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Preface-Addressing challenges to advance sediment fingerprinting research

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1 SPECIAL ISSUE: ADVANCES IN SEDIMENT FINGERPRINTING

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3 **Preface — Addressing challenges to advance sediment fingerprinting research**

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32 **1 Background**

33 Scientists and managers require information on the patterns and processes
34 associated with fine sediment and related contaminant dynamics in river catchments. A
35 fundamental part of this understanding of contemporary river systems relates to the
36 sources of fine sediment mobilised, transported and temporarily stored in river catchments.
37 Such information is particularly sought after because it has considerable value for the
38 targeting of management resources to reduce excess fine sediment supply and its very
39 significant impacts on water resources and aquatic ecosystems (Owens et al. 2005; Bilotta
40 and Brazier 2008). Sediment source information is also needed to complement other
41 measurement or modelling techniques used in catchment-scale studies of fine sediment and
42 associated contaminant redistribution (Evrard et al. 2011; Smith et al. 2011).

43 Fine sediment source fingerprinting procedures have significant potential to address
44 these requirements. Referred to henceforth as 'sediment fingerprinting', this technique
45 involves the discrimination of sediment sources and apportionment of contributions from
46 those sources to fine-grained sediment (typically <63 μm) transported within river
47 catchments. The approach requires the selection of physical and chemical tracer properties
48 that discriminate source materials combined with the use of statistical procedures to un-mix
49 the unknown contributions from these sources to the mixture of sediment delivered
50 downstream. Catchment sources widely considered by sediment fingerprinting studies
51 include agricultural land uses, geological zones and subsoil sources (e.g. channel banks,
52 gullies), as well as specific features such as road verges, urban surfaces and farm tracks. To
53 discriminate these sources, a diverse range of tracer properties may be employed,
54 comprising geochemical, radionuclide, mineral magnetic, stable isotopes, organic
55 compounds, and colour properties (Foster and Lees 2000; Guzman et al. 2013).

56 Sediment fingerprinting originated from studies into the use of mineral magnetic and
57 geochemical properties as source tracers in the 1970s and 1980s and has since expanded
58 rapidly (see the review by Walling 2013). Some studies focused on the use of pre-selected
59 tracer properties that were known to discriminate targeted sources, such as fallout
60 radionuclides (Wallbrink and Murray 1993). In contrast, multi-tracer sediment fingerprinting
61 studies rely on statistical selection of a subset of tracer properties that discriminate sources.
62 Approaches to address uncertainty in predicted source contributions based on Monte Carlo

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63 sampling methods were also developed (Franks and Rowan 2000) and became an important
64 part of the sediment fingerprinting procedure. Early fingerprinting studies focused on land
65 use and channel bank sources in agricultural catchments (Peart and Walling 1986; Walling
66 1993). The subsequent growth of sediment fingerprinting research extended applications
67 across a range of catchment sizes, sediment sources and environments, including urban and
68 forest settings. However, agricultural catchments remain the dominant environment for
69 sediment fingerprinting studies, reflecting the demand from management agencies for
70 information to support measures for reducing sediment pollution from agriculture (Gellis
71 and Walling 2011).

72 Against this background, this special issue of the Journal of Soils and Sediments on
73 ‘Advances in Sediment Fingerprinting’ was conceived to bring together a set of publications
74 at the forefront of recent developments across this field of research. The papers were
75 largely drawn from the session HS9.7 ‘Revisiting techniques for quantifying sources and
76 travel times of fine sediment from catchment to coast’ which was held during the European
77 Geosciences Union (EGU) General Assembly in 2014. This followed a similar previous session
78 held during the EGU Assembly in 2013. The impetus for the organisation of these sessions
79 originated from discussions between the authors of this preface and others concerning
80 challenges and progress in the development of sediment fingerprinting techniques.

81

82 **2 Challenges**

83 Over the last 40 years, very considerable progress has been made in the
84 development of sediment fingerprinting techniques. However, in the previous two years,
85 there has been a move to challenge and test underlying assumptions of the fingerprinting
86 technique along with aspects of data processing and the treatment of uncertainty. This has
87 been in response to studies that present sediment fingerprinting as a highly transferable
88 technique that delivers accurate and precise estimates of source contributions across a
89 range of environments. In contrast, recent findings suggest that such a view of the present
90 state of sediment fingerprinting as a fully functioning scientific and management tool is
91 misplaced. Here, we briefly review contributions from this special issue and the wider
92 literature that address these recent challenges.

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94 **2.1 Source and sediment sampling**

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95 Combining independent sources of information may have particular value for the
1 2 96 design of source and sediment sampling protocols used by sediment fingerprinting studies.
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4 97 For example, Wilkinson et al. (2015, this issue) employed modelling of soil erosion across a
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6 98 large catchment to guide stratified sampling of surface soils to reflect the disproportionate
7
8 99 effect that highly eroding areas will exert on the concentrations of tracers used to
9
10 100 characterise surface sources. Wethered et al. (2015, this issue) demonstrated the value of
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12 101 coupling sediment source tracing with geomorphic analysis based on various channel
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14 102 metrics to understand downstream changes in sediment contributions from topsoil and
15
16 103 subsoil sources. The investigation of longitudinal changes in sediment source contributions
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18 104 was also an important part of the study by Palazón et al. (2015, this issue), while Haddadchi
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20 105 et al. (2015, this issue) found that, within the 18 month timescale of their study, proximal
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22 106 sources tended to make a higher contribution to a given river sampling point than more
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24 107 distal sources in a nested catchment study based on geological source areas. The large
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26 108 reported changes in proportional source contributions with distance downstream were
27
28 109 attributed to changes in land use, erosion processes and the effect of sediment storage
29
30 110 opportunities in reducing the proportion of upstream sediments that reached downstream
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32 111 measurement sites. The findings from these various studies show that selection of both the
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34 112 source and sediment sampling locations within catchments can have important implications
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36 113 for the interpretation of results from sediment fingerprinting studies.

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38 115 **2.2 Source discrimination and tracer selection**

40 116 Recent criticisms of sediment fingerprinting draw attention to the use of statistical
41
42 117 procedures to select tracer properties without identifying the basis for source
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44 118 discrimination. Careful consideration of the factors leading to source discrimination,
45
46 119 whether by pedogenic, anthropogenic or other environmental processes, is required to
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48 120 support the selection of tracers used to discriminate sources, in addition to statistical
49
50 121 methods (Koiter et al. 2013a; Smith et al. 2013; Smith and Blake 2014). Effective source
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52 122 discrimination relies on differences between sources exceeding those differences that may
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54 123 result from other landscape factors affecting soil properties (Smith and Blake 2014). Small
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56 124 contrasts between tracer concentrations in different sources were observed to correspond
57
58 125 with larger uncertainty in source apportionment results (Pulley et al. 2015), emphasising the
59
60 126 importance of tracer selection for predicting source contributions with confidence.

127 A major theme of studies in the special issue relates to tracer selection. The standard
1 approach for selecting tracers seeks to minimise the number of tracers for use in source un-
2 128 mixing. Sherriff et al. (2015, this issue) found that uncertainty in source predictions was
3
4 129 actually reduced by increasing rather than minimising the number of tracers used in source
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6 130 un-mixing. Laceby et al. (2015, this issue) used prior geochemical knowledge of geological
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8 131 source areas and statistical processing as a basis for element selection, in contrast to many
9
10 132 previous fingerprinting studies that rely of statistical analysis alone. In some instances, the
11
12 133 two approaches selected different elements which led to divergent source apportionment
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14 134 results. Pulley et al. (2015, this issue) compared predicted source contributions based on
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16 135 data from radionuclide, geochemical and mineral magnetic tracer groupings and found large
17
18 136 differences in source predictions between groups. These studies clearly demonstrate that
19
20 137 the choice of tracers can exert an important effect on predicted source contributions.
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25 140 **2.3 Tracer non-conservative behaviour**

27 141 Non-conservative behaviour of tracer properties during transport and storage can
28
29 142 confound sediment fingerprinting studies and requires attention (Koiter et al. 2013b). The
30
31 143 non-conservative behaviour of even a single tracer property included in source un-mixing
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33 144 may result in large changes to predicted source contributions, as was shown by Sherriff et
34
35 145 al. (2015, this issue) using synthetic data. This effect on source contributions was found to
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37 146 be greatly reduced by increasing the number of tracer properties used for un-mixing.
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39 147 Kraushaar et al. (2015, this issue) adopted an expanded tracer selection procedure for
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41 148 fingerprinting reservoir sediments to reduce the possible effect of non-conservative
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43 149 behaviour. In addition to statistical selection, this approach combined a literature review
44
45 150 with analysis of reservoir water chemistry to identify tracers that may be susceptible to
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47 151 dissolution during transport from the terrestrial to the aquatic environment. Few sediment
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49 152 fingerprinting studies explicitly account for such changes in environmental conditions that
50
51 153 may affect the conservative behaviour of selected tracer properties.

52 154 The sensitivity of tracer concentrations and predicted source contributions to non-
53
54 155 conservative behaviour of particle size and organic matter content has been a focus of
55
56 156 recent work (Koiter et al. 2013a; Smith and Blake 2014). Differences between sources and
57
58 157 downstream sediments may arise from selective transport, the effect of which was captured
59
60 158 quantitatively by Koiter et al. (2015, this issue) using a flume experiment to show that
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159 increasing channel gravel depth led to preferential deposition of larger particles. Haddadchi
160 et al. (2015, this issue) highlighted the important effect that particle size can exert, with
161 predicted source contributions varying between three size fractions analysed. These authors
162 emphasised the significance of identifying the size fraction of most interest as part of
163 sediment fingerprinting studies, the selection of which should be guided by the research or
164 management problem under investigation.

166 **2.4 Mixing models and data treatments**

167 The type and structure of statistical mixing models used in sediment fingerprinting
168 can significantly impact on source apportionment results, a point largely overlooked in the
169 fingerprinting literature until recently. The emergence of Bayesian mixing models in
170 sediment fingerprinting (e.g. Fox and Papanicolaou 2008; D'Haen et al. 2013; Nosrati et al.
171 2014) represents a departure from the widely employed frequentist based approaches,
172 which typically use optimization to minimize the sum of squared residuals to obtain
173 predictions of catchment source contributions (e.g. Collins et al. 1997). The study by Cooper
174 et al. (2014) showed that the choice of mixing model can significantly affect estimates of
175 source contributions, with variations in median contributions of up to 21 % between tested
176 model versions, which included both Bayesian and frequentist model structures. Likewise,
177 Haddadchi et al. (2014) found considerable variation in source apportionment results from
178 various mixing model formulations compared to data from artificial mixtures. Notably, the
179 least accurate model tested by these authors was that which applied weightings to tracer
180 data.

181 Various correction factors and weightings have often been employed in mixing
182 models. Correction factors have been used to account for differences in particle size and
183 organic matter content between sources and target sediments (e.g. Collins et al. 1997).
184 Tracer-specific weightings have also been applied based on relative differences in: (i) the
185 proportion of sources classified correctly using Discriminant Function Analysis; and (ii) the
186 level of within-source variability for individual tracer properties (e.g. Collins et al. 2010).
187 These weightings were intended to ensure tracers with higher discriminatory power or
188 lower with-in source variability exerted a greater influence during the optimisation
189 procedure used to obtain predictions of source contributions.

190 The use of these correction factors and weightings has since been challenged in the
1 literature. Smith and Blake (2014) found that inconsistent relationships between particle
2 191 size, organic matter content and tracer concentrations undermined the basis for the use of
3
4 192 widely applied correction factors. They recommended against the use of organic matter
5
6 193 corrections, while corrections for particle size should be considered on a case-by-case basis.
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8 194 Laceby and Olley (2014) used artificial mixtures of catchment sources to test the effect of
9
10 195 different data treatments on source apportionment. These authors found that the use of
11
12 196 tracer weightings did not improve the accuracy of source apportionment results, and in the
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14 197 case of the weighting based on within-source variability, actually significantly reduced the
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16 198 accuracy of results compared to known artificial mixtures. It follows that a transparent and
17
18 199 data-based justification is required to support any changes or weightings that are applied to
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20 200 tracer datasets used in source un-mixing.
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25 203 **3 Recommendations and future needs**

27 204 The findings from recent research together with the papers published in this special
28
29 205 issue challenge underlying assumptions, data processing and mixing modelling approaches
30
31 206 widely used in sediment fingerprinting research. Combined, this highlights a desire by many
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33 207 researchers to address key methodological aspects of sediment fingerprinting to support a
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35 208 renewal of research progress in this field. Ultimately, for sediment fingerprinting research to
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37 209 advance, we need to open the 'black box' linking catchment source inputs to sediment
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39 210 outputs (Koiter et al. 2013b). This prevailing 'black box' also extends beyond the catchment
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41 211 processes affecting tracer development to encompass a severe lack of transparency in
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43 212 aspects of data presentation, processing and caveats that have been a feature of too many
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45 213 sediment fingerprinting papers. Nevertheless, we consider that there has been considerable
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47 214 research progress in the last two years, as revealed by a growing diversity of questions,
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49 215 approaches and groups actively pursuing sediment fingerprinting research. Such diversity
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51 216 can only be of benefit to the field. Here, we outline recommendations and future needs to
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53 217 support the continued development of sediment fingerprinting.

54 218 Recent work has clearly established the need to combine both environmental and
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56 219 statistical approaches to tracer selection. Statistical selection of tracers should be
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58 220 complemented by: (i) information on the environmental or anthropogenic basis for source
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60 221 discrimination; and (ii) review of literature pertaining to the selected tracers and the
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222 likelihood that they may undergo transformations in the study environment (Koiter et al.
223 2013b; Kraushaar et al. 2015, this issue). Prior to undertaking any statistical treatment of
224 tracer data, the range in source and sediment tracer datasets should be compared to check
225 for evidence of non-conservative behaviour (Martínez-Carreras et al. 2010). This latter point
226 is now a feature of tracer selection in most recent fingerprinting studies.

227 Statistical tracer selection procedures that minimise the number of tracers passed to
228 the mixing model require re-examination. There is growing evidence to suggest that
229 reducing the number of tracers increases the likelihood of errors in source ascription by
230 allowing a single erroneous tracer to exert a greater influence on predicted source
231 contributions (Sherriff et al. 2015, this issue) or through selection of tracers that may
232 strongly discriminate only a subset of sources (Smith and Blake 2014). The latter may lead to
233 mixing model optimisation that favours those tracers that exhibit the largest differences
234 between some but not necessary all sources. Selecting tracers that maximise contrasts in
235 tracer concentrations between all sources is likely to be an important factor in reducing
236 source prediction uncertainty (Pulley et al. 2015).

237 Recent work criticised unjustified corrections to tracer data for organic matter
238 content (Smith and Blake 2014) and the use of tracer weightings that actually reduced
239 model accuracy (Lacey et al. 2015). On this basis, corrections for organic matter and tracer
240 weightings based on discrimination ability or within-source variability should not be used in
241 source un-mixing. In contrast, it has been shown that including elemental correlations
242 improves the accuracy of predicted source contributions compared to artificial mixtures
243 (Lacey et al. 2015). Therefore, future studies should consider element correlations as part
244 of the procedure for sampling individual source tracer distributions to ensure that
245 relationships between individual elements are preserved (Lacey et al. 2015).

246 The choice of mixing model type and structure constitutes a significant future
247 research challenge for sediment fingerprinting. The growth of research investigating the
248 effect of mixing model structure and treatment of uncertainty (cf. Cooper et al. 2014;
249 Nosrati et al. 2014) has opened up a set of choices for source un-mixing, not least of which
250 includes the selection of Bayesian or frequentist mixing model structures. Certainly more
251 work is needed in this area to determine whether there is a preferred approach that can be
252 applied more generally. The use of artificial mixtures would form a useful basis for testing
253 the performance of different model structures and error treatments.

254 There are several other aspects of sediment fingerprinting that warrant further
1 attention. Sediment fingerprinting requires pre-identification of all major sources
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3 contributing to downstream sediment. Evidence of an un-sampled source may be expected
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5 to emerge when comparing source and sediment tracer datasets, however, this cannot be
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7 assumed as the un-sampled source may not be captured by the suite of tracers used.
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9 Greater justification is needed for the pre-selection of sources in future sediment
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11 fingerprinting studies. Another challenge relates to balancing laboratory costs against the
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13 data demands of mixing models, particularly Bayesian models that require larger input
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15 datasets. The collection of greater sample numbers per source should be seen as desirable,
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17 particularly with increasing catchment size and diversity of land use or geology, which are
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19 likely to enhance the heterogeneity of physical and chemical properties of source materials,
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21 thereby hindering source discrimination. The viability of future studies may hinge
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23 increasingly on the need for sampling levels that are commensurate with catchment and
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25 source heterogeneity.
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28 29 269 **4 Concluding remarks**

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31 270 The use of the sediment fingerprinting technique to identify the sources of fine
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33 271 sediment (and associated nutrients and contaminants) in river catchments has grown
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35 272 rapidly in the last 5 to 10 years (Koiter et al. 2013b; Walling 2013). This has resulted in
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37 273 scientists from a variety of disciplines new to this field (e.g. inorganic and organic chemistry,
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39 274 ecology, statistics and mathematics), working alongside those from disciplines that have
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41 275 traditionally tackled catchment sediment problems (e.g. soil science, hydrology,
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43 276 geomorphology), to develop new fingerprinting approaches and operational procedures.
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45 277 Interdisciplinary cooperation is vital to increase the robustness of the technique and
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47 278 confidence in model outputs to support catchment managers and policy makers. This is an
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49 279 exciting time for scientists across disciplines to be engaged in sediment fingerprinting and
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51 280 we hope that the discussion above and the papers in this special issue contribute to this
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53 281 growing research momentum.

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