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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 km<sup>2</sup>). Fallout radionuclides (Beryllium-7 -  
15 <sup>7</sup>Be, unsupported Pb-210 -<sup>210</sup>Pb<sub>xs</sub>, and Cesium-137 -<sup>137</sup>Cs) 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 <sup>7</sup>Be-labelling  
18 method were validated by implementing experiments in the laboratory (i.e., rainwater <sup>7</sup>Be  
19 sorption to soil particles) and in the field (i.e., <sup>7</sup>Be: <sup>210</sup>Pb<sub>xs</sub> 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 <sup>7</sup>Be, 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 <sup>7</sup>Be-labelled particles derived from collapsed riverbanks  
29 to sediment export was also demonstrated. This pilot study supports the use of fallout <sup>7</sup>Be  
30 and <sup>210</sup>Pb<sub>xs</sub> 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 (Riboldi 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 ( $^7\text{Be}$ ) 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  $^7\text{Be}$  and  $T_{1/2} = 22.3$  years for  $^{210}\text{Pb}_{\text{xs}}$ ) are particularly relevant to  
57 differentiate between fresh sediment tagged with  $^7\text{Be}$  and older remobilized sediment  
58 depleted in  $^7\text{Be}$ . 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 20<sup>th</sup>  
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  $^7\text{Be}$  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 (Riboldi 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 km<sup>2</sup> catchment) and is continuously monitored at S10 (draining a  
102 11.6 km<sup>2</sup> 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 (Riboldi 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-m<sup>2</sup> 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  $^{222}\text{Rn}$  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 cm<sup>3</sup> (GWL-220-15 Ortec<sup>®</sup>). Most samples were analysed within less  
166 than 53 days (<sup>7</sup>Be 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 <sup>7</sup>Be, <sup>137</sup>Cs and <sup>210</sup>Pb 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<sup>®</sup> and Ortec<sup>®</sup>) at LSCE. <sup>210</sup>Pb<sub>xs</sub>, was calculated by  
174 subtracting the supported activity from the total <sup>210</sup>Pb activity (measured at 46.5 keV) using  
175 two <sup>226</sup>Ra daughters, i.e. <sup>214</sup>Pb (average count at 295.2 and 351.9 keV) and <sup>214</sup>Bi (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<sup>-1</sup>. Activities were also decay corrected to  
180 the sampling date. Counting time reached a maximum of ca. 13 x 10<sup>4</sup> s for rainwater  
181 samples and ca. 25 x 10<sup>4</sup> 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 <sup>7</sup>Be energy. Uncertainties on radionuclides  
185 activities were ca. 10% for <sup>210</sup>Pb<sub>xs</sub>, 20% for <sup>7</sup>Be and up to 30% for <sup>137</sup>Cs.

### 186 3.3. Estimates of fresh sediment (F) and surface soil ( $\alpha$ ) 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  $^7\text{Be}$  and  $^{210}\text{Pb}_{\text{xs}}$  activities in  
193 suspended sediment ( $\text{Bq kg}^{-1}$ ) and  $A_0$  and  $B_0$  are the  $^7\text{Be}$  and  $^{210}\text{Pb}$  activities in rainfall ( $\text{Bq}$   
194  $\Gamma^{-1}$ ). Although the spatio-temporal variability of  $^7\text{Be}$  and  $^{210}\text{Pb}$  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  $\alpha$  is the percentage of particles derived from surface soil,  $C_{\text{Ssample}}$  is the  $^{137}\text{Cs}$  activity  
202 in the sample,  $C_{\text{Ssubsurf.}}$  is the mean  $^{137}\text{Cs}$  activity in the subsurface soils and  $C_{\text{Ssurf.soil}}$  is the  
203 mean  $^{137}\text{Cs}$  activity in the surface soils (top 0-2 cm).

#### 204 3.4. Checking the field recovery procedure and the assumptions underpinning the $^7\text{Be}$ 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  $150^\circ\text{C}$  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  $^7\text{Be}$  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 ( $4.4 \text{ g L}^{-1}$ ) and centrifuged at high velocity  
217 ( $25,000 \text{ rpm}$ , *ca.*  $78650 \times g$ ) using the Beckman Coulter<sup>®</sup> 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  $1\text{-m}^2$  experimental plot and of a *ca.*  $8\text{-m}^2$  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  $\Delta S_t$  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  $\text{Bq m}^{-2}$ ).

### 238 3.5. Particle size distribution measurements

239 Particle size distribution (PSD) was analysed after a 48-h rehydration of TSS samples ( $\approx 1$   
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<sup>®</sup> 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  $\mu\text{m}$ . 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  $\mu\text{m}$ ) 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<sup>®</sup> 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  $^7\text{Be}$ . 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  $^7\text{Be}$  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  $^7\text{Be}$  and  $^{210}\text{Pb}_{\text{xs}}$   
275 activity with time. The  $^7\text{Be}$  activity in rainwater used in the experiment was  $15 \pm 2 \text{ Bq kg}^{-1}$   
276 and the initial  $^7\text{Be}$  activity was null for soil particles. Mean activities of  $10 \pm 3 \text{ Bq kg}^{-1}$  (range:  
277  $7 \pm 2$  to  $13 \pm 3 \text{ Bq kg}^{-1}$ ) for  $^7\text{Be}$  and  $29 \pm 3 \text{ Bq kg}^{-1}$  (range:  $25 \pm 4$  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  $^7\text{Be}$  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,  $^7\text{Be}$ - 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  $^7\text{Be}$  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-m<sup>2</sup> 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<sup>-1</sup> for  $^7\text{Be}$  and  
292 37 - 166 ± 30 mBq L<sup>-1</sup> 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<sup>-1</sup> for  $^7\text{Be}$  and 60 ± 2 Bq kg<sup>-1</sup> for  $^{210}\text{Pb}_{\text{xs}}$ . Overland  
294 flow collected during rainfall displayed activities ranging 31 - 219 mBq L<sup>-1</sup> for  $^7\text{Be}$  and 11 -  
295 345 mBq L<sup>-1</sup> for  $^{210}\text{Pb}_{\text{xs}}$ . Related activities in suspended sediments ranged 24 - 95 ± 10 Bq  
296 kg<sup>-1</sup> for  $^7\text{Be}$  and 10 - 100 ± 10 Bq kg<sup>-1</sup> for  $^{210}\text{Pb}_{\text{xs}}$ . By comparison of radionuclide inputs and  
297 exports (expressed in Bq m<sup>-2</sup> min<sup>-1</sup>) 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  $^7\text{Be}$  ( $r^2 = 0.75$ ; Fig. 2a),  $^{210}\text{Pb}_{\text{xs}}$   
303 ( $r^2 = 0.89$ ; Fig. 2b) and  $^{137}\text{Cs}$  ( $r^2 = 0.88$ ; Fig. 2c), confirming that radionuclides were  
304 adsorbed by particles. Rainfall  $^7\text{Be}$  and  $^{210}\text{Pb}_{\text{xs}}$  inputs compensated the exports by TSS  
305 loads below 2 g L<sup>-1</sup> in the overland flow. Rainwater brought a total amount of 1.02 Bq m<sup>-2</sup> for  
306  $^7\text{Be}$  and 1.01 Bq m<sup>-2</sup> for  $^{210}\text{Pb}_{\text{xs}}$  (ratio: 1.0 ± 0.2). In the same time, overland flow exported  
307 1.30 Bq m<sup>-2</sup> for  $^7\text{Be}$  and 1.59 Bq m<sup>-2</sup> 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<sup>-2</sup>, -0.59 Bq m<sup>-2</sup> and -0.02 Bq m<sup>-2</sup>  
309 during the event for  $^7\text{Be}$ ,  $^{210}\text{Pb}_{\text{xs}}$  and  $^{137}\text{Cs}$ , 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  $85 \text{ mm h}^{-1}$   
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  $80 \text{ mm h}^{-1}$ . 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 ( $24 \text{ g L}^{-1}$ )  
372 was recorded just before the second Q peak whereas the second and the third TSS peaks  
373 (respectively  $25$  and  $22 \text{ g L}^{-1}$ ) 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 ( $\alpha$ ) contributions

388 Rainfall activities ranged 0.11-0.33 Bq L<sup>-1</sup> and 0.04-0.12 Bq L<sup>-1</sup> for <sup>7</sup>Be and <sup>210</sup>Pb<sub>xs</sub>  
389 respectively, with a mean <sup>7</sup>Be/<sup>210</sup>Pb<sub>xs</sub> of ca. 2.8. Weight fractions of fresh sediment were  
390 calculated for all TSS samples using this latter value. As expected, <sup>137</sup>Cs was not detected  
391 in rainfall. No <sup>7</sup>Be activity (<3 Bq kg<sup>-1</sup>) 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, <sup>137</sup>Cs activity in TSS increased with water discharge from ca. 1.0 Bq kg<sup>-1</sup>  
395 during the rising stage, then peaked near its maximum level (1.5-2.0 Bq kg<sup>-1</sup>) and remained  
396 nearly constant (ca. 1.5 ± 0.6 Bq kg<sup>-1</sup>) 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, <sup>137</sup>Cs  
398 activity in TSS was rather stable (ca. 0.8 ± 0.2 Bq kg<sup>-1</sup>) with intermediate values between  
399 surface soil signature and <sup>137</sup>Cs-depleted particles, found in gullies and stream banks  
400 (Table 1). In contrast, <sup>210</sup>Pb<sub>xs</sub> 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<sup>-1</sup>, 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 <sup>210</sup>Pb<sub>xs</sub> activities (up to 106 ± 3 Bq kg<sup>-1</sup>).  
406 Contrary to <sup>137</sup>Cs, <sup>7</sup>Be activities (and corresponding <sup>7</sup>Be/<sup>210</sup>Pb<sub>xs</sub>) 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 <sup>137</sup>Cs activities – originating from  
410 subsurface sources like collapsed riverbanks or deep rills / gully floor erosion (Hancock et  
411 al., 2014) – but tagged with <sup>7</sup>Be and <sup>210</sup>Pb<sub>xs</sub> supplied by recent rainfall and (ii) sediments

412 initially originating from surface soils with high  $^{137}\text{Cs}$  but low  $^7\text{Be}$  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  $^{137}\text{Cs}$   
422 (ca.  $0.4 \text{ Bq kg}^{-1}$ , Table I), with (2) 75-80% of  $^{137}\text{Cs}$ -labelled surface soil particles (ca.  $2.2 \text{ Bq}$   
423  $\text{kg}^{-1}$ , Table I), would provide estimates of ca.  $1.8 \text{ Bq kg}^{-1}$ , consistent with the  $^{137}\text{Cs}$  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,  $^7\text{Be}$  activities (in  $\text{Bq L}^{-1}$ ) 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  $^7\text{Be}$  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,  $^7\text{Be}$  activities in TSS (in  $\text{Bq kg}^{-1}$ ; Fig. 5g) decrease  
431 when TSS loads increase (Fig. 5a). This trend reflects that particles tagged with  $^7\text{Be}$  that  
432 are exported from S10 at the beginning of the flood are then diluted by  $^7\text{Be}$ -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  $^7\text{Be}$ -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  $^{137}\text{Cs}$  activity measured at S10  
439 (ca.  $1.0 \text{ Bq kg}^{-1}$ ) was lower than at S1 ( $1.4 \text{ Bq kg}^{-1}$ ) and S4 ( $1.6 \text{ Bq kg}^{-1}$ ), 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  $^{137}\text{Cs}$ -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  $^{137}\text{Cs}$  and  $^7\text{Be}$  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  $\alpha$  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  $\mu\text{m}$ ) was recorded at S4 at the end of the flood whereas, the highest  
469 value (12  $\mu\text{m}$ ) 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  $\mu\text{m}$ , 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  $\mu\text{m}$  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<sup>-1</sup>) 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 20<sup>th</sup> century <sup>137</sup>Cs 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 <sup>137</sup>Cs 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 <sup>137</sup>Cs 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  $^7\text{Be}$  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  $^7\text{Be}$ -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  $^{137}\text{Cs}$  inventories  
550 (Huon et al., 2013). This behavior may be responsible for the global trend to progressive  
551 decrease of  $^7\text{Be}$  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 km<sup>2</sup>) 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:  $^7\text{Be}$ ,  $^{210}\text{Pb}_{\text{xs}}$  and  $^{137}\text{Cs}$  stock variations vs. total suspended sediment (TSS) concentration in overland flow exported from the 1-m<sup>2</sup> experimental plot during each of the 45 minutes of the June 1 event. Error bars represent 1 $\sigma$  uncertainty.

Fig. 3: Evolution of rainfall intensity, stream discharge (Q, thicker solid line), total suspended sediment (TSS) concentration, electric conductivity (EC), median particle size ( $d_{50}$ ),  $^{137}\text{Cs}$ ,  $^{210}\text{Pb}_{\text{xs}}$  and  $^7\text{Be}$  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 $\sigma$  uncertainty.

Fig. 4: Evolution of rainfall intensity, stream discharge (Q, thicker solid line), total suspended sediment (TSS) concentration, electric conductivity (EC), median particle size ( $d_{50}$ ),  $^{137}\text{Cs}$ ,  $^{210}\text{Pb}_{\text{xs}}$  and  $^7\text{Be}$  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 $\sigma$  uncertainty.

Fig. 5: Evolution of rainfall intensity, stream discharge (Q, thicker solid line), total suspended sediment (TSS) concentration, electric conductivity (EC), median particle size ( $d_{50}$ ),  $^{137}\text{Cs}$ ,  $^{210}\text{Pb}_{\text{xs}}$  and  $^7\text{Be}$  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 $\sigma$  uncertainty.

Fig. 6: Correlations between total suspended sediment (TSS) concentration and  $^7\text{Be}$  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 ( $\alpha$ ) 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  $1\sigma$  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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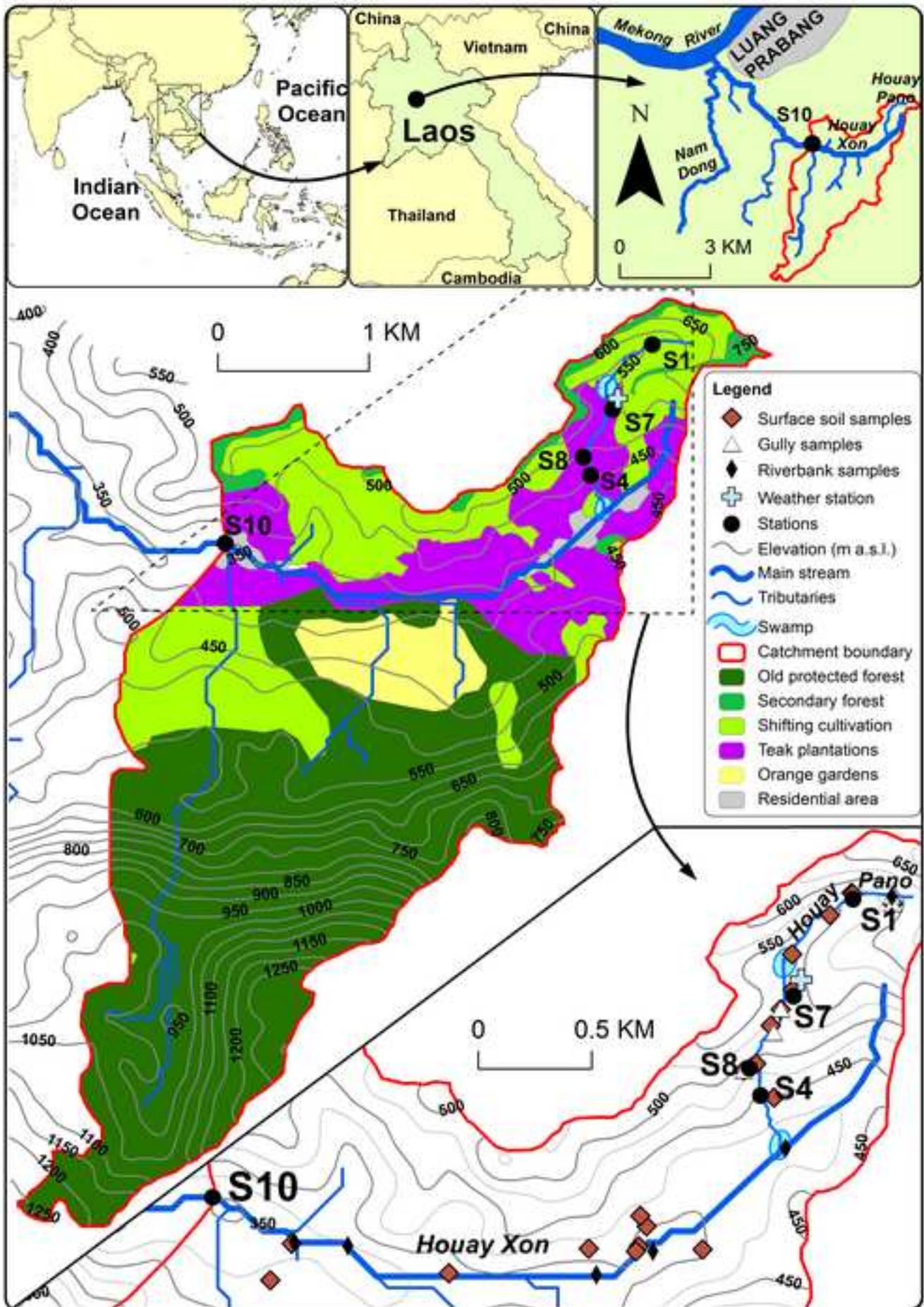


Figure2

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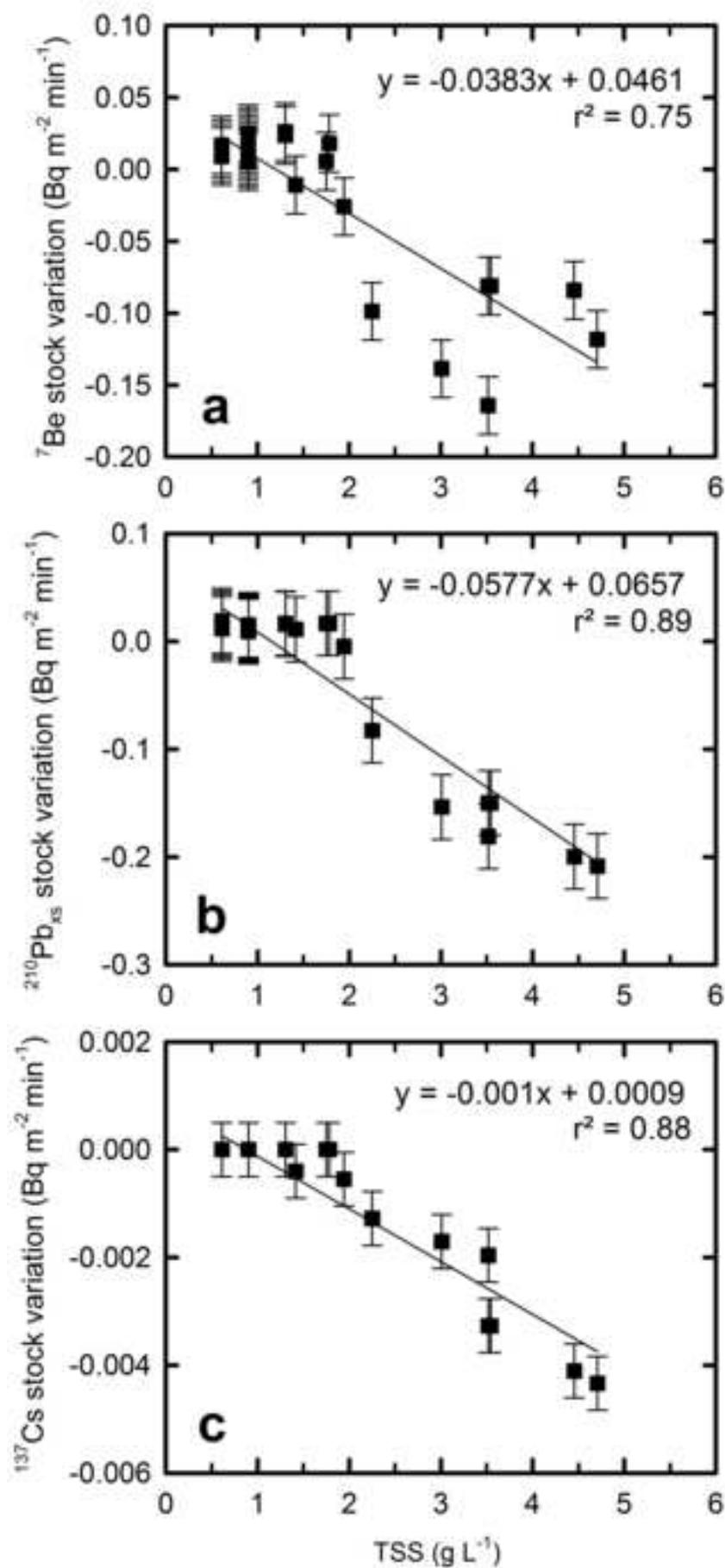


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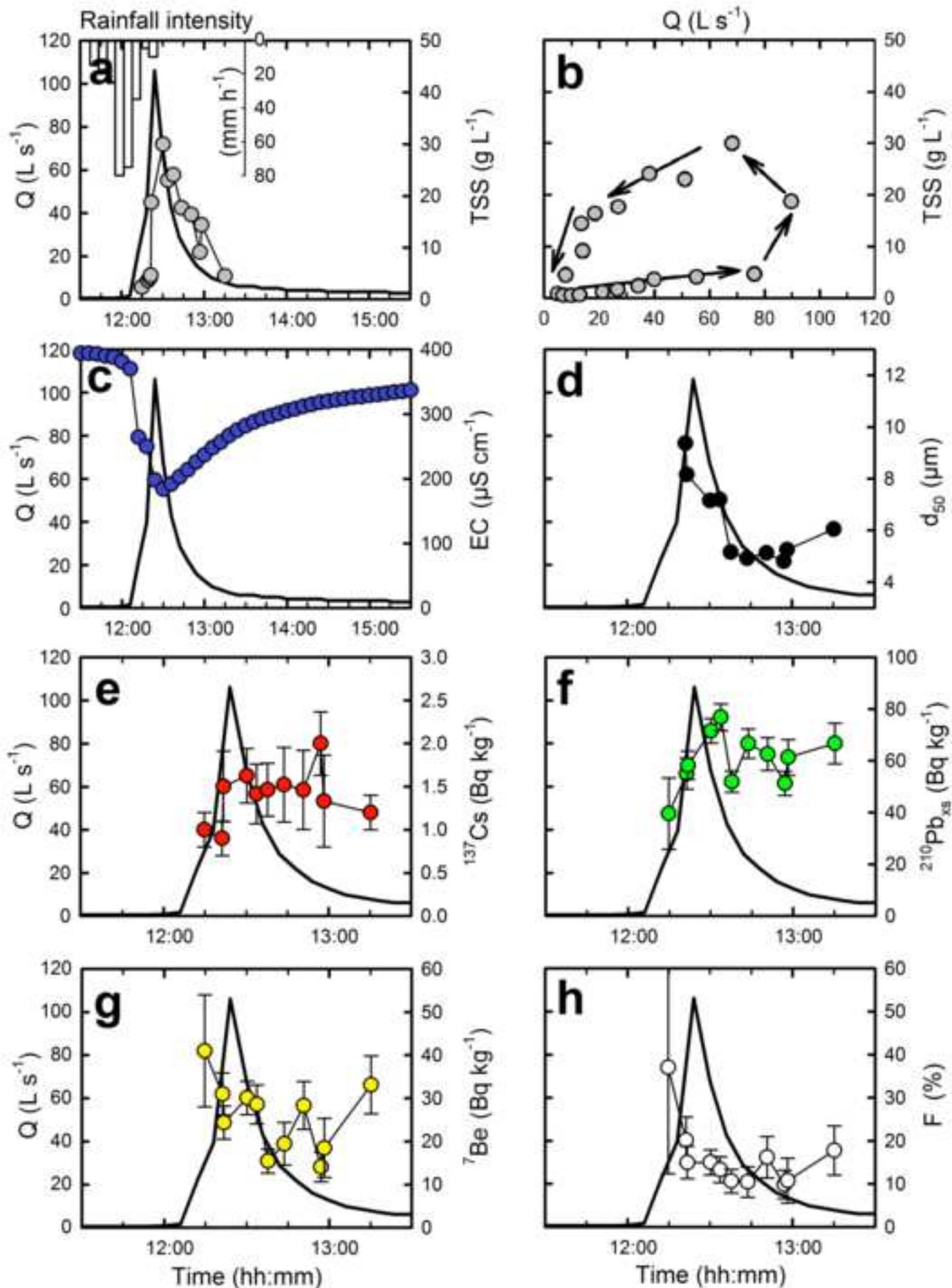


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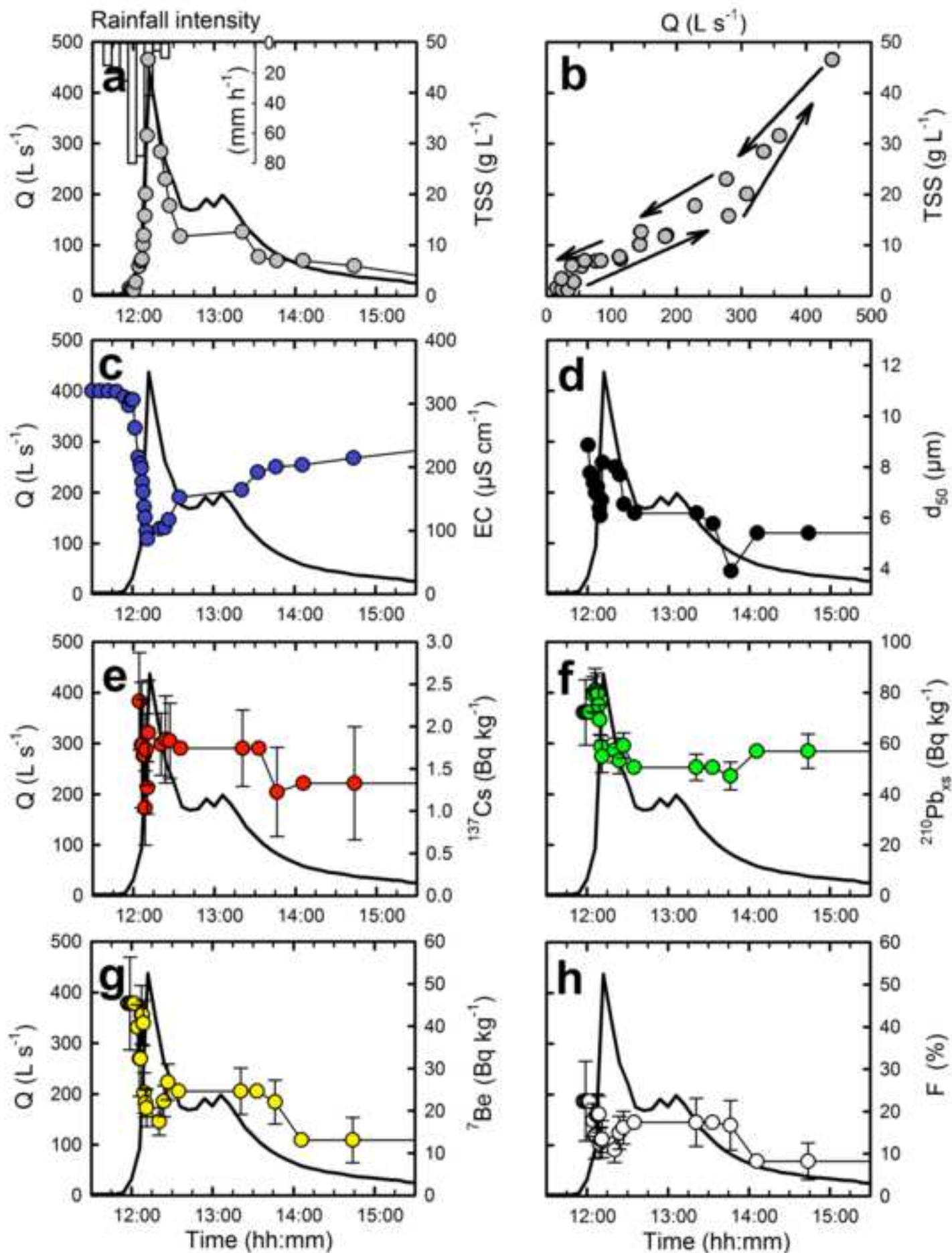


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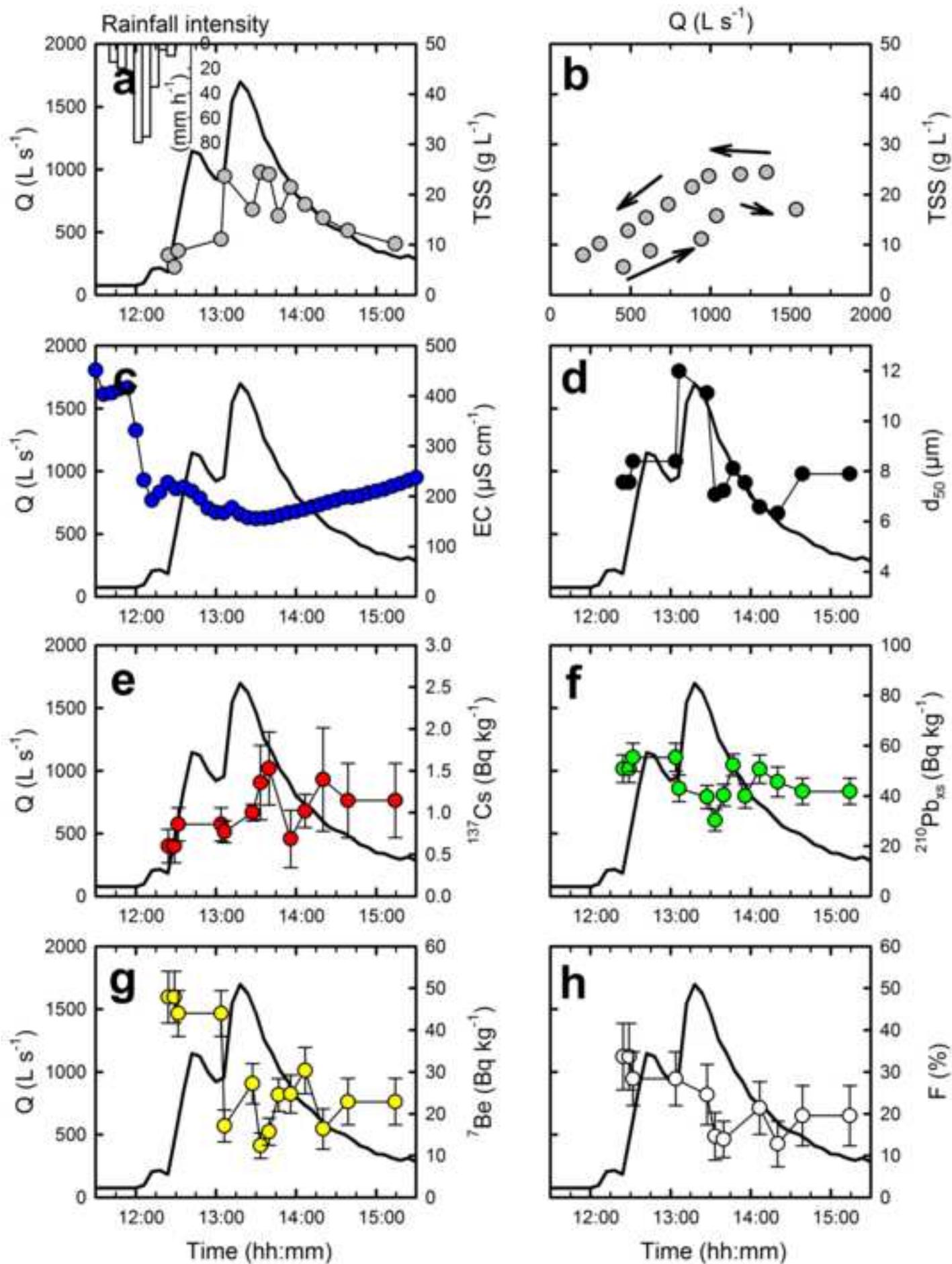


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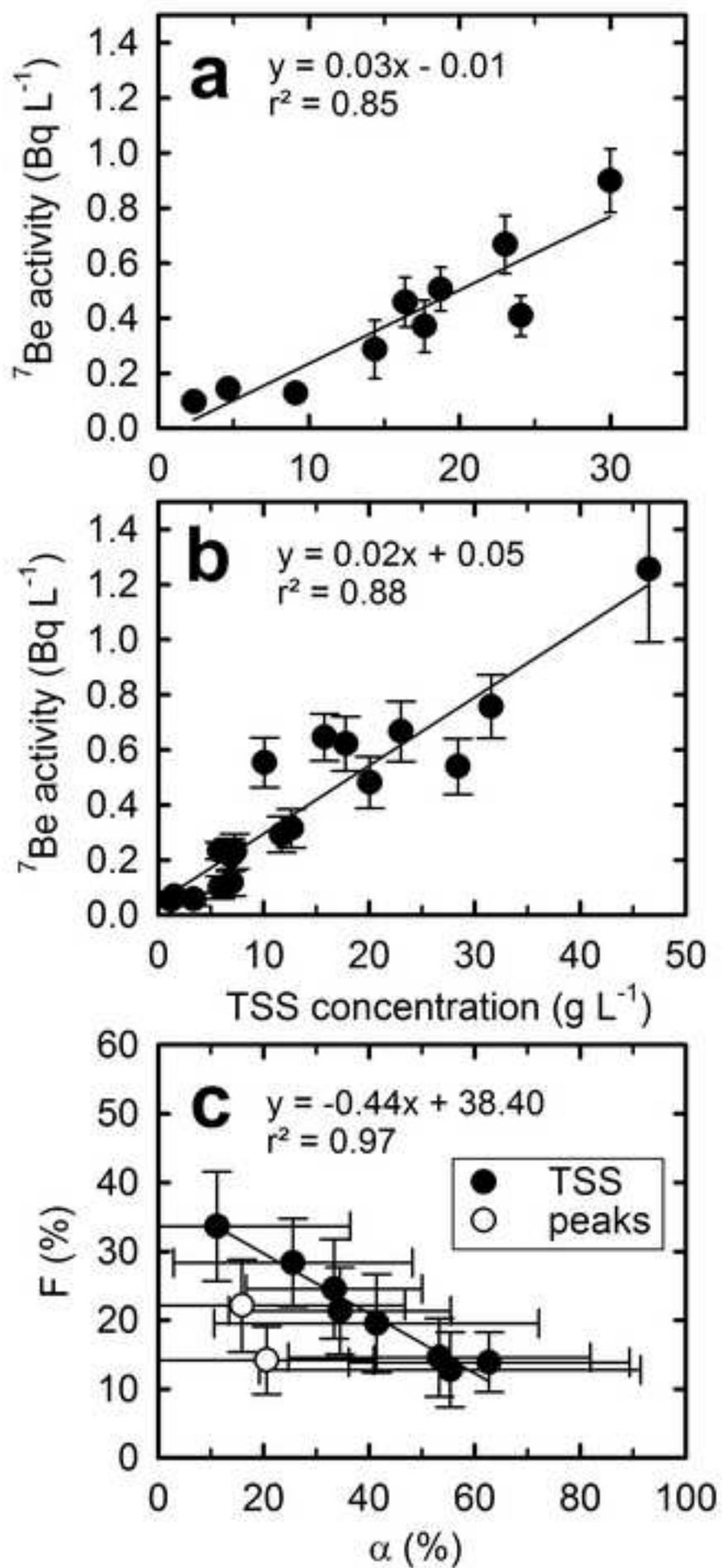
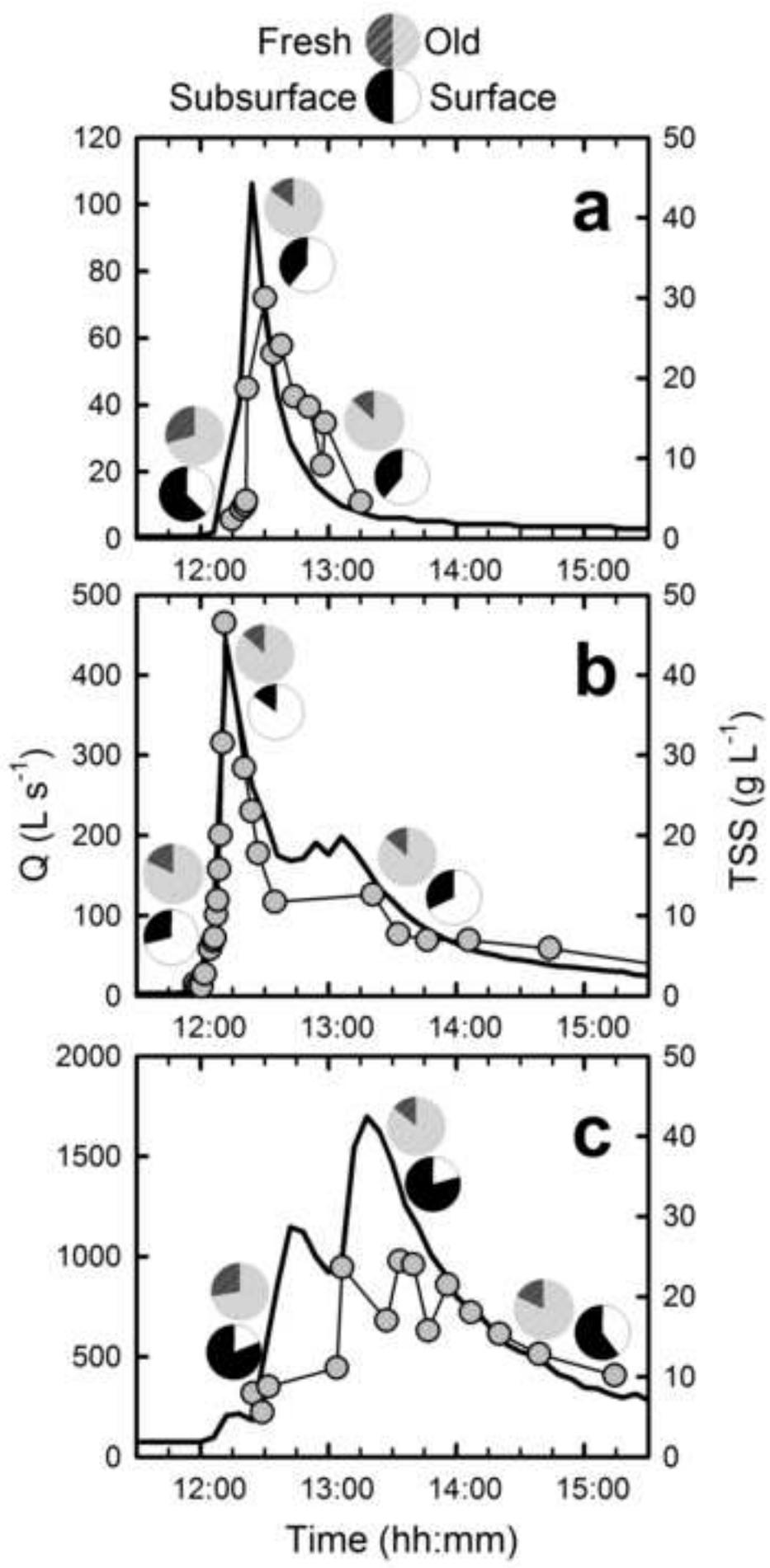


Figure7

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

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

Location	Number of samples	$^{137}\text{Cs}$ (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 <sup>2</sup> )	0.20	0.60	11.6
Sediment yields (Mg km <sup>-2</sup> )	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 <sup>2</sup> )	1 storm event	2.8	0.2 – 1.0	8 – 37 %
Evrard et al., 2010	Central Mexico	3 subcatchments (3 – 12 km <sup>2</sup> )	8 – 18 storms (6 months)	7- 27	2 – 14	10 – 80 %
Bonniwell et al., 1999	ID, USA	Catchment (390 km <sup>2</sup> ) - 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 <sup>2</sup> )	Individual storms (1 year)	13 - 17	1.0 – 1.7	6 – 14 %
Huisman et al., 2013	WI, USA	Catchment (12.4 km <sup>2</sup> )	7 campaigns (6 months)	3 - 12	0.1 – 5.5	1 – 100 %

*n.a.* = not available

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