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### ► **To cite this version:**

O. Evrard, J. Patrick Laceby, Gentile Francesco Ficetola, Ludovic Gielly, Sylvain Huon, et al.. Environmental DNA provides information on sediment sources: A study in catchments affected by Fukushima radioactive fallout. *Science of the Total Environment*, 2019, 665, pp.873-881. 10.1016/j.scitotenv.2019.02.191 . hal-02049144

**HAL Id: hal-02049144**

**<https://hal.science/hal-02049144>**

Submitted on 12 May 2020

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1 **Environmental DNA provides information on sediment sources: A study in catchments affected by**  
2 **Fukushima radioactive fallout**

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36 **Abstract**

37 An excessive supply of sediment is observed in numerous rivers across the world where it leads to  
38 deleterious impacts. Information on the sources delivering this material to waterbodies is required to  
39 design effective management measures, and sediment tracing or fingerprinting techniques are  
40 increasingly used to quantify the amount of sediment derived from different sources. However, the  
41 current methods used to identify the land use contributions to sediment have a limited  
42 discrimination power. Here, we investigated the potential of environmental DNA (eDNA) to provide  
43 more detailed information on the plant species found in sediment source areas as a next generation  
44 fingerprint. To this end, flood sediment deposits ( $n=12$ ) were collected in 2017 in two catchments  
45 impacted by the Fukushima radioactive fallout along differing river sections draining forests, cropland  
46 or a mix of both land uses. Conventional fingerprints (i.e. fallout radionuclides and organic matter  
47 properties) were also measured in these samples. The conventional fingerprint model results showed  
48 that most sediment samples contained a dominant proportion of subsoil material. Nevertheless, the  
49 eDNA information effectively discriminated the three above-mentioned groups of sediment, with the  
50 dominance of tree, shrub and fern species in sediment sampled in rivers draining forests versus a  
51 majority of grass, algae and cultivated plant species in sediment collected in rivers draining cropland.  
52 Based on these encouraging results, future research should examine the potential of eDNA in mixed  
53 land use catchments where the contribution of topsoil to sediment dominates and where the  
54 cultivation of land has not been abandoned in order to better characterize the memory effect of  
55 eDNA in soils and sediment.

56

57 **Keywords:** Sediment fingerprinting; catchment; sediment source; FDNPP accident; ancient plant DNA  
58 (aDNA).

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## 61 Introduction

62 Soil erosion is often exacerbated in agricultural catchments where erosion rates may be an order of  
63 magnitude greater on cropland relative to grassland and forested landscapes (Cerdan et al., 2010;  
64 Montgomery, 2007). Although particle detachment processes are generally reduced in areas with  
65 dense vegetation or an abundant litter layer, soil erosion regularly occurs on these natural  
66 landscapes (Fukuyama et al., 2010; Laceby et al., 2016b). Soil erosion rates are accelerating  
67 worldwide (Amundson et al., 2015), resulting in deleterious on-site and off-site consequences (Evrard  
68 et al., 2007).

69 One major off-site consequence of soil erosion are elevated suspended sediment loads that reduce  
70 fish spawning habitats (Owens et al., 2005), decrease reservoir life spans (Foucher et al., 2014) and  
71 degrade water quality (Gateuille et al., 2014). Accordingly, it is important to quantify the relative  
72 contribution of the major sources supplying suspended sediment to river networks in order to  
73 develop effective best management practices that mitigate deleterious downstream effects of  
74 accelerated soil erosion.

75 Sediment source fingerprinting is increasingly used (Walling, 2013) to estimate the relative source  
76 contributions of detached particles through the analysis of physical or biogeochemical properties, or  
77 ‘fingerprints,’ in source soils and their downstream sediments (Walling et al., 1979). A variety of  
78 fingerprinting properties can be used to estimate sediment source contributions, including fallout  
79 radionuclides (e.g.  $^{137}\text{Cs}$ ,  $^{210}\text{Pb}_{\text{xs}}$ ,  $^7\text{Be}$ ) (Evrard et al., 2013), element geochemistry (Douglas et al.,  
80 2003), mineral magnetics (Walden et al., 1997), colour (Legout et al., 2013) or organic parameters  
81 (TN, TOC) and their isotopes (Gourdin et al., 2015), amongst others.

82 There are three fundamental approaches to sediment source fingerprinting. First, researchers can  
83 investigate the dominant erosion processes (e.g. surface or subsurface erosion) contributing material  
84 to the river network (Wallbrink et al., 1998). Second, researchers can examine temporal dynamics, to  
85 verify whether sediment transiting the river network consists of recently or previously eroded

86 material (Evrard et al., 2016a). Third, researchers can determine the dominant spatial source areas  
87 contributing particulate material. For this latter approach, the dominant spatial areas often  
88 investigated include lithological regions (e.g. bedrock or surficial geology)(Le Gall et al., 2017), soil  
89 types (Lepage et al., 2016), and land use or land cover (Tiecher et al., 2017).

90 To investigate sediment derived from different land uses or land covers, researchers are increasingly  
91 tracing the vegetation signatures from sources areas with carbon and nitrogen elemental  
92 concentrations (TOC and TN) and their stable isotope ratios ( $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ ) (Gourdin et al., 2015;  
93 Huon et al., 2013; Lacey et al., 2016c). As the discrimination power of these properties is limited,  
94 particularly in areas with a mixture of  $\text{C}_3$  and  $\text{C}_4$  plants (Evrard et al., 2013), researchers are  
95 expanding their toolbox, for example, by tracing the fatty acids in sediments to their source soils with  
96 Compound-Specific Stable Isotope (CSSI) analysis (Gibbs, 2007). These approaches provide detailed  
97 information regarding the vegetative source signature of the sediments, including potentially  
98 different land uses (e.g., permanent pasture, woodland, cultivated land) or even agricultural  
99 practices (e.g. maize and stubble)(Blake et al., 2012). Nevertheless, the discrimination potential of  
100 these organic tracers is typically limited to the broad land uses (Bravo-Linares et al., 2018) and  
101 debate remains regarding the conservative behavior of these organic tracers during their transport  
102 across the landscape (Koiter et al., 2013; Reiffarth et al., 2016). Therefore, other fingerprints are  
103 required that provide more specific and potentially more conservative source information.

104 Environmental DNA (eDNA) is the DNA obtained from environmental samples such as water and  
105 sediment. The DNA of plants and animals living in soil and above the soil is transferred to the river  
106 network by erosion processes, and was shown to be preserved in deposited particulate matter  
107 (Giguet-Covex et al., 2014; Parducci et al., 2017; Pedersen et al., 2015). The preservation of eDNA is  
108 much higher in sediment – including in sediment dating back to the Holocene or the Pleistocene  
109 (Willerslev et al., 2003) – than in freshwater ecosystems (Dejean et al., 2011; Thomsen et al., 2012).  
110 With recent advances in High Throughput Sequencing, eDNA offers a novel approach that could

111 provide far more detailed sediment source information than CSSI tracers. In other fields, eDNA has  
112 already provided highly detailed information regarding specific plant types and/or land management  
113 practices with the analysis of eDNA in ancient sediment in lacustrine sediment cores (Ficetola et al.,  
114 2018; Pansu et al., 2015; Sjogren et al., 2016). However, to the best of our knowledge, eDNA has  
115 never been analysed in contemporary sediments with the objective of determining the relative  
116 contribution of different sediment sources through the improved discrimination of different  
117 vegetation species.

118 In catchments of Northeastern Japan impacted by radioactive fallout from the Fukushima Dai-ichi  
119 Nuclear Power Plant (FDNPP) accident in March 2011 (Evrard et al., 2015), the quantification of the  
120 land use contributions to sediment transiting the rivers to the Pacific Ocean is imperative in order to  
121 prioritize remediation works. Previous fingerprinting research based on the measurement of carbon  
122 and nitrogen elemental concentrations and their stable isotope ratios showed that subsoils were the  
123 dominant sources ( $45\pm 26\%$ ) of particulate matter to the rivers, followed by cultivated land ( $38\pm 19\%$ )  
124 and forests ( $17\pm 10\%$ ) including evergreen and deciduous species (Lacey et al., 2016b). However, the  
125 identification of the land use contributions was provided with limited resolution so far. Although  
126 areas cultivated with rice (*Oryza sativa* L.) were shown to supply a disproportionate amount of  
127 radiocesium contaminated sediment to the region's waterbodies (Yoshimura et al., 2016),  
128 uncertainties remain regarding the specific contribution of paddies compared to that of other  
129 cultivated fields or that of forested areas to sediment transiting these rivers.

130 Accordingly, the objective of this study is to explore the potential of eDNA as a next generation  
131 sediment source fingerprint. As this is, to our knowledge, the first application of eDNA in a sediment  
132 source fingerprinting context, our goal is to compare the discrimination potential of the source  
133 information provided by eDNA to that achieved with carbon and nitrogen properties (TOC, TN,  $\delta^{13}\text{C}$   
134 and  $\delta^{15}\text{N}$ ) and fallout radionuclides ( $^{137}\text{Cs}$ ). Our main objective is to demonstrate the potential utility  
135 of eDNA as a sediment source fingerprint, through highlighting its higher source discrimination

136 potential relative to conventional sediment fingerprinting approaches. Our secondary objective is to  
137 overview the challenges and opportunities of developing eDNA as a sediment fingerprinting  
138 parameter of the future.

## 139 **Materials and Methods**

### 140 ***Study area***

141 This research was conducted in the Niida (275 km<sup>2</sup>) and Ota (77 km<sup>2</sup>) catchments (Fig. 1), draining the  
142 main radioactive contamination plume of the Fukushima Prefecture, Northeastern Japan. This region  
143 is exposed to a very erosive climate (mean annual rainfall of ~1400 mm), with the occurrence of  
144 spring floods and typhoons being the main hydro-sedimentary events that may lead to significant  
145 erosion and sediment transfer in rivers (Chartin et al., 2017; Laceby et al., 2016a). The Niida and Ota  
146 River catchments are characterized by land uses that are representative of those found in the fallout  
147 impacted region, with a dominance of forests (75%) and cropland (21%).

148 The main catchment features include an upstream coastal mountain range (<900 m) and a broad,  
149 more densely inhabited, coastal plain (i.e. <100 m). Dams are installed at the transition between both  
150 zones on the Ota River (Yokokawa Dam) and on the main Niida River tributary (Takanokura Dam) in  
151 the coastal plain (i.e., Mizunashi River) to provide water for drinking, and irrigation to the local  
152 communities. These dams create sediment disconnectivities between upper and lower catchment  
153 areas, with dams storing sediment (Chartin et al., 2017). The Niida River mainstream is devoid of  
154 dam.

### 155 ***Sediment sampling***

156 Sediment samples were collected in June 2017 at sites draining only forests ( $n=5$ ), a dominance of  
157 cropland ( $n=4$ ), or a mix of both land uses ( $n=3$ ; Fig. 1). These samples comprise particulate material  
158 that settled on channel banks, inset benches and floodplains during the falling limb of the last  
159 significant hydro-sedimentary event. One soil sample (0 – 5 cm surface layer) was also collected in a

160 paddy field located near the Ota River for comparison (Table 1). For fallout radionuclide and  
161 carbon/nitrogen measurements, 5 subsamples (~5 g per subsample) of recently deposited material at  
162 each site were taken with a plastic spatula over a 5 m reach and composited into one sample. For the  
163 eDNA analyses, 5 subsamples (~ 5 g per subsample) were collected at each site with a trowel,  
164 sterilized with a blowtorch, mixed and homogenized. Approximately 15 g of material were stored in a  
165 sealed container with silica gel bags for in-situ drying.

#### 166 ***Fallout radionuclide analysis and modelling***

167 Prior to analyses, samples were dried at 40°C for ~48 h, sieved to 2 mm, ground to a fine powder in  
168 an agate mortar, and pressed into 15 mL polyethylene containers for analysis. Fallout radionuclide  
169 activities were determined with gamma spectrometry using coaxial N- and P-type HPGe detectors  
170 (Canberra/Ortec).  $^{137}\text{Cs}$  activities were measured at the 662 keV emission peak.  $^{134}\text{Cs}$  activities were  
171 calculated as the mean of the counts found at the 605 and 796 keV emission peaks. All radionuclide  
172 activities were decay-corrected to March 14, 2011, the date of the main radionuclide fallout  
173 deposition (Kinoshita et al., 2011).

174 Based on the  $^{137}\text{Cs}$  analyses, the relative contributions of contaminated (high  $^{137}\text{Cs}$ ) versus depleted  
175 (low  $^{137}\text{Cs}$ ) sources of radiocesium contaminated sediment were modelled in the Niida catchment.  
176 The model was run with the source sample data from Evrard et al. (2016b) which included 118  
177 samples of contaminated source soil and 42 samples of depleted source material sampled in the  
178 Niida catchment. A distribution modelling approach estimated the relative contribution of these two  
179 end-members (Lacey and Olley, 2015). The objective was to estimate whether the sediment  
180 samples in the Niida catchment were derived from upstream contaminated sources (e.g. forested  
181 reaches) versus cropland or other areas in the coastal plains that did not receive significant fallout.  
182 Although the source dataset has been published in Evrard et al. (2016b), the sediment dataset is  
183 original and has not been published elsewhere.

184



185 ***Carbon and nitrogen analysis and modelling***

186 A subsample of the material analysed for fallout radionuclides was analysed for TOC, TN,  $\delta^{13}\text{C}$  and  
187  $\delta^{15}\text{N}$  with a continuous flow Elementar® VarioPyro cube analyzer coupled to a Micromass® Isoprime  
188 Isotope Ratio Mass Spectrometer (EA-IRMS) at the Institute of Ecology and Environmental Science  
189 (iEES) in Paris. First, TOC and  $\delta^{13}\text{C}$  were measured with a set of tyrosine standards (Coplen et al.,  
190 1983; Girardin and Mariotti, 1991). Second, TN and  $\delta^{15}\text{N}$  were analysed with sample weights  
191 optimized after the carbon analyses. Oxygen for combustion was injected during 70s ( $30\text{ mL min}^{-1}$ )  
192 and temperatures were set at  $850^\circ\text{C}$  and  $1120^\circ\text{C}$  for the reduction and combustion furnaces,  
193 respectively (Agnihotri et al., 2014).

194 The carbon and nitrogen parameters were used to trace land use/land cover sources with a source  
195 data set from Laceby et al. (2016b). This included TOC, TN,  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  data for 46 forest, 28  
196 cultivated and 25 subsoil source samples analysed with identical methods at iEES. A concentration  
197 dependent distribution modelling approach estimated the relative contribution of these two end-  
198 members (Laceby et al., 2015) and provide the foundation for comparing the eDNA analyses with  
199 modeled source contributions that differentiate agricultural sources from those anticipated from  
200 forests and subsoil/decontaminated sources in the Fukushima coastal region that was impacted by  
201 the FDNPP accident. Although the source dataset is published in Laceby et al. (2016b), the sediment  
202 dataset is original and has not been published elsewhere.

203 ***eDNA analysis and interpretation***

204 Sediment eDNA was extracted following the protocol described in Pansu et al. (2015), targeting  
205 extra-cellular eDNA. For each sample,  $\sim 15\text{ g}$  of sediment was mixed with  $15\text{ ml}$  of saturated  
206 phosphate buffer ( $\text{Na}_2\text{HPO}_4$ ;  $0.12\text{ M}$ ,  $\text{pH} \approx 8$ ) for  $15\text{ min}$ . Two ml of the mixture was centrifuged  
207 ( $10\text{min}$  at  $10000\text{ g}$ );  $400\text{ }\mu\text{l}$  of resulting supernatant was kept as starting material for extraction using  
208 the NucleoSpin® Soil kit (Macherey-Nagel, Düren, Germany), skipping the cell lysis step and following

209 manufacturer's instructions (Taberlet et al., 2012). The extracted DNA was eluted in 100 µl of SE  
210 buffer and used as PCR template. Seven extraction controls were also conducted. The eDNA of  
211 vascular plants was amplified with the g-h primers, which amplify a short variable region on  
212 chloroplast DNA and have very limited mismatches with sequences of vascular plants (Taberlet et al.,  
213 2018). Additionally, six PCR controls containing PCR mix and no DNA template, along with six positive  
214 PCR controls were performed (Parducci et al., 2017). Each sediment sample and control was  
215 amplified in four PCR replicates (Ficetola et al., 2015). The sequencing was conducted by 2 × 125–  
216 base pair pair-end sequencing on an Illumina HiSeq 2500 platform. The sequences of DNA were  
217 filtered with OBITools software (Boyer et al., 2016). Plant sequences were assigned with a database  
218 of the vascular plants found in Japan. Sequences with a >97% match with a plant genus were kept,  
219 providing the table of the Molecular Taxonomic Units (MOTUs) to analyse. For each sample, the sum  
220 of the reads of the four replicates was calculated. Those MOTUs associated with a total number of  
221 reads lower than 1000 were removed from further analysis. The logarithm of the reads of those  
222 sequences kept for analysis +1 was then calculated as a measure of relative abundance (Yoccoz et al.,  
223 2012). A Principal Component Analysis (PCA) was conducted in order to discriminate various groups  
224 of sediment based on the significant MOTUs detected in the samples. A hierarchical classification of  
225 the retained MOTUs was also performed and, finally, a Permutational Multivariate Analysis of  
226 Variance Using Distance Matrices (PERMANOVA) was conducted to assess the amount of variation of  
227 the eDNA explained by the sources.

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## 233 **Results and discussion**

### 234 ***Fallout radionuclides***

235 Sediment in lag deposit samples draining forest soils had both significantly higher and more variable  
236  $^{137}\text{Cs}$  activity concentrations (mean (M): 23,436 Bq kg<sup>-1</sup>; SD: 17,810 Bq kg<sup>-1</sup>) compared to rivers  
237 draining mixed land uses (both forest and cropland) (M: 3238 Bq kg<sup>-1</sup>; SD: 3156 Bq kg<sup>-1</sup>) and cropland  
238 only (M: 2962 Bq kg<sup>-1</sup>; SD: 2002 Bq kg<sup>-1</sup>). The  $^{137}\text{Cs}$  activity concentration measured in the paddy field  
239 soil was low (1384 ±12 Bq kg<sup>-1</sup>), although it remained in the range of values found in sediment  
240 collected in rivers draining cultivated land.  $^{134}\text{Cs}/^{137}\text{Cs}$  activity ratios for all three sediment sample  
241 types were in a narrow range (0.95–1.02) consistent with the ratio expected from fallout from the  
242 FDNPP accident (Kobayashi et al., 2017).

243 Sediment  $^{137}\text{Cs}$  activity concentrations were modeled in the Niida catchment to investigate whether  
244 they were derived primarily from contaminated regions in the catchments or those that have been  
245 decontaminated and/or not impacted by the initial fallout. On average, 6% (SD 7%) of sediment in  
246 the Niida catchment was modeled to be derived from sources contaminated by the initial fallout (e.g.  
247 upper catchment areas and surface soils). The forest sample from the Mizunashi reach had the  
248 highest contribution of fallout-contaminated landscapes (14%, Model Error (ME) 1.2%). The paddy  
249 field sample farther downstream on this reach had a 5% (ME 1%) contribution from contaminated  
250 areas. In contrast, on the main stem, three samples had a less than 2% contribution from  
251 contaminated areas. Interestingly, the sediment sample collected the farthest downstream in the  
252 Niida catchment had an increased contaminated area contribution of again ~14%. Overall, most of  
253 the sediment sampled was derived from sources that were not contaminated with fallout from the  
254 FDNPP (e.g. subsoils, decontaminated soils, or regions near the coast that did not receive fallout).

### 255 ***Carbon and nitrogen composition***

256 The TN content was too low for accurate measurement in 2 of the 13 samples analyzed (FNL1243 and  
257 FOL1233). These samples also had the lowest TOC (<0.6%) of all analyzed samples indicating that  
258 they are mainly comprised of coarser material, most likely provided by subsoil sources. The sediment  
259 collected in the forested catchments had lower mean  $\delta^{13}\text{C}$  (M -28.4‰, SD 0.8‰) and  $\delta^{15}\text{N}$  (M 1.8‰,  
260 SD 1.6‰) compared to those sampled in cultivated catchments (M -27.1‰, SD 0.9‰ and M -4.3‰,  
261 SD 0.3‰ respectively). Three of the forested catchment sediment samples, and one of the cultivated  
262 catchment sediment sample (FNL1245) had low TOC (<1.2%) and low TN (<0.08%) suggesting higher  
263 contributions of subsoil sources that are known to be low in organic matter content. The three  
264 remaining cultivated catchment sediment samples had slightly higher TOC (M 1.4%, SD 0.2%) and TN  
265 (M 0.11%, SD 0.03%). The remaining forest catchment sample (FOL1239) had very high TOC (19.3%)  
266 and TN (1.1%). The carbon and nitrogen parameters suggest that the sediment collected in forested  
267 and cultivated catchments likely have significant subsoil source material contributing to the low TOC  
268 and TN content, with the exception of slightly elevated material derived from cultivated sources, and  
269 also one sample, owing to its very high organic contents, dominated by forest material.

270 Sediment in three of the mainly forested catchments were modelled to be almost entirely derived  
271 from subsoil sources (M 97%, SD 3%), whereas the high TOC sample was modelled to be entirely  
272 from forested sources (100%). Importantly, there was a 0% contribution of cultivated land modelled  
273 for the forested sites, supporting that this model is providing relevant predictions as there are  
274 essentially no rice paddy fields in these forested regions. For the cultivated catchments, 86% (SD 8%)  
275 of sediment was modelled to be derived from subsoil sources, only 13% (SD 8%) from rice paddy  
276 fields or other agricultural sources, and with a mean of 2% (SD 1%) from forest sources. One of the  
277 two mixed catchment sediment samples (FNL1242) was modelled to be derived entirely from subsoil  
278 sources (100%) whereas the other (FNL1245), was modelled to be derived from 44% cultivated land,  
279 18% forest and only 38% subsoil sources, the lowest subsoil source contribution modelled. Indeed,  
280 subsoil erosion processes dominate the supply of sediment across these catchments (M 77%, SD  
281 33%). Furthermore, the sampling location FNL1245 behaved differently than the other samples

282 again. The higher <sup>137</sup>Cs and contaminated source contribution in this sample relative to the others on  
283 this reach is likely due to the higher forest and cropland source contributions, which are diluted on  
284 the main stem of the Niida in downstream direction across the coastal plain.

#### 285 **eDNA**

286 After filtering the data, 67 abundant MOTUs were kept for analyses (Appendix A). When examining  
287 those sequences individually (Fig. 2), woodland species including mosses, ferns and shrubs were  
288 almost exclusively found in sediment collected in rivers draining forested areas or mixed land uses. In  
289 contrast, grasses and rice (*Oryza sativa* L.) dominated in the samples collected in rivers draining  
290 cropland and mixed land uses. In addition, species and/or families of cultivated plants were also  
291 found in these latter sediment samples, including Brassicaceae (e.g. cabbage), Fabaceae, Apiaceae  
292 (e.g. celery, carrot) or rice. These results demonstrate the potential of eDNA to provide a very  
293 detailed discrimination of those land use sources supplying sediment, with the identification of  
294 targeted crop types for instance.

295 To facilitate their analysis, the MOTUs were aggregated in 6 different groups: grasses, wetland  
296 grasses and cultivated plants (MOTUs 1 to 4); algae (MOTUs 5 to 14); mosses (MOTUs 15 to 29); ferns  
297 (MOTUs 30 to 35); trees and shrubs (MOTUs 36 to 50) and other plants including cultivated crops  
298 (MOTUs 51 to 67). The PCA (Fig. 3) using these 67 MOTUs showed a clear discrimination between  
299 samples corresponding to (i) the paddy field soil (group A) and (ii) those 3 classes of sediment  
300 identified during sampling: material collected in rivers draining cultivated land (group AA), in rivers  
301 draining mixed land uses (group B) and in watercourses draining forests (group C).

302 The hierarchical classification shows that the above-mentioned groups of MOTUs are well  
303 discriminated by the eDNA information (Fig. 4). Results from the PERMANOVA analysis showed that  
304 grouping explains a very large amount of variation of the eDNA composition ( $R^2 = 0.47$ ), with a  
305 strongly significant effect ( $p < 0.001$ ).

306 ***Combining the information provided by fallout radionuclides, organic matter composition and***  
307 ***eDNA***

308 For logistical reasons, the current research focused on the analysis of sediment deposits collected  
309 from different types of tributaries in catchments of the Fukushima region. The analysis of fallout  
310 radionuclides and organic matter properties of these samples, and the associated modeling showed  
311 that the majority of these samples contained significant proportions of subsoil material. Various  
312 sources may supply large quantities of subsoil material to the rivers in this region (Fig. 5). These  
313 include the mobilization of deep soil horizons under forests as a result of landsliding, gullying or road-  
314 side erosion (Mizugaki et al., 2008). Channel bank erosion may also provide a significant quantity of  
315 material to the rivers, in un-channeled reaches (Fig. 5b). Accordingly, soil particles are not only  
316 transferred by overland flow, they are also generated by erosion of deeper soil horizons.  
317 Nevertheless, this deeper soil material is still tagged with eDNA. A significant number of MOTUs (67)  
318 were identified in these samples containing high proportions of subsoil and the analysis of this eDNA  
319 provided the discrimination of the different sediment sample groups identified during fieldwork. As a  
320 result, a dominance of tree, fern and moss species were found in sediment collected in rivers  
321 draining forests, whereas a majority of grass and cultivated species were detected in sediment  
322 collected in watercourses draining cropland.

323 ***Implications for future research and concluding remarks***

324 Although the Fukushima case study was expected to be a relatively straightforward approach for  
325 discriminating land use-derived sediment sources in different rivers draining either forested areas or  
326 cropland, the situation was more complex owing to the large proportion of subsoil material transiting  
327 these river systems (Fig. 5). This is also likely due to the fact that our sampling approach targeted  
328 sediment deposited in the river channel after flooding events, rather than suspended matter  
329 transiting rivers. However, this approach was guided by the precautions required to avoid the  
330 biological contamination of the samples during their collection in the field and the necessity to

331 sterilize tools to collect the lag deposit samples rather than filter large volumes of river water  
332 containing lower quantities of suspended matter.

333 Furthermore, cultivated land was abandoned by farmers after the nuclear accident in March 2011.  
334 During the sampling campaign conducted in 2017, agricultural activities had not restarted in the  
335 study area. This situation likely explains the dominance of subsoil material transiting these rivers as a  
336 dense vegetation cover rapidly covered the abandoned fields and protected them against erosion,  
337 thereby limiting the contribution from the erosion of cultivated topsoils. Furthermore, this situation  
338 outlines the need to further investigate the memory effect of eDNA of cultivated plants in soils after  
339 6 years of abandonment, although the presence of plants cultivated in the region before the nuclear  
340 accident (e.g. cabbage, rice, mulberry) was demonstrated in the current research.

341 Based on these preliminary, encouraging results, future applications should be preferentially  
342 conducted in mixed land use catchments where the contribution of surface material to sediment  
343 transiting rivers is dominant and where agricultural activities have not been abandoned for several  
344 years. The systematic collection of both soil and sediment samples should be conducted in order to  
345 examine the memory effect of eDNA in soils and the potential changes in signatures from the soil to  
346 the sediment, including samples collected in rivers and in deposition areas (e.g. ponds or lakes).

347 Although the design of un-mixing models to quantify sediment source contributions to sediment is  
348 the ultimate objective of this research, multiple important phases of research are required before we  
349 can model eDNA effectively. This has been recently demonstrated by many reviews in the sediment  
350 tracing literature (Koiter et al., 2013; Lacey et al., 2017; Owens et al., 2016). For example, the  
351 conservative behavior of eDNA must be thoroughly researched to evaluate its persistence on the  
352 particles during mobilization, transport and deposition processes. Indeed, prior to modelling we need  
353 to understand if eDNA properties are maintained during these processes, or vary in a predictable way.  
354 Furthermore, we need to investigate whether there is an affinity for eDNA to be bound to different particle  
355 size fractions or even different soil and or mineral types. If eDNA indeed behaves conservatively

356 during sediment generation and deposition processes, without significant particle size impacts,  
357 researchers will have to then investigate what is the most appropriate approach to using eDNA in  
358 end member mixing models to quantitatively apportion source contributions to material transiting  
359 river networks. The analysis of artificial mixtures – in which known proportions of the different  
360 sources are combined in the laboratory – will likely be necessary to develop the best approach to  
361 incorporate eDNA into end member mixing models. We believe that the quantitative use of eDNA in  
362 sediment source fingerprinting models will be extremely beneficial for managing the degradation of  
363 our riverine, lacustrine and potentially estuarine environments through providing a more detailed  
364 source information than many of the currently employed fingerprints.

### 365 **Acknowledgements**

366 This work has been supported by the French National Research Agency (ANR) in the framework of  
367 the AMORAD project (ANR-11-RSNR-0002). The authors are grateful to Véronique Vaury for  
368 conducting the carbon and nitrogen parameter analyses.

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527 **Figure captions**

**Figure 1.** A) Location of the radioactive contamination plume and the selected Niida and Ota

catchments in the Fukushima Prefecture, Japan. B) The background map corresponds to the total radiocesium initial contamination of the soils (5-cm topsoil layer) decay-corrected to the 14 March, 2011 (Chartin et al., 2013). C) Sampling design in the Niida and Ota catchment along with land use.

**Figure 2.** Number of reads associated with a selection of plant types identified in the analysed samples collected in rivers draining cropland [C], forests [F] or a mix of cropland and forests [M].

528

529 **Figure 3.** Results of the Principal Component Analysis conducted to discriminate different groups of  
530 sediment samples based on the 67 significant MOTUs detected: group A – paddy field soil (in black);  
531 group AA – sediment collected in rivers draining cultivated land (in red); group B – sediment collected  
532 in rivers draining mixed land uses (in green); group C – sediment collected in rivers draining forests  
533 (in blue).

534

535 **Figure 4.** Results of the hierarchical classification conducted on the 67 significant MOTUs found in the  
536 analysed soil and sediment samples. Only those variables with  $\cos > 0.3$  were shown on the graph.

537

538 **Figure 5.** Example of sources delivering subsoil material to the rivers in the study site: (a) subsoil  
539 erosion in forests; (b) exposed channel bank in an un-channeled river section; (c) shallow landsliding  
540 on a roadside nearby a paddy field; (d) deeper landslide on a forested hillslope.

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**Table 1.** Location and characteristics of the sediment sampling sites, concentrations in fallout radionuclides ( $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$ ) and associated  $^{134}\text{Cs}/^{137}\text{Cs}$  activity

Sample ID	Basin	Sub-basin	Type	Y	X	Sampling date	$^{137}\text{Cs}$ (Bq kg <sup>-1</sup> )	$^{134}\text{Cs}$ (Bq kg <sup>-1</sup> )	$^{134}\text{Cs}/^{137}\text{Cs}$	TN (%)	TOC (%)	TOC/TN	$\delta^{13}\text{C}$ (‰)	$\delta^{15}\text{N}$ (‰)
FOL1233	Ota	Ota	Cultivated	37.60223	140.992928	28/06/2017	875	860	0.98	-	0.6	-	-26.3	-
FOS1235	Ota	Soil	Paddy	37.603681	140.955704	28/06/2017	1385	1360	0.98	0.26	3.2	12.2	-23.8	5.4
FOL1234	Ota	Ota	Cultivated	37.603668	140.953856	28/06/2017	3210	3265	1.02	0.14	1.62	11.6	-25.9	4.4
FOL1236	Ota	Ota	Cultivated	37.597836	140.925567	28/06/2017	6335	6260	0.99	0.09	1.22	13.6	-27.4	3.9
FOL1237	Ota	Ota	Forest	37.556682	140.874831	29/06/2017	17380	17080	0.98	0.05	0.75	15.0	-27.6	3.4
FOL1238	Ota	Ota	Forest	37.564072	140.872601	29/06/2017	20530	20560	1.00	0.06	0.91	15.2	-28.0	2.4
FOL1239	Ota	Ota	Forest	37.578823	140.874961	29/06/2017	48730	48855	1.00	1.10	19.29	17.5	-29.3	-0.4
FNL1240	Niida	Mizunashi	Forest	37.627235	140.875257	29/06/2017	7105	7050	0.99	0.08	1.2	15.3	-28.8	1.7
FNL1241	Niida	Mizunashi	Cultivated	37.639792	140.934335	29/06/2017	3435	3465	1.01	0.11	1.2	11.3	-28.1	4.2
FNL1242	Niida	Niida	Mix	37.651152	140.958309	29/06/2017	1895	1870	0.99	0.06	0.7	11.3	-26.1	6.3
FNL1243	Niida	Niida	Mix	37.661327	140.911026	29/06/2017	980	965	0.98	-	0.3	-	-25.1	-
FNL1244	Niida	Niida	Cultivated	37.664088	140.899675	29/06/2017	960	935	0.97	0.06	0.6	10.7	-27.0	4.5
FNL1245	Niida	Niida	Mix	37.651603	140.889993	29/06/2017	6845	6535	0.95	0.35	4.5	12.7	-26.8	3.3

ratio, organic matter properties in the soil/sediment samples.

**Table 2**[Click here to download Table: Table 2.docx](#)

**Table 2:** Dilution model results for the Niida catchment including the Cs-137 activities and modeled contribution from upstream areas that were contaminated by fallout by the FDNPP accident

<b>Sample ID</b>	<b>Basin</b>	<b>Sub-basin</b>	<b>Type</b>	<b>Cs-137 Bq kg<sup>-1</sup></b>	<b>Error Bq kg<sup>-1</sup></b>	<b>Contam. Source (%)</b>	<b>Error (%)</b>
FNL1240	Niida	Mizunashi	Forest	7104	25	14.0%	1.2%
FNL1241	Niida	Mizunashi	Cultivated	3435	18	4.6%	1.0%
FNL1242	Niida	Niida	Mix	1893	15	1.4%	1.0%
FNL1243	Niida	Niida	Mix	977	9	0.0%	1.0%
FNL1244	Niida	Niida	Cultivated	957	9	0.0%	1.0%
FNL1245	Niida	Niida	Mix	6843	41	13.7%	1.2%

**Table 3**[Click here to download Table: Table 3.docx](#)

**Table 3:** Results from the Carbon and nitrogen parameter model for the Niida and the Ota catchments including the Cs-137 activities and modeled contribution from upstream areas that were contaminated by fallout by the FDNPP accident

Sample ID	Basin	Sub-basin	Type	Forest (%)	ME (%)	Crop (%)	ME (%)	Subsoil (%)	ME (%)
FOS1235	Ota	Soil	Paddy	4%	1%	64%	2%	32%	3%
FOL1234	Ota	Ota	Cultivated	2%	2%	21%	3%	76%	10%
FOL1236	Ota	Ota	Cultivated	3%	2%	8%	12%	89%	20%
FOL1237	Ota	Ota	Forest	0%	1%	0%	5%	100%	15%
FOL1238	Ota	Ota	Forest	1%	3%	0%	5%	98%	17%
FOL1239	Ota	Ota	Forest	100%	1%	0%	1%	0%	1%
FNL1240	Niida	Mizunashi	Forest	6%	3%	1%	3%	93%	15%
FNL1241	Niida	Mizunashi	Cultivated	1%	2%	16%	3%	82%	16%
FNL1242	Niida	Niida	Mix	0%	1%	0%	4%	100%	9%
FNL1244	Niida	Niida	Cultivated	0%	1%	4%	2%	96%	15%
FNL1245	Niida	Niida	Mix	18%	1%	44%	3%	38%	16%

Figure 1  
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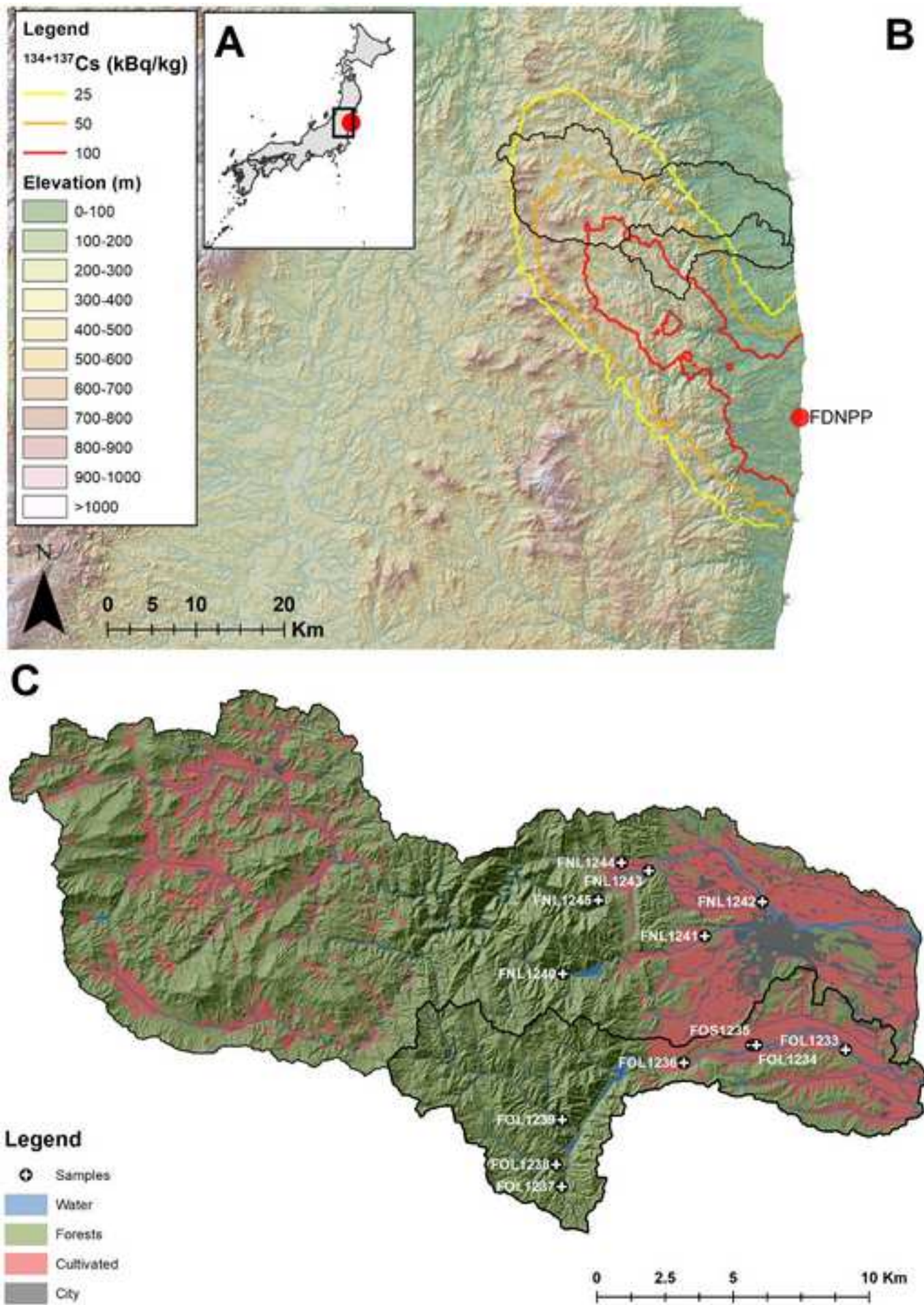




Figure 2

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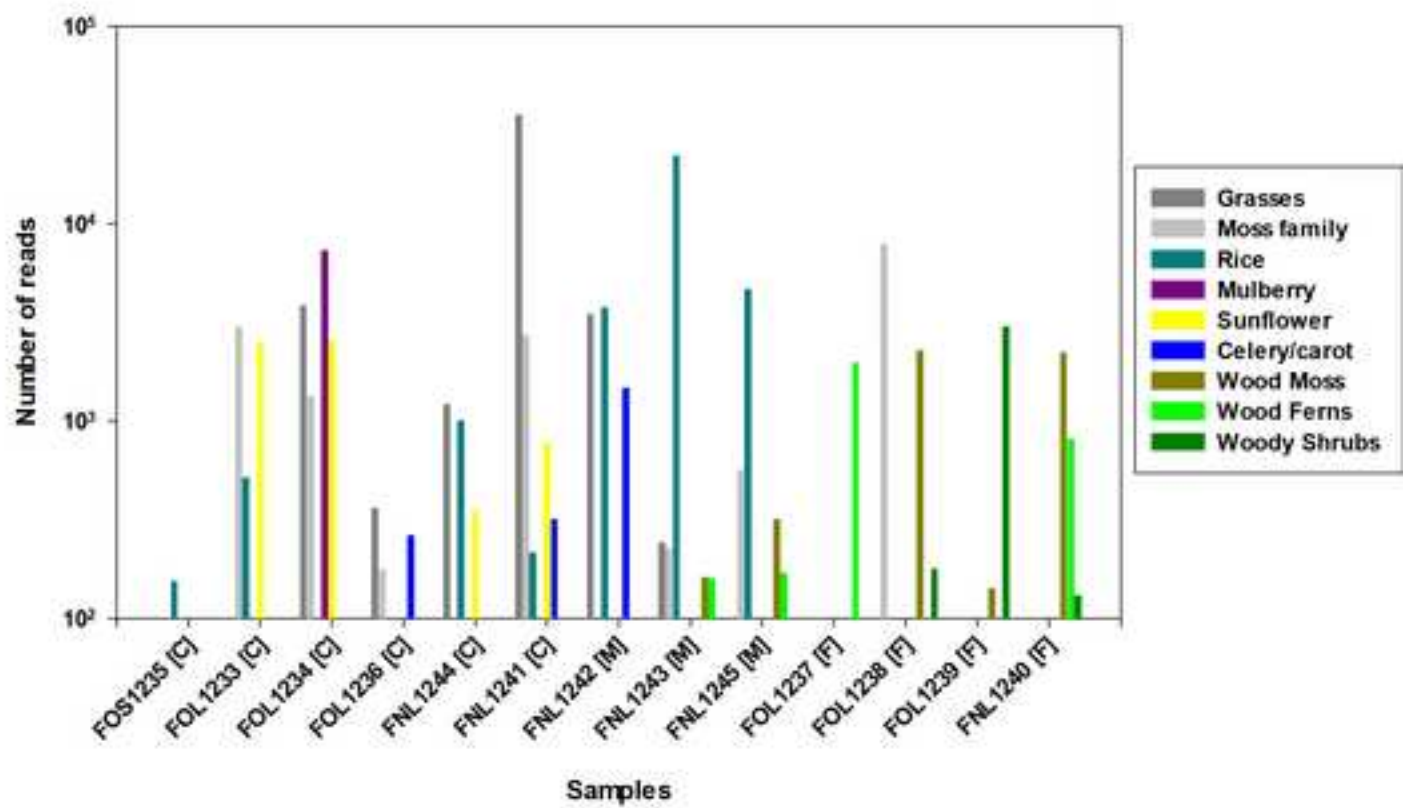
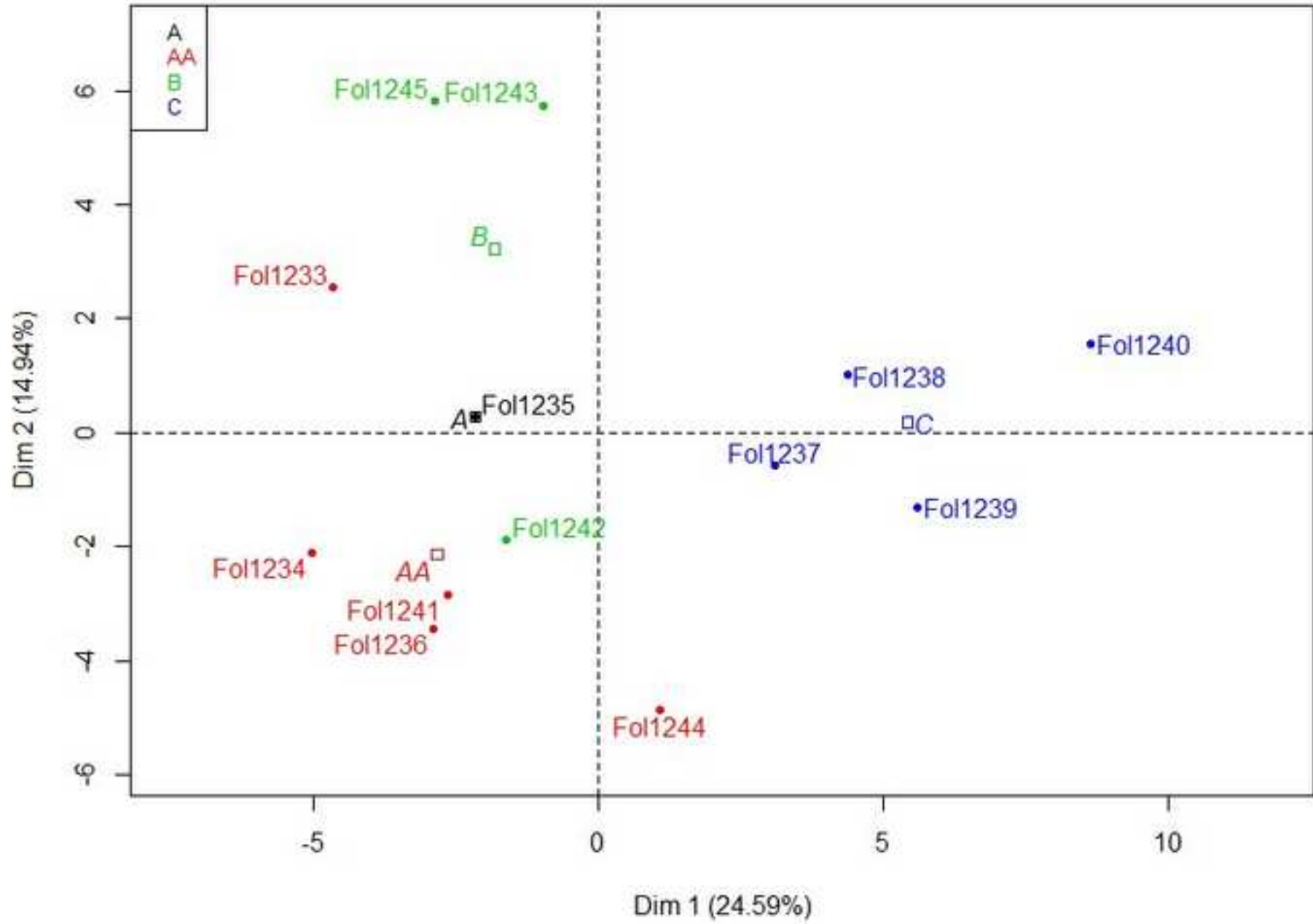


Figure 3  
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**Figure 5**  
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(a)



(b)



(c)



(d)



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