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1 SEDIMENT FINGERPRINTING IN THE CRITICAL ZONE

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3 **Using spectrophotometry to trace sediment source dynamics in coastal catchments draining the**
4 **main Fukushima radioactive pollution plume (2011–2017)**

5

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8

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21

22 **Abstract**

1
2 23 *Purpose.* Spectrocolorimetric measurements provide a relatively inexpensive, quick and non-
3 24 destructive alternative to the analysis of geochemical and organic matter properties. When analyzed
4 25 on sediments and their potential sources, these colour parameters may provide important information
5 26 on the dominant processes (i.e. erosion) occurring in the Critical Zone. Here, they are used to
6 27 investigate whether eroded sediment is derived from forest (i.e. natural), cultivated (i.e.
7 28 anthropogenic) or subsoil sources in order to assess their potential to monitor the effect of
8 29 decontamination in regions impacted by fallout from the Fukushima Dai-ichi Nuclear Power Plant
9 30 (FDNPP) accident.

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14 31 *Materials and methods.* Fifteen spectrocolorimetric properties (L^* , a^* , b^* , C^* , h , x , y , z , L , a , b , u^* , v^* ,
15 32 u' , v') were measured in potential source ($n=37$) and sediment ($n=400$) samples collected during 13
16 33 campaigns from 2011–2017 after major flood events in two catchments (total surface area of 450 km²)
17 34 draining the main Fukushima Dai-ichi Nuclear Power Plant (FDNPP) radioactive pollution plume.
18 35 Potential sources included topsoil from forest and cultivated sources along with subsoil material
19 36 originating from landslides, channel banks and the decontamination of cultivated areas. The optimum
20 37 set of parameters used in the mixed linear model to calculate the sediment source contributions was
21 38 obtained through the use of a range test, the Kruskal-Wallis H -test, and a linear discriminant analysis.

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25 39 *Results and discussion.* Nine selected colour parameters correctly classified 100% of the source
26 40 samples (i.e., forest, subsoil and cultivated sources). The results illustrate that cultivated landscapes
27 41 were the main source of sediment to these river systems (mean 56%, SD 34%) followed by subsoil
28 42 (mean 26%, SD 16%) and forest sources (mean 21%, SD 24%). However, these contributions varied
29 43 strongly over time, with a peak of subsoil contributions (mean 57%, SD 17%) in Fall 2015, coinciding
30 44 with the occurrence of a typhoon after the remediation works. These results were consistent with
31 45 monitoring studies conducted in the same area that showed the major impact of typhoon Etai in
32 46 September 2015 on sediment and radiocesium fluxes.

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37 47 *Conclusions.* These original results demonstrate that spectrocolorimetric measurements may
38 48 contribute to the routine monitoring of the effectiveness of remediation works in this post-accidental
39 49 context. Owing to the inexpensive, rapid and non-destructive analyses, spectrocolorimetric-based
40 50 tracing methods have significant potential to provide information on the dominant erosion processes
41 51 occurring in the Critical Zone.

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47 53 **Keywords** Colour • FDNPP • Radiocesium • Sediment fingerprinting • Soil erosion • Spectroscopy
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54 **1 Introduction**

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2 55 Obtaining quantitative information on the main sources delivering sediment to river networks is
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4 56 required to improve our knowledge of hydro-sedimentary processes occurring in the Critical Zone and
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7 57 also to implement effective soil erosion management programs (Owens et al. 2005). Information on
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10 58 sediment provenance is necessary to reduce the deleterious effects associated with excessive
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12 59 sediment loads to downstream aquatic ecosystems (Minella et al. 2008, Navas et al. 2014, Le Gall et
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14 60 al. 2017). In particular, it is important to understand the origin of the fine-grained fraction of sediment,
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16 61 which is chemically-active and thus facilitates the downstream transfer of contaminants and nutrients
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19 62 (Lacey et al. 2017). Among these contaminants, fallout radionuclides (i.e. radiocesium) are
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21 63 preferentially bound to fine-grained sediment (He & Walling 1996) and predominantly transferred
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24 64 downstream in particulate form following nuclear accidents (Evrard et al. 2015).

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26 65 Fluvial fingerprinting techniques provide sediment source information, based on the measurement of
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28 66 conservative biogeochemical and/or physical parameters in both source and sediment material
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31 67 (Haddadchi et al. 2013, Walling 2013, Owens et al. 2016). In general, the choice of tracing properties,
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33 68 or fingerprints, is guided by the sources requiring discrimination. Geochemical measurements are
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36 69 often used to discriminate between different soil types or lithologies (Douglas et al. 2003, Lacey et al.
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38 70 2015), whereas organic matter composition may trace different land use sources (Thothong et al. 2011,
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40 71 Garzon-Garcia et al. 2017). Many of these conventional tracing properties often require destructive
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42 72 biogeochemical measurements. Furthermore, the preparation of the samples prior to these analyses
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45 73 may have a significant impact on the source apportionment results. For example, the total or partial
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47 74 dissolution of solid samples before geochemical measurements with Inductively Coupled Plasma-Mass
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50 75 Spectrometry (Dabrin et al. 2014) or the destruction of carbonates before carbon and nitrogen isotope
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52 76 analysis with Isotope-Ratio Mass Spectrometry (Harris et al. 2001) may affect sediment fingerprinting
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54 77 parameters.

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57 78 In contrast, the analyses of physical sediment parameters are generally non-destructive and do not
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59 79 require pre-treatments that may affect source apportionment results. Among these parameters, the

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80 colour of material, corresponding to the radiation reflected by the material components in the visible
81 region of the electromagnetic spectrum (400–700 nm), has long been used to describe soils (Viscarra
82 Rossel et al. 2006) and material transiting river networks (Strunk 1992). In early sediment fingerprinting
83 research, this tracer was used to provide qualitative sediment source information (Grimshaw & Lewin
84 1980). The improvement of spectroscopic techniques then resulted in an increase in the number of
85 studies using this relatively quick and inexpensive technique quantitatively to trace sediment sources
86 (Debret et al. 2011). For example, sediment has been traced with the visible (Pulley & Rowntree 2016),
87 mid-infrared or near-infrared regions of the spectra (Poulenard et al. 2009), or a combination of these
88 regions (Brosinsky et al. 2014). Furthermore, colour parameters have also been combined with
89 conventional sediment source tracers, such as geochemical elements (Tiecher et al. 2015).

90 Different approaches have been developed to include the colour-derived parameters quantitatively in
91 sediment source fingerprinting research. Colour coefficients from the visible spectra have been used
92 in mixed linear models to predict sediment source contributions (Martínez-Carreras et al. 2010a). An
93 alternative consisted in using the raw spectra in the visible range to estimate the concentrations of
94 geochemical properties, which were then used in an optimized mixing model to quantify sediment
95 provenance (Martínez-Carreras et al. 2010b). Other studies used visible spectroscopy to estimate the
96 proportions of minerals, such as hematite and goethite (Tiecher et al. 2015) or determine the type of
97 organic matter found in sediment based on the analysis of the first derivative reflectance spectra
98 (Debret et al. 2011). Researchers have also estimated source contributions based on partial least-
99 square regression (PLSR) models taking information from the entire spectra into account, after
100 calibrating these models with experimental artificial mixtures containing fixed proportions of sources
101 prepared in the laboratory (Poulenard et al. 2012, Evrard et al. 2013).

102 The use of colour-derived properties in sediment fingerprinting research is most effective in areas with
103 drastic colour differences between sources, for example, black marls, white limestones and coloured
104 molasses in the French Alps (Legout et al. 2013). In Japan, a major colour contrast occurs in coastal
105 catchments exposed to radioactive fallout following the Fukushima Dai-ichi Nuclear Power Plant

106 (FDNPP) accident. The main soil types in this region are dark-coloured Cambisols and Andisols, or light-
107 coloured Fluvisols (Fig. 1). Moreover, decontamination works in this region consisted of removing the
108 radiocesium contaminated topsoil (upper 5 cm) and replacing it with a new substrate consisting of a
109 light-coloured crushed granite extracted from local quarries (Yasutaka & Naito 2016).

110 In this post-nuclear accident context, identifying sources supplying the material transiting river
111 systems, and the spatial and temporal dynamics of sediment sources, is fundamental to quantifying
112 the effectiveness of remediation strategies and for defining future priorities for the management of
113 radiocesium-contaminated environments. Previous studies based on the destructive analysis of
114 geochemical and organic matter properties identified cultivated sources (e.g. paddy fields) and
115 Fluvisols as the main sources of sediment (Lepage et al. 2014, Lacey et al. 2016b, Huon et al. 2018).
116 However, the replication of these analyses on a large number of samples is time-consuming and may
117 result in insufficient material for future analyses, in particular for sediment samples already analyzed
118 by multiple destructive techniques. As a significant amount of funds are invested in the management
119 of this post-fallout environment, we require multiple lines of evidence to support initial results
120 regarding the dominant source of radiocesium-contaminated particulate matter in this region.
121 Accordingly, the utility of a rapid, low-cost, non-destructive colour tracing technique, combined with
122 non-destructive radiocesium measurements, was investigated in order to provide further
123 understanding of the main spatial and temporal sediment sources dynamics in two catchments
124 draining the main radioactive contamination plume of the FDNPP accident.

125 126 **2 Materials and methods**

127 **2.1 Study site**

128 This study was conducted in the Niida (275 km²) and Mano (175 km²) catchments (Fig. 2) in the
129 Fukushima Prefecture, Japan. The upper catchment areas are mainly covered with forests, although
130 paddy fields are found in the floodplains along the river channel. These upper catchment areas were
131 heavily contaminated by radiocesium fallout (¹³⁴Cs + ¹³⁷Cs), with initial activities ranging from 20 to 150

132 kBq kg⁻¹. The middle section of these catchments is exclusively forested and characterized by very
1 steep slope gradients (Chartin et al. 2013). Downstream, the coastal plains are densely cultivated and
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4 134 inhabited with initial fallout contamination from radiocesium not exceeding 20 kBq kg⁻¹ (Lepage et al.
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6 135 2014). In the headwaters, the main soil types are Cambisols and Andisols, whereas in the coastal plains,
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8 136 Fluvisols are ubiquitous. The mean annual rainfall in the region, located within a 100 km radius from
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10 137 the FDNPP, is 1420 mm (Lacey et al. 2016a). The frequent occurrence of typhoons or tropical storms
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12 138 were found to drive sediment generation and radiocesium export from upper catchment areas to the
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14 139 Pacific Ocean (Chartin et al. 2017). In the upper catchment areas, the local population was evacuated
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16 140 after the nuclear accident during Spring in 2011, and these zones were only reopened to inhabitants
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18 141 early on April 1, 2017. However, it is estimated that only ~10% of the population that lived in these
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20 142 areas before the accident had returned home by December 2018 (Iitate Village authorities,
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22 143 unpublished data).

28 144 **2.2 Source and sediment sampling**

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30 145 Soil samples ($n=37$) were collected in areas representative of the main potential sediment sources in
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32 146 the region, including forests ($n=12$), cultivated soils ($n=8$), subsoil sources ($n=6$) (i.e. channel bank, mass
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34 147 movements) and decontaminated material ($n=11$). Source sampling was restricted to those areas
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36 148 draining to the uppermost river sections, where erosion processes are the most active. Furthermore,
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38 149 soil erosion remains very limited in the flat coastal plain, and those very steep and forested areas
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40 150 located in the middle section of the catchment are very difficult to access. Samples were collected with
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42 151 a plastic trowel, compositing 10 grab subsamples (top 1–2 cm) into one well-mixed and homogenized
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44 152 sample for analysis. Fine sediment samples ($n=400$) were collected at 20 sites on the Niida River and
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46 153 19 sites on the Mano River during twelve fieldwork campaigns occurring every ~6 months after the
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48 154 main hydro-sedimentary events (e.g. spring snowmelt, and typhoons) that occurred in the region
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50 155 between November 2011 and November 2017. Fine deposited material was scraped (i.e. the top 1 cm
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52 156 layer) using a plastic spatula and each sample comprised of ~10 subsamples collected along a 5 m
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157 reach. All the soil and sediment samples were oven-dried at 40°C for ~48 h, sieved to 2 mm prior and
158 pressed into polyethylene boxes (~15 mL) for analyses.

159 For this research, we explicitly analyzed tracer parameters in the <2 mm particle size fraction. In the
160 Fukushima region, the abundance of ¹³⁷Cs fallout resulted in radiocesium being bound to multiple
161 fractions, including fine and coarse sands. For example, Fan et al. (2014) reported a stronger
162 correlation for radiocesium and coarse grain sediment relative to the fine grain fraction. As the
163 objective of this research is to test the potential of colorimetric parameters to monitor the dynamics
164 of sediment contaminated with radiocesium in this post-accidental context, this current research will
165 focus on the <2mm fraction. This choice is supported by measurements of the specific surface area on
166 a selection of source soils ($n=30$) and sediment ($n=162$) (Lepage et al. 2016). Differences in particle size
167 were found not to be statistically significant (Normality Test (Shapiro Wilk): $p < 0.05$; Mann-Whitney
168 U-test: $U = 2006$, $p = 0.142$).

170 **2.3 Radionuclide measurements**

171 Gamma-emitting radionuclide activities were determined with gamma spectrometry using coaxial
172 HyperPure Germanium detectors (Canberra/Ortec) at the *Laboratoire des Science et de*
173 *l'Environnement*. ¹³⁷Cs activities were measured at the 662 keV emission peak. ¹³⁴Cs activities were
174 calculated as the mean of activities measured at both 604 keV and 795 keV emission peaks. As the
175 initial radionuclide deposits contained similar ¹³⁴Cs and ¹³⁷Cs activity concentrations, with an initial
176 ¹³⁴Cs/¹³⁷Cs activity ratio of ~1, only ¹³⁷Cs results were presented in the current research (Chartin et al.
177 2013). ¹³⁷Cs activities were decay-corrected to March 14, 2011, the date of the main radionuclide
178 fallout deposition on soils of the region (Kinoshita et al. 2011).

180 **2.4 Spectrocolorimetric measurements**

181 Measurements were taken within a 3-mm target radius using a portable diffuse reflectance
182 spectrophotometer (Konica Minolta CM-700d). All samples were measured in the same polyethylene

183 boxes used for gamma spectrometry (volume 15 mL) with a minimum sample quantity of 0.2 g.
184 Although the analysis was systematically made at the same location within the boxes, three
185 measurements were conducted on each sample to take potential heterogeneities into account.
186 Spectral reflectance was obtained with a 10-nm resolution between 360 and 740 nm.
187 All measurements were taken with the D65 standard illuminant, the 10° angle observer and with the
188 specular component excluded. The raw data was comprised of the spectral reflectance percentage for
189 each of the 39-wavelength classes, from which eight components of various colorimetry models were
190 derived (Viscarra Rossel et al. 2006). XYZ tristimulus values were calculated based on the colour-
191 matching functions defined in 1931 by the International Commission on Illumination (CIE 1931) that
192 were then transformed into the CIE L*a*b* and the CIE L*u*v* Cartesian coordinate systems based on
193 the equations provided in CIE (1978). The spectrophotometer was calibrated before each set of
194 measurements by making a zero and a white calibration. In total, 15 colour metric parameters were
195 quantified for the source and sediment samples (L, L*, a, a*, b*, C*, h, x, y, z, b, u*, v*, u^l, and v^l).

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197 **2.5 Analyses and modelling**

198 First, the selection of discriminant colorimetric parameters was performed following three steps: (i)
199 a range test, (ii) the Kruskal–Wallis *H*-test (KW *H*-test), and (iii) a discriminant function analysis (DFA).
200 For the range test, the variables with sediment concentrations lying outside the range of sources ± 1
201 standard deviation were excluded from further analysis, as recommended by Smith and Blake (2014).
202 Then, the KW *H*-test was performed to test the null hypothesis ($p < 0.05$) that the sources are belonging
203 to the same population.

204 A multivariate discriminant function analysis (DFA) was then performed to determine the minimum
205 number of variables that maximized the discrimination between the sources. DFA is based on the
206 Wilks' lambda (Λ^*) value from the analysis of variance, where the criterion used by the statistical model
207 is the minimization of Λ^* . A Λ^* value of 1 is found when all the group means are the same whilst a low
208 Λ^* value means that the variability within the groups is small compared to the total variability. The

209 DFA was performed in the backward mode, with 0.01 as the maximum or minimum significance of F
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7 212 Second, a modified version of the classical Solver-based mixing model determined the relative
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9 213 contribution of different sources to in-stream sediment through simultaneously minimizing the mixing
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11 214 model difference (MMD):
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$$MMD = \sum_{i=1}^n \left(\left(C_i - \left(\sum_{s=1}^m P_s S_{si} \right) \right) / C_i \right)^2 \quad (1)$$

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20 215 where n is the number of parameters in the model chosen by the three-step selection process; C_i is the
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22 216 in-stream sample parameter (i); m is the number of sources; P_s is the contribution of source (s); S_{si} is
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24 217 the mean of parameter (i) in source (s). The proportional contribution from each source (P_s) was
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26 218 modelled by solving Equation 1 with the Solver Function in Microsoft Excel with P_s being between 0
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28 219 and 1 and the sum of all P_s estimations equalling 1. The GRG Non-Linear solving method was used with
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30 220 automatic scaling in Solver, ignoring integer constraints, with a maximum run time of 5000 and
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32 221 allowing for 2500 iterations. A multi-start population size of 2500 was used along with the same
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34 222 random seed for each of the model runs while requiring bounds on the variables. A constraint precision
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36 223 and convergence of 0.000001 were selected.
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43 225 **3 Results**

44 226 **3.1 Comparing colour parameters between sources and target sediments**

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47 227 All 15 colorimetric parameters analyzed systematically plotted within the range of values observed in
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49 228 the three main potential sources of riverine material (Fig. 3). The range test demonstrates that the
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51 229 values of our sediment parameters plot within our source range and thus it is possible to use end-
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53 230 member mixing models to quantify the source contributions to target sediment samples. The fact that
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231 all the sediment samples plot within the source range further infers that there may not be any
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2 232 significant particle size impacts affecting the colorimetric parameters nor missing sources.

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7 234 **3.2 Colorimetric parameters selection for modelling**

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9 235 When considering all three potential sources of sediment, all 15 colorimetric parameters were selected
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11 236 as potentially discriminant by the KW *H*-test (Table 1). Among these parameters, 13 were selected by
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13 237 the backward DFA as the best combination to discriminate between the sources (L^* , a^* , b^* , C^* , h , x , y ,
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15 238 v' , L , a , b , u' , v^* ; Table 2). Although the forest and cultivated source samples were well classified using
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17 239 this approach, only 75 to 88% of the subsoil and decontaminated source samples were attributed to
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19 240 the correct class. Moreover, the uncertainty of classification for these two sources was high (13-26%;
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21 241 Table 2). This illustrates the difficulty in discriminating between these two subsoil sources (i.e.
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23 242 decontaminated or gully/channel bank).

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26 243 In contrast, when grouping these two classes into a single 'subsoil' category, only nine colorimetric
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28 244 parameters were selected by the DFA (L^* , a^* , b^* , v^* , h , a , b , u' , L ; Table 2). When considering three
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30 245 sources only (i.e. cultivated, forested, and subsoil sources), the DFA is able to classify correctly 100%
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32 246 of the samples. Furthermore, the uncertainty of the classification of these sources was lower than
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34 247 0.05% (Table 2). The improvement in discrimination when merging decontaminated or gully/channel
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36 248 bank into one subsoil source is highlighted in Fig. 4.

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39 250 **3.3 Source apportionment modelling**

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41 251 The modelling results demonstrate that cultivated soils were the main source of sediment to these
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43 252 river systems between Fall 2011 (mean 57%) and Spring 2015 (mean 58%; Fig. 5). Thereafter, their
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45 253 contribution decreased sharply in Fall 2015 (mean 33%), before increasing again in 2016–2017 (mean
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47 254 range 58–68%) and exceeding the levels observed between 2011–2015. The contribution of subsoils
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49 255 remained relatively constant during the entire period (mean range 19–25%), except in Spring 2015

256 with a peak of 57%. The supply of sediment originating from forests was higher between 2011–2015
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2 257 (mean range 19–29%) than in 2016–2017 (mean range 11–18%).
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4 258 Despite a clear decrease over the study period (from a mean of 21,920 Bq kg⁻¹ in 2011 vs. a mean of
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6 259 5950 Bq kg⁻¹ in 2017), ¹³⁷Cs activities strongly varied in sediment collected at the different sampling
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8 260 locations, between 30–51,590 Bq kg⁻¹ in the Niida River, and between 880–27,350 Bq kg⁻¹ in the Mano
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10 261 River for the entire study period. A negative correlation ($r = -0.28$) was observed between the subsoil
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12 262 contributions to sediment sampled in the current research in the upper catchment areas and the ¹³⁷Cs
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14 263 activities measured in these deposits (Fig. 6). However, this relationship was associated with a low
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16 264 determination coefficient ($R^2 = 0.08$) illustrating the complexity of the processes occurring in the study
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26 267 **4 Discussion**

28 268 **4.1 Impact of land use and remediation works on sediment sources**

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30 269 Cultivated soils were modelled to be the dominant source of sediments in the Niida and Mano
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32 270 catchments, followed by subsoil and forest sources. Previous studies demonstrated that soil erosion
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34 271 strongly varied depending on the land use in the Fukushima fallout impacted area, with the highest
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36 272 rates of soil loss and ¹³⁷Cs wash-off being measured in farmland compared to the much lower rates
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38 273 observed under grassland and forests (Yoshimura et al. 2014).
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41 274 In the Fukushima region, the top 5 cm of the soil profile contains upwards of 99% of the radiocesium
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43 275 contamination (Kato et al. 2012, Lepage et al. 2015). Accordingly, the remediation of radiocesium
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45 276 contamination involves the clearcutting of vegetation and the removal of the 5-cm topsoil. Initially, a
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47 277 light-coloured crushed granite layer corresponding to the ‘decontaminated soil’ was used to replace
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49 278 the top soil layer prior to this layer being replaced with an organic rich soil. This remediation
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51 279 predominantly occurred on agricultural landscapes (e.g. paddy fields) resulting in the exposure of
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53 280 subsoils and decontaminated soil to the erosive power of rainfall in this region subject to frequent
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55 281 tropical cyclones.
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282 The impact of these remediation works on sediment sources was particularly evident in Fall 2015 after
283 the occurrence of typhoon Etau (Chartin et al. 2017), which was the most intense typhoon observed
284 after decontamination had started late in 2013 on the upper section of the Niida River. This may
285 explain the sharp increase of the subsoil contribution of sediment between Spring 2015 (mean 20%,
286 SD 15%) and Fall 2015 (mean 57%, SD 17%) in both Niida and Mano Rivers as a result of the extensive
287 landslides, the collapse of channel banks and the erosion of subsoil material exposed in
288 decontaminated fields that occurred during this particularly heavy event. However, as cropland
289 remained abandoned in the study area after remediation works have been completed, vegetation
290 regrew quickly and covered these decontaminated soils, which protected them against further erosion.
291 This probably corresponds to subsoil contributions to sediment decreasing again after this period, in
292 both Fall 2016 (mean 22%, SD 17%) and Fall 2017 (mean 20%, SD 14%).
293 This phenomenon likely explains the much lower radiocesium concentrations measured in sediment
294 transiting these rivers in 2015 compared to the 2011–2014 period (Evrard et al. 2016). These results
295 were confirmed by Osawa et al. (2018), who demonstrated that radiocesium levels decreased
296 significantly between 2013–2016 in the Mano and the Hiso Rivers. They also highlight the massive
297 sediment and radiocesium fluxes transported during typhoon Etau, in September 2015 (Chartin et al.
298 2017), which likely explains the very different sediment source contributions calculated in the current
299 research for sediment deposits collected in Fall 2015, after this particularly extreme flooding event.
300 The negative correlation observed between subsoil contributions to sediment and ¹³⁷Cs activities was
301 expected, as material originating from decontaminated zones and other subsoil material were not
302 exposed to the initial radioactive fallout in 2011, and their contribution therefore diluted the
303 radiocesium contamination supplied to the river by cultivated or forest soils (Evrard et al. 2016). The
304 low determination coefficient associated with this relationship may be explained by the spatial
305 heterogeneity of the initial fallout deposition, with higher levels of ¹³⁷Cs observed in upper parts of the
306 Niida River catchment (Fig. 2) compared to those observed in upper parts of the Mano River catchment

307 (Chartin et al. 2013). Furthermore, the local hydro-sedimentary dynamics may also explain the
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2 308 variability in these contamination levels and source contributions at individual sites.

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6 7 310 **4.2 Land use-based source contributions to sediment**

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9 311 The results of the current research may also be compared to those obtained by previous studies
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11 312 quantifying the land use contributions to sediment transiting the Niida and Mano Rivers. Laceby et al.
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13 313 (2016b), using carbon and nitrogen parameters, calculated that cropland provided the most sediment
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15 314 to the Niida River whereas subsoil was the dominant source of material in the Mano River. When
16
17 315 comparing the results obtained for those samples analysed by both Laceby et al. (2016) for the <2mm
18
19 316 fraction of sediment and the current research ($n=34$), similar conclusions may be reached for the Niida
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21 317 River. In this catchment, there were similar dominant contributions of cultivated sources to sediment
22
23 318 between November 2012 and November 2014 (mean 55%, SD 31% in the current research vs. mean
24
25 319 46%, SD 6% in Laceby et al. 2016). In contrast, the dominant proportion of subsoil contributions found
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27 320 by Laceby et al. (2016) in the Mano River (mean 62%, SD 12%) during this period were not
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29 321 systematically confirmed in the current research (mean 22%, SD 12%). The analysis of experimental
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31 322 mixtures prepared in the laboratory and containing known proportions of the individual sources would
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33 323 provide a powerful way of comparing both approaches and investigate whether the colorimetric
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35 324 parameters or the carbon and nitrogen elemental concentrations and stable isotope ratios provide a
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37 325 better approach to trace sediment derived from these different erosion sources.
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47 327 **4.3 Perspectives for the future monitoring of the impact of remediation**

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49 328 Although the evolution of radiocesium levels in rivers draining the main radioactive plume in
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51 329 Northeastern Japan has been monitored at several river stations across the Fukushima Prefecture by
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53 330 the Japanese authorities (Fukushima Prefecture 2019, JAEA 2019) and individual research groups (e.g.
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55 331 Osawa et al. 2018), these hydrological and/or hydro-sedimentary records have not been coupled with
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57 332 sediment fingerprinting results. Accordingly, the identification of the sources supplying suspended
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333 matter transiting these rivers and their spatial and temporal variations could be significantly improved.
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2 334 In this context, the utility of using colour as a discriminant property of sediment sources is that it is
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4 335 easily understandable by local communities and water managers, as colour parameters may act as
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7 336 visual indicators in catchments impacted by the deleterious effects of excessive soil erosion and
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9 337 sedimentation. In the Fukushima Prefecture, changes in sediment colour are associated with a change
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11 338 in the dominant source of material. These changes are easy to identify by the local inhabitants, given
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13 339 the strong colour contrast occurring between the black-coloured sediment originating from forests or
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15 340 paddies and the yellow-coloured material supplied by subsoil (Fig. 7). This tool is therefore appropriate
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17 341 to discuss with the local communities, which are invited to return to this region with the re-opening of
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19 342 the decontaminated areas since April 2017. Local residents could potentially compare the results of
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21 343 this research with their own observations in the field, as it is easy for them to discriminate between,
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23 344 for instance, the dark material originating from forests from the light-coloured sediment supplied by
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25 345 the decontaminated cultivated sources to the rivers. The installation of low-cost colour sensors that
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27 346 may be deployed across a network of monitoring stations in the region could provide more detailed
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29 347 information on the dynamics of contaminated particulate material in this region. Therefore, there is
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31 348 potential to develop a colour based monitoring program to trace the efficacy of decontamination
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33 349 programs in this post nuclear accident context.
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42 351 **4.4 Perspectives for future tracing research**

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44 352 In the future, other spectrophotometric approaches based on the analysis of first derivative values of
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46 353 the reflectance in the visible part of the spectra should be tested (Tiecher et al. 2015). The use of Partial
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48 354 Least Square regression models calibrated and validated based on the analysis of experimental
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50 355 mixtures prepared in the laboratory (Legout et al. 2013) could also be implemented to further test the
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52 356 potential of these techniques based on the analysis of colorimetric properties for sediment tracing.
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54 357 Future research should also combine spectrophotometric methods with geochemical indicators, such
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56 358 as X-Ray Fluorescence devices, as geochemistry was shown to provide discrimination between
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359 Fluvisols found in the local floodplains and Cambisols/Andisols found under forests and cultivated land
360 in these catchments (Lepage et al. 2016). Identifying an additional geochemical property, capable of
361 discriminating between subsoil associated with decontamination works versus subsoil originating from
362 other sources, would provide an unambiguous tracer of remediation and facilitate the development
363 of a more comprehensive approach to understand the efficacy of remediation programs in the
364 Fukushima region.
365 Furthermore, future research should focus on forests that received the majority of radiocesium fallout
366 after the accident. Importantly, there currently are no plans to decontaminate forested landscapes in
367 the fallout impacted region. In contaminated forests, radiocesium was shown to have almost
368 completely (80–95%) migrated in 7 years from the litter layer to the mineral layers of the soil, although
369 differences were observed depending on the forest type (Takahashi et al. 2018). Novel tracing
370 approaches, including those based on environmental DNA (Evrard et al. in review) that may
371 discriminate between different plant species, may be beneficial for understanding the impact of forests
372 on in-stream radiocesium contamination.

376 **5 Conclusions**

377 Spectrocolorimetric methods were demonstrated to have significant potential to investigate the main
378 sources of sediment (e.g. cultivated, forest or subsoils) in Fukushima coastal catchments draining the
379 main radiocesium contamination plume. However, to fully achieve this goal, these methods should be
380 combined with geochemical measurements, to provide an unambiguous tracer of the decontaminated
381 soil contribution to sediment. This approach will provide results that may help guide the design of
382 programs to monitor the efficacy of decontamination in this post-accidental context as quantifying the
383 subsoil contribution provides straightforward information on the radiocesium content of sediment, as
384 subsurface material was sheltered from the initial radioactive fallout. In the future, these techniques

385 could be applied to other types of sediment material types including suspended matter and material
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2 386 accumulated in dam reservoirs in order to check for the consistency of these results for different
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4 387 sample types and compare the current sediment source contributions, including the subsoil
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7 388 proportion, to those reconstructed based on longer term sediment archives. This work is particularly
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9 389 important as the local population has been invited to return to this region and it will likely start to re-
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11 390 cultivate cropland, which will likely further change the sediment source contributions and hence their
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14 391 potential radiocesium content to these rivers.

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29 398 **References**

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31 **Figure captions**

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 33 **Figure 1** – Illustration of the drastic colour differences between potential sediment sources in coastal
 34 548 catchments affected by fallout from the FDNPP-accident including (a) dark-coloured paddy fields on
 35 549 Andisols; (b) light-coloured subsurface material mobilized by landslides; (c) light-coloured crushed
 36 550 granite extracted from local quarries for the decontamination works.
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39 552 **Figure 2** – Location of the investigated catchments in Japan (A) with ¹³⁴Cs/¹³⁷Cs concentrations
 40 553 modelled from Chartin et al. (2013) mapped with elevation (B) along with land use and the location
 41 554 of the sediment and source samples modelled in this current study (C).
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44 555 **Figure 3** – Box-plots of several visible-color parameters measured in the potential sources and in
 45 556 suspended sediment samples. The box indicates the location of the first and third quartiles; the line
 46 557 indicates the median value; whiskers indicate the non-outlier range; circles indicate values outside of
 47 558 the range of $\pm 1SD$.
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50 559 **Figure 4** –Two-dimensional scatterplots of the first and second discriminant functions provided by
 51 560 the stepwise discriminant function analysis (DFA) applied using four (a) and three (b) potential
 52 561 sources of sediment, respectively.
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55 562 **Figure 5** – Mean (and standard deviations) of the source contributions to the sediment transiting
 56 563 both the Mano and Niida Rivers during each fieldwork campaign, from Fall 2011 to Fall 2017,
 57 564 according to the conventional linear un-mixing model.
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565 **Figure 6** – Correlation between the subsoil contributions to sediment calculated in the current
1 566 research and the ¹³⁷Cs activities measured in sediment collected before 2011–2017 in upper sections
2 567 of the Mano and Niida Rivers. All ¹³⁷Cs activities were decay-corrected to March 14, 2011 (main
3 568 fallout period on Japanese land).
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6 569 **Figure 7** – Mean soil colour for the different potential land use sources compared to the
7 570 darkest/mean/lightest colours observed in sediment analysed in the current research.
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561 **Figure captions**

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3 563 catchments affected by fallout from the FDNPP-accident including (a) dark-coloured paddy fields on
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5 565 granite extracted from local quarries for the decontamination works.
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8 566 **Figure 2** – Location of the investigated catchments in Japan (A) with ^{134}Cs + ^{137}Cs concentrations
9 567 modelled from Chartin et al. (2013) mapped with elevation (B) along with land use and the location
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25 577 both the Mano and Niida Rivers during each fieldwork campaign, from Fall 2011 to Fall 2017,
26 578 according to the conventional linear un-mixing model.
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30 580 research and the ^{137}Cs activities measured in sediment collected before 2011–2017 in upper sections
31 581 of the Mano and Niida Rivers. All ^{137}Cs activities were decay-corrected to March 14, 2011 (main
32 582 fallout period on Japanese land).
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(a)



(b)



(c)

Figure 2

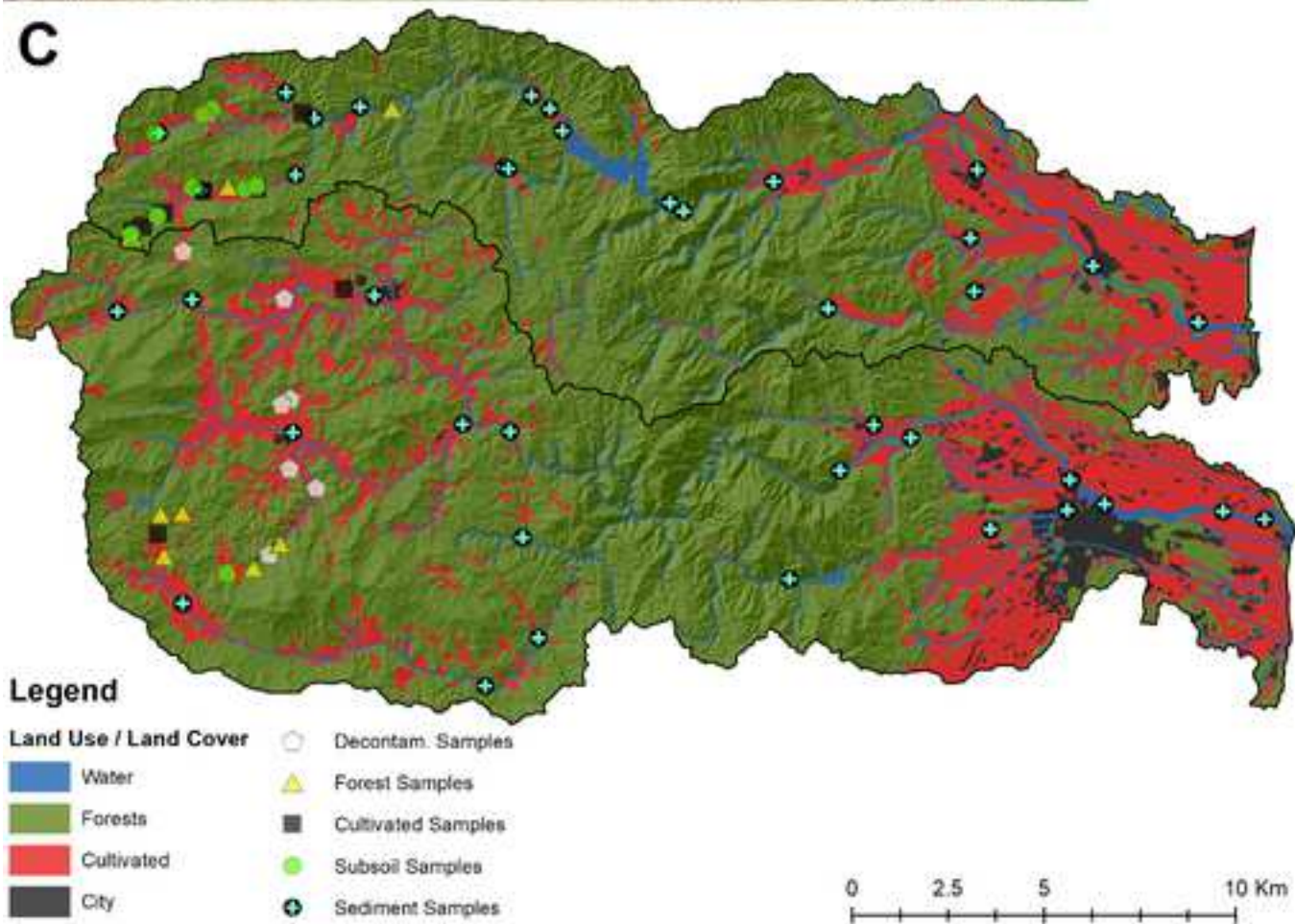
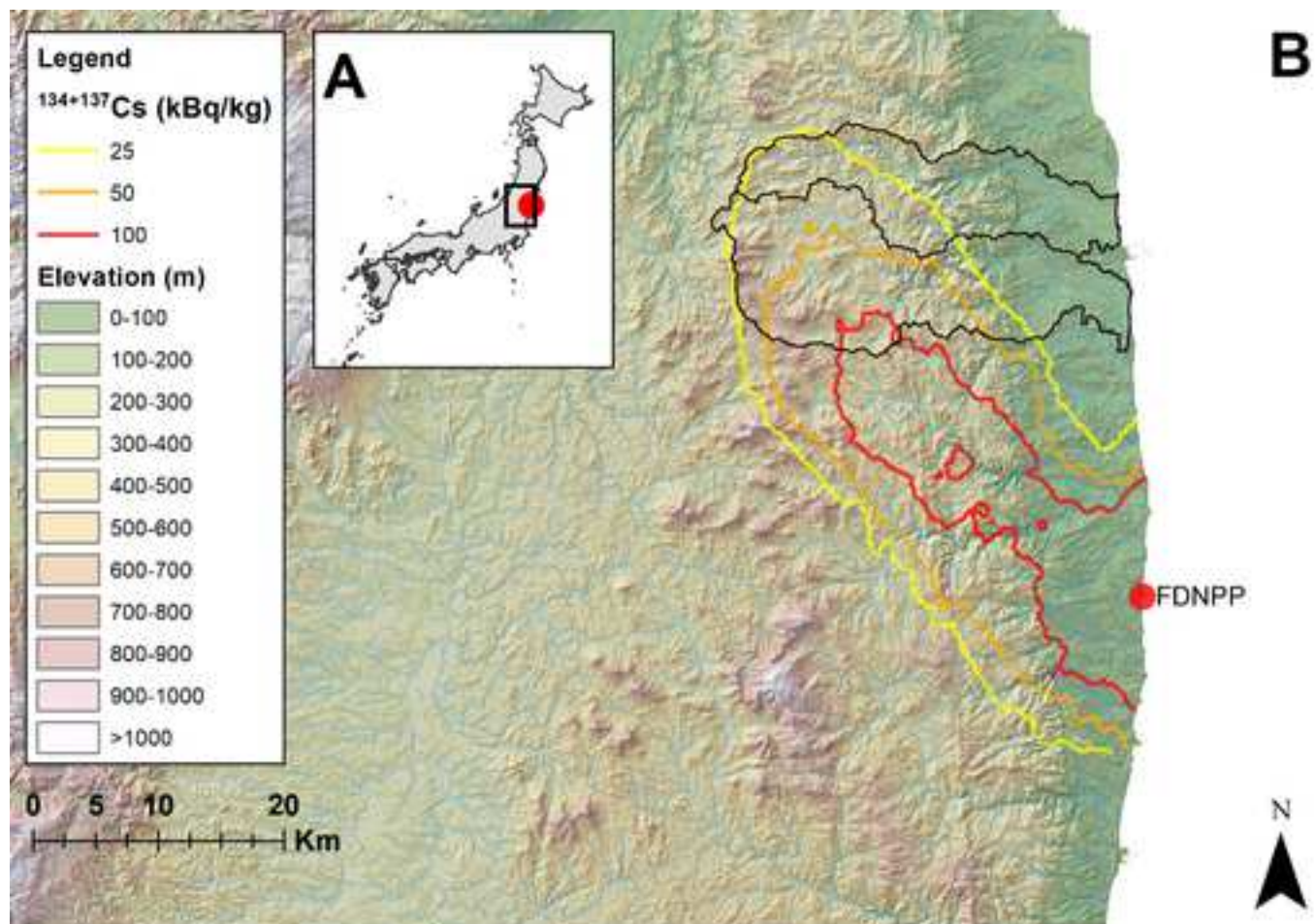


Figure 3

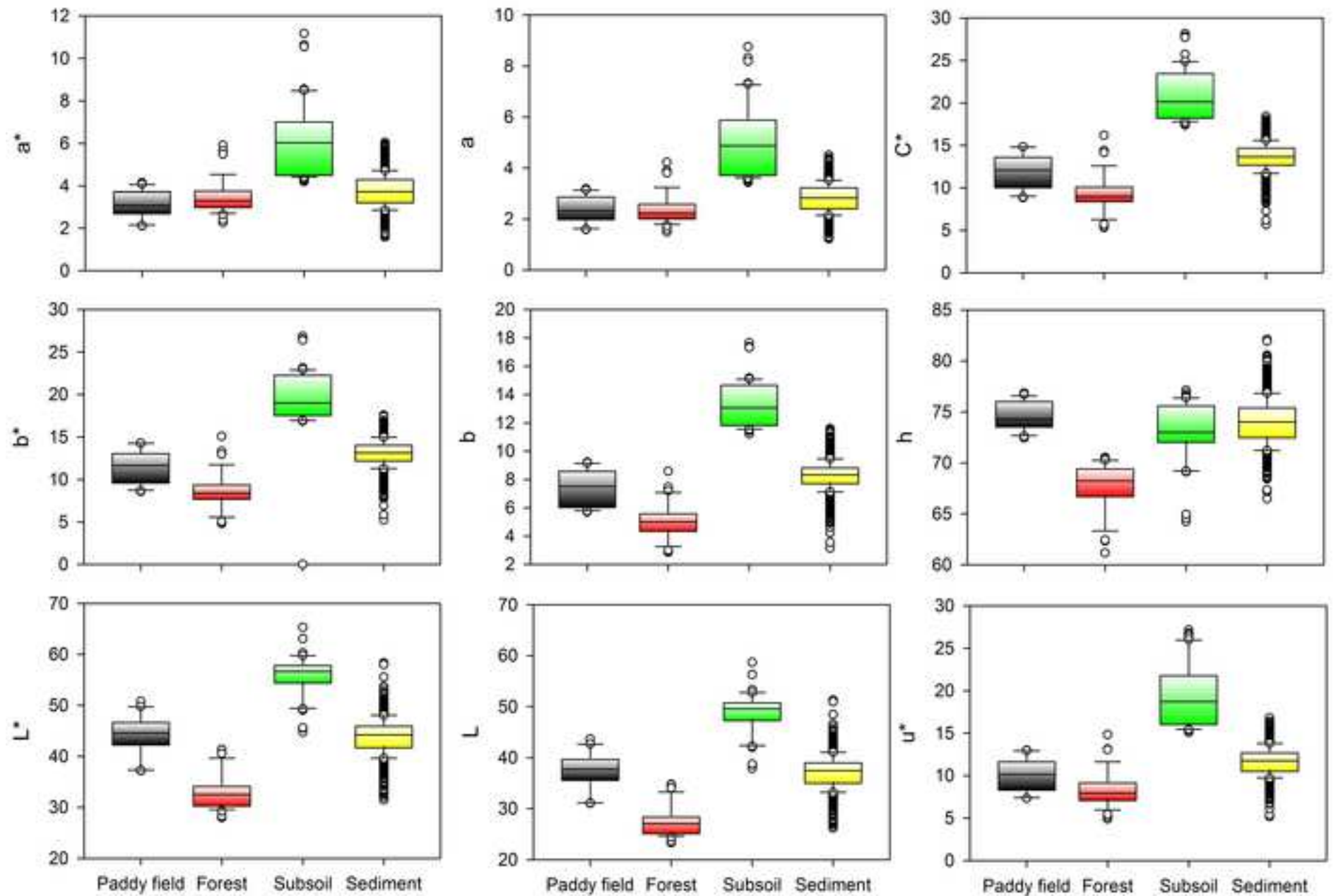


Figure 3 continued

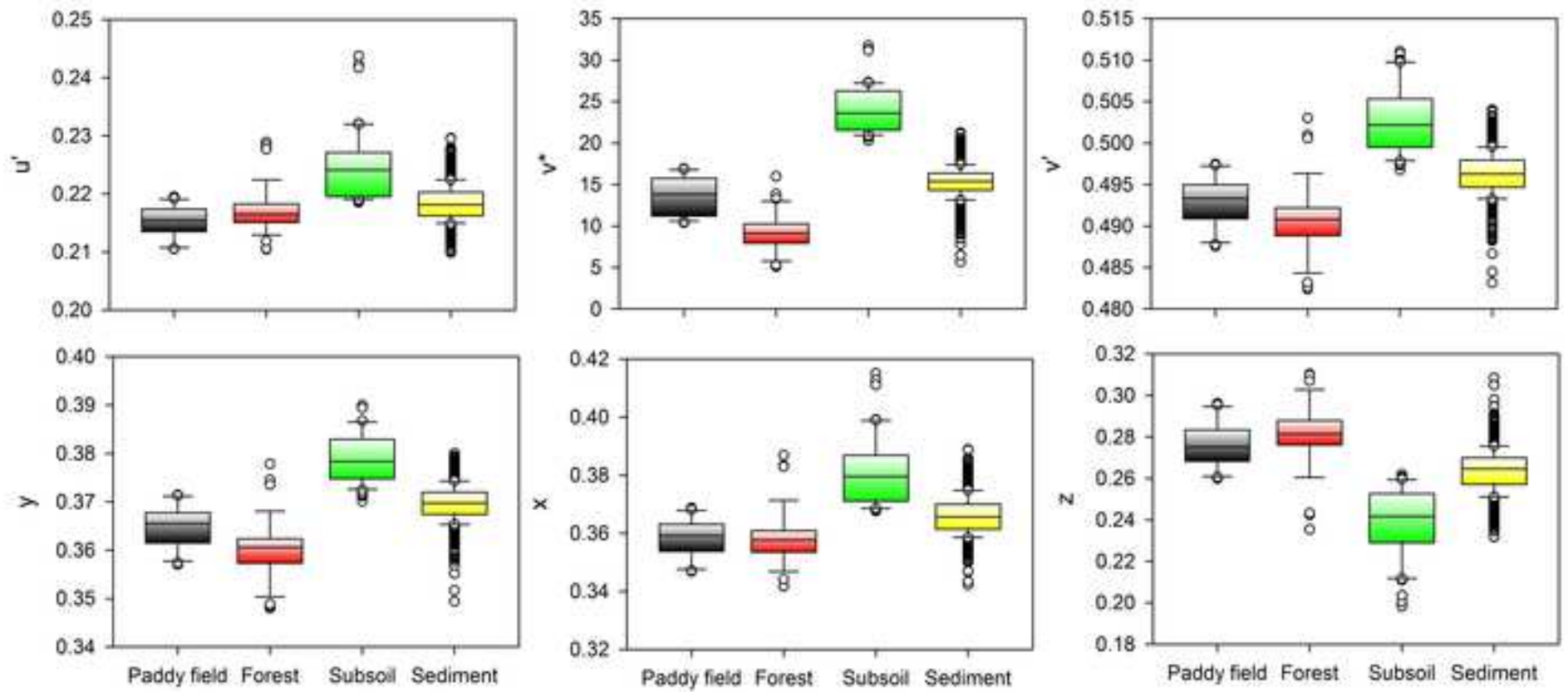


Figure 4

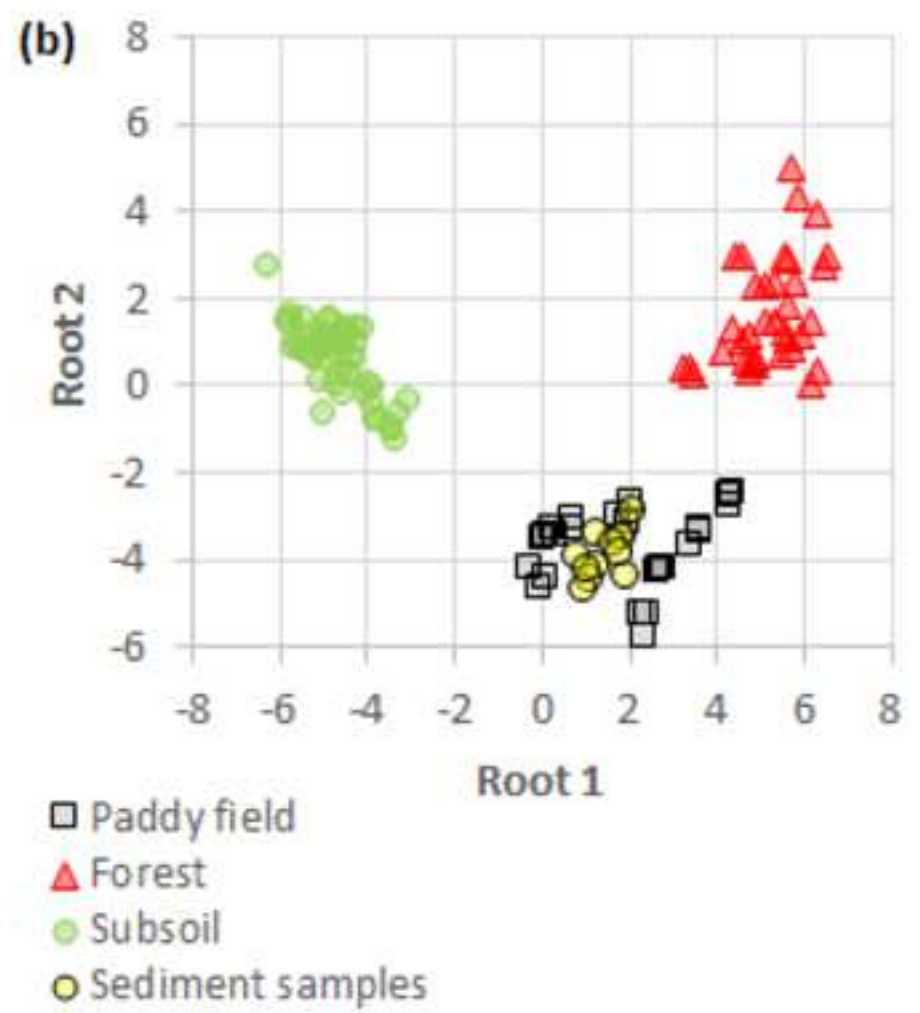
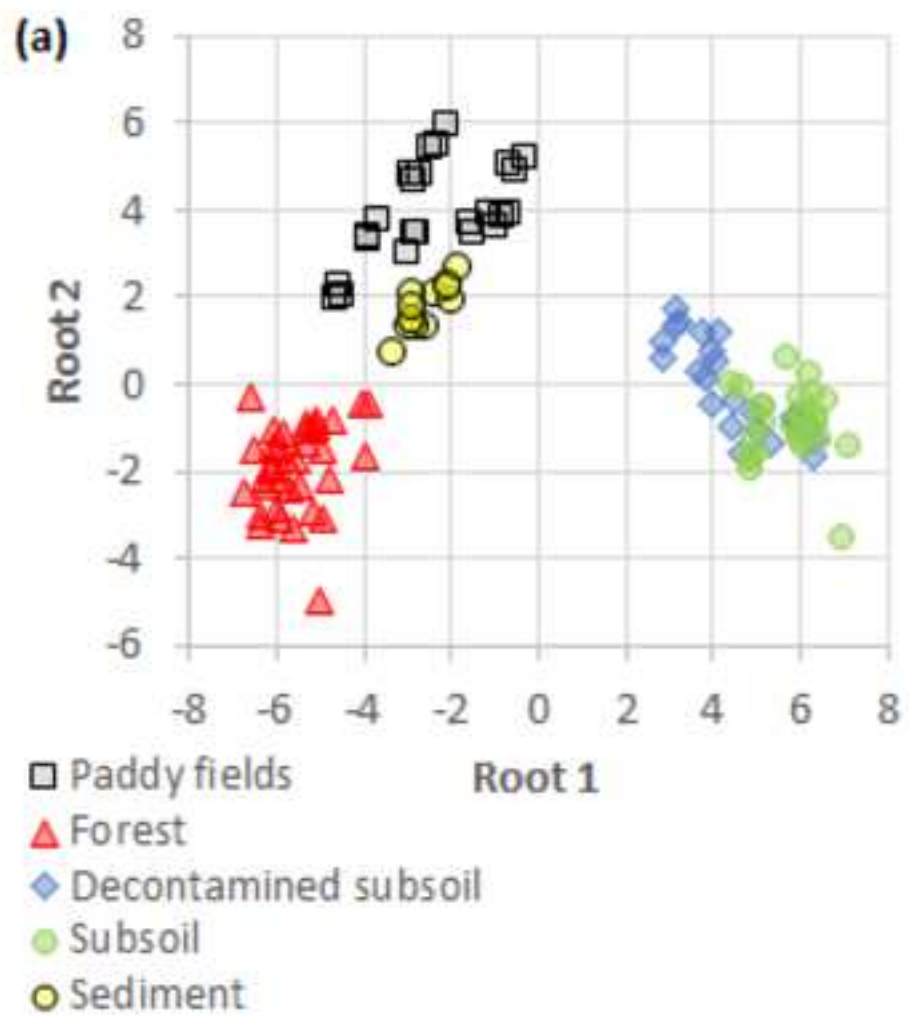


Figure 5

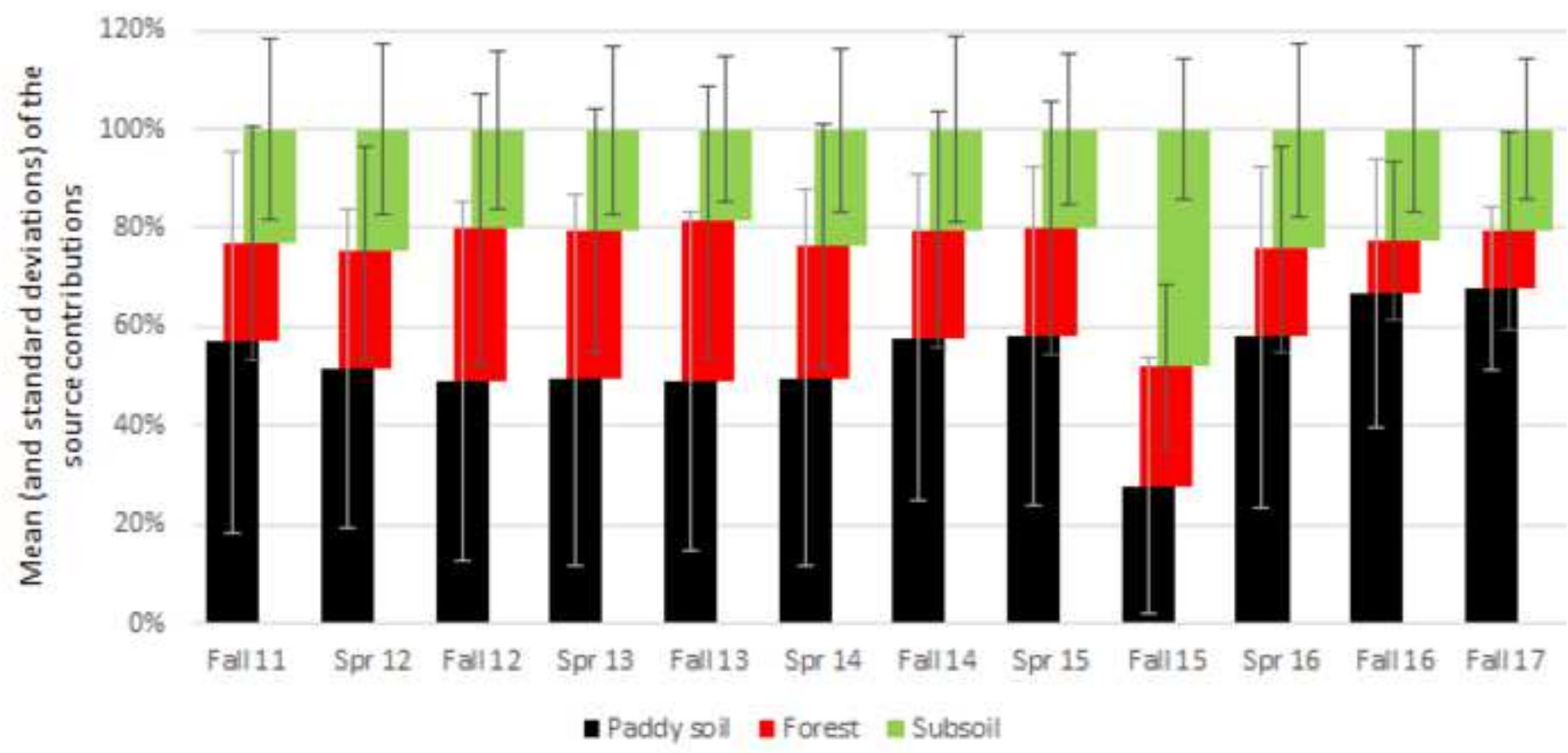


Table 1.

Kruskal-Wallis H test results (p-values) examining the occurrence of significant differences between sources based on colorimetric parameters.

Colour parameter	Four source approach		Three source approach	
	Kruskal-Wallis test		Kruskal-Wallis test	
	H-value	p-value	H-value	p-value
L*	90.6	<0.001	90.5	<0.001
a*	79.4	<0.001	75.6	<0.001
b*	90.4	<0.001	87.7	<0.001
C*	90.4	<0.001	87.7	<0.001
h	70.6	<0.001	63.5	<0.001
x	73.8	<0.001	71.4	<0.001
y	82.8	<0.001	80.7	<0.001
z	76.6	<0.001	74.2	<0.001
L	90.6	<0.001	90.5	<0.001
a	81.0	<0.001	77.0	<0.001
b	92.7	<0.001	90.7	<0.001
u*	88.6	<0.001	85.3	<0.001
v*	92.6	<0.001	90.6	<0.001
u'	68.7	<0.001	65.4	<0.001
v'	78.7	<0.001	76.3	<0.001

Table 2. Results of the discriminant function analysis with four potential sources in the Mano and Niida catchments.

DFA parameters	Four source approach	Three source approach	
<i>Selected tracers</i>	L*, a*, b*, C*, h, x, y, v', L, a, b, u', v*	L*, a*, b*, v*, h, a, b, u', L	
<i>Source type samples correctly classified (%)</i>			
Forest	100	Forest	100
Subsoil	88	Subsoil	100
Decontaminated subsoil	75	Paddy fields	100
Paddy fields	100	-	-
Total	92	Total	100
<i>Uncertainty associated with the discrimination of the source (%)</i>			
Forest	7.41E-04	Forest	9.82E-03
Subsoil	13.61	Subsoil	1.72E-06
Decontaminated subsoil	23.23	Paddy fields	5.30E-02
Paddy fields	0.18	-	-

