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Sediment fingerprinting in agricultural catchments: A critical re-examination of source discrimination and data corrections

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ABSTRACT

Fine sediment source fingerprinting techniques have been widely applied in agricultural river catchments. Successful source discrimination in agricultural environments depends on the key assumption that land-use source signatures imprinted on catchment soils are decipherable from those due to other landscape factors affecting soil and sediment properties. In this study, we re-examine this critical assumption by investigating (i) the physical and chemical basis for source discrimination and (ii) potential factors that may confound source un-mixing in agricultural catchments, including particle size and organic matter effects on tracer properties. The study is situated in the River Tamar, a predominantly agricultural catchment (920 km^2) in south-west England that has also been affected by mining. Source discrimination focused on pasture and cultivated land uses and channel banks. Monthly, time-integrated suspended sediment samples were collected across seven catchments for a 12-month period. Physical and chemical properties measured in source soils and sediment included fallout radionuclides (^{137}Cs, excess ^{210}Pb), major and minor element geochemical constituents, total organic carbon and particle size. Source discrimination was entirely dependent on differences in tracer property concentrations between surface and sub-surface soils. This is based on fallout radionuclide concentrations that are surface-elevated, while many geochemical properties are surface-depleted due to weathering and pedogenetic effects, although surface soil contamination can reverse this trend. However, source discrimination in the study catchments was limited by (i) rotation of cultivated and pasture fields resulting in reduced differences between these two sources, and (ii) the cultivated source signature resembling a mix of the pasture and channel bank sources for many tracer properties. Furthermore, a combination of metal pollution from abandoned historic mines and organic enrichment of sediment from upland areas of peaty soils resulted in the non-conservative behaviour of some tracer properties in several catchments. Differences in the particle size and organic carbon content of source soils could explain much of the variation in these properties in downstream sediment, rather than selective transport effects. Inconsistent relationships between particle size, organic carbon and tracer property concentrations further undermined the basis for the use of widely applied corrections to tracer datasets. Sensitivity analysis showed that correcting source tracer data for differences in organic matter can produce large changes to source contribution estimates that cannot be justified, and such corrections should not be used. Confounding factors related to poor source discrimination and non-conservative behaviour are highly likely to affect sediment fingerprinting studies in many agricultural catchments. As a result, estimates of source contributions in many fingerprinting studies may contain significant unquantified errors.

1. Introduction

Fine sediment fingerprinting involves the discrimination of sediment sources based on differences in source material properties and quantification of the relative contributions from these sources to sediment delivered downstream in river catchments. The fingerprinting procedure employs statistical testing of a range of source material tracer properties to select a subset that discriminate sources (Collins and Walling, 2002). These tracers may include geochemical, radionuclide, mineral magnetic, organic constituent, stable isotope and colour properties (Foster and Lees, 2000). Source un-mixing requires solutions to a set of linear equations that represent the value of an individual tracer property in sediment as a function of the sum of the values of that tracer for each source multiplied by the unknown proportional contribution from each source. Solutions are obtained using optimisation techniques that minimise the sum of errors associated with the equations (Yu and Oldfield, 1989; Collins et al., 1997; Walden et al., 1997).

Multi-parameter sediment source fingerprinting techniques were initially developed in agricultural catchments, and sought to discriminate pasture, cultivated and forest land uses as well as channel bank sources (Peart and Walling, 1986; Walling et al., 1993; Walling and

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Woodward, 1995). Other early approaches to determine fine sediment sources were based exclusively on selected properties such as fallout radionuclides (Wallbrink and Murray, 1993; Wallbrink et al., 1998) or mineral magnetic measurements (Oldfield et al., 1979; Yu and Oldfield, 1989). The key difference between these early approaches was the a priori selection of specific tracer properties based on well-established source differences, in contrast to multi-parameter source fingerprinting which relies on statistical selection of a subset of properties to discriminate sources. There remains a notable divide in the literature between the multi-parameter fingerprinting studies and those that use only pre-selected tracer properties, most commonly the fallout radionuclides ^{137}Cs and excess ^{210}Pb.

Since the initial studies there has been a rapid expansion in research output based on sediment source fingerprinting techniques (see reviews by Walling, 2005; Mukundan et al., 2012). Besides land use or channel bank sources, other fingerprinting studies have focused on discriminating sources according to geological zones based on soil geochemical as well as spectral–colorimetric properties (e.g. Douglas et al., 2009; Evrard et al., 2011; D'Haen et al., 2012; Legout et al., 2013). Fingerprinting studies have also examined sediment sources in urban environments (Carter et al., 2003; Poleto et al., 2009), forest environments including harvest areas and roads (Motha et al., 2003), and in forest areas disturbed by wildfire, where most studies use fallout radionuclide tracers to discriminate hillslope and channel bank sources (e.g. Wilkinson et al., 2009; Smith et al., 2013).

However, source fingerprinting techniques continue to be most widely applied in agricultural catchments (e.g. Owens et al., 2000; Gruszowski et al., 2003; Stutter et al., 2009; Collins et al., 2010a; Martínez-Carreras et al., 2010b; Blake et al., 2012). This reflects demand from land management agencies for information on fine sediment sources and the need to target resources to reduce elevated sediment pollution from agriculture (Gellis and Walling, 2011). In this context, source discrimination continues to focus on land use, while recent studies have sought to extend this to include sources such as damaged road verges, urban street dust and farm track surfaces (Collins et al., 2010a, 2012). With the demand for greater levels of detail and hence inclusion of additional sources related to highly specific landscape features, it is very important to consider the physical and chemical basis for source discrimination that underpins the entire multi-parameter fingerprinting method.

Recent studies tend to present fine sediment source fingerprinting as a robust and highly transferable technique that can deliver accurate estimates of source apportionment with high precision in a range of catchment environments. However, there is a need for further investigation of the extent to which land use-based sources of fine sediment within agricultural catchments can be reliably discriminated and apportioned. A critical assumption underpinning the widespread use of source fingerprinting in agricultural catchments is that land-use source signatures imprinted on catchment soils are decipherable from those due to other landscape factors affecting soil and sediment properties, such as differences in geology, soil type or previous land-use effects (e.g. historic mining). This key underlying assumption for discriminating and apportioning land-use sources has not been adequately acknowledged or challenged in the literature to date.

In this study, we re-examine the application of the fine sediment source fingerprinting procedure to discriminate land use and channel bank sources in agricultural river catchments. The study focuses on the discrimination of pasture, cultivated and channel bank sources of suspended sediment. The objectives are (i) to identify the physical and chemical basis for source discrimination by tracer properties selected using the fingerprinting procedure in agricultural catchments, and (ii) to re-examine the treatment of tracer data for particle size and organic matter effects. It is essential to establish that land use and channel bank source categories can be discriminated and apportioned with confidence, given that these sources form part of all fingerprinting studies in agricultural catchments. Additionally, because source fingerprinting produces proportional results, a large error in the estimated contribution for one source must affect the results for one or more other sources. Therefore, it is very important that errors in source discrimination and apportionment are constrained, and preferably contextualised using catchment sediment load data to assess source-specific mass contributions.

2. Methods

2.1. Study catchments

The study was situated in the River Tamar, a predominantly agricultural catchment located in south-west England (Fig. 1). The river forms the main boundary between the counties of Devon and Cornwall and drains south into the Tamar Estuary at Plymouth. All source soil and river sediment sampling were conducted above the tidal limit. River monitoring sites were situated at 7 locations comprising 6 sub-catchments nested within the main Tamar catchment (920 km^2) upstream of the village of Gunnislake, the study catchment outlet. The monitored sub-catchments include the Rivers Carey (67 km^2), Inny (97 km^2), Kensey (38 km^2), Lyd (219 km^2), Ottery (124 km^2), and a second measurement site on the main channel of the Tamar in the upper catchment (Tamar Upper; 238 km^2).

The topography of the River Tamar is characterised by short, steep hillslopes in the lower and mid-catchment and lower relief, undulating

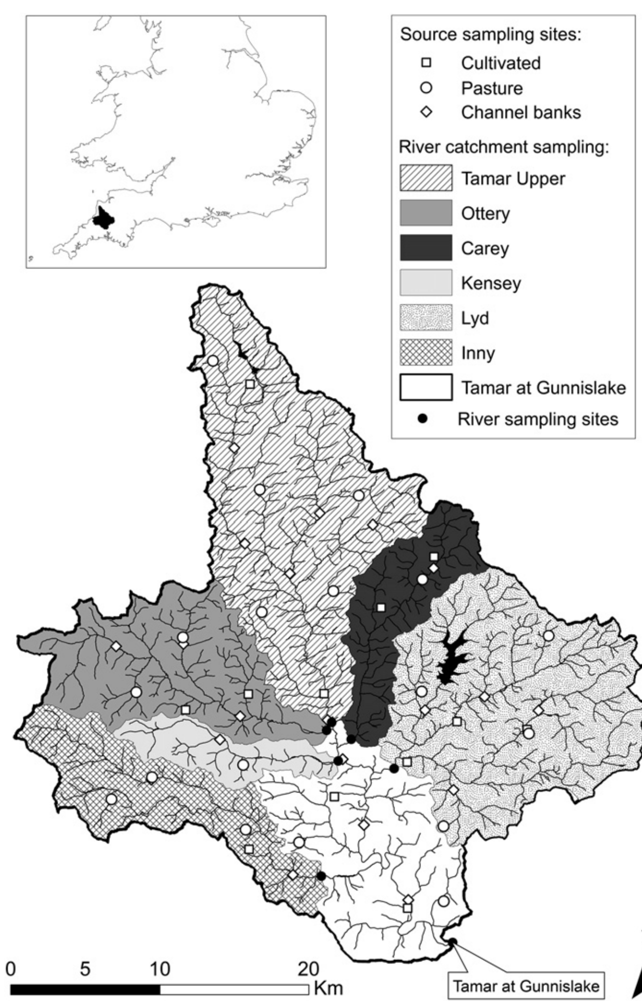

Fig. 1. River Tamar catchment with source soil and river sediment sampling locations displayed.