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16 **Temporal evolution of plutonium concentrations and isotopic ratios in**
17 **the Ukedo - Takase Rivers draining the Difficult-To-Return Zone in**
18 **Fukushima, Japan (2013–2020)**
19

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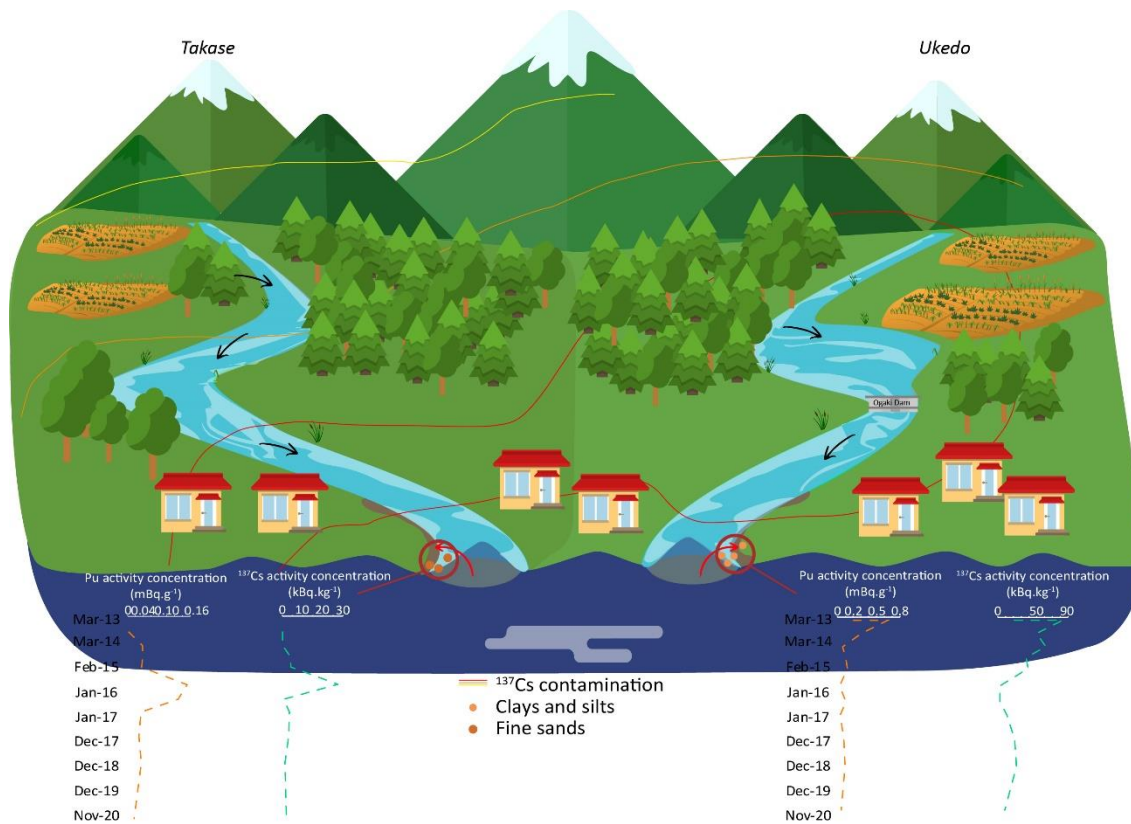
26
27 **Key words:** ²⁴⁰Pu/²³⁹Pu isotopic ratios, Fukushima nuclear power plant accident, flood sediment
28 deposits, radiocesium.
29

30 **Abstract**

31 In 2011, the Fukushima Dai-Ichi Nuclear Power Plant (FDNPP) accident released significant
32 quantities of radionuclides into the environment. Japanese authorities decided to progressively reopen
33 the Difficult-To-Return Zone after the decontamination of priority reconstruction zones. These areas
34 include parts of the initially highly contaminated municipalities located to the north of the FDNPP,
35 including Namie Town, an area drained by the Ukedo and Takase Rivers. Eleven years after the accident,
36 research focused on the spatial distribution of plutonium (Pu) and radiocesium (Cs) isotopes at
37 contrasted individual locations. To complement previous results, the current research was conducted
38 on flood sediment deposits collected at the same locations after major flooding events during eleven
39 fieldwork campaigns organised between 2013 and 2020 at the outlet of the Ukedo and Takase Rivers
40 (n = 22).

41 The results highlighted a global decrease of the Pu and ¹³⁷Cs contents in sediment with time during
42 the abandonment phase in the region, from 2013 (238.20 fg.g⁻¹) to 2020 (4.28 fg.g⁻¹). Furthermore,
43 based on the analysis of the ²⁴⁰Pu/²³⁹Pu isotopic ratios, the plutonium transiting these rivers (range:
44 0.166 - 0.220) essentially originated from the global fallout (0.180 ± 0.014 (Kelley et al., 1999)).
45 Sediment showed contrasted properties in the two investigated rivers, which is likely mainly the result
46 of the occurrence of Ogaki Dam on upper sections of the Ukedo River as it strongly impacts the material
47 supply from this river to the Pacific Ocean. A statistical analysis highlighted the strong correlation
48 between Pu activity concentrations and ¹³⁷Cs activities in both rivers, confirming that both
49 radionuclides are transported with a similar pathway. Despite it was detected early after the accident

50 (2011 – 2013), the current research demonstrates that plutonium originating from FDNPP is no longer
 51 detected in these rivers draining the Difficult-To-Return Zone at the onset of the reopening of the area
 52 to its former inhabitants.



53

54 Introduction

55 Following the accident that occurred at Fukushima Dai-Ichi Nuclear Power Plant (FDNPP) on March
 56 11, 2011, large quantities of radionuclides were emitted into the environment, with a total activity
 57 estimated to 520 PBq, which deposited mainly in regions of Northeast Japan (Steinhauser et al., 2014).
 58 Models showed that ~ 80 % of radionuclide releases occurred over the ocean (Mathieu et al., 2018;
 59 MEXT, 2011). These emissions mainly consisted in volatile or semi-volatile fission products (¹³⁷Cs, ¹³⁴Cs,
 60 ¹³¹I, ...) although amounts of actinides (U, Pu) from the reactor fuel were also released (Steinhauser et
 61 al., 2014). The most significant contaminant released into the environment was radiocesium (¹³⁴⁺¹³⁷Cs),
 62 with initial emissions estimated to 10 PBq of ¹³⁷Cs (Onda et al., 2020). In contrast, the releases of
 63 ²³⁹⁺²⁴⁰Pu were estimated to range from 1 GBq to 3.5 GBq, i.e. one million times lower than for
 64 radiocesium (Schneider et al., 2013; Shinonaga et al., 2014; Yamamoto et al., 2014; Zheng et al., 2013).
 65 From 2011 onwards, scientists focused on the characterisation of the spatial distribution of radioactive
 66 fallout as a prerequisite to investigate the potential health and environmental impacts of this accident
 67 (Adachi et al., 2013; Aliyu et al., 2015; Evrard et al., 2015, 2013; Onda et al., 2020). The primary means
 68 of radiocesium transport were shown to be soil erosion and transfer of particle-bound radiocesium

69 in river systems. In the case of actinides, some studies demonstrated their occurrence in the
70 environment under the form of microparticles (Igarashi et al., 2019; Kurihara et al., 2020; Martin et al.,
71 2019, 2016; Ochiai et al., 2018). Nevertheless, results suggest that their transportation in rivers may
72 follow similar pathways as radiocesium (Evrard et al., 2014). At the *bulk* scale, research on the actinide
73 isotopic characterization in sediment has shown that the $^{235}\text{U}/^{238}\text{U}$ does not allow to discriminate its
74 source (Jaegler et al., 2019), which justifies the need to analyse Pu atom ratios to characterize its origin
75 (Cao et al., 2016; Evrard et al., 2014; Jaegler et al., 2018; Johansen et al., 2021; Oikawa et al., 2015;
76 Schneider et al., 2013; Shinonaga et al., 2014; Yamamoto et al., 2014). In particular, $^{240}\text{Pu}/^{239}\text{Pu}$ and
77 $^{241}\text{Pu}/^{239}\text{Pu}$ isotopic ratios are powerful tools to identify the plutonium source. In the northern
78 hemisphere, the main Pu source is linked to the atmospheric nuclear weapon tests performed from
79 1945 to 1980 (Bu et al., 2015a; Danesi et al., 2008; Muramatsu et al., 2001; Yamamoto et al., 2002).
80 During this period, plutonium isotopes were globally spread in the stratosphere and they can be
81 detected since then in the environment all around the world. The $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio of the global
82 fallout in the Northern Hemisphere is estimated to 0.180 (\pm 0.014) (Kelley et al., 1999). Since the
83 atmospheric nuclear tests, other emissions of plutonium into the environment occurred as a result of
84 the Chernobyl Nuclear Power Plant and the FDNPP accidents. A specific labelling of soils in Northern
85 Japan with the FDNPP Pu isotopic signature has also been outlined (Zheng et al., 2013). The plutonium
86 isotopic composition in the fuel of FDNPP at the moment of the accident was reconstructed by
87 Nishihara et al. (2012) based on model simulations. The $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio was shown to vary
88 depending of the reactor of emission, with values of 0.339, 0.315 and 0.351, for reactors 1, 2 and 3,
89 respectively.

90 As highlighted by Wu et al. (2022), only few studies investigated the Pu signature in sediment
91 samples. These studies demonstrated the occurrence of specific Pu fallout due to FDNPP in samples
92 collected between 2011 and 2013 (Evrard et al., 2014; Jaegler et al., 2019; Schneider et al., 2013;
93 Steinhauser et al., 2015; Yamamoto et al., 2014) although, to the best of our knowledge, longer-term
94 records and analyses on samples collected more recently near the FDNPP have not been conducted to
95 detect potential further changes in these Pu signatures.

96 Accordingly, the objective of this study is to investigate the temporal changes in Pu signatures
97 between 2013 and 2020 in flood sediment deposits collected in the Ukedo and the Takase Rivers
98 draining a part of the Difficult-to-Return Zone, in Fukushima Prefecture, and to identify potential
99 factors controlling these changes. To this end, the current research focused on (1) the characterization
100 of the Pu activity concentrations in sediment and their evolution, (2) the potential changes in sediment
101 Pu isotopic signatures within and between rivers throughout time, and (3) the identification of the
102 potential control factors of the Pu activity concentrations and signatures using a simple statistical
103 approach. This represents to the best of our knowledge the first decadal time series of Pu analyses in

104 river sediment samples collected at the same locations within the Difficult-To-Return Zone since the
105 FDNPP accident.

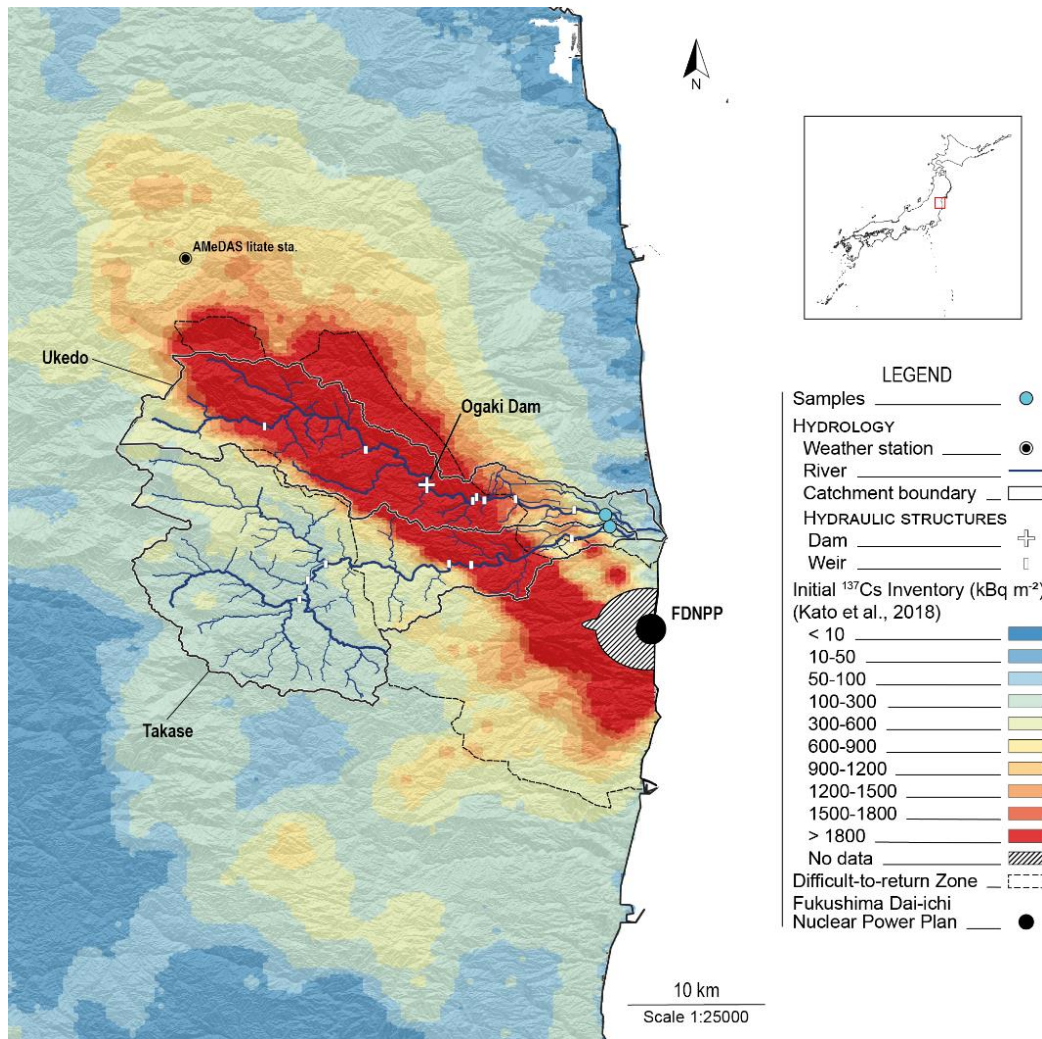
106 **Materials and methods**

107 **Study region**

108 This study was conducted in the Ukedo River catchment located to the north of the FDNPP and
109 draining a surface area of 534.8 km². It may be subdivided into two sub-catchments, i.e. Ukedo branch
110 (201.6 km²) and Takase branch (333.2 km²). The Ukedo and Takase catchments are mainly covered
111 with forests (84.1 – 88.5 % resp.) and paddy fields (0.6 – 0.8 % resp.), other cropland (12.1 - 9.2 % resp.),
112 urban areas (1.7 – 0.6 % resp). This catchment drains the surfaces that received the highest levels of
113 radiocesium (¹³⁴⁺¹³⁷Cs) fallout in March 2011, although very heterogeneous radiocesium contamination
114 levels are found across the catchment along the Ukedo and Takase rivers, where they range from 50
115 kBq.kg⁻¹ to 1800 kBq.kg⁻¹ (**Fig. 1.**) (JAXA, 2022; Kato and Onda, 2018). The upper part of these
116 catchments is currently under decontamination, while the lower part has been fully decontaminated
117 since March 2017 (Ministry of Environment, 2018).

118 **Samples**

119 The twenty-two samples investigated in the current research were collected in the downstream
120 sections of the Ukedo (37.496692°N; 141.001488°E) and Takase (37.490175°N; 141.004735°E) rivers,
121 just before their confluence (**Fig. 1.**). These lag deposit samples were comprised of fine particulate
122 material that settled on channel banks, inset benches and floodplains during the falling limb of the last
123 significant hydro-sedimentary event (Evrard et al., 2021). Sampling occurred bi-annually (i.e. in autumn
124 after the typhoon season and in spring, after the snowmelt run-off) between May 2013 and November
125 2016. Then, the campaigns occurred only after the typhoon season late in October or early in
126 November between 2017 and 2020 (Table S1) (Evrard et al., 2021). Samples were dried in an oven at
127 40°C for 72h, sieved to 2 mm, grounded to a fine powder using an agate mortar and stored at room
128 temperature after preparation. This <2 mm fraction was used for analysis.



129
 130 **Fig. 1.** Background ^{137}Cs level map in soils of the Fukushima Prefecture after (Kato and Onda, 2018)
 131 with location of the Fukushima Dai-Ichi Nuclear Power Plant, the investigated river catchments, the
 132 location of the sediment lag deposit samples (blue dot), and the Difficult-to-return Zone.

133 **Gamma spectrometry**

134 Radionuclide activities (^{134}Cs , ^{137}Cs) in sediment samples were determined by gamma
 135 spectrometry using HPGe detectors. All samples (10 to 15 g of material depending on the density) were
 136 pressed into 15mL polyethylene Petri dish containers for analysis. Counting time of sediment samples
 137 varied between 5 and 8×10^4 s, and activities were expressed in Bq per kg of dry weight ($\text{Bq}\cdot\text{kg}^{-1}$) and
 138 decay-corrected to January 2021. Quality assurance was conducted using certified International
 139 Atomic Energy Agency (IAEA) reference materials (i.e. IAEA-444, IAEA-375) as well as a multi-gamma
 140 resin produced by the IRSN (Institut de Radioprotection et de Sûreté Nucléaire, France), with elevated
 141 ^{137}Cs ($160 \text{ kBq}\cdot\text{kg}^{-1}$; reference date: 7 January 2011) and ^{134}Cs activities ($330 \text{ kBq}\cdot\text{kg}^{-1}$) prepared in the
 142 same containers as the samples.

143 **Samples chemical preparation and purification for plutonium analysis**

144 Samples were divided into 4 analytical batches containing 5 to 8 samples and 3 analytical blanks.
145 For each sample, ~5 gram-aliquots were transferred in Pyrex® beakers covered with watch glasses and
146 carbonized at 450°C for 12 hours in an electric furnace to decompose organic matter. After cooling,
147 samples were transferred in 60 mL Savillex® beakers by adding HNO₃ (10 mL) and H₂O (5 mL, 3 times),
148 and an evaporation to dryness was carried out. A first leaching on hot plate (150°C) with HNO₃ (20 mL)
149 and evaporation to dryness was performed. After cooling, a limited amount of ²⁴⁴Pu (~100 fg) was
150 added to the samples as isotopic dilution tracer for plutonium quantitative analysis. The ²⁴⁴Pu tracer
151 was used because this isotope was not found in the samples and therefore allows for plutonium
152 quantification. The other steps of separation and purification of plutonium followed the method used
153 by Jaegler et al. (2019, 2018).

154 **ICP-MS measurements**

155 Pu isotopic composition, activity concentration and concentrations were measured with a multi-
156 collection ICP-MS (“Neptune Plus”, Thermo-Fisher, Bremen, Germany). Instrumental settings used to
157 perform the isotopic plutonium measurements were the same as those detailed by Jaegler et al. (2019,
158 2018). The measured isotopes are ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu and ²⁴²Pu. All plutonium activity concentrations
159 and isotopic ratios presented in the current research were corrected by the average of analytical blanks
160 for each batch. The validity of the analytical measurements and the correction method were verified
161 by analysing the IAEA-385 certified material for the ²⁴⁰Pu/²³⁹Pu ratio. The certified value is 0.168 (±
162 0.016) and the measured value is 0.168 (± 0.011) with a confidence level of 95 % for each value. During
163 the analyses, the blank values varied between 4.0 (± 0.8) fg and 12.1 (± 8.6) fg and the chemical
164 efficiency between 26 % (± 3 %) and 100 % (± 2 %) (Detailed in Table S1).

165 **Organic matter property measurements**

166 Both total carbon and organic carbon fractions, from the bulk sediment were analysed in all
167 samples. Approximately 15 to 20 mg of sediment was weighed in tin cups for analysis (with a precision
168 of 1 µg). The sample was combusted in an elementary analyser (FlashEA1112, Thermo Fisher Scientific),
169 and the carbon content determined using the Eager software. A standard was inserted every 10
170 samples. The inorganic carbon content in the bulk sediment was calculated by assuming that mineral
171 carbon exists only as CaCO₃. The results are reported in %weight of carbonate/bulk sediment and
172 in %weight of organic carbon/bulk sediment.

173 **Carbon stable isotopic signatures**

174 Analysis was performed online using a continuous flow EA-IRMS coupling, i.e. a FlashEA1112
175 Elemental Analyser coupled to a ThermoFinnigan Delta+XP Isotope-Ratio Mass Spectrometer. Three in-

176 house standards (Hobo5 sediment - $\delta^{13}\text{C} = -13.4 \text{ ‰}$, oxalic acid 2 - $\delta^{13}\text{C} = -16.7 \text{ ‰}$, and GCL - $\delta^{13}\text{C} =$
177 -26.7 ‰) were inserted every five samples. Each in-house standard was regularly checked against
178 international standards. The results are reported in the δ notation: $\delta^{13}\text{C} = (R_{\text{sample}}/R_{\text{standard}} - 1)$, where
179 R_{sample} and R_{standard} are the $^{13}\text{C}/^{12}\text{C}$ ratios of the sample and the international standard, Vienna Pee Dee
180 Bee (VPDB), respectively.

181 Particle size analyses

182 Particle size analyses were performed on all sediment samples by laser granulometry
183 (Mastersizer[®] 3000, 194 Malvern Instruments, Ltd., UK) This technology provides the advantage of
184 providing data on the particle size distribution between 0.01 and 3500 μm and the corresponding
185 particle size distribution at 10 % (D10), the median particle size (D50), and at 90 % (D90). The Specific
186 Surface Area (SSA) was also determined during the analysis assuming that the particles are spherical.

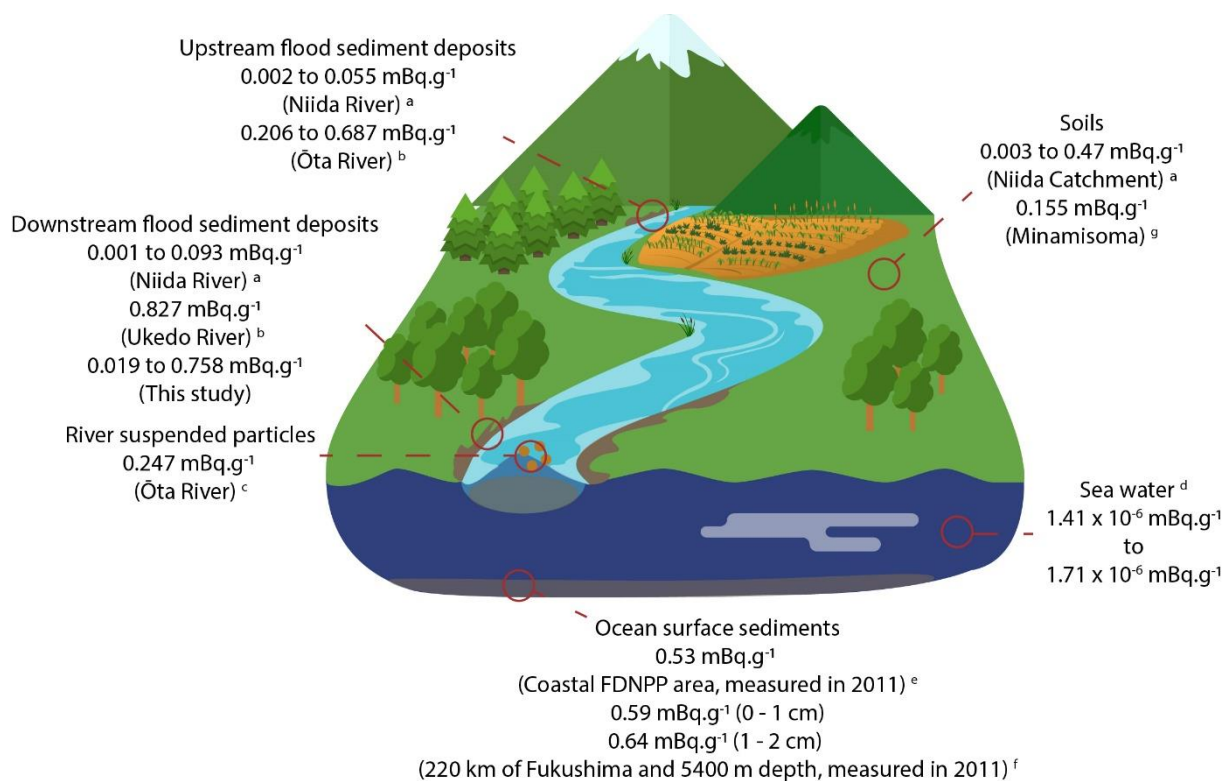
187 Statistical treatment

188 In order to better understand the distribution and control factors of Pu in each river, correlations
189 with other radionuclide concentrations and sediment properties were assessed. The Pearson (linear)
190 and Spearman (non-linear) correlations were calculated using the *cor* function from *stats* package (R
191 Core Team, 2021). Significance of correlations was assessed using the *cor.mtest* function from *corrplot*
192 package (Wei and Simko, 2021) for each type of correlation. Only correlations with a significance test
193 p-value below $\alpha = 0.05$ were considered as significant and were retained. When both linear and non-
194 linear correlations were significant, only the correlation with the highest value was retained.
195 Correlations with an absolute value above 0.50 were represented with a correlogram.

196 Results and discussion

197 Changes in Pu activity concentrations in river sediment from 2013 to 2020

198 Pu activity concentrations were found to vary from 0.031 to 0.758 mBq.g^{-1} in the Ukedo River
199 sediment and from 0.015 to 0.138 mBq.g^{-1} in the Takase River sediment (Table 1). Globally, the
200 plutonium activity concentrations were higher in the Ukedo River (mean value: 0.173 mBq.g^{-1}) than in
201 the Takase River (mean value: 0.049 mBq.g^{-1}). However, a global decrease in concentrations was
202 observed with time with some sporadic increases induced by typhoons. Several studies have quantified
203 the Pu activity concentrations in different environments, all along the entire continent-to-ocean
204 continuum (**Fig. 2**).



205
 206 **Fig. 2.** Sketch illustrating the Pu activity concentration variations (all values were decay-corrected to
 207 March 2021) along the continent-to-ocean continuum. **a.** (Jaegler et al., 2018) ; **b.** (Evrard et al., 2014) ;
 208 **c.** (Cao et al., 2016) ; **d.** (Men et al., 2018) ; **e.** (Oikawa et al., 2015) ; **f.** (Bu et al., 2015b) ; **g.** (Schneider
 209 et al., 2013).

210
 211 Previous studies indicate a strong decreasing activity concentration gradient from the upstream
 212 sources in the soils to the marine sediment where contaminated material deposits (Bu et al., 2015b;
 213 Cao et al., 2016; Evrard et al., 2014; Jaegler et al., 2018; Men et al., 2018; Oikawa et al., 2015; Schneider
 214 et al., 2013). On the continent (soils and flood sediment deposits), activity concentrations up to ~1.5
 215 millibecquerel per gram (from 1.56 to 0.015 mBq.g⁻¹) were measured with a global decrease over time
 216 (0.007 in 2011 in Niida sediment to 0.001 in 2020 in Takase sediment). In contrast, the lowest activity
 217 concentrations, in the nanobecquerel per gram range ((1.41 - 1.71) x 10⁻⁶ mBq.g⁻¹), were obtained in
 218 seawater. Ocean surface sediment show intermediate activity concentrations between 0.53 mBq.g⁻¹
 219 and 0.64 mBq.g⁻¹. Lower plutonium activity concentrations are observed in the marine environment,
 220 probably due to the dilution of particles induced by the strong Kuroshio current (Buesseler et al., 2015;
 221 Men et al., 2018). In these studies, a similar decrease and dilution effect of ¹³⁷Cs from the Ukedo River
 222 and Takase River was observed in the marine environment, and it was found that the total contribution
 223 of the ¹³⁷Cs from these rivers represented only 7 % of the total annual inventory supplied to seabed
 224 sediments (Misonou et al., 2022). Therefore, it is expected that a similar contribution of plutonium

225 may be supplied by these rivers to marine environments but its source is not necessarily identical to
 226 the cesium one because a strong contribution from global fallout is observed.

227 **Changes in plutonium isotopic signatures in river sediment**

228 In this study, we focused the interpretation on the $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio, because ^{241}Pu and
 229 ^{242}Pu are below the detection limits (concentrations presented in Table S2). This represents a major
 230 change compared to earlier publications (Evrard et al., 2014; Jaegler et al., 2018) that detected ^{241}Pu
 231 and demonstrated thereby a significant supply of Pu from the FDNPP accident. Similarly as for ^{241}Pu ,
 232 ^{242}Pu is not used for the interpretation in the current research, as this isotope was systematically found
 233 below the analytical detection limits (around 0.1 fg for Pu isotopes) whereas it had been detected by
 234 Jaegler et al. (2018). The Ukedo lag deposit referred to as FEL385 sampled in May 2013 was also
 235 analysed by Evrard et al. (2014) and the value found in the current research (0.181 ± 0.006) remains
 236 very close and not significantly different from that found in 2014 (0.172 ± 0.012). In the current
 237 research, the $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratios were measured for all Ukedo and Takase flood sediment
 238 deposit samples. Results are provided in **Table 1**, and the temporal evolution of this isotopic ratio is
 239 shown in **Fig. 2.A.** for the Ukedo River and in **Fig. 2.B.** for the Takase River.

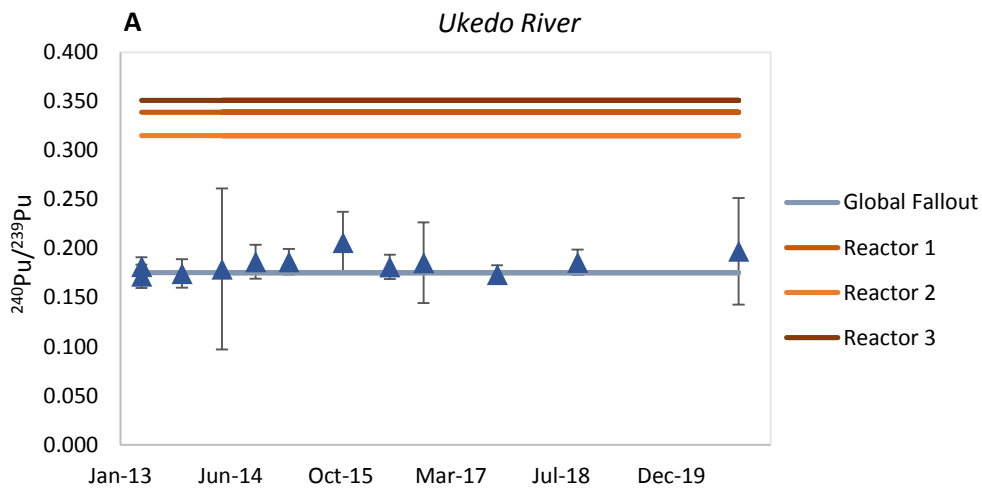
240
 241 **Table 1:** Plutonium activity concentrations and $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratios measured in lag deposit
 242 samples collected in the Ukedo River and Takaser River from 2013 to 2020. The grey cell corresponds
 243 to the value obtained on a sample already analysed by a previous study. All uncertainties are combined
 244 expanded uncertainties with a confidence level of 95 %.

Sample ID	Sampling date	Location	$^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio	$^{239+240}\text{Pu}$ activity concentration ($\text{mBq}\cdot\text{g}^{-1}$)
FEL_383	09/05/2013	Ukedo	0.182 ± 0.010	0.229
FEL_385	09/05/2013		0.172 ± 0.012	0.758
FEL385 Evrard et al. (2014)	09/05/2013		0.181 ± 0.006	0.827
FEL_519	02/11/2013		0.175 ± 0.014	0.353
FEL_616	10/05/2014		0.179 ± 0.024	0.066
FEL_648	30/10/2014		0.187 ± 0.017	0.114
FEL_762	26/03/2015		0.187 ± 0.013	0.144
FEL_932	05/11/2015		0.206 ± 0.031	0.025
FEL_1004	23/06/2016		0.181 ± 0.012	0.119
FEL_1043	02/11/2016		0.186 ± 0.041	0.031
FEL_1322	25/10/2017		0.174 ± 0.009	0.087
FEL_1416	31/10/2018		0.186 ± 0.013	0.104
FEL_1521	30/10/2020		0.197 ± 0.054	0.045
FEL_615	05/10/2014	Takase	0.168 ± 0.031	0.033

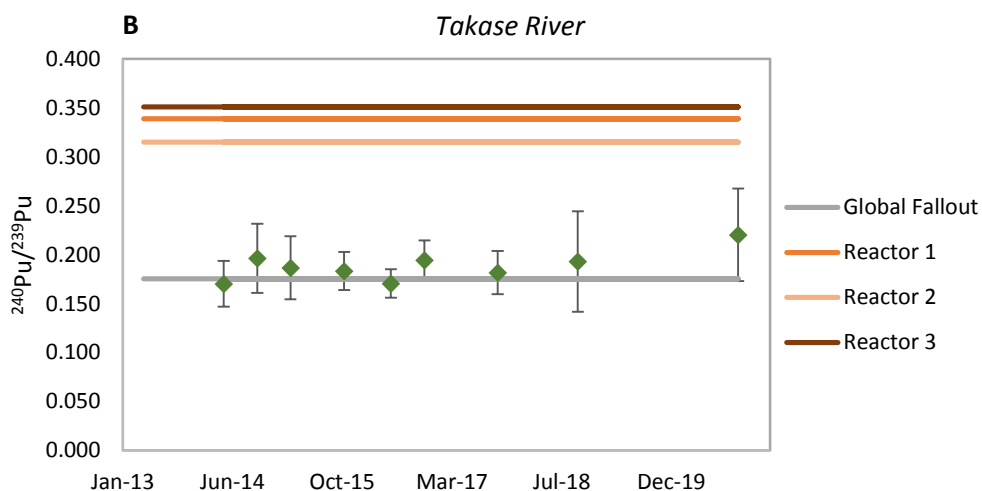
FEL_647	30/10/2014	0.196 ± 0.035	0.034
FEL_761	26/03/2015	0.187 ± 0.032	0.031
FEL_931	05/11/2015	0.183 ± 0.019	0.138
FEL_1003	23/06/2016	0.171 ± 0.015	0.115
FEL_1042	02/11/2016	0.194 ± 0.020	0.030
FEL_1321	25/10/2017	0.182 ± 0.022	0.025
FEL_1417	31/10/2018	0.193 ± 0.051	0.024
FEL_1522	30/10/2020	0.220 ± 0.047	0.015

245

246 The $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio and its changes with time provide information on the origin and fate
 247 of plutonium found in the Ukedo and Takase rivers. The $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio values may be
 248 compared to those of global fallout in the northern hemisphere as determined by Kelley et al. (1999)
 249 (0.180 (± 0.014)), and to the signatures of the FDNPP reactors estimated by the model simulations
 250 conducted by Nishihara et al. (2012) (reactor 1 : 0.339 ; reactor 2 : 0.315 ; reactor 3 : 0.351) (**Fig. 3**).
 251 For the Ukedo lag deposits, the $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio ranged between 0.172 (± 0.012) and 0.206 (±
 252 0.031) (see **Fig. 3.A**). For the Takase lag deposits, the $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio ranged between 0.168
 253 (± 0.031) and 0.220 (± 0.047) (see **Fig. 3.B**).



254



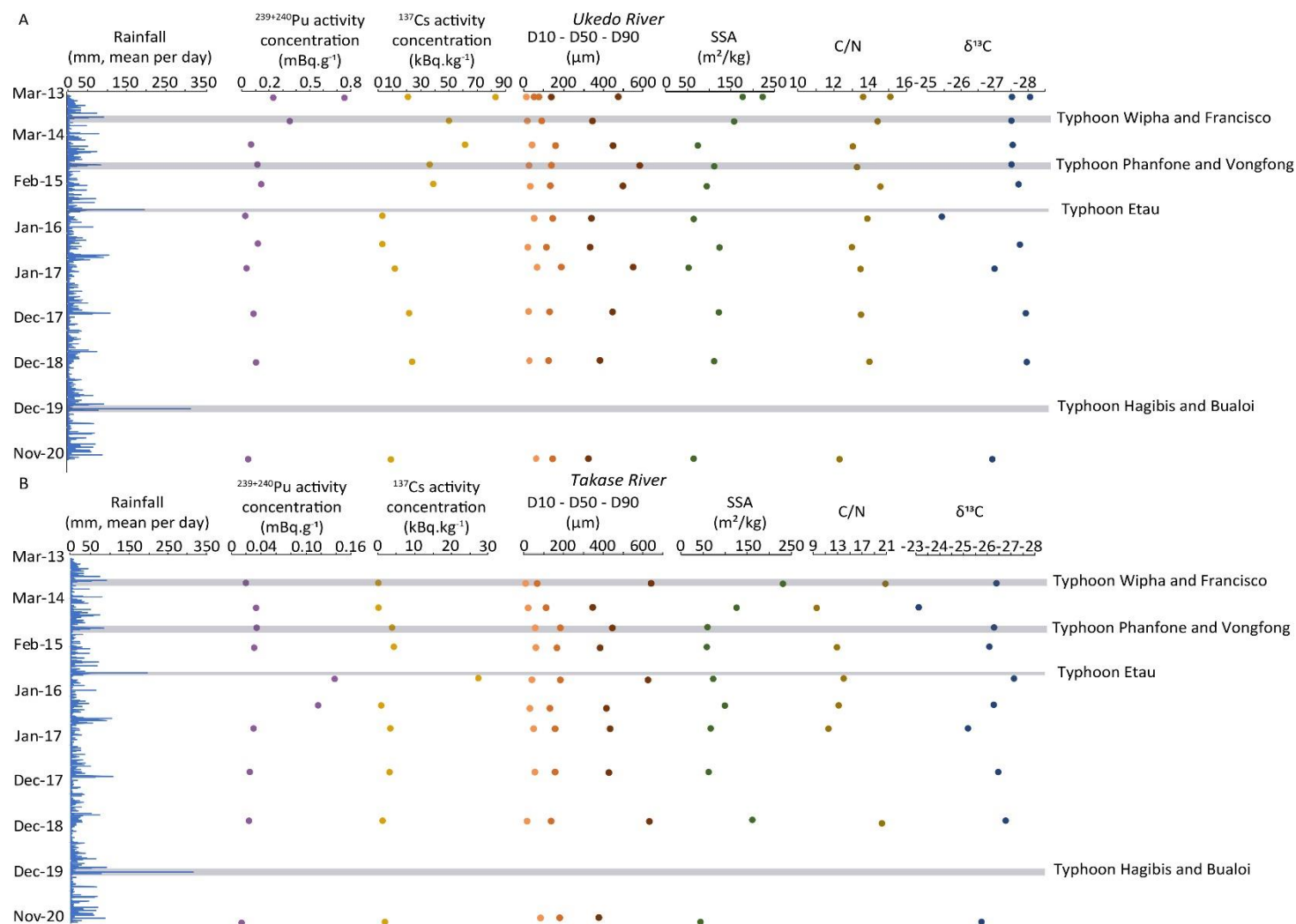
255
 256 **Fig. 3.** Evolution of $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratios between 2013 – 2020 in the (A) Ukedo River sediment
 257 and (B) the Takase River sediment. The grey line corresponds to the mean of the global fallout signature
 258 in the northern hemisphere (Kelley et al., 1999). The orange lines correspond to the estimated source
 259 values of the FDNPP reactors (Nishihara et al., 2012). All uncertainties are combined expanded
 260 uncertainties with a confidence level of 95%.

261
 262 $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratios measured in this study are not significantly different from those
 263 associated with the global fallout. Values of this isotopic ratio in Japanese soils, sediments and lag
 264 deposits were reported in several publications (Evrard et al., 2014; Jaegler et al., 2019, 2018; Schneider
 265 et al., 2017; Steinhauser et al., 2015; Yang et al., 2017; Zheng et al., 2013, 2012), and, for those samples
 266 collected until 2013, some of the measured values were much closer to the Fukushima Pu source
 267 signature (Evrard et al., 2014; Jaegler et al., 2019; Schneider et al., 2017; Yang et al., 2017). Yang et al.
 268 (2017) analysed the $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio in soils sampled in 2011 in the Fukushima Prefecture with
 269 values ranging between 0.245 (± 0.014) and 0.312 (± 0.044). Evrard et al. (2014) collected lag deposits
 270 from Niida, Ota and Ukedo Rivers between November 2011 and May 2013, and obtained $^{240}\text{Pu}/^{239}\text{Pu}$
 271 isotopic ratios between 0.150 (± 0.005) and 0.281 (± 0.012). Jaegler et al. (2019) analysed soil samples
 272 collected between November 2011 and November 2012 in the Mano River catchment located further
 273 to the North from FDNPP compared to the Ukedo River catchment. $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratios values
 274 in these samples ranged between 0.177 (± 0.014) and 0.285 (± 0.049). This isotopic ratio was also
 275 investigated by Steinhauser et al. (2015) in soil samples collected in Minami-soma on September 7,
 276 2014, and values ranged between 0.160 and 0.220 (Figure S3). Then, in contrast, values obtained on
 277 environmental samples collected after 2013 remained in the range of signatures attributed to the
 278 global fallout. In the current research, these values ranged between 0.168 (± 0.031) and 0.220 (± 0.047).
 279 Schneider et al. (2017) studied soil samples from Minami-soma City and Okuma Town collected in June

280 2013 and May 2015. $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratios ranged between 0.18 and 0.48 (± 0.15) in 2013, whereas
281 the samples taken in 2015 showed rather constant values close to 0.18. Overall, the current results
282 show that (1) the global fallout provides the main source of Pu to sediment whatever the sampling
283 year after the FDNPP accident, and (2) the actual negligible contribution of the FDNPP accident of Pu to
284 the sediment. Accordingly, the current research provides a useful complement to the conclusions
285 made by Jaegler et al. (2018) who estimated very low FDNPP contributions (ranging between 1.2 % (\pm
286 1.4 %) and 10.4 % (± 3.1 %)) to plutonium supply in sediment of the Niida River.

287 Evolution of sediment properties from 2013 to 2020

288 The temporal evolution of rainfall and some sediment properties such as ^{137}Cs activity
289 concentrations, median particle size (D50), SSA, C/N ratios and $\delta^{13}\text{C}$ values was considered in order to
290 improve our understanding of the radionuclide control factors in sediment (All the data are available
291 online: <https://doi.org/10.5281/zenodo.7233904> (Diacre et al., 2022)). Sediment properties are
292 provided in **Fig. 4**, and their temporal evolution is shown in **Fig. 4.A.** for the Ukedo River and in **Fig. 4.B.**
293 for the Takase River. Pu concentrations and the $^{239+240}\text{Pu}/^{137}\text{Cs}$ ratio are available in Table S4.



294

295 **Fig. 4.** ^{137}Cs activity concentrations, D10 - D50 – D90, SSA, C/N and $\delta^{13}\text{C}$ analysed in this study. **A.** Results for the Ukedo River sediment, and **B.** for the Takase
 296 River sediment. Main typhoons that occurred during this period are reported (Evrard et al., 2020; Laceby et al., 2016a). The mean daily rainfall is plotted for
 297 the station Iitate (37.665N, 140.727E) (Japanese Meteorological Agency, 2021).

298 First, the ^{137}Cs activities show a global decrease and different patterns of temporal variations
299 between the two rivers. In the case of the Ukedo River, ^{137}Cs activities ranged between 2.5 kBq.kg^{-1}
300 and 83.2 kBq.kg^{-1} , with the highest ^{137}Cs activities recorded in Spring in 2013 (**Fig. 4.A.**). The evolution
301 of the ^{137}Cs activity in lag deposits collected at this location was not impacted by the occurrence of
302 heavy rainfall events (typhoons and tropical storms). In contrast, the ^{137}Cs activities measured in the
303 Takase River were lower than those found in the Ukedo River sediment, as they ranged between 0.1
304 kBq.kg^{-1} and 27.5 kBq.kg^{-1} (**Fig. 4.B.**), with the highest value observed after Typhoon Etau in 2015. The
305 difference in ^{137}Cs activity levels and variations between both rivers may be related to (1) the different
306 levels of initial contamination in their respective drainage areas, as observed in **Fig. 1**, and (2) a
307 difference in decontamination phases in the watershed, which may controls inputs especially during
308 strong variations (Feng et al., 2022; Onda et al., 2020). The absence of correlation between rainfall
309 depths and ^{137}Cs activities in sediment in the Ukedo River is likely related to the presence of Ogaki Dam
310 18 km upstream of the sampling location (Yamada et al., 2015). It was estimated that only 10% of the
311 sediment transported by the Ukedo River upstream of Ogaki Dam reaches the river outlet (Kitamura
312 et al., 2014). However, according to the literature, the overall decrease in ^{137}Cs shows a similar trend
313 upstream and downstream of the dam (Feng et al., 2022). For the Ukedo River and Takase River, the
314 ^{137}Cs activity evolution shows the same trend as for the Pu concentration (**Fig. 4**).

315 Then, granulometric parameters were also considered to characterize the sediment properties
316 carrying the radionuclides of interest. In both rivers, the same inter-event variations of the D50 were
317 observed, and similar mean particle size values of $126 \mu\text{m}$ and $146 \mu\text{m}$ were found for both Ukedo and
318 Takase Rivers, respectively (**Fig. 4**). These mean D50 values show that the flood deposit sediments are
319 mainly composed of fine sands, although the Ukedo River sediment contained a higher proportion of
320 fine minerals (i.e. silt and clay minerals) (Figure S5). Logically, a similar interpretation can be made for
321 the SSA parameter, with mean values of 115 and $98 \text{ m}^2/\text{kg}$ for the Ukedo River and Takase River,
322 respectively (**Fig. 4**). An anti-correlation was observed between the D50 and the SSA, with the latter
323 being correlated to the finest particles, which likely explains why the lowest value of this parameter
324 was recorded during the heaviest rainfall events until 2015. Typhoon Etau in 2015 likely flushed most
325 of the contaminated material stored in the river channels or in its close vicinity since 2011 as
326 demonstrated in previous research (e.g. Chartin et al. (2017); Evrard et al. (2021)).

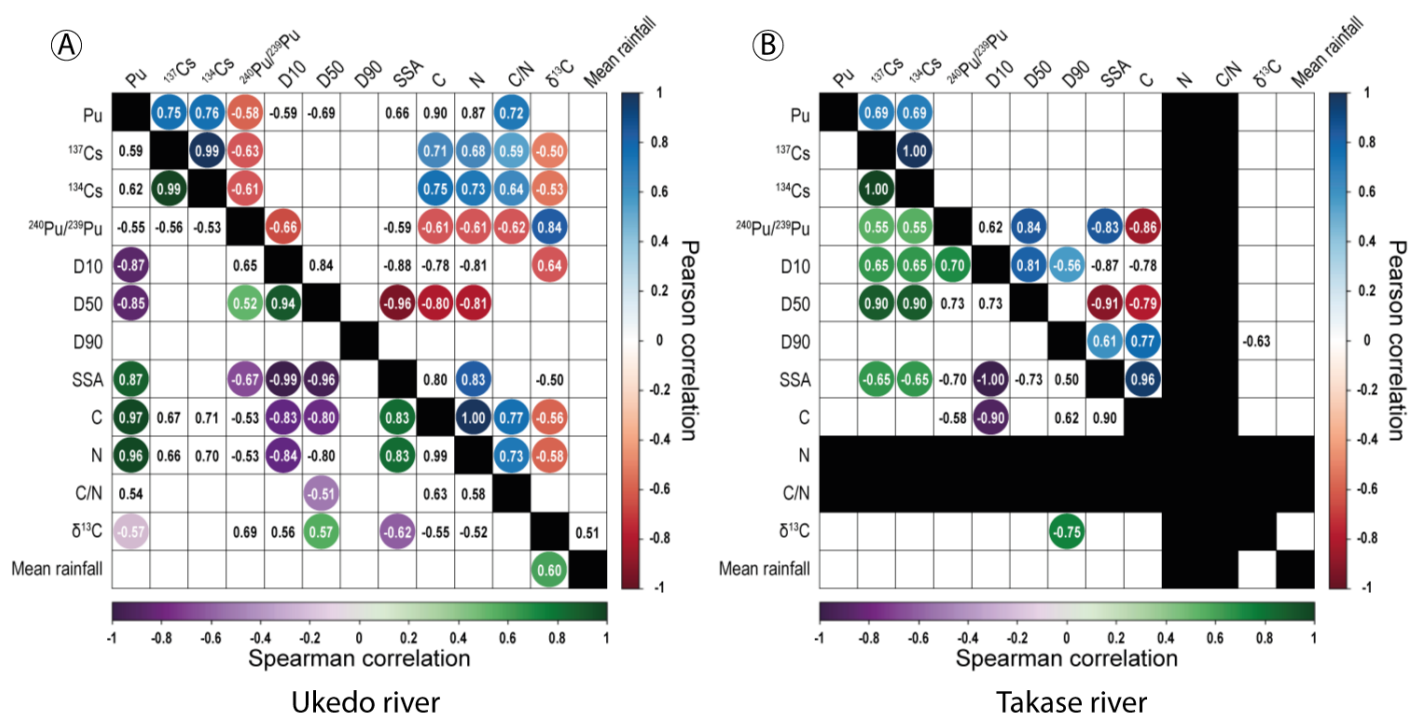
327 Finally, the $\delta^{13}\text{C}$ and the C/N parameters allowed identifying the main source of the organic matter
328 of sediment transiting both rivers. A $\delta^{13}\text{C}$ difference is observed between the Ukedo and Takase rivers
329 although their catchment drainage areas have similar land use distributions, which likely indicates that
330 different sediment sources are mobilised in both catchments (Nakanishi et al., 2021). The entire $\delta^{13}\text{C}$
331 value ranges observed over the period 2013–2020 are comprised between -25.42 ‰ and -28.07 ‰ for
332 the Ukedo River, compared to an interval comprised between -23.10 ‰ and -27.11 ‰ for the Takase

333 River (**Fig. 4**). These $\delta^{13}\text{C}$ values show that the organic matter found in sediment mainly originates from
334 terrestrial plants and particularly from C_3 plants (Lamb et al., 2006). Based on sediment source data
335 from neighbouring catchments, material fluxes from the Ukedo River are therefore mainly supplied by
336 soils under forest ($\delta^{13}\text{C}$ values ranging between -28 and -26 ‰), while those from Takase River are
337 likely mainly delivered by cultivated soils or subsoils ($\delta^{13}\text{C}$ values ranging between -26.5 and -22 ‰)
338 (Lacey et al., 2016b). This diagnosis is further supported by the analysis of C/N ratios for both the
339 Ukedo and Takase river sediment, with mean respective values of 13.4 and 13.7 (**Fig. 4**). The mean
340 value is representative for the only source for the Ukedo River, while for the Takase River values ranged
341 between 9.5 and 20.8, which is likely indicative of the occurrence of two main sediment sources
342 (cultivated soils or subsoils). The C/N values exceeding 12 correspond to organic matter of plants
343 composed of lignin and cellulose, as cedar, pine, and oak characteristic of forest soils (Lamb et al.,
344 2006). This source clearly dominated the contributions of the Ukedo River organic matter inputs, while
345 it partly contributed to those of the Takase River. In the latter, the rest of the material was supplied by
346 sources with C/N values ranging between 10 and 12 and corresponding to vascular plant sources, as
347 cultivated plants, as observed in the Takase River sediment collected in May 2014 and November 2016.
348 The interpretations made based on C/N values are in agreement with those made based on $\delta^{13}\text{C}$ values.

349 Statistical approach to determine the control factors of the Pu concentration evolution

350 In the previous section, we observed that Pu concentration variations tend to follow that of Cs
351 although their emission origins are distinct. This demonstrates the same mode of transport of
352 radionuclides regardless of their source. Their input to the outlet is therefore mainly controlled by
353 fluctuations in precipitation i.e. runoff, erosion. To complement this finding and derive further
354 information about the behaviour of these radionuclides, a statistical approach plotting the Pearson
355 correlation coefficients calculated between radionuclide concentrations and other sediment
356 properties was conducted under the form of a heatmap.

357 The results show that the two investigated rivers show differences in the dynamics of Pu
358 transportation as inferred from the correlations between the tested parameters (**Fig. 5**).



360 **Fig. 5.** Significant Pearson (linear; upper part) and Spearman (non-linear; lower part) correlation
 361 matrices between radionuclide and sediment properties above 0.5, for (A) Ukedo river ($n = 12$) and (B)
 362 Takase river ($n = 10$). The highest correlation and anti-correlation value between both modes (i.e.,
 363 linear and non-linear) are indicated with a coloured circle.

364

365 In the case of the Ukedo River, Pu concentrations showed significant and high positive linear
 366 correlations with ^{137}Cs and ^{134}Cs activities (r of 0.75 and 0.76 resp.) and C/N ratio (r of 0.72).
 367 Furthermore, the analysis showed high non-linear correlations with sediment properties with, on the
 368 one hand, anti-correlations with grain size parameters (D10: $r = -0.87$; D50: $r = -0.87$) and, on the
 369 other hand, positive correlations with SSA ($r = 0.87$) and with organic matter parameters (C: $r = 0.97$;
 370 N: $r = 0.95$) (Fig. 5.A). These results suggest that Pu was transported in association with organic matter
 371 or fine minerals (high anti-correlation with D10 and D50), while the high correlation with SSA suggests
 372 its strong affinity for clay materials, as it was demonstrated for ^{137}Cs (Feng et al., 2022; Hirose, 2022;
 373 Onda et al., 2020). Other radionuclides (i.e. ^{137}Cs and ^{134}Cs), showed the same trend in their
 374 correlations, although only linear correlations were observed with them. Based on the literature, ^{137}Cs
 375 is mainly sorbed onto the surface of minerals (i.e. mainly clays minerals), and its transport also shows
 376 some level of land use dependency (Feng et al., 2022; Hirose, 2022; Onda et al., 2020). The non-linear
 377 relationship of Pu with grain size and organic matter, in contrast to ^{137}Cs and ^{134}Cs , suggests another
 378 way of transport in addition to sorption. According to the literature, the $^{239+240}\text{Pu}$ activity concentration
 379 increases in the lowest particle size fractions as well as ^{137}Cs . However, in contrast to ^{137}Cs , $^{239+240}\text{Pu}$ is

380 known to be associated not only with clays but also with organic matter and oxides (for
381 example through organic and inorganic colloids. Furthermore, their stability in solution increases
382 when they are multiphase (organic + inorganic)) (Alewell et al., 2017; Kersting, 2013; Romanenko and
383 Lujanienė, 2023; Xu et al., 2017). Finally, Romanenko and Lujanienė, (2023) showed that Pu(IV) reacts
384 actively with colloids and suspended solids while Pu(V) reacts with carbonate particles. In the case of
385 this study, it is probably Pu(IV) since it is associated with fine particles (clays, silts, organic matter). The
386 presence of the Ogaki dam upstream (Kitamura et al., 2014) may impact Pu transport. Therefore, the
387 transport of material downstream is likely controlled by the outflow discharge from the dam with only
388 10% of the inflow being transferred downstream (Nakanishi et al., 2021). Moreover, as it was shown
389 that sands are deposited preferentially upstream of Ogaki Dam, silt deposits would occur across the
390 reservoir and clays would remain in suspension, deposit nearby the dam and part of this fine material
391 may eventually be transferred to the downstream river section where the samples analysed in the
392 current research were collected, which highlights the major role of the dam (Yamada et al., 2015).

393 Similarly, Pu concentrations in the Takase River sediment were found to be highly linearly
394 correlated to ^{137}Cs and ^{134}Cs activities (**Fig. 5.B**) ($r = 0.69$). However, in contrast with what was observed
395 for the Ukedo River, no sediment property was found to be correlated with Pu concentrations. In
396 addition, ^{137}Cs and ^{134}Cs activities were showed to be non-linearly correlated with sediment properties
397 while they were linearly correlated with these properties in the Ukedo River.

398 Although these two rivers are geographically very close, they showed different sediment dynamics
399 and characteristics. Overall, the Ukedo River flood deposit sediment contained finer particles (silts,
400 clays, fine sands) than in Takase River (Figure S5). These different properties with coarser particles
401 transiting in the Takase River compared to the Ukedo River is likely related to the presence of Ogaki
402 Dam along the latter, leading to a preferential transport of the finest fractions from upper to lower
403 catchment parts. In contrast, the flow of the Takase River is not regulated and the river drains less
404 contaminated soils ($\pm 47\%$ of the catchment surface), which likely explains why the statistical analysis
405 led to less clear results for Pu in this second river.

406 **Conclusions**

407 In the current research, Pu concentrations were measured in flood deposit sediments collected in
408 lower sections of the Ukedo and the Takase rivers from 2013 to 2020. For both Ukedo and Takase
409 rivers, a global decrease in Pu concentrations (following that of ^{137}Cs activity concentrations) was
410 observed throughout time possibly in response to the abandonment of the region after the accident
411 and the progressive exhaustion of the easily mobilized contaminated particles stored in the main river
412 channels and in their vicinity after 2011 and until typhoon Etou in 2015. This situation allowed
413 vegetation to re-grow, which protected the contaminated soils from surface erosion, and subsurface

414 erosion mobilized instead the deeper soil layers that were not contaminated by the FDNPP accident or
415 forest soils that are mainly found on steeper slopes in the case of the Ukedo River. In addition to the
416 quantification of Pu concentrations, the determination of Pu isotopic ratios provided additional
417 information on this actinide source. Contrary to studies conducted early after the FDNPP accident on
418 environmental samples collected until 2013, in the current research, flood deposit sediments showed
419 a $^{240}\text{Pu}/^{239}\text{Pu}$ signature (ranging between 0.168 and 0.220) coinciding with that of the global fallout
420 (around 0.180 in the Northern hemisphere). Accordingly, in sediment collected between 2013 and
421 2020, the impact of the FDNPP fallout on sediment contamination with Pu had become negligible
422 despite the occurrence of multiple heavy rainfall events in the region during this period and the fact
423 that these rivers are located in the vicinity of FDNPP and drain a significant part of the Difficult-to-
424 Return Zone. Differences of the hydrosedimentary properties found in both rivers was also highlighted,
425 probably induced by the presence of the Ogaki dam on the Ukedo River, that highly impacted the
426 radionuclide transfer from the Difficult-to-Return zone to the Pacific Ocean.

427 Currently, decontamination work is underway in the area, and it would be interesting to investigate
428 variations in ^{137}Cs activities, Pu concentrations and sediment sources during this major land cover
429 change process. Indeed, a similar study could be conducted across the area after its reopening in 2023,
430 in order to detect potential changes, including those related to a potential Pu remobilization, after
431 remediation that may expose contaminated soil layers that were protected by vegetation regrowth
432 after land abandonment until the recent decontamination works.

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