

1 Variability of Fallout Radionuclides
2 (FRNs) in River Channels: Implications
3 for Sediment Tracing

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20 **Abstract**

21 Purpose: To assess the sources of variability of particulate FRNs (^7Be , $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs) in
22 river channels, the influence of sediment properties such as particle size distribution (PSD) and organic
23 matter (OM) on FRN distributions, and to discuss the implications for sediment tracing in rivers.

24 Methods: Suspended and channel bed sediment samples were collected in the River Avon
25 (Devon, UK) during five strategic surveys involving a wide variation of river flows, including flood
26 conditions. Particulate matter was analysed for ^7Be , $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs by gamma spectrometry, PSD by
27 laser diffraction, and organic constituents, total organic carbon (TOC) and total nitrogen (TN) by
28 elemental analysis.

29 Results: FRNs activity concentrations vary significantly both spatially, characterised by
30 changes in activity concentrations within and between locations, and temporally, with changes during
31 the storm hydrograph and between seasons. Variability was attributed to changes in sediment sources
32 and, on some occasions, to significant correlation of activity concentrations with sediment properties.
33 The results also highlighted the influence of changes in channel characteristics and the magnitude and
34 frequency of floods on FRN distributions.

35 Conclusion: In the context of sediment tracing, attention should be given to seasonal changes
36 in riverine conditions that have the potential to affect ^7Be and ^{137}Cs conservativeness. Application of
37 FRNs in sediment fingerprinting studies should be accompanied by appropriate temporal
38 characterisation of potential sediment sources. Finally, potential contribution of ^7Be -depleted sediment
39 from channel resuspension to the suspended load should also be considered.

40 **Keywords**

41 Radionuclides, Sediments, Channel bed, Gravel bed, Sediment tracing.

42 **Statements and declarations**

43 **Author contribution**

44 All authors contributed to the study conception and design. Material preparation and data
45 collection were carried out by Enrique Muñoz-Arcos and Richard Hartley. Data analyses were
46 performed by Enrique Muñoz-Arcos. The first draft of the manuscript was written by Enrique Muñoz-

47 Arcos and all authors commented on previous versions of the manuscript. All authors read and
48 approved the final manuscript.

49 **Competing interests**

50 The authors have no competing interests to declare that are relevant to the content of this
51 article.

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61 1 Introduction

62 Fine sediment plays a key role in the healthy functioning of river ecosystems as it provides
63 nutrients and connectivity throughout the river basin soil-sediment cascade (Fryirs 2013; Wohl 2015).
64 However, excess sediment supply in river corridors can have detrimental effects on water quality and
65 river ecology (Owens et al. 2005; Wohl 2015; Wharton et al. 2017; Owens 2020). Sediment also acts
66 as a vector for contaminants, namely radionuclides (Millward and Blake 2023), trace metals (Bravo-
67 Linares et al. 2024), pesticides (Gellis et al. 2017), polycyclic aromatic hydrocarbons (Froger et al. 2019;
68 Van Metre et al. 2022) and microplastics (Hurley et al. 2018; Gerolin et al. 2020). Improved
69 understanding of fine particle dynamics in riverine ecosystems, specifically in river channels, is critical
70 to inform targeted sediment remediation strategies and basin-wide sediment management practices to
71 tackle point and diffuse riverine pollution.

72 Fallout radionuclides have been widely used as particle tracers in aquatic systems (Wallbrink
73 and Murray 1993; Blake et al. 2002; Wilkinson et al. 2009; Du et al. 2011; Kaste and Baskaran 2011;
74 Matisoff 2014; Muñoz-Arcos et al. 2022). The basis for the use of FRNs as tracers is their strong affinity
75 with particles as shown by their high reported distribution coefficients, K_d , of the order of 10^4 - 10^5 L kg⁻¹
76 (Olsen et al. 1986; Hawley et al. 1986; Van Hoof and Andren 1989). The application of these
77 radionuclides has improved our understanding of sediment dynamics in river systems. For instance,
78 FRNs have been used to estimate sediment residence and storage times (Jweda et al. 2008; Fisher et
79 al. 2010; Gartner et al. 2012; Froger et al. 2018), resuspension rates (Fitzgerald et al. 2001; Jweda et
80 al. 2008) and the contribution of catchment erosion sources to riverine sediments (Hancock et al. 2014;
81 Evrard et al. 2016). However, recent literature assessments have identified several challenges and
82 research needs with regards to the application of FRNs as sediment tracers in river systems, including
83 consideration of the short-lived ⁷Be radionuclide (Taylor et al. 2013; Walling 2013; Muñoz-Arcos et al.
84 2022). Some of the identified challenges are related to the variability of FRNs in river channels and the
85 processes contributing to this variation. Highlighted challenges include, but are not restricted to: 1) the
86 effect of changing sediment properties such as PSD and OM on FRN distributions; 2) the effects of
87 activity concentration dilution due to contribution of FRN-depleted sediment sources, particularly
88 decreasing particulate ⁷Be activity concentrations; and 3) the contribution of direct fallout onto the river
89 channel. Sediment tracing applications using FRNs, such as sediment fingerprinting, have focused on
90 the assessment of the FRN conservativeness between potential sources and sediments by evaluating

91 differences in sediment properties (Estrany et al. 2016; Smith et al. 2018; Muñoz-Arcos et al. 2021).
92 However, little research has addressed the conservativeness of FRN tracers due to variations imposed
93 by in-channel physical and chemical processes.

94 The effects of PSD and OM on FRNs in sediment tracing studies have been widely discussed
95 in the literature (He and Walling 1996; Jweda et al. 2008; Fisher et al. 2010; Smith and Blake 2014;
96 Singleton et al. 2017; Laceby et al. 2017; Smith et al. 2018; Gaspar et al. 2022). To address the potential
97 effects on radionuclide activities due to enrichment or depletion by changes in PSD and/or OM, several
98 approaches have been applied such as sample fractionation (Blake et al. 2009), particle size/organic
99 matter correction factors (Collins et al. 1997; Koiter et al. 2018) and normalisation with ^{210}Pb or ^{228}Th
100 (Fisher et al. 2010; Foucher et al. 2015). However, it is not only overcorrection of FRN activity and
101 geochemical element concentrations that has been reported (Smith and Blake 2014; Koiter et al. 2018)
102 but also that the specific surface area (SSA) might not exert significant controls on FRN activity
103 concentrations. Rather, the presence and amount of acid-extractable grain coatings (comprised
104 predominantly of Fe, Mn, Al, and Ca) may be more significant in controlling FRN distributions in fluvial
105 sediments (Singleton et al. 2017). Moreover, recent research has reported that ^{137}Cs has increased
106 mobility in catchments with soils characterised with high OM content (Fan et al. 2014), and its mobility
107 in sediments may be enhanced due to weaker interaction of ^{137}Cs with clay minerals caused by the
108 blocking effect of the adsorbed humic substances onto fine particles (Suga et al. 2014; Takahashi et al.
109 2017). Beryllium-7, on the other hand, has been reported not to be controlled by grain size sorting
110 effects in well sorted sandy channel beds (Kaste et al. 2014). However, potential for mobility under
111 reducing conditions in sediment storage zones (Taylor et al. 2013) and scavenging by channel bed
112 particles from the water column during low flow conditions has been reported (Fisher et al. 2010; Kaste
113 et al. 2014). Consequently, an improved understanding of FRN variability, both spatially and temporally,
114 in river channel sediments is necessary.

115 This contribution aims to address the research gaps identified above through the following
116 specific objectives: 1) to assess the spatial and temporal variability of FRNs (^7Be , $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs) in
117 river channels (suspended and channel bed sediments); 2) to examine their relationship with sediment
118 properties i.e. PSD and OM (TOC and TN); and 3) to discuss the implications for sediment tracing
119 studies in light of our findings.

120 **2 Material and methods**

121 **2.1 Study site**

122 The River Avon (Devon, UK, Fig. 1) is a 40 km long gravel-bed river with a catchment area 110
123 km². The mean annual flow is 3.7 m³ s⁻¹ and is moderated by periodically managed discharges from an
124 upland reservoir upstream. The source of the River Avon is at 475 m above sea level on southern
125 Dartmoor, and the elevation in the middle and lower parts of the catchment ranges between 150 and
126 200 m above sea level. The geology of the catchment includes Carboniferous granitic intrusions, Middle
127 Devonian slates, and Early Devonian sandstones and argillaceous rock that dominate in the upper,
128 middle and lower part of the catchment, respectively (Wang et al. 2021). Land use in the upper
129 catchment is characterised by rough hillslope and grazing areas, whereas the middle and lower
130 catchment is dominated by mixed arable and pasture lands. The major land uses in the Avon catchment
131 are pasture, arable land, woodland, wet moorland and housing, which comprise 66, 18, 8, 5 and 3 % of
132 the catchment respectively (Wang et al. 2021).

133 **2.2 Sampling site selection and river monitoring**

134 A 5 km river reach was selected to study FRN variability in the middle part of the Avon
135 catchment. This was located above a flow monitoring station (Environment Agency, UK) and the Normal
136 Tidal Limit (NTL) near to the lower reach (Fig. 1b). Three established and submerged mid-channel bars
137 located in the upper, middle and lower reach of the river were selected as riverine geomorphological
138 features of interest because they are subjected to changes in sediment composition/dynamics reflecting
139 variations in streamflow, channel characteristics and sediment loads. An additional monitoring station
140 was set up in the upper reach to record river stage and turbidity to subsequently model river flow,
141 suspended sediment concentrations (SSCs) and suspended sediment loads (SSLs).

142 **2.3 Sampling and sample preparation**

143 Channel bed sediments (CBS, n = 106) were sampled using the stilling-well resuspension
144 method (Lambert and Walling 1988) during five field surveys and in response to flood events. A
145 bottomless bin was placed on the riverbed to isolate the resuspendable sediments from the river flow
146 and in three transects along submerged mid-channel bars (i.e. head, middle and tail, see Table S1.1)
147 to account for the spatial variability therein. Then, a battery-operated drill connected to a stainless-steel
148 helix was used to vigorously disturb the surface of the bar thereby facilitating resuspension of fine
149 particles. After resuspension, ~3 seconds of dwell time were allowed to let coarser grains settle before

150 sampling the fine fraction. The suspended sediments were then collected using a plastic jar and quickly
151 poured into acid rinsed 10 L HDPE containers.

152 Suspended sediments (SS, n = 26) were collected using time-integrated sediment traps
153 (Phillips et al. 2000) placed at each sampling point (i.e. close to each channel bar). Sediment traps were
154 usually emptied in the field on the same days as resuspension sampling. The trapped suspended solids
155 were carefully poured into an acid rinsed 10 L HDPE container. Using small volumes of river water,
156 remnants of particles were recovered from the trap and transferred to the sample container. The tubes
157 were rinsed with river water and replaced at the same location. Suspended sediments (n = 4) were also
158 collected (ca. hourly) during a single storm event at the upper river reach using an in-situ continuous-
159 flow centrifuge to assess the variability of FRN activity concentrations throughout the storm hydrograph.

160 Samples were stored at 4°C until further processing. Suspensions were allowed to settle
161 overnight and dewatered, after which the sediment slurry was centrifuged at 4000 RPM for 10 minutes
162 and the supernatant discarded. Particles were subsequently freeze-dried until complete dryness was
163 attained. Dried samples were disaggregated using a pestle and mortar and sieved using a clean
164 stainless-steel sieve to isolate the < 63 µm fraction. Samples were packed in plastic polyethylene bags
165 for further analyses.

166 **2.4 Sample analyses**

167 2.4.1 Particle size distribution

168 Subsamples (~ 1 g) of dried and sieved particles were transferred into 12 ml vials. The process
169 was repeated to prepare five replicates for every sample. Then, 2 – 3 ml of 6% Hydrogen Peroxide
170 (H₂O₂) was added to each vial and allowed to stand overnight. Samples were then placed in a water
171 bath at 80°C for 2 h and then removed and allowed to cool. An additional aliquot of 6% of H₂O₂ was
172 added for every sample to assess if any effervescence takes place. Samples were then placed into a
173 water bath at 80°C for an additional 2 – 4 hours to check for complete removal of organic matter and
174 then cooled at room temperature. Sodium hexametaphosphate was added to every sample to a
175 concentration of 0.1 % v/v to aid dispersion and avoid particle flocculation. Particle size distributions
176 were assessed using a Malvern Mastersizer-2000 laser diffraction particle size analyser with a wet
177 sample Hydro-G unit. PSD results were statistically checked for large variability between replicates. A
178 Relative Standard Deviation (RSD) of < 5% between selected percentiles (i.e. 5, 25, 50 -median, 75

179 and 95 percentiles) was defined as an acceptable result. The SSA was obtained from PSD data
180 assuming that particles are both spherical and nonporous.

181 2.4.2 Total Organic Carbon

182 Total Organic Carbon analysis was carried out on subsamples varying from 0.1 to 1.0 g of
183 sieved material depending on the amount of sample available. An aliquot of diluted HCl (0.5 M) was
184 added to each sample to ensure complete removal of carbonates. Subsequently, particles were dried
185 in an oven at 60°C for 48 h. Approximately 20 mg of acid-treated and non-acid treated samples were
186 weighed into tin capsules and analysed for C composition (%) and N composition (%) respectively using
187 a CHN elemental analyser (Carlo Erba EA1110). The instrument was calibrated using an acetanilide
188 analytical standard (Sigma-Aldrich) of known C, H and N compositions and correction factors were
189 applied through analysis of six method blanks. Additionally, three method replicates were run every ten
190 samples to check for method precision (i.e. repeatability) with an RSD < 10% obtained for all sample
191 runs.

192 2.4.3 Gamma spectrometry

193 Sediment samples were packed and sealed into aluminium containers, or 4 mL vials in the case
194 of low mass samples and allowed to incubate for at least 21 days to promote the development of
195 equilibrium between ^{222}Rn and its parent ^{226}Ra . Gamma counting was carried out at the ISO9001
196 certified Consolidated Radioisotope Facility at University of Plymouth. Samples were counted for
197 ~170,000 s using two calibrated HPGe gamma spectrometers (ORTEC planar detector model GMX50-
198 83-LB-C-SMN-S and ORTEC well detector model GWL-170-15-S for low mass samples). ^7Be and ^{137}Cs
199 were determined from gamma emissions at 477 and 662 keV, respectively. $^{210}\text{Pb}_{\text{ex}}$ was determined by
200 subtraction of ^{226}Ra activity using ^{214}Pb gamma emissions (295 and 352 keV) from total ^{210}Pb (46.5
201 keV). Activity concentrations were decay-corrected to the sample collection date for CBS samples, or
202 to the date of the first flood for SS samples to correct for *in situ* decay of ^7Be inside the trap. Activity
203 concentrations are reported with 2σ uncertainty, and $^{210}\text{Pb}_{\text{ex}}$ uncertainty was obtained by propagation
204 of total ^{210}Pb and ^{214}Pb uncertainties. The detector was calibrated using low radioactivity background
205 soil spiked with a certified and traceable mixed radioactive standard (80717-669; Eckert & Ziegler, GA,
206 USA) packed in containers of the same geometry as the samples. Quality control was carried out at
207 each batch of analysis using IAEA moss soil certified reference material (CRM IAEA-447, employed in
208 the IAEA proficiency test IAEA-CU-2009-03). Obtained values were in good agreement in terms of both

209 precision and accuracy for ^{137}Cs with CRM certified values in both detectors (RSD < 3 % and percentage
210 of measured concentration between 98 to 100 %, Table S2.1 and S2.2). Total ^{210}Pb showed more
211 variation in the planar detector (RSD = 5.6 %) and a higher percentage of measured concentration in
212 both detectors (115 and 124% in the well and planar detectors, respectively. See Table S2.1 and S2.2)
213 which was considered acceptable. The minimum detectable activity concentrations under the conditions
214 of analysis for the five surveys (i.e. background, counting time, geometry and efficiency calibration) for
215 ^7Be , ^{137}Cs and ^{210}Pb were 14, 3.3 and 32 Bq kg⁻¹ for the GWL and 6.0, 1.9 and 25 Bq kg⁻¹ for the GMX,
216 respectively. In order to assure inter-detector comparability (i.e. between well and planar detectors) a
217 cross-check validation experiment was carried out. For this purpose, five sediment samples from the
218 first survey were analysed in both detectors for which no statistically significant differences were found
219 for ^7Be (paired t-test, p-value > 0.05, n = 5) and $^{210}\text{Pb}_{\text{ex}}$ (paired t-test, p-value > 0.05, n = 5), whereas
220 differences between detectors were statistically significant for ^{137}Cs (paired t-test, p-value < 0.05, n =
221 5) with reported ^{137}Cs concentrations higher for the well detector. A correction was applied to the
222 samples from the first survey by subtracting the mean difference (2.6 ± 1.2 Bq kg⁻¹) obtained from the
223 cross-check experiment results.

224 **2.5 Statistical analyses**

225 A similar approach as in Smith et al. (2018), Munoz-Arcos et al. (2021) and Gaspar et al. (2022)
226 was adopted to address the potential influence of sediment properties on FRN concentrations.
227 Correlation analysis was carried out to assess the strength and direction of the association between
228 activity concentrations and sediment properties seasonally. If significant relationships are found, and
229 depending on the direction, enrichment or depletion in tracer properties due to particle sorting and/or
230 organic matter is evidenced (Smith et al. 2018). Here, association between FRNs and sediment
231 properties (D50, SSA, TOC and TN) were assessed using the Pearson correlation coefficient (i.e. R).
232 The main reason for using the Pearson correlation was to assess whether there was a linear association
233 between radionuclide activity concentrations and the measured sediment properties on the basis that a
234 linear association assumption may be reasonable on narrow ranges of SSA, particularly below 1.0 m²
235 g⁻¹ (Smith and Blake 2014). The relationship between OM properties and FRNs may be more complex
236 and dependant on individual FRN chemical characteristics as well as type of OM. Scatterplots of FRNs
237 versus sediment properties were used to assess the overall distribution of the samples and statistically
238 significant correlation coefficients reported at the 95% confidence level. Due to the small number of SS

239 samples per season, correlation analysis was carried out for the whole study period for these samples.
240 Principal Component Analysis (PCA) was conducted to visualise SS and CBS groupings in the
241 ordination plot along the first two main principal components, retained based on their percentage of
242 explained variance.

243 **3 Results**

244 **3.1 Fallout radionuclide distributions**

245 Median ^7Be activity concentrations in SS were higher in spring and summer (367 and 372 Bq
246 kg^{-1} , respectively) and significantly lower in autumn 2022 and winter 2023 (248 and 212 Bq kg^{-1} ,
247 respectively, Fig. 2a). In turn, median ^7Be concentrations in CBS samples were higher in winter and
248 spring 2022 (204 and 197 Bq kg^{-1} , respectively) and lower in summer and autumn 2022 and winter 2023
249 (145, 154 and 127 Bq kg^{-1} , respectively). Beryllium-7 activity concentrations were substantially higher
250 in SS compared to CBS. Excess ^{210}Pb showed higher median activity concentration in SS samples in
251 autumn 2022 (214 Bq kg^{-1}) and lower median concentration in winter samples (110 and 135 Bq kg^{-1} in
252 winter 2022 and 2023 samples, respectively). Median activity concentrations of $^{210}\text{Pb}_{\text{ex}}$ in CBS,
253 however, do not vary significantly between seasons (interquartile ranges between 50 and 150 Bq kg^{-1} ,
254 Fig. 2). Similarly, $^{210}\text{Pb}_{\text{ex}}$ shows consistently high activity concentrations in SS samples compared to
255 CBS throughout all seasons, except in winter 2022 samples (Fig. 2b). Differences in ^{137}Cs activity
256 concentrations between the two sediment types are negligible, however, they vary throughout the
257 seasons. Higher median activity concentration was found in winter 2022 in both SS and CBS (15.0 and
258 15.4 Bq kg^{-1} , respectively) while lower concentrations occurred in summer of the same year (11.2 and
259 11.9 Bq kg^{-1} in CBS and SS, respectively).

260 Substantial variation in FRN activity concentrations were found, both spatial and temporal, in
261 CBS and SS samples. For example, ^7Be activity concentrations in CBS samples throughout seasons
262 presented coefficients of variation (CV) that ranged from 16 to 30 %, 1 to 31 % and 9 to 28 % in the
263 upper, middle and lower river reaches, respectively (Table S3.1). Similarly, $^{210}\text{Pb}_{\text{ex}}$ activity concentration
264 variation ranged from 4 to 25 %, 8 to 21 % and 8 to 33 % in the same locations, and ^{137}Cs variation
265 within sites ranged from 6 to 20 %, 7 to 23 % and 11 to 32 % in the upper, middle and lower reaches,
266 respectively (Table S3.1). Notably lower variation is observed in SSs. Activity concentrations of ^7Be
267 (corrected to the date of first flood to account for *in situ* decay inside the trap) in SS samples along the

268 river reach varied 4, 15, 23, 15 and 13 % in winter, spring, summer, autumn 2022 and winter 2023,
269 respectively, $^{210}\text{Pb}_{\text{ex}}$ varied 19, 14, 13, 17 and 22 %, and ^{137}Cs varied 18, 7, 15, 15 and 12 % during the
270 same seasons (Table S3.2).

271 Fallout radionuclide concentrations in suspended sediment also varied within the storm
272 hydrograph (Fig. 3). Beryllium-7 for instance showed a u-shaped trend with the lowest concentrations
273 ($144 \pm 39 \text{ Bq kg}^{-1}$) occurring during the hydrograph's falling limb and highest SSC. Interestingly, the
274 highest activity concentration ($274 \pm 48 \text{ Bq kg}^{-1}$) occurred during the rising limb of the following storm,
275 but was characterised by significantly lower SSCs (Fig. 3g). Excess ^{210}Pb shows a similar trend with
276 the lowest concentration at the end of the falling limb ($58 \pm 26 \text{ Bq kg}^{-1}$) and highest sediment
277 concentration ($94 \pm 30 \text{ Bq kg}^{-1}$, Fig. 3h) at the rising limb of the following storm. Caesium-137, however,
278 shows steady conditions until it increases slightly from the falling limb trough and remains high during
279 low SSC conditions ($10 \pm 3 \text{ Bq kg}^{-1}$, Fig. 3i), although this trend must be interpreted cautiously since the
280 highest activity concentrations do not depart significantly from the counting error of the lowest
281 concentrations. Total organic carbon (Fig.3f) is the dominant control on ^7Be and $^{210}\text{Pb}_{\text{ex}}$ changes
282 whereas PSD (Fig. 3d and e) are closely related to variations in river flow, highlighting the influence of
283 turbulence during storm flow.

284 **3.2 Sediment property distribution in channel sediments**

285 Particle size distribution parameters were not markedly different between CBS and SS
286 samples, except in summer 2022, where SS exhibited significantly lower D50 values and consequently
287 higher SSA values compared to CBS (Fig. 4). However, seasonal differences in their distribution were
288 noticeable. For example, low D50 values were found in spring 2022 and in winter 2023 (median = 12.5
289 and 13.5 μm) and high values in winter 2022 (median = 16.7 μm) in CBS samples. Suspended
290 sediments, in turn, showed highest D50 values in winter 2022 and lowest in summer 2022 i.e. median
291 = 18.8 and 10.5 μm , respectively. On the other hand, TOC and TN showed consistently higher values
292 in SS compared to CBS and they showed important differences in their distribution between seasons
293 (Fig. 4). It is noteworthy that differences in TOC and TN concentrations between SS and CBS samples
294 are maximised in autumn 2022 samples with CBS samples showing the lowest median concentrations
295 (4.3 and 0.4 % for TOC and TN, respectively) and with SS samples exhibiting the highest median
296 concentrations (8.9 and 0.86 % for TOC and TN, respectively). On the contrary, lowest values of TOC

297 and TN were found in winter 2023 for SS (median = 5.8 and 0.6 %, respectively) whereas higher values
298 were found in summer for CBS (median = 6.3 and 0.7 %, respectively).

299 **3.3 Relationships between FRNs and sediment properties**

300 Correlations between ^7Be and ^{137}Cs activity concentrations and sediment properties in SSs
301 during the study period were not significant ($R < 0.4$, $p\text{-value} > 0.05$, see Fig. 5). Excess $^{210}\text{Pb}_{\text{ex}}$
302 presented a strong and statistically significant association with TOC and TN across the five surveys (R
303 = 0.6 and $p\text{-value} < 0.001$). Association between sediment properties and FRNs in CBSs is highly
304 influenced by seasonality. For example, ^7Be and ^{137}Cs only showed moderate to strong significant
305 correlation, although with opposite slopes, with D50 and SSA in winter 2023 samples ($R = -0.46$ and -
306 0.57 with D50 and 0.60 and 0.56 with SSA for ^7Be and ^{137}Cs , respectively, $p\text{-value} < 0.05$, Table S4.2),
307 whereas $^{210}\text{Pb}_{\text{ex}}$ association with these variables was only significant in spring 2022 samples ($R = -0.57$
308 and 0.52 with D50 and SSA, respectively, $p\text{-value} < 0.05$, Table S4.2). A strong linear, positive and
309 statistically significant correlation between ^7Be activity concentrations and % TOC was found in summer
310 and autumn 2022 samples ($R = 0.88$, $p\text{-value} < 0.001$ and $R = 0.71$, $p\text{-value} < 0.05$ for summer and
311 autumn 2022, respectively). Moderate but significant correlation was also observed in winter 2022
312 samples ($R = 0.44$, $p\text{-value} < 0.05$). Caesium-137 showed strong linear and positive association with
313 TOC in spring samples ($R = 0.74$, $p\text{-value} < 0.01$) and moderate association, although statistically
314 significant, in spring 2022 ($R = 0.44$, $p\text{-value} < 0.05$) and winter 2023 ($R = 0.47$, $p\text{-value} < 0.05$) surveys.
315 Notably, ^{137}Cs showed a moderate negative relationship with TOC in summer samples ($R = -0.4$, $p\text{-value} < 0.05$).
316 Similar trends were found with % TN. A strong positive association between ^7Be and TN
317 in spring and autumn 2022 samples was found ($R = 0.8$, $p\text{-value} < 0.01$), and a moderate but significant
318 correlation between $^{210}\text{Pb}_{\text{ex}}$ and % TN in winter 2023 survey ($R = 0.44$, $p\text{-value} < 0.05$). Caesium-137
319 showed strong and statistically significant correlation, although with opposite slopes, with TN in summer
320 and autumn samples ($R = -0.56$, $p\text{-value} < 0.01$ in summer 2022 and $R = 0.6$, $p\text{-value} < 0.05$ in autumn
321 2022). In winter 2023 correlation between these two variables was moderate but statistically significant
322 ($R = 0.44$, $p\text{-value} < 0.05$).

323 Principal Component Analysis was carried out to assess the distribution of samples along
324 potential influencing environmental variables. TOC and TN loadings plot closely together as a result of
325 their high and positive correlation. The same is true for D50 and SSA, but, because these variables are
326 inversely correlated, they plot along the same components with opposite direction. The first component

327 in the PCA for SS samples (Dim1 in Fig. 7a) accounts for 38 % of the variation, mainly influenced by
328 the TOC, TN and $^{210}\text{Pb}_{\text{ex}}$. This supports the finding that organic matter changes are the dominant control
329 on the distribution of $^{210}\text{Pb}_{\text{ex}}$ in SSs in the River Avon, as also supported by correlation analysis (see
330 $^{210}\text{Pb}_{\text{ex}}$ in Fig. 5). Samples grouping together along this component include those from autumn, spring
331 and winter 2023. Changes in D50 and SSA are the dominant variables along the second component,
332 accounting for 30 % of variation. These loadings affect the distribution of winter and summer 2022
333 samples, which consistently show the highest and lowest median particle size, respectively (see Fig. 4)
334 and the opposite for SSA values. In the case of the CBS samples (Fig. 7b), the first component is mainly
335 driven by PSD properties and ^{137}Cs concentrations, accounting for 33 % of the explained variance. The
336 main groupings along this component include samples from winter and autumn 2022, and to a smaller
337 degree, but with significant influence of SSA values, winter 2023 samples. The second component is
338 mainly associated with increasing values of TOC, TN and to a smaller extent $^{210}\text{Pb}_{\text{ex}}$, explaining 29 %
339 of the variation. The main groups along this component include summer and spring 2022 samples and,
340 to a small degree but characterised by low values of TOC and TN, winter 2023 samples.

341 **4 Discussion**

342 **4.1 Variability of FRN in river channels**

343 Fallout radionuclide activity concentrations in SS and CBS in the study reach vary both spatially,
344 characterised by changes in activity concentrations within and between locations, and temporally, with
345 changes during the storm hydrograph (only SS) and between seasons.

346 4.1.1 Suspended sediments

347 Beryllium-7 was observed to have less spatial variation during winter months (4 to 13 %, Table
348 S3.2) with high coefficients of variation (CV) during summer and marked changes in concentration
349 between seasons. Excess ^{210}Pb also show significant variation between locations and pronounced
350 changes in concentration throughout the seasons. The same applies to ^{137}Cs concentrations. On the
351 one hand, spatial variation in FRN activity concentrations can be attributed to local sediment source
352 dynamics. For instance, low-order feeder streams carrying substantial amounts of sediment from the
353 catchment hillslopes to the main channel close to sampling sites were observed during several storm
354 events. These streams might have a local influence on increasing suspended sediment concentrations
355 at a given site, but with subsequent dilution the effect can be buffered or negligible at downstream sites.

356 On the other hand, seasonal variation in FRNs can be attributed to changes in dominant basin-wide
357 suspended sediment sources. For example, high activity concentrations of ^7Be can be observed during
358 winter, spring and summer 2022 surveys, with a sharp decrease in autumn 2022 and winter 2023, while
359 $^{210}\text{Pb}_{\text{ex}}$ is gradually increasing during the same period reaching its mean maximum concentration (278
360 $\pm 44 \text{ Bq kg}^{-1}$) during autumn 2022 followed by a sharp decrease in winter 2023 (Fig. 2). This suggests
361 a change in the dominant suspended sediment source during autumn 2023. We attribute this shift from
362 sediments delivered from the catchment surface (with moderate changes in relative contribution
363 between pasture to arable lands as can be evidenced by a decline in ^{137}Cs activity concentration) to
364 resuspension from the channel. High activity concentrations of both ^7Be and $^{210}\text{Pb}_{\text{ex}}$ in SS during
365 previous surveys suggest that erosion from the catchment surface is the dominant suspended sediment
366 source. However, the decrease in ^7Be during autumn suggests that ^7Be has undergone significant
367 dilution from a ^7Be -depleted source. The increase in $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs concentrations in the same period
368 can only be attributed to resuspension from the channel because any other sediment source depleted
369 in ^7Be , such as channel banks and/or subsoil erosion from the catchment, would have lower $^{210}\text{Pb}_{\text{ex}}$ and
370 ^{137}Cs as these potential sediment sources tend to have low activity concentrations of these FRNs
371 (Wallbrink and Murray 1993; Blake et al. 2002; Hancock et al. 2014). This inference was supported by
372 observations during a field visit after flooding in November 2022. Here, the mid-channel bars showed
373 significant changes in shape and composition owing to gravel movement and fine sediment washout
374 caused by the flow competence of the high magnitude flood events. However, this switch only occurred
375 during the exceptional events of autumn 2022, as consistent decline in these three radionuclides
376 suggests that significant channel bank erosion subsequently took place during winter 2023 due to high
377 river flows, which was also observed *in situ*, provoking overbank flooding in the upper reach monitoring
378 site peaking between $25 - 30 \text{ m}^3 \text{ s}^{-1}$, a two-fold increase compared to $15 \text{ m}^3 \text{ s}^{-1}$ during storm events in
379 winter 2022 (Fig. 1b).

380 Variations in FRN concentrations during the storm hydrograph were characterised by increased
381 ^7Be and $^{210}\text{Pb}_{\text{ex}}$ concentrations during the rising limb to the peak and a significant decrease during the
382 falling limb that remained steady, although $^{210}\text{Pb}_{\text{ex}}$ decreased slightly before increasing again during the
383 rising limb of the next storm. Caesium-137, however, showed slight increases along storm hydrograph.
384 The general u-shaped trend in ^7Be concentration has also been observed in previous studies under
385 flood conditions, i.e. in the river Clyst, Devon, UK with similar land use regime to the present study

386 (Blake et al. 2002) and in the Houay Pano catchment in Laos (Evrard et al. 2016). Blake et al. (2002)
387 and Evrard et al. (2016) postulated that elevated FRNs concentrations along with increasing SSC during
388 the rising limb of the storm hydrograph were associated with significant erosion from the catchment
389 surface followed by incision into the shallow ^7Be depth profile (Wallbrink and Murray 1993). This is also
390 applicable to the River Avon as the middle and lower parts of this catchment are dominated by arable
391 and pasture lands (Wang et al. 2021). A decline in ^7Be , $^{210}\text{Pb}_{\text{ex}}$ and suspended sediment concentrations
392 during the falling limb can be attributed to a change in suspended sediment sources. Here, two
393 possibilities have previously been described: a switch from 1) the catchment surface to fine sediment
394 remobilised from the channel bed (Blake et al. 2002); or 2) the catchment surface to 'subsurface'
395 sources (Evrard et al. 2016). The decrease in ^7Be and $^{210}\text{Pb}_{\text{ex}}$ concentrations and the slight increase in
396 ^{137}Cs suggest that the dominant sediment source changed from a source that must be low in both ^7Be
397 and $^{210}\text{Pb}_{\text{ex}}$ but presumably constant in ^{137}Cs e.g. ploughed soil profile. A significant contribution from
398 channel banks does not explain this trend as they have been reported to have very low, sometimes
399 negligible, concentrations of these FRNs (Walling and Woodward 1992; Wallbrink and Murray 1993;
400 Walling 2013; Hancock et al. 2014). Additionally, sediment contribution from channel resuspension
401 might not be significant during this event as peak flow reached only $6 \text{ m}^3 \text{ s}^{-1}$ (Fig. 3a). A minor event
402 like this is less likely to resuspend a significant amount of fine sediment from the channel bed in contrast
403 to later events that season with peaks between $25 \text{ m}^3 \text{ s}^{-1}$ and $> 30 \text{ m}^3 \text{ s}^{-1}$. If resuspension would have
404 been dominant, then $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs activity concentrations would have remained relatively high as
405 decay of these radionuclides during summer would have been negligible. Therefore, a third possibility
406 seems more plausible to explain this trend: initial sheet wash overland flow from arable fields was
407 followed by significant rill incision (common in this landscape), resulting in a significant decrease in ^7Be
408 and slight decrease in $^{210}\text{Pb}_{\text{ex}}$, but not ^{137}Cs . During this time of the year, arable fields are ploughed to
409 prepare the land for the next crop season mixing the radionuclides into the plough layer and reducing
410 their activity concentration in the surface, relative, for example, to an undisturbed soil. Then, with
411 precipitation, fallout ^7Be and $^{210}\text{Pb}_{\text{ex}}$ will adsorb onto the exposed soil surfaces adding to the current
412 inventory of these radionuclides but not ^{137}Cs . Finally, as for the period of following the next storm,
413 sediment concentration remains low, but a sharp increase in ^7Be and $^{210}\text{Pb}_{\text{ex}}$ - and not ^{137}Cs - is
414 observed. We attribute these changes to a particle concentration effect, i.e. partition coefficients have
415 been reported to be inversely proportional to suspended particulate matter concentration in natural

416 systems (O'Connor and Connolly 1980; Hawley et al. 1986; You et al. 1989; Benoit 1995). After a period
417 of less intense rain, another storm started to increase river flow but not SSC, as can be seen in Fig. 3b.
418 Under this scenario, particles in suspension were more efficient in scavenging ^7Be and ^{210}Pb from the
419 water column in addition to direct fallout into the channel, highlighting the importance of direct channel
420 precipitation for the ^7Be and $^{210}\text{Pb}_{\text{ex}}$ sediment budget (Karwan et al. 2016, 2018).

421 4.1.2 Channel bed sediments

422 The distribution of FRNs in CBS was characterised by substantial spatial variation within and
423 between mid-channel bars (i.e. locations) throughout seasons within the study reach. It is recognised
424 that the resuspension procedure utilised in this study for bed sediment sampling is subjected to
425 uncontrolled variation due to the penetration depth during sediment remobilisation, especially for ^7Be
426 inventories. Although the sampling program was carried out considering this source of uncertainty by
427 consistently and carefully resuspending the shallow surface of the channel bars, variation in ^7Be
428 concentrations in CBS samples was higher compared to $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs (CV = 32, 22 and 23 % in
429 the 5 surveys for ^7Be , $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs , respectively). The fact that $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs showed similar
430 degrees of variation confirms that penetration depth might be a potential source of variability for ^7Be
431 aside from random variation during sample processing and analysis. Notwithstanding the above,
432 significant conclusions can be drawn from the distribution of these radionuclides in channel bed
433 sediments. For example, variation in activity concentrations within channel bars can be attributed to
434 their changing particle composition along their length. Generally, the head was dominated by sand while
435 the middle and tail were dominated by gravel (see Table S1.1). Often, more fine particles were
436 recovered from the sandy bar heads than gravel-dominated tails. However, no clear trend could be
437 established. Variability of FRNs between sites can be attributed to local channel characteristics and
438 mid-channel bar sizes (Table S1.1). For example, the upper reach bar was smaller and formed in a
439 narrow part of the channel with a stronger current, while the middle and lower reach channel bars were
440 found in wider areas of the channel characterised by lower currents. Therefore, it is expected that fine
441 sediment exchange in the upper bar would be more efficient compared to the middle and lower reach
442 bars. This was supported by particle recovery data from resuspension (Table S1.2) in which middle and
443 lower bars generally resulted in higher fine sediment recovery. These local channel characteristics
444 might have influenced turnover of $^{210}\text{Pb}_{\text{ex}}$, and to a lesser extent ^{137}Cs , which consistently showed higher
445 activity concentrations in the upper reach except during autumn 2022 (see Table S3.1) where, as has

446 been already discussed, significant resuspension took place. As to seasonal variability, relatively high
447 FRN concentrations in winter and spring 2022 (see Fig. 2) suggest significant replenishment of fine
448 sediments in the surface of the channel bars from catchment surface soil erosion. The decline in activity
449 concentrations in summer suggest that although ^7Be has undergone significant decay it must have been
450 replenished by significant erosional inputs from the catchment, otherwise, it would have decreased its
451 concentration by half due to decay since the last flood (late March 2022). Furthermore, the slight decline
452 in both $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs suggests that the channel bed was replenished with fine sediment depleted in
453 activity concentration from these radionuclides. The monitoring station located in the lower reach
454 recorded two small storm events that took place on 4th and 29th June. These events were followed by a
455 substantial increase in suspended sediment within the channel, but they did not significantly increase
456 river flow (Fig. 1b). It is likely that exposed arable soil surfaces during the harvesting season, relatively
457 depleted in ^{137}Cs due to mixing during ploughing, but not significantly depleted in $^{210}\text{Pb}_{\text{ex}}$ and in ^7Be due
458 to fresh fallout, had been eroded during these events contributing to the decline in their concentrations
459 in bed sediments. Similar FRN activity concentrations in the autumn suggest two possibilities: 1)
460 replenishment from a sediment source depleted in ^7Be ; or 2) significant fine sediment removal from the
461 bar surfaces due to remobilisation/resuspension. In this case, observational evidence supports the
462 latter. First, field observations suggest that significant gravel movement took place after the autumn
463 2022 floods (Fig. 1b), and in some cases significant change in the gravel composition of the bars was
464 also noted. Second, low amounts of sediment were recovered after resuspension from these bars
465 compared to previous surveys (about 70 % less material, see Table S1.2) and only eight samples were
466 above the minimum detectable activity for ^7Be (see 'n' in Fig. 2). Third, as explained in section 4.1.1,
467 complementary evidence from $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs in SS suggest significant resuspension during this
468 period as their activity concentrations increased significantly in SS, which would not be the case if
469 significant erosion from subsurface soils or channel banks had occurred. Nevertheless, continued
470 decline in these FRNs during winter 2023 can be attributed to significant inputs from channel banks as
471 significant flood events and overbank flooding occurred during December and January 2023.

472 **4.2 Influence of sediment properties on FRNs**

473 The relationship between sediment properties and radionuclide activity concentrations in the
474 context of sediment redistribution studies is well documented in soils, and empirical relationships have
475 been derived to account for potential influence on FRN activity concentrations from variations in PSD

476 and/or changes in OM (He and Walling 1996; Taylor et al. 2014; Laceby et al. 2017). However, the
477 assessment of these relationships has received less attention in the context of potential changes in
478 fluvial sediment properties. Moreover, the effects of changes in sediment properties on FRN
479 concentrations under channel storage conditions have rarely been considered (Singleton et al. 2017).
480 Here we discuss different potential environmental controls on FRN activity concentrations for fluvial
481 suspended (SS) and channel bed (CBS) sediments.

482 4.2.1 Suspended sediments

483 Associations between FRNs and D50 and SSA in SS were not significant during the study
484 period, suggesting that particle size selectivity, i.e. the preferential adsorption of trace elements and
485 radionuclides to finer particles (Horowitz and Elrick 1987; He and Walling 1996), might not always have
486 a significant influence on FRN activity concentrations in suspended sediment. The associations
487 between $^{210}\text{Pb}_{\text{ex}}$ and TOC and TN were strong. High and usually positive association between these
488 radionuclides and OM can be attributed to changes in dominant sediment source. As discussed in
489 section 4.1.1, evidence supports change of dominant sediment source between seasons, with surface
490 erosion dominant during the first three surveys, i.e. winter, spring and summer 2022, with a switch to
491 significant resuspension from the channel in autumn 2022, and dominant input from channel bank
492 erosion in winter 2023. It is therefore expected that OM varies in a similar fashion to $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs
493 when discriminating between surface soils and subsoils sediment sources, as both TOC and TN have
494 been reported to decrease with depth (Laceby et al. 2017). This is not the case for ^7Be as its occurrence
495 is only limited to the top two centimetres of the soil profile where significant changes in OM are less
496 likely to occur. Based on PCA results (Fig. 7a), OM properties co-varying with ^{210}Pb along the first
497 component suggest that changes in OM exert slightly more influence in characterising seasonal SS
498 compared to PSD as the first component of explained variance is 8 % higher.

499 4.2.2 Channel bed sediments

500 On the whole, the association between FRNs and PSD properties were not of significance in
501 CBS, except for an inverse, statistically significant correlation of both ^7Be and ^{137}Cs with PSD properties
502 in winter 2023 samples. Increased activity concentrations with decreasing D50 (or increasing SSA)
503 indicates potential for particle selectivity during this period. It can be postulated that these radionuclides
504 were significantly affected by particle sorting during the high flow events that occurred during winter
505 2023 (see Fig. 1b) which interestingly did not affect $^{210}\text{Pb}_{\text{ex}}$ in the same way.

506 On the other hand, OM properties showed positive and significant linear relationships with ^7Be
507 in winter, summer and autumn 2022. Of note, correlation between these two variables was highly
508 significant in summer ($p < 0.0001$, see Table S4.2). As explained in section 4.1.2., significant sediment
509 input from catchment erosion took place during this period, as it could be seen from the notable increase
510 in SSC (Fig. 1b). Consequently, it is likely that ^7Be enrichment due to an increase in particulate organic
511 matter had occurred. However, if sediment inputs from the catchment were the sole driver of this
512 association, similar trends would have been observed for $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs . The fact that no clear
513 association between $^{210}\text{Pb}_{\text{ex}}$ and TOC and a negative correlation between ^{137}Cs and TOC were
514 observed further indicates that occurrence of these radionuclides in the channel bed might be affected
515 by changes in organic matter content and composition. Research suggests that rivers during dry
516 seasons with low rainfall and river flow induce a lacustrine-like condition leading to the OM composition
517 being more aquagenic and flocculation-favourable (Lee et al. 2019). This could be observed during the
518 field survey in summer 2022, where OM flocculation was evident on the surface of the sampled bars. A
519 significant increase in TOC in summer 2022 samples compared to previous surveys also supports this
520 (Fig. 4). With increasing water temperature, algae growth and autochthonous OM production is
521 favoured. Organic matter then may be adsorbed onto fine sediments causing either stabilisation or
522 flocculation (Lee et al. 2019). Under this scenario, ^7Be is delivered as Be^{+2} in slightly acidic rainfall which
523 is highly competitive for cation exchanges sites (Kaste et al. 2002) thus highly particle-reactive following
524 fallout in the channel. Baskaran et al. (1997) reported a positive correlation between ^7Be K_d and
525 particulate organic carbon ($R = 0.61$, $p\text{-value} < 0.05$), although this relationship resulted from data
526 obtained in a rather different environment, i.e. a shallow, high dissolved organic carbon, turbid estuary.
527 Caesium-137, on the other hand, has been reported to have increased mobility in catchments with soils
528 characterised with high OM content (Dumat and Staunton 1999; Fan et al. 2014), and its mobility in
529 sediments may be enhanced due to weaker interaction of ^{137}Cs with clay minerals caused by the
530 blocking effect of the adsorbed humic substances onto fine particles (Suga et al. 2014; Takahashi et al.
531 2017). Clearly, there is potential for FRN alteration due to long particle residence time in the channel
532 under changing river water conditions, especially in summer season. The PCA of CBS samples also
533 supports this as the significant influence of OM properties on summer 2022 samples is clearly
534 evidenced. In addition, the potential for reducing conditions in sediment storage zones, such as the
535 channel bed and waterlogged soils, have been reported as a potential cause of desorption of ^7Be (Taylor

536 et al. 2012, 2013). This does not seem to be the case in the River Avon, however, as sediment turnover
537 in the channel bed is highly responsive to flood conditions.

538 **5 Conclusion and Implications for sediment tracing in rivers**

539 Activity concentrations of FRNs (^7Be , $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs) were quantified in fine sediments from
540 time-integrated sediment traps, channel bars and one storm hydrograph. Activity concentrations in SSS
541 varied spatially along the river reach which was attributed to contribution of local sediment sources as
542 observed from substantial suspended sediments transported by low-order feeder streams during storm
543 events, and seasonally due to changes in dominant suspended sediment sources. In addition,
544 suspended sediments showed variation in FRN activity concentrations during the storm hydrograph
545 which were attributed to changes in dominant sediment sources during the rising and falling limbs and
546 also to a particle concentration effect. The latter occurred at the beginning of another storm where river
547 flow increased significantly but SSC remained low, favouring radionuclide scavenging onto particles.
548 Fallout radionuclides in CBS varied spatially both within the channel bars, with changes associated to
549 the gravel bar composition, and between channel bars, reflecting changes in channel width and bar
550 size. Seasonal variation in FRN activity concentrations is closely related to trends observed in SS
551 samples reflecting changes in dominant sediment sources. Furthermore, PSD did not show significant
552 correlation with FRNs in both SS and CBS samples (except for ^7Be and ^{137}Cs in CBS winter 2023
553 samples). Organic matter properties, on the other hand, showed strong significant correlation with
554 $^{210}\text{Pb}_{\text{ex}}$ in SS during the whole study period, attributed to co-variations with OM due to changes in
555 dominant sediment sources. Positive and significant correlation of ^7Be and OM, and inverse but
556 significant correlation of ^{137}Cs and OM in summer 2022 samples, were attributed to riverine
557 autochthonous organic matter production that increased particulate OM content and changed its
558 composition.

559 The variability of FRNs in river channels can give significant insights into catchment-wide
560 processes defining sediment dynamics and sources, however, significant spatial and temporal variation
561 adds complexity to their use as sediment tracers. For instance, when using FRNs in the context of
562 sediment fingerprinting, continuous characterisation of temporally relevant sediment sources is highly
563 recommended, as variations in suspended sediment activity concentrations are not only responsive to
564 changes in dominant sediment sources but also to potential OM changes through time (e.g. seasons).

565 Additionally, when using channel bed sediments as target/mixture sediments, attention should be given
566 to potential ^7Be enrichment and mobility of ^{137}Cs due to OM production under storage conditions
567 affecting their conservativeness as sediment tracers. Beryllium-7 activity concentration can decrease
568 by significant contribution of ^7Be -depleted sources such as subsurface erosion and channel banks,
569 however, resuspension of sediment depleted in ^7Be from the channel can also contribute significantly
570 to ^7Be depletion in suspended sediments. Therefore, sediment transit/residence time studies focusing
571 on suspended sediments should consider potential sediment resuspension from the channel as a key
572 part of the sediment source framework. If resuspension is significant then there is potential for
573 overestimation of sediment transit/residence times. As highlighted above, ^7Be enrichment due to in-situ
574 growth of OM can also affect sediment storage time estimations, and potential enrichment through OM
575 scavenging could result in underestimation of storage times. Finally, continuous river monitoring can
576 further aid interpretation of FRN temporal variability and sediment dynamics, and we recommend that
577 sediment tracing studies employing FRNs should routinely be accompanied by river monitoring to permit
578 hydrological and sediment flux dynamics to be integrated into interpretations.

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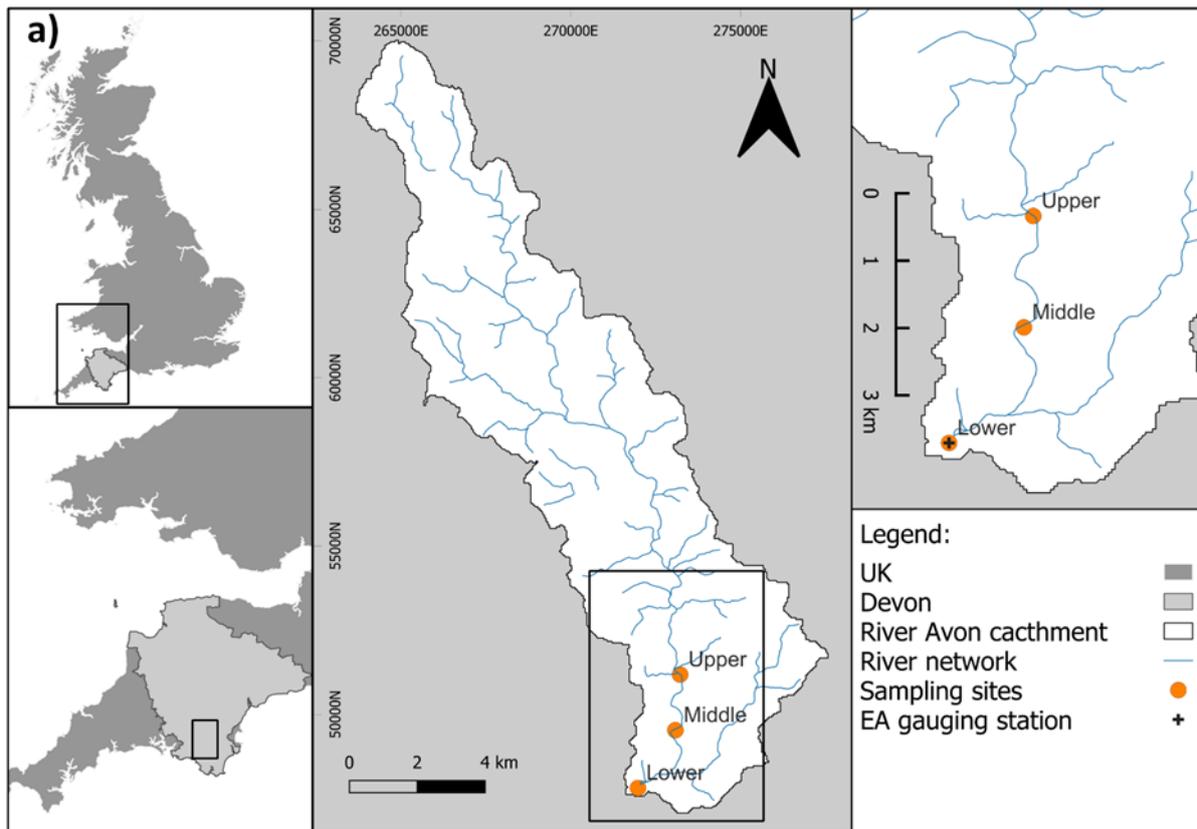
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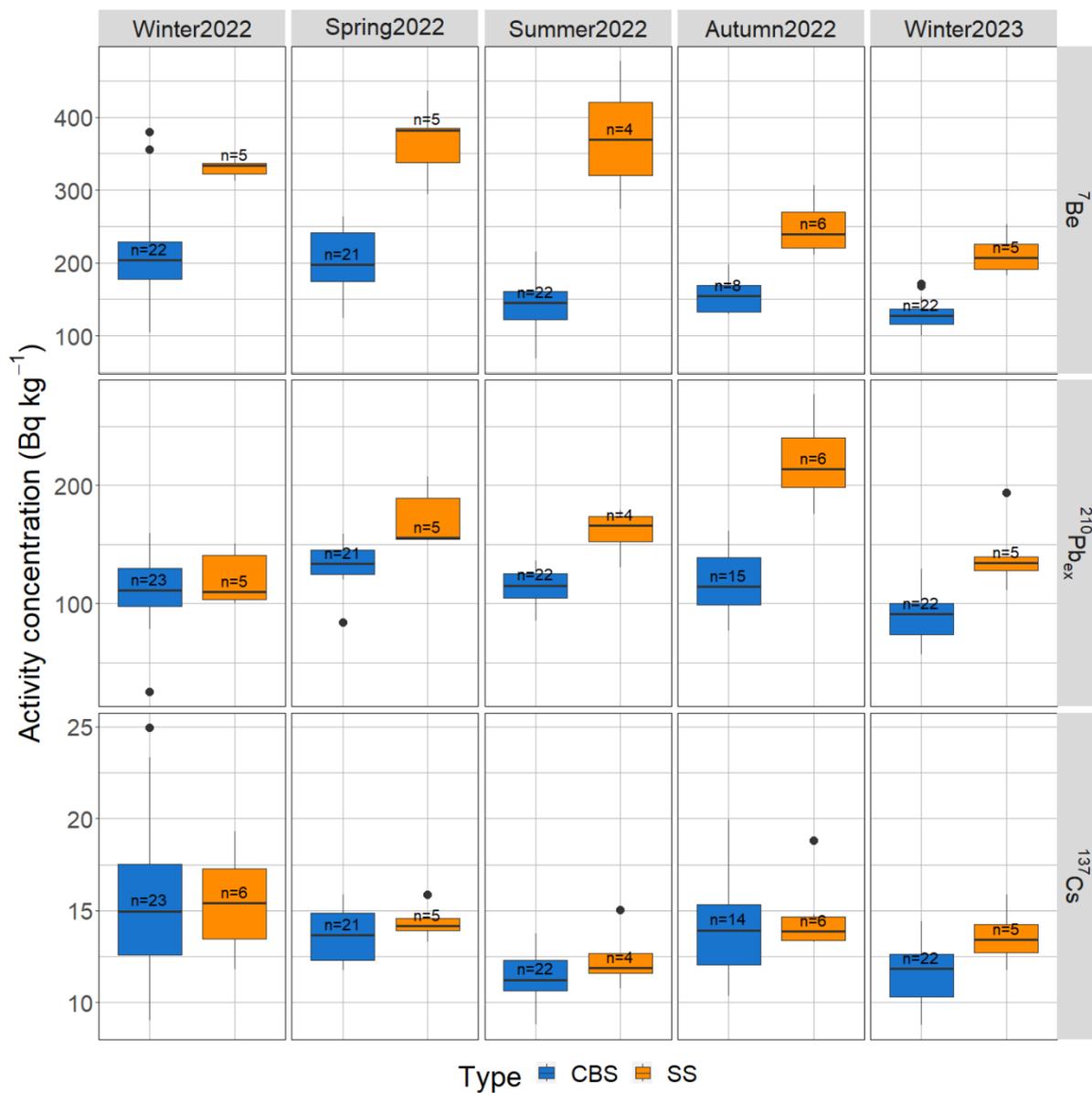


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763 **Fig. 1** Map of the a) the River Avon catchment and sampling sites in the selected river reach; and b)
 764 river flow and suspended sediment concentration (SSC). River flow was obtained for the Loddiswell
 765 gauging station managed by the Environment Agency, UK from the Hydrology Data Explorer at
 766 <https://environment.data.gov.uk/hydrology/explore> and SSC obtained from turbidity records at a

767 monitoring station set up at the lower reach sampling site (note that lack of data in September and
 768 October was due to probe battery failure). Dashed black lines indicates the date of the sampling.

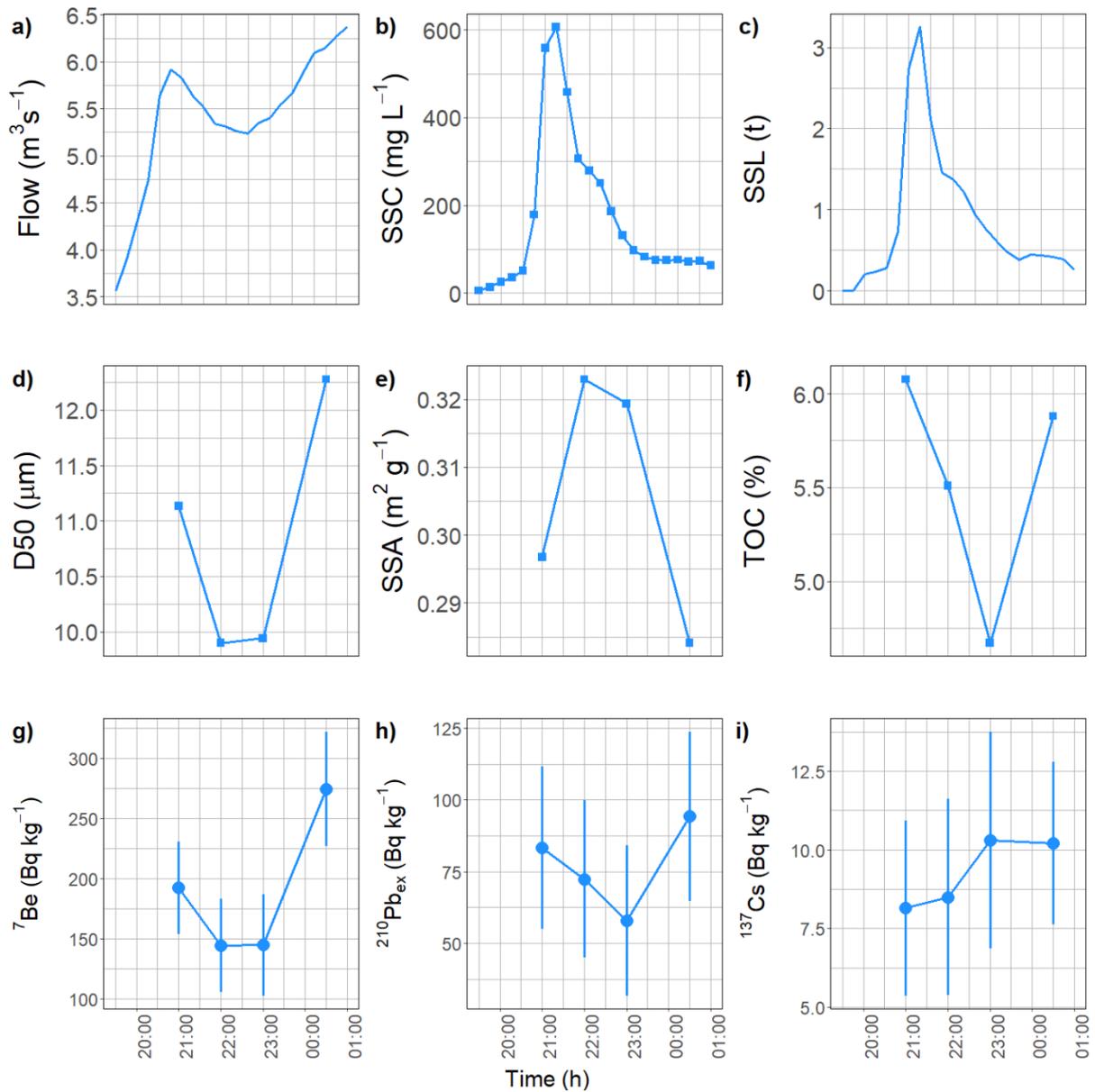
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771 **Fig. 2** Seasonal distributions of ⁷Be, ²¹⁰Pb_{ex} and ¹³⁷Cs in CBS and SS samples.

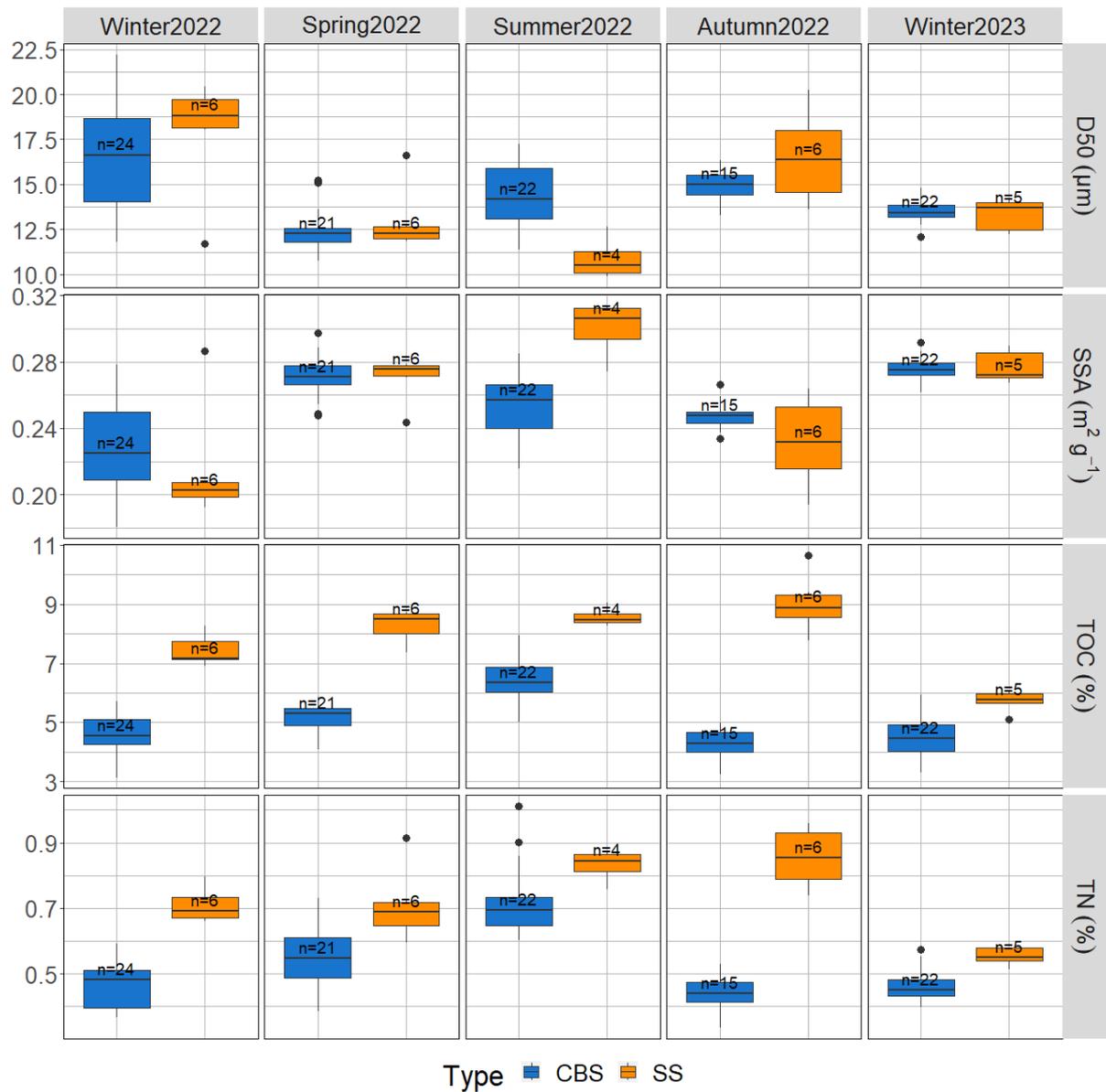
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774 **Fig. 3** Changes in FRN activity concentrations along the storm hydrograph in samples collected in the
 775 upper part of the study reach on 02/11/2022. a) River flow, b) suspended sediment concentration (SSC),
 776 and c) suspended sediment loads (SSL) obtained from a monitoring station on site. d) Median particle
 777 size (D50), e) specific surface area (SSA), f) total organic carbon (TOC), g) ^7Be , h) $^{210}\text{Pb}_{\text{ex}}$ and i) ^{137}Cs .
 778 Uncertainty bars in activity concentrations indicate 2σ error from counting statistics. Note that
 779 suspended particles from continuous flow separation were sieved to $< 63 \mu\text{m}$.

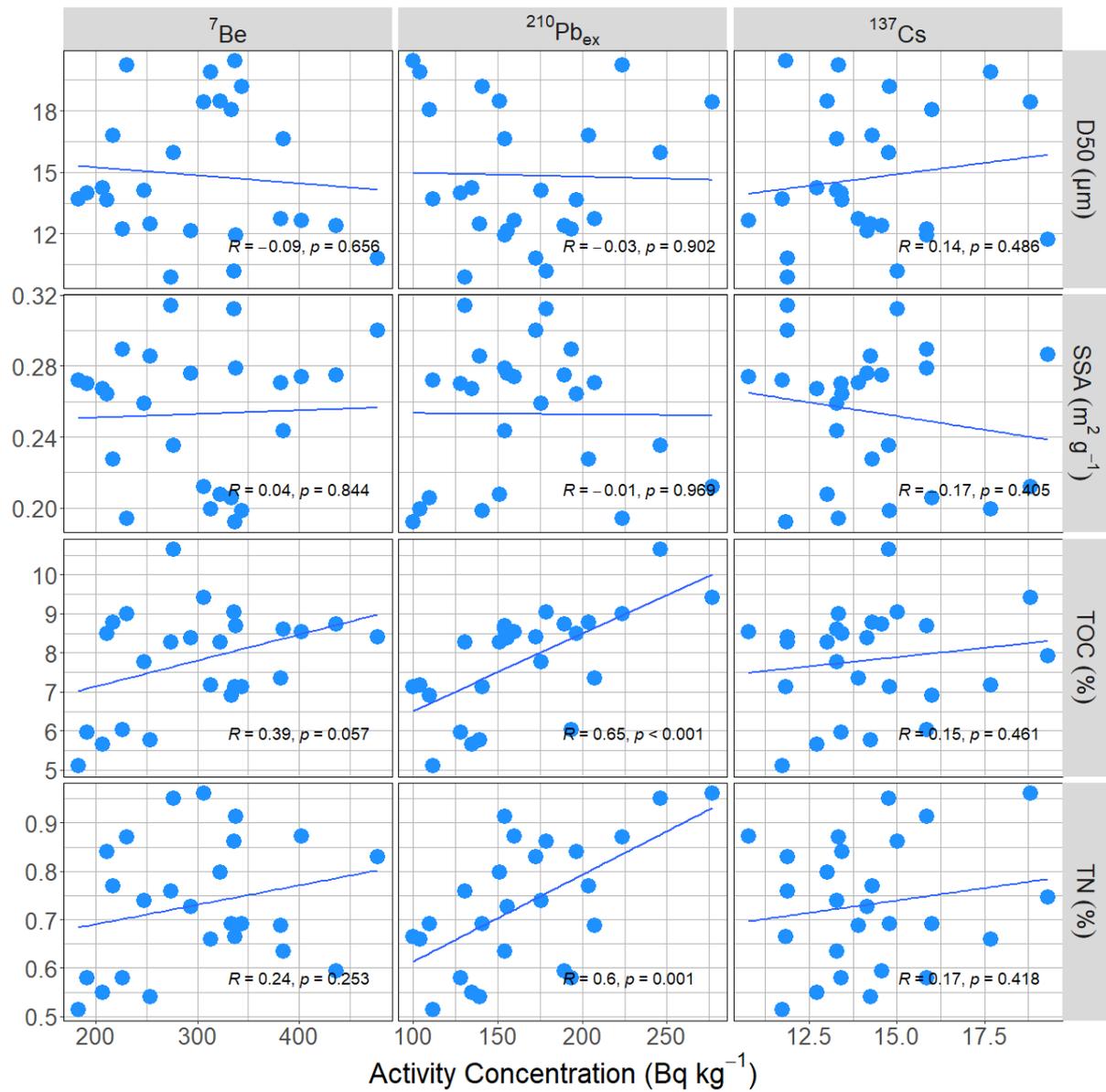
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782 **Fig. 4** Seasonal distribution of D50, SSA, TOC and TN values in SS and CBS samples.

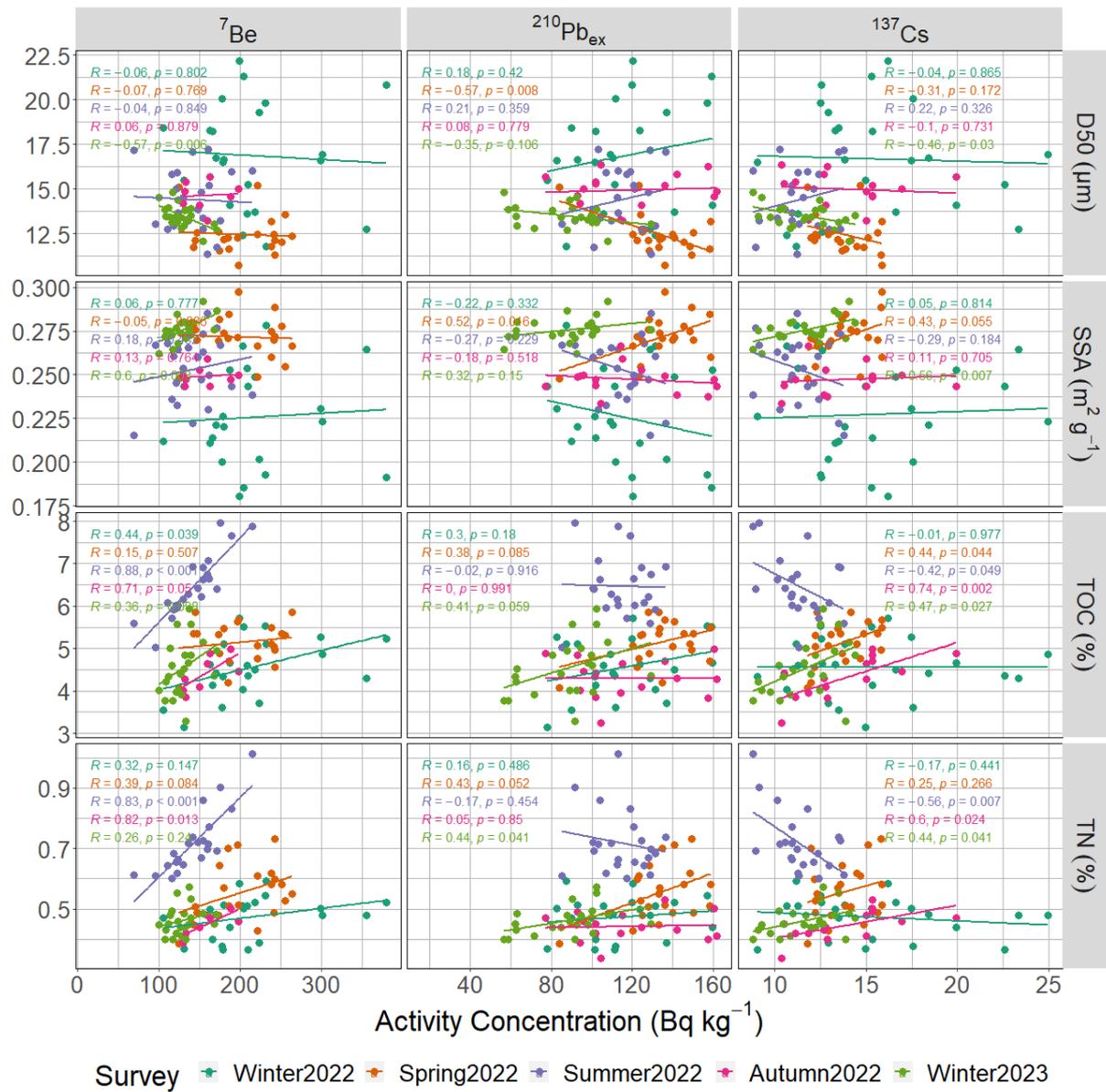
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785 **Fig. 5** Scatter plots, including Pearson correlation coefficients and p-values, between ${}^7\text{Be}$, ${}^{210}\text{Pb}_{\text{ex}}$ and
 786 ${}^{137}\text{Cs}$ and D50, SSA, TOC and TN in suspended sediments (SS) collected from traps.

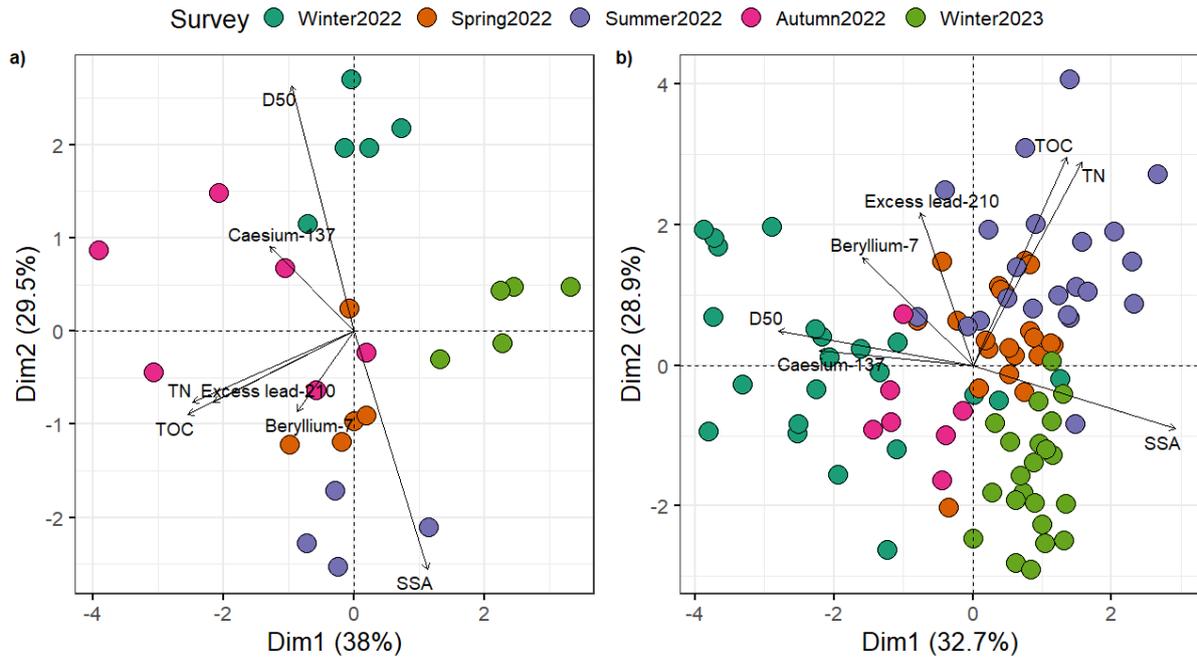
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789 **Fig. 6** Scatter plots, including Pearson correlation coefficients and p-values, between ${}^7\text{Be}$, ${}^{210}\text{Pb}_{\text{ex}}$ and
 790 ${}^{137}\text{Cs}$ and D50, SSA, TOC and TN in samples CBS samples.

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793 **Fig. 7** PCA score (samples) and loading (variables) plots of a) SS and b) CBS samples.



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