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**Sediment fingerprinting as a tool to identify temporal and spatial variability of sediment sources and transport pathways in agricultural catchments**

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1 Sediment fingerprinting as a tool to identify temporal and spatial variability of sediment sources and  
2 transport pathways in agricultural catchments

3

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14 Abstract

15 Management strategies to reduce soil loss and sediment delivery from agricultural land requires an  
16 empirical understanding of sediment sources. Sediment fingerprinting is a technique to apportion  
17 sources to a downstream sediment sample which, when applied at high spatial and temporal resolutions,  
18 can offer insights into catchment sediment dynamics. However, developing an over-arching tool can be  
19 hindered due to indeterminate interactions such as, for example, landuse, soil and geological conditions  
20 and multiple sediment source pressures. To address this, a multi-proxy sediment fingerprinting  
21 approach was used in three catchment observatories in Ireland, characterised and referred to by their  
22 predominant soil drainage and land use characteristics: poorly-drained grassland, well-drained arable  
23 and moderately-drained arable. Potential sediment source groups: channels, field topsoils, and roads,  
24 were sampled. Target sediment samples were collected from six sites within each catchment over  
25 approximately two-years from May 2012 to May 2014. Geochemical, mineral magnetic and  
26 radionuclide tracers were measured in source and target sediment samples and, following justified tracer  
27 selection, source proportions were estimated using an uncertainty inclusive un-mixing model. Overall,  
28 the poorly-, well- and moderately-drained catchments exported 828, 421 and 619 tonnes, respectively  
29 ( $36, 19$  and  $33 \text{ t km}^{-2} \text{ yr}^{-1}$ ). Estimated source contributions from channel, field topsoil and road groups  
30 were overall, 67%, 27% and 4% in the poorly-drained grassland, 53%, 24% and 24% in the well-drained  
31 arable and 9%, 82% and 8% in the moderately-drained arable catchment outlets. Sub-catchment source  
32 estimates were generally consistent with the outlet over space and time. Short-term activation of  
33 previously unidentified transport pathways were detected, for example, field sources transported by the  
34 road network in the well-drained catchment. In catchments with high hydrological surface connectivity  
35 (moderate and poor soil drainage), exposed soils were most sensitive to soil erosion and sediment  
36 delivery. Where groundcover is maintained on these soils, sediment connectivity was diminished and  
37 flow energy is transferred to the stream network where channel bank erosion increased. In the well-  
38 drained catchment, sub-surface flow pathways dominated and consequently channel sources, broadly  
39 representative of subsoil characteristics, were the largest sediment source. Sediment connectivity  
40 contrasted in the studied agricultural catchments according to source availability, and erosion, transport

41 and delivery processes. Effective sediment management strategies in intensive and intensifying  
42 agricultural catchments must consider sediment loss risk resulting from catchment specific sediment  
43 connectivity and emphasise mitigation strategies accordingly.

44

45 Keywords:, soil erosion, water quality, agriculture, catchment management, connectivity

## 46 **1 Introduction**

47 Intensive agricultural systems, resulting in enhanced soil erosion and sediment delivery can  
48 pose risks to aquatic ecosystems such as rivers and lakes (Collins and Zhang, 2016; Borelli et  
49 al., 2017; Tiecher et al., 2017; Vanwalleggem et al., 2017). In agricultural catchments,  
50 fluctuations in groundcover due to arable cultivation or livestock poaching (soil structural  
51 damage due to animal trampling) exposes the soil surface to erosional processes, thereby  
52 increasing their sensitivity to soil erosion and subsequent sediment loss (Haygarth et al., 2006).  
53 Land management, such as installation of artificial drainage, promotes aeration and alleviates  
54 excess soil moisture, thereby increasing the productivity of soils (Ibrahim et al., 2013). This  
55 also increases the efficiency of hydrological transfers from hillslopes to channels and runoff  
56 ratios (Shore et al., 2013). Moreover, landscape modifications interact with local heterogeneous  
57 catchment attributes (landscape position, slope, soil drainage, antecedent conditions) and  
58 rainfall to alter the distribution of soil erosion and sediment delivery, i.e., sediment connectivity  
59 (Sherriff et al., 2016).

60

61 In waterbodies, augmented supply of sediments to the channel bed can cause degradation of  
62 aquatic habitats resulting in reduced species diversity, as specifically noted in Ireland (Davis  
63 et al., 2018) and France (Descloux et al., 2013), and extensively reviewed throughout the world  
64 by Kjelland et al. (2015). High suspended sediment concentrations in aquatic ecosystems, for  
65 example following rainfall events, also reduce habitat quality for example resulting in increased  
66 drifting of invertebrates, commonly used as bioindicators (Kjelland et al., 2015; Béjar et al.,  
67 2017). Overall, reduction of ecological diversity challenges the achievement of ecologically  
68 “good” status as required under the EU Water Framework Directive (WFD; 2000/60/EC,  
69 Official Journal of the European Communities, 2000). Catchment management strategies  
70 require identification of sediment sources and an understanding of the spatial and temporal

71 dynamics of physical processes to cost-effectively target and reduce on-farm soil loss and off-  
72 farm downstream sediment supply (Walling et al., 2008).

73

74 There are difficulties in fully defining catchment sediment risks and monitoring mitigations.  
75 Firstly, auditing individual soil erosion and sediment storage components into a catchment  
76 sediment budget demands considerable investigation time and resources (Walling and Collins,  
77 2008). Secondly, establishing an evidence-base, relating specific agricultural practices to  
78 different sediment sources and delivery pathway fluctuations over multiple seasons, requires a  
79 representatively long study period with observations at an appropriate resolution (Sherriff et  
80 al., 2015a). Alternative catchment-scale techniques such as sediment fingerprinting have,  
81 therefore, emerged as an effective management tool in river catchments (Gruszowski et al.,  
82 2003, Rowan et al., 2012; Thompson et al., 2013; Lamba et al., 2015).

83

84 The sediment fingerprinting approach assumes that physico-chemical properties of  
85 minerogenic sediment, the inorganic component, can be conserved along a transport pathway,  
86 providing the numerical basis to ‘unmix’ the the composite-signatures of suspended sediments  
87 samples during flood events or from sediment stores such as channel beds, floodplains and  
88 lakes (Pulley et al., 2015) and to apportion the relative contribution to their respective upstream  
89 sources (Haddadchi et al., 2013). The upstream catchment is subdivided into potential sources  
90 (or source group types) that can be distinguished by their properties, for example, according to  
91 land use (Gruszowski et al., 2003; Blake et al., 2012), lithology (Collins et al., 1998), or  
92 erosional processes (Fox and Papandicolou, 2008).

93

94 Sediment tracers typically employed include geochemistry, mineral magnetics and  
95 environmental radionuclides and are potentially numerous considering the availability of

96 modern analytical equipment (Pulley and Rowntree, 2016). However, selected tracers must be  
97 conservative (resistant to chemical transformation) and their environmental significance  
98 justified in terms of the ability to discriminate between environmentally relevant sources  
99 (Koiter et al., 2013). Furthermore, it is assumed the impact of physical processes (erosion,  
100 transport, deposition, and re-entrainment) on tracer concentrations due to particle size  
101 selectivity and organic matter variation, can be numerically corrected. Simple correction  
102 factors are commonly used (Collins et al., 2001), but the appropriateness of these is now  
103 disputed (Smith and Blake, 2014) and more refined approaches involving particle size  
104 fractionation are an alternative (Motha et al., 2004; Small et al., 2004). Sediment contributions  
105 from each source are determined using statistically-based un-mixing algorithms, frequently  
106 accompanied by uncertainty estimates (Franks and Rowan, 2000; Sherriff et al., 2015b).

107

108 Sediment fingerprinting studies have been applied across a range of scales designed to explore  
109 the variability of sediment sources to a single area of impact, e.g., lake, degraded gravel habitat,  
110 or streams (Pulley et al., 2015). Particular advances have included assessing high-resolution  
111 temporal changes in sediment sources and investigations across hydrological regimes (Cooper  
112 and Krueger, 2017; Rose et al., 2018; Tiecher et al., 2018). However, the negative impacts of  
113 excessive sediment transport and/or deposition may extend far upstream of a catchment outlet  
114 (Fryirs et al., 2007). As such, there is a need to define sub-catchment variability of sediment  
115 sources to overcome the indeterminate potential of interacting land use, soil/geology and source  
116 variability issues that exist with sediment dynamics in catchments. This can facilitate  
117 interrogation of catchment hydrological and sediment connectivity processes inferred at the  
118 catchment scale, and how they relate to the spatially heterogeneous pattern of land use (e.g.,  
119 crop type, animal grazing) and land management (e.g., riparian vegetation, arrangement of farm  
120 tracks – Sherriff et al., 2016).

121

122 Correct identification of sediment sources and disentangling the processes controlling soil  
123 erosion, sediment entrainment, transfer and deposition will provide an evidence base for  
124 application of targeted on- and off-farm sediment management strategies (Rowan et al., 2012).  
125 This is particularly important in catchments with contrasting physical and agricultural land  
126 management characteristics where targeted strategies may be different (Sherriff et al., 2016).  
127 Appropriate source-based mitigation measures are necessary to prevent off-farm nutrient and  
128 sediment supply downstream (Evrard et al., 2007; Deasy et al., 2010). Successful application  
129 of suitable mitigation measures are essential to reduce on-farm nutrients and soil losses through  
130 the preservation of chemical, physical and biological soil quality (Cerdà et al., 2017). These  
131 are important considerations to reduce the environmental impact of intensive agriculture and  
132 also to offset the likely changes occurring as land becomes more intensively managed.  
133 Increased or changing land use (crop types, animal numbers), soil drainage and increased  
134 machine trafficking are all likely to occur under scenarios of agricultural intensification in  
135 Ireland, Europe and worldwide (Ewert et al., 2005; Coyle et al., 2016; Teshager et al., 2016).

136

137 The overall aim of this study was to use sediment fingerprinting to define the spatial and  
138 temporal variability of sediment sources of instream sediments in intensive or intensifying  
139 agricultural catchments. The sediment fingerprinting methodology used a multi-proxy suite of  
140 environmental radionuclides, geochemistry and mineral magnetism within a statistically based  
141 un-mixing framework. This method was applied in three catchment observatories in order to  
142 fulfil two objectives. Firstly, to assess relative magnitudes of sediment sources between  
143 catchments with contrasting land use and dominant soil drainage characteristics. Secondly, to  
144 assess the spatial and temporal variability of sediment sources within each catchment. This



145 analysis was used to recommend catchment and source specific measures to reduce the soil and  
146 sediment loss from land.

147 **2 Methods**

148 *2.1 Catchment observatories*

149 Sediment fingerprinting studies were focussed on three lowland intensive agricultural  
150 catchments in Ireland. Consistent with Sherriff et al. (2016), these catchments are named  
151 according to their dominant soil drainage and predominant land use types; poorly-drained  
152 grassland, well-drained arable and moderately-drained arable (Fig. 1).

153

154 The poorly-drained grassland catchment (11.0 km<sup>2</sup>) with median slopes of 3°, is located in  
155 south-east Ireland. It is geologically permeable owing to Ordovician Volcanics and  
156 metasediments of the Campile formation (Tietzsch-Tyler et al., 1994). However, overlying  
157 Groundwater Gley soils in the lowlands (Luvic Stagnosol, sandy loam - World Reference Base  
158 classification for soils, Creamer, 2014; 2016), influenced by the Irish Sea till subsoil, dominate  
159 the catchment area and consequently impede drainage once the well-drained upper horizons  
160 are saturated. Well-drained Brown Earths (Haplic Cambisol, loam) are found in limited areas  
161 of the uplands but overall surface flow hydrological pathways dominate in this catchment  
162 (Mellander et al., 2012). Consequently, artificial surface (open drainage ditches) and sub-  
163 surface (piped) drainage networks are widespread across the landscape. Grassland agriculture  
164 for sheep, beef and dairy grazing (77%) is the predominant land use, with arable crops (12%)  
165 primarily spring cereals contained to upland well-drained soils.

166

167 The well-drained arable catchment (11.2 km<sup>2</sup>) with median slopes of 4° is located in south-east  
168 Ireland. The catchment is geologically composed of slate and siltstones of the Oaklands  
169 Formation (Tietzsch-Tyler at al., 1994). This provides poor primary permeability but secondary  
170 productivity creates opportunities for water transfer through fissure flow. Overlying soils are  
171 predominantly well-drained Brown Earths (Haplic Cambisol, loam) with limited areas of

172 poorly-drained Groundwater Gleys (Haplic Gleysol, clay loam) in the eastern stream corridor  
173 leading to a dominance of sub-surface hydrological pathways in this catchment. Artificial  
174 drainage networks are limited to the poorly-drained catchment areas which are primarily  
175 utilised for permanent grassland (39% of catchment) for beef cattle and sheep grazing. Arable  
176 crops (54% of catchment), mostly spring barley, are supported by well-drained soils which  
177 undergo limited rotation between years.

178

179 The moderately-drained arable catchment (9.5 km<sup>2</sup>) is located in north-east Ireland and has  
180 median slopes of 3°. Geologically, it is characterised by calcareous greywacke sandstones and  
181 banded mudstones (McConnell et al., 2001) which results in a poorly productive aquifer  
182 (Mellander et al., 2012). The overlying soils are categorised as moderate and poorly-drained  
183 dominated by A-horizon loams and B-horizon clay loams underlain by fine till containing  
184 siliceous stones and fluvio-glacial sediments in the channel corridor (Haplic Cambisols, loam  
185 and Gleyic Fluvisol, silt). Surface hydrological pathways dominate in the catchment but a  
186 greater sub-surface hydrological influence has been detected during winter months (Melland  
187 et al., 2012; Mellander at al., 2012). Artificial drainage networks are dominant in poorly-  
188 drained areas but generally widespread across the catchment. A large proportion of mixed  
189 arable crops (42%) including winter-sown cereals, maize, potatoes among others are supported  
190 in the catchment and the other land use is permanent grassland (48%) to support grazing of  
191 sheep and cattle for dairy and beef.

192

### 193 *2.2 Fingerprinting sample collection*

194 Reconnaissance surveys identified six primary potential sediment sources in the study  
195 catchments: grassland topsoils, arable topsoils, damaged road verges, farm tracks, eroding  
196 channel banks and eroding ditch banks, which were sampled for analysis. Field sample

197 locations were randomly selected using a spatial dataset of fields stratified by their general land  
198 use, grassland or arable. The proportion of fields sampled from each land use approximately  
199 reflected the overall proportion of that land use contained within each catchment. Each field  
200 topsoil sample (0-5 cm) was collected with an auger and comprised a composite of multiple  
201 sub-samples within each field. In the poorly-drained grass, well-drained arable and moderately-  
202 drained arable catchments, 22 fields (grassland n=16; arable n=6), 24 fields (grassland n=9,  
203 arable n=16) and 30 fields (grassland n=19, arable n=11) were sampled, respectively.

204

205 Channel and drainage ditches were sampled during winter 2013 and 2014 when vegetation  
206 cover was low. Samples targeted actively eroding areas from which a composite sample of  
207 vertical bank section was collected with a trowel (from a stream reach no greater than the  
208 adjacent field width), with opposite river banks collected separately. In total, 62 channel  
209 samples were collected from the poorly-drained grass catchment, 15 from the well-drained  
210 arable catchment and 14 from the moderately-drained arable catchment. Time constraints  
211 associated with radionuclide isotope analysis resulted in analysis of a randomly selected sub-  
212 set of 30 samples (from the 62) which were deemed appropriate, based on the number of  
213 samples used to represent other potential sources, to characterise the channel sediment source  
214 in the poorly-drained grass catchment (Fig. 1). Active erosion of ditch (open field drain)  
215 channels was observed only in in the poorly-drained grassland catchment (n=4) and was  
216 sampled consistently with channel banks.

217

218 Surface scrapings of damaged road verges and farm tracks were collected with a trowel along  
219 a maximum road or track length of approximately 200 m and compositing, where relevant,  
220 both sides of the road into one sample (Fig. 1). Twenty-three, eight and 12 samples were

221 collected in the poorly-drained grass, well-drained arable and moderately-drained arable  
222 catchments, respectively.

223

224 River sediment samples were collected using time integrated suspended sediment (TISS)  
225 samplers (Phillips et al., 2000) at multiple locations within each study catchment from May  
226 2012 to May 2014 (Fig. 1). The samplers are constructed of a main body (98 mm internal  
227 diameter PVC pipe, 1 m length) capped with an upstream facing funnel and a closing  
228 downstream cap. Small diameter inlet/outlet tubes (4 mm diameter, approximately 20 cm  
229 length) facilitate the flow of water through the sampler. Suspended sediment particles are  
230 deposited inside the main body when leaving the inlet tube due to the reduction in velocity  
231 coinciding with the increase in diameter. Sediment samples were collected at 6-12 week  
232 intervals to assess seasonal changes in sediment sources (Table 1). Missing data were attributed  
233 to equipment malfunctions or insufficient sample quantity for analysis (using label locations  
234 and sample times in Fig. 1 and Table 1: MD3 – T14, MD1 and MD5 – T10, PD4 – T11, WD4  
235 – T3, WD6 – T7, T8 and T11). Site PD3 was removed following vandalism and a replacement  
236 site, PD1, established upstream to coincide with the deployment of samplers on 13/05/2013.  
237 At WD5, channel reconfiguration (following T10) resulted in cessation of sample collection at  
238 this location. Short term site inaccessibility (due to inundated channels) prevented the  
239 collection of the MD3 and WD1-WD5 at the end of T9. These samples were retrieved at the  
240 next collection occasion and were therefore representative of the two periods T9 and T10,  
241 combined.

242

### 243 *2.3 Suspended sediment load estimation*

244 Catchment outlet suspended sediment loads were estimated using turbidity (Solitax, Hach-  
245 Lange, Germany) and water level (m) data collected from a vented pressure-transducer (OTT

246 Orpheus-mini, OTT, Germany) located inside a stilling well at 10-min resolution from May  
247 2012 to May 2014. Turbidity data were converted to suspended sediment concentrations (SSC)  
248 using detailed cross-sectional turbidity-SSC calibrations (Sherriff et al., 2015a). The velocity-  
249 area rating method for gauging instantaneous discharge was used to calculate discharge over a  
250 non-standard Corbett flat-v weir (Corbett Concrete, Cahir, Ireland) using WISKI-SKED  
251 software. Meteorological data; 10-min rainfall, air temperature, relative humidity, radiation and  
252 wind-speed were collected from lowland weather stations in each catchment (BWS200,  
253 Campbell Scientific).

254

#### 255 *2.4 Laboratory analysis*

256 Geochemical, radionuclide and mineral magnetic analysis was conducted on soil and sediment  
257 samples. Geochemical elements (Cd, Co, Cr, Cu, Mn, Ni, Pb and Zn) were analysed using an  
258 Agilent ICP-OES (Santa Clara, US) following microwave assisted acid digestion (USEPA,  
259 1996) to obtain total concentrations ( $\text{mg kg}^{-1}$ ). Radionuclide activity mass concentrations ( $\text{Bq}$   
260  $\text{kg}^{-1}$ ) of  $^{210}\text{Pb}$ ,  $^{234}\text{Th}$ ,  $^{235}\text{U}$ ,  $^{214}\text{Pb}$ ,  $^{137}\text{Cs}$ ,  $^{228}\text{Ac}$ ,  $^{40}\text{K}$  were measured using a low background Ortec  
261 HPGe gamma spectrometer detector (Model no. GEM-FX7025-S) after samples were radon-  
262 sealed inside 55 mm petri dishes for a minimum of 30 days to determine the unsupported  
263 fraction ( $^{210}\text{Pb}_{\text{unSUPP}}$ ), of  $^{210}\text{Pb}$  activity (Foster et al., 2007; Rowan et al., 2012). Detector  
264 calibration was achieved using a National Physics Laboratory mixed-gamma standard (R08-  
265 03) within standardised mass/geometries (1, 5, 10 g). Mineral magnetic measurements, the  
266 mass-specific low field susceptibility ( $\chi_{\text{LF}} - 10^{-6} \text{ m}^3 \text{ kg}^{-1}$ ), high field susceptibility ( $\chi_{\text{HF}} - 10^{-6}$   
267  $\text{m}^3 \text{ kg}^{-1}$ ), frequency-dependent susceptibility ( $\% \chi_{\text{FD}}$ ), anhysteretic remanence magnetisation  
268 ( $\chi_{\text{ARM}} - 10^{-7} \text{ Am}^2 \text{ kg}^{-1}$ ), saturation isothermal remanent magnetisation (SIRM at 1 T  $- 10^{-5} \text{ Am}^2$   
269  $\text{kg}^{-1}$ ), backfield IRM measurements  $\text{IRM}_{\text{soft}}$  and  $\text{bIRM}_{\text{hard}}$  were completed, and ratios  
270  $\text{SIRM}/\chi_{\text{LF}}$ ,  $\text{SIRM}/\chi_{\text{ARM}}$ ,  $\chi_{\text{ARM}}/\chi_{\text{LF}}$  and the H-ratio ( $0.5 * (\text{SIRM}-\text{bIRM}_{40})$ ) calculated.

271

272 The specific surface area (SSA –  $\text{m}^2 \text{kg}^{-1}$ ) of soil and sediment samples were measured using a  
273 Malvern Mastersizer Hydro 2000G (range 0.02 to 2000  $\mu\text{m}$ ) following organic matter removal  
274 and chemical/physical dispersion (Fenton et al., 2015). Total carbon (TC) and total organic  
275 carbon (TOC – following acid treatment of the inorganic fraction with hydrochloric acid  
276 (Massey et al., 2013)) were analysed on a LECO Truspec CN analyser (LECO Corporation,  
277 Michigan, USA) as a proxy for organic matter content. Samples were individually corrected  
278 for particle size (corrected tracer concentration = measured tracer concentration/SSA –  
279 Gruszowski et al., 2003) and organic matter content (corrected tracer concentration = particle  
280 size corrected concentration/ % organic carbon).

281

#### 282 *2.4 Statistical analysis*

283 The capability of an individual tracer to distinguish between sources was assessed using the  
284 Kruskal-Wallis test (SPSS v. 22.0; IBM, USA) –  $p < 0.05$ ), with tracers that differed  
285 significantly between sources being retained for further analysis. This was followed by  
286 pairwise analysis using the Dunn-Bonferroni test (adjusted  $p < 0.05$ ) to determine significant  
287 differences between groups. Under spatial interrogation, source sample tracer characteristics  
288 did not display sub-catchment trends within each study catchment; therefore, all source  
289 samples, regardless of their catchment location, were utilised for un-mixing suspended  
290 sediment samples. Non-conservative behaviour of tracers in suspended sediment samples was  
291 identified by values exceeding the range defined by source samples were removed from further  
292 analysis (Mukundan et al., 2010; Smith and Blake, 2014). Removal of  $^{210}\text{Pb}_{\text{unSUPP.}}$  and  $^{137}\text{Cs}$   
293 where concentrations were  $0 \text{ Bq kg}^{-1}$  in suspended sediment samples occurred as, although  
294 tracers were not non-conservative, their impact on result predictions and uncertainty were  
295 considerable, consistent with Sherriff et al. (2015b). All remaining tracers were subsequently

296 interrogated to justify their environmental significance (Koiter et al., 2013). Tracer values were  
297 entered into Multiple Discriminant Analysis (MDA) to determine the discrimination capability  
298 of the resulting tracer set for each outlet and sub-catchment suspended sediment sample (Table  
299 5). Subsequently, source contributions were un-mixed using the uncertainty inclusive FR2000  
300 model (Franks and Rowan, 2000), with uncertainties determined on probability distributions  
301 from the input dataset of tracer values and target sediments. For simplification, the distance  
302 between the upper and lower uncertainty values, representing the 95<sup>th</sup> percentile for each  
303 source, were combined for each target sediment sample.



### 304 **3 Results and discussion**

#### 305 *3.1 Tracer selection*

306 The six sampled source groups could not be discriminated using MDA when deploying the full  
307 tracer arrays in all three study catchments (Yu and Rhoads, 2018). However, three composite  
308 ‘parent’ groups were identified and attributed to similar soil loss processes:

- 309 • Channels: comprising channel banks and ditches
- 310 • Field topsoils: comprising arable and grassland topsoils
- 311 • Roads: comprising road verges and tracks.

312 Source group tracer summaries are shown in Table 2 – poorly-drained grassland, Table 3 –  
313 well-drained arable and Table 4 – moderately-drained arable and hereafter the use of ‘channel’,  
314 ‘field’ and ‘road’ refers to parent groups unless specified. Tracers that were statistically non-  
315 significant between at least two sources (Kruskal Wallis  $p > 0.05$ ) were removed from analysis  
316 (Tables 2-4).

317

318 In all catchments, road samples were generally elevated compared to other sources for mineral  
319 magnetic tracers ( $\chi_{LF}$ ,  $\chi_{HF}$ ,  $\chi_{ARM}$ , SIRM) and/or metallic elements Cu, Pb and Zn, reflecting  
320 possible inputs from vehicle exhausts (Rose et al., 2018). This trend was not reflected in the  
321 well-drained arable catchment likely due to greater ferrimagnetic minerals (high  $IRM_{soft}$ ) due  
322 to iron-rich geology. Surficial sources were well defined by  $^{137}Cs$  in the poorly-drained  
323 grassland (Table 2) and well-drained arable catchments (Table 3). In the moderately-drained  
324 arable catchment (Table 4), however, this tracer failed to distinguish between any sources.

325

326 All catchments showed higher  $\% \chi_{FD}$  in field topsoils soils than channel sources. Maher and  
327 Taylor (1988) attributed elevated  $\% \chi_{FD}$  in topsoils to the production of magnetite grain coatings  
328 in the surface of poorly-drained soils. Similarly elevated values but less group variability in the

329 well-drained catchment was attributed to higher background ferromagnetic material rather than  
330 that produced in-situ. Higher concentrations of Cu and Ni in channel sources compared to  
331 topsoils were explained in the poorly-drained grassland and well-drained catchments by  
332 reduced weathering (and therefore depletion of element concentration) relative to topsoils  
333 (Smith and Blake, 2014). Soil heterogeneity prevented this trend in the moderately-drained  
334 arable catchment. Channel samples could be considered a good representation of other sub-  
335 surface sources such as drains, gullies and tracks which lose subsoil material (Collins et al.,  
336 2010; Cooper and Krueger, 2017). In the poorly-drained grassland catchment, a thick and low-  
337 permeability marine clay subsoil present at 1.5 – 2 m (below surface) was successfully  
338 characterised in this source group due to occasional exposure at the base of channel bank  
339 sections (Mellander et al., 2015).

340

341 The suspended sediment samples showed consistent tracer non-conservativeness (more than  
342 36% of samples within a catchment) in two tracers, Cd and Mn, from the poorly-drained  
343 grassland catchment; and four tracers,  $SIRM/\chi_{ARM}$ , Cd, Cr, and Mn from the moderately-  
344 drained arable catchment which were consequently removed from analysis. Removal of non-  
345 conservative tracers in individual suspended sediment samples were similarly removed from  
346 their associated source dataset. This did not impact significantly on the cumulative  
347 discriminatory power as determined by the MDA. The source groupings of the maximal tracer  
348 set (Fig. 2) and the distribution of discriminatory power for all suspended sediment samplers  
349 (Table 5) qualified against other acceptable values reported elsewhere (Lamba et al., 2015;  
350 Theuring et al., 2015).

351

352 *3.2 Catchment outlet export and sediment source predictions over time*

353 Total suspended sediment export over the study period was 828, 619 and 421 tonnes in the  
354 poorly-drained grassland, well-drained arable and moderately-drained arable catchments,  
355 respectively (equivalent to 36, 19 and 33 t km<sup>-2</sup> yr<sup>-1</sup>, respectively). These were higher than the  
356 longer-term average yields (hydrological years 2009-2012) previously measured, 25, 12 and  
357 24 t km<sup>-2</sup> yr<sup>-1</sup> in the poorly-drained grassland, well-drained arable and moderately-drained  
358 arable catchments, respectively, but did not exceed the maximum annual sediment yields  
359 (Sherriff et al., 2015a).

360

361 Load specific un-mixing using median predictions indicated channels were the dominant  
362 sediment sources in the poorly-drained grassland catchment outlet samples (PD6 – Fig. 3a)  
363 which overall accounted for 67% (range 42-77%), or 554 t, of the suspended sediment load  
364 (SSL). This confirms previous studies suggesting that proximal sediment sources, likely  
365 channel banks, were primary contributors to the SSL here (Sherriff et al., 2016). Field topsoils  
366 overall contributed 27% (range 19-49%) to the SSL and did not increase with greater sediment  
367 export (Sep-12 to Apr-13 and Dec-13 to Mar-14) confirming that, despite good connectivity,  
368 hillslope sediment loss risk was largely reduced by permanent pasture groundcover. Roads  
369 were a negligible sediment source in this catchment (total 4%, range 1-17%) related to their  
370 distal location relative to the stream network. Additionally, roads were frequently bordered by  
371 road-side ditches designed to divert and store surface water. Previous analysis in these  
372 catchments has indicated that disconnecting ditches from the stream network provides a  
373 sediment sink to prevent subsequent transport to watercourses (Shore et al., 2013).

374

375 Sediment sources were predominantly channel derived at the well-drained arable catchment  
376 outlet – WD6 (53%, range 0-69%), with smaller proportions attributed to field topsoils (24%,  
377 range 9-40%) and roads (24%, range 18-47%) over the study period (Fig. 3b). Higher average

378 sediment exports (greater than  $\sim 2 \text{ t day}^{-1}$ ) were less frequent compared to the other catchments  
379 due to the less-flashy hydrological response and dependence of event-scale rainfall  
380 characteristics on sediment connectivity (Sherriff et al., 2016). The stream network is largely  
381 (approximately 66%) contained by a woodland riparian corridor thereby stabilising channel  
382 bank soils through root networks and reducing the potential for bank erosion (Polvi et al.,  
383 2014). Despite this, observed and reported representative channel composite sources are from  
384 localised drainage programmes (two drainage projects occurred during January 2014 on the  
385 north-south tributary with banks without woodland), subsurface sediment sources to tile drain  
386 flow (e.g. Deasy et al. 2009), and other efficient subsurface hydrological pathways supported  
387 by fractured bedrock (Warsta et al., 2013; Sherriff et al., 2016).

388

389 Field topsoils dominated sediment sources in the moderately-drained arable catchment,  
390 accounting for 82% of the total yield (range 59-93%) with 9% attributed to channels (range 0-  
391 17%) and 8% to roads (range 0-23%) over the study period (Fig. 3c). Foucher et al. (2014)  
392 similarly reported the dominance of surface sources in a row crop arable catchment in France  
393 with widespread sub-surface drainage. Previous inferences using hysteresis analysis (Sherriff  
394 et al., 2016) assigned sediment export risk to bare arable fields with good hillslope connectivity  
395 (resulting from predominately moderately- and poorly-drained soils), which these data  
396 confirm. Similarly to the well-drained catchment, increased bank stabilisation from the  
397 extensive root networks of riparian woodland likely reduced the risk of bank erosion for much  
398 of the stream network.

399

### 400 *3.3 Sub-catchment sediment sources and uncertainty over space and time*

401 In all study catchments, sub-catchment sediment sources were generally consistent with outlet  
402 predictions (Fig. 4). This was most evident in the poorly-drained grassland catchment whereby

403 channels were consistently the dominant sources throughout the catchment. This indicates the  
404 processes of soil erosion and sediment transfer were consistent. Higher field topsoil  
405 contributions occurred on average in the lower poorly-drained grassland catchment sites (PD4,  
406 PD5 and PD6) indicating that arable topsoils primarily located on well-drained soils in the  
407 upper catchment were not contributing significantly. There were some elevated field  
408 contributions in samples T9 and T10 but low catchment outlet sediment export here suggests  
409 minor impacts on the load particularly in T9 where result predictions were associated with  
410 greater uncertainty (Fig. 5).

411

412 The well-drained arable sub-catchment samples showed, similarly to the outlet, channel  
413 sources dominated. However, greater field topsoil contributions occurred during periods of  
414 increased SSL (T4, T5 and T12) reflecting greater sediment connectivity during wetter periods.  
415 Sediment source estimations at sites WD1 and WD4, which drain the north-south tributary,  
416 were consistently elevated for field topsoils relative to other sites. This suggests sediment  
417 connectivity was greater here, where hillslopes were steeper with a narrow stream corridor and  
418 a reduced riparian woodland cover (Wischmeier and Smith, 1978; Gurnell, 2014). Increased  
419 field topsoils here coincided with greater road contributions suggesting runoff over  
420 impermeable surfaces is an important delivery mechanism for field eroded soils. Road verges,  
421 in addition would contribute as a separate sediment source (Gruszowski et al., 2003). At WD6  
422 during T13, channel sources were negligible and showed inconsistencies over time (WD6) and  
423 space (T13 samples) – but with high uncertainty in the predictions (Fig. 5).

424

425 In the moderately-drained arable catchment, field topsoils contributed on average greater than  
426 80% across all sub-catchment sites over time and showed less variability relative to other study  
427 catchments. The availability of sediment sources and surficial mechanisms of soil erosion,

428 sediment transport and delivery were, therefore, consistent across the catchment and over time  
429 (Sherriff et al., 2016). Similarly to the downstream sites in the well-drained arable catchment,  
430 MD5 and MD6 in this catchment show higher channel sediment contributions from T1-T7.  
431 These samples corresponded with greater sediment loads suggesting increased inundation of  
432 the channel and short term channel bank source availability, for example, stock access  
433 degrading channel banks or channel management (during summer months) or increased  
434 cumulative flow. The estimated proportion of road contributions was inconsistent at sub-  
435 catchment sites but showed a relatively consistent elevation at MD6. Site MD1 reported high  
436 road group contributions in T2, T5, T7 and T9, however, the corresponding uncertainty for  
437 these predictions were relatively high also.

438

439 Despite relatively high spatial resolution source sampling relative to small catchment sizes  
440 ( $\sim 10 \text{ km}^2$ ), the variability of tracer values between sources was frequently lower than the  
441 variability of single sources. This prevented the discrimination of the intended potential sources  
442 ( $n=6$ ) identified at the outset of the investigation (Rowan et al., 2012; Rose et al., 2018).  
443 Definition of additional sources (improved dimensionality), such as land use type within each  
444 catchment (arable versus grassland), would improve the resolution of source provenance and,  
445 in turn, improve the understanding of the interactions between sediment sources and catchment  
446 processes, and onward risk assessment and mitigation. Gruszowski et al. (2003) successfully  
447 distinguished grassland and arable topsoils using tracers  $\chi_{\text{HF}}$ ,  $\chi_{\text{ARM}}$ ,  $\text{IRM}_{880}$ , Fe, Al, Na and Cu.  
448 Equivalent tracers measured in this study were, however, not capable of discriminating arable  
449 and grassland topsoils and may be attributed to greater arable and grassland crop rotation.  
450 Measurement of additional tracers such as soil enzymes and crop-specific compound specific  
451 stable isotopes may be useful to provide greater dimensionality (Nosrati et al., 2011; Blake et  
452 al., 2012). Despite this, the sediment fingerprinting approach validated, and are consequently

453 validated by, the catchment characteristics shown by SSC-discharge hysteresis analysis in these  
454 study catchments (Sherriff et al., 2016). The confirmation is a significant finding as storm event  
455 discharge-suspended sediment hysteresis analysis compared to sediment fingerprinting  
456 strategies requires less in terms of analytical sophistication but more in terms of longer-term  
457 high resolution river-side monitoring.

458

### 459 *3.4 Implications for catchment management*

460 Despite high agricultural intensity (more than 90% utilisation area and high stocking  
461 density/crop yields – Sherriff et al., 2015a), source provenance results indicated channel  
462 derived sediments were dominant in some settings. These were likely accelerated by field-scale  
463 agricultural management such as the presence and configuration of drainage systems which  
464 can increase channel flow velocities. These landscape modifications (aimed at reducing excess  
465 soil moisture and increasing the utilisable area for agriculture on hillslopes) may occur in  
466 combination with bank destabilisation which are also responsible for increased stream energy  
467 and consequently bank erosion. Accordingly, mitigation measures designed to reduce flow  
468 velocities may dissipate stream power and encourage deposition of entrained sediments. For  
469 example, field wetlands positioned at the edge of field or catchment outlets have been shown  
470 to retain  $0.01 - 6 \text{ t ha}^{-1} \text{ yr}^{-1}$  of sediment and on-farm buffer strips have reported 2 – 50% efficacy  
471 (Ockenden et al., 2012; Collins et al., 2018).

472

473 Sediment delivery from field topsoils was greater where low ground cover coincided with good  
474 hydrological connectivity (consistently in catchments with poor- or moderate soil drainage or  
475 sporadically following extreme rainfall events on well-drained soils). Consistently, low risk  
476 soils have also found to be a minor contributor to catchment sediments loads in Brazil (LeGall  
477 *et al.*, 2017). Strategies emphasising soil conservation or dis-connecting hydrological pathways

478 may contribute to reduced sediment delivery. On-field, prevention of low groundcover periods,  
479 for example by retaining crop residues or increasing surface roughness through conservation  
480 tillage practices, should be considered to reduce soil erodibility and efficacy of hydrological  
481 transport across soil surfaces (Deasy et al., 2010). In grassland fields, prevention of soil  
482 structure degradation and over-grazing may reduce soil erosion risk. Measures to reduce  
483 hillslope-to-channel sediment connectivity to encourage deposition of entrained sediments,  
484 such as abovementioned field wetlands, buffer-strips or unploughed margins are potential  
485 sediment mitigation measures (Rickson, 2014),

486

487 Management of the public and farm road network to intercept road derived or road transported  
488 sediments at road-stream intersections is a source and/or pathway requiring greater study.  
489 Although the proportion of road-derived sediments were generally low in the present study,  
490 their subsequent elevation with field topsoil contributions has implications for regulations to  
491 minimise soiled water transfers from farm roadways to waters by 2021 (European Union,  
492 2017). Similarly to field ditches, roadside ditches can be designed and managed to encourage  
493 deposition of associated particulates transported in the flow which consequently reduces  
494 sediment delivery to watercourses (Shore et al., 2016). However, installing such features in  
495 established agricultural areas may not be practical.

496

497 The separation of sediment load into three source groups, defined here in three catchments with  
498 contrasting land use and soil drainage combinations, provides a blueprint for sediment transfer  
499 vulnerability in agricultural catchments. Expanding the spatial resolution of source provenance  
500 shows good consistency of soil erosion and sediment transfer mechanisms within each  
501 catchment but also reveals additional processes which are undetectable at the catchment scale.  
502 Previous research in the study catchments (Shore et al., 2014; Mellander et al., 2015; 2016;



503 Sherriff et al., 2016, Thomas et al., 2016) and elsewhere (Fryirs et al., 2007; Dupas et al., 2015)  
504 has confirmed the influence of hydrological connectivity on the delivery of sediment and  
505 nutrients at catchment outlets and the importance of their interactions for in-stream ecology  
506 (Davis et al., 2018). Identification and mitigation of hillslope sediment losses must target  
507 critical source areas to maximise the success and cost-efficiency of mitigation measures (Shore  
508 et al., 2013; Thompson et al., 2013; Thomas et al., 2016). Controls on bank erosion of natural  
509 and artificial channels are required and have been typically overlooked in agri-environmental  
510 policy, at least at a European level (Collins and Anthony, 2008). The potential trade-off  
511 between reducing hillslope soil moisture to sustain or increase agricultural production and the  
512 initiation and acceleration of erosion at the field edge must be fully considered.

#### 513 **4 Conclusions**

514 The successful multi-proxy partitioning of parent sediment sources (field topsoil, channel and  
515 road) indicated contrasting hillslope versus channel influences in the catchment observatories,  
516 according to source availability and transport pathways. The main conclusions are:

- 517 • The poorly-drained grassland and moderately-drained arable catchments exported  
518 overall greater sediment load overall, 828 and 619 tonnes, respectively, than the well-  
519 drained arable catchment, 421 tonnes, due to a greater likelihood of sediment  
520 connectivity correlated with impeded soil drainage (or lack of infiltration capacity);
- 521 • At the catchment outlets, channel, field topsoil and road contributions were 67%, 27%  
522 and 4% in the poorly-drained grassland catchment, 53%, 24% and 24% in the well-  
523 drained arable catchment and 8%, 82% and 9% in the moderately-drained arable  
524 catchment.
- 525 • Greater sediment export in the well-drained catchment during three periods (T4, T5 and  
526 T12) corresponded with greater contributions from field topsoils, 29% and 24%,  
527 respectively, which were attributed to the establishment of surface hydrological  
528 connectivity and consequent surface erosion and overland flow transport following  
529 extreme rainfall events;
- 530 • Spatially, sediment sources were less variable in predominately poorly-drained  
531 grassland or moderately-drained arable catchments due to the consistency of flow  
532 pathways and respective source availability (poorly-drained grassland catchment –  
533 channels, moderately-drained arable catchment – hillslopes);
- 534 • Contributions from roads sources were higher (total 24%) where road-stream  
535 intersections were more frequent. This effect was seemingly diminished where a runoff  
536 ditch broke this direct connectivity; catchments with ditches at the roadside had lower

537 estimated road sediment contributions (4 and 9%) suggesting that when designed and  
538 managed correctly they are a useful management tool to encourage sediment  
539 deposition;

540 In terms of wider application, successful sediment mitigation measures must consider hillslope  
541 and riparian areas in intensive agricultural catchments and consider the likely impact of  
542 hydrological or land use modifications on downstream sediment loss risk. Emphasis on certain  
543 measures according to setting and process is likely to reduce the burden of mitigation required  
544 but also likely to target the most vulnerable sediment source areas. Targeted considerations  
545 will be necessary to decrease the environmental impacts of intensive agriculture from sediment  
546 loss to meet water quality objectives and also offset the likely increases of these impacts as  
547 agricultural management is further intensified.

548

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798 Tables

799 Table 1. Sub-catchment areas and sample collection dates.

Catchment/ Sample location	Area (km <sup>2</sup> )	Sampling periods													
		T1	T2	T3	T4	T5	T6	T7	T8	T9	T10	T11	T12	T13	T14
Poorly-drained grass															
PD1	0.75														
PD2	1.76					19-21/02/2013									
PD3	2.64	23/07/2012					04/04/2013								
PD4	7.76		24/09/2012					13/05/2013							
PD5	9.81			11/12/2012					24/06/2013						
PD6	11.88				24/01/2013					26/09/2013					22/05/2014
Well-drained arable															
WD1	2.07														
WD2	3.30	11/07/2012													
WD3	4.10		01/10/2012												
WD4	4.23			09/01/2013											
WD5	6.36				12/02/2013										
WD6	11.16					05/04/2013									
Moderately-drained arable															
MD1	1.16														
MD2	2.08	27/07/2012													
MD3	3.10		10/10/2012												
MD4	4.13			11/12/2012											
MD5	6.81				15/01/2013										
MD6	9.48					07/02/2013									

800 PD: suspended sediment sample locations in the poorly-drained grass catchment, WD: suspended  
 801 sediment sample locations in the well-drained arable catchment, MD: moderately-drained arable  
 802 suspended sediment locations in the moderately-drained arable catchment.

803 Table 2. Summary of tracer source data in the poorly-drained grass catchment.

Tracer	Channel banks & ditches		Field topsoils		Road verges & tracks	
	Mean	Std Dev	Mean	Std Dev	Mean	Std Dev
$\chi_{LF}$ ( $10^{-6}m^3kg^{-1}$ ) <sup>1)</sup>	1.46 <sup>b</sup>	0.40	1.92 <sup>b</sup>	2.55	6.43 <sup>a</sup>	3.05
$\chi_{HF}$ ( $10^{-6}m^3kg^{-1}$ ) <sup>1)</sup>	1.43 <sup>b</sup>	0.40	1.78 <sup>b</sup>	2.27	6.24 <sup>a</sup>	2.97
$\chi_{FD}$ (%)	2.07 <sup>b</sup>	1.46	4.43 <sup>a</sup>	2.24	5.10 <sup>ab</sup>	8.05
$\chi_{ARM}$ ( $10^{-7}Am^3kg^{-1}$ )	0.06 <sup>b</sup>	0.03	0.12 <sup>b</sup>	0.18	0.22 <sup>a</sup>	0.12
SIRM ( $10^{-5}Am^3kg^{-1}$ )	248.76 <sup>b</sup>	122.81	168.25 <sup>c</sup>	152.83	1210.55 <sup>a</sup>	717.62
bIRM <sub>soft</sub> ( $10^{-5}Am^3kg^{-1}$ )	102.34 <sup>b</sup>	46.90	103.83 <sup>b</sup>	116.67	685.86 <sup>a</sup>	373.48
bIRM <sub>hard</sub> ( $10^{-5}Am^3kg^{-1}$ )	23.38 <sup>a</sup>	6.15	13.95 <sup>b</sup>	4.33	24.54 <sup>a</sup>	13.48
SIRM/ $\chi_{LF}$	139.73 <sup>b</sup>	33.70	80.02 <sup>c</sup>	21.96	182.11 <sup>a</sup>	61.92
SIRM/ $\chi_{ARM}$	3882.79 <sup>b</sup>	2545.56	1577.60 <sup>c</sup>	814.41	5805.23 <sup>a</sup>	3286.54
$\chi_{ARM}/\chi_{LF}$ <sup>†</sup>	0.04	0.01	0.04	0.02	0.03	0.01
H-ratio ( $10^{-5}Am^3kg^{-1}$ )	225.38 <sup>b</sup>	118.11	154.31 <sup>c</sup>	149.03	1186.01 <sup>a</sup>	713.57
Cd* (mg kg <sup>-1</sup> )	0.20	0.11	0.08	0.04	0.18	0.14
Co (mg kg <sup>-1</sup> )	13.45 <sup>a</sup>	2.82	6.17 <sup>b</sup>	1.16	11.85 <sup>a</sup>	3.16
Cr (mg kg <sup>-1</sup> )	23.75 <sup>a</sup>	8.38	17.65 <sup>b</sup>	2.74	22.54 <sup>a</sup>	7.96
Cu (mg kg <sup>-1</sup> )	20.11 <sup>b</sup>	5.96	12.30 <sup>c</sup>	3.89	28.89 <sup>a</sup>	7.36
Mn* (mg kg <sup>-1</sup> )	1233.14	647.86	511.19	211.98	1285.03	464.69
Ni (mg kg <sup>-1</sup> )	24.31 <sup>a</sup>	5.24	12.73 <sup>b</sup>	2.37	22.02 <sup>a</sup>	6.36
Pb (mg kg <sup>-1</sup> )	15.70 <sup>b</sup>	2.91	17.75 <sup>b</sup>	3.74	44.63 <sup>a</sup>	59.61
Zn (mg kg <sup>-1</sup> )	62.57 <sup>b</sup>	12.52	41.35 <sup>c</sup>	6.71	122.07 <sup>a</sup>	41.48
<sup>234</sup> Th (Bq kg <sup>-1</sup> )	36.94 <sup>a</sup>	14.80	21.65 <sup>b</sup>	5.74	29.75 <sup>a</sup>	10.21
<sup>235</sup> U (Bq kg <sup>-1</sup> )	47.11 <sup>a</sup>	16.00	29.68 <sup>b</sup>	11.38	50.97 <sup>a</sup>	20.55
<sup>228</sup> Ac (Bq kg <sup>-1</sup> )	27.22 <sup>a</sup>	7.41	17.02 <sup>b</sup>	4.74	26.55 <sup>a</sup>	8.72
<sup>137</sup> Cs (Bq kg <sup>-1</sup> )	0.90 <sup>b</sup>	1.60	5.84 <sup>a</sup>	2.45	8.96 <sup>a</sup>	8.05
<sup>40</sup> K (Bq kg <sup>-1</sup> )	804.67 <sup>a</sup>	13.76	634.55 <sup>b</sup>	89.85	860.75 <sup>a</sup>	140.24
<sup>210</sup> Pb <sub>unSUPP.</sub> (Bq kg <sup>-1</sup> )	10.72 <sup>b</sup>	7.59	10.08 <sup>b</sup>	10.79	60.97 <sup>a</sup>	31.88

804 Tracers removed from analysis: <sup>†</sup>tracer failed Kruskal-Wallis test (p>0.05) showing no statistically  
805 significant differences in tracer values between source groups; statistically significant differences  
806 between source tracer groups (as determined by pairwise Dunn-Bonferroni testing (adjusted p < 0.05)  
807 are denoted by different letters). \* frequent non-conservativeness displayed in river sediment samples.



808 Table 3. Summary of source tracer data in the well-drained arable catchment.

Tracer	Channel banks & ditches		Field topsoils		Road verges & tracks	
	Mean	Std Dev	Mean	Std Dev	Mean	Std Dev
$\chi_{LF}$ ( $10^{-6}\text{m}^3\text{kg}^{-1}$ )	1.81 <sup>b</sup>	1.21	13.33 <sup>a</sup>	8.69	12.51 <sup>a</sup>	2.15
$\chi_{HF}$ ( $10^{-6}\text{m}^3\text{kg}^{-1}$ )	1.73 <sup>b</sup>	1.06	11.87 <sup>a</sup>	7.70	11.70 <sup>a</sup>	2.10
% $\chi_{FD}$ (%)	2.28 <sup>b</sup>	1.84	7.31 <sup>a</sup>	0.96	6.39 <sup>a</sup>	1.66
$\chi_{ARM}$ ( $10^{-7}\text{Am}^3\text{kg}^{-1}$ )	0.08 <sup>b</sup>	0.07	0.86 <sup>a</sup>	0.54	0.59 <sup>a</sup>	0.14
SIRM ( $10^{-5}\text{Am}^3\text{kg}^{-1}$ )	121.15 <sup>b</sup>	57.29	700.69 <sup>a</sup>	484.98	1257.31 <sup>a</sup>	410.67
IRM <sub>soft</sub> ( $10^{-5}\text{Am}^3\text{kg}^{-1}$ )	77.53 <sup>b</sup>	45.88	579.05 <sup>a</sup>	420.24	865.06 <sup>a</sup>	242.54
IRM <sub>hard</sub> ( $10^{-5}\text{Am}^3\text{kg}^{-1}$ )	13.56 <sup>b</sup>	18.35	12.85 <sup>ab</sup>	6.03	17.04 <sup>a</sup>	5.51
SIRM/ $\chi_{LF}$	58.10 <sup>a</sup>	15.51	36.40 <sup>b</sup>	6.94	99.03 <sup>a</sup>	30.20
SIRM/ $\chi_{ARM}$	1532.81 <sup>a</sup>	561.87	545.66 <sup>ab</sup>	86.74	2334.86 <sup>a</sup>	1083.26
$\chi_{ARM}/\chi_{LF}$	0.03 <sup>b</sup>	0.01	0.05 <sup>a</sup>	0.01	0.05	0.01
H-ratio ( $10^{-5}\text{Am}^3\text{kg}^{-1}$ )	107.59 <sup>b</sup>	61.23	687.84 <sup>a</sup>	480.56	1240.27 <sup>a</sup>	406.31
Cd ( $\text{mg kg}^{-1}$ )	0.14 <sup>b</sup>	0.08	0.14 <sup>b</sup>	0.07	0.50 <sup>ab</sup>	0.43
Co ( $\text{mg kg}^{-1}$ )	16.29 <sup>a</sup>	6.23	9.47 <sup>b</sup>	1.40	12.68 <sup>a</sup>	1.30
Cr ( $\text{mg kg}^{-1}$ )	30.78 <sup>a</sup>	5.34	19.27 <sup>b</sup>	2.88	25.89 <sup>a</sup>	5.69
Cu ( $\text{mg kg}^{-1}$ )	24.36 <sup>a</sup>	7.49	14.75 <sup>b</sup>	2.93	32.13 <sup>a</sup>	14.38
Mn ( $\text{mg kg}^{-1}$ )	969.36 <sup>b</sup>	263.5	1140.24 <sup>ab</sup>	247.92	1344.56 <sup>a</sup>	224.16
Ni ( $\text{mg kg}^{-1}$ )	31.72 <sup>a</sup>	6.85	15.55 <sup>b</sup>	2.70	24.20 <sup>a</sup>	3.17
Pb ( $\text{mg kg}^{-1}$ )	20.75 <sup>b</sup>	6.60	17.50 <sup>b</sup>	2.58	32.91 <sup>a</sup>	12.31
Zn ( $\text{mg kg}^{-1}$ )	76.62 <sup>a</sup>	19.79	58.45 <sup>b</sup>	8.66	132.66 <sup>a</sup>	51.62
$^{234}\text{Th}^\dagger$ ( $\text{Bq kg}^{-1}$ )	212.77	60.25	147.07	39.23	173.82	53.14
$^{235}\text{U}^\dagger$ ( $\text{Bq kg}^{-1}$ )	126.63	43.46	102.55	29.92	99.89	47.52
$^{228}\text{Ac}$ ( $\text{Bq kg}^{-1}$ )	141.91 <sup>a</sup>	45.13	111.74 <sup>b</sup>	32.02	135.44 <sup>ab</sup>	63.69
$^{137}\text{Cs}$ ( $\text{Bq kg}^{-1}$ )	19.42 <sup>b</sup>	18.37	92.55 <sup>a</sup>	40.23	126.90 <sup>a</sup>	43.40
$^{40}\text{K}$ ( $\text{Bq kg}^{-1}$ )	948.11 <sup>a</sup>	157.55	662.97 <sup>b</sup>	102.46	849.42 <sup>a</sup>	111.10
$^{210}\text{Pb}_{\text{unsupp.}}$ ( $\text{Bq kg}^{-1}$ )	0 <sup>c</sup>	0	11.41 <sup>b</sup>	4.83	32.69 <sup>a</sup>	26.31

809 Tracers removed from analysis: <sup>†</sup>tracer failed Kruskal-Wallis test ( $p>0.05$ ) showing no statistically  
810 significant differences in tracer values between source groups; statistically significant differences  
811 between source tracer groups (as determined by pairwise Dunn-Bonferroni testing (adjusted  $p < 0.05$ )  
812 are denoted by different letters).

813 Table 4. Summary of source tracer data in the moderately-drained arable catchment.

Tracer (units defined in text)	Channel banks & ditches		Field topsoils		Road verges & tracks	
	Mean	Std Dev	Mean	Std Dev	Mean	Std Dev
$\chi_{LF}$ ( $10^{-6}\text{m}^3\text{kg}^{-1}$ )	2.55 <sup>a</sup>	1.62	1.56 <sup>b</sup>	0.67	3.76 <sup>a</sup>	1.49
$\chi_{HF}$ ( $10^{-6}\text{m}^3\text{kg}^{-1}$ )	2.45 <sup>a</sup>	1.46	1.48 <sup>b</sup>	0.61	3.66 <sup>a</sup>	1.48
% $\chi_{FD}$ (%)	2.56 <sup>b</sup>	1.66	3.74 <sup>a</sup>	1.44	2.90 <sup>ab</sup>	0.95
$\chi_{ARM}^\dagger$ ( $10^{-5}\text{Am}^3\text{kg}^{-1}$ )	0.13	0.05	0.13	0.07	0.16	0.04
SIRM ( $10^{-5}\text{Am}^3\text{kg}^{-1}$ )	339.21 <sup>a</sup>	133.33	177.31 <sup>b</sup>	80.41	510.37 <sup>a</sup>	229.26
IRM <sub>soft</sub> ( $10^{-5}\text{Am}^3\text{kg}^{-1}$ )	216.48 <sup>a</sup>	96.64	111.69 <sup>b</sup>	54.78	323.92 <sup>a</sup>	142.46
IRM <sub>hard</sub> ( $10^{-5}\text{Am}^3\text{kg}^{-1}$ )	13.38 <sup>a</sup>	4.10	10.70 <sup>a</sup>	2.95	14.21 <sup>a</sup>	5.22
SIRM/ $\chi_{LF}$	144.99 <sup>a</sup>	42.78	92.14 <sup>b</sup>	15.30	123.66 <sup>a</sup>	21.12
SIRM/ $\chi_{ARM}^*$	2569.42	752.73	1100.94	211.93	2880.03	997.97
$\chi_{ARM}/\chi_{LF}$	0.06 <sup>b</sup>	0.01	0.07 <sup>a</sup>	0.01	0.04 <sup>b</sup>	0.01
H-ratio ( $10^{-5}\text{Am}^3\text{kg}^{-1}$ )	325.83 <sup>a</sup>	130.99	166.61 <sup>b</sup>	78.22	496.16 <sup>a</sup>	225.38
Cd* (mg kg <sup>-1</sup> )	0.50	0.12	0.40	0.09	0.44	0.08
Co (mg kg <sup>-1</sup> )	9.96 <sup>a</sup>	1.21	7.42 <sup>b</sup>	1.11	10.49 <sup>a</sup>	0.77
Cr* (mg kg <sup>-1</sup> )	26.88	5.04	22.50	1.90	27.54	3.27
Cu (mg kg <sup>-1</sup> )	20.61 <sup>b</sup>	2.98	19.57 <sup>b</sup>	3.60	31.42 <sup>a</sup>	6.66
Mn* (mg kg <sup>-1</sup> )	787.22	290.03	486.47	116.21	674.62	87.45
Ni (mg kg <sup>-1</sup> )	33.05 <sup>a</sup>	5.60	23.58 <sup>b</sup>	3.15	34.91 <sup>a</sup>	2.55
Pb (mg kg <sup>-1</sup> )	35.10 <sup>ab</sup>	5.08	30.68 <sup>b</sup>	6.21	37.61 <sup>a</sup>	9.17
Zn (mg kg <sup>-1</sup> )	69.60 <sup>b</sup>	12.66	61.40 <sup>b</sup>	10.98	111.54 <sup>a</sup>	29.39
<sup>234</sup> Th <sup>†</sup> (Bq kg <sup>-1</sup> )	30.04	9.09	32.67	10.21	27.11	8.00
<sup>235</sup> U <sup>†</sup> (Bq kg <sup>-1</sup> )	36.51	6.92	42.81	10.15	43.77	10.78
<sup>228</sup> Ac <sup>†</sup> (Bq kg <sup>-1</sup> )	26.46	6.54	23.77	5.01	25.30	4.42
<sup>137</sup> Cs <sup>†</sup> (Bq kg <sup>-1</sup> )	11.54	12.33	8.62	2.67	9.50	4.89
<sup>40</sup> K (Bq kg <sup>-1</sup> )	922.79 <sup>a</sup>	133.77	731.64 <sup>b</sup>	100.58	886.23 <sup>a</sup>	70.27
<sup>210</sup> Pb <sub>unSUPP.</sub> (Bq kg <sup>-1</sup> )	12.08 <sup>b</sup>	6.30	12.46 <sup>ab</sup>	6.32	22.58 <sup>a</sup>	13.78

814 Tracers removed from analysis: <sup>†</sup>tracer failed Kruskal-Wallis test (p>0.05) showing no statistically  
815 significant differences in tracer values between source groups; statistically significant differences  
816 between source tracer groups (as determined by pairwise Dunn-Bonferroni testing (adjusted p < 0.05)  
817 are denoted by different letters). \* frequent non-conservativeness displayed in river sediment samples.

818

819

820 Table 5. Source group discrimination (expressed as percent of original grouped cases correctly  
 821 classified) of tracer arrays for target suspended sediment samples in the study catchments.

822

<b>Catchment</b>	<b>% of original grouped cases correctly classified</b>	<b>Number of suspended sediment samples</b>
Poorly-drained grassland	89.5	1
	91.9	2
	93.0	2
	94.2	1
	95.3	12
	96.5	6
	97.7	39
Well-drained arable	90.9	4
	93.2	4
	95.5	8
	97.7	46
Moderately-drained grassland	84.2	1
	89.5	2
	91.2	2
	93.0	4
	94.7	9
	96.5	17
	98.2	47

823

824 Figure captions

825 Figure 1. Study catchment locations, outlet coordinates and source and stream sediment sampling  
826 locations in the poorly-drained grassland (PD), well-drained arable (WD) and moderately-drained  
827 arable catchments (MD).

828

829 Figure 2. Canonical Discrimination Functions (output from Multiple Discriminant Analysis) of the full  
830 source sample tracer datasets collected from the, a) poorly-drained grassland, b) well-drained arable,  
831 and c) moderately-drained arable catchments categorised by source group.

832

833 Figure 3. Load specific un-mixing of median source predictions of outlet suspended sediment samples  
834 in the, a) poorly-drained grass, b) well-drained arable and c) moderately-drained arable catchments over  
835 the study period.

836

837 Figure 4. Median source predictions at sub-catchment and outlet locations in the three study catchments.  
838 Site names are denoted by catchment abbreviation: PD – poorly-drained grass, WD – well-drained  
839 arable and MD – moderately-drained arable with increasing numbers representing sites with increasing  
840 sub-catchment areas. Time period, T, represents the period of deployment of time-integrated suspended  
841 sediment samplers between May 2012 and May 2014 (see table 1). Grey squares indicate no un-mixed  
842 data.

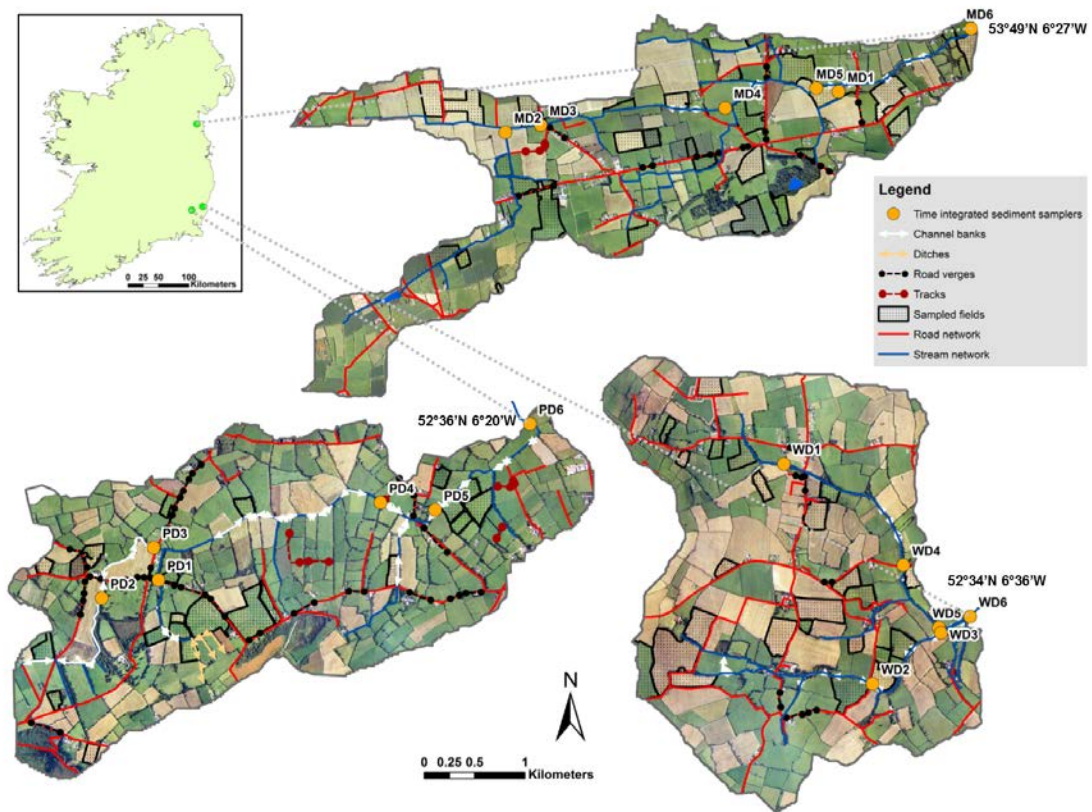
843

844 Figure 5. Combined 95-percentile uncertainty envelopes for sediment source predictions in three study  
845 catchments. Site names are denoted by catchment abbreviation: PD – poorly-drained grass, WD – well-  
846 drained arable and MD – moderately-drained arable with increasing numbers representing sites with  
847 increasing sub-catchment areas. Time period, T, represents the period of deployment of time-integrated

848 suspended sediment samplers between May 2012 and May 2014 (see table 1). Grey squares indicate no  
849 un-mixed data.

850 Figures

851 Figure 1



852

853

