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Suspended sediment dynamics in a Southeast Asian mountainous catchment: Combining river monitoring and fallout radionuclide tracers

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5 Title:

6 Suspended sediment dynamics in a Southeast Asian mountainous catchment: combining
7 river monitoring and fallout radionuclide tracers

8

9 Abstract

10 Soil erosion is intense in mountainous tropical regions where heavy storms result in the
11 supply of large quantities of sediment to rivers. The origin and dynamics of suspended
12 sediment were analysed in a catchment located in northern Laos during the first erosive
13 flood of the rainy season in May 2012. The catchment was equipped with 4 successive
14 gauging stations (draining areas ranging 0.2 - 11.6 km²). Fallout radionuclides (Beryllium-7 -
15 ⁷Be, unsupported Pb-210 -²¹⁰Pb_{xs}, and Cesium-137 -¹³⁷Cs) were determined on rainfall,
16 overland flow, stream water, suspended sediment, soil surface and subsurface samples
17 (with n = 3, 19, 75, 75, 65 and 14 respectively). Assumptions underpinning the ⁷Be-labelling
18 method were validated by implementing experiments in the laboratory (i.e., rainwater ⁷Be
19 sorption to soil particles) and in the field (i.e., ⁷Be: ²¹⁰Pb_{xs} activity ratio evolution in rainwater
20 and related overland flow during a natural storm event). Radionuclide analyses provided a
21 way to quantify variations in sediment dynamics and origin throughout the flood: (1) a
22 proportion of recently eroded sediment (labelled by ⁷Be, and referred to as “fresh
23 sediment”) ranging between ca. 8 - 35% in suspended loads; (2) higher contributions of
24 fresh sediment at the beginning of the flood rising stage; (3) a progressive dilution of fresh
25 sediment by particles remobilised from the riverbed / channel; (4) the dominance of
26 particles originating from the soil surface (ca. 70 - 80% of total sediment load) in upper
27 parts and a much larger contribution of subsurface material (ca. 64%) at the downstream
28 station. The original contribution of ⁷Be-labelled particles derived from collapsed riverbanks
29 to sediment export was also demonstrated. This pilot study supports the use of fallout ⁷Be
30 and ²¹⁰Pb_{xs} in tropical catchments to constrain sediment dynamics. It also puts forward the
31 need to better characterize the sources of sediment in order to avoid possible
32 misinterpretations.

33 1. Introduction

34 Soil erosion is particularly intense in mountainous subtropical regions where heavy storms
35 may result in the supply of large quantities of suspended sediment to streams (Descroix et
36 al., 2008; Valentin et al., 2008). Large exports of suspended matter by mountain rivers lead
37 to numerous problems downstream (Syvitski et al., 2005). Sediments can accumulate
38 behind dams, which results in the siltation of water reservoirs (Downing et al., 2008;
39 Thothong et al., 2011). Suspended matter also contributes to water quality degradation
40 (Tanik et al., 1999) and conveys biological compounds, playing thereby a major role in
41 global nutrient biogeochemical cycles (Quinton et al., 2010). It also constitutes a potential
42 vector for various pollutants such as metals, polycyclic aromatic hydrocarbons or faecal
43 bacteria (Ribolzi et al., 2010; Gateuille et al., 2014).

44 In order to limit those negative impacts, sediment supply to rivers needs to be controlled.
45 Design and implementation of appropriate management procedures require a better
46 understanding of suspended matter dynamics in mountainous catchments. Their behaviour
47 should be better constrained in time, and particularly during floods, as most riverine
48 sediments are exported during those short periods (Meybeck et al., 2003; Mano et al.,
49 2009). To this end, tracers that are preferentially sorbed or contained in the fine mineral and
50 organic suspended fractions (i.e., clays and fine silts, He and Walling, 1996) may be used
51 to follow sediment pathways across catchments (Koiter et al., 2013).

52 Radionuclides that are supplied to the soil surface by rainfall, i.e. beryllium-7 (^7Be) and
53 unsupported or excess lead-210 ($^{210}\text{Pb}_{\text{xs}}$) are used to estimate soil erosion rates at the
54 hillslope scale (Schuller et al., 2006; Sepulveda et al., 2008), or to characterize the
55 temporal transfer of sediment in larger river systems (Bonniwell et al., 1999). Their different
56 half-lives ($T_{1/2} = 53$ days for ^7Be and $T_{1/2} = 22.3$ years for $^{210}\text{Pb}_{\text{xs}}$) are particularly relevant to
57 differentiate between fresh sediment tagged with ^7Be and older remobilized sediment
58 depleted in ^7Be . Based on this simple principle, Matisoff et al. (2005) proposed to calculate
59 the $^7\text{Be} : ^{210}\text{Pb}_{\text{xs}}$ activity ratio ($^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$) in both rainwater and riverine sediment to

60 estimate fresh sediment percentages in rivers and infer transfer times or transport
61 distances. Alternative approaches used radionuclide mass-balance models such as the one
62 proposed by Dominik et al. (1987) and improved by Le Cloarec et al. (2007), or associated
63 both methods (Evrard et al., 2010). However, several limitations may arise regarding the
64 assumptions underpinning those methods. The validity of radionuclides as tracers of
65 sediment fluxes in large rivers has been recently questioned (Walling, 2012; Taylor et al.,
66 2013). The main criticism focussed on the potential difference of ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{xs}}$ activity ratio
67 value in rainwater and in fresh sediment. This may occur when particles are tagged with
68 radionuclides from successive storms and not with the event of investigation alone. Another
69 concern arises from a possible misinterpretation of ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{xs}}$ variations measured in
70 sediment, as low values may result from various processes: radionuclide decay; desorption
71 (when sediment remained buried in the riverbed) or changes in the source of sediment with
72 the supply of subsurface particles (depleted in fallout radionuclides; e.g., Whiting et al.,
73 2005). In order to reduce those uncertainties, a third fallout radionuclide, cesium-137 (${}^{137}\text{Cs}$;
74 $T_{1/2} = 30.2$ years) proved to be useful to distinguish between particles originating from soil
75 surface and exposed to atmospheric fallout of bomb tests during the second half of the 20th
76 century (Ritchie and McHenry, 1990) and particles from the subsurface (below ca. 30 cm
77 depth), protected from ${}^{137}\text{Cs}$ and ${}^7\text{Be}$ fallout (e.g. Olley et al., 1993; Ben Slimane et al.,
78 2013; Evrard et al., 2013; Hancock et al., 2014).

79 In this study, experiments were carried out in the Houay Pano - Houay Xon nested
80 catchments located in Laos and exposed to summer monsoon, to quantify the respective
81 contributions of surface and subsurface soil to suspended sediment loads during an erosive
82 flood event that took place at the beginning of the rainy season in May 2012. The fallout
83 ${}^7\text{Be}$ activity of the previous rainy season should have sufficiently decayed during the 6-
84 months dry period to become negligible compared to their recent supply at the onset of the
85 wet season. Every compartment of the erosional system, from rainwater to stream

86 sediment, was sampled for fallout radionuclide analyses. Adsorption experiments were also
87 conducted for ^7Be at the microplot's scale under natural rainfall and in the laboratory.

88 2. Study site

89 The Houay Pano catchment, located 10 km south of Luang Prabang in northern Laos (Fig.
90 1), has been part of the MSEC (Monitoring Soil Erosion Consortium) network since 1998
91 (Valentin et al., 2008). The tropical monsoon climate of the region is characterized by the
92 succession of dry and wet seasons with *ca.* 80% of rainfall occurring during the rainy
93 season from May to October (Ribolzi et al., 2008). The Houay Pano stream has an average
94 base flow of $0.4 \pm 0.1 \text{ L s}^{-1}$ and is equipped with 2 gauging stations that subdivide the
95 catchment into nested subcatchments. These stations, S1 and S4, draining 20 ha and 60
96 ha respectively, are located along the main stem of the stream. Between S1 and S4
97 stations, water flows through a swamp (0.19 ha), supplied with water by a permanent
98 groundwater table (Fig. 1). Only temporary footslope and flood deposits can be found along
99 this narrow section of the stream and the swamp represents the major sediment
100 accumulation zone in the Houay Pano catchment. The Houay Pano stream flows into the
101 Houay Xon River (22.4 km² catchment) and is continuously monitored at S10 (draining a
102 11.6 km² catchment), located 2.8 km downstream of S4. The Houay Xon is a tributary of the
103 Nam Dong River, flowing into the Mekong River within the city of Luang Prabang (Ribolzi et
104 al., 2010).

105 The geological basement of the Houay Pano catchment is mainly composed of pelites,
106 sandstones and greywackes, overlaid in its uppermost part by Carboniferous to Permian
107 limestone cliffs. Soils consist of deep (>2 m) and moderately deep (>0.5 m) Alfisols
108 (UNESCO, 1974), except along crests and ridges where Inceptisols can be found (Chaplot
109 et al., 2009). Soils have a low cation exchange capacity and a low pH ranging between 4.9
110 –5.5 across the catchment. Native vegetation consisted of lowland forest dominated by
111 bamboos that were first cleared to implement shifting cultivation of upland rice at the end of

112 the 1960s (Huon et al., 2013). Elevation across the catchment ranges *ca.* 272–1300 m.a.s.l.
113 As cultivation takes place on steep slopes (ranging between 3-150%), the catchment is
114 prone to soil erosion (Chaplot et al., 2005; Ribolzi et al., 2011). Due to the decline of soil
115 productivity triggered by soil erosion over the years (Patin et al., 2012) and to an increasing
116 labour need to control weed invasion (Dupin et al., 2009), farmers progressively replaced
117 rice fields by teak plantations in the catchment (Fig. 1). During the present study, main land
118 uses in the Houay Pano catchment were teak plantations (36% of total area), rotating
119 cropping land (35%), Job's tears (10%), banana plantations (4%) and upland rice fields
120 (3%); the forest covering less than 9% of the area. The land use was different in the larger
121 area drained by S10, with 56% of the surface covered with forests, 15% under teak
122 plantations and 23% under cropland.

123 [Fig. 1]

124 3. Materials and methods

125 3.1. Sample and data collection

126 Rainfall, stream and overland flow waters were sampled during the May 23 flood in 2012.
127 Rainfall intensity was monitored with an automatic weather station (elevation: 536 m.a.s.l.)
128 and stream discharge was calculated from water level continuous recording and rating
129 curves. Rainfall was sampled with three cumulative collectors, located in the village near
130 the confluence between Houay Pano and Houay Xon streams, near a teak plantation on the
131 hillslopes located just upstream of the village and within the Houay Pano catchment.
132 Overland flow was collected at the outlet of 1-m² experimental plots. Stream water was
133 collected in plastic bottles after each 20-mm water level change by automatic samplers
134 installed at each gauging station. Fifty-six total suspended sediment (TSS) samples were
135 collected at the three stations (S1, S4, S10). Samples were dried shortly after collection in
136 an oven ($t \approx 100^\circ\text{C}$) for 12-48 h. In addition, sediment deposited upstream of S4 in the river

137 channel (top 0-1 cm of in-channel deposits collected using a plastic trowel) was sampled
138 the day before the May 23 flood to document the initial radionuclide activity. Surface soil
139 samples (top 0–5 cm; n=65) were collected using plastic trowels on the hillslopes
140 connected to the Houay Pano Stream and the Houay Xon River (Fig. 1) during three
141 campaigns conducted in July 2002 (Huon et al., 2013), May 2012 and December 2012.
142 Additional gully (n = 6) and riverbank (n = 8) samples were also collected in December
143 2012 to document the characteristics of the potential subsurface sources of sediment to the
144 river.

145 Cumulative TSS exports were calculated at each station by summing the TSS masses
146 exported between two successive sample collections. As no rating curve could be
147 determined between TSS and Q (hysteresis patterns) during the entire event (absence of
148 causal relationship between both parameters), TSS concentration was considered to vary
149 linearly between successive measurements. Considering the relatively high number of
150 successive samples collected at each station and the resulting high temporal resolution of
151 TSS measurements, this assumption appeared to be reasonable. Sediment yields were
152 calculated by dividing the cumulative TSS exports by the corresponding sub-catchment
153 area.

154 3.2. Sample preparation and radionuclide analyses

155 To reduce the volume of rainwater (0.6 - 18 L) that would have been required to conduct
156 direct gamma spectrometry analyses, fallout radionuclide recovery was performed in the
157 field by co-precipitation with aluminium hydroxides (Ciffroy et al., 2003; Evrard et al., 2010).
158 Samples were prepared by adding 1.5 g of aluminium chloride hexahydrate. Co-
159 precipitation was achieved by addition of 1-N NaOH solution until pH attained 8.5-9.0. After
160 5 hours, the supernatant was removed and the precipitates were placed in an aluminium
161 tray and dried in an oven. All residues were placed in polypropylene tubes and sealed
162 airtight to contain ^{222}Rn and allow in-growth of its decay products. Counting was conducted

163 at the Laboratoire des Sciences du Climat et de l'Environnement (LSCE) in Gif-sur-Yvette
164 by gamma spectrometry using a low-background, high-efficiency, well-type Ge detector with
165 a crystal volume of 220 cm³ (GWL-220-15 Ortec[®]). Most samples were analysed within less
166 than 53 days (⁷Be half-life) following the rainfall event.

167 Radionuclide activities were measured in a total of 45 individual or composite (depending
168 on the quantity of material recovered) suspended sediment samples (0.2 – 15.1 g), 1
169 riverbed sediment sample (72 g) and 29 soil samples (48 – 78 g). Samples were packed
170 into 15-60-ml (depending on the quantity available) polyethylene specimen cups and sealed
171 airtight. The ⁷Be, ¹³⁷Cs and ²¹⁰Pb activities were determined at 477.6 keV, 661.6 keV and
172 46.5 keV, respectively, by gamma spectrometry using the very low-background coaxial N-
173 and P- type GeHP detectors (Canberra[®] and Ortec[®]) at LSCE. ²¹⁰Pb_{xs}, was calculated by
174 subtracting the supported activity from the total ²¹⁰Pb activity (measured at 46.5 keV) using
175 two ²²⁶Ra daughters, i.e. ²¹⁴Pb (average count at 295.2 and 351.9 keV) and ²¹⁴Bi (609.3
176 keV). When insufficient matter was available (<5 g), counting was performed with the same
177 well-type Ge detector as for rainfall analyses. All measurements were corrected for
178 background level determined every two months as well as for detector and geometry
179 efficiencies. All results were expressed in Bq kg⁻¹. Activities were also decay corrected to
180 the sampling date. Counting time reached a maximum of ca. 13 x 10⁴ s for rainwater
181 samples and ca. 25 x 10⁴ s for soil and sediment samples, to optimize counting statistics.
182 Counting efficiencies and reliability were conducted using internal and certified International
183 Atomic Energy Agency (IAEA) standards prepared in the same specimen cups as the
184 samples. Efficiencies were interpolated for ⁷Be energy. Uncertainties on radionuclides
185 activities were ca. 10% for ²¹⁰Pb_{xs}, 20% for ⁷Be and up to 30% for ¹³⁷Cs.

186 3.3. Estimates of fresh sediment (F) and surface soil (α) contributions to suspended loads

187 Respective proportions of (1) fresh sediment vs. particles isolated from recent fallout and,
188 (2) surface soil -derived particles and subsurface particles (mobilized from gullies and

189 riverbanks) were estimated in TSS load. The fresh sediment proportion in TSS load was
190 estimated following the method (Eq. 1) proposed by Matisoff et al. (2005):

$$191 \quad F = 100 \times [(A/B) / (A_0/B_0)] \quad (1)$$

192 where F is the percentage of fresh sediment, A and B are the ^7Be and $^{210}\text{Pb}_{\text{xs}}$ activities in
193 suspended sediment (Bq kg^{-1}) and A_0 and B_0 are the ^7Be and ^{210}Pb activities in rainfall (Bq
194 Γ^{-1}). Although the spatio-temporal variability of ^7Be and ^{210}Pb wet deposition may be
195 important for longer events or for successive storms, this study focused on a single event of
196 short duration and the use of a single value to characterize $^7\text{Be}/^{210}\text{Pb}$ in rainfall was shown
197 to be meaningful (Gourdin et al., 2014).

198 Proportion of surface soil -derived particles in a given sediment sample was estimated with
199 Eq. 2 (e.g., Brigham et al., 2001; Olley et al., 2012):

$$200 \quad \alpha = 100 \times [(C_{\text{Ssample}} - C_{\text{Ssubsurf.}}) / (C_{\text{Ssurf.soil}} - C_{\text{Ssubsurf.}})] \quad (2)$$

201 where α is the percentage of particles derived from surface soil, C_{Ssample} is the ^{137}Cs activity
202 in the sample, $C_{\text{Ssubsurf.}}$ is the mean ^{137}Cs activity in the subsurface soils and $C_{\text{Ssurf.soil}}$ is the
203 mean ^{137}Cs activity in the surface soils (top 0-2 cm).

204 3.4. Checking the field recovery procedure and the assumptions underpinning the ^7Be 205 method

206 In order to determine the efficiency and reproducibility of radionuclide recovery by co-
207 precipitation, experiments were carried out at LSCE. Six aliquots (2 L each) of rainwater
208 were prepared by adding aluminium chloride hexahydrate. Three of them were completely
209 evaporated using heating plates at 150°C for two days, assuming that 100% yields are
210 obtained by total evaporation (Cazala et al., 2003). Co-precipitation was conducted on the
211 three other aliquots using the procedure described in section 3.2. Supernatants were also

212 evaporated to determine the residual radionuclide activity that might still be present in the
213 solution after precipitation.

214 A simple experiment was carried out to assess ^7Be adsorption kinetics on soil particles
215 during rainfall. Five aliquots of a composite topsoil sample collected in the Houay Pano
216 catchment in Laos were mixed with rainwater (4.4 g L^{-1}) and centrifuged at high velocity
217 ($25,000 \text{ rpm}$, *ca.* $78650 \times g$) using the Beckman Coulter[®] J-26 XP air-cooled centrifuge
218 facility at UMR Bioemco. The total contact time between soil particles and rainwater,
219 including acceleration and deceleration phases of the centrifuge, were 13, 23, 31, 41 and
220 72 min. After removal of supernatants, recovered sediments were evaporated and analysed
221 by gamma spectrometry as described in section 3.2.

222 In order to check the underlying assumption that fresh sediment labelling is characterized
223 by a $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$ similar to that of rainwater, ratios were compared in both rainfall and
224 overland flow water. To this end, overland flow and rainfall samples were collected
225 simultaneously at the outlet of a 1-m^2 experimental plot and of a *ca.* 8-m^2 rain-collector
226 during the June 1 rainfall event at the field site. The experiment was conducted on a fallow
227 soil with 33% slope and 60% vegetation cover (*ca.* 10 cm high). The rain collector was
228 installed at 1.8 m height from the soil surface to avoid splash contamination. Rainwater and
229 overland flow samples were collected in plastic bottles at the outlet of the plot and of the
230 rain collector. Radionuclide activities were determined in TSS samples dried in an oven,
231 and radionuclide recovery of rainfall samples was realized by co-precipitation as described
232 in section 3.2. Radionuclide stock variations for the experimental plot were calculated with 1
233 min- steps during the event using Eq. 3:

$$234 \quad \Delta S_t = I_t - E_t \quad (3)$$

235 Where ΔS_t is the stock variation of the plot, I_t is the amount of radionuclide supplied by
236 rainfall to the plot and E_t is the amount of radionuclide exported from the plot by overland
237 flow, between time $t-1$ and time t (expressed in Bq m^{-2}).

238 3.5. Particle size distribution measurements

239 Particle size distribution (PSD) was analysed after a 48-h rehydration of TSS samples (≈ 1
240 g) in 50 mL of distilled water, followed by a 5-min immersion in a Branson 2510 ultrasonic
241 cleaning bath. We used the laser diffraction system (Malvern[®] Mastersizer 2000) coupled to
242 a liquid dispersing unit (Hydro 2000G) both available at the Earth Science Department
243 (University Paris-Sud, Orsay, France). As sediment samples were dried and rehydrated,
244 these PSD do not correspond to the “effective” PSD (Jouon et al., 2008). However, as no
245 dispersing agent was used, the distributions provided here also differ from “absolute” PSD.
246 The protocol was adjusted in order to ensure particle suspension without breaking all the
247 aggregates and allowing flocs’ formation in the presence of organic matter, which is
248 assumed to occur naturally in the stream. Replicate measurements were realized to check
249 reproducibility (*ca.* $\pm 0.5 \mu\text{m}$), and the timing of analysis was adapted to optimize signal
250 stability. The PSD of each sample was obtained for 100 grain size classes ranging between
251 0.02 and 2000 μm . The parameter chosen for comparison between the particle size
252 distributions is d_{50} , corresponding to the median diameter of sediment particles (expressed
253 in μm) with 50% of total volume of particles in the sample below this grain size (e.g. Jouon
254 et al., 2008; Grangeon et al., 2012).

255 3.6. Water electrical conductivity measurement

256 In order to characterize stream / river baseflow dilution by storm event water, water
257 electrical conductivity was monitored every 6-min at the inlet of each gauging station using
258 Schlumberger in situ CTD probes, and additional measurements were conducted using an
259 YSI[®] 556 probe on each collected sample.

260 4. Results

261 4.1. Checking methodological assumptions

262 4.1.1. Radionuclide recovery procedure

263 Reproducibility and efficiency of fallout radionuclide recovery procedures used in this study
264 are presented for ^7Be . Reproducibility was slightly better when conducting the total
265 evaporation procedure (as described in Cazala et al., 2003), with a deviation of ca. 0.5%
266 between triplicates, than when achieving co-precipitation (5% deviation). Differences
267 induced by both treatments were lower than the 10% analytical uncertainty associated with
268 ^7Be activities measured by gamma spectrometry. The recovery was equivalent for both co-
269 precipitation ($90 \pm 6 \text{ mBq L}^{-1}$) and evaporation ($86 \pm 6 \text{ mBq L}^{-1}$) procedures. Activities
270 measured in the supernatant (co-precipitation procedure) remained below the lower
271 instrumental detection limits (<3% of co-precipitated sample activity) and a ca. 100%
272 recovery of fallout radionuclides can therefore be assumed.

273 4.1.2. Fallout radionuclide adsorption kinetics

274 The adsorption kinetics experiment did not show any significant variation in ^7Be and $^{210}\text{Pb}_{\text{xs}}$
275 activity with time. The ^7Be activity in rainwater used in the experiment was $15 \pm 2 \text{ Bq kg}^{-1}$
276 and the initial ^7Be activity was null for soil particles. Mean activities of $10 \pm 3 \text{ Bq kg}^{-1}$ (range:
277 7 ± 2 to $13 \pm 3 \text{ Bq kg}^{-1}$) for ^7Be and $29 \pm 3 \text{ Bq kg}^{-1}$ (range: 25 ± 4 to $32 \pm 4 \text{ Bq kg}^{-1}$) for
278 $^{210}\text{Pb}_{\text{xs}}$ were determined in particles. Taking into account all analytical uncertainties, total
279 rainwater ^7Be adsorption was fulfilled at ca. 75-100% for all samples. The supply of $^{210}\text{Pb}_{\text{xs}}$
280 by rainfall ($2.0 \pm 0.2 \text{ Bq kg}^{-1}$) was low compared to its initial content in the soil ($26 \pm 2 \text{ Bq kg}^{-1}$
281 1). However, the activity in $^{210}\text{Pb}_{\text{xs}}$ measured in the soil after the experiment ($28 \pm 2 \text{ Bq kg}^{-1}$)
282 remained consistent with the occurrence of additional sorption. Furthermore, ^7Be - and
283 $^{210}\text{Pb}_{\text{xs}}$ - sorption by soil particles occurred in less than 13 min, corresponding to the shortest
284 contact time between water and soil particles that could be achieved during the experiment.

285 ^7Be and $^{210}\text{Pb}_{\text{xs}}$ labelling of soil particles by rainfall occurred very quickly as shown in other
286 studies (e.g. Taylor et al., 2012).

287 4.1.3. Fresh sediment labelling by rainfall

288 During the 1-m² plot experiment conducted on June 1, a 45 min- storm event with 11-mm
289 cumulative rainfall occurred. It triggered the export of 8.5 L of overland flow and 20 g of
290 fresh sediment. TSS samples (n = 13) were mixed together to form 3 successive composite
291 samples. Rainfall was characterised by activities ranging 57 - 171 ± 30 mBq L⁻¹ for ^7Be and
292 37 - 166 ± 30 mBq L⁻¹ for $^{210}\text{Pb}_{\text{xs}}$. Initial activities in surface soil (5 upper mm) collected on
293 May 31 before rainfall were 8 ± 1 Bq kg⁻¹ for ^7Be and 60 ± 2 Bq kg⁻¹ for $^{210}\text{Pb}_{\text{xs}}$. Overland
294 flow collected during rainfall displayed activities ranging 31 - 219 mBq L⁻¹ for ^7Be and 11 -
295 345 mBq L⁻¹ for $^{210}\text{Pb}_{\text{xs}}$. Related activities in suspended sediments ranged 24 - 95 ± 10 Bq
296 kg⁻¹ for ^7Be and 10 - 100 ± 10 Bq kg⁻¹ for $^{210}\text{Pb}_{\text{xs}}$. By comparison of radionuclide inputs and
297 exports (expressed in Bq m⁻² min⁻¹) from the experimental plot, stocks of radionuclides were
298 calculated for each time step using Eq. 3. The evolutions of these stocks during each of the
299 45 minutes of the event are plotted versus the TSS concentration in the corresponding
300 overland flow exported from the experimental plot on Fig. 2.

301 [Fig. 2]

302 Stock variations were correlated with TSS concentration for ^7Be ($r^2 = 0.75$; Fig. 2a), $^{210}\text{Pb}_{\text{xs}}$
303 ($r^2 = 0.89$; Fig. 2b) and ^{137}Cs ($r^2 = 0.88$; Fig. 2c), confirming that radionuclides were
304 adsorbed by particles. Rainfall ^7Be and $^{210}\text{Pb}_{\text{xs}}$ inputs compensated the exports by TSS
305 loads below 2 g L⁻¹ in the overland flow. Rainwater brought a total amount of 1.02 Bq m⁻² for
306 ^7Be and 1.01 Bq m⁻² for $^{210}\text{Pb}_{\text{xs}}$ (ratio: 1.0 ± 0.2). In the same time, overland flow exported
307 1.30 Bq m⁻² for ^7Be and 1.59 Bq m⁻² for $^{210}\text{Pb}_{\text{xs}}$ (ratio: 0.8 ± 0.2). Comparing total cumulative
308 inputs and exports, stock depletions were -0.27 Bq m⁻², -0.59 Bq m⁻² and -0.02 Bq m⁻²
309 during the event for ^7Be , $^{210}\text{Pb}_{\text{xs}}$ and ^{137}Cs , respectively. However, as no soil sample was

310 collected after the event, control of the stock balance could not be achieved. Overland flow
311 particles F were estimated to $81 \pm 20\%$, consistent with the assumption that freshly labelled
312 sediments have ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{xs}}$ similar to rainfall. Taking into account analytical errors
313 associated with gamma - counting and their impact on ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{xs}}$ estimates, this result
314 strengthens the reliability of this ratio to fingerprint fresh sediment supply at the onset of the
315 rainy season.

316 4.2. Application of the tracing method to a flood

317 4.2.1. Composition of potential sediment sources within the catchment

318 Mean radionuclide characteristics of surface soils, gullies and stream banks materials
319 collected in the catchment are reported in Table 1.

320 [Table 1]

321 Surface and subsurface (stream banks and gullies) sources were best discriminated by
322 their ${}^{137}\text{Cs}$ activity that is higher in surface soils. Mann-Whitney *U*-tests indicate statistically
323 significant differences between surface and subsurface sources samples at $p < 0.001$ for
324 both ${}^{137}\text{Cs}$ and ${}^{210}\text{Pb}_{\text{xs}}$ activities.

325 4.2.2. Hydro-sedimentary characteristics of the May 23 flood

326 The particles exported during a flood at the onset of the rainy season 2012 in the Houay
327 Xon catchment were collected successively all along the event at nested stations (Fig. 1)
328 and analysed to investigate their sources and dynamics. The main characteristics of this
329 event are described thereafter. The studied flood was triggered by a storm that occurred on
330 May 23 2012 between 11:36 am and 12:24 pm. Rainfall intensity reached 85 mm h^{-1}
331 between 11:54 am and 12:00 am, and cumulated 27 mm rainfall in 48 min. This event was
332 below 0.01 y return period value (34.7 mm daily rainfall), according to Bricquet et al. (2003)
333 for the 1950-2000 period. It was the first significant erosive event of the rainy season and

334 the first event with rainfall intensity exceeding 80 mm h^{-1} . Rainfall samples collected during
335 the event displayed ${}^7\text{Be}$ activities ranging between $110 - 330 \text{ mBq L}^{-1}$. The main hydro-
336 sedimentary characteristics of the flood are reported for the three gauging stations in Fig. 3-
337 4-5.

338 [Fig. 3-4-5]

339 The lag time between stream discharge (Q) and rainfall intensity peaks differed at each
340 station. Q increased 10 min after the rainfall peak and reached its maximum 10 min later at
341 S1 (Fig. 3a), whereas at S4, Q rise started during the rainfall peak, and the Q peak
342 occurred *ca.* 15 min later (Fig. 4a), i.e. 5 min before S1. This behaviour suggests an earlier
343 beginning of rainfall on hillslopes located upstream of S4, with a progressive displacement
344 of raincloud toward the location of the automatic weather station and the upstream S1
345 draining area (Fig. 1). Downstream at S10, the lag time between rainfall and Q peaks
346 increased to 70 min (Fig. 5a). The evolution of TSS concentration as a function of stream
347 discharge (Fig. 3b-4b-5b) displayed counterclockwise hysteresis dynamics (Williams, 1989)
348 in the three subcatchments. Even though Q increased faster than TSS concentration at the
349 beginning of the flood, water EC decreased concomitantly in the three stations (Fig. 3c-4c-
350 5c). This evolution of stream EC suggests the progressive mixing of highly mineralized pre-
351 event water (PEW, i.e. groundwater - high EC) with a low TSS concentration by weakly
352 mineralized event water (EW, i.e. overland flow - low EC) with high sediment loads, the
353 proportion of the EW increasing with decreasing EC (e.g., Nakamura, 1971; Pilgrim et al.,
354 1979; Sklash and Farvolden, 1979; Ribolzi et al., 1997; Collins and Neal, 1998). Despite
355 relatively common TSS-Q trends, major differences between the three stations were
356 observed. At S1 the TSS maximum occurred 10 minutes after the water discharge peak
357 and differed markedly between the water rising and recessing stages. During a second
358 stage, TSS increased rapidly at the onset of Q decrease, reflecting the contribution of
359 overland flow loaded with sediments originating from remote areas of the subcatchment.

360 During a third stage, TSS and Q decreased together. Station S4 showed the fastest
361 response to rainfall. In contrast to S1, S4 displayed three discharge peaks. The first (and
362 main) one likely corresponds to the contribution, upstream of the station, of hillslopes
363 relatively close and well connected to the stream channel. The second and/or third peaks
364 rather result from later arrival (at 12:55) of water flow originating from remote parts of the
365 catchment (possibly including that exported from S1 30 min before). The evolution of Q vs.
366 TSS during the rising and falling water stages followed relatively similar pathways (Fig. 4b).
367 At S10, downstream of S4, Q increased with a time-lag of 22 min after the rainfall peak and
368 the maximum Q was reached 45 min later. Two main successive water discharge peaks
369 (12:42 and 13:17) were related to three successive TSS peaks (13:07, 13:33, 13:57; Fig.
370 5a), reflecting contributions to the river from distinct parts of the catchment. Each of the two
371 first TSS peaks occurred *ca.* 30 min after the related Q peak. The first TSS peak (24 g L^{-1})
372 was recorded just before the second Q peak whereas the second and the third TSS peaks
373 (respectively 25 and 22 g L^{-1}) occurred during the recessing stage, 22 and 27 min later.

374 Overall, high TSS concentrations ($> 5 \text{ g L}^{-1}$) were maintained during the recession phase at
375 the three monitoring stations. The amounts of sediment (calculated as described in section
376 3.1) exported from the three subcatchments are summarized in Table 2.

377 [Table 2]

378 Unfortunately, no sample was collected at the highest Q at S1. Therefore, these sediment
379 exports and yields estimates (Table 2) could be slightly underestimated. The highest
380 sediment yield was calculated at S4. It might be related to the larger area covered by teak
381 plantations sensitive to soil erosion (32%) in this subcatchment, that is two-fold higher than
382 in the drainage areas of S1 (14%) and S10 (15%). The river channel morphology and the
383 hillslope-to-river connectivity varied across the area, as mentioned above in the description
384 of hydro-sedimentary characteristics. The higher connectivity between cultivated hillslopes
385 and the river upstream of S4 might also explain the higher sediment yield from this station.

386 4.2.3. Radionuclide measurements and estimates of fresh sediment (F) and surface-derived
387 particle (α) contributions

388 Rainfall activities ranged 0.11-0.33 Bq L⁻¹ and 0.04-0.12 Bq L⁻¹ for ⁷Be and ²¹⁰Pb_{xs}
389 respectively, with a mean ⁷Be/²¹⁰Pb_{xs} of ca. 2.8. Weight fractions of fresh sediment were
390 calculated for all TSS samples using this latter value. As expected, ¹³⁷Cs was not detected
391 in rainfall. No ⁷Be activity (<3 Bq kg⁻¹) could be determined for the deposited sediment
392 sample collected just before the flood (May 22) upstream S4 confirming the almost
393 complete decay of the previous year fallout.

394 At S1 and S4, ¹³⁷Cs activity in TSS increased with water discharge from ca. 1.0 Bq kg⁻¹
395 during the rising stage, then peaked near its maximum level (1.5-2.0 Bq kg⁻¹) and remained
396 nearly constant (ca. 1.5 ± 0.6 Bq kg⁻¹) during the falling stage period (Fig. 3e-4e). The
397 evolution was more variable at S10 (Fig. 5e). During the beginning of the rising stage, ¹³⁷Cs
398 activity in TSS was rather stable (ca. 0.8 ± 0.2 Bq kg⁻¹) with intermediate values between
399 surface soil signature and ¹³⁷Cs-depleted particles, found in gullies and stream banks
400 (Table 1). In contrast, ²¹⁰Pb_{xs} activities measured at S1, S4 and S10 (Fig. 3f-4f-5f) were
401 generally higher than the average level measured in catchment soils (ca. 40 Bq kg⁻¹, Table
402 1), in particular during the rising stage of the flood at S4. This little enrichment may result
403 from the preferential export of fine-grained particles – enriched in fallout radionuclides – by
404 overland flow (e.g., Walling and He, 1999; Matisoff, 2014). However, the values found in
405 TSS remained in the range of bulk surface soils ²¹⁰Pb_{xs} activities (up to 106 ± 3 Bq kg⁻¹).
406 Contrary to ¹³⁷Cs, ⁷Be activities (and corresponding ⁷Be/²¹⁰Pb_{xs}) were higher during the
407 rising stage and then started to drop at peak flow maximum (Fig. 3g-4g-5g), following a
408 dilution pattern consistent with the behaviour of water EC (Fig. 3c-4c-5c). The overall trend
409 in S10 consisted in the mixing of: (i) particles with low ¹³⁷Cs activities – originating from
410 subsurface sources like collapsed riverbanks or deep rills / gully floor erosion (Hancock et
411 al., 2014) – but tagged with ⁷Be and ²¹⁰Pb_{xs} supplied by recent rainfall and (ii) sediments

412 initially originating from surface soils with high ^{137}Cs but low ^7Be activities (which suggests
413 that they were immersed under water in deposition areas such as in swamps – see Huon et
414 al., 2013 – and isolated from recent fallout labelling before being resuspended during the
415 investigated flood).

416 A F value of ca. 10-30% was estimated during flood peaks and falling water stages (Fig. 3h-
417 4h-5h). Values in $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$ were not significantly different in most samples, except during
418 the beginning of the flood rise in S10, resulting in higher F (Fig. 5h). Those large analytical
419 uncertainties were either due to low sediment yields or to the short gamma counting time to
420 allow for analysing the entire sample set. However, proportional calculations show that
421 mixing of (1) 20-25% of fresh sediment originating from stream banks, depleted in ^{137}Cs
422 (ca. 0.4 Bq kg^{-1} , Table I), with (2) 75-80% of ^{137}Cs -labelled surface soil particles (ca. 2.2 Bq
423 kg^{-1} , Table I), would provide estimates of ca. 1.8 Bq kg^{-1} , consistent with the ^{137}Cs activities
424 measured in TSS at S1 and S4 (Fig. 3e-4e). The contribution of fresh sediment was more
425 important at S10, up to ca. 35% at the beginning of the flood (20% of TSS export during the
426 flood). At S1 and S4, ^7Be activities (in Bq L^{-1}) were positively correlated with TSS
427 concentrations ($r^2 = 0.85$ and 0.88 , respectively; Fig. 6a-6b) during the entire flood. This
428 behaviour confirms that ^7Be is transported by the solid phase in the stream samples of the
429 Houay Pano upstream catchment. However, for S10, no clear trend was observed on a
430 similar plot (not shown). Furthermore, ^7Be activities in TSS (in Bq kg^{-1} ; Fig. 5g) decrease
431 when TSS loads increase (Fig. 5a). This trend reflects that particles tagged with ^7Be that
432 are exported from S10 at the beginning of the flood are then diluted by ^7Be -depleted TSS
433 loads, which highlights a different behaviour in the downstream part of the Houay Xon
434 catchment compared with upstream stations of the Houay Pano subcatchment.

435 [Fig. 6]

436 Results obtained at S10 suggest a mixing between a ^7Be -labelled source of fresh sediment
437 and remobilized sediment from the river channel. In addition, variations in the origin of

438 suspended sediment were observed at this station. Mean ^{137}Cs activity measured at S10
439 (ca. 1.0 Bq kg^{-1}) was lower than at S1 (1.4 Bq kg^{-1}) and S4 (1.6 Bq kg^{-1}), revealing a larger
440 contribution of particles originating from subsurface sources at this station, which is
441 consistent with our field observations. Vegetated riverbanks are less sensitive to erosion
442 upstream of S4 where the stream bed does not deeply incise the bedrock. Using Eq. 2
443 (section 3.3), we estimated a contribution of ca. 90% of ^{137}Cs -depleted sediment at the
444 beginning of the flood at S10. A potential explanation would be that those particles were
445 initially supplied to the river by stream bank erosion or collapse during the former wet
446 season(s). At the end of the previous rainy season, i.e. 6 months before the studied flood,
447 this sediment was deposited in the river channel. As the river level decreased during the dry
448 season, subsequent exposure to atmospheric fallout took place at the beginning of the wet
449 season. The morphology of the riverbed upstream of S10 is consistent with the formation of
450 such deposits. The remainder of sediments depleted in both ^{137}Cs and ^7Be were likely
451 “older” particles originating from riverbanks or gullies and deposited in the river channel
452 during the previous years. Furthermore, when plotting estimated F and α for S10, nearly all
453 data points (except 2) are aligned (Fig. 6c). The two outlying samples correspond to the first
454 and the third TSS peaks, which are likely associated with the export of material with a
455 different origin, suggesting the contribution of a third type of source. Both samples are
456 located below the regression line, reflecting their depletion in fallout radionuclides, which
457 suggests that they were supplied by deep gully walls or riverbank erosion processes (Olley
458 et al., 1993, 2012; Hancock et al., 2014) that were not dominant during the rest of the
459 event. When excluding those two samples implying this third secondary source, a negative
460 linear correlation ($r^2=0.97$; Fig. 6c) is observed during the flood. High contributions of fresh
461 sediment are associated with exports of particles originating from subsurface soils, whereas
462 remobilized sediments are mainly originating from surface soils. This observation outlines
463 the existence of a source of fresh sediment, derived from collapsed riverbanks, that
464 represents a significant proportion of the TSS load conveyed at this station.

465 4.2.4. Particle size distribution of suspended sediments

466 The relationship between TSS grain size and water discharge was investigated by
467 comparing d_{50} (median particle size) values for the three stations (Fig. 3d-4d-5d). The
468 lowest d_{50} value (4 μm) was recorded at S4 at the end of the flood whereas, the highest
469 value (12 μm) was measured during the discharge maximum at S10. Mean d_{50} at S1, S4
470 and S10 reached 6.3, 6.9 and 8.1 μm , respectively, reflecting the increasing discharge and
471 the higher competence of the river in downstream direction.

472 All three stations presented high d_{50} during the peaks of discharge that corresponded to the
473 transport of both freshly eroded and remobilized particles. During the recessing stage of the
474 flood, transport was progressively replaced by deposition of particles on the streambed, as
475 d_{50} decreased to 5, 5.5 and 6 μm at S1, S4 and S10 respectively.

476 5. Discussion

477 5.1. Sediment sources and dynamics along the river continuum

478 The time-lag between water and sediment peaks observed at S10 may result from the
479 presence of a dense vegetation cover on both riverbed and banks that represent obstacles
480 to flow propagation (Gurnell, 2007). Although it did not stop completely the transport of
481 upstream particles as the discharge was high, it may have slowed them down and delayed
482 their arrival compared to water flow propagation, by increasing Houay Xon River channel
483 hydraulic roughness (Manning, 1889) in a similar way as the so-called "grassed waterways"
484 (GWW) installed in agricultural lands to combat muddy runoff generated on cultivated
485 hillslopes (Evrard et al., 2008). Furthermore, this effect should increase with the distance of
486 transportation (Heidel, 1956) as S4 sediment exports had to travel approximately 3 km
487 before reaching S10. As reported by Williams (1989), such counterclockwise hysteresis
488 dynamics may occur in highly erodible catchments submitted to prolonged erosion. Similar
489 lagging sediment peaks were also observed in larger catchments receiving the successive

490 contributions from areas characterized by low and high specific sediment yields (Yun-Liang
491 et al., 1985). Inversely, Whiting et al. (2005) reported clockwise hysteresis dynamics for
492 suspended sediment concentration and fallout radionuclide activity (in Bq L⁻¹) recorded at
493 successive stations along the Yellowstone River (samples collected during seven different
494 floods between April and July 2000). Smith and Dragovitch (2009) reported several
495 counterclockwise hysteresis events at the upstream station of nested catchments affected
496 by severe gully and riverbank erosion in south-eastern Australia's uplands. However,
497 clockwise hysteresis patterns were mostly observed in these catchments, and they were
498 interpreted as resulting from sediment exhaustion effects, particularly during multi-rise
499 events.

500 [Fig. 7]

501 The changes in suspended sediment signatures during the flood at the upstream stations
502 (S1 and S4; Fig. 7a-b) indicate that they were mostly derived from surface soils (tagged by
503 20th century ¹³⁷Cs fallout), which is consistent with previous observations made in the
504 Houay Pano catchment (Huon et al., 2013). Previous works by Chaplot et al. (2005)
505 reported the formation of gullies and rills on hillslopes upstream of S4 and S1 in 2001.
506 However, most of those linear features formed during a rainfall event of higher intensity (90
507 mm cumulative rainfall; return period > 2 yrs). This storm took place in August, i.e. at the
508 period of the rainy season with the lowest infiltrability and highest mean monthly rainfall
509 (Patin et al., 2012). During the 23 May 2012 event, no active gully was observed in the field.
510 Furthermore, we could not quantify specifically the contribution of rill erosion to sediment
511 exports for this event (Evrard et al., 2010; Ben Slimane et al., 2013). Nevertheless,
512 suspended material conveyed at the downstream station contained lower ¹³⁷Cs activities
513 (Fig. 5e), suggesting a switch in the source of particles at this scale with the likely
514 contribution of collapsed riverbanks (e.g. Nagle and Ritchie, 2004). Decreasing ¹³⁷Cs fluxes
515 with increasing drainage areas had been previously reported by Whiting et al. (2005) and

516 interpreted as resulting from bank erosion increase in downstream direction. These authors
517 also reported the dominance of new sediment at upstream stations and early in the
518 hydrograph. Indeed, the global trend observed at S10 (Fig. 7c) corresponded to the arrival,
519 at the beginning of the flood, of particles from collapsed riverbanks originating from the
520 Houay Xon River channel section, associated with a first peak of discharge. Then, during
521 the main discharge peak, more particles mobilized from remote surface soils by overland
522 flow were exported. Those materials were mixed with remobilized sediment from the river
523 channel that diluted the fresh sediment input signal. Finally, as Q decreased, remobilized
524 and eroded particles from most remote sources were exported and progressively deposited.

525 Cumulative exports of fresh sediment were estimated to *ca.* 0.3, 3 and 26 Mg for S1, S4
526 and S10, respectively. They represented respectively *ca.* 13, 12 and 20% of the total
527 suspended sediment exports previously estimated (see section 4.2.2; Table 2).

528 Corresponding estimates in individual suspended sediment samples ranged *ca.* 10 - 60% of
529 surface soil-derived particles. Furthermore, the mean contribution of surface-derived
530 particles was estimated to *ca.* 60 and 76% at S1 and S4, respectively, whereas it amounted
531 to only 29% at S10, reflecting the importance of subsurface sources contribution
532 downstream of S4 (Fig. 7).

533 5.2. Methodological assumptions and prospects

534 Due to the absence of pre-event ^7Be labelling (Appendix) the May 23 flood event appeared
535 to be the first major erosive flood of the 2012 rainy season. Therefore, the hypotheses
536 underpinning the use of the $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$ method (Matisoff et al., 2005; Schuller et al., 2006)
537 were simplified. At the plot scale, $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$ in rainwater and fresh sediment showed a
538 strong similarity during a comparable storm to that of May 23. As this study focused on a
539 single event of short duration (*ca.* 1h rainfall with 60% total cumulative water depth in 12
540 min) the use of a single integrated value for rainfall $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$ proved to be meaningful.
541 However, for longer lasting events, progressive decrease of radionuclide content in

542 rainwater may occur (Wallbrink and Murray, 1994; Ioannidou and Papastefanou, 2006;
543 Gourdin et al., 2014). In order to estimate radionuclide signature of fresh sediment inputs to
544 the rivers, overland flow could be collected and analysed instead of rainfall. Indeed, as
545 showed by Chaplot and Poesen (2012), only a limited proportion of soil-detached (and
546 freshly ^7Be -labelled) particles may reach the Houay Pano stream channel and be
547 transported downstream. The transport of those particles by overland flow progressively
548 decreased with rainfall intensity and most of mobilized materials were deposited and
549 remained on hillslopes. Comparable conclusions could be drawn from soil ^{137}Cs inventories
550 (Huon et al., 2013). This behavior may be responsible for the global trend to progressive
551 decrease of ^7Be activity in TSS observed at all stations during the event.

552 Studies investigating $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$ variations in catchments and rivers grew in number during
553 the last years (Taylor et al., 2013). However, they were conducted over a wide range of
554 time (1h–1yr) and spatial scales (0.7–390 km²) for different environmental and climatic
555 contexts, and their results may not be easy to compare to our study. The main $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$
556 values and related F estimates found in the literature are summarized in Table 3.

557 [Table 3]

558 A large seasonal variability was observed for these ratios depending, among other factors,
559 on the origin of air masses evolving throughout the year and across regions (Bourcier et al.,
560 2011). We could, nevertheless, compare our results with those obtained for a composite
561 sediment sample collected during the first erosive flood of the rainy season in central
562 Mexico (Evrard et al., 2010) where similar conditions prevailed. A comparable contribution
563 of fresh sediment (*ca.* $25 \pm 4\%$) was supplied to the stream, suggesting the dominance (75–
564 87%) of processes remobilizing “older” material at the beginning of the wet season.
565 However in both cases, more detailed spatial information is needed to characterize the
566 origin (surface vs. subsurface) of both freshly eroded and remobilized sediment. In this
567 study, we could determine the sources supplying suspended sediment (mainly derived from

568 surface soils) during this early monsoon event and constrain their dynamics. The bulk of
569 exported particles was remobilized from previous year deposits accumulated in the
570 stream/river channel system.

571 The chosen nested approach provided a way to outline changes in the succession of
572 dominant processes along the river system, from headwaters to the outlet with contrasting
573 sensitivities to erosion along the stream path (Table 2) and a variable connectivity between
574 hillslopes and the main river channel. Whilst hillslopes were directly connected to the
575 stream in upper parts of the catchment, the connection between surface sources and the
576 Houay Xon River was less direct in the downstream sections, characterized by a gentler
577 topography and the presence of depositional areas including a swamp and a wider river
578 channel. Furthermore, the variations in land uses and covers characterized by varying
579 sensitivities to erosion observed across the catchment also partly explain the differences in
580 sediment yields calculated in the drainage areas of the 3 stations. The higher sediment
581 yields calculated at S4 might therefore be explained by the presence of well-connected
582 hillslopes covered by teak plantations. Indeed, this particular land use has been detected as
583 generating large quantities of runoff, especially when teak age exceeds 10 years (Patin et
584 al., 2012).

585 Our results suggest that information on sediment sources (i.e., surface vs. subsurface)
586 should be systematically provided when using $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$ to avoid misinterpretations on
587 their variations. Furthermore, given the rather large uncertainties associated with their
588 results due to logistical and analytical constrains that are difficult to reduce, interpretation of
589 these ratios should remain cautious (e.g. by mentioning proportions of fresh sediment
590 instead of sediment ages in days). It should also focus on the identification of sources and
591 processes of sediment export during the main flood stages. Under those conditions, the
592 $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$ method will provide useful constrains on the processes controlling sediment

593 dynamics in rivers and support design and implementation of efficient soil conservation
594 measures to limit erosion.

595 6. Conclusions

596 An early monsoon flood was monitored at three nested stations in the Houay Xon
597 catchment (Laos) and suspended sediment content in fallout radionuclides was analysed
598 throughout the event. Our results showed that rainwater fallout radionuclides were quickly
599 (< 13 min) bound to surface soil particles. Furthermore, freshly mobilized sediments in
600 overland flow displayed a ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{xs}}$ similar to the one measured in rainwater.

601 Consequently, fresh sediment contributions were estimated for each of the three nested
602 sub-catchments. During this first erosive flood of the rainy season, remobilized particles
603 represented the main type of sediment exported, whatever the spatial scale considered.
604 Contribution of sediments originating from surface soils was dominant upstream (69-78%)
605 whereas they only represented 36% of suspended load downstream, highlighting the key
606 role played by land use and hill slope connectivity on sediment delivery to the river in the
607 different subcatchments.

608 Furthermore, fallout radionuclides provided a mean to identify the contribution of ${}^7\text{Be}$ -
609 labelled particles originating from collapsed riverbank sediments, deposited on aerial
610 exposed areas in the river channel.

611 This study highlights the interest of combining ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{xs}}$ measurements with additional
612 parameters providing information on sediment origin in order to avoid misinterpretation of
613 their dynamics. Further work should attempt to use similar combinations of tracers applied
614 to a longer river continuum (integrating more or larger nested subcatchments). Tracking the
615 downstream flood propagation would provide an opportunity to outline the evolution of
616 dominant processes and sources in larger mountainous tropical catchments where
617 excessive erosion results in critical problems. Providing such information appears crucial to

618 design efficient conservation measures in upstream catchments to prevent an excessive
619 supply of sediment to the rivers.

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Figure captions:

Fig. 1: Location of the Houay Xon catchment (top). Houay Xon S10 subcatchment sampling stations and main land uses areas during the study (centre). Location of surface soil, gully and riverbank samples, swamp areas and weather station (bottom).

Fig. 2: ^7Be , $^{210}\text{Pb}_{\text{xs}}$ and ^{137}Cs stock variations vs. total suspended sediment (TSS) concentration in overland flow exported from the 1-m² experimental plot during each of the 45 minutes of the June 1 event. Error bars represent 1 σ uncertainty.

Fig. 3: Evolution of rainfall intensity, stream discharge (Q, thicker solid line), total suspended sediment (TSS) concentration, electric conductivity (EC), median particle size (d_{50}), ^{137}Cs , $^{210}\text{Pb}_{\text{xs}}$ and ^7Be activities and calculated percentage of fresh sediment (F; see text) at upstream station S1 (Houay Pano Stream) during the May 23 flood. River samples: grey circles. Error bars represent 1 σ uncertainty.

Fig. 4: Evolution of rainfall intensity, stream discharge (Q, thicker solid line), total suspended sediment (TSS) concentration, electric conductivity (EC), median particle size (d_{50}), ^{137}Cs , $^{210}\text{Pb}_{\text{xs}}$ and ^7Be activities and calculated percentage of fresh sediment (F; see text) at intermediate station S4 (Houay Pano Stream) during the May 23 flood. River samples: grey circles. Error bars represent 1 σ uncertainty.

Fig. 5: Evolution of rainfall intensity, stream discharge (Q, thicker solid line), total suspended sediment (TSS) concentration, electric conductivity (EC), median particle size (d_{50}), ^{137}Cs , $^{210}\text{Pb}_{\text{xs}}$ and ^7Be activities and calculated percentage of fresh sediment (F; see text) at downstream station S10 (Houay Xon River) during the May 23 flood. River samples: grey circles. Error bars represent 1 σ uncertainty.

Fig. 6: Correlations between total suspended sediment (TSS) concentration and ^7Be activity at S1 (a) and S4 (b); c: relation between calculated percentage of fresh sediment (F) and calculated percentage of particles derived from surface soil (α) at S10 during the May 23 flood. Regression lines only consider black filled circles (c: peaks samples -white filled circles- are excluded from the regression). Error bars represent 1σ uncertainty.

Fig. 7: Evolution of stream discharge (Q, thicker solid line), total suspended sediment (TSS) concentration (small grey circles), calculated proportions of (1) particles derived from surface/subsurface soil (white/black pie chart) and (2) old/fresh sediment (light-grey/dark-grey pie chart) at S1 (a), S4 (b) and S10 (c) during each stage of the May 23 flood (rise-peak-recession).

Figure1
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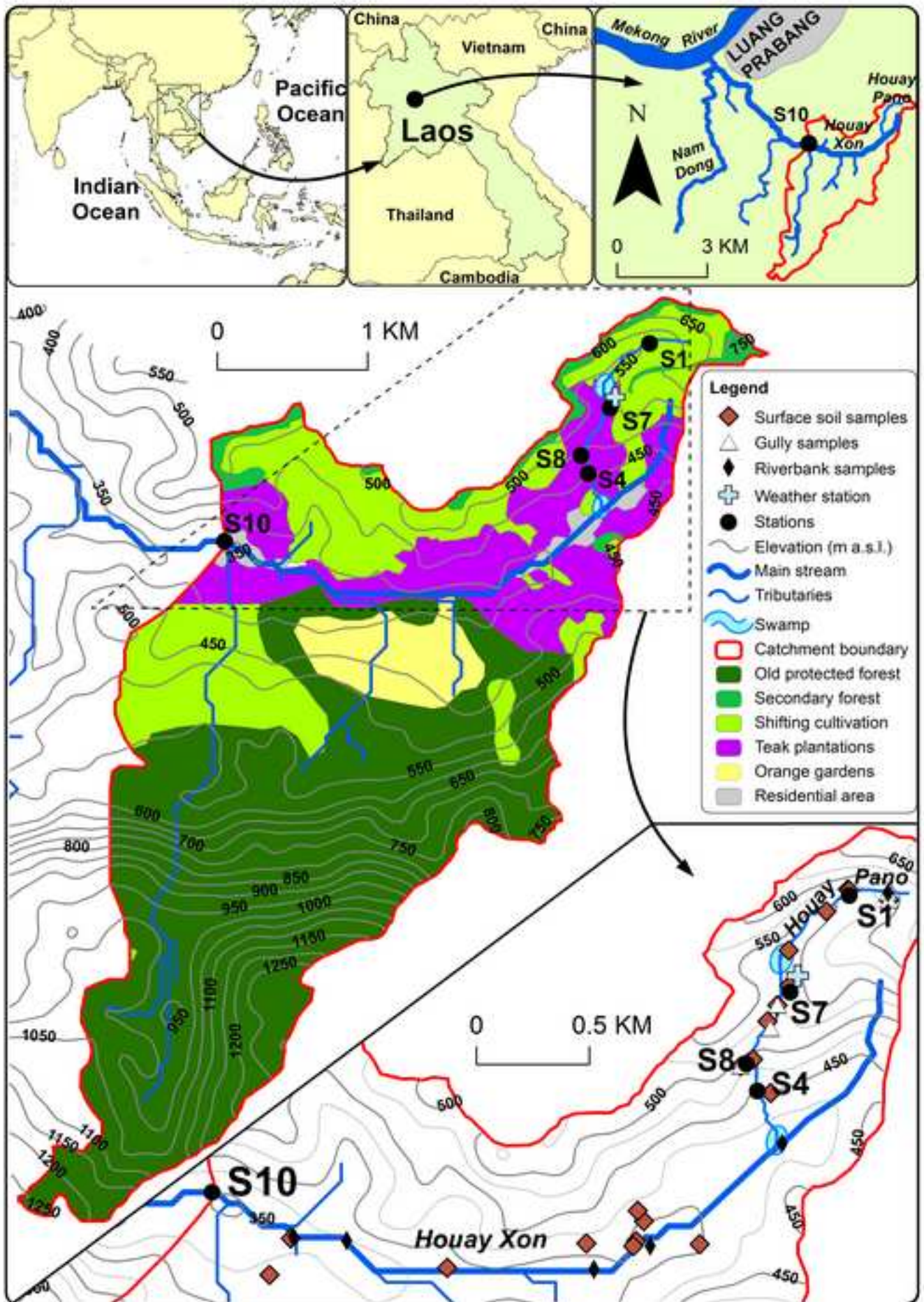


Figure2

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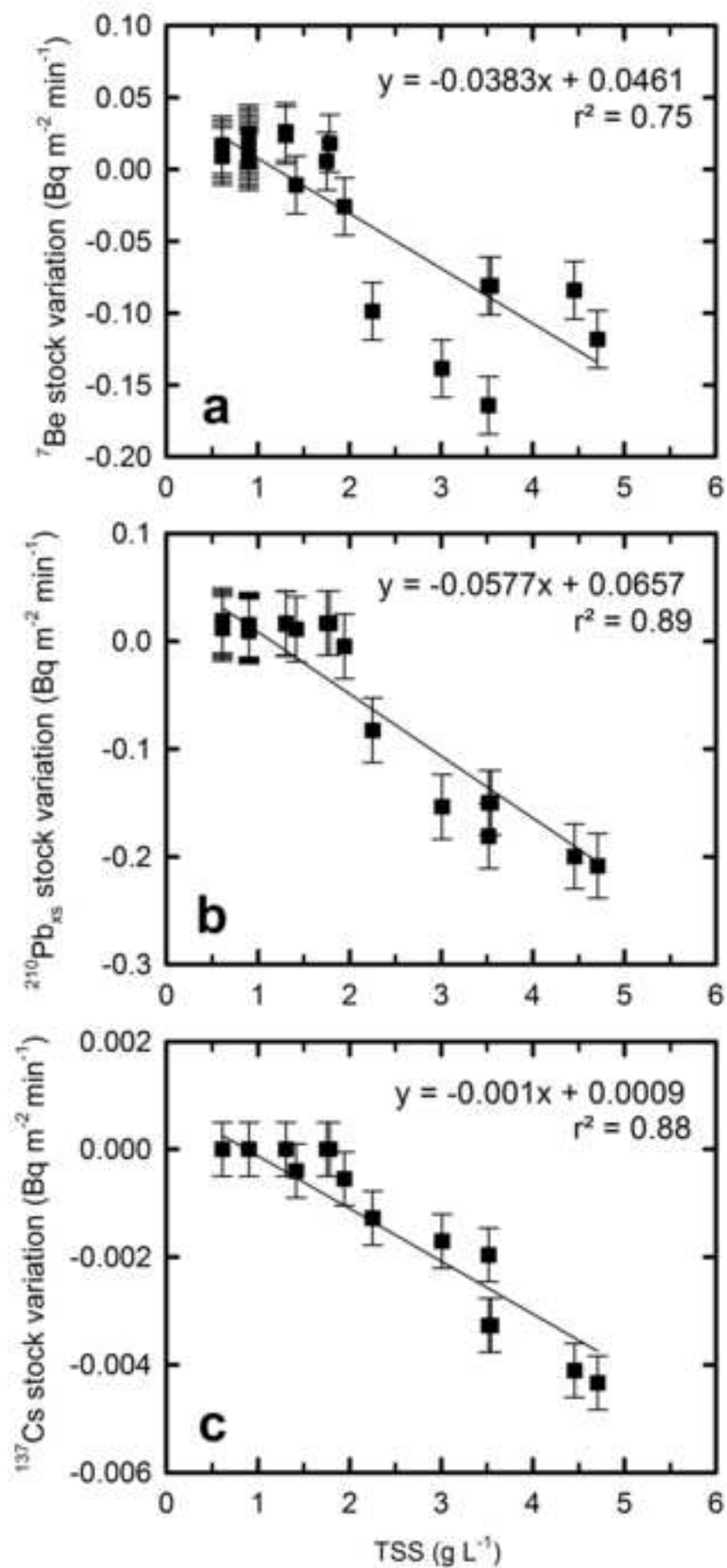


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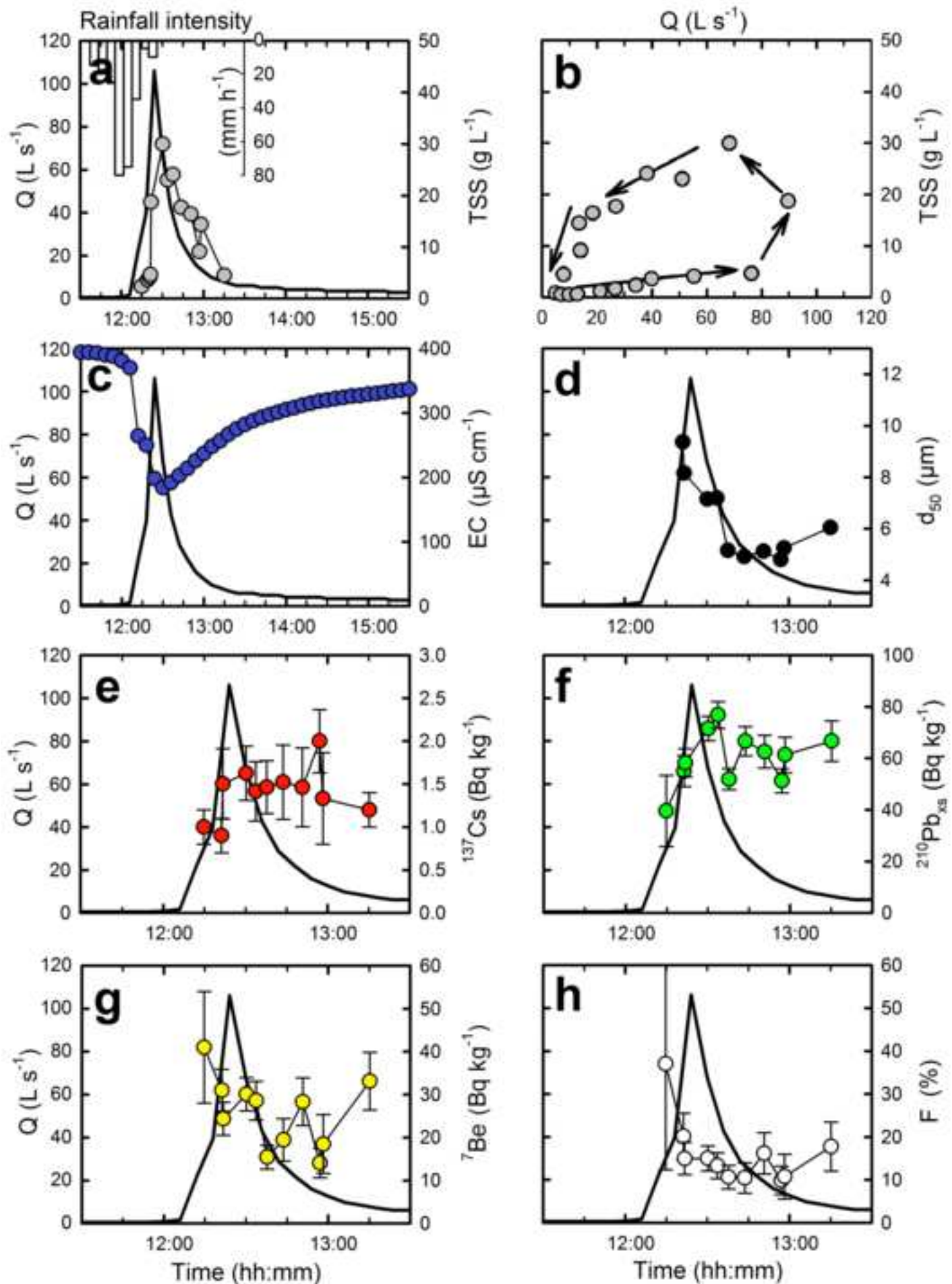


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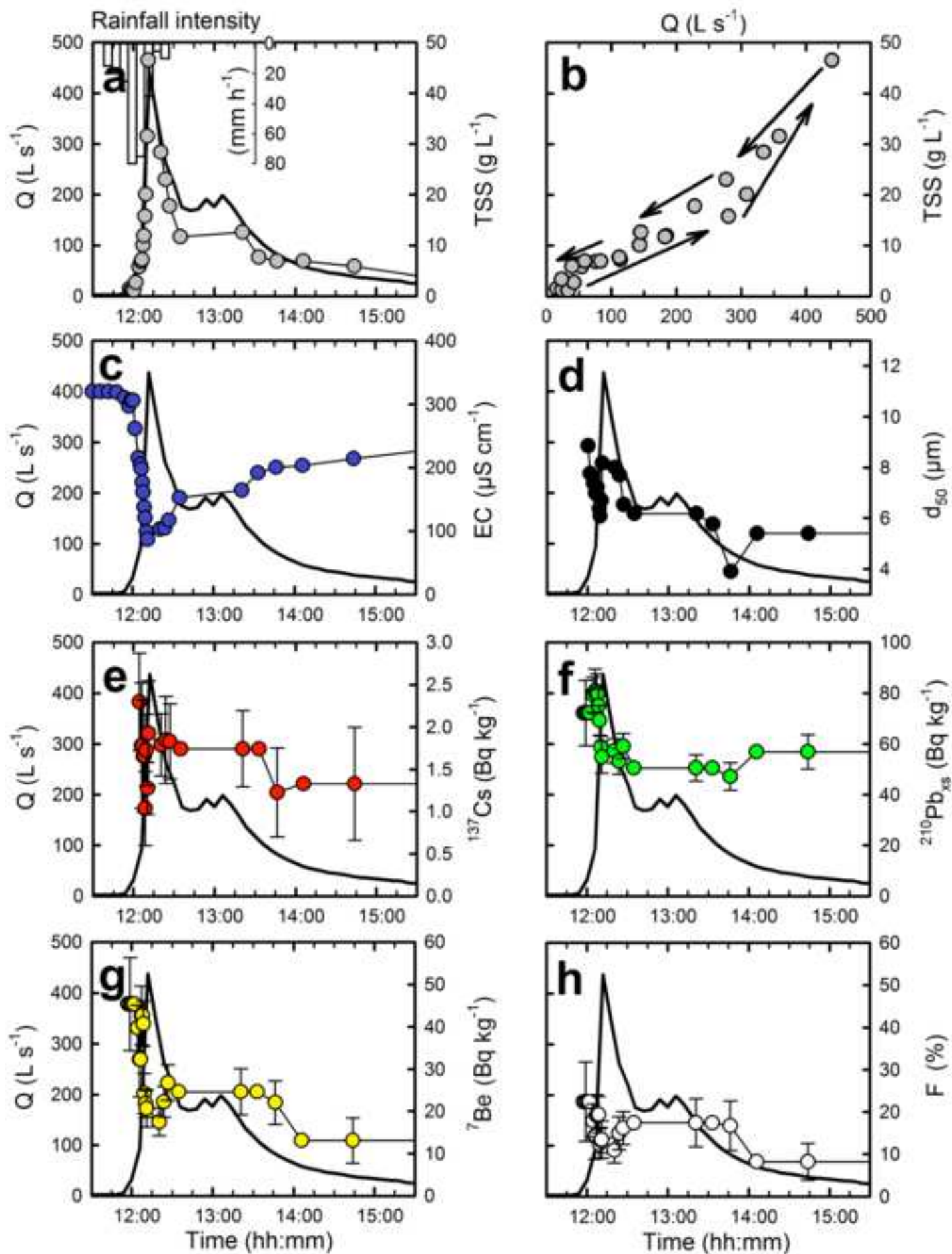


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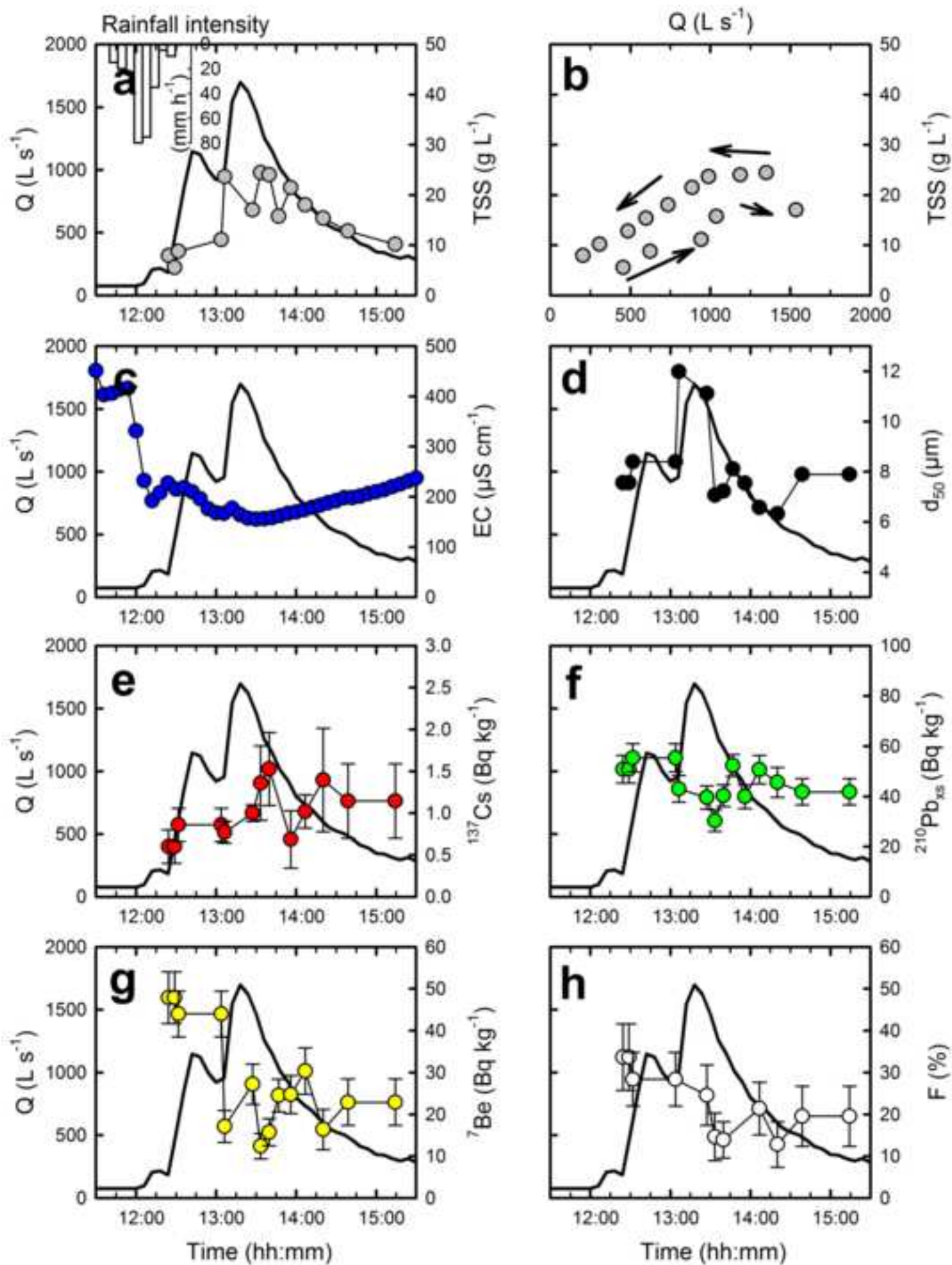


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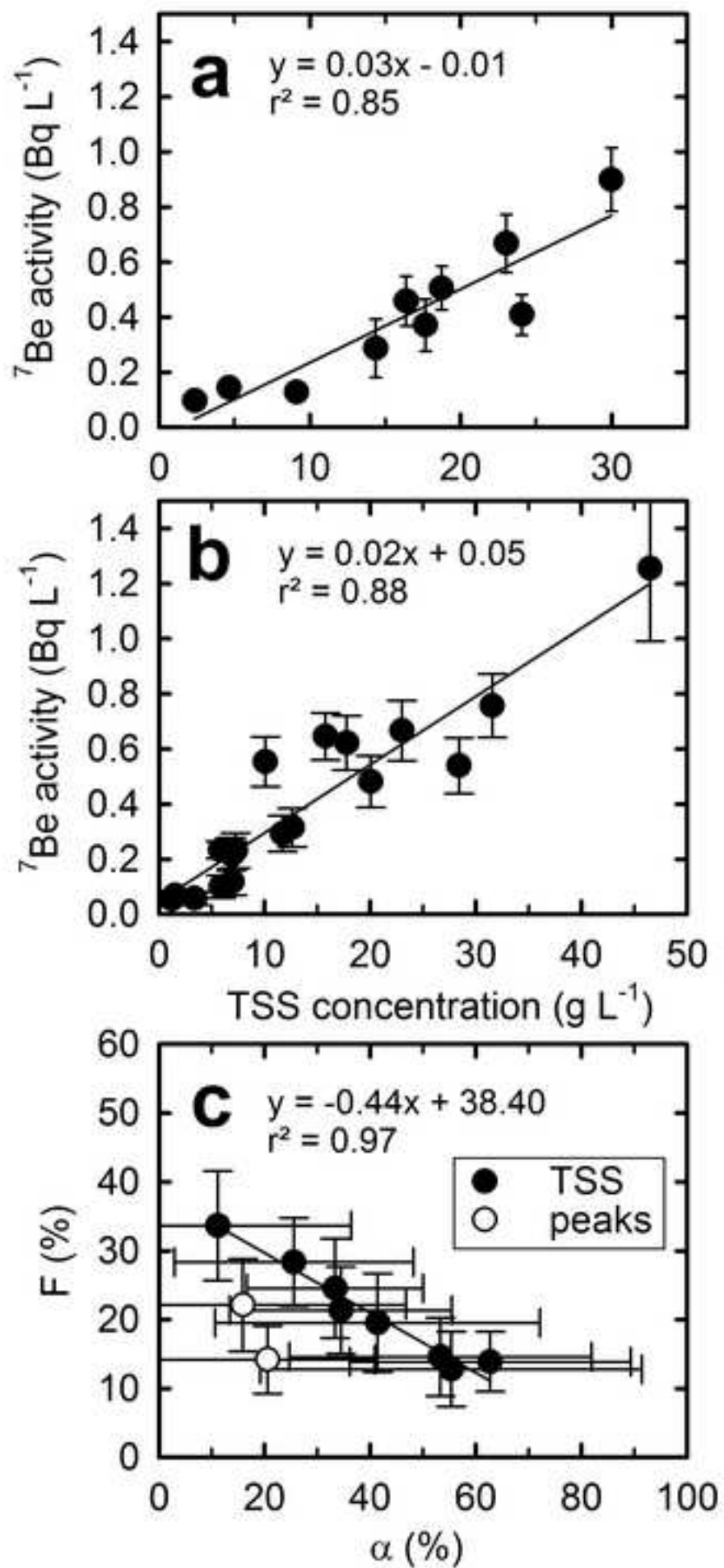


Figure7

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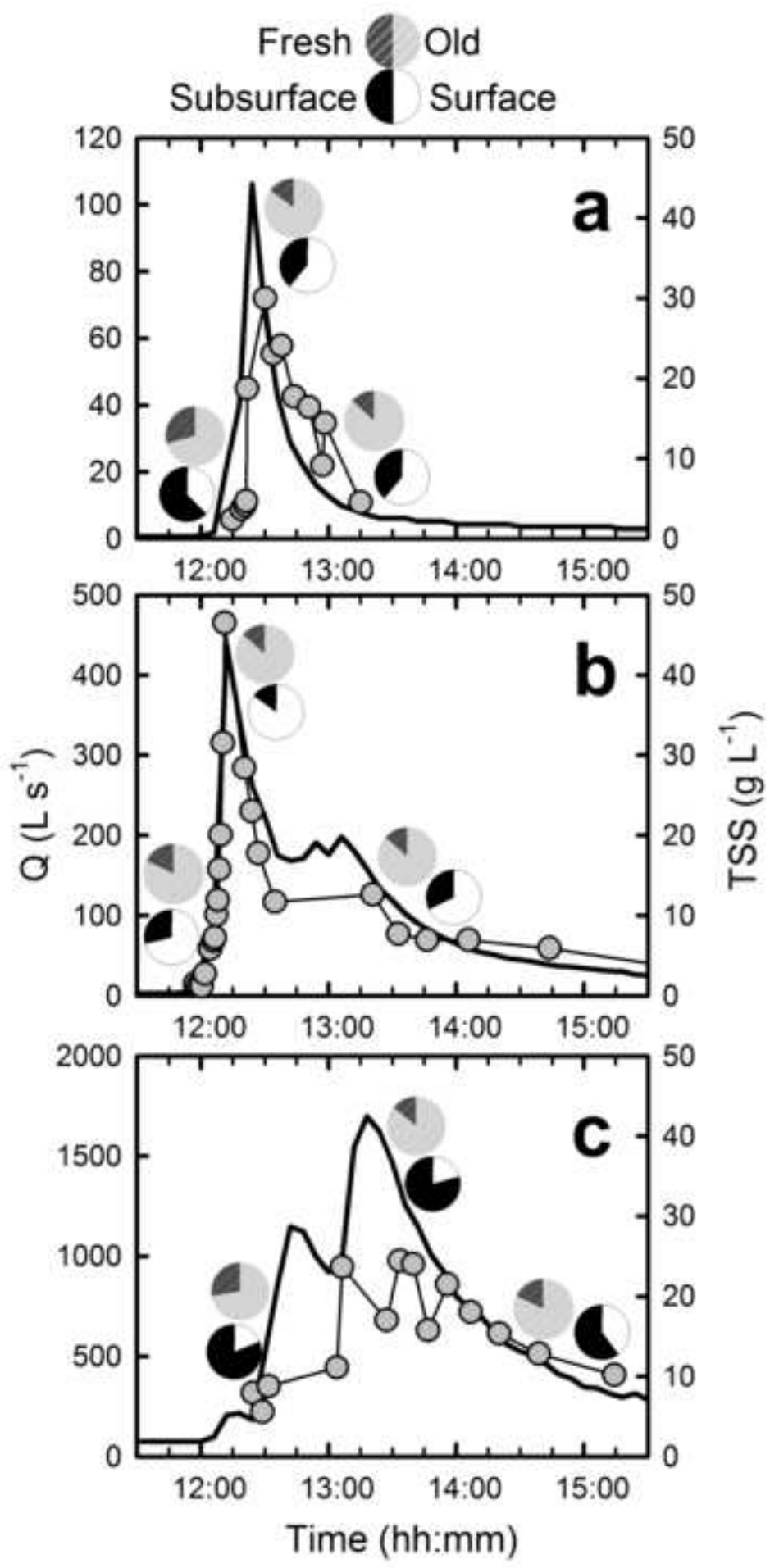


Table1[Click here to download Table: Table 1.doc](#)

Table 1: Mean radionuclide activity (± 1 standard deviation) for surface soils, gullies and stream bank samples in the Houay Pano and Houay Xon catchments

Location	Number of samples	^{137}Cs (Bq kg $^{-1}$)	$^{210}\text{Pb}_{\text{xs}}$ (Bq kg $^{-1}$)
Surface soils*	65	2.2 \pm 0.9	38 \pm 19
Stream banks**	8	0.4 \pm 0.3	14 \pm 11
Gullies**	6	0.4 \pm 0.3	21 \pm 27

*Data from Huon et al. (2013) decay-corrected to 2012 and this study (2012), **this study (2012).

Table 2: Sediment budget for the May 23 flood event

Gauging station	S1	S4	S10
Suspended sediment exports (Mg)	2.3	26	130
Catchment surface (km ²)	0.20	0.60	11.6
Sediment yields (Mg km ⁻²)	11.5	43.3	11.2

Table3

[Click here to download Table: Table 3.doc](#)Table 3: Comparison of ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{xs}}$ and estimates of fresh sediment proportion (F) compared to literature data

References	Location	Spatial scale	Temporal scale	${}^7\text{Be}/{}^{210}\text{Pb}_{\text{xs}}$		F
				rainfall	sediment	
This study	Northern Laos	3 nested subcatchments (0.2-11.6 km ²)	1 storm event	2.8	0.2 – 1.0	8 – 37 %
Evrard et al., 2010	Central Mexico	3 subcatchments (3 – 12 km ²)	8 – 18 storms (6 months)	7- 27	2 – 14	10 – 80 %
Bonniwell et al., 1999	ID, USA	Catchment (390 km ²) - River (ca. 30 km reach)	3 months	<i>n.a.</i>	0.3 – 3.8	18 – 96 %
Matisoff et al., 2005	OH; AL; OR, USA	3 catchments (70 km ²)	Individual storms (1 year)	13 - 17	1.0 – 1.7	6 – 14 %
Huisman et al., 2013	WI, USA	Catchment (12.4 km ²)	7 campaigns (6 months)	3 - 12	0.1 – 5.5	1 – 100 %

n.a. = not available

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