1 Authors' Response

2 The detailed responses to the general and specific comments of the referees are first provided

3 here together with corresponding changes applied to the revised manuscript, which follows

4 (marked-up version: pages 17-54) in the second part of this document.

5 **Point-by-point response to the reviews**

6 Response to Referee #1

7

8 Small mountain river catchments are thought to play an important role in the erosion of particulate 9 organic carbon (POC) and clastic sediment from the continents, delivering these materials to large 10 river catchments and/or the coastal ocean. However, it is likely much of this POC flux occurs during 11 storm events, which are challenging to sample. In order to better understand the processes operating 12 in these catchments (and therefore what sets the rates of carbon export) it is necessary to examine 13 these flashy events at high temporal resolution. Gourdin et al., contribute a detailed dataset, which 14 examines the erosion of POC during an individual storm event in a small, tropical mountain river. In 15 my opinion, the paper provides at least two novel insights which warrant publication and should 16 interest the readership at Biogeosciences. First, the authors combine hydrological tracers (d18O) to 17 quantify water sources and overland flow contribution. This allows them to interrogate the POC 18 dataset in light of erosion pathways and processes. Second, it consists of three nested gauging 19 stations, sampled at high resolution during the same storm event, providing the opportunity to track 20 POC from hillslope to catchment-scale.

However, the paper was somewhat lacking in its analysis of the tracers of POC source. In my opinion, the work could be much improved by more detailed assessment of the stable isotope measurements (d13C and d15N) and element ratios (C/N) (points 1 and 2). In addition, I felt the two novel aspects I list above could be brought out more, especially the nested stations which weren't really discussed. I've outlined my thoughts on these points below. I've also provided some other comments, which I hope the authors find useful when making revisions.

1. d13C and C/N data: This data is very interesting, and not enough is made of it in the current manuscript. The description of the changes through the hydrograph are rather qualitative, and there is much more information to be gained. To illustrate that point, I've plotted d13C versus N/C for the data in the manuscript (see Figure 1) (N/C is chosen because the inverse of concentration versus its isotope ratio will reveal a binary mixture, or fractionating process, as a linear trend). The patterns are fascinating. They show at least two things which need further discussion. First, in S1 and S10, a trend

33 from a C3 'soil-like' signature (d13C \sim -26permil, N/C \sim 0.1) to a 'vegetation-like' signature with a higher 34 N/C. This has a heavier isotopic signature, perhaps suggesting a C4 plant input (or could it be 35 petrogenic? Or carbonate?). Material with this composition has not been collected in the catchment 36 (Table 1). Second, why are the samples from S4, which is downstream of S1 and upstream of S10, 37 not showing that 13C-enriched signal? Is this a sampling issue (different grain sizes?) or a real 38 observation of fluctuating sources downstream? This point links to 3 below. The N/C vs d13C figure 39 should be used to discuss these aspects in the paper. Related to this point, I think the authors need a 40 more careful discussion of rock-derived ('petrogenic' or 'fossil') POC given the outcrops of 41 sedimentary rocks in the catchment. Without 14C measurements, it is difficult to rule out its 42 contribution with such high TSS (>1g/L is very high) given high petrogenic contributions are seen in 43 other catchments where TSS reach that high (see the Taiwan work, and recent work in the Andes by 44 Clark et al., 2013). The N/C and d13C data do suggest that C4 plant debris with low N/C is important,

but they do not rule out a marine rock-source (d13C~-21permil). Perhaps the high TOC% rule out a
significant petrogenic source (which is likely to be more dilute? Unless a black shale?). A more careful
discussion, aware of the caveats without 14C data, should be considered.

48 **Response to point 1**

49 The different particulate organic matter (POM) compositions recorded by δ^{13} C measurements for the

50 two sampling stations of the Houay Pano sub-catchment (S1 and S4) are most likely due to changes

51 in the composition of sediment sources within the catchment and, downstream, to sorted sediment

1 2 transport. Upstream of S1, i.e. the uppermost sampling station, the stream is directly connected to hillslopes cultivated with C₄ plants (Job's tears) surrounding the gauging weir and the stream channel $\overline{3}$ located immediately above is covered with Napier Grass (another C₄ plant). Therefore it is likely that 4 suspended sediment loads are naturally labelled by ¹³C-enriched vegetation debris (higher C/N) and 5 by topsoil organic matter (lower C/N) supplies during the water level rising stage. At peak flow, ¹³C-6 depleted compositions reflect the dominant contribution of remote fields, mainly cultivated with C₃-7 plants (i.e.: -25.5 ± 1.4 ‰ in the 0-10 cm topsoil layer; Huon et al., Agriculture, Ecosystems and 8 9 Environments, 169, 43-67, 2013) that dilute the "C₄-signal" at the sampling station. In contrast, S4, i.e., the outlet gauging station of the Houay Pano sub-catchment, is located ca. 1.4 km downstream of S1 10 and drains a mosaic of steep cultivated fields with dominant C_3 plant covers (upland rice) and tree plantations (teaks). The depleted composition of POM sampled during the flood is consistent with the 11 12 land cover of the middle - lower part of the sub-catchment. However, between S1 and S4, the Houay 13 Pano stream flows through a 0.19 ha swampy area (Fig. 1 in the submitted manuscript), permanently 14 covered with Taro (a C₃ plant) at its inlet and with Napier grass (a C₄ plant) in its central and outlet 15 parts. The swamp acts as sediment filter (Huon et al., 2013) and a part of the suspended load is 16 trapped and does not reach S4, except during high magnitude floods. For low to intermediate 17 magnitude floods, like the one sampled in this study, downstream export mainly involves sediment conveyed during high water discharge periods. It is why the "C4-signal" is weakly recorded 18 19 downstream at S4 and only during the water level rising stage (see Fig. 3 in the submitted article). A 20 similar process can be observed further downstream between S4 and S10 (close to S4, Figure 1 in the 21 submitted article) where another swampy area covered by Napier grass is found. The δ^{13} C increase 22 slightly (up to -23‰) due to the contribution of vegetation and weakly mineralized C₄ labelled 23 particulate organic matter (higher C/N). In the revised version of the manuscript we will provide these 24 additional information in the result and discussion sections. We will also further discuss the transfer of 25 particulate organic matter along the three nested stations in order to take better into account the 26 filtering role of swamps along the course of the stream as outlined in previous articles published for 27 the same catchment (i.e., Huon et al., 2013).

28 -----

2. Role of carbonate: This is an important point, which needs more open discussion in the methods and results/discussion. The river suspended sediment samples weren't acidified to remove carbonate. This has some benefit, as the inorganic carbon removal protocol is known to attack some of the labile POC (Galy et al., 2007). However, it could severely bias the d13C (carbonate at -5 permil to +5 permil) and C/N (carbonate very high C/N). I think the N/C vs d13C plot points towards carbonate not being responsible for the 13C-enrichment, because the intercept at low N/C (the carbonate end member) is isotopically light. Still, there needs to be some commentary on this.

36 Response to point 2

37 We agree that the occurrence of particulate carbonate grains in suspended sediment loads may induce a problem for the interpretation of ¹³C-enrichments in particulate organic matter with respect to 38 C_3/C_4 -plant derived sources. We tested the possible presence of fine carbonate grains by adding a 39 40 few drops of a 30 % HCl solution on suspended sediment separates collected at S1 and S4 during the 41 water level rising stage and at peak flow but we did not observe any CO₂ bubbling, typical for Cacarbonate dissolution. It is likely that carbonate grains, "if present", do not represent a significant 42 43 fraction of the sediment. Other arguments can be inferred from a previous study conducted in the 44 swamp located between S1 and S4 (Huon et al., 2013). A 1.9 m-long sediment core (corresponding to 45 the sediment sequence accumulated during the last 60 years) was sampled at the inlet of the swamp 46 and the composition of sedimentary organic matter was analysed for each 10 cm depth interval and following the same analytical procedure as in this study. The δ^{13} C averaged -26.2 ± 0.7 ‰ and we did not observe any ¹³C-enrichment that could be explained by the deposition of carbonates. If they were 47 48 49 present, we should observe a "shift" of δ^{13} C values in sediment deposits at the inlet of the swamp where water flow velocity drops down. Therefore the ¹³C-enrichment of suspended organic matter 50 51 composition can be attributed to the cultivation of C₄ plants in upper parts of the catchment. A last 52 argument against the presence of significant carbonate contribution is that soils of the Houay Pano 53 catchment have low pH (4.4 - 5.5, Chaplot et al., SSSAJ 73, 3, 769-779, 2009) indicating that CaCO₃ 54 cannot occur in the soil, even in the upper part of the catchment. Accordingly, soil erosion cannot 55 supply significant amounts of particulate carbonate in suspended sediment loads. It is not the case for 56 dissolved loads that are characterized by high water electrical conductivities controlled by carbonate 57 dissolution and water residence time in upstream aquifers. As recommended, this information will be 58 synthesised and added to the revised version of the manuscript in the "Materials and methods" 59 section.

In the submitted manuscript, we thought that displaying the temporal evolution and trends of particulate organic matter - $\delta^{13}C$ (together with several other parameters) during the flood was 1 2 3 sufficient to illustrate the main changes in the sources of suspended sediments and their related thresholds, i.e., (1) the δ^{13} C trend at flow peak that indicates that suspended organic matter 4 5 6 7 composition evolves towards the average composition of catchment soils or (2) the mixing of C₃-C₄ plant debris and soil-derived organic matter that is mainly found in the upstream section (S1) during the water level rising stage. We are aware that mixing diagrams can be built up from our data and may 8 provide a comprehensive picture of the mixing processes as suggested by the reviewer. We added a 9 figure (new Fig. 6) and further discuss the contribution of another possible source of particulate organic matter, i.e. "fossil organic carbon" (not relevant for our study). We did not perform bedrock analyses in this study as we have shown using fallout ¹³⁷Cs, ²¹⁰Pb_{xs} and ⁷Be activities (Gourdin et al., Journal of Hydrology 519, 1811-1823, 2014) that surface soil and riverbank sources of sediment 10 11 12 13 explain the observed mixing trends. In a new figure (Fig. 6) we reported literature data from Hilton et 14 al. (Geochimica et Cosmochimica Acta 74, 3164–318, 2010) and Kao and Liu (Global Biogeochemical 15 Cycles 14, 189-198. 2000) for tropical mountainous catchments in Taiwan. The main information 16 provided by this new figure is that our sediment loads do not match the published bedrock 17 compositions (they can also be removed from the final figure if requested).

18 -----

19 3. Nested gauging stations: Very little seemed to be made of the nested gauging station and 20 downstream transmission of sources. Particularly in the light of Figure 1 plotted here. This is a 21 novel aspect which could be expanded upon.

- 22 **Response to point 3**
- 23 This question has been addressed in our response to the first comment.
- 24 ------
- 25 All other specific comments and suggestions provided by the reviewer are addressed in the following

26 Comment #1: P9343-L12: Specify what subsurface samples, soil?

Reply to Comment #1: Subsurface soil samples were collected on the walls of gullies and
 riverbanks.The sentence was modified to specify the nature of collected samples.

29 Changes made: P17-L29-30: "soil surface and subsurface samples" was replaced by "soil surface 30 (first 2 cm) and soil subsurface (gullies and riverbanks) samples".

31 -----

32 **Comment #2: P9343-L22-25:** Really, can you say that. Is it not more likely that the previous dataset 33 don't properly account for high flows? Rather than significant changes in land use?

34

35 Reply to Comment #2: The two last sentences of the abstract were rephrased in order to remove the 36 former "too speculative" information and report information regarding the downstream transmission of 37 suspended sediment loads (nested catchments).

Changes made: P18-L10-14: "Swampy areas located along the main stream that acted as sediment filter upstream and sediment sources downstream also controlled the composition of suspended organic matter. Despite the low magnitude of the flood, total organic carbon specific yields were high as this event was the first erosive of the rainy season, following the period of slash and burn in the catchment.

- 43 -----
- 44 **Comment #3: P9344-L11:** why mention South American rivers here? Instead, is it useful to give some 45 indication of the proportion of the global flux contributed? And which rivers you are referring to?
- 46
- 47 **Reply to Comment #3:** corrections made in the text (Asian tropical rivers)

48 Changes made: P18-L28-31: The sentence has been rephrased as follows: "Degens et al. (1991) 49 identified the Asian tropical rivers (e.g., Mekong, Indus/Ganges/Brahmaputra) as the main contributors

50 of dissolved (*ca.* 94 x 10¹² gC yr⁻¹) and particulate (*ca.* 128 x 10¹² gC yr⁻¹) organic matter to world

1 oceans, accounting for more than 50% of global organic carbon inputs (about 335 x 10^{12} gC yr⁻¹ 2 excluding Australia)."

3 -----

Comment #4: P9344-L15: I find this 'dilution' explanation potentially misleading. It is true that the %TOC can be reduced by adding inorganic (or mineral) sediments with low %TOC. However, at the same time, the total mass of suspended sediment increases (g/L), and therefore, the total mass of particulate organic carbon (g/L) also increases. You show this in your dataset pretty convincingly in Figure 5. I would suggest to rephrase this.

- 9
- 10 **Reply to Comment #4:** rephrasing done

11 Changes made: P19-L3-6: The sentence "indicating that particulate organic matter is diluted by high 12 concentrations of mineral matter that is supplied to the rivers" was rephrased as follows "indicating 13 that particulate organic matter is associated with higher concentrations of mineral matter in high TSS 14 loads that are supplied to the rivers through erosion and sediment remobilization processes taking 15 place along the river courses"

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17 **Comment #5: P9344-L27-28:** This sentence jumps in logic, the thereby is misplaced. Please 18 rephrase.

19

20 **Reply to Comment #5:** rephrasing done

Changes made: P19-L14-15: the sentence was rephrased as follows: "Suspended organic matter also
 contributes to water quality degradation (Tanik et al., 1999) and plays a major role in nutrient
 biogeochemical cycles (Quinton et al., 2010)".

24 ------

25 Comment #6: P9345-L3-6: It seemed to me, the benefit of studies like this are to better understand 26 the processes which mobilise organic matter (C and N) from soils and export them from river 27 catchments. Surely this is the main contribution? I think this can be better explained.

- 28
- 29 **Reply to Comment #6:** sentence rephrased.

Changes made: P19-L20-22: The sentence "The design and implementation of appropriate management procedures require the identification of suspended organic matter sources and dynamics" was rephrased as follows: "The design and implementation of appropriate management procedures require the identification of the processes that mobilise organic matter from soils and export suspended organic matter to rivers."

- 35 -----
- 36 **Comment #7: P9345-L20-22:** Linked to the previous comment, why are you doing this? This could be 37 better explained.
- 38
- 39 **Reply to Comment #7:** sentences rephrased and information added.

40 Changes made: P20-L1-5: the sentence has been cut in two and the second half has been rephrased 41 as follows: "The aim was to: (1) estimate the overland flow contribution to stream water and 42 investigate its role for soil organic matter export, and (2) discriminate the respective contributions of 43 soil and stream channel sediment supplies in order to identify the main processes responsible for 44 particulate organic matter delivery at different nested spatial scales."

The sentence immediately following was also completed: "This study is complementary to a previous one, dedicated to the quantification of sediment dynamics from fallout radionuclide measurements (Gourdin et al., 2014), that highlighted the binary contribution of soil and stream channel sediment

48 sources during the same erosive flood event."

1 ------

Comment #8: P9346-L23: Is 'cliffs' needed here. What do you know about the %TOC content (and ideally C/N, d13C and d15N) of the sedimentary rocks? Somewhere in this section I feel you need to explain that the soil and gully samples were not collected from the steepest, highest elevation part of the S10 sub-catchment. Also, if rock samples were not collected, you should be upfront about that, and that you don't know their composition.

7

8 Reply to Comment #8: The term "cliffs" indicates that these vertical rock bars represent a very small area in the uppermost part of the catchment. The carbon content and the isotopic composition of basement rocks of the catchment were not studied as soil covers the entire catchment. These limestone bars (and some narrow stream channel sections) are the only outcrops of basement rocks. A sentence has been added in the text and precisions about the possible contribution of lithic sources of sediments will be addressed in section 3.1.

- 14 Changes made: P21-L9-11: sentence added: "Except for the limestone cliffs and some sections of the 15 narrow streambed, soils or flooded soils cover the entire catchment".
- P23-L8-9: sentence added :" No soil sample was collected in the south-eastern part of the catchmentof S10."
- 18 -----

19 **Comment #9: P9348-L13:** It's normally the case that particulate organic matter samples are filtered 20 through 0.2-0.7micron filters. Was this not the case?

21

Reply to Comment #9: No, it was not the case. Total suspended sediment loads were recovered by total evaporation using a gas oven in the field. Temperature was *ca*. 60-80°C rather than 100°C (water ebullition) as written in the manuscript. Precisions will be added in the text in order to clarify this point.

25 Changes made: P22-L25-P23-L2: The sample preparation is detailed in a new paragraph: "Shortly 26 after collection all samples were dried in 1 L aluminium trays in a gas oven (ca. 60-80°C) for 12-48 h. 27 Preliminary studies carried out in 2002-2007 showed that dissolved organic carbon concentrations in 28 the Houay Pano stream water are commonly low, 1.8 ± 0.4 mg L⁻¹ (n = 74) and 2.0 ± 0.7 mg L⁻¹ (n = 29 65), at base flow and discharge peak, respectively. With high-suspended sediment loads (see further 30 in the Results section), a 3 mgC L⁻¹ content for dissolved organic carbon would represent 1-10 wt% of 31 the total (dissolved and particulate) organic carbon load. In average 97 ± 3 % of the total matter 32 recovered is made of particulate matter, 90-95% during the water rising stage and 95-99% for the 33 other water levels. We are confident that all measurements account for particulate organic matter with 34 negligible dissolved loads and that the dynamics of organic compounds during the flood refers to 35 particulate matter."

36 -----

37 Comment #10: P9349-L6: Just a few pages ago you mention carbonate 'cliffs'. How can you be sure 38 there is no carbonate? The TSS values are very high in your rivers (almost reaching Taiwan-like 39 levels). It may be a logical assumption to make in acidic soils, but I'm not convinced that detrital 40 carbonate is not playing a role (see also comments above).

41

42 **Reply to Comment #10:** The absence of carbonate in suspended sediment samples collected for 43 different stages of the flood at S1 and S4 has been checked by adding drops of a 30% HCl solution 44 and no CO₂-bubbling (that would have occurred in the presence of carbonate grains) could be 45 observed. We therefore assume that, if present, this contribution is not significant and may be 46 neglected. Precisions on the characteristics of the soils were added in the description of the study site 47 (section 2, see above comment #6). The description of the HCl test has also been added in the 48 material and method section 3.2.

49 Changes made: P21-L13-16: The following sentence has been added: "The soils have low pH ranging

4.4-5.5 (Chaplot et al., 2009) indicating that carbonate precipitation is not favoured in soils, even in the
 upper part of the catchment and, accordingly, cannot supply particulate inorganic carbon to
 suspended sediment loads."

P23-L25-30: The following sentence has been added: "The possible occurrence of carbonate minerals (or carbonate rock fragments) in TSS samples, collected at different stages of the flood at stations S1 and S4, was checked by pouring drops of a 30 % HCl solution on dry sample aliquots. No CO₂bubbling, typical for carbonate dissolution, was observed. Therefore, common carbonate minerals such as calcite do not represent a detectable fraction of the suspended sediment loads and could be neglected."

7

8 **Comment #11: P9350-L17:** The d15N values seem very high for tropical vegetation and soil (see 9 compilations from Martinelli et al., 1999 and recent work in Taiwan, Hilton et al., 2013). Is this worth 10 commenting on?

11

Reply to Comment #11: We only measured soil $\delta^{15}N$ (typically enriched in ¹⁵N with respect to 12 13 vegetation) and Martinelli et al. (1999) reported an average $\delta^{15}N$ of 8 ‰ in bulk soils, consistent with our data. In contrast, Hilton et al. (2013) displayed δ^{15} N values ranging between 0.7 ‰ and 6.5 ‰ but 14 15 this study accounted for soil and lithic sources of organic matter with variable mineralization status. The later should provide lower δ^{15} N than for soils as preservation of organic matter in sedimentary and 16 low metamorphic grade rocks takes place at "high temperature" (lower ¹⁵N/¹⁴N fractionation) whereas 17 18 incorporation and stabilization of organic matter in soils occurs at "low (surface) temperature" (higher 19 ¹⁵N/¹⁴N fractionation). The paper by Amundson et al. (Global patterns of the isotopic composition of 20 soil and plant nitrogen, Global Biogeochemical Cycles 17, 1031, 2003) provides δ^{15} N estimates for the 21 first 50 cm of tropical soils covering our study area (ca. 6-10 ‰) that match our soil and suspended 22 particulate organic matter data. High δ^{15} N is also an argument against important occurrence of rock-23 derived organic matter in suspended sediment loads.

24 Changes made: P25-L4-7: The following sentences were added: "Soil surface and subsurface sources 25 can also be distinguished by their δ^{15} N values that are slightly lower for the former (**Table 1**). The 26 overall values reflect high 15 N/ 14 N fractionation during incorporation and mineralization of plant tissues 27 in soils, typical for tropical environments (e.g., Amundson et al., 2003)."

28 -----

29 **Comment #12: P9351-L4:** these TSS values are very high. It is perhaps worth comparing to 30 measurements made on other small tropical rivers.

31

Reply to Comment #12: This value corresponds to the maximum TSS concentration measured in overland flow at the 1m² scale. It is consistent with average values reported by Chaplot and Poesen (2012) at the same scale and in the same catchment (ranging between 2.8 and 8.4 g L⁻¹, respectively, from backslope to hillslope shoulder). At plot scale suspended loads are much higher up slope than down slope due to the deposition of soil-detached particles along catchment's slopes. It is difficult to provide direct comparisons with suspended loads of other small tropical rivers: physiographic setting, flood magnitude, land use and cultivation practise are too different.

- 39 Changes made: no change made in the revised manuscript.
- 40 -----

41 **Comment #13: P9351-L10:** This observation has been made in small, steep catchments in other 42 tropical settings (Clark et al., 2013) and temperate settings (Smith et al., 2013) and might be worth 43 commenting on at this point.

44

45 Reply to Comment #13: Comments were added in order to compare the observed dynamics to the46 behaviours previously reported in the literature.

47 Changes made: P25-L25-29: The following sentence has been added: "Similar behaviours were 48 reported by Clark et al. (2013) in the tropical Andes and interpreted as a greater contribution of non-49 fossil POC during the rising stage and the peak discharge and in the Swiss Alps by Smith et al. (2013) 50 who interpreted the initial decrease of POC during the rising stage as resulting from in-channel 51 clearing."

52 -----

- 1 **Comment #14: P9351-L24:** it might be useful to refer to 2012 when using 23 May in the text (and subheadings) for clarity.
- 34 Reply to Comment #14: precision added.
- 5 Changes made in the revised manuscript as "23 May 2012 flood".
- 6 -----
- 7 **Comment #15: P9356-L1:** why is this sentence a separate paragraph?
- 9 **Reply to Comment #15:** The sentence has been grouped with the following paragraph.

10 Changes made: The line feed has been deleted.

11 -----

12 **Comment #16: P9357-L8:** This is a very long paragraph, and contains some novel and interesting 13 observations. I'd recommend splitting it. Also, I think the discussion of organic matter sources needs to 14 be more careful. The attached Figure 1 shows co-variation of N/C and d13C that needs to be 15 discussed (see main comment above).

16

8

17 Response to Comment #16: This section has been rewritten and split into two paragraphs: "5.2.1 18 Sources of suspended organic matter in the catchment" and "5.2.2 Dynamics of suspended organic matter". The co-variation between TN/TOC and δ^{13} C is shown in a new Fig. 6 placed in section 5.2.2 19 20 together with the composition of the possible mixing end-members determined for the catchment. 21 Because carbonate and "fossil" organic matter sources of carbon in suspended sediment loads could 22 be excluded or minimized, the co-variation is best explains by the mixing of surface soil and 23 subsurface soil organic matter defined by the end-members. Considering the limited bedrock outcrops, 24 the low magnitude of the flood, the absence of mass transport along catchment slopes and high $\delta^{15}N$ 25 values in TSS loads (see above in the first reply), the correlation between TN/TOC and δ^{13} C highlights 26 the mixing of C_3 - C_4 labelled organic matter.

27

Changes made: major changes were made for this section. The former sentences were either
 completed or rewritten in the revised manuscript.

30 -----

31 **Comment #17: P9357-L14:** I think it would be useful to link these concentration measurements to 32 other places where 'fossil' POC has been observed, e.g. in the Andes (Clark et al., 2013) and Taiwan 33 (e.g. Hilton et al., 2010).

34

Reply to Comment #17: Comparisons to literature data were added in the revised manuscript and
 reported in the new Fig. 6 (see above comment #16).

37 Changes made: P31-L1-4: together with new sentences the following was added "In the Andes, Clark 38 et al. (2013) identified fossil POC contributions associated with TSS concentrations above 1 g L⁻¹. In a 39 Taiwanese river, Hilton et al. (2011) reported suspended sediment concentrations up to *ca.* 30 g L⁻¹ 40 leading to fossil POC concentrations up to *ca.* 0.1 g L⁻¹."

- 41 -----
- 42 Comment #18: P9357-L20: What about the much larger, steeper Houay Xon river catchment (S10)?
 43 Where landslides/mass wasting processes occurring there?

44

45 Reply to Comment #18: The Houay Xon river catchment is larger but its channel is not steeper than 46 the Houay Pano. In fact its slope is gentler and the connectivity of hillslopes with the main channel is 47 lower. It is true that the S10 station drains the SE area that includes the highest elevations of the

48 catchment, but this area is covered with old protected forests and no major tributary flows from it into

the Houay Xon River. Considering land use and topography, sediments possibly generated by erosion in the upper part of the drainage area are deposited before reaching the main stream and the intermittent streams located upstream of S10 did not flow during the storm (Figure 1).

Changes made: P20-L30-P21-L5: the missing information was added in the revised manuscript in section 2 (study site): "The Houay Xon river catchment is larger but its channel is not steeper than for the Houay Pano subcatchment. Its slope is gentler and the connectivity of hillslopes with the main stream is lower. The drainage basin that includes the highest elevations of the catchment is covered with old protected forests but no major tributary flows into the Houay Xon River. Sediments generated by erosion in the drainage area can settle before reaching the main stream due to a decline of topography above the left bank of the river (**Fig. 1**). The intermittent streams located upstream of S10 did not flow during the 23 May 2012 flood."

- 12 -----
- 13 **Comment #19: P9357-L24:** Is this a good point to split this long, but important paragraph?
- 14

15 Reply to Comment #19: Done

- 16 Changes made: Line feed added
- 17 -----

18 Comment #20: P9359-L4: I wasn't convinced this section was useful, especially as this 137Cs data is 19 not shown here.

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21 Reply to Comment #20: As outlined at the end of the introduction section this study is 22 complementary of a paper recently published (Gourdin et al., Journal of Hydrology 519, 1811-1823, 23 2014). The radionuclide measurements were carried out on the same sample aliquots (no 24 transformation due to y-counting) collected during the same flood than in this study. The main interest of referring to these data is that when fine suspended matter is tagged by ¹³⁷Cs, its origin can only be soils in surface position. Radionuclide fallout took place in the 1950-1970's during the period of 25 26 27 maximum atmospheric nuclear bomb tests (maximum supply in 1963) with limited migration with soil 28 depth. This radionuclide did not exist before atmospheric bomb tests started. Sediment labelling acts the same way ¹⁴C does with organic matter and because soil organic matter is bound to fine soil clay particles, bedrocks are not labelled by ¹³⁷Cs. We think that mentioning this paper (where a full 29 30 31 description and discussion is provided) in our discussion section will strengthen our interpretation as 32 we already have a good idea of the contributing sources of sediments in the catchment.

Changes made: P30-33: we rephrased our discussion in this direction in the revised version of themanuscript in section 5.2.1 and 5.2.2.

35 -----

36 **Comment #21: P9360-L6:** How do these storm specific POC yields relate to other measurements in 37 tropical catchments? There are quite a lot of estimates from work in Taiwan.

38

Response to Comment #21: we reduced this part in the revised manuscript as we found out that the conclusions we could draw from the comparison between suspended organic matter collected a single low magnitude flood event (our study) with annual carbon budgets in the catchment (Chaplot and Poesen, 2012) or elsewhere were too speculative.

43 Changes made: several paragraphs were removed and the related modifications were made in the 44 abstract, discussion and conclusion sections of the revised manuscript.

45 -----

46 **Comment #22: P9361-L13:** It seems the discussion of the larger S10 catchment has been forgotten

47 at this stage. I think the novelty of this study is the nested approach, which should come out in the 48 conclusions.

49

Reply to Comment #22: we fully agree with this comment and the missing information and discussion were added in the revised version of the manuscript. Additional information for S10 was added in section 2 (study site) see above for comment #18. Information for the "nested catchment approach" is reported in a new rewritten section 5.2.3 reported below. Other changes were made in the concluding remarks section.

6 Changes made: "Nested approach". Changes were made in last part of the discussion section: 5.2.3 7 Suspended sediment and particulate organic carbon delivery. The following sentences were added . 8 9 and replace the former. "Compared to the 2002-2003 annual sediment yield at S4 (2090 kg ha⁻¹ yr⁻¹) and S10 (540 kg ha⁻¹ yr⁻¹) reported by Chaplot and Poesen (2012), the 23 May 2012 flood represented 10 ca. 21% of the total annual exports recorded for both stations. These deliveries are high for a single 11 event of moderate intensity. However, fallout radionuclide measurements (Gourdin et al., 2014) 12 indicate that this flood was the first important erosive event of the 2012 rainy season and mainly 13 consisted of remobilized river channel sediments (ca. 80%). The TSS yield (Sy) of ca. 433 kg ha⁻¹ (8.3 14 kgC ha⁻¹) at S4 is greater than at S1 and S10 (Table 3) and consistent with higher specific runoff and 15 runoff coefficient values (Table 2). With a low value at S1, the succession of nested catchments was 16 not related to a decrease in specific delivery when drainage area increased. This unusual behaviour is 17 best explained by the occurrence of swamp areas along the main stream. In the upper part of the 18 catchment, a natural swamp acts as a filter for sediments conveyed during low to medium magnitude 19 floods (Fig. 1). Napier grass, the main aquatic plant forms dense masses of litter that reduce stream 20 flow velocity during the rainy season. Nearly 33 Mg of soil-derived organic carbon was thus accumulated since the early 1960's (Huon et al., 2013). This swamp played a key role with respect to 21 22 downstream export of suspended sediment during the 23 May 2012 flood. It also explains why the 23 high δ^{13} C values of TSS loads, observed during the rising water stage upstream of the swamp (at S1), 24 were only partly transmitted to S4. Soil-derived organic matter supplied by overland flow replaced the 25 major part of the TSS during the rising stage, downstream of the swamp. A comparable picture can be 26 drawn for the wetlands located at the outlet of the village. However in contrast this swampy area 27 where streambanks are also encroached by Napier grass contributed to a rise of the δ^{13} C values of 28 suspended organic matter at the monitoring station S10. This shift fingerprinted the extent of 29 streambank sediment retention and mobilization processes taking place downstream in accordance 30 with radionuclide activity measurements carried out for the same samples (Gourdin et al., 2014)."

31

1 Response to Referee #2

2 Gourdin et al. present a detailed characterization of riverine organic carbon composition during a 3 storm event in a small catchment of Northern Laos. The authors use gauging data, water isotope and 4 electric conductivity to characterize the hydrological response of the river system to the storm event. 5 6 7 They then use riverine particulate organic carbon characterization (%C, %N, d13C, d15N) along with a characterization of soil organic matter at the plot scale to propose a model of organic carbon mobilization during the storm event. The combination of detailed characterization of . 8 9 hydrological/erosion processes and particulate organic carbon flux and composition assessments is interesting as it has the potential to provide a mechanistic understanding of particulate organic carbon 10 mobilization during storms. In that regards the paper would deserve publication in Biogeosciences. 11 That said, I am a bit disappointed by 1) the lack of in depth interpretation of the organic carbon composition (d13C, d15N, C/N) and, 2) the speculative nature of the last part of the discussion (i.e. 12 comparison of the May 2012 organic carbon yields with previous estimates). As a result, I recommend 13 14 that the paper be revised before publication. Detailed comments follow.

15

16 General comments

17

18 1) The authors claim that carbonate minerals are absent (and thereby justify not performing any acid treatment prior to organic carbon characterization) yet they mention the existence of "limestone cliffs" in the catchment. Even trace quantities of carbonates can severely affect the measured d13C, therefore the authors must demonstrate the complete absence of carbonate minerals.

23

24 **Response to point 1**

25 We agree that the occurrence of particulate carbonate grains in suspended sediment loads may induce a problem for the interpretation of ¹³C-enrichments in particulate organic matter with respect to 26 27 C_{4} -plant derived soil sources. We tested the possible presence of fine carbonate grains by adding 28 drops of a 30 % HCl solution on suspended sediment separates collected at S1 and S4 during the 29 water level rising stage and at peak flow but we did not observe any CO₂ bubbling, typical for Ca-30 carbonate dissolution. It is likely that carbonate grains, "if present", do not represent a significant 31 fraction of the sediment. Other arguments can be inferred from a previous study conducted in the 32 swamp located between S1 and S4 (Huon et al., Agriculture, Ecosystems and Environments, 169, 43-33 67, 2013). A 1.9 m-long sediment core (corresponding to the sediment sequence accumulated during 34 the last 60 years) was sampled at the inlet of the swamp and the composition of sedimentary organic 35 matter was analysed for each 10 cm depth interval, following the same analytical procedure as in this 36 study. The δ^{13} C averaged -26.2 ± 0.7 ‰ and we did not observe any 13 C-enrichment that could be explained by the deposition of carbonates. If they were present, we should observe a "shift" of δ^{13} C 37 38 values in sediment deposits at the inlet of the swamp where water flow velocity drops down. Therefore 39 the ¹³C-enrichment of suspended organic matter composition can be attributed to the cultivation of C4 40 plants in upper parts of the catchment. A last argument against the presence of significant carbonate 41 contribution is that soils of the Houay Pano catchment have low pHs (4.4-5.5, Chaplot et al., SSSAJ 42 73. 3. 769-779, 2009) indicating that CaCO₃ cannot occur in the soil, even in the upper part of the 43 catchment. Accordingly, soil erosion cannot supply significant amounts of particulate carbonate in 44 suspended sediment loads. It is not the case for dissolved loads that are characterized by high water 45 electrical conductivities, controlled by carbonate dissolution and water residence time in upstream 46 aquifers.

As recommended, this information was synthesised and added to the revised version of the
 manuscript in the Materials and methods section (3.2. Particulate organic matter composition
 measurements).

50 "The possible occurrence of carbonate minerals (or carbonate rock fragments) in TSS samples, 51 collected at different stages of the flood at stations S1 and S4, was checked by pouring drops of a 30% HCl solution on dry sample aliquots. No CO₂-bubbling, typical for carbonate dissolution, was 53 observed. Therefore, common carbonate minerals such as calcite do not represent a detectable 54 fraction of the suspended sediment loads and could be neglected. No additional treatment was 55 applied." 1 -----

2 2) The authors claim that rock-derived organic carbon is not present in their samples, yet they 3 do not demonstrate it. There is quickly growing body of literature dealing with the 4 concentration, composition and dynamics of rock-derived organic carbon in rivers and as such 5 the authors need to seriously consider this component of the organic carbon pool.

6

7 Response to point 2

We are aware that lithic sources of particulate organic matter (so called « fossil carbon », Meybeck, 8 9 Advances in Soil Science, CRC Press, 2006) may represent a significant part of the particulate 10 organic carbon exported, as shown in other catchments (e.g., Kao and Liu, Global Biogeochemical Cycles 14, 189-198, 2000; Huon et al., Advances in Soil Science, CRC Press 2006; Hilton et al., 11 12 Geochimica et Cosmochimica Acta 74: 3164-318, 2010). However, evidence for such a supply is 13 often difficult to put forward. Sedimentary and meta-sedimentary rocks provide total organic matter 14 δ^{13} C that may overlap soil organic matter values. This approach is more relevant in large river systems 15 (Galy et al., Geochimica et Cosmochimica Acta 72: 1767-1787, 2008; Clark et al. Geochemistry, 16 Geophysics, Geosystems. 14: 1644-1659, 2013) and for simple binary mixtures with sediments 17 originating from geographically distinct locations with contrasted geomorphic evolution or when these 18 different areas are covered with contrasted vegetation (e.g., using C3 vs. C4 vegetation natural 19 fingerprinting). It is not the case for the Houay Pano catchment covered by a mosaic of cultivated 20 fields. The use of $\delta^{15}N$ may help solving the problem. It is likely that $\delta^{15}N$ in sedimentary and metasedimentary rocks should provide lower values than soil organic matter that underwent low temperature $^{15}\text{N}/^{14}\text{N}$ fractionation processes during mineralization in soils. However only scarce $\delta^{15}\text{N}$ 21 22 measurements have been reported for lithic sources of nitrogen and these measurements account for 23 24 both organic matter and mineral bound ammonium contributions (e.g., Boudou et al., Fuel 63: 1508-25 1510, 1984; Bebout and Fogel, Geochimica et Cosmochimica Acta, 56: 2839-2849, 1992; Kao and 26 Liu, previously cited 2000; Huon et al., previously cited, 2006). Field information do not support the 27 hypothesis of an important supply of lithic sources of organic matter. We did not observe any mass 28 movements along hillslopes of the catchment that could have induced a massive input of lithic 29 material. This flood, representative of most of the events taking place in the region during the rainy 30 season, was of low to intermediate magnitude and could be easily sampled and monitored. Finally, 31 ¹³⁷Cs and ²¹⁰Pb_{xs}/⁷Be labelling of suspended sediment during the same flood indicate that suspended 32 sediment in the stream was only supplied by surface (sub-surface) soil sources (Gourdin et al., 33 Journal of Hydrology 519, 1811-1823, 2014).

Section 5.2 (P30-34) has been rewritten in order to better account for the fossil rock-derived source of organic matter together with a new **Fig. 6** (see in the revised manuscript and in comment 3 below).

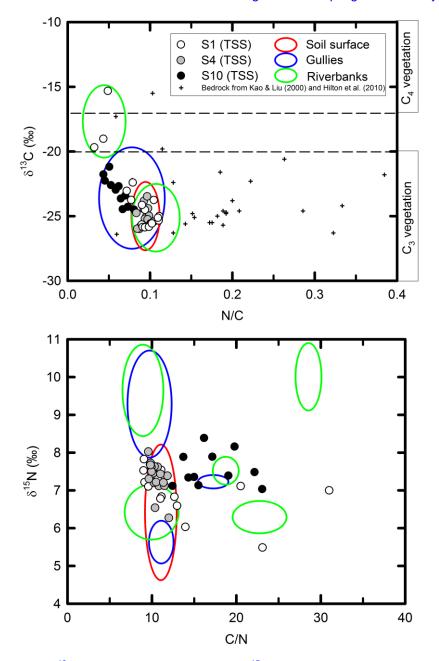
- 36
- 37 -----

3) The authors did not make the most out of their organic matter characterization. They
 measured 3 conservative tracers (d13C, d15N and C/N) but haven't really exploited these data.
 For instance I would like to see cross plots such as d13C vs. N/C and d15C vs C/N. These
 should be very informative regarding the source/nature of the organic matter.

42

43 **Response to point 3**

44 In the submitted manuscript, we thought that displaying the temporal evolution and trends of particulate organic matter - δ^{13} C (together with several other parameters) during the flood was 45 46 sufficient to picture the main changes in the sources of suspended sediments and their related thresholds, i.e., (1) the δ^{13} C trend at flow peak that indicates that suspended organic matter 47 48 composition evolves towards the average composition of catchment soils or (2) the mixing of C_3 - C_4 49 plants and soil-derived organic matter that is mainly found in the upstream section during the water 50 level rising stage. We are aware that mixing diagrams can be built up from our data and may provide a 51 more comprehensive picture of the mixing processes as suggested by the reviewer. We added a new 52 figure (**Fig. 6**, δ^{13} C vs. TN/TOC) in the revised manuscript and further discuss the possible contribution 53 of "fossil organic carbon". We also reported literature data from Hilton et al. (Geochimica et 54 Cosmochimica Acta 74: 3164-318, 2010) and Kao and Liu (Global Biogeochemical Cycles 14: 1891 198. 2000) for tropical mountainous catchments in Taiwan. We did not perform bedrock analyses in this study as we have shown using fallout ¹³⁷Cs, ²¹⁰Pb_{xs} and ⁷Be activities (Gourdin et al., Journal of Hydrology, cited above) that surface soil and riverbank sources of sediment explain the observed mixing trends. The main information provided by this new **Fig. 6** is that our sediment loads do not match the published bedrock compositions (they can also be removed from the final figure). Plot of $\delta^{15}N$ vs. TOC/TN (Fig. A below) does not provide any new insight with respect to **Fig. 6**. The composition of TSS is well constrained for S1 and S4 with some deviation from the identified endmembers for S10 due to insufficient « riverbank » and « gullies » sampling in the vicinity of S10.



9



Figure A – Plots of δ^{13} C vs. TN/TOC (upper) and δ^{15} N vs. TOC/TN (lower) for total suspended sediment loads (TSS) showing the possible sources of sediments determined in the catchment (not displayed in the revised manuscript and replaced by new **Fig. 6**).

- 14
- 15 -----

4) The discussion of the temporal variation of the specific yields (section 5.2.3) is speculative.
 While it is possible that land use change explains the higher yield for the May 23rd storm the

18 authors do not demonstrate it and other explanations (e.g. specific characteristics of the

- 1 storm) cannot be excluded. I recommend deleting this paragraph.
- 2

3 Response to point 4

We agree with the reviewer's suggestion but for a slightly different reason. Much more sediment data and land use information are required to strengthen the comparison between previously published 2002-2003 annual sediments yields (Chaplot and Poesen, Catena 88: 46–56, 2012) and the flood sampled in 2012 (work in progress). This section has been shortened in the revised version of the manuscript (5.2.3. Suspended matter and particulate organic carbon deliveries) and new information on the transmission of suspended loads downstream with respect to the filtering role of swampy areas along the main stream replaces this former section. Related modifications have been made in the abstract and the concluding remarks (section 6).

12 P33-L25: New section 5.2.3: "Total suspended sediment exports are summarized in Table 3 for S1, 13 S4 and S10 sub-catchments. Compared to the 2002-2003 annual sediment yield at S4 (2090 kg ha yr⁻¹) and S10 (540 kg ha⁻¹ yr⁻¹) reported by Chaplot and Poesen (2012), the 23 May 2012 flood 14 15 represented ca. 21% of the total annual exports recorded for both stations. These deliveries are high 16 for a single event of moderate intensity. However, fallout radionuclide measurements (Gourdin et al., 17 2014) indicate that this flood was the first important erosive event of the 2012 rainy season and mainly 18 consisted of remobilized river channel sediment (*ca.* 80%). The TSS yield (S_y) of *ca.* 433 kg ha⁻¹ (8.3 19 kgC ha⁻¹) at S4 is greater than at S1 and S10 (**Table 3**) and consistent with higher specific runoff and 20 runoff coefficient values (Table 2). With a low value at S1, the succession of nested catchments was 21 not related to a decrease in specific delivery when drainage area increased. This unusual behaviour is 22 best explained by the occurrence of swamp areas along the main stream. In the upper part of the 23 catchment, a natural swamp acts as a filter for sediments conveyed during low to medium magnitude 24 floods (Fig. 1). Napier grass, the main aquatic plant forms dense masses of litter that reduce stream 25 flow velocity during the rainy season. Nearly 33 Mg of soil-derived organic carbon was thus 26 accumulated since the early 1960's (Huon et al., 2013). This swamp played a key role with respect to 27 downstream export of suspended sediment during the 23 May 2012 flood. It also explains why the 28 high δ^{13} C values of TSS loads, observed during the rising water stage upstream of the swamp (at S1), 29 were only partly transmitted to S4. Soil-derived organic matter supplied by overland flow replaced the 30 major part of the TSS during the rising stage, downstream of the swamp. A comparable picture can be 31 drawn for the wetlands located at the outlet of the village. However in contrast this swampy area 32 where streambanks are also encroached by Napier grass contributed to a rise of the δ^{13} C values of 33 suspended organic matter at the monitoring station S10. This shift fingerprinted the extent of streambank sediment retention and mobilization processes taking place downstream in accordance 34 35 with radionuclide activity measurements carried out for the same samples (Gourdin et al., 2014)."

36 -----

5) It appears to me that there is a large (relative to the size of the catchment) tributary that flows into the Houay Xon just upstream of S10. Given that the vegetation and land use in this sub catchment is drastically different (mostly old protected forest, see fig 1) one can imagine that the signal at S10 is dominantly controlled by the mixing proportions between the Houay Xon and this tributary. Interpreting the organic carbon composition at S10 would then require to at least characterize the composition of the organic matter in the tributary.

43

44 **Response to point 5**

Our figure does not provide information on the contribution of this minor ungauged stream that did not
 convey significant sediment loads during the flood. We added the following information in the site
 description (section 2) of the revised version of the manuscript.

P20-L30-P21-L5: "The Houay Xon river catchment is larger but its channel is not steeper than for the Houay Pano subcatchment. Its slope is gentler and the connectivity of hillslopes with the main stream is lower. The drainage basin that includes the highest elevations of the catchment is covered with old protected forests but no major tributary flows into the Houay Xon River. Sediments generated by erosion in the drainage area can settle before reaching the main stream due to a decline of topography above the left bank of the river (Figure 1). The intermittent streams located upstream of S10 did not flow during the 23 May 2012 flood".

55 -----

1 Specific comments

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3 Comment #1: P9346 L21-23: any idea what the C concentration in the sandstones and greywackes is ?

6 **Reply to comment #1:** No, we did not perform any measurement for basement rocks that only 7 scarcely outcrop: limestone cliffs in the uppermost NE part of the upper catchment and locally for a 8 few 2-5 m sections of the streambed. The catchment is covered by soil and/or flooded soils in the 9 swampy areas.

10 Changes made: P21-L7-10: The sentence has been rephrased as follows: "The geological basement 11 of the Houay Pano upper catchment is mainly composed of pelites, sandstones and greywackes (not 12 sampled), overlaid in its uppermost NE part by Carboniferous - Permian limestone cliffs (not sampled) 13 that only cover a very small area in the catchment."

- 14 -----
- 15 Comment #2: P9347 L1 (and throughout the ms): please use degrees for slopes, not %
- 16
- 17 Reply to comment #2: Slopes were converted from percentages to degrees has requested.

Changes made: P21-L19-20 "between 3 and 150%" was replaced by "between 2 and 57°" and P22L16: "with 33% slope" was replaced by "with 18° slope".

- 20 -----
- 21 **Comment #3: P9348 L10:** give sample volume
- 22
- 23 **Response to Comment #3:** The volume of the river water samples was 0.65 L.
- 24 Changes made P22-L22: "0.65 L" was added before "polyethylene bottles".
- 25
- 26 -----
- Comment #4: P9348-L11: what does 100 degrees C do to the organic matter? There is a possibility
 that the most labile compounds would degrade at this temperature.
- 29

Reply to comment #4: The sample preparation procedure was rewritten and details were added in
 the text in section 3.1. The temperature of the gas oven was lower than 100°C (no ebullition), likely *ca.* 60-80°C range) and labile dissolved organic phases were likely degraded.

33 Changes made: The sample preparation is detailed in a new paragraph: "Shortly after collection all 34 samples were dried in 1 L aluminium trays in a gas oven (ca. 60-80°C) for 12-48 h. Preliminary studies 35 carried out in 2002-2007 showed that dissolved organic carbon concentrations in the Houay Pano stream water are commonly low, 1.8 ± 0.4 mg L⁻¹ (n = 74) and 2.0 ± 0.7 mg L⁻¹ (n = 65), at base flow 36 and discharge peak, respectively. With high-suspended sediment loads (see further in the Results 37 38 section), a 3 mgC L⁻¹ content for dissolved organic carbon would represent 1-10 wt% of the total 39 (dissolved and particulate) organic carbon load. In average 97 ± 3 % of the total organic matter 40 recovered is made of particulate organic matter, 90-95% during the water rising stage and 95-99% for 41 the other water levels. We are confident that all measurements account for particulate organic matter with negligible dissolved loads and that the dynamics of organic coumpounds during the flood refers to 42 43 particulate matter."

44 -----

45 **Comment #5: P9349:** Please provide propagated uncertainty (i.e. taking into account precision and 46 accuracy) for samples. Precision better than 0.1‰ for d15N is not common, please provide data 47 supporting that statement.

48

Reply to comment #5: We fully agree with this comment. In fact there is a mistake in the text (an inopportune "copy and paste" of information relevant to a dual-inlet MS no longer used). One must read: precision is better than ± 0.2 ‰ for δ^{13} C and ± 0.3 ‰ for δ^{15} N. The accuracy of MS measurements is based on repeated measurements of a 99% pure tyrosine standard (Girardin and Mariotti, 1991) still used in the lab. For each batch of 50 samples, 8 tyrosines are measured at the beginning and 10-12 additional tyrosines are placed between samples (every 5 sample intervals) to control the possible instrumental drift. However, some samples were repeated several times so that we can consider that precision can be lower than the assumed uncertainties ("is better than").

9

10 Change made P23-L20-25: The sentences: "Analytical precision was better than ± 0.1‰ vs. PDB-AIR standards (Coplen et al., 1983) and 0.1 mg g⁻¹ (equivalent to 0.01 wt.%) for δ^{13} C- δ^{15} N and TOC-TN, 11 12 respectively. Data reproducibility was checked by replicate analyses of selected samples and of a 13 tyrosine laboratory standard (Girardin and Mariotti, 1991)" were replaced by: "Analytical precision was 14 better than $\pm 0.2 - 0.3 \text{ }$ ws. PDB-AIR standards (Coplen et al., 1983) and 0.1 mg g⁻¹ (equivalent to 15 0.01 wt.%) for δ^{13} C- δ^{15} N and TOC-TN, respectively. Reproducibility was checked by replicate analyses of a 99% pure tyrosine laboratory standard (Girardin and Mariotti, 1991) using 18 tyrosines 16 17 per batch of 50 samples. Selected sample measurements were also repeated during the course of the 18 study.

- 19
- 20 -----
- 21 **Comment #6: P9350 L17-19:** why are d15N values lower for surface organic matter ?
- 22

Reply to comment #6: surface soil organic matter is depleted in ¹⁵N compared to deeper soil 23 horizons because mineralization of plant tissues progressively enriches in ¹⁵N the residual organic 24 matter during its incorporation and burial in the soil. The lighter ¹⁴N isotope is preferentially consumed 25 26 and degraded by soil's microorganisms (e.g., Nadelhoffer and Fry, 1988. SSSAJ 52, 1633-1640; Martinelli et al., 1999, Biogeochemistry 46, 45-65). The δ^{15} N of soil organic matter increases with soil 27 28 depth (e.g., between +1.1 and +4.2 ‰ at tropical sites, Martinelli et al., 1999) and mainly in the first 50 29 cm. Accordingly, the most superficial soil layer is less degraded and displays δ^{15} N values closer to the 30 vegetation composition (typically ca. 0%; e.g., Martinelli et al., 1999).

- 31 Changes made: none
- 32 -----
- 33 **Comment #7: P9351 L8-9:** "that match topsoil organic matter composition" true at the beginning of 34 the event, not so much by the end.
- 35
- 36 **Reply to comment #7:** some precision was added in the revised manuscript

Changes made P25-L22-23: the sentence has been completed: "with a slight evolution towards the composition of deeper superficial layers (1-3 cm) at the end of the event".

39 -----

- 40 **Comment #8: P9351 L9-10:** "likely results from the preferential export of vegetation debris" is this supported by high C/N ratios?
- 42
- 43 **Response to Comment #8:** vegetation debris contain more C and N than soil organic matter due to 44 the bacterial mineralization of plant tissues. Due to different mineralization rates C-rich compounds are 45 more easily degraded than N-rich compounds and the TOC:TN ratio decreases during mineralization. 46 Therefore, non-degraded vegetation or weakly mineralized soil organic matter should provide higher 47 TOC/TN than soil organic matter. In contrast δ^{13} C and δ^{15} N increase due to preferential ¹⁴N uptake 48 during mineralization and accordingly non-degraded vegetation debris display lower values.
- 49 Changes made: none
- 50

1 **Comment #9: P9354 L26-27:** "high contribution of OF at this station" this is not that obvious when 2 looking at the d18O – EF plot.

Reply to Comment #9: Yes, pre-event δ^{18} O values are missing, so the beginning of the dilution trend is not represented on this plot. However, we can see a major dilution pattern (EC decreased by more than 200 µS cm⁻¹ in less than 10 min) at the beginning of the event (**Fig. 3IIIc**).

6 Changes made: P28 L25-27: the sentence has been rephrased as follows: "The EC values, 7 decreasing from 450 to 155 μ S cm⁻¹ at the beginning of the event (**Fig. 3IIIc**), suggest a high 8 contribution of OF at this station".

9

10

Sources and export of particle-borne organic matter during a monsoon flood in a catchment of northern Laos

- 3
- E. Gourdin¹, S. Huon², O. Evrard¹, O. Ribolzi³, T. Bariac⁴, O. Sengtaheuanghoung⁵ and
 S. Ayrault¹
- 6 [1]{Laboratoire des Sciences du Climat et de l'Environnement (LSCE), UMR 8212 (CEA-
- 7 CNRS-UVSQ-IPSL), Domaine du CNRS, avenue de la Terrasse, 91198 Gif-sur-Yvette cedex,
- 8 France}
- 9 [2]{Université Pierre et Marie Curie (UPMC), UMR 7618 iEES (UPMC-CNRS-IRD-INRA-
- 10 Université Paris 7-UPEC), case 120, 4 place Jussieu. 75252 Paris cedex 05, France}
- 11 [3]{IRD, Géosciences Environnement Toulouse (GET), UMR 5563 (CNRS-UPS-IRD), 14
- 12 avenue Edouard Belin, 31400 Toulouse, France}
- 13 [4]{CNRS, UMR 7618 iEES (UPMC-CNRS-IRD-INRA-Université Paris 7-UPEC), campus
- 14 INRA AgroParisTech, Bâtiment EGER, 78550 Thiverval Grignon, France}
- 15 [5]{Department of Agriculture Land Management (DALam), P.O. Box 4199, Ban
- 16 Nogviengkham, Xaythany District, Vientiane, Lao PDR}
- 17 Correspondence to: E. Gourdin (<u>elian.gourdin@lsce.ipsl.fr</u>)
- 18

19 Abstract:

20 Tropical rivers of Southeast Asia are characterized by high specific carbon yields and supplies 21 to the ocean. The origin and dynamics of particulate organic matter were studied in the Houay 22 Xon River catchment located in northern Laos during the first erosive flood of the rainy 23 season in May 2012. The partly cultivated catchment is equipped with three successive 24 gauging stations draining areas ranging between 0.2 and 11.6 km² on the main stem of the 25 permanent stream, and two additional stations draining 0.6 ha hillslopes. In addition, the 26 sequential monitoring of rainwater, overland flow and suspended organic matter compositions 27 was realized at 1-m² plot scale during a single storm. The composition of particulate organic matter (total organic carbon, and total nitrogen concentrations, $\delta^{13}C$ and $\delta^{15}N$) was 28 determined for suspended sediment, soil surface (first 2 cm) and soil subsurface (gullies and 29 30 riverbanks) samples collected in the catchment (n = 57, 65 and 11 respectively). Hydrograph

separation of event water was conducted using water electric conductivity and $\delta^{18}O$ data 1 2 measurements measured for rainfall, overland flow and river water base flow (n = 9, 30 and)57, respectively). The composition of particulate organic matter indicates that upstream 3 4 suspended sediments were mainly derived from cultivated soils labelled by their C₃ vegetation 5 cover (upland rice, fallow vegetation and teak plantations) but that collapsed riverbanks, characterized by C₄ vegetation occurrence (Napier grass), significantly contributed to 6 7 sediment yields in particular during water level rise-and at the downstream station. The highest runoff coefficient (11.7%), sediment specific yield (433 kg ha⁻¹), total organic carbon 8 specific yield (8.3 kgC ha⁻¹) and overland flow contribution (78-100%) were found for the 9 reforested areas covered by teak plantations. Swampy areas located along the main stream 10 11 that acted as sediment filter upstream and sediment sources downstream also controlled the 12 composition of suspended organic matter. Despite the low magnitude of the flood, Ttotal organic carbon specific yields were up to 2.6-fold high as this event was the first erosive of 13 14 the rainy season, following the period of slash and burn in the catchment.er (at downstream station) than the annual ones calculated 10 years earlier, before the expansion of teak 15 16 plantations in the catchment. They may be attributed both to the sampling period at the onset of the rainy season (following field clearing by slash and burn) and to the impact of land use 17 18 change during the past decade.

19

20 1 Introduction

21 Soil is the largest terrestrial reservoir of carbon, exceeding biosphere and atmosphere storage 22 capacities (e.g., Sarmiento and Gruber, 2002). Although tropical soils account for ca. 30% of the total carbon storage (e.g., Dixon et al., 1994; Zech et al., 1997), high intensity storms 23 24 (e.g., Goldsmith et al., 2008, Thothong et al., 2011) as well as deforestation and land use 25 change are responsible for high soil carbon losses and deliveries by rivers. For example 26 Houghton (1991) estimated that deforestation in Laos, the sixth most affected tropical country according to the FAO / UNEP (1981), released *ca*. 85 x 10^{12} gC yr⁻¹ to the atmosphere from 27 1979 to 1989. Degens et al. (1991) identified the Asian tropical rivers (e.g., Mekong, 28 <u>Indus/Ganges/Brahmaputra</u>) as the main contributors of dissolved (*ca.* 94 x 10^{12} gC yr⁻¹) and 29 particulate (*ca.* 128 x 10^{12} gC yr⁻¹) organic matter to world oceans, accounting for more than 30 50% of global organic carbon inputs (about 335 x 10¹² gC yr⁻¹ excluding Australia), far before 31 South American rivers. More recently, Huang et al. (2012) estimated that tropical rivers of 32 Asia have the highest specific total-carbon, inorganic and organic, dissolved and particulate 33

carbon yield in which ca. 25% of the delivery is made of particulate organic matter (POM). 1 2 This latter component does not vary linearly with total suspended sediment load (Ludwig et al., 1996), indicating that particulate organic matter is diluted associated withby higher 3 4 concentrations of mineral matter in high TSS loads that is are supplied to the rivers through 5 soil and/or riverbank erosion and sediment remobilization processes taking place along the river courses. Small mountainous headwater catchments play a key role in the delivery pattern 6 7 because they are characterized by high specific discharges and sediment loads (Milliman and 8 Syvitski, 1992). In this context, processes that control organic matter export from tropical 9 catchments should be better understood and constrained, as they account for a significant 10 component in the drawdown or emission of carbon dioxide (Lal, 2003).

11 Tropical storms may also result in the supply of large quantities of suspended sediment to 12 streams (Descroix et al., 2008; Evrard et al., 2010) and lead to numerous problems 13 downstream (Syvitski et al., 2005). Sediments can accumulate behind dams, which results in 14 the siltation of water reservoirs (Downing et al., 2008; Thothong et al., 2011). Suspended 15 organic matter also contributes to water quality degradation (Tanik et al., 1999) and playsing thereby a major role in nutrient biogeochemical cycles (Quinton et al., 2010). It also 16 17 constitutes a potential vector for various contaminants such as metals, polycyclic aromatic hydrocarbons or faecal bacteria (Ribolzi et al., 2010; Gateuille et al., 2014). In order to reduce 18 19 the extent of these negative impacts, sediment delivery by rivers needs to be monitored and 20 controlled. The design and implementation of appropriate management procedures require the identification of the processes that mobilise organic matter from soils and export suspended 21 22 organic matter to riverssources and dynamics. To this end, total organic carbon (TOC) concentration measurements as well as natural ¹⁵N/¹⁴N (e.g., Mariotti et al, 1983; Kao and 23 Liu, 2000, Huon et al., 2006) and ¹³C/¹²C (e.g., Masiello and Druffel, 2001; Hilton et al., 24 25 2010; Smith et al., 2013) stable isotope fingerprinting methods may be used on particulate 26 material collected from hillslopes to rivers, either independantly or in combination with 27 fallout radionuclides to document variations in sediment sources and pathways across 28 catchments (e.g., Ritchie and McCarty, 2003; Ellis et al., 2012; Schindler Wildhaber et al., 29 2012; Ben Slimane et al., 2013; Koiter et al., 2013). In addition, complementary information on sediment conveyed to the river by runoff and overland flow can also be inferred from 30 water tracers such as ¹⁸O natural abundance (for a review see Klaus and McDonnell, 2013). 31

In this study, rainwater, stream water, overland flow and suspended sediment loads were sampled in the partly cultivated headwater catchment of the Houay Xon river, a small tributary of the Mekong River in Laos, during an erosive flood event that took place at the

beginning of the 2012 rainy season. The aim was in order to: (1) estimate the overland flow 1 2 contribution to stream water and and investigate its role for soil organic matter export, and (2) 3 discriminate the respective contributions of surface-soil and stream channel or material to particulate organic matter exportsediment supplies in order to identify the main processes 4 responsible for particulate organic matter delivery at different nested spatial scales. This study 5 6 is complementary to a previous one dedicated to the quantification of sediment dynamics 7 during the same erosive flood event from fallout radionuclide measurements (Gourdin et al., 8 under review2014), that highlighted the binary contribution of soil and stream channel 9 sediment sources during the same erosive flood event.

10

11 2 Study site

The Houay Pano catchment, part of the MSEC (Monitoring Soil Erosion Consortium)
network since 1998 (Valentin et al., 2008), is located 10 km south of Luang Prabang in
northern Laos (19.84°N - 102.14°E; Fig. 1).

15 **[Fig. 1]**

The tropical monsoon climate of the region is characterized by the succession of dry and wet 16 seasons. Almost 80% of annual rainfall (1960-2013 average: 1302 ± 364 mm vr⁻¹) occurs 17 during the rainy season, from May to October (Ribolzi et al., 2008). The Houay Pano 18 permanent stream has an average base flow of 0.4 ± 0.1 L s⁻¹ and is equipped with 5 gauging 19 stations that subdivide the catchment into nested sub-catchments. Two of these stations, S1 20 21 and S4, draining 20 ha and 60 ha respectively, are located along the main stem of the stream. Two additional stations (S7 and S8) draining two hillslopes (0.6 ha each) connected to the 22 23 main stream between S1 and S4 were also monitored. Between S1 and S4, water flows through a natural swamp (0.19 ha), fed by a permanent groundwater table (Fig. 1). Only 24 25 temporary foot slope and flood deposits can be found along the narrow section of the stream and the swamp represents the major-main sediment accumulation zone in the upper 26 27 catchment. The Houay Pano stream flows into the Houay Xon River (22.4 km² catchment) 28 and crosses another swampy area (ca. 3 ha), partly occupied by fishponds (ca. 0.6 ha) at the 29 outlet of the village. Its discharge is continuously monitored at S10 (draining a 11.6 km² subcatchment), located 2.8 km downstream of S4. The Houay Xon River catchment is larger but 30 its channel is not steeper than for the Houay Pano sub-catchment. Its slope is gentler and the 31 connectivity of hillslopes with the main stream is lower. The drainage basin that includes the 32

highest elevations of the catchment is covered with old protected forests but no major
tributary flows into the Houay Xon River. Sediments generated by erosion in the drainage
area can settle before reaching the main stream due to a decline of topography above the left
bank of the river (Fig. 1). The intermittent streams located upstream of S10 did not flow
during the 23 May 2012 flood. The Houay Xon is a tributary of the Nam Dong River, flowing
into the Mekong River within the city of Luang Prabang (Ribolzi et al., 2010).

7 The geological basement of the Houay Pano upper catchment is mainly composed of pelites, 8 sandstones and greywackes (not sampled), overlaid in its uppermost NE part by 9 Carboniferous - Permian limestone cliffs (not sampled) that only cover a very small area in the catchment. Except for the limestone cliffs and some sections of the narrow streambed, 10 soils or flooded soils cover the entire catchment. They Soils consist of deep (>2 m) and 11 moderately deep (>0.5 m) Alfisols (UNESCO, 1974), except along crests and ridges where 12 13 Inceptisols can be found (Chaplot et al., 2009). The soils have low pH ranging between 4.4 14 and 5.5 (Chaplot et al., 2009) indicating that carbonate precipitation is not favoured in soils, 15 even in the upper part of the catchment and, accordingly, cannot supply particulate inorganic carbon to suspended sediment loads. Native vegetation consisted of lowland forest dominated 16 17 by bamboos that were first cleared to implement shifting cultivation of upland rice at the end of the 1960s (Huon et al., 2013). Elevation across the Houay Xon catchment ranges between 18 19 272 and 1300 m a.s.l. As cultivation takes place on steep slopes ranging between 3-2 and 20 $150\%57^{\circ}$, land use evolution in the catchment is prone to soil erosion (Chaplot et al., 2005; 21 Ribolzi et al., 2011). Due to the decline of soil productivity triggered by soil erosion over the 22 years (Patin et al., 2012) and to an increasing labour need to control weed invasion (Dupin et 23 al., 2009), farmers progressively replaced rice fields by teak plantations in the catchment (Fig. 1). In 2012 the Houay Pano catchment was covered by teaks (36%), rotating cropping lands 24 25 under fallow (35%), Job's tears (10%), bananas (4%), upland rice (3%) and secondary forest 26 (<9%). The vegetation cover was different in the larger area drained by the Houay Xon River, 27 with 56% of forests, 15% under teak plantations and 23% croplands.

28

29 **3** Materials and methods

30 3.1 Sample and data collection

31 Rainfall, stream and overland flow waters were sampled during the <u>23</u> May 23 flood in 2012.

32 Rainfall intensity (I) was monitored with an automatic weather station (elevation: 536 m a.s.l.;

Fig. 1) and stream discharge was calculated from water level continuous recording and rating curves. Estimates of event water discharge (EWD), defined here as the total water volume exported from each sub_catchment during the event minus the base flow discharge, were calculated by adding sequential water volumes corresponding to the average discharge between two water level measurements. Specific runoff (SR, in mm) was obtained by dividing EWD by the corresponding sub-catchment area (Chow et al., 1988).

7 Rainfall was sampled with three cumulative collectors located: in the village near the 8 confluence between the Houay Pano and Houay Xon streams, near a teak plantation on the 9 hillslopes located just upstream of the village and within the Houay Pano catchment (Fig. 1). The runoff coefficient (RC) corresponds to the fraction of total rainfall that was exported from 10 the catchment during the event. Overland flow was collected at the outlet of $1-m^2$ 11 experimental plots (OF_{1m^2}) designed for runoff studies (Patin et al., 2012). For one of them 12 13 (Fig. 2) the evolution of rainwater, overland flow and suspended organic matter composition 14 was monitored during a rainfall event (June 1, 2012), simultaneously at its outlet and for a *ca*. 15 8-m² rain-collector set-up located a few meters apart. The experiment was conducted on a soil with 33%18° slope and *ca*. 60% fallow vegetation cover (*ca*. 10 cm high; Fig. 2a). The rain 16 17 collector was installed at 1.8 m above soil surface to avoid splash contamination. Four samples were collected in the first 3 cm of a soil profile (0-5 mm; 6-10 mm; 11-20 mm; 21-30 18 19 mm) within a ca. 400-cm² area adjacent to the experimental plot to estimate the composition 20 of organic matter in the topsoil layer (Fig. 2b).

21 [Fig. 2]

22 River water was collected in 0.65 L polyethylene bottles for each 20-mm water level change 23 by automatic samplers installed at each gauging station. Sixty-nine total suspended sediment (TSS) samples were collected for five stations, S1, S4 and S10 on the main stem and S7 - S8 24 25 for hillslopes drained by temporary tributaries (Fig. 1). Shortly after collection all samples were dried in 1 L aluminium trays in a gas oven (ca. 60-80°C) for 12-48 h. Preliminary 26 studies carried out in 2002-2007 showed that dissolved organic carbon concentrations in the 27 Houay Pano stream water are commonly low, 1.8 ± 0.4 mg L⁻¹ (n = 74) and 2.0 ± 0.7 mg L⁻¹ 28 (n = 65), at base flow and discharge peak, respectively. With high-suspended sediment loads 29 (see further in the Results section), a 3 mgC L^{-1} content for dissolved organic carbon would 30 represent 1-10 wt% of the total (dissolved and particulate) organic carbon load. In average 97 31 \pm 3 % of the total organic matter recovered is made of particulate organic matter, 90-95% 32 during the water rising stage and 95-99% for the other water levels. We are confident that all 33

measurements account for particulate organic matter with negligible dissolved loads and that 1 2 the dynamics of organic compounds during the flood refers to particulate matter. Samples were dried shortly after collection in an oven (t $\approx 100^{\circ}$ C) for 12-48 h. To complete the topsoil 3 data set available for the catchment (Huon et al., 2013), additional soil cores were collected 4 5 on hillslopes connected to the Houay Pano stream and the Houay Xon River (Fig. 1) in May 6 and December 2012. Sampling was further completed with several gully (n = 5) and riverbank 7 (n = 6) samples in December 2012 to document the characteristics of the potential subsurface 8 sources of sediment to the river. No soil sample was collected in the south-eastern part of the 9 catchment of S10.

10 Cumulated suspended sediment yields (SSY) were calculated at each station by adding the 11 total suspended sediment (TSS) masses exported between two successive samples. The TSS 12 concentration was considered to vary linearly between successive measurements. Specific 13 sediment yields (S_Y) were calculated by dividing the cumulated SSY by the corresponding 14 drainage area.

15 **3.2** Particulate organic matter composition measurements

All samples were finely grounded with an agate mortar, weighed and packed into tin capsules 16 17 (5 x 9 mm) for analysis. Total organic carbon (TOC) and total nitrogen (TN) concentrations, and ¹³C/¹²C and ¹⁵N/¹⁴N stable isotopes were measured using the Elementar[®] VarioPvro cube 18 analyzer on line with a Micromass[®] Isoprime Isotope Ratio Mass Spectrometer (IRMS) 19 facility (IEES, Paris). Analytical precision was better than $\pm 0.42 - 0.3\%$ vs. PDB-AIR 20 standards (Coplen et al., 1983) and 0.1 mg g⁻¹ (equivalent to 0.01 wt.%) for δ^{13} C- δ^{15} N and 21 TOC-TN, respectively. Data reproducibility was checked by replicate analyses of selected 22 23 samples and of a 99% pure tyrosine laboratory standard (Girardin and Mariotti, 1991) using 24 18 tyrosines per batch of 50 samples. Selected sample measurements were also repeated 25 during the course of the study. The possible occurrence of carbonate minerals (or carbonate 26 rock fragments) in TSS samples, collected at different stages of the flood at stations S1 and 27 S4, was checked by pouring drops of a 30% HCl solution on dry sample aliquots. No CO₂bubbling, typical for carbonate dissolution, was observed. Therefore, common carbonate 28 minerals such as calcite do not represent a detectable fraction of the suspended sediment loads 29 30 and could be neglected. Due to the absence of carbonate minerals in sediments and suspended loads, nNo additional treatment was requirapplied. For the entire flood, total particulate 31 organic carbon yields (C_{SSY}) were calculated by summing the successive TOC contents 32 associated with suspended sediments (SSY multiplied by TOC concentration). The TOC 33

1 concentration of particulate organic matter was assumed to vary linearly between successive 2 samples. Specific TOC yields (C_Y) were calculated by dividing the cumulated C_{SSY} by the 3 corresponding drainage area.

4 3.3 Water δ^{18} O and electrical conductivity measurements

Water aliquots were recovered in 30-mL glass flasks from stream, overland flow and rain 5 samples (see section 3.1 for details) and filtered using $<0.2 \ \mu m$ acetate filters. Stable ${}^{18}O/{}^{16}O$ 6 7 isotope measurements were carried out using the standard CO₂ equilibration method (Epstein and Mayeda, 1953) and determined with a VG Optima® mass spectrometer (IEES, Thiverval-8 Grignon). Isotopic ratios are reported using the δ^{18} O notation, relative to the Vienna-Standard 9 Mean Ocean Water (V-SMOW; Gonfiantini, 1978) with an analytical precision better than \pm 10 0.1‰. Water electrical conductivity (EC) was monitored every 6-min at the inlet of each 11 gauging station using Schlumberger in situ CTD probes. Additional measurements were 12 conducted using an YSI[®] 556 probe for manually collected samples. Hydrograph separation 13 was carried out with end-member mixing equations using water electrical conductivity and 14 δ^{18} O measurements (Sklash and Farvolden, 1979; Ribolzi et al., 2000; Ladouche et al., 2001). 15

16

17 **4 Results**

4.1 Composition of the potential sources of particulate organic matter in the catchment

The mean organic matter characteristics are reported in **Table 1** for surface soils, gullies and stream banks collected in the catchment, together with ¹³⁷Cs activity determined on the same sample aliquots (Huon et al., 2013; Gourdin et al., <u>under review2014</u>). In contrast to the high ¹³⁷Cs activities measured in surface soil samples, gully and riverbank sites are depleted in this radioisotope (**Table 1**).

25 [Table 1]

Surface (soils) and subsurface (stream banks and gullies) sources of particulate organic matter are best discriminated by their TOC content that is higher in surface soils. The dominance of C_3 photosynthetic pathway plants across the catchment is reflected by low δ^{13} C values in soils (-25.5 ± 1.4‰). However, soil-originating particles accumulated in sediments of the swamp provide ¹³C-enriched compositions, up to *ca.* -15‰, that are explained by the strong contributioninput of particulate organic matter derived from C₄ photosynthetic pathway plant tissues. The latter are mainly Napier grass growing in the swamp and along limited sections of the stream channel, and to a much lower extent, Job's tears and maize cultivated on nearby hillslopes (Huon et al., 2013). Soil surface and subsurface sources can also be distinguished by their δ^{15} N values that are slightly lower for surface sources the former (**Table 1**). The overall values reflect high 15 N/ 14 N fractionation during incorporation and mineralization of plant tissues in soils, typical for tropical environments (e.g., Amundson et al., 2003).

8 9

4.2 Monitoring water and particulate organic matter exports at the microplot scale during a rainfall event

10 The distribution of organic matter composition with soil depth is displayed on Fig. 2b. The TOC content decreases exponentially with depth together with TN (not plotted), leading to a 11 nearly constant TOC : TN ratio of *ca.* 10 (Fig. 2b). Both δ^{13} C and δ^{15} N increase with soil 12 depth from -26.3 to -24.7‰ and from 6.6 to 8.6‰, respectively, reflecting the contribution of 13 fallow vegetation debris depleted in ¹³C and ¹⁵N with respect to soil organic matter (Balesdent 14 15 et al., 1993). Overland flow samples (OF) were collected continuously at the outlet of the experimental plot during the June 1^{st} storm that lasted for 45-min. Cumulated rainfall was *ca*. 16 11 mm and its intensity reached 30 mm h⁻¹ during 20 min. Suspended sediment concentration 17 increased to a maximum of 4.7 g L^{-1} (Fig. 2c-d). The estimated runoff coefficient was 77% 18 during the entire storm with an average infiltration rate of 3.3 mm h⁻¹, assuming no 19 evaporation during rainfall. As shown on Fig. 2c, suspended sediments exported from the 20 experimental plot were characterized by TOC, TOC/TN, δ^{13} C and δ^{15} N values that match 21 22 topsoil organic matter composition (Fig. 2b), with a slight evolution towards the composition of deeper superficial layers (1-3 cm) at the end of the event. The higher TOC and lower $\delta^{13}C$ 23 and δ^{15} N recorded at the beginning of the storm likely result from the preferential export of 24 vegetation debris fine soil organic matter. Similar behaviours were reported by Clark et al. 25 26 (2013) in the tropical Andes and interpreted as a greater contribution of non-fossil POC 27 during the rising stage and the peak discharge, and in the Swiss Alps by Smith et al. (2013) 28 who interpreted the initial decrease of POC during the rising stage as resulting from inchannel clearing. The evolution of rainwater and OF δ^{18} O is shown on Fig. 2d. At the 29 beginning of the storm, both displayed a similar decreasing δ^{18} O trend (from -3.8 to -5.5%) 30 with increasing rainfall intensity, concomitant to a rise of the suspended load. Overland flow 31 32 EC averaged $20 \pm 6 \ \mu\text{S cm}^{-1}$ (range: 15 - 36 $\ \mu\text{S cm}^{-1}$, n = 17). The values are consistent with

the ones of two other cumulated OF samples, 21 and 43 μ S cm⁻¹, collected in the Houay Pano catchment during the <u>May 2323 May 2012</u> event (see section 4.3). Contrasted increasing trends were also observed for rain- and OF-¹⁸O contents (reaching -1.7‰ and -4.0‰, respectively) during the falling water stage. They reflected the mixing of progressively ¹⁸Oenriched rainwater with former ¹⁸O-depleted rainwater temporarily stored in the topsoil. It is likely that OF that triggers soil detachment and suspended sediment export will better reflect the contribution of event water to the main stream than rainwater.

8

4.3 Hydro-sedimentary characteristics of the May 2323 May 2012 flood

9 Thise May 23 flood was triggered by a 48 min storm that brought 27 mm of cumulated 10 rainfall between 11:36 am and 12:24 pm. According to Bricquet et al. (2003), this event has a 11 return period of *ca*. 0.01 year (34.7 mm day⁻¹). It was the first significant erosive event of the 12 2012 rainy season and the first event with rainfall intensity exceeding 80 mm h⁻¹ (6-min time 13 steps). The main hydro-sedimentary characteristics of the flood are reported for the three 14 gauging stations in **Fig. 3 I-II-IIIa-b-c-d**.

15 [**Fig. 3**]

16 The lag time between stream discharge (Q) and rainfall intensity peaks differed at the 17 successive stations. Q increased 10 min after the rainfall peak and reached its maximum 10 18 min later at S1 (Fig. 3Ia), whereas both peaks were synchronous at S4 (Fig. 3IIa). Downstream, the lag time between rainfall and Q peaks increased to 70 min at S10 (Fig. 19 **3IIIa**). The evolution of TSS concentration that peaked at 24-47 g L^{-1} (Fig. 3I-II-IIIb) 20 displayed counterclockwise hysteresis dynamics (Williams, 1989; Lenzi and Marchi, 2000) at 21 22 the three stations. Even though Q increased faster than TSS concentration at the beginning of 23 the flood, water EC decreased concomitantly at the three stations (Fig. 3I-II-IIIc). This 24 behaviour suggests the progressive mixing of pre-event water (i.e. groundwater) with a low 25 TSS load by weakly mineralized event water (i.e. overland flow) with high sediment loads, the proportion of the latter increasing with decreasing EC. Pre-event EC values measured in 26 the stream just before the flood were 394, 320 and 450 $\mu S~cm^{-1}$ at S1, S4 and S10, 27 28 respectively (Fig. 3I-II-IIIc) in contrast with the low values determined for OF (see above). 29 As expected, the highest values were recorded at S10, which is located downstream of riparian villages (Ribolzi et al., 2010) where high EC wastewaters are directly released into 30 the river. In contrast, upstream of this village, stream waters exclusively originate from 31 cultivated lands. Pre-event water ¹⁸O content was estimated to -7.1‰ at station S4 with 32

samples collected before peak flow rise (**Fig. 3IId**). However, for S1 and S10, automatic sampling only took place during the water rising stage and the composition of pre-event water had to be estimated. At S1, a δ^{18} O value of -8‰ corresponding to a maximum EC of 394 µS cm⁻¹ was estimated by fitting the correlative trend (see **section 5**). Pre-event and event waters could not be distinguished with δ^{18} O signatures at S10. Overall, despite the limited number of samples collected, the composition of cumulated rainwater remained rather constant in the catchment (-5.1, -5.5 and -5.6‰), averaging -5.4 ± 0.3‰.

8 4.4 Particulate organic matter export at catchment scales during the May 2323 May 9 2012 flood

10 Large-Important variations in suspended organic matter composition were recorded at S1 with TOC concentration (20-70 mgC g⁻¹, Fig. 3Ie), TOC/TN (8-31, Fig. 3If), δ^{13} C (-26 to -15‰, 11 Fig. 3Ig) and δ^{15} N (5.5-8.0‰. Fig. 3Ih) measurements. They all indicate changes in the 12 source delivering suspended organic matter during the rising water stage. The δ^{13} C signature 13 14 of suspended organic matter reach the average composition (-25.5 \pm 1.4%; Table 1) of 15 topsoil organic matter in the catchment at peak flow and during the recession stage (Fig. 3I-16 IIg). Due to larger and more heterogeneous areas drained at S4 and S10, the temporal evolution of TOC/TN, δ^{13} C and δ^{15} N in TSS (Fig. 3II-III, e-f-g-h) were less contrasted than at 17 S1. At S10, the mean TOC/TN was higher (17.0 \pm 3.2) than at S1 (13.1 \pm 5.9) and S4 (10.3 \pm 18 19 0.9), reflecting a greater contribution of vegetation debris and / or weakly mineralized organic 20 matter downstream than in upper parts of the catchment (Table A1). Furthermore, the highest 21 TOC/TN (23; Fig. 3IIIf) was obtained during the water discharge peak at S10 whereas it was recorded at the beginning of the rising stage at S1 (31; Fig. 3If). 22

23

24 **5** Interpretation and discussion

25 **5.1** Overland flow contribution to stream discharge

As overland flow is the main supply of eroded particulate organic matter to the streams during the flood, hydrograph separation in pre-event groundwater and event water contributions using end-members mixing equations should provide information on water dynamics and suspended sediment sources during the flood. However, several questions may arise regarding the relevance of using water mass tracers to constrain end-members signatures and provide reliable estimates of overland flow contribution.

1 5.1.1 Evolution of water composition during the flood

Water electrical conductivity and δ^{18} O measurements conducted on rainwater, overland flow and stream water highlight in-channel mixing processes between base flow groundwater (preevent water) and event water characterized by contrasted signatures (**Fig. 4I**).

5 [Fig. 4]

At S1 (Fig. 4Ia), all samples are aligned between the PEW and OF_{1m^2} end-members during 6 both rising and recessing stages, suggesting that the composition of corresponding source 7 8 remained constant during the event. This condition is one of the assumptions underpinning 9 hydrograph separation procedures (e.g. Buttle, 1994; Ribolzi et al., 2000; Klaus and 10 McDonnell, 2013). At S4 (Fig. 4Ib), the evolution of stream water composition during the 11 flood displays a more complex pattern, with the succession of three phases characterized by distinct behaviours. During the rising stage, a similar trend between PEW and OF_{1m^2} is 12 observed as for S1. Near peak flow, stream water EC and δ^{18} O concomitantly decrease 13 towards the signature of cumulated rainwater samples (Fig. 4Ib) until the dilution of PEW by 14 15 EW reaches its maximum. This behaviour likely reflects the progressive depletion of rainwater in ¹⁸O during the storm, as observed during the microplot experiment (**Fig. 2d**), 16 17 following a Rayleigh-type distillation process (Dansgaard, 1964). The decrease of EC in 18 stream water is also consistent with the supply of weakly mineralized overland flow water 19 mixing rainwater and pre-event soil water with low and high dissolved loads, respectively. A 20 remarkable point is that the water composition supplied by S7-S8 sub-catchments, referred to as $OF_{0.6ha}$ (Fig. 4Ib), closely matches the composition of stream water during this period. 21 22 Finally, during the third phase corresponding to the recession period, the composition of the 23 river water evolved towards the "initial" PEW signature along a third mixing line. At S10, stream water composition displayed large variations in EC but limited changes in δ^{18} O (range: 24 from -6.0 to -5.2‰, Fig. 4Ic). The EC values, decreasing from 450 to 155 μ S cm⁻¹ at the 25 beginning of the event (Fig. 3IIIc)ranging between 155 and 450 µS cm⁻¹, suggest a high 26 27 contribution of OF at this station.

28 5.1.2 Catchment hydrological characteristics inferred from hydrograph separation

As highlighted by Klaus and McDonnell (2013), high-frequency analyses of rainfall-runoff are necessary to record end-members intra-event signature variations and reduce uncertainties on hydrograph separation. The microplot experiment previously described recorded such temporal variations during a single storm event (**Fig. 2d**). The OF signature displayed lower

1 variations (-5.5 to -3.7‰) than rainwater (-5.6 to -1.7‰) as a result of mixing between rain 2 and soil water. Although samples could not be taken during the May 2323 May 2012 flood, a similar intra-storm evolution magnitude of *ca*. 2‰ for OF- δ^{18} O was assumed. In order to 3 estimate event water contribution to total water discharge monitored at each station, this 4 5 possible intra-storm variation of rainwater and overland flow signature must be taken into account, as suggested by McDonnell et al. (1990). The very close δ^{18} O values of the three 6 7 rainwater samples collected on May 2323 May 2012 across the catchment remain consistent 8 with the first assumption formulated by Harris et al. (1995) regarding spatial uniformity of 9 cumulated rainwater isotopic signature. However, the behaviour of stream water during peak discharge at S4 (Fig. 3IId-4Ib) suggests the evolution of the OF end-member signature 10 towards low $\delta^{18}O$ (as recorded for $OF_{0.6ha}$ in Fig. 4Ib), consistent with a Rayleigh-type 11 distillation of rainwater. Pre-event soil water signature, likely enriched in ¹⁸O by evaporation 12 at the onset of the rainy season (e.g., Hsieh et al., 1998), could not be characterized. Its higher 13 δ^{18} O range can be assumed to be responsible for the higher δ^{18} O observed for OF_{1m²} during 14 the May 2323 May 2012 flood (-3.9 to -2.5%; Fig. 4Ib). The higher EC values recorded for 15 $OF_{0.6ha}$ compared to OF_{1m^2} likely result from dissolved elements loading by runoff due to 16 interactions between rainwater, vegetation, and soil particles along slopes. As the temporal 17 evolution of rainwater and of the resulting $OF-\delta^{18}O$ values could not be measured during the 18 May 2323 May 2012 flood, we used EC only to provide estimates of overland flow 19 20 contribution, taking into account the potential variation of this end-member's signature, from 20 to 150 μ S cm⁻¹, during the event (**Fig. 4II**). 21

22 [Table 2]

23 Estimates of event water discharge (EWD), specific runoff (SR) and runoff coefficient (RC)
24 are summarized in Table 2.

25 **[Table 2]**

Runoff coefficients are rather low in most parts of the catchment (4.0 and 3.9% at S1 and S10, respectively), except at S4 which displayed a higher value of 11.7% (**Table 2**). Overall, those low runoff coefficients remained consistent with the high infiltration rates reported by Patin et al. (2012) in the same area (>100 mm h⁻¹). Chaplot and Poesen (2012) reported an annual runoff coefficient of *ca*. 13% for twelve 1-m² plots monitored in this catchment. The v¥alues decrease both with hillslope downward position of the experimental plots and for increasing drainage area, down to 6% for S4 and 1.5% for S10. Estimates of the OF contribution to total

1 water discharge, based on the evolution of water EC, are displayed on Fig. 4II. At discharge 2 peak, OF was lower at S1 (53-80%) than at S4 (78-100%) and S10 (67-95%). The highest 3 value was obtained at S4 where the highest runoff coefficient was also recorded. This 4 behaviour likely results from a different soil cover in this sub-catchment. Indeed, teak 5 plantations prone to soil erosion and low infiltrability conditions (Patin et al., 2012) covered 6 32% of this sub-catchment area in 2012, whereas it had a two-fold smaller extension in the 7 drainage areas of S1 (14%) and S10 (15%). Moreover, the annual runoff coefficients reported 8 by Chaplot and Poesen (2012) at S4 and S10 were lower than those reported in this study, but 9 they were measured when teak plantations covered a much lower part of the catchment (2002-10 2003, Chaplot et al., 2005). Overall, it is likely that teak plantations will enhance overland 11 flow and soil erosion at least during the years following land use conversion.

12 **5.2** Particulate organic matter delivery <u>during the 23 May 2012 flood</u>

13 5.2.1 Sources and dynamics of suspended organic matter in the catchmentduring the 14 May 23 flood

15 Variations in the composition of particulate organic matter reflect changes in the source 16 supplying suspended sediment in the catchment during the flood. For S1 and S4, this evolution follows hyperbolic trends with suspended sediment loads for TOC, $\delta^{13}C$ and $\delta^{15}N$ 17 18 and tends to reach the mean composition of catchment surface soils during the main transport 19 phase (Fig. 5I-IIa-b). As reported by Bellanger et al. (2004) in the Venezuelan Andes, this 20 behaviour indicates that sheet erosion iwas likely the dominant process. Due to the absence of 21 particulate inorganic carbon that could have biaised the measurements (see above in section 3.2), the composition of suspended sediments is consistent with the supply of carbonate free 22 soil-detached particles exported from catchment's soils. Furthermore, the ¹³⁷Cs activity of 23 suspended sediments (Gourdin et al., under review) remains within the range of surface soil 24 activities (>1 Bq kg⁻¹; Table 1) and supports this interpretation. 25

26 **[Fig. 5]**

However, Meybeck (1993) outlined that hyperbolic trends may indicate that a significant fraction of particulate organic matter exported from mountainous regions by rivers may be supplied by the direct erosion of sedimentary – metamorphic bedrocks (the so-called "fossil carbon" pool) and pointed out that neglecting this source induces a bias in carbon budgets. Fossil carbon may account for 90-100% of total particulate organic matter exported in rivers with average annual suspended loads exceeding 5 g L⁻¹ (Meybeck, 2006), in the range

1 recorded for this study. In the Andes, Clark et al. (2013) identified fossil POC contributions associated with TSS concentrations above 1 g L⁻¹. In a Taiwanese river, Hilton et al. (2011) 2 reported suspended sediment concentrations up to ca. 30 g L^{-1} leading to fossil POC 3 concentrations up to *ca*. 0.1 g L^{-1} . Fossil particulate organic carbon contributions have been 4 identified using ¹⁴C natural abundance and C-N stable isotope measurements in various 5 studies (e.g., Kao and Liu, 1997; Raymond and Bauer, 2001; Copard et al., 2007; Graz et al., 6 7 2012; Smith et al., 2013). In our The present study, does we could not support the hypothesis 8 of a identify any significant supply of rock-derived fossil carbon. This export is, often 9 associated with important sediment exports originating from gully systems (Duvert et al., 10 2010), landslides and mass movements that (Huon et al., 2006), which were not observed in 11 the Houay Pano catchment during this medium magnitude flood. Bedrock outcrops are scares and not directly connected to the stream. Moreover the highest $\delta^{15}N$ values rather reflect the 12 occurrence of soil derived organic matter than fossil organic matter (e.g. Huon et al., 2006). 13 The later should theoretically provide lower $\delta^{15}N$ than for soils as preservation of organic 14 matter in sedimentary and low metamorphic grade rocks takes place at "high temperature" 15 (low ¹⁵N/¹⁴N fractionation with respect to vegetation) whereas incorporation and stabilization 16 of organic matter in soils should occur at "low temperature" (high ¹⁵N/¹⁴N fractionation). 17 Fossil particulate organic carbon contributions have been identified using ¹⁴C natural 18 abundance and C-N stable isotope measurements in various studies (e.g., Kao and Liu, 1997; 19 20 Raymond and Bauer, 2001; Copard et al., 2007; Graz et al., 2012; Smith et al., 2013). For the Houay Xon catchment, the ¹³⁷Cs activity of suspended sediments measured on the same 21 sample aliquots are in the range of surface soil activities (above 1 Bq kg⁻¹; **Table 1**: Gourdin 22 et al., 2014). Paleozoic bedrocks could not be tagged by fallout ¹³⁷Cs whose supply only took 23 24 place in the 1960-1970's (Ritchie and McHenry, 1990)Furthermore, suspended sediments are labelled by ¹³⁷Cs activities that reflect the dominant contribution of soil surface sources 25 (Gourdin et al., under review). 26

27 **5.2.2 Dynamics of suspended organic matter**

At S1, the ¹³C-enriched compositions (**Fig. 5IIa**) first reflect the supply of organic matter derived from C₄ photosynthetic pathway plants (Huon et al., 2013)as observed in the field. With increasing water discharge, suspended sediments progressively incorporate ¹³C-depleted organic matter originating from soils covered by C₃ photosynthetic pathway plants that dominate in the drainage area. Decreasing TOC/TN (increasing TN/TOC) and increasing δ^{15} N trends during the flood are best explained by the re-suspension of weakly mineralized (low 1 δ^{15} N) C₄-plant debris (high TOC/TN), followed by their mixing with soil organic matter 2 exported from cultivated fields and supplied by overland flow to the main stream (low 3 TOC/TN, high δ^{15} N). <u>Plot of δ^{13} C *vs*. TN/TOC shows that the composition of suspended 4 sediment loads matches that of the main pools of particulate organic matter in the catchment, 5 i.e., surface soils and subsurface soils (gullies and river banks, **Fig. 6**). Mixing between the 6 two end-members is pictured by correlative behaviours for S1 and S10.</u>

7 [Fig. 6]

8 It is worth noticing that bedrock source compositions available from literature for tropical catchments (i.e., Kao and Liu, 2000, Hilton et al., 2010) fall outside the observed mixing 9 10 trends. In additionHowever, the occurrence of light density charcoal fragments produced by 11 slash-and-burn cultivation might have slightly increased TOC/TN with respect to soil organic 12 matter (Soto et al., 1995; Rumpel et al., 2006). Overland flow supply of particulate organic 13 matter exported from soils that are currently or were previously cultivated with upland rice, is largely dominant at S4, compared to S1 (Fig. 5I-IIa-b). Fields cropped with C_4 -plants only 14 cover small areas in the catchment and their imprint on soil organic matter composition is 15 therefore limited (Huon et al., 2013). The δ^{13} C recorded during and after the water discharge 16 peak were similar (-25.7%; Fig. 5IIa-b) to those of surface soils, reflecting the dominance of 17 18 surface vs. subsurface sources in Houay Pano catchment. At S8, located close to S4 (Fig. 1), δ^{15} N increased noticeably from 6.5 to 8.3% during the storm, indicating that ¹⁵N-depleted 19 organic matter (i.e., vegetation debris) was first exported and that erosion progressively 20 affected deeper ¹⁵N-enriched layers of the topsoil (**Table 1**). In contrast to the two other 21 22 stations, the maximum TOC/TN (23) recorded downstream at S10 occurred during the water 23 discharge peak (Figs. 3IIIf and 5Ic). Fresh organic matter characterized by high ratios is 24 exported with a time lag due to the remote location of its source (Gurnell, 2007). Suspended organic matter transported at the beginning of the flood (range: from -23 to -21‰; Table A1, 25 Fig. 5IIc) is enriched in ¹³C and ¹⁵N compared to the mean surface soil $(-25.5 \pm 1.4\%)$ and 26 27 matches subsurface soil signatures (stream banks and gullies, Table 1). This observation 28 validatesupports previous findings showing the dominance of riverbank erosion 29 suggest characterized by the depletion in fallout radionuclides measured for sediments collected at this station (Gourdin et al., under review2014). Positive correlative trends 30 between soil TOC and ¹³⁷Cs inventories suggest that a similar process, i.e. erosion and 31 erosion-induced carbon depletion, controlled their concomitant decrease since the onset of 32 33 cultivation in the 1960's (Huon et al., 2013). Smith and Blake (2014) reported similar

correlations for riverine sediments in parts of their study sites. No such relationships could be
 put forward during the 23 May 2012 flood (data not shown).

3 Contribution of overland flow to stream water discharge derived from hydrograph separation 4 can be linked to the source of suspended organic matter (Fig. 5 III) as well as to the extent of 5 particulate organic matter transfer (Fig. 5 IV). In terms of water - sediment dynamics, high OF contributions (above ca. 50%) supply large quantities of soil organic matter (fingerprinted 6 7 by lower TOC contents and enriched isotopic compositions compared to fresh vegetation 8 debris) to the river. In contrast, low OF contributions may indicate the dominance of 9 riverbank erosion and remobilization of material-sediment deposited on the riverbed 10 duringafter previous floods. Based on this hydrograph separation, it is then possible to draw 11 sediment and particulate organic carbon budgets at the catchment's scale in areas where 12 surface soil erosion dominates.

13 **5.2.2** Suspended sediment TOC-¹³⁷Cs relationships

Positive correlative trends between soil TOC and ¹³⁷Cs inventories suggest that a similar 14 process, i.e. erosion and erosion induced carbon depletion, controlled their concomitant 15 decrease since the onset of cultivation in the 1960s (Huon et al., 2013). Similar positive 16 17 correlations were reported by Smith and Blake (2014) for riverine sediments in parts of their 18 study sites. We could not derive such relationships for suspended sediment loads during the May 23 flood. This observation may reflect selective detachment and transport, with respect 19 20 to cultivated soils, of small size mineral-bound organic matter to the rivers. It could however also result from the local contribution of channel bed organic matter, degraded with time, 21 22 inducing TOC depletion and ¹³C enrichment. This later interpretation is supported by the large proportion of remobilized sediments fingerprinted by the low ⁷Be:²¹⁰Pb_{xs} activity ratios 23 measured in suspended sediment loads (Gourdin et al. under review). 24

5.2.3 Suspended sediment and <u>particulate organic</u> carbon deliver<u>iesy at catchment</u> scale

Total suspended sediment exports are summarized in **Table 3** for S1, S4 and S10 subcatchments.

29 **[Table 3]**

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30 The sediment yield (S_Y) of *ca.* 433 kg ha⁻¹ at S4 is greater than at S1 and S10 (**Table 3**) and 31 consistent with higher specific runoff and runoff coefficient values (**Table 2**). Due to the low

Sx-observed at S1, the succession of nested catchments was not related to a decrease in 1 2 specific delivery when drainage area increased. Compared to the 2002-2003 annual sediment deliveries-yield at S4 (2090 kg ha⁻¹ yr⁻¹) and S10 (540 kg ha⁻¹ yr⁻¹) reported by Chaplot and 3 Poesen (2012), the May 2323 May 2012 flood representeds ca. 21% of the total annual 4 5 exports recorded for both stations. These deliveries are very high for a single event of moderate intensity. However, fallout radionuclide measurements (Gourdin et al., under 6 7 review2014) indicate that thise May 23 flood was the first important erosive event of the 2012 8 rainy season and that exported matter mainly consisted of remobilized river channel sediments (ca. 80%). that may not have been fully taken into account in the previous study. 9 The TSS yield (S_v) of *ca*. 433 kg ha⁻¹ (8.3 kgC ha⁻¹) at S4 is greater than at S1 and S10 10 (Table 3) and consistent with higher specific runoff and runoff coefficient values (Table 2). 11 With a low value at S1, the succession of nested catchments was not related to a decrease in 12 13 specific delivery when drainage area increased. This unsual behaviour is best explained by the occurrence of swamp areas along the main stream. In the upper part of the catchment, a 14 natural swamp acts as a filter for sediments conveyed during low to medium magnitude floods 15 16 (Fig. 1). Napier grass, the main aquatic plant forms dense masses of litter that reduce stream 17 flow velocity during the rainy season. Nearly 33 Mg of soil-derived organic carbon was thus accumulated since the early 1960's (Huon et al., 2013). This swamp played a key role with 18 19 respect to downstream export of suspended sediment during the 23 May 2012 flood. It also explains why the high δ^{13} C values of TSS loads, observed during the rising water stage 20 21 upstream of the swamp (at S1), were only partly transmitted to S4. Soil-derived organic 22 matter supplied by overland flow replaced the major part of the TSS during the rising stage, 23 downstream of the swamp. A comparable picture can be drawn for the wetlands located at the outlet of the village. However in contrast this swampy area where streambanks are also 24 encroached by Napier grass contributed to a rise of the δ^{13} C values of suspended organic 25 matter at the monitoring station S10. This shift fingerprinted the extent of streambank 26 27 sediment retention and mobilization processes taking place downstream in accordance with 28 radionuclide activity measurements carried out for the same samples (Gourdin et al., 2014). Carbon specific deliveries (C_Y) suggest a higher erodibility of the S4 draining area, exporting 29 ca. 8.3 kgC ha⁻¹ of soil organic carbon, i.e. more than twice the quantity exported from S1 and 30 S10 (2.9-3.7 kgC ha⁻¹; Table 3). This behaviour may be related to higher RC (11.7%; Table 31 2) and OF contribution at discharge peak (78-100%; Fig. 4IIb) estimated at this station. As

for sediment delivery, we calculated a much higher carbon flux exported by the catchment 33 than previously reported by Chaplot and Poesen (2012), i.e., 8.5 and 1.4 kgC ha⁴-yr⁴ for S4 34

32

and S10, respectively. To the best of our knowledge, this discrepancy may be explained by 1 2 the different TOC concentrations used for carbon flux calculation in both studies. In this study the values are almost 5-fold higher for S4 (25 vs. 4.1 mgC g⁻¹) and 14-fold higher for S10 (36 3 vs. 2.6 mgC g⁻¹). As the same analytical method was used in both studies, these differences 4 could also be explained by a greater contribution of deep soil layers through linear erosion 5 (gullies and riverbanks) during the 2002 rainy season, responsible for the export of sediments 6 7 with low TOC content ($<5 \text{ mgC g}^{-1}$) in all catchments draining a surface exceeding 0.6 ha. 8 Total organic carbon and total nitrogen concentrations of riverine sediments are usually 9 higher than in soils due to preferential mobilisation and transport of fine and light soil organic fractions after aggregates destruction (Stoltenberg and White, 1953). However, Chaplot and 10 Poesen (2012) observed rather similar TOC enrichment factors (close to 1), suggesting a good 11 stability of soil aggregates in their study. The low carbon yields (8.5 and 1.4 kgC ha⁻¹ yr⁻¹, for 12 13 S4 and S10 respectively) reported by these authors indicate that nearly all soil particulate organic matter deposited on hillslopes before reaching the river channel. The connectivity of 14 15 hillslopes to the stream channel may also have changed since 2002-2003 due to the 16 replacement of cultivated plots by teak plantations initiated in 2009 in the catchment. Teaks 17 are characterized by large leaves that concentrate rainwater and enhance raindrop erosivity, soil crusting and runoff, especially after 10 years (Patin et al., 2012). However, their impact 18 19 on soil erosion and organic matter export is still poorly understood (C. Valentin, personal communication) and should be further investigated. 20

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6 Concluding remarks

The composition of suspended organic matter and stream water, monitored during the first erosive <u>– medium magnitude flood</u> event of the 2012 rainy season in a cultivated catchment of northern Laos, provided an efficient way to quantify the evolution of particulate organic matter sources along a network of nested gauging stations.

In <u>the upper parts</u> of the <u>drainage basin (Houay Pano sub-</u>catchment), the composition of
suspended organic matter exported <u>washows that sediment</u> mainly originateding from inchannel and nearby sources during the rising stage, and from cultivated surface soils at peak
flow and during the recessing stage.

31 Downstream, the composition of suspended and deposited organic matter in the Houay Xon
 32 <u>River</u> reflected the dominant supply of subsurface sources (riverbanks and gullies) and a

subsequent dilution of the soil-derived organic matter delivery by channel - bank mixing and
 remobilization processes.

3 Wetlands and swampy areas played a key role in the process by trapping sediment upstream

4 in the steep part of the catchment and by remobilizing riverbank sediment downstream in the

5 <u>floodplain as highlighted by changes in the composition of suspended organic matter.</u>

6 The results of this study suggest that relationships between water flow and suspended
7 sediment load as well asnd hydrograph separation procedures at the outlet of catchments
8 wouldcan be better constrained using high-resolution monitoring of overland flow than direct
9 rainfall as shown in this study.

Finally, as higher suspended organic matter exports than in previous studies were determined, these results indicate that both the sampling period, at the onset of the rainy season, (following field clearing by slash and burn in this study), and the impact of land use change played a key role for explains the important sediment delivery observed at the outlet of the catchment for a medium magnitude flood.

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16 Acknowledgements

17 The authors would like to thank the Lao NAFRI (National Agriculture and Forestry Research 18 Institute in Vientiane) and the MSEC project (Multi-Scale Environment Changes) for their 19 support. They are also grateful to Keo Oudone Latsachack, Bounsamai Soulileuth and 20 Chanthamousone Thammahacksa for their help in the field, to Véronique Vaury (iEES-Paris) for organic matter composition measurements, and to Patricia Richard (iEES-Thiverval 21 Grignon) for δ^{18} O measurements of water samples. Elian Gourdin received a PhD fellowship 22 from Paris-Sud University, Orsay, France. This work received financial support from the 23 24 French CNRS EC2CO / BIOHEFECT program (Belcrue project).

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Table 1. Mean organic matter composition and ¹³⁷Cs activity (\pm 1 standard deviation) for surface soils (n=64), gullies (n=5) and stream bank (n=6) samples in the Houay Pano and Houay Xon catchments. For ¹³⁷Cs activity measurements, see Gourdin et al. (<u>under review2014</u>).

Location	TOC	TN	TOC/TN	$\delta^{13}C$	$\delta^{15}N$	¹³⁷ Cs
	$(mgC g^{-1})$	$(mgN g^{-1})$		(‰)	(‰)	$(Bq kg^{-1})$
Surface soils*	25 ± 5	2.1 ± 0.5	11.6 ± 2.0	-25.5 ± 1.4	6.7 ± 1.3	2.2 ± 0.9
Stream banks**	13 ± 6	1.1 ± 0.3	12.4 ± 7.7	-23.2 ± 4.4	8.6 ± 1.9	0.4 ± 0.3
Gullies**	14 ± 7	1.4 ± 0.6	9.6 ± 0.8	-22.7 ± 0.8	8.7 ± 2.1	0.4 ± 0.3

*Data from Huon et al. (2013) and this study (2012), **this study (2012).

Station	Drainage area	EWD*	SR**	RC***	
	(km²)	$(x \ 10^6 L)$	(mm)	(%)	
S 1	0.2	0.215	1.1	4.0	
S 4	0.6	1.88	3.2	11.7	
S10	11.6	12.2	1.1	3.9	

Table 2. Estimates of event water discharge (EWD) and related specific runoff (SR) and runoff coefficient (RC) for the three stations during the <u>May 2323 May 2012</u> flood.

* EWD = total water discharge minus baseflow discharge

** SR = EWD / drainage area

*** RC = 100 x (SR / rainfall) assuming an homogeneous cumulative rainfall of 27 mm

Table 3. Total suspended sediment yield (SSY), total particulate organic carbon yield (C_{SSY}), specific total suspended sediment yield (S_Y) and specific total organic carbon yield (C_Y) for the May 2323 May 2012 flood.

Station	SSY	C _{SSY}	S_Y^*	C _Y **
	(Mg)	(kg)	(kg ha ⁻¹)	(kgC ha ⁻¹)
S 1	2.3	58	115	2.9
S 4	26	496	433	8.3
S10	130	4346	112	3.7

* $S_y = 10 \times SSY / drainage area in Table 2$ ** $C_Y = 10^{-2} \times C_{SSY} / drainage area in Table 2$

Label	Time*	TSS*	Q*	EC*	$\delta^{18} O^*$	TOC*	TN*	TOC/TN*	$\delta^{13}C^*$	$\delta^{15}N^{\ast}$
	(hh:mm)	(g L ⁻¹)	(L s ⁻¹)	$(\mu S \text{ cm}^{-1})$	(‰ vs. V- SMOW)	$(mgC g^{-1})$	(mgN g ⁻¹)		(‰ vs. PDB)	(‰ vs. AIR)
Station S1										
LS0101	12:08	0.86	5	335	-7.2	42.0	2.0	20.5	-15.3	7.1
LS0102	12:09	0.56	7	317	-7.1	60.3	2.6	23.1	-19.0	5.5
LS0103	12:09	0.53	10	317	-7.0	-	-	-	-	-
LS0104	12:10	0.61	13	299	-6.8	66.2	2.1	31.0	-19.7	7.0
LS0105	12:10	-	16	299	-	-	-	-	-	-
LS0106	12:11	1.23	21	282	-6.9	32.2	2.3	14.0	-23.0	6.0
LS0107	12:13	1.70	27	262	-6.6	26.2	2.0	13.0	-23.7	6.6
LS0108	12:14	2.37	34	259	-6.2	25.0	2.0	12.7	-22.4	6.8
LS0109	12:19	3.65	40	241	-6.0	23.1	2.1	10.8	-24.4	7.2
LS0110	12:20	4.17	55	233	-6.1	23.4	2.2	10.7	-24.5	7.3
LS0111	12:21	4.65	76	224	-5.9	22.5	2.0	11.1	-24.1	7.5
LS0112	12:21	18.74	90	215	-5.8	27.4	2.0	11.1	-25.6	6.8
LS0113	12:30	29.98	68	184	-5.8	25.7	2.1	11.0	-25.8	6.8
LS0114	12:33	23.02	51	188	-5.5	25.8	2.2	10.7	-25.9	7.5
LS0115	12:37	24.05	38	194	-5.4	23.3	2.0	10.1	-25.8	7.5
LS0116	12:43	17.67	27	205	-5.6	20.8	2.5	9.8	-25.6	7.7
LS0117	12:50	16.38	18	218	-5.7	19.3	2.3	9.0	-25.3	7.8
LS0118	12:57	9.13	14	232	-5.8	18.7	2.4	9.0	-25.0	7.5
LS0119	12:58	14.37	13	233	-6.1	18.6	2.3	9.1	-25.1	7.2
LS0120	13:15	4.50	8	262	-6.2	19.3	2.1	9.6	-23.8	7.1
Station S4										
LS0403	11:57	1.53	15	297	-6.9	-	-	-	-	-
LS0404	11:58	1.21	24	306	-6.7	-	-	-	-	-
LS0403-4**	-	-	-	-	-	27.7	2.6	10.8	-23.8	7.1
LS0405	12:00	1.16	33	306	-6.5	29.9	2.9	10.4	-23.5	6.5
LS0406	12:01	2.71	42	262	-6.1	24.8	2.4	10.2	-24.4	7.2
LS0407	12:04	5.83	54	216	-5.5	22.6	2.2	10.5	-25.0	7.2
LS0408	12:05	6.83	76	205	-5.2	-	-	-	-	-
LS0409	12:06	7.25	114	198	-5.3	-	-	-	-	-
LS0408-9**	-	-	-	-	-	21.2	2.1	10.1	-25.0	7.5
LS0410	12:07	10.07	144	177	-4.7	22.1	2.1	10.7	-25.2	7.6
LS0411	12:07	11.89	185	161	-4.7	20.8	2.0	10.3	-25.2	7.6
LS0412	12:08	15.75	280	138	-4.5	19.2	1.9	9.9	-25.6	7.6
LS0413	12:09	20.05	309	121	-4.9	-	-	-	-	-
LS0414	12:10	31.56	358	99	-5.1	19.6	2.1	9.6	-25.4	8.0
LS0415	12:11	46.51	440	87	-5.2	21.1	2.1	10.0	-25.6	7.5

Appendix A: Table A1.	Summary of data for	stations S1, S4 and S1	0 during the May	23<u>23 May 2012</u> flood.
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Label	Time*	TSS*	Q*	EC*	$\delta^{18}O^{\ast}$	TOC*	TN*	TOC/TN*	$\delta^{13}C\ast$	$\delta^{15}N^{\ast}$
	(hh:mm)	(g L ⁻¹)	(L s ⁻¹)	$(\mu S \text{ cm}^{-1})$	(‰ vs. V- SMOW)	$(mgC g^{-1})$	(mgN g ⁻¹)		(‰ vs. PDB)	(‰ vs. AIR)
LS0416	12:20	28.40	335	103	-5.4	24.5	2.1	11.5	-26.0	7.1
LS0417	12:23	23.00	277	105	-5.4	24.7	2.2	11.4	-25.8	7.2
LS0418	12:26	17.76	228	117	-5.6	24.7	2.1	11.8	-26.0	7.4
LS0419	12:34	11.70	183	152	-5.6	-	-	-	-	-
LS0420	13:20	12.62	145	164	-5.6	-	-	-	-	-
LS0419-20**	-	-	-	-	-	22.5	2.0	11.0	-25.8	7.4
LS0421	13:32	7.71	112	192	-5.8	19.0	1.9	9.8	-25.6	7.7
LS0422	13:46	6.92	84	201	-6.1	19.6	2.0	9.8	-25.6	7.7
LS0423	14:05	6.93	59	203	-6.0	-	-	-	-	-
LS0424	14:43	5.89	39	214	-6.2	-	-	-	-	-
LS0425	15:46	3.37	23	230	-6.2	-	-	-	-	-
LS0423-25**	-	-	-	-	-	20.9	2.2	9.6	-25.5	7.3
Station S10										
LS1002	12:24	7.94	204	227	-5.5	-	-	-	-	-
LS1003	12:28	5.57	455	220	-5.5	-	-	-	-	-
LS1002-3**	-	-	-	-	-	39	2.0	19.8	-21.2	8.2
LS1004	12:31	8.77	623	215	-5.9	-	-	-	-	-
LS1005	13:03	11.10	943	167.5	-5.6	-	-	-	-	-
LS1004-5**	-	-	-	-	-	36	1.9	19.1	-22.6	7.4
LS1006	13:06	23.63	990	167	-5.7	44	1.9	23.1	-21.8	7.0
LS1007	13:27	17.02	1535	156	-5.4	44	2.0	22.2	-22.3	7.5
LS1008	13:33	24.43	1350	155.5	-5.7	29	1.8	16.2	-22.7	8.4
LS1009	13:39	24.00	1187	157	-5.6	31	1.8	17.2	-23.0	7.9
LS1010	13:46	15.74	1038	160	-5.6	25	1.8	13.7	-24.3	7.9
LS1011	13:55	21.47	886	167.5	-5.7	27	1.9	14.3	-23.4	7.3
LS1012	14:06	18.01	735	174	-5.6	29	1.9	15.0	-24.5	7.4
LS1013	14:20	15.35	597	184	-5.8	24	2.0	12.4	-24.5	7.1
LS1014	14:38	12.80	485	198	-5.7	-	-	-	-	-
LS1015	15:14	10.17	308	222	-5.9	-	-	-	-	-
LS1014-15**	-	-	-	-	-	30	1.9	15.5	-23.6	7.1

* = Time of collection, total suspended sediment load (TSS), stream discharge (Q), water electric conductivity (EC) and δ^{18} O, total organic carbon in TSS (TOC), total nitrogen in TSS (TN), δ^{13} C and δ^{15} N for TSS, ** = composite sample, - = no value.

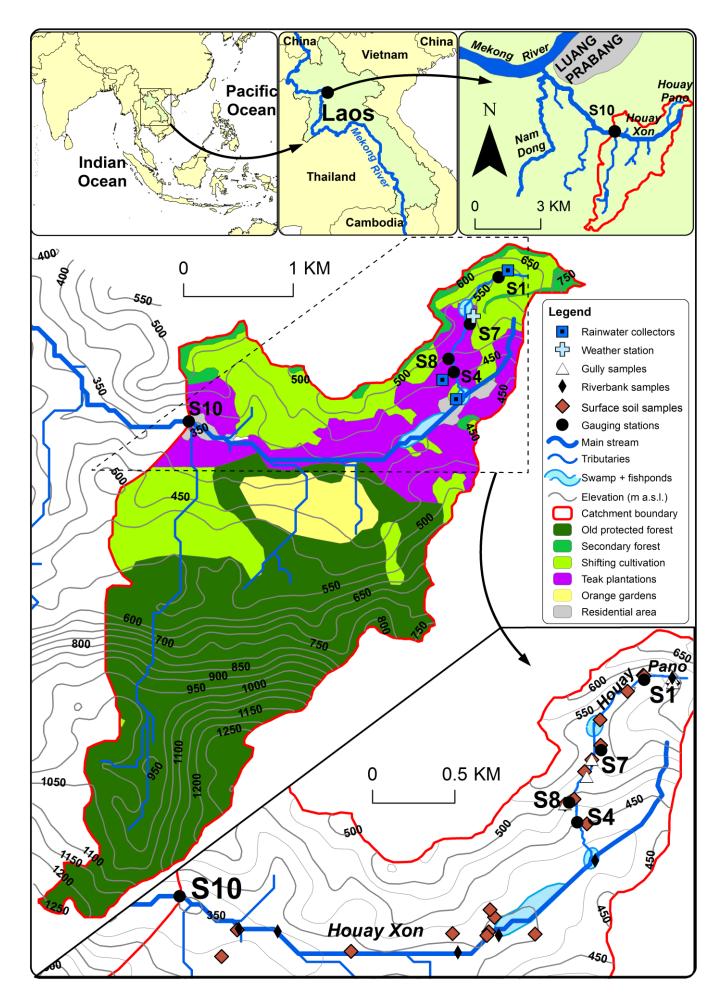


Fig. 1. Location of the Houay Xon River catchment in SE Asia (a). Topographic and land use map of the Houay Xon S10 sub-catchment in 2012 with location of the gauging stations (S1, S4, S7, S8, S10), rainwater collectors and automatic weather station (b), surface soil, gully and riverbank sampling locations (c).

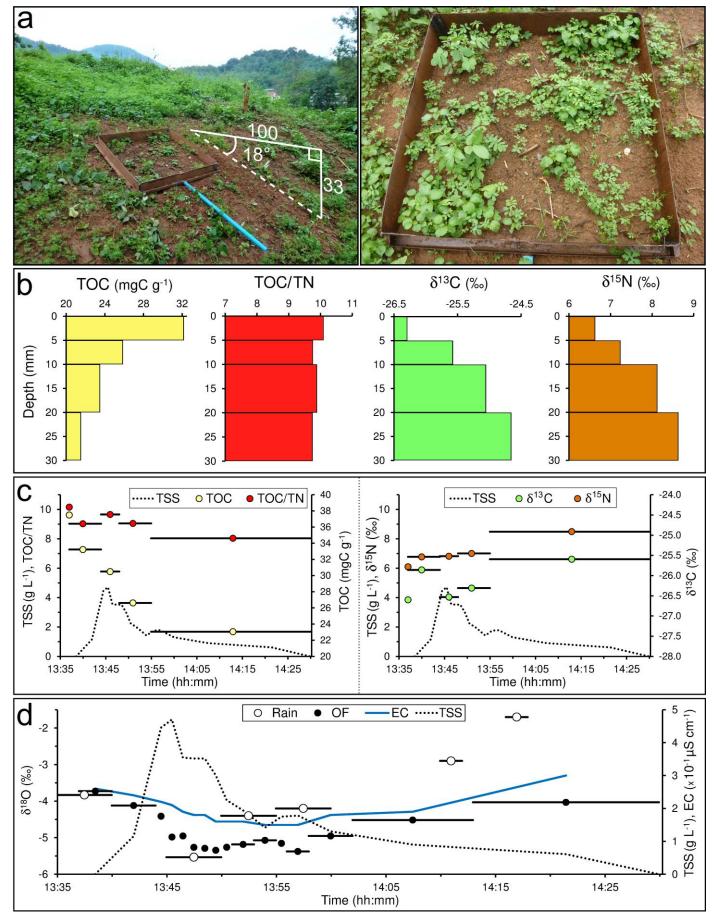


Fig. 2. Microplot experiment: (a) presentation of the 1-m² collecting system and its vegetation cover; (b) Distribution of topsoil total organic carbon (TOC) concentration, total organic carbon : total nitrogen ratio (TOC/TN), δ^{13} C and δ^{15} N with soil depth; (c) temporal evolution of the total suspended sediment load (TSS) plotted with TOC and TOC/TN in TSS (left) and with δ^{13} C and δ^{15} N in TSS (right) during the June 1st storm and (d) temporal evolution of the overland flow TSS load with rainwater- δ^{18} O (Rain), overland flow- δ^{18} O (OF) and overland flow electric conductivity (EC) during the June 1st storm.

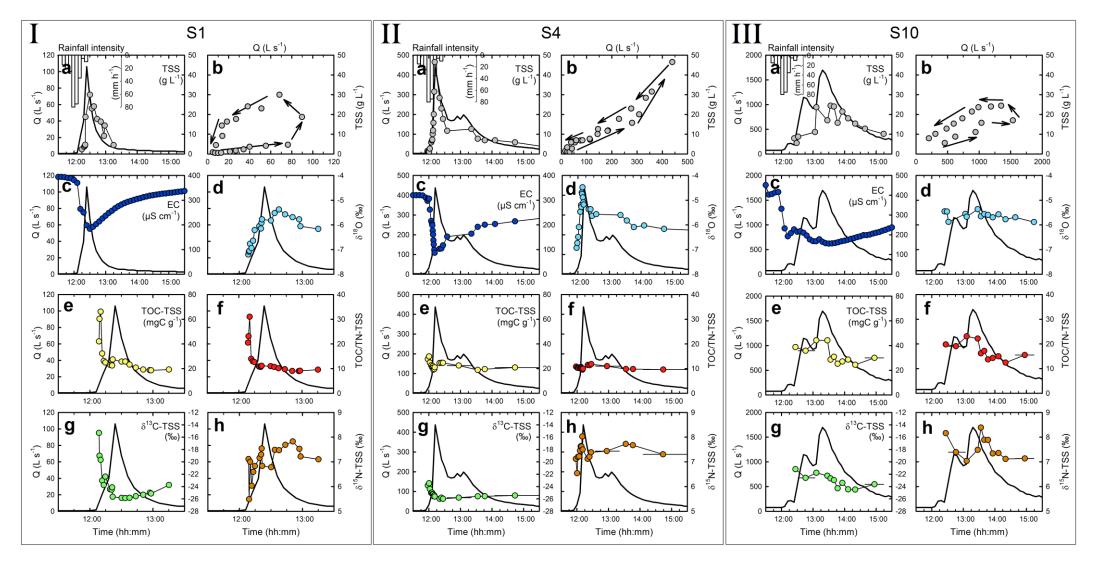


Fig. 3. Plots of the temporal evolution of (a) rainfall intensity, stream discharge (Q, thicker solid line), (b) total suspended sediment load (TSS), (c) water electric conductivity (EC), (d) streamwater- δ^{18} O, (e) total organic carbon concentration in the TSS (TOC-TSS), (f) total organic carbon : total nitrogen ratio in the TSS (TOC/TN-TSS), (g) δ^{13} C-TSS, (h) δ^{15} N-TSS for : (I) the upstream station S1, (II) the intermediate station S4, and (III) the downstream station S10, during the May 2323 May 2012 flood. Horizontal bars represent sampling period for composite samples.

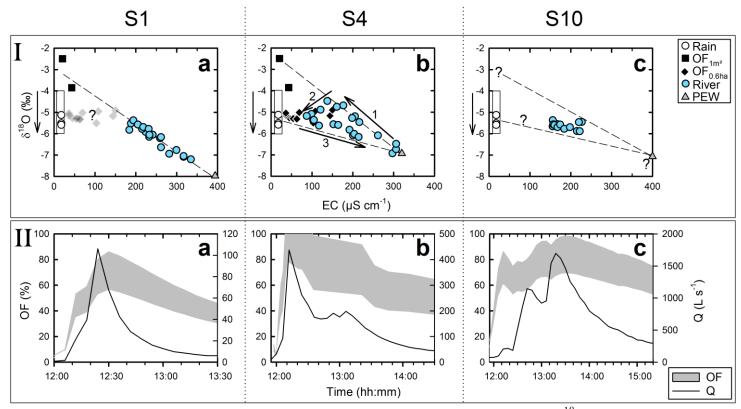


Fig. 4. Plots of: (I) relationships between water electric conductivity (EC) and water δ^{18} O, and (II) temporal evolution of stream water discharge (Q) with overland flow contribution estimates (OF) for (a) the upstream station S1, (b) the intermediate station S4, and (c) the downstream station S10, during the <u>May-2323 May</u> 2012 flood. In (I), open circles correspond to rainwater, filled squares to cumulative overland flow obtained with 1-m² plots (OF_{1m²}), filled diamonds to overland flow from S7 and S8 hillslopes (OF_{0.6ha}), filled colored circles to stream water, triangles to pre-event water (PEW). The rectangle areas and vertical arrows represent the potential temporal variability of rainwater- δ^{18} O during the storm. In (II), the shaded area corresponds to the variability range for the estimated overland flow contribution.

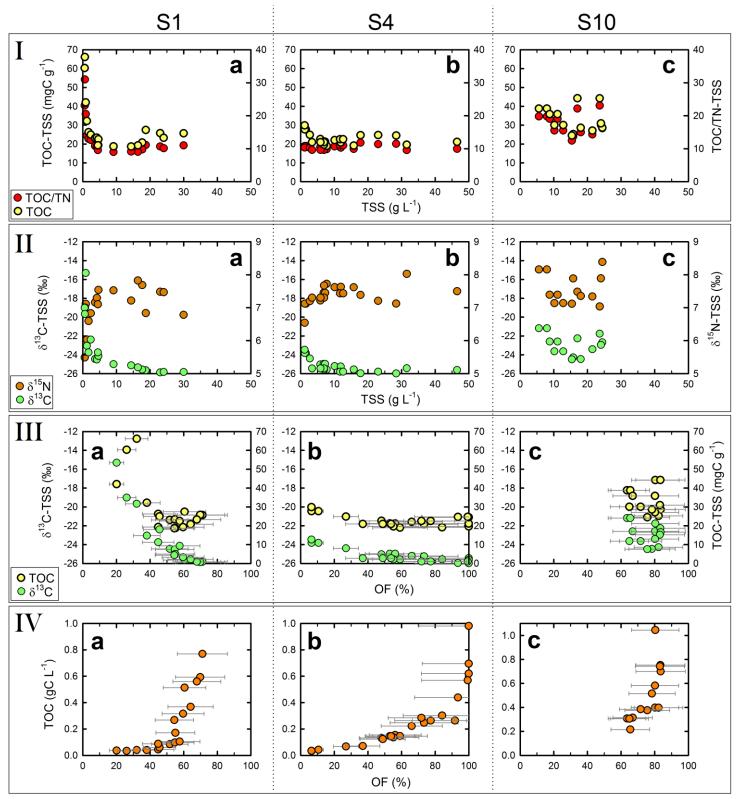


Fig. 5. Relationships between total suspended sediment load (TSS), total organic carbon concentration in the TSS (TOC-TSS), total organic carbon : total nitrogen ratio in the TSS (TOC/TN-TSS), δ^{13} C-TSS, δ^{15} N-TSS, total organic carbon load (TOC) and overland flow contribution estimates (OF): (a) at upstream station S1 (Houay Pano Stream), (b) at intermediate station S4, and (c) at downstream station S10, during the <u>May 2323</u> <u>May 2012</u> flood. In (III) and (IV), circles represent the median values of the variability range (horizontal bars) of estimated OF contribution.

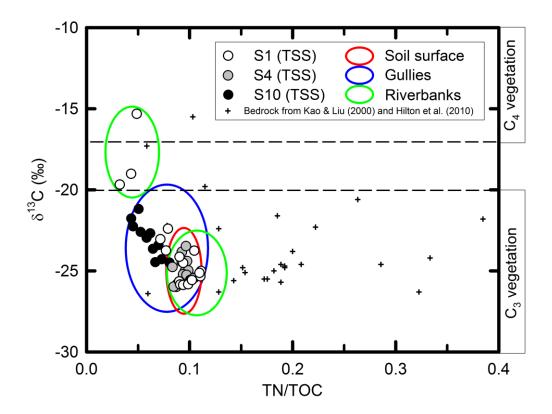


Fig. 6. Plot of δ^{13} C vs. TN/TOC for total suspended sediment loads (TSS) collected at S1 (open circles), S4 (grey circles) and S10 (closed circles) during the 23 May 2012 flood and for the potential sources of sediment (Soil surface: red area; Gullies: blue area; Riverbanks: green areas) determined in the catchment. Bedrock data (plus signs) are taken from literature (Kao and Liu, 2000; Hilton et al., 2010).