded material to a channel is also highly spatially and temporally variable, with the effective catchment area contributing sediment to a channel changing over time (Fryirs, 2013). The transfer of sediment between areas of a catchment is dependent on their morphology and the energy and materials flowing through them (Chorley, 1971; Schumm, 1981). For example, gully or rill formation can rapidly change the area of a catchment contributing sediment to the river channel (Foster *et al.*, 2012).

Sediment source groups may, in some ways, represent geomorphic processes, with poaching dominating sediment generation in grassland and sheet or rill erosion affecting cultivated land (Evans *et al.*, 2016), or with a different geology being present on valley floors affected by gullying compared to hillslopes which are affected by sheet erosion (e.g. van der Waal *et al.*, 2015). The major problem with variability in sediment source properties is that it is likely that only a small number of the source samples retrieved will be from an area that is directly contributing sediment to a river at any sample collection time. Therefore, the tracer distributions of the source groups input into an un-mixing model will rarely match the distributions of the actual sources of sediment, which may explain why results derived using different tracer groups can often be very different (Pulley *et al.*, 2015a). This uncertainty can be incorporated into modelling outputs by thorough source sampling and the representation in an un-mixing model of distributions of tracer properties on the basis of measured data and the associated tracer property locations (e.g. mean or median) and scale (e.g. standard deviation or median absolute deviation). However, this often results in a large range of uncertainty associated with apportionment results. It may be questioned how useful contribution estimates with uncertainties typically in excess of 30% (on a 0 -100% scale) are for catchment management purposes (Collins *et al.*, 2014).

To improve source discrimination and reduce the effects of tracer variability, two options are available. Firstly, to only use tracers such as ^{137}Cs which are known to discriminate robustly between certain sediment source groups or which can be clearly demonstrated to robustly discriminate using existing empirical data. However, as previously mentioned, such tracers are rare and can be unreliable in agricultural landscapes with land use rotation. Alternatively, sediment source groups can be created to best fit the measured tracers, by using methods like cluster analysis (Walling *et al.*, 1993; Walling and Woodward 1995; Pulley *et al.*, 2016a). These groups must, however, also be useful for management purposes. Source groups heavily fragmented into small areas scattered around the catchment will likely be of little use for targeting management and difficult to interpret by end users. It can therefore be argued that whilst classification into sediment source groups makes sense from a catchment management standpoint, it often makes little sense in terms of catchment geomorphology or tracer suitability. Therefore, questions arise as to if sediment source tracing be conducted without dividing a catchment into pre-defined specific source groups, and if the suitability of potential source groups can be assessed as an additional methodological step to determine if groups fit the tracers used. To address these questions this paper revisits three catchments where sediment source fingerprinting was previously conducted. In doing so, an attempt is made to establish if a direct comparison between the tracer concentrations of each individual source sample and a target sediment sample can yield useful sediment provenance information to supplement existing tracing methodologies and reduce uncertainties.

3. Study sites

Three study catchments were examined; two in the East Midlands of the UK and one in the Eastern Cape of South Africa. Sediment in the River Nene (1634 km^2) basin, UK, was originally traced by Pulley *et al.* (2015a) who found that different tracer types (magnetic, geochemical, fallout radionuclude and lithogenic radionuclide) produced very different sediment provenance results. The catchment has a low sediment yield (13 – 18 t.km^2 yr^{-1}) and a fairly low (by UK standards) average annual rainfall of 638 mm (Pulley and Foster, 2016). Land use is 56% cultivated land, 22% improved grassland and 9% urban, with the remaining 13% composed of woodland, rough grassland and surface water (Morton *et al.*, 2011). This has changed from the catchment being dominated by pasture in the 1930s (Stamp, 1931). The geology comprises Jurassic mudstones, sandstones and ironstones dominating in the west and in valley bottoms and Quaternary diamicton and Jurassic limestones dominating on hilltops and in the east. Both the ironstone and limestone have highly distinctive tracer signatures. An examination of the ^{137}Cs activities in sediment showed that channel banks are its dominant source (Pulley and Foster 2016). It has, however, not been possible, thus far, to identify the spatial areas of the catchment contributing most sediment directly to the river.

Sywell reservoir (7.84 km^2) is in the centre of the River Nene basin. It was constructed in 1906 and was cored in 2011 as described by Pulley *et al.* (2015b). Its land use and geology are comparable to that of the River Nene basin as a whole with 54% of land cultivated, 23% used for sheep grazing, and 23% covered by woodland. Since the 1930s, land use has changed from being dominated by grassland (Stamp, 1931). Despite this change, there is no evidence of an increase in catchment sediment yield over time (Pulley and Foster, 2016). The geology is diamicton in the upper catchment, ironstone in the central part of the catchment and mudstones in the lower catchment. Soils are freely draining brown earths over ironstone, poorly draining clays over diamicton and mudstones. Results reported by Pulley *et al.* (2015b) suggest that channel banks are the dominant source of sediment to the reservoir, albeit with a high corresponding uncertainty.

The Vuvu (65 km^2) catchment is in the north-east of the Eastern Cape of South Africa and forms a tributary of the Thina River and the larger Umzimvubu River. Average annual rainfall is 707 to 928 mm, with lower rainfall in valley bottoms (~920 masl) and increased rainfall at higher altitudes (up to 2100 masl) (Nel *et al.*, 2010). High intensity storms occur in the summer months (Nel, 2008). The catchment is located on the escarpment of the Southern Drakensburg and is underlain by Drakensberg Group basalts, dolerites, and Clarens Formation sandstones in its upper half, overlying Elliot Formation mudstones. Topsoils on basalt, dolerite and sandstone hillslopes are typically shallow (~20 cm) and poorly developed. Valley floors are partially covered by Quaternary colluvium and alluvium up to 6 m thick

(Fey *et al.*, 2010). Valley bottom soils are highly dispersive and degraded with soil pipes, rills and gullies found extensively (van der Waal *et al.*, 2015), as has been described for other areas in the Mzimvubu catchment by Beckedahl and Dardis (1988). The upper catchment over the Drakensberg formation igneous geology is mostly utilised for grazing; in contrast, the valley bottoms over the Elliot formation mudstones and Quaternary colluvium are much more intensively utilised for cultivation, human habitation and livestock grazing. van der Waal *et al.*, (2015) determined that sedimentary sources from the lower catchment dominate the provenance of most deposited flood bench sediments, but in high flow events which inundate flood benches elevated 2-4m above the channel, distal igneous sources are important.

4. Methods

4.1. Sediment and source data

The data used for the River Nene basin were collected and described by Pulley *et al.* (2015a). Samples of overbank sediment were washed from riparian vegetation along the length of the main channel and its tributaries immediately after flood waters receded after four flood events in April, July, October and November 2011. Source samples were retrieved from the cultivated land, improved grassland, urban roads and exposed channel banks in the catchment and sieved to <63µm to match the particle size distribution approximately of the retrieved sediments. Mineral magnetic, radionuclide and geochemical tracers were measured for all samples. Where particle size is referred to it was measured using a Malvern Mastersizer 2000 laser granulometer after pre-treatment using hydrogen peroxide.

The samples for Sywell reservoir were those described by Pulley *et al.,* (2015b). A core was retrieved from the centre of the reservoir using a mini Mackereth pneumatic corer. Samples of this core were dated using ^{210}Pb and ^{137}Cs and source apportioned after calculating mean tracer concentrations for four sections between 0-15(2011-1989), 15-25(1989-1967), 25-35(1989-1947) and 35-45cm (1906-1947) depth. A range of source samples were retrieved from topsoils, channel banks and damaged road verges and each source sample was sieved to <63µm for analysis. Mineral magnetic, lithogenic radionuclide and geochemical tracers were measured.

The tracing of the Vuvu catchment sediments used four cores retrieved from upper flood benches (2-4m above the channel) and lower benches (1-2m above the channel) in the lower catchment, and surface and subsurface source samples collected by van der Waal *et al.,* (2015). Unlike in the original publication, all source and sediment core samples were sieved to <32μm to minimise the potential impacts of particle size on tracer properties. Six mineral magnetic signatures were measured for each sample. As there was little down-core variability in tracer concentrations in any core, the mean value for the cores from the upper and lower benches was used to represent the target sediment. An organic matter dilution correction was used with the mineral magnetic signatures in this study (Lees, 1999). No other organic matter and particle size data corrections were used in any investigated site in this study, or in the originally published studies.

4.2. Tracing methodology

The tracing methodology used was a simple comparison between the concentrations of all tracers measured for each source sample and a single target sediment sample. Each source sample is examined independently without prior regard to its land use, geology, soil type or other potential groupings. Prior to beginning the analysis, the source and sediment tracer values were normalised to between 0 and 1 by dividing each value by the maximum found for that tracer in the source dataset. The absolute difference between the concentration of each tracer measured for the target sediment sample and each individual source sample was calculated and expressed as a percentage of the concentration in the sediment sample. The result for each source sample was summarised as the mean percentage difference for all measured tracers. This procedure was repeated for every target sediment sample collected. The likelihood of a given source sampling point having contributed to the target sediment is based on a small mean percentage difference between it and the target sediment when compared to the differences calculated for the other source samples. If sediment was evenly contributed from all areas of the catchment, a uniform percentage difference would be expected for all source samples. Deviation from a uniform difference identifies areas less or more likely to have contributed sediment, as well as potential source groupings of samples with similarly large or small mean differences.

The mean difference for each tracer between all source samples and all sediment samples was calculated. In this way, any tracer with a disproportionally large difference between the source and sediment samples could be examined in the context of representing a specific sediment source or being affected by non-conservatism processes. A disproportionally large difference was defined as being greater than 1 standard deviation above the mean for all tracers. It was also examined if each sediment tracer concentration fell within the full range of the source samples as a basic range test for conservatism; this test was satisfied for all tracers in all samples. The final results of the tracing were presented as a map of the catchments showing the source samples and their mean percentage difference from a target sediment sample. Each map represents a single sediment sample or section of a sediment core.

5. Results

5.1. Overbank sediment in the River Nene basin

The overall mean difference for ^{137}Cs appears high due to the low concentrations found in the sediment samples compared to many of the source samples (Table 1). Zinc has moderately high concentrations in the source samples yet very large differences between potential sources and the target sediments. ^{235}U also had high differences between sources and sediments.

Land use has little effect on the mean differences between the tracer concentrations of the source samples and a sediment sample retrieved from Ditchford in April 2011, except for urban road dust, which shows little similarity to the target sediment (Figure 1). The presence of the ironstone geology with its distinctive tracer signature (with high concentrations of almost all tracer types) also results in large differences between sources and sediments; however, limestone which also has a distinctive tracer signature with high Ca, Mg and Sr concentrations does not have particularly pronounced differences. There is no observable contrast between the differences derived for channel banks and surface topsoil sources.

In the Kislingbury arm of the Nene there is a good consistency between the spatial trends in mean percentage difference between sources and sediments found for the five sediment samples taken from its tributary sub-catchments and the sample taken from its outlet (Figure 2). Much of this consistency is caused by the presence of ironstone with large differences between its overlying soils and the target sediment. However, the observed trends are also similar with the other source samples, with specific samples being comparable to the sediment both at the tributary scale and at the arms outlet.

Overbank target sediment samples retrieved from three later floods in 2011 (Figure 3) showed the same general trend in which source samples have the most similar tracer concentrations to the sediment as the samples retrieved in April, with source samples in areas such as in the centre of the Weedon 1 and Heyford sites being consistently more similar to the sediment than most other source samples. The sample retrieved in November is notably less similar to all sources than samples retrieved in other floods.

5.2. Sywell Reservoir

For the Sywell catchment, the mean difference for IRM at -100mT is higher than most other tracers (Table 2). The high mean difference for cu is caused by its high concentrations in road verges and several outlying topsoil samples predominantly located over the ironstone geology. It is of note that the mean difference between sources and target sediment for all tracers and all samples is around half (65%) that of the whole River Nene basin (120%).

All sources apart from damaged road verges have comparable tracer concentrations to the sediment (Figure 4). Generally, sediment sources closest to the river channel or reservoir are most similar to the target sediment with channel banks often being the most comparable. There is no major difference between the results derived using sediments from the different layers of the core apart from a greater dominance of channel banks at 15 – 25 cm, suggesting no major change in sediment source over time (Figure 5). Unlike in the Nene basin as a whole, the source samples retrieved from the ironstone geology are not more different to the target sediment than sources over other geologies.

5.3. Vuvu floodplain benches

Overall, the differences found (Table 3) are comparable to those found for the magnetic tracers in the River Nene basin. χfd has pronounced differences between sources and the target sediments which are considerably higher than for other tracers. The average difference for all tracers apart from χARM is higher in the lower core than the upper core.

Differences between the upper and lower bench cores (Figure 6) appear to reflect the greater contribution of sediment from the upper igneous catchment during the high flow events capable of inundating the upper bench. There is generally little difference between the results derived using the surface and subsurface source samples, apart from at the boundary of the igneous and sedimentary geologies, where subsurface sources appear to reflect the igneous geology at a lower altitude.

6. Discussion

In all three catchments, the method used revealed key information regarding sediment provenance and the link between potential sediment source groups and the tracers used. In the overbank sediments of the Nene, urban areas and ironstone geology proved to be the most distinctive source groups with large differences between their properties and the target sediment. The failure of Pulley *et al.* (2015a) to trace surface and subsurface inputs to the Nene was likely in part due to the ironstone sources not being treated as a separate group and therefore increasing the within-source group variability of the source groups adopted and increasing uncertainty. There was also a consistent trend as to which of the other source samples were most similar to the target sediment when examining different sediment samples obtained from different locations and flood events. As these source areas could not be linked to specific source groups, this finding could not be determined by the conventional source fingerprinting approach. It was not possible, however, to firmly identify that channel banks were the dominant sediment source (Pulley and Foster, 2016) by examining the individual source samples; the only indication was that mean differences were often lowest for the bank samples in a given tributary sub-catchment. Pulley and Foster (2016) identified that channel banks were the dominant sediment source in the Nene using only ^{137}Cs. It may therefore be optimal to use the method presented in this paper in combination with the separate examination of robust tracers with a physical basis for source discrimination, such as ^{137}Cs, to identify both the spatial location of sediment inputs and to confirm if inputs are from surface or subsurface sources. Pulley *et al.* (2016b) also identified inputs of urban sediments from the town of Northampton and their subsequent down-stream dilution using pb, cu and zn, as well as a lack of sediment inputs from soils over the catchment ironstone geology using arsenic which is found in high concentrations in this geology. There is, therefore, considerable potential for the identification of contributions from easily discriminated sources using robust tracers with low associated uncertainties and a more general identification of source areas using the method presented here.

In Sywell reservoir, little indication of a change in sediment provenance was found in the four sections of the core. In all sections, damaged road verge source samples had far higher differences between their tracer concentrations and those of the target reservoir sediments than the other potential source samples. Unlike in the Nene basin as a whole, ironstone topsoils had similar tracer properties to the target sediments suggesting significant inputs from this source as well as other topsoil and subsurface sources. The mean difference between sources and target sediment for all tracers and all samples was around half (65%) that of the whole River Nene basin (120%) which is likely linked to the absence of ironstone as a non-contributing source. There was an indication from the results that samples close to the river channel and especially channel banks had tracer concentrations most similar to the target sediment. The fact that the sediments traced were deposited over the last ~100 years means that neither ^{137}Cs nor ^{210}Pb can be used to ascertain if contributions are from surface or subsurface sources. Instead, it may be optimal in this case to combine the method used with a conventional fingerprinting approach to ascertain the relative importance of surface and subsurface sources, given that there is some indication that channel banks are the most comparable to the target sediments and may therefore be effectively discriminated. The tracing of these reservoir sediments using a conventional fingerprinting approach by Pulley *et al.* (2015b) suggested that banks were the most important sediment source.

In the Vuvu catchment, the differences between sediment provenance on the upper and lower flood benches was primarily shown by the presence of a greater amount of highly magnetic sediment originating from the igneous upper catchment in the upper bench, which is only inundated during large flood events. This finding supports the conclusions determined using the conventional fingerprinting approach by van der Waal *et al.* (2015). Field observations in this catchment suggested that a large proportion (61 – 83 %) of gullies, which are primarily concentrated in the lower catchment, are still eroding, whereas the majority (79 – 86 %) of areas with sheet erosion have stabilised. Therefore, whilst the method used provides valid information on sediment provenance, in this case it provided little new information over the more conventional approach, other than identifying that at the boundary of the igneous and sedimentary geologies, subsurface sources appear to reflect the igneous geology at a lower altitude than surface sources. This might be expected if igneous surface material from upslope forms a blanket over the local sedimentary valley fill. In this case study, the source groups of igneous and sedimentary geologies reflect the differing erosion processes on hillslopes and valley bottoms well and the much stronger magnetic signatures of igneous sources provides a robust justification for the tracers used.

Mean differences between individual tracers for all sediment and source samples were able to reveal information about the ability of specific tracers to discriminate between sources as well as about their possible non-conservatism. In the Nene basin, ^{137}Cs had large overall differences in its activities between sources and sediments; this is likely a result of the dominance of subsurface sediment sources to sediment inputs (Pulley and Foster, 2016), which is not well reflected by most of the other tracers. Zinc had moderately high concentrations in the source samples yet very large differences between potential sources and the target sediments, likely due to its very high concentrations in urban road dusts. ^{235}U also had large differences between sources and sediments which may be linked to its high activities in the ironstone geology or possibly due to tracer non-conservatism. In the Sywell catchment, the mean difference for IRM at -100mT was high, likely due to its high concentrations in damaged road verges compared to other sources. A high mean difference for cu was also exhibited and likely caused by several outlying topsoil samples predominantly located over the ironstone geology which may make cu an unsuitable tracer when used with many potential source groupings. In the Vuvu catchment, χfd had pronounced differences between sources and the target sediments. This may represent the possible dissolution of ultra-fine superparamagnetic grains in the deposited flood bench sediments.

When compared to the conventional fingerprinting approach the method trialled here has the disadvantage of not quantifying inputs from the sediment sources, meaning that numerical and easily conveyed results cannot be provided to catchment managers. It also has the disadvantage that a source sample with comparable properties to the target sediment may not necessarily be contributing to the river. However, this limitation is also associated with the conventional *a priori* source group based approach as erosion and connectivity are likely to vary both spatially and temporally, meaning that it is unlikely that sediment will have originated from areas close to all source samples within a source group in any given high flow event. It is therefore suggested that more may be learned by combining the methodological step reported here with the latest decision-trees for conventional fingerprinting (e.g. Collins *et al.*, 2017) or the use of robust tracers to further refine sediment provenance. As with the conventional fingerprinting approach, the method trialled is also likely to be affected by changes to sediment particle size and organic matter concentration. The good consistency between the spatial trends in source – sediment differences found for many different overbank sediment samples in the Nene and for the different core layers in the Sywell and Vuvu catchments suggest, however, that neither factor was a consistent significant source of uncertainty in this study. It is possible, however, that the high differences between all source samples and the November 2011 Nene overbank sediment sample was caused by its coarse median particle size of 33.04 µm compared to 17.37 µm (standard deviation 10.95 µm) for all sediment samples.

The method reported herein could potentially be improved by source sampling according to heavily eroding areas in the catchment, which is likely to result in the source sampling being more representative of sediment reaching the river (van der Waal et al. 2015; Wilkinson et al. 2015). If source samples are also retrieved from actively eroding locations with clear connectivity to the river channel (Gellis and Noe, 2013) the resulting map of differences is likely to be more informative than the collection of source samples solely according to a set number of samples from each land use or geology. However, this method of sampling does require thorough sampling to account for spatial and temporal variability in the source areas. A randomised source sampling strategy would also be preferred over sampling on the basis of land use if the method trialled here is used independently of the conventional fingerprinting methodology. However, when the methodological step reported here is viewed in the context of potential source groups, it provides much needed insight into their suitability for a given study area, as well as preliminary insight into sediment provenance independently of source group and the application of an un-mixing model for generating quantitative source apportionment with uncertainties. The methodological step discussed herein may also be combined with the use of a cluster analysis based source group classification (e.g. Pulley et al., 2016a) to attempt to achieve the optimal spatial resolution of results and use of available tracers.

7. Conclusions

The simple tracing approach applied in this paper shows great potential for further exploring catchment sediment dynamics. Whilst this method provides only qualitative results, it could be used to supplement existing quantitative tracing methods and has the advantage of not pre-determining what source category discrimination might be possible. The method used acts as both a 'reality check' to determine if the sediment source groups are likely to be representative of the actual catchment sources and sediment tracer concentrations, and as a way to refine sediment source area identification. The extra methodological step may be particularly beneficial when combined with the targeted quantitative use of robust tracers such as ^{137}Cs which have clearly defined reasons for potentially effective discrimination between sources. In this way, maximum value can be extracted from a dataset without the potential for uncertainty associated with conventional (e.g. management orientated) source groups and a need for a 'black box' type utilisation of tracer data.

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Table 1: Mean and standard deviation percentage differences between tracer concentrations in all River Nene basin source and all sediment samples.

Tracer	Mean	Standard deviation	Tracer	Mean	Standard deviation	Tracer	Mean	Standard deviation
χlf	179	342	^{234}Th	168	183	Fe	104	117
χARM	148	227	^{235}U	226	254	Mg	53	89
IRM1T	169	310	^{212}Pb	59	50	Ni	40	39
IRM -100mT	195	352	Al	52	46	Pb	60	79
HIRM	155	424	As	96	128	V	56	75
^{210}Pb	37	54	Ba	47	51	Zn	421	840
^{226}Ra	185	159	Ca	98	153	Zr	33	29
^{137}Cs	399	641	Co	39	32			
^{228}Ac	68	63	Cr	45	40			
^{40}K	37	31	Cu	79	149			

Table 2: Mean and standard deviation percentage differences between tracer concentrations in Sywell Reservoir source and the sediment samples.

	0-15 cm		15-30 cm		30-45 cm		45-60 cm	
	Average	Standard deviation	Average	Standard deviation	Average	Standard deviation	Average	Standard deviation
χlf	113	*107*	95	*89*	97	*91*	126	*121*
χARM	63	*24*	64	*23*	66	*22*	59	*25*
IRM1T	71	*64*	63	*51*	64	*53*	90	*92*
IRM-100mT	199	*78*	185	*67*	186	*68*	231	*103*
HIRM	160	*253*	111	*152*	111	*150*	124	*184*
^{226}Ra	53	*25*	46	*24*	49	*24*	30	*20*
^{228}Ac	26	*17*	27	*17*	25	*17*	14	*11*
^{40}K	14	*12*	13	*10*	12	*9*	15	*10*
^{234}Th	42	*26*	50	*29*	33	*24*	19	*16*
^{235}U	36	*29*	33	*27*	27	*22*	26	*22*

^{212}Pb	22	*14*	26	*15*	19	*13*	11	*9*
Al	20	*13*	20	*13*	20	*18*	20	*19*
As	78	*67*	80	*68*	68	*61*	59	*54*
Ba	33	*36*	33	*38*	33	*38*	33	*36*
Ca	86	*17*	84	*16*	78	*22*	73	*60*
Co	45	*48*	36	*41*	32	*37*	31	*35*
Cr	57	*48*	52	*45*	59	*48*	41	*39*
Cu	204	*311*	199	*304*	207	*316*	260	*397*
Fe	35	*29*	34	*28*	33	*27*	40	*32*
Mg	70	*97*	71	*101*	80	*129*	96	*162*
Ni	75	*62*	63	*57*	54	*51*	47	*47*
Pb	64	*137*	59	*124*	62	*132*	78	*163*
V	55	*45*	47	*42*	44	*40*	30	*29*
Zn	70	*101*	63	*91*	72	*103*	97	*128*
Zr	37	*27*	24	*20*	24	*19*	21	*17*

Table 3: Mean and standard deviation percentage differences between tracer concentrations in Vuvu floodplain core and the sediment samples.

Upper Bench	**χlf**	**χfd**	**χARM**	**SIRM**	**IRM-100mT**	**HIRM**
Average	141	244	80	116	75	69
Standard deviation	177	353	22	136	45	31
Lower Bench	**χlf**	**χfd**	**χARM**	**SIRM**	**IRM-100mT**	**HIRM**
Average	206	382	71	167	231	117
Standard deviation	227	443	26	174	265	137

Figure 1: The mean percentage difference between tracer concentrations in source and target (overbank) sediment retrieved in April 2011 from Ditchford in the River Nene.

Figure 2: The mean percentage difference between tracer concentrations in source and target(overbank) sediment samples retrieved in April 2011 from the Kislingbury arm of the River Nene.

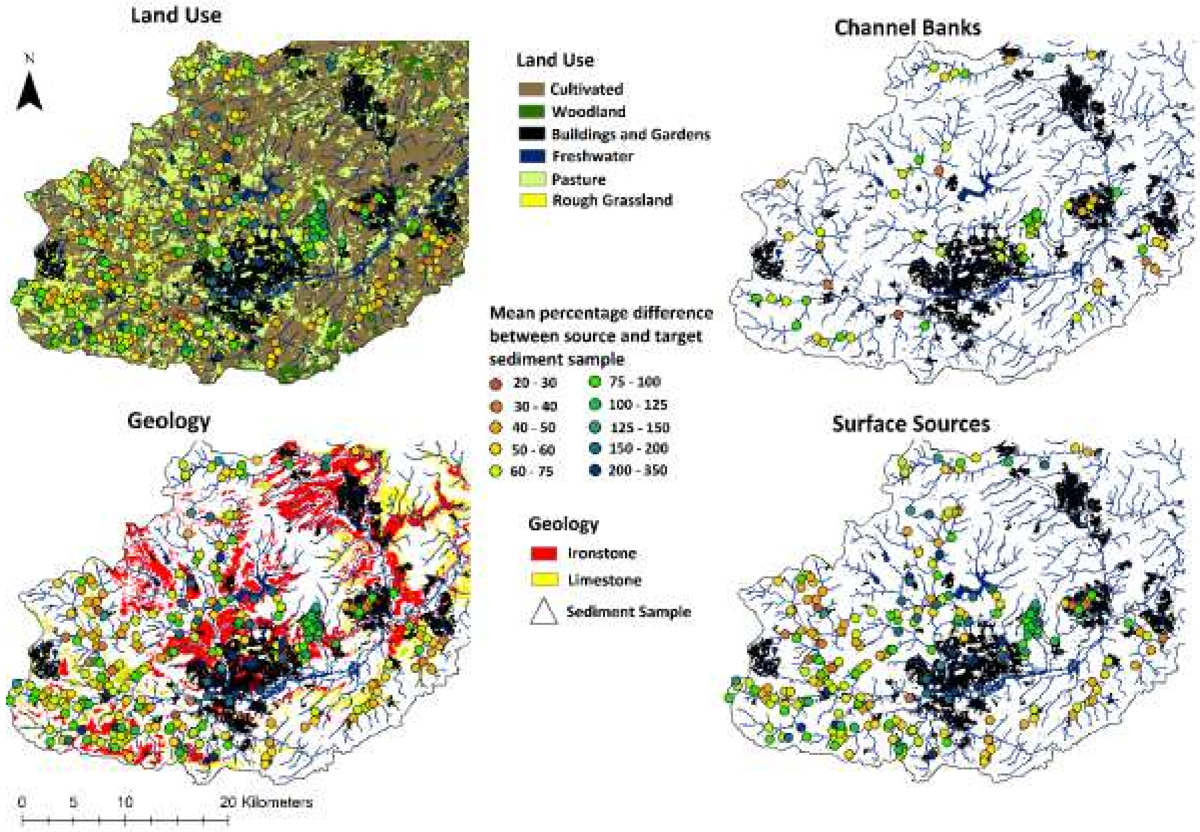
Figure 3: The mean percentage difference between tracer concentrations in source and target (overbank) sediment samples retrieved in July, October and November 2011 from the upper River Nene basin.

Figure 4: The mean percentage difference between tracer concentrations in source and target (Sywell reservoir) sediment samples between 0 and 15cm depth by land use.

Figure 5: The mean percentage difference between tracer concentrations in source and target (Sywell reservoir) sediment samples with increasing depth.

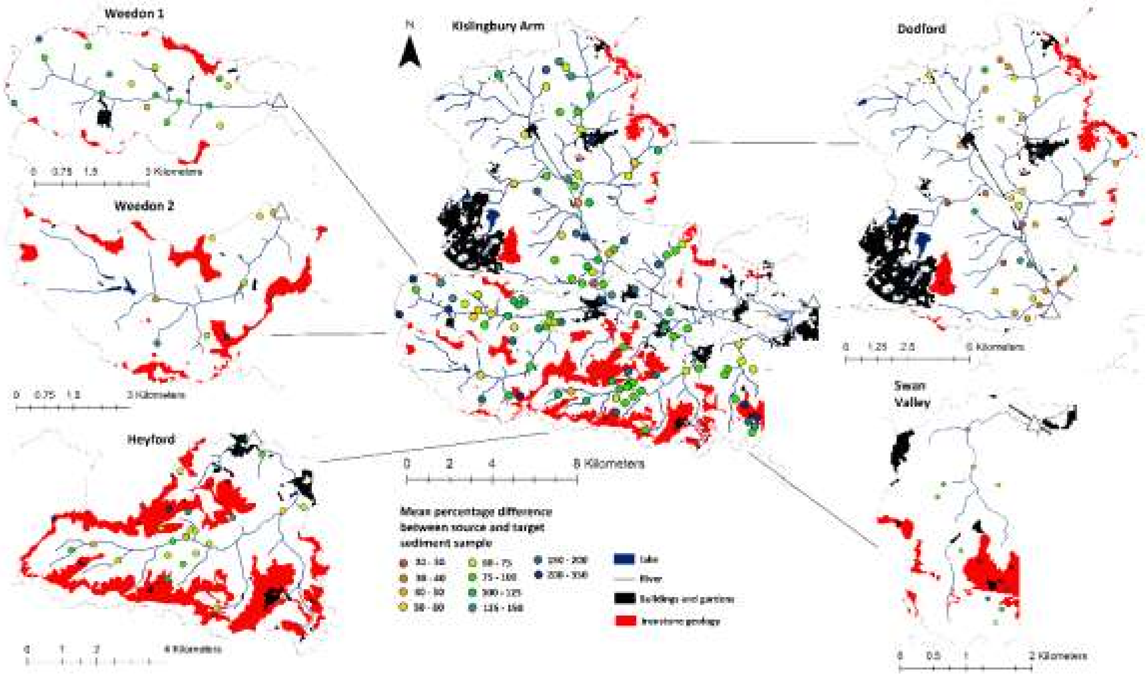
Figure 6: The mean percentage difference between tracer concentrations in surface and subsurface sources in upper and lower target (Vuvu flood bench) sediment samples.

Fig 1



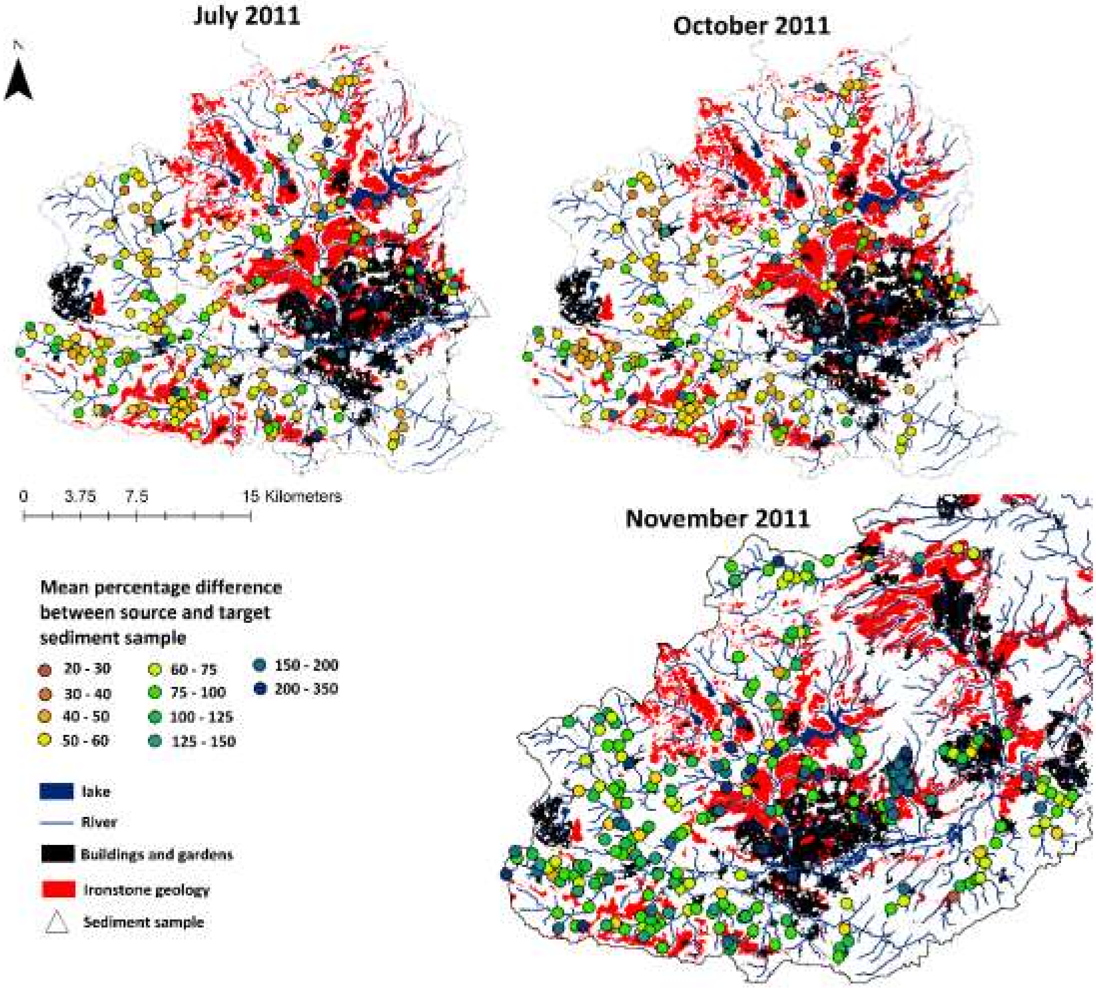
Land Use
Channel Banks
Geology
Surface Sources
N
Land Use
Cultivated
Woodland
Buildings and Gardens
Freshwater
Pasture
Rough Grassland
Mean percentage difference between source and target sediment sample
20 - 30
30 - 40
40 - 50
50 - 60
60 - 75
75 - 100
100 - 125
125 - 150
150 - 200
200 - 350
Geology
Ironstone
Limestone
Sediment Sample
0 5 10 20 Kilometers

Fig 2



Weedon 1
0 0.75 1.5 3 Kilometers
Weedon 2
0 0.75 1.5 3 Kilometers
Heyford
0 1 2 4 Kilometers
N
Kislingbury Arm
0 2 4 8 Kilometers
Mean percentage difference between source and target sediment sample
75 - 100
125 - 150
Lake
River
Buildings and gardens
Ironstone geology
Dodford
0 1.25 2.5 5 Kilometers
Swan Valley
0 0.5 1 2 Kilometers

Fig 3



July 2011
October 2011
November 2011
0
3.75
7.5
15 Kilometers
Mean percentage difference between source and target sediment sample
20 - 30
30 - 40
40 - 50
50 - 60
60 - 75
75 - 100
100 - 125
125 - 150
150 - 200
200 - 350
lake
River
Buildings and gardens
Ironstone geology
Sediment sample

Fig 4

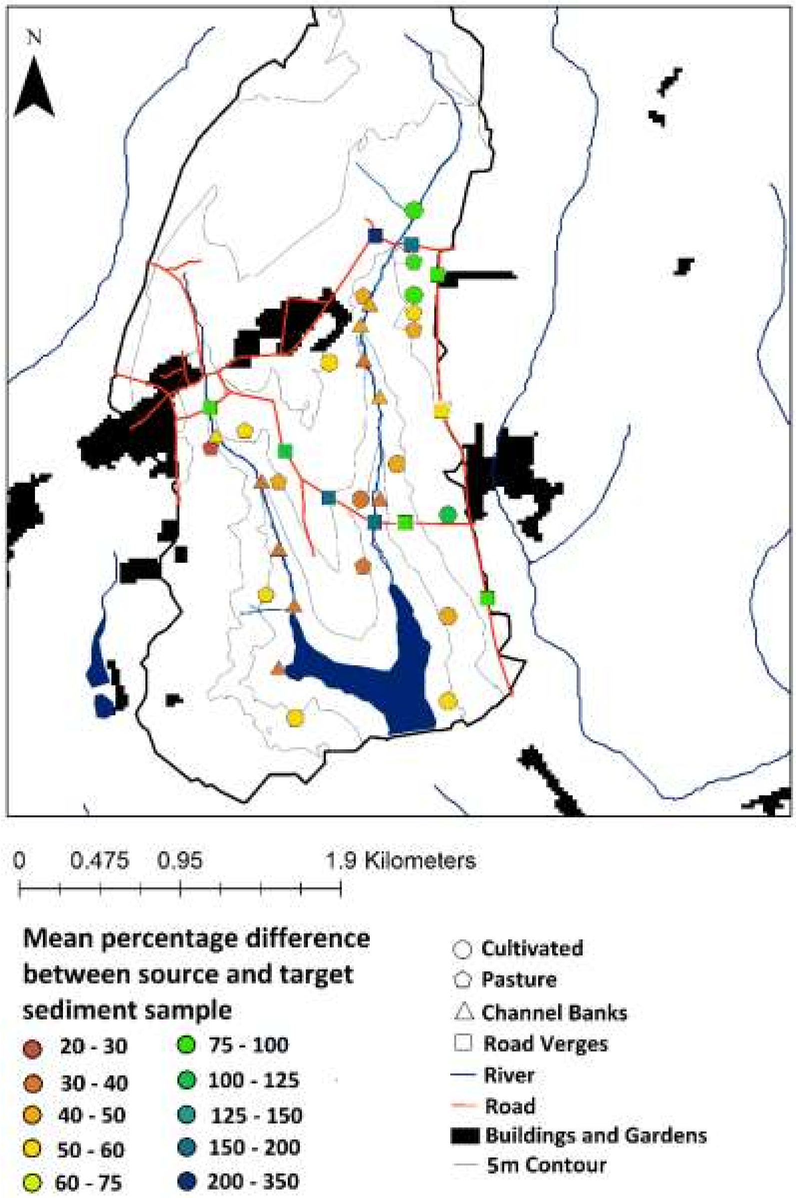

N
0 0.475 0.95 1.9 Kilometers
Mean percentage difference between source and target sediment sample
20 - 30
30 - 40
40 - 50
50 - 60
60 - 75
75 - 100
100 - 125
125 - 150
150 - 200
200 - 350
Cultivated
Pasture
Channel Banks
Road Verges
River
Road
Buildings and Gardens
5m Contour

Fig 5

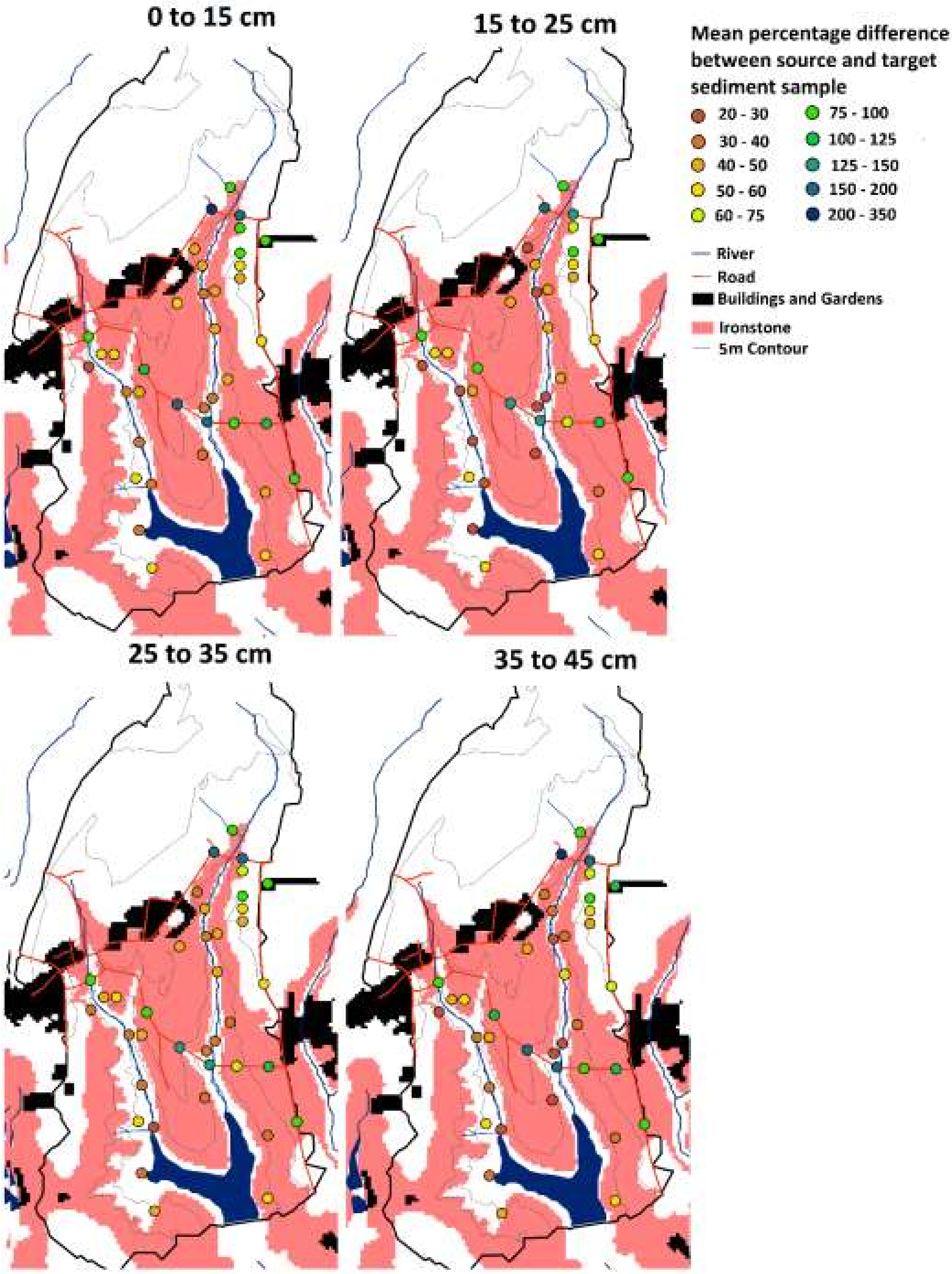
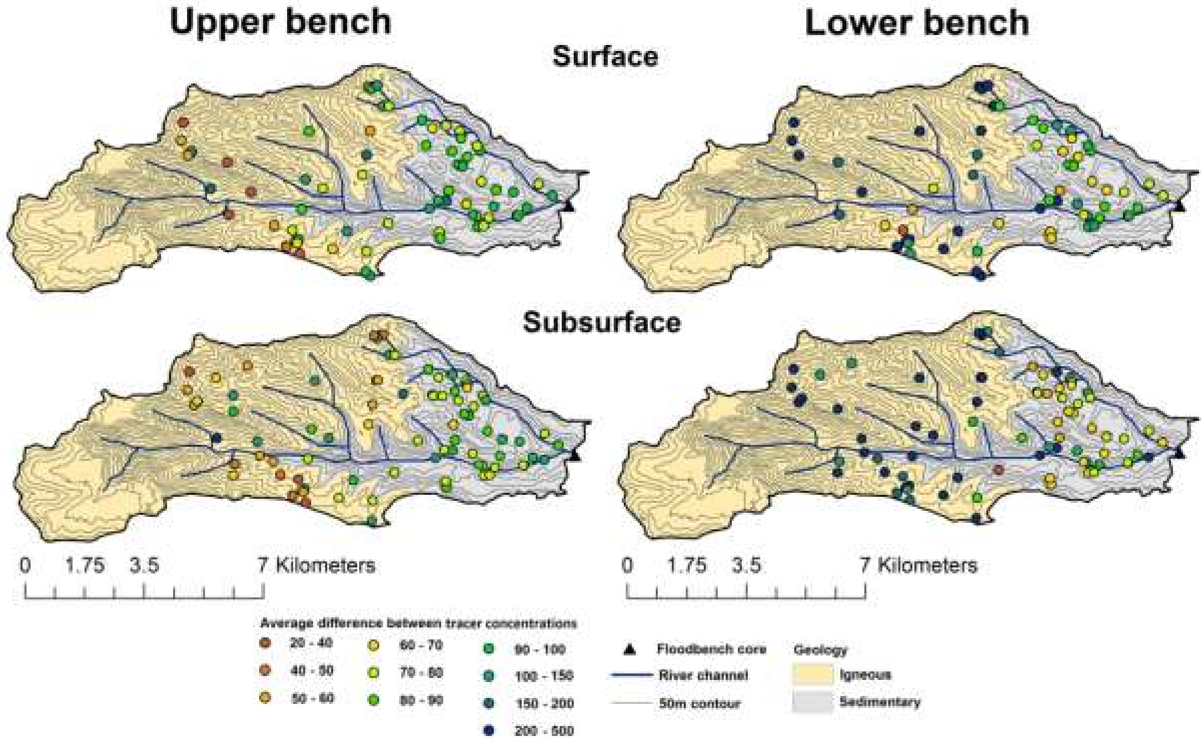

0 to 15 cm
15 to 25 cm
25 to 35 cm
35 to 45 cm
Mean percentage difference between source and target sediment sample
20 - 30
30 - 40
40 - 50
50 - 60
60 - 75
75 - 100
100 - 125
125 - 150
150 - 200
200 - 350
River
Road
Buildings and Gardens
Ironstone
5m Contour

Fig 6


Upper bench
Lower bench
Surface
Subsurface
0 1.75 3.5 7 Kilometers
0 1.75 3.5 7 Kilometers
Average difference between tracer concentrations
20 - 40
40 - 50
50 - 60
60 - 70
70 - 80
80 - 90
90 - 100
100 - 150
150 - 200
200 - 500
Floodbench core
River channel
50m contour
Geology
Igneous
Sedimentary