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Combining multiple fallout radionuclides (^{137}Cs , ^7Be , $^{210}\text{Pbxs}$) to investigate temporal sediment source dynamics in tropical, ephemeral riverine systems

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4 SEDIMENTS, SEC 3 • HILLSLOPE AND RIVER BASIN SEDIMENT DYNAMICS • RESEARCH ARTICLE
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8 **Combining multiple fallout radionuclides (^{137}Cs , ^7Be , $^{210}\text{Pb}_{\text{xs}}$) to investigate temporal sediment**
9 **source dynamics in tropical, ephemeral riverine systems**
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4 **Abstract**
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6 *Purpose:* Land use change and the associated acceleration of soil erosion have increased sediment supply to rivers
7 worldwide. To effectively manage sediment loads, it is important to understand sediment dynamics. Fallout
8 radionuclides (i.e. Be-7, excess Pb-210, Cs-137) separately provide spatial and temporal sediment source
9 information. However, their combined application has been limited by an assumption that subsurface sources do
10 not receive ⁷Be fallout. Here, we examine this assumption and investigate the utility of combining these
11 radionuclides to simultaneously model spatial and temporal sediment source dynamics.
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15 *Materials and methods:* Soil and subsurface material was sampled to characterize potential sediment sources in a
16 12 km² catchment in Northern Laos. This included material that was directly sampled from surface and subsurface
17 sources ($n = 65$) and also in-situ sediment source samples that were collected in ephemeral flow on hillslopes ($n =$
18 19). Suspended sediment ($n = 16$) was sampled at two monitoring stations. A distribution modelling approach
19 quantified the relative contributions of surface and subsurface sources, as well as the proportion of material
20 labelled with ⁷Be. The results from these two end-member models were compared to a four end-member model
21 examining contributions from recently eroded surface, recently eroded subsurface, re-suspended surface and re-
22 suspended subsurface sources.
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28 *Results and discussion:* Approximately 80 % of sediment was modelled to be supplied by sources labelled with ⁷Be.
29 Subsurface sources were modelled to contribute ~75 % of sediment. Accordingly, there was ~55 % more sediment
30 modelled from subsurface sources than expected if subsurface sources were sheltered from ⁷Be fallout.
31 Differences between subsoil and ⁷Be labelled source contributions modelled by the two and four end-member
32 models were, with one exception (0.3 %), within the range of model uncertainty, further supporting the existence
33 of re-suspended subsurface sources. At the upstream sampling location (S4), surface sources contributed the
34 majority of sediment (55 %) whereas subsurface sources dominated the supply of sediment downstream (S10 –
35 74 %). Importantly, re-suspended subsurface sources, labelled with ⁷Be, were a significant sediment source at the
36 catchment outlet (S10 – 60 %) during the investigated event.
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43 *Conclusions and perspectives:* This study demonstrates the utility of combining multiple radionuclides when
44 investigating spatial and temporal sediment source dynamics in tropical, ephemeral catchments. In the future, this
45 approach should be tested with larger source datasets during the entire wet season and in larger catchments. This
46 research furthers our understanding of sediment propagation in tropical catchments, contributing to the
47 implementation of efficient soil conservation measures to reduce the deleterious effects of suspended sediment
48 loads.
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55 **Keywords** Beryllium-7 • Caesium-137 • Fingerprinting • Laos • Lead-210 • Sediment tracing
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1 Introduction

An excessive supply of fine sediment is degrading freshwater riverine environments (Owens et al. 2005) and is playing a major role in the transfer of particle-bound contaminants, such as metals (Turner et al. 2008), radionuclides (Chartin et al. 2013), organic contaminants (Gateuille et al. 2014) and bacterial pathogens (Falbo et al. 2013). In tropical developing countries, these negative consequences are exacerbated when local populations directly use riverine water resources, potentially triggering infectious disease or intoxication by contaminants (Ribolzi et al. 2011). Knowledge of the relative contribution of different sources supplying sediment to riverine systems is a crucial prerequisite to implementing efficient best practices necessary to limit off-site impacts of excessive sediment delivery (Walling 2005; Belmont et al. 2011; Haddadchi et al. 2013; Koiter et al. 2013).

In agricultural catchments, sediment is generally provided by two main source types, i.e. topsoil from cropland/grassland and subsurface material from gullies or channel banks (Ben Slimane et al. 2013; Evrard et al. 2013). The relative contributions of both sources can be estimated by modelling source and sediment caesium-137 (^{137}Cs , $T_{1/2} = 30$ y) activities (Mabit et al. 2008). In tropical regions, ^{137}Cs fallout activity levels are exclusively associated with the atmospheric nuclear weapon testing that peaked in the 1960s. Currently, ^{137}Cs activity levels are low in tropical topsoil and virtually absent in subsurface material (Huon et al. 2013; Olley et al. 2013). Tracing sediment sources with ^{137}Cs provides quantitative information fundamental to catchment management by indicating whether soil conservation measures should be targeted on improving farming practices in cultivated land or on stabilizing gullies and channel banks (Collins et al. 2011).

Quantifying sediment sources with ^{137}Cs allows for the modelling of only two source end-members. Accordingly, the modelling and analyses of this single tracer does not incorporate chronological information about sediment sources. For example, sediment supplied to a watercourse during an event

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4 is either exported from the catchment or deposited and potentially re-suspended during subsequent
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6 events (Duvert et al. 2010). Other tracers are thus required to understand temporal sediment dynamics.
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8 Although they have been previously used for source discrimination (e.g., Wallbrink and Murray 1996;
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10 Wilkinson et al. 2009), the potential use of two shorter-lived radionuclides, beryllium-7 (${}^7\text{Be}$, $T_{1/2}= 53$ d)
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12 and excess lead-210 (${}^{210}\text{Pb}_{\text{xs}}$, $T_{1/2}= 22$ y) as chronometers of sediment transfer in rivers has attracted
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14 increasing attention (Bonniwell et al. 1999; Le Cloarec et al. 2007; Taylor et al. 2013). Both radionuclides
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16 are continuously supplied to the soil surface by rainfall. Once they contact the soil, they are strongly and
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18 almost irreversibly bound to soil particles (Taylor et al. 2012). One temporal application of these
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20 radionuclides generalizes the age of suspended sediment transiting the stream network by quantifying
21
22 the relative proportions of ‘new’ (${}^7\text{Be}$ -enriched) and ‘old’ (${}^7\text{Be}$ -depleted) sediment based on comparisons
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24 of the ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{xs}}$ ratio in both rainfall and suspended sediment (Matisoff et al. 2005; Evrard et al. 2010).
25
26 In a critique of this approach, Walling (2013) suggested that this application of sediment fingerprinting
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28 techniques to quantify temporal source information involved a number of assumptions which “greatly
29
30 oversimplify reality”. One significant challenge when incorporating ${}^7\text{Be}$ as a temporal tracer is its short
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32 half-life and associated rapid decay in sediment which must be addressed by rapid analyses. A second
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34 challenge identified was that recently mobilized sediment will be characterized by a ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{xs}}$ ratio
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36 similar to that of rainfall. Gourdin et al. (2014a) demonstrated that this limitation may be addressed by
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38 analyzing sediment collected in overland flow to characterize potential sources.
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40 A third critique was that the ${}^7\text{Be}$ activity of recently mobilized sediment is controlled by the source of
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42 sediment, particularly with subsurface material being theoretically sheltered from atmospheric fallout
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44 (e.g. ${}^7\text{Be}$) (Walling 2013). Although vertical subsurface soils eroded during a rainfall event may be
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46 sheltered from direct atmospheric labelling, subsurface sources eroded during previous events that
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48 remain stored as shallow sediment deposits near their origin or downstream within the stream network
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50 may indeed receive direct atmospheric fallout.
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4 To investigate this latter limitation, we postulate that, in a monsoonal climatic region, ^7Be from the
5 previous year's wet season had completely decayed and material stored within the stream network,
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7 from either subsurface or surface sources, are initially depleted in ^7Be . Contrary to the approach where
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9 all old sediments transiting the system are depleted in ^7Be , we hypothesize that there is an additional
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11 temporal class of shallow sediment deposits in ephemeral catchments, subsurface re-suspended
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13 sediment, that are enriched in ^7Be , owing to the location of their deposition in the stream network
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15 above the low-water level (e.g. on channel benches or floodplains).
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21 A distribution modelling approach is presented that quantifies the relative contributions of sediment
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23 from four different end-members (recently eroded surface, re-suspended surface, recently eroded
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25 subsurface, and re-suspended subsurface) during the first erosive flood of the monsoon in an upstream
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27 cultivated catchment of Northern Laos. Understanding these different source contributions is important
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29 to inform catchment management. Ultimately, the goal of this research is to further develop the
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31 application of multi-fallout radionuclide tracing that simultaneously quantifies spatial and temporal
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33 sediment source dynamics.
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40 **2 Materials and methods**

41 42 2.1 Study site

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44 The Houay Pano catchment is located 10 km south of Luang Prabang in northern Laos (Fig. 1). The
45
46 tropical monsoon climate of the region is characterized by the succession of dry and wet seasons with
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48 80 % of rainfall occurring from May to October (Ribolzi et al. 2008). Water and sediment fluxes have
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50 been monitored in the area since 1998 (Valentin et al. 2008). The main stem of the Houay Pano stream
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52 is equipped with two gauging stations, S1 and S4, draining 20 ha and 60 ha, respectively (Fig. 1). Two
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54 additional stations (S7 and S8) draining two hillslopes (0.6 ha each) connected to the main stream
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56 between S1 and S4 were also monitored. The Houay Pano stream flows into the Houay Xon River (22.4
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4 km² catchment) where it is continuously monitored at station S10, which drains an 11.6 km² catchment
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7 (Fig. 1).

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9 The geology of the Houay Pano catchment mainly consists of pelites, sandstones and greywackes.
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11 Limestone cliffs of Carboniferous to Permian ages are found in the uppermost, north-east boundary of
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13 the catchment. Soils consist of deep (>2 m) and moderately deep (>0.5 m) Alfisols, except along crests
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15 and ridges where Inceptisols can be found (Chaplot et al. 2009). The main land uses in the 11.6 km²
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17 catchment drained by the S10 monitoring station are forests (covering 56 % of the total surface area),
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19 teak plantations (15 %) and cropland (23 %). In the upstream catchment drained by S4 (0.6 km²),
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21 cropland (52 %) and teak plantations (36 %) dominate and are actively eroding (Gourdin et al. 2014a).
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23 Accordingly, there is some channel aggradation in river sections flowing through teak plantations. The
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25 Houay Pano and Houay Xon catchments do not have extensive gully erosion or badlands (Gourdin 2014).
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28 Periodic landslides providing potential 'horizontal' sources of subsurface materials may occur after
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30 heavy rainfall (mainly upstream of S4), whereas channel erosion is evident between S4 and S10 stations
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32 (Gourdin et al. 2014a). Although horizontal sediment sources have been acknowledged as a significant
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34 sediment source in Australian catchments (Hancock et al. 2014), this periodic landslide activity is not
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36 directly connected to the stream network and thus it is only considered to be an important sediment
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38 source during heavy rainfall events. Sediment storage occurs in marshes located upstream of S4 station
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40 and between S4 and S10 stations (Fig. 1). However, resuspension of sediment trapped in these marshes
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42 is unlikely during low-to-intermediate magnitude storms (Huon et al. 2013).
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51 2.2 Soil and sediment sampling

52 Composite surface (top 0–2 cm; $n = 32$) and subsurface samples (gullies $n = 11$, channel banks $n = 21$)
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54 were collected to characterize potential sources of sediment (Fig. 1) (Gourdin et al. 2014a). Each sample
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56 was composed of five subsamples collected within an area of 10 m² with surface sources sampled from a
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4 range of land uses (i.e. cropland, teak plantations). Channel bank samples were collected by scraping the
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6 material from the exposed vertical face (1-2 cm) of the bank. To comprehensively characterize source
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8 samples, in light of the critique of Walling (2013), suspended sediment, hereafter referred to as in-situ
9
10 sediment-source samples ($n = 11$), was collected during an ephemeral flow that occurred on hillslopes at
11
12 the S7 and S8 stations during the June 16, 2014 flood. This event was the first erosive flood of the
13
14 monsoon season. Lag deposit samples from ephemeral flows on hillslopes ($n = 7$) plus an additional
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16 sediment sample from the S8 station were included from a flood on May 23, 2012. All samples were
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18 oven dried (~48 h) and sieved to 1 mm prior to analyses to remove coarse grains and debris.
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23 As soil erosion and sediment transport processes often result in an enrichment of radionuclides in fine
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25 particle size classes, often sediment tracing and fingerprinting researchers isolate a particle size fraction
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27 (e.g. $<10 \mu\text{m}$ or $<63 \mu\text{m}$) or apply particle size corrections (Collins et al. 1997; Olley and Caitcheon 2000).
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29 Indeed, there has been a recent focus on the impacts of particle size on sediment tracing modelling
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31 results, particularly the non-linear responses and potential challenges of effective particle size
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33 corrections (Smith and Blake 2014).
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37 A third approach to addressing particle size selectivity may be derived from the tributary sediment
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39 tracing technique (Olley and Caitcheon 2000). For example, in tributary sediment tracing approaches,
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41 sediment is compared from upstream to downstream reaches, negating the major impacts of particle
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43 sorting on tracing results (Olley and Caitcheon 2000; Laceby et al. 2015). The approach taken in this
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45 current research combines both techniques through comparing in-situ sediment samples, taken from
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47 hillslope auto-samplers, to standard source samples to address potential particle size enrichment.
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49 When directly comparing sediment obtained with hillslope auto-samplers to standard source samples
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51 (Fig. 2), it becomes evident that there is no significant fallout radionuclide enrichment in the $<1 \text{ mm}$
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53 material eroded from hillslopes. Based on this direct comparison of hillslope and sediment samples,
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4 analyzing the <1 mm particle size fraction in this study likely incorporates particle sorting activities
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6 similarly to sieving the <63 μm or <10 μm fraction.
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9 Stream discharge was calculated from the continuous monitoring of water level and from rating curves
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11 built for each of the three stations for the June 16 flood. Eighteen suspended sediment samples were
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13 collected for this analysis during the June 16 flood at two stations (S4, $n = 10$; S10, $n = 7$). Samples were
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15 oven dried for ~ 48 h. For each sampling station, the first sample was omitted owing to insufficient
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17 material for analyses.
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23 2.3 Radionuclide analyses

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25 Radionuclide activities (^7Be , ^{137}Cs and ^{210}Pb) were measured at the Laboratoire des Sciences du Climat et
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27 de l'Environnement (Gif-sur-Yvette, France) following the methods detailed in Gourdin et al. (2014a). All
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29 results are expressed in Bq kg^{-1} with activities decay-corrected to the sampling date. Uncertainties on
30
31 radionuclides activities were ca. 10 % for $^{210}\text{Pb}_{\text{xs}}$, 20 % for ^7Be and up to 30 % for ^{137}Cs . All samples were
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33 measured within 50 days of sampling to ensure ^7Be was detectable.
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40 2.4 Distribution modelling

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42 A distribution modelling approach was used to incorporate distributions throughout the entire
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44 modelling framework, including the source contribution terms, i.e. not only source and in-stream
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46 components (Lacey and Olley 2015). To examine the potential of subsurface sources being labelled
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48 with ^7Be , distribution models were initially used to quantify the relative contributions from two end-
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50 members, first surface and subsurface sources (^{137}Cs), and second, sources with high and low ^7Be
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52 activities. Next, four end-members (i.e. recently eroded surface, recently eroded subsurface, re-
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54 suspended surface and re-suspended subsurface) were modelled to quantify the relative contributions
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56 of these individual sources, which were then compared to their relative global contributions derived
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4 from the two-end member model. A summary of radionuclide activities characterizing these four
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6 sediment types is provided in Table 1. In particular, re-suspended subsurface material corresponds to
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8 material (depleted in ^{137}Cs) stored in the alluvial plain above the water level, whereas re-suspended
9
10 surface material consists of sediment (originating from the soil surface and labelled with ^{137}Cs) that was
11
12 deposited on the riverbed below the water level prevailing at the onset of the wet season.
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16 Prior to modelling, the direct source samples were plotted with the in-situ sediment-source samples
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18 collected from the hillslopes. The latter were examined for their source potential within the four source
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20 quadrants and then re-classified based on their ^7Be and ^{137}Cs content. Mann-Whitney U-tests were used
21
22 to quantify the significance of source discrimination for all radionuclides in the two and four source end-
23
24 member models (Table 2).
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26

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28 For the two-source model, it is assumed that sediment samples from the S4 and S10 stations constitute
29
30 a discrete mixture of their sources, with the source contribution of A being x , and the contribution of B
31
32 being $1-x$:
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$$35 \quad Ax + B(1 - x) = C \quad (1)$$

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37
38 Where C is the in-stream sediment distribution; A and B are the two source distributions; and x is
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40 modelled as a truncated normal distribution ($0 \leq x \leq 1$) with a mixture mean (μ_m) and standard deviation
41
42 (σ_m) following Caitcheon et al. (2012). The model is solved by minimizing the median difference between
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44 the distributions of both sides of Eq. 1 (i.e. C and $Ax + B(1 - x)$) with the Optquest algorithm in Oracle's
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46 Crystal Ball software (2013) with a randomly generated mixture mean (μ_m) and standard deviation (σ_m)
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48 (more details provided below).
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53 The relative contribution of the four source end-members to in-stream sediment is determined by
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55 simultaneously minimizing the median mixing model difference (MMD):
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$$58 \quad MMD = \sum_{i=1}^n \left| \left(C_i - \left(\sum_{s=1}^m P_s S_{si} \right) \right) / C_i \right| \quad (2)$$

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4 where n is the number of radionuclides included in the model; C_i is the normal distribution of
5 radionuclide i from the in-stream sediment; m represents the number of sources; P_s is the contribution
6 distribution from source (s); S_{si} is the normal distribution of radionuclide i in source s . P_s is solved as a
7 truncated normal distribution ($0 \leq x \leq 1$) derived with a randomly generated mixture mean (μ_m) and
8 standard deviation (σ_m) with the absolute contribution of all proportional contributions (P_s) summing to
9 1. Absolute values were used in Eq. 2 rather than summing the squares as Laceby and Olley (2015)
10 demonstrated this approach produced slightly improved model accuracy.

11
12 In-stream sediment (C or C_i) samples, from the two monitoring stations (S4, S10), were first modelled as
13 two distinct in-stream sample groups. The mean and standard deviations of these sample groups were
14 used to derive normal distributions of sediment sampled at S4 and S10. Thereafter, each sediment
15 sample from these monitoring stations was individually modelled similarly to Wilkinson et al. (2015). To
16 derive normal distributions for these individual samples, their analytical uncertainty was substituted for
17 the standard deviation centered on the sediment radionuclide activity mean. This allows for
18 distributions to be modelled throughout the modelling framework, even for individual in-stream
19 sediment samples, in order to examine the spatial and temporal variability in sources delivering
20 sediment in this catchment. This also allows for a direct comparison between model results from sample
21 groupings and individual sediment samples. The statistical dependence or covariance between
22 radionuclides in each source was directly incorporated into the mixing model following the approach of
23 Laceby and Olley (2015).

24
25 The Optquest algorithm in Oracle's Crystal Ball software (2013) modelled the radionuclide distributions
26 throughout a Monte Carlo style modelling framework. Non-negative constraints were applied to all
27 radionuclides. The μ_m and σ_m for each source's contribution distribution (x or P_s) were repeatedly varied
28 in the Optquest algorithm to minimize the median MMD in Eq. 2, or the distribution difference in Eq. 1,
29 when simultaneously solving these equations 2500 times with 2500 random samples selected with a
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4 Latin Hypercube technique (500 bins) drawn from each in-stream (C or C_i) and source (A , B , or S_{si})
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6 distribution. This model simulation and solving process, that determined one optimal source
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8 contribution distribution (x or P_s), was then repeated 2500 times with the median proportional source
9
10 contribution (x or P_s), from these 2500 additional simulations, reported as the contribution from each
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12 source.
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16 Model uncertainty around the proportional contribution for each source was determined by summing
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18 the modelled standard deviation of the mixture mean (σ_m), plus the median absolute deviation (MAD) of
19
20 this modelled standard deviation for the additional 2500 model simulations, plus the MAD of the
21
22 individual sources median proportional contribution, again for the additional 2500 simulations (Lacey
23
24 et al. 2015). This approach to reporting model uncertainty sums the actual standard deviation modelled
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26 for each source's contribution distribution along with the MAD of this standard deviation and the MAD
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28 of the actual source contribution distribution for the 2500 additional simulations.
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33 Modelling sediment results individually for each sediment sample provides only the relative
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35 contributions for different sources in relation to the particular volume of sediment sampled at an
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37 individual temporal period. The modelling results were event-weighted to incorporate differences in
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39 suspended sediment concentrations throughout the hydrograph. The event-weighting was calculated by
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41 multiplying river water discharge with suspended sediment concentrations to quantify instantaneous
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43 sediment fluxes corresponding to individual stage samples.
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49 **3 Results**

50 51 3.1 Source and target material analysis

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53 When comparing direct source samples (i.e. surface/subsurface) to the in-situ sediment-source samples,
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55 it is quickly evident that the latter have elevated ^7Be activities (Fig. 3). In fact, these in-situ sediment-
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57 source samples plot across a range of ^7Be concentrations and surface/subsurface source types. Based on
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4 previous research conducted in this catchment (Huon et al. 2013; Gourdin et al. 2014a), a threshold ^{137}Cs
5 activity of 1 Bq kg^{-1} was only used to discriminate surface and subsurface sources for the in-situ
6 sediment source samples. Furthermore, a threshold of $\sim 16 \text{ Bq kg}^{-1}$, deduced from residual activities that
7 may remain from early non-erosive storms, evidently separates source samples with high and low ^7Be
8 activities. These thresholds are illustrated with dashed lines in Fig. 3. The wide range of ^7Be values is
9 likely due to spatial variability in delivery and adsorption of ^7Be (Gourdin et al. 2014b). Scavenging of ^7Be
10 by particles in the water column is estimated to be negligible in the well-mixed shallow sheetflow
11 occurring in this catchment after rainfall, and recently labelled sediment was shown to display a similar
12 $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$ activity ratio as rainfall (Gourdin et al. 2014a).

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There were six in-situ sediment-source samples below the ^7Be threshold (all $< 5.6 \text{ Bq kg}^{-1}$) compared to
14 samples above. For ^{137}Cs , six samples were again below the activity threshold, three with high ^7Be
and three with low ^7Be . The discrimination between these samples is also evident for $^{210}\text{Pb}_{\text{xs}}$, although to
a lesser extent; likely the result of local soil type heterogeneities. The range of the in-situ sediment
source samples, including high and low ^7Be samples, indicates that this approach to characterizing
sources incorporates the direct in-stream labelling of sediment with ^7Be and the spatial or temporal
heterogeneity of sediment sources.

Four sources were established that combine the direct source samples and in-situ sediment-source
samples. Recently eroded surface sources are comprised of in-situ sediment source samples with ^{137}Cs
activities $> 1 \text{ Bq kg}^{-1}$ and ^7Be activities $> 16 \text{ Bq kg}^{-1}$. Recently eroded subsurface sources include the
directly sampled subsoil samples and the in-situ sediment source samples with ^{137}Cs activities $< 1 \text{ Bq kg}^{-1}$
and ^7Be activities $< 16 \text{ Bq kg}^{-1}$. Re-suspended surface sources include the direct surface source and the in-
situ sediment source samples with ^{137}Cs activities $> 1 \text{ Bq kg}^{-1}$ and ^7Be activities $< 16 \text{ Bq kg}^{-1}$. This source
depicts previously eroded surface material that was deposited below the dry-season low-water level and
is remobilized during subsequent events. Re-suspended subsurface sources, comprised of in-situ

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4 sediment source samples with ^{137}Cs activities $<1 \text{ Bq kg}^{-1}$ and ^7Be activities $>16 \text{ Bq kg}^{-1}$, depict recently
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6 eroded subsurface material that was deposited and stored in the drainage network above the dry-
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8 season low-water level.
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11 Although there are clear differences between source radionuclide activities, is it important to determine
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13 whether the discrimination between source combinations is significant. Out of 18 potential source
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15 combinations, only five (or 28 %) did not provide significant discrimination (Table 2). As expected, ^{137}Cs
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17 did not discriminate between the two surface sources, or between the two subsurface sources. ^7Be did
18
19 not discriminate between recently eroded surface and re-suspended subsurface sources, both with
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21 elevated ^7Be activities. Interestingly, $^{210}\text{Pb}_{\text{xs}}$ did not discriminate between re-suspended subsurface and
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23 re-suspended surface sources, or between re-suspended subsurface and recently eroded subsurface
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25 sources.
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30 As $^{210}\text{Pb}_{\text{xs}}$ behaves similarly to ^7Be , albeit with a longer half-life, it may indeed trace soils differently and
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32 this complex behavior warrants future investigation. Nonetheless, for each potential source
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34 combination, at least two of the fallout radionuclides provided significant discrimination, with the
35
36 exception of re-suspended subsurface and recently eroded surface sources where only ^7Be
37
38 discriminated. Accordingly, all fallout radionuclides were incorporated into the distribution model. The
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40 modelled within source radionuclide correlations are presented in Table S1 in the Electronic
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42 Supplementary Material and all data used in these analyses are included in Table S2 (Electronic
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44 Supplementary Material).
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51 3.2 Evolution of radionuclide activities in sediment during the flood

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53 Following a prolonged dry period, the first flood event of the 2014 monsoon season was characterized
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55 by a moderate intensity, with up to 27 mm rainfall (at the S10 station) in 43 minutes. This intense rainfall
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57 event did not have driving rain and windy conditions that could potentially label vertical subsurface
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4 channel bank sources. The recurrence interval of this storm was <1 year. The shape of the hydrograph
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6 was similar at S4 and S10 stations, with the peak sediment discharge occurring just before the peak of
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8 water discharge (Fig. 4). Peak discharge was more than 10-fold higher at S10 (543 L s^{-1}) compared to S4
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10 (33 L s^{-1}). S4 drains a subcatchment of 0.6 km^2 , whereas the surface area drained by S10 is 11.6 km^2
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12 (ratio of 19). As the corresponding ratio between peak discharges recorded at both stations is 16, it is
13
14 probable that the entire catchment experienced a relative similar amount of precipitation.
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18 The maximum sediment concentration reached 2.1 g L^{-1} at S10 compared to 1.2 g L^{-1} at S4. The ^{137}Cs
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20 activities in suspended sediment were very low at S4 at the beginning of the flood, but they rapidly
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22 exceeded 1 Bq kg^{-1} , ranging from $1.0\text{--}1.6 \text{ Bq kg}^{-1}$ during the rising stage of the storm and maintained
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24 these activities after peak discharge (Fig. 4). This elevated peak in ^{137}Cs activities indicates an increase in
25
26 the surface source contributions during the event. Although ^{137}Cs activities varied in suspended material
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28 at S10, the activities were below those recorded at S4 and near or below 1 Bq kg^{-1} , indicative of elevated
29
30 subsurface source contributions.
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35 At S4, ^7Be activities varied through the hydrograph, with the highest activities ($40\text{--}70 \text{ Bq kg}^{-1}$) sampled in
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37 rising and falling stages, contrasting lower activities during peak discharge ($\sim 35 \text{ Bq kg}^{-1}$; Fig. 4). In
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39 comparison, at S10, ^7Be activities in suspended sediment remained moderate and stable throughout the
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41 flood ($37\text{--}55 \text{ Bq kg}^{-1}$). Similarly to ^{137}Cs , more variance was observed at S4 and S10. This trend was also
42
43 evident with $^{210}\text{Pb}_{\text{xs}}$. At S4, $^{210}\text{Pb}_{\text{xs}}$ varied between 10 and 64 Bq kg^{-1} compared to a range of 33 and 48 Bq
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45 kg^{-1} at S10 (Fig. 4).
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49 During both events, radionuclide activities measured in suspended sediment samples remained within
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51 the range of values measured in source material (Fig. 3). This finding supports the absence of a
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53 significant grain size effect between source and target material during this flood. Further, these plots
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55 indicate that most suspended sediment was likely supplied by sources with significant ^7Be labelling,
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57 including surface and subsurface sources.
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3.3 Source contribution analyses

There was significant discrimination between subsurface and surface sources with ^{137}Cs (Mann-Whitney $p < 0.001$) and between source samples with elevated and low ^7Be activities (Mann-Whitney $p < 0.001$).

The two-end member mixing models indicate that the majority of sediments are derived from subsurface materials with elevated ^7Be activities. At the upstream sampling station (S4) 59 % \pm 8 % of sediment were modelled to be derived from subsurface sources. This subsoil contribution increased downstream to 75 % \pm 7 % at S10. The mean subsurface contribution from all individually modelled samples at S4 was 56 % (σ 26 %) with results varying from 22 % (\pm 8 %) to 95 % (\pm 6 %) during different stages. At S10, the mean subsurface contribution from the individually monitored stations was 66 % (σ 23 %) with contributions ranging from 56 % (\pm 6 %) to 99 % (\pm 6 %). Source contributions and uncertainties from all distribution models are listed in Table 2.

Although there was an increase in subsurface contributions at the downstream site, there was a decrease in contributions from sources with elevated ^7Be activities. At the upstream site (S4), 85 % (\pm 6 %) of sediment was modelled to be derived from sources with elevated ^7Be concentrations compared to 79 % (\pm 5 %) at S10. When sediment from S4 were individually modelled, the mean contribution was 78 % (σ 17 %) ranging from 55 % (\pm 7 %) to 100 % (\pm 6 %). At S10, the mean contribution from individual stage samples was 80 % (σ 11 %) ranging from 72 % (\pm 6 %) to 100 % (\pm 6 %).

If subsurface sources were sheltered from ^7Be fallout and only surface sources were labelled, ~80 % of sediment would be expected to be supplied by surface sources. As ~75 % of sediment is supplied by subsurface sources, there is ~55 % more sediment derived from subsurface sources than expected, indicating the potential presence of a re-suspended subsurface source in this catchment that is labelled with ^7Be fallout.

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4 One approach to further investigating the presence of this re-suspended subsurface material is to
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6 compare the contributions from the two-end member model to the summed, global contributions from
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8 the four-end member model. The global subsurface contributions from the four-end member model
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10 indicate that 59 % of the sediment are derived from subsurface sources at S10 and 76 % at S4, differing
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12 by only 0.2 % and 1.7 % from the two-end member model. For the individual stage samples, the mean
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14 difference between models increased to 4.5 % (σ 0.6 %) at S4, and 9.5 % (σ 4.1 %) at S10. For ^7Be
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16 labelled sources, the models differed by 8.6 % at S4 and 3.1 % at S10 for the stage sample groupings. For
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18 the individual stage samples, the models differed by a mean of 5.1 % (σ 5.0 %) at S4 and 10.0 % (σ 9.2 %)
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20 at S10. Aside from the minor exception (0.3 %) for the individual models at S10, the mean differences
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22 were less than the mean uncertainty for all models (9.7 %). The fact that the subsurface and ^7Be labelled
23
24 source contributions do not differ greater than the mean uncertainty for these models further supports
25
26 the likelihood of re-suspended subsurface derived material transiting the Houay Pano catchment.
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32 The difference between modelling all sediment samples as a grouped distribution for each station
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34 compared to the mean from modelling individual stage samples at S4 (5.5 %) and S10 (8.6 %) was below
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36 the mean model uncertainty (9.9 %). This validates the use of analytical error as a substitute for a
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38 standard deviation to model distributions for individual sediment samples (Wilkinson et al. 2005) and
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40 allows for examining the weighted contributions of samples collected during different stages of the
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42 hydrograph.
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46 47 48 49 3.4 Evolution of weighted-source contributions during the flood

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51 To thoroughly understand the sediment contributions from each source, detailed evolutions of source
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53 contributions to sediment flux were reconstructed for each stage sample at the S4 and S10 stations (Fig.
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55 5). Source contributions from the sediment-weighted modelling results are listed in Table 3. At S4, the
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57 first two stage samples were comprised of sediment primarily originating from re-suspended subsurface
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4 sources (>90 %). Thereafter, recently eroded surface sources contributed ~50 % of the sediment for the
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6 remainder of the stage samples. Based on the event weighting, recently eroded surface sources
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8 contributed 46 % of the total sediment load, compared to 24 % for re-suspended subsurface sources, 20 %
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10 for recently eroded subsurface sources, and 8 % for re-suspended surface sources. The weighting of the
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12 source contributions reduced the re-suspended subsurface contribution by 19 % with an increase of 10 %
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14 for the recently eroded surface sources, 6 % for the recently eroded subsurface sources, and 3 % for the
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16 re-suspended surface sources. When determining source contributions with sediment from rising stage
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18 samplers, it may indeed be important to similarly report weighted values.
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23 At the downstream S10 station, the relative contributions of sources were less variable, with a dominant
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25 supply of sediment originating from subsurface material (49-100 %) for the entire flood. Most of this
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27 subsurface material (μ 77 %, σ 17 %) consisted of re-suspended particles labelled with ^7Be . In total, the
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29 re-suspended subsurface sources contributed 60 % to the material sampled throughout the hydrograph
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31 compared to 14 % from the recently eroded subsurface sources. Less than 25 % of sediment was
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33 modelled to be derived from recently eroded surface (16 %) and re-suspended surface (9 %) sources,
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35 with the main contributions from these sources occurring in the rising and falling stages of the
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37 hydrograph. Although the sources of sediment were variable through time at S10, their relative
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39 weighted contributions were similar to the non-weighted contributions with a mean difference of only
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44 2.5 % (μ 1.8 %).
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49 **4 Discussion**

50 51 4.1 Spatial-temporal tracing

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53 Although Walling (2013) indicated that subsurface material should be sheltered from atmospheric ^7Be
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55 fallout, in the Houay Pano catchment there is a significant supply of subsurface material depleted in
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57 ^{137}Cs but labelled with ^7Be transiting the river. Most likely, at the downstream station (S10), this
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4 sediment was originally supplied to the river by subsurface sources and has remained stored in the river
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6 channel, above the water level, where it was exposed to ^7Be fallout. This hypothesis was supported by
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8 analyzing material that was collected in the channel before the flood. Radionuclide activities measured
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10 in this sediment confirmed its subsurface origin ($0.4 \pm 0.2 \text{ Bq kg}^{-1}$ of ^{137}Cs) and showed it was depleted in
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12 ^7Be ($< 0.2 \text{ Bq kg}^{-1}$) and $^{210}\text{Pb}_{\text{xs}}$ ($4.9 \pm 4.5 \text{ Bq kg}^{-1}$). This possibility is supported by the lack of horizontal
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14 subsurface sources that could be labelled directly with ^7Be between S4 and S10 or significant driving
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16 wind during this event that could label vertical exposed subsoil. In contrast, at S4, sediment depleted in
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18 ^{137}Cs and labelled with ^7Be may also originate from a nearby landslide exposed to rainfall. However, as
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20 the storm was of low intensity, it is unlikely that extensive erosion took place in this area during the
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22 event. Indeed, sediment deposited at the foot of the landslide and exposed to rainfall during the event
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24 may have been remobilized during the flood.
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30 The similarity of model results from the two and four end-member models supports the existence of this
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32 re-suspended subsurface material, validating the potential of combining multiple radionuclides to model
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34 spatial-temporal sources (i.e. recently eroded surface, recently eroded subsurface, re-suspended
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36 surface and re-suspended subsurface). Accordingly, ^7Be may be an effective spatial-temporal tracer
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38 when used in combination with ^{137}Cs and $^{210}\text{Pb}_{\text{xs}}$ in similar tropical ephemeral environments.
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42 These results demonstrate that the type and approach to sediment discrimination must be adapted to
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44 the environmental context of the research. For instance, in catchments of Eastern Australia where
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46 subsurface sediment source contributions largely dominate in river sediment (Olley et al. 2013), ^{137}Cs ,
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48 ^7Be and $^{210}\text{Pb}_{\text{xs}}$ activities were shown to discriminate between, on the one hand, ‘horizontally aligned’
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50 subsurface erosion sources where rilling and intense sheet erosion or ‘scalds’ had exposed B-horizon
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52 soils and, on the other hand, ‘vertical’ erosion sources (i.e. channel banks and gully walls) (Hancock et al.
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54 2014). In contrast, in a semi-arid catchment of Northern Mongolia, the same combination of
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56 radionuclides was used to discriminate between cultivated topsoil and channel bank sources (Theuring
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4 et al. 2013). In both cases, the tracer set including ^7Be was used to calculate the contribution of
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6 sediment sources and not to investigate their transfer times. In Australia for instance, it is argued that
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8 the travel time of suspended sediment in the river during floods is short compared to the half-life of ^7Be
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10 and that fine sediment storage in the channel is negligible, justifying why ^7Be can be used to characterize
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12 the source of riverine material (Hancock et al. 2014). Although this may be potentially the case in
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14 Australia with $<10\mu\text{m}$ sediment, future research using ^7Be to characterize sediment sources must be
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16 cognizant of the potential labelling of material transiting the stream network, particularly where old
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18 material is deposited and temporarily stored above the low-water level. Other approaches with
19
20 different combinations of radionuclides (including long-lived ^{10}Be and $^{239+240}\text{Pu}$) may be beneficial to
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22 further discriminate the specific contribution of material remobilized from floodplains to sediment
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24 transiting the rivers (Smith et al. 2012; Belmont et al. 2014). In particular, other sediment properties are
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26 required to effectively discriminate between re-suspended subsurface and horizontal subsurface
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28 sediment sources.
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35 In Laos, the specific context observed at the beginning of the wet season must be taken into account to
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37 avoid the misinterpretation of the results that may arise from the examination of fresh sediment
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39 percentages or the calculation of their age following the method proposed by Matisoff et al. (2005).
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41 According to this approach, sediment collected before the flood at S4-2 would be 73 % fresh with 25
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43 days of residence time in the river. Contrarily, our results illustrate a different situation at the beginning
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45 of the monsoon, with the dominance of re-suspended subsurface material, exposed to ^7Be fallout within
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47 the stream network (S4 - 100 %). This material is likely preferentially transported due to its location in
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49 the channel network, immediate availability, and high connectivity.
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56 4.2 Management implications and perspectives 57 58 59 60 61 62 63 64 65

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4 Most studies applying the ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{xs}}$ ratio strictly investigate sediment transit times, and not their
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6 sources (Bonniwell et al. 1999; Matisoff et al. 2005; Evrard et al. 2010). Studies addressing both
7
8 sediment dynamics and source apportionment started in recent years (Huisman et al. 2013; Gourdin et
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10 al. 2014a). However, these authors considered temporal and spatial components separately. The results
11
12 of this current research indicate that there is a potential to combine these techniques into a single
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14 approach to derive important management information.
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18 At the upstream station (S4), re-suspended subsurface material that was likely available for transport on
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20 the riverbed, was first re-mobilized during the flood event's rising stage. The relative contribution of re-
21
22 suspended subsurface sources to river sediment decreased during the flood, with the arrival of surface
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24 material originating likely from cropland located on hillslopes connected to the stream during peak flow
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26 and flood recession stage. In the upper area of the catchment, the surface source contributions (both
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28 recently eroded and re-suspended) are clearly evident during the hydrograph peak suggesting in smaller
29
30 catchments that management should target well-connected surface sources. Importantly at S4, there
31
32 were differences between the weighted and non-weighted sediment source contributions indicating the
33
34 importance of weighting sediment tracing results when modelling sediment obtained similarly with
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36 auto-samplers.
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40 At the catchment outlet (S10 station), most sediment was derived from re-suspended subsurface
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42 sources, likely corresponding to material originating from channel bank erosion that was deposited on
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44 the riverbed or floodplains when the water level decreased following the previous wet season. During
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46 this first erosive event of the monsoon, this subsurface material was then exposed to ${}^7\text{Be}$ atmospheric
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48 fallout before the increase in water level (Fig. 6). The remainder of the sediment was derived from
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50 recently eroded subsurface (15 %), recently eroded surface (16 %), and re-suspended surface sources
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52 (9 %).
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4 When managing larger catchment areas, subsurface sources likely become increasingly significant and
5
6 require direct attention. More importantly, the prevalence of readily erodible alluvial material near the
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8 channel is important to consider at larger catchment scales. This research highlights the importance of
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10 understanding potential scale effects when translating sediment source contributions into potential
11
12 management practices. In upper parts of the catchment, where surface sources are the main supply of
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14 sediment to the rivers, priority should be given managing hillslopes sediment sources. For instance,
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16 farmers should be encouraged to stop burning the understorey vegetation in teak plantations to
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18 maintain soil cover. In contrast, in downstream river reaches, management should prioritize the
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20 stabilization of channel banks.
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25 One of the challenges with the multi-tracer approach to quantifying spatial-temporal sediment
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27 dynamics is effectively sampling re-suspended subsurface sources. New sampling approaches are likely
28
29 required to obtain sufficient samples of this source material. The evidence of re-suspended subsurface
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31 material that is labelled with ^7Be on inset channel benches and possibly floodplains likely indicates there
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33 is a similarly deposited surface material. Furthermore, there is likely a subsurface material that is
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35 deposited below the low-water level that does not receive ^7Be labelling. In other catchments with
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37 significant gully erosion, badlands and extensive subsurface erosion, there is the likelihood of horizontal
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39 sediment sources. Future research should examine whether these sources exist, are significant, and
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41 whether other sediment properties, such as potentially nitrogen stable isotopes, may provide further
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43 discrimination. Increasing the sample size of all sources, so their distributions may be succinctly
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45 modelled (e.g. log-normal, gamma) would provide more certainty to the source apportionments. An
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47 expanded analyses of the applicability of in-situ source sampling would further validate the approach of
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49 using in-situ source samples to address particle size selectivity in sediment tracing research.
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59 **5 Conclusions**

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4 In this study, we proposed a unique modelling approach to quantify the respective proportions of
5 recently eroded surface, recently eroded subsurface, re-suspended surface, and re-suspended
6 subsurface material during the first erosive flood of the monsoon season in a nested catchment of Laos.
7
8 Variations of these contributions during a succession of events throughout the wet season should be
9 investigated in future. Understanding these different source contributions is important to guide
10 catchment management by indicating whether management should target surface (e.g. cultivated soils –
11 i.e. at S4) or subsurface (e.g. channel bank) sources, or material stored within the channel that may
12 contain sediment-sorbed pathogens and contaminants (i.e. at S10).
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23 This research suggests that the use of a mixing model to quantify the relative contributions of sediment
24 provided by these sources avoids the limitations identified in previous studies regarding the use of ^7Be
25 as a tracer of recently eroded sediment. This is an important development in the application of multi-
26 fallout radionuclide tracing approaches that simultaneously quantifies spatial and temporal sediment
27 dynamics. Among the fallout radionuclides, ^7Be is a unique tracer of recent erosion/sedimentation,
28 although $^{210}\text{Pb}_{\text{xs}}$ with its longer half-life likely warrants more in-depth examination.
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Fig. 1 Study catchment location in Southeast Asia along with land use, elevation, in-stream monitoring stations (S1, S4 and S10), direct source sample locations, and hillslope in-situ sediment-source sampling stations (S7 and S8) (Gourdin et al. 2014)

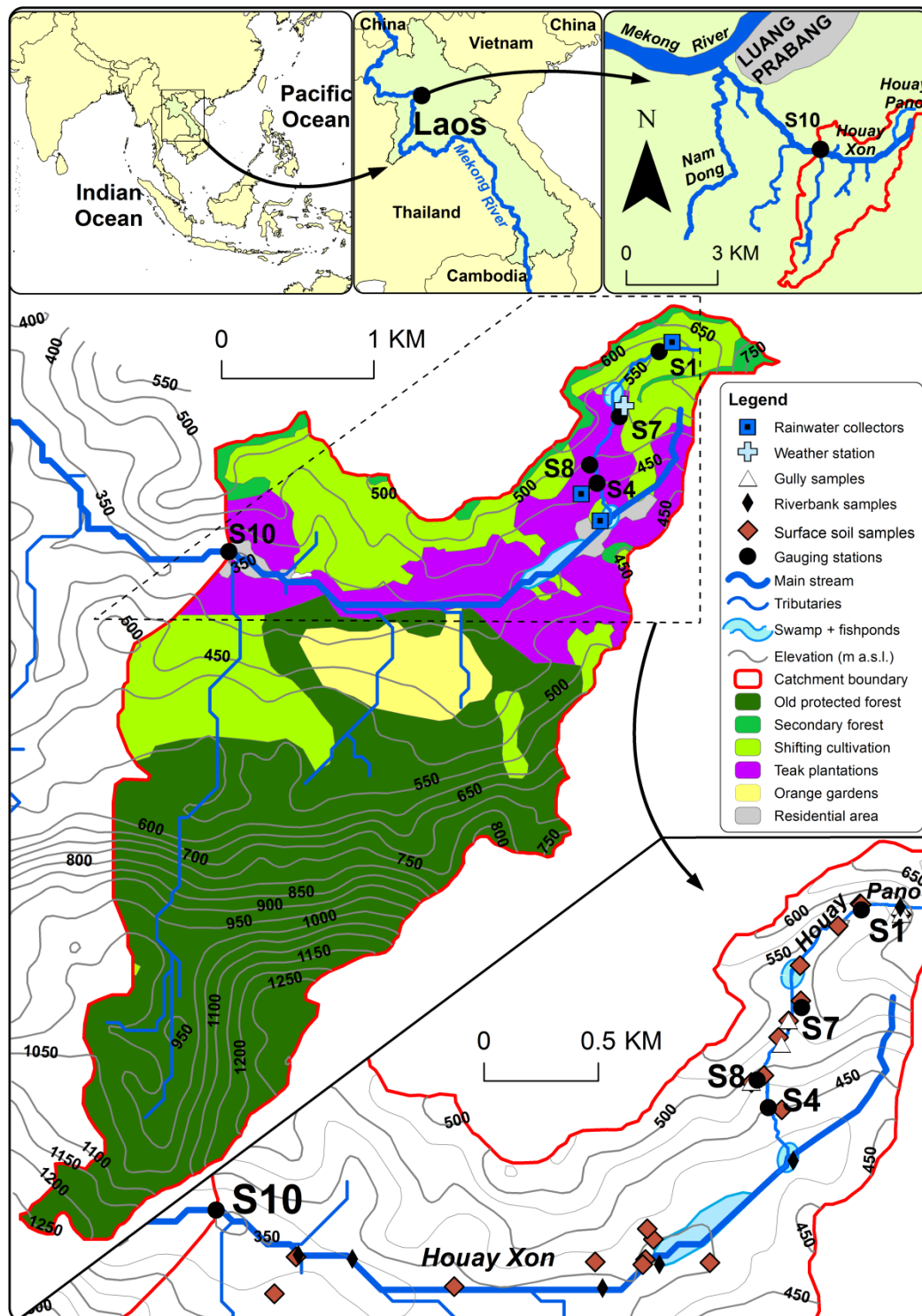


Fig. 2 Scatter plots of radionuclide activities (Bq kg^{-1}) measured in soil samples and in the in-situ sediment source samples

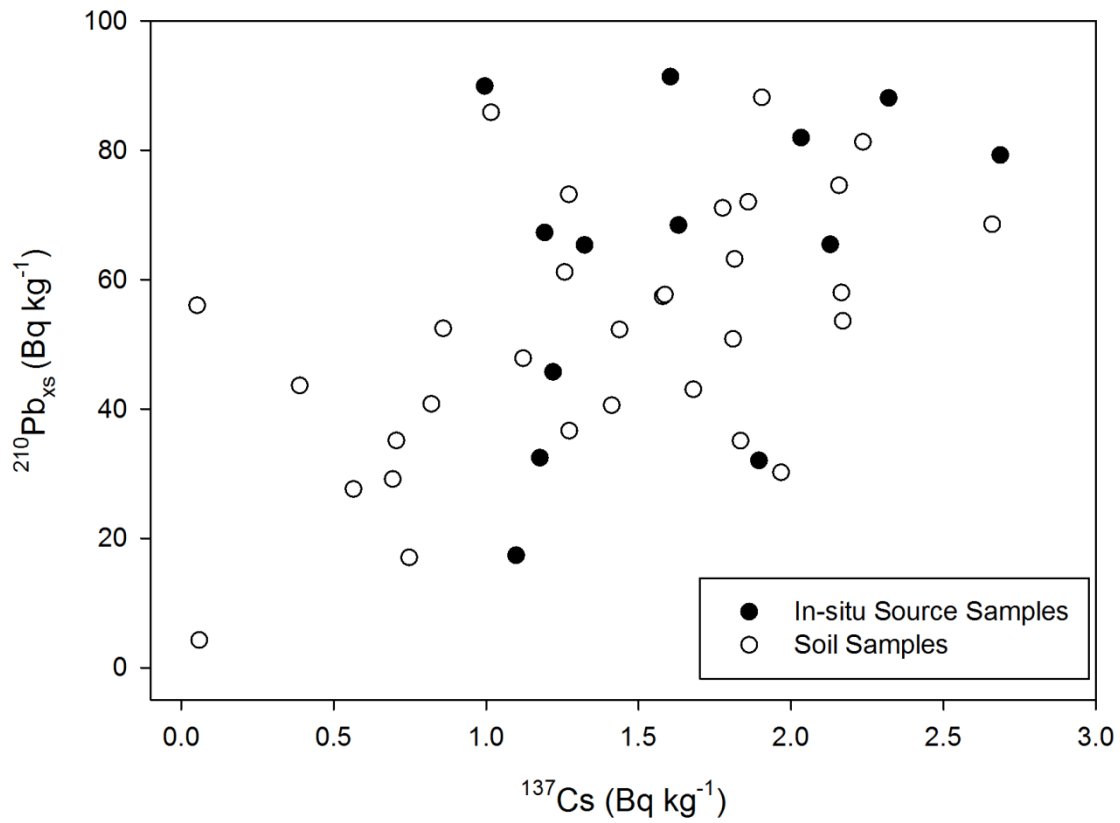


Fig. 3 Scatter plots of radionuclide activities (Bq kg^{-1}) measured directly from source samples and also the in-situ sediment source samples (Plots A & B – left side). The four source quadrants are delineated via the dashed lines on plots A and C. The regrouped source samples and suspended sediment samples from S10 and S4 are plotted the right side (C & D)

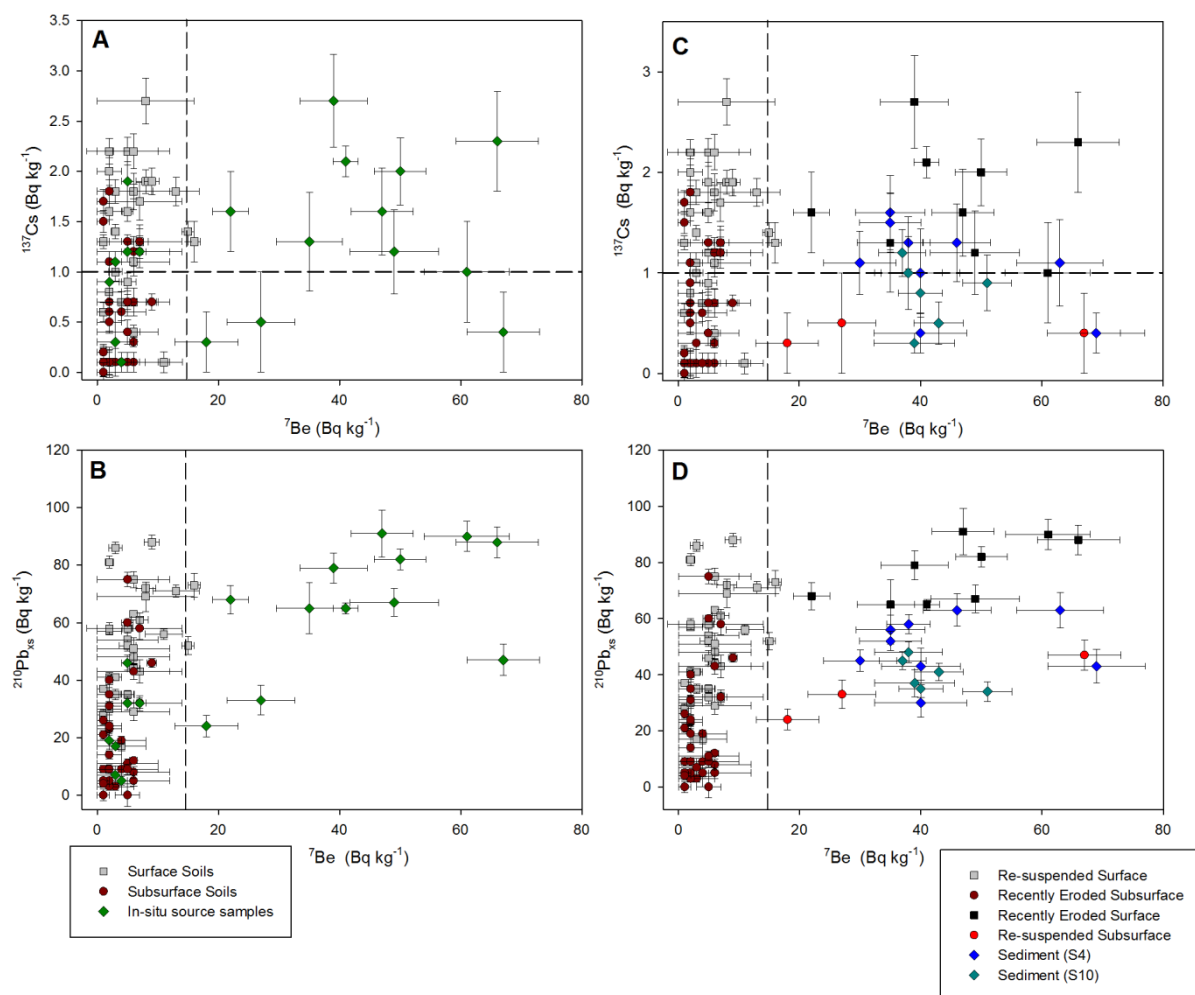


Fig. 4 Hydro-sedimentary characteristics and evolution of mean radionuclide activities in suspended sediment collected at S4 and S10 stations during the flood

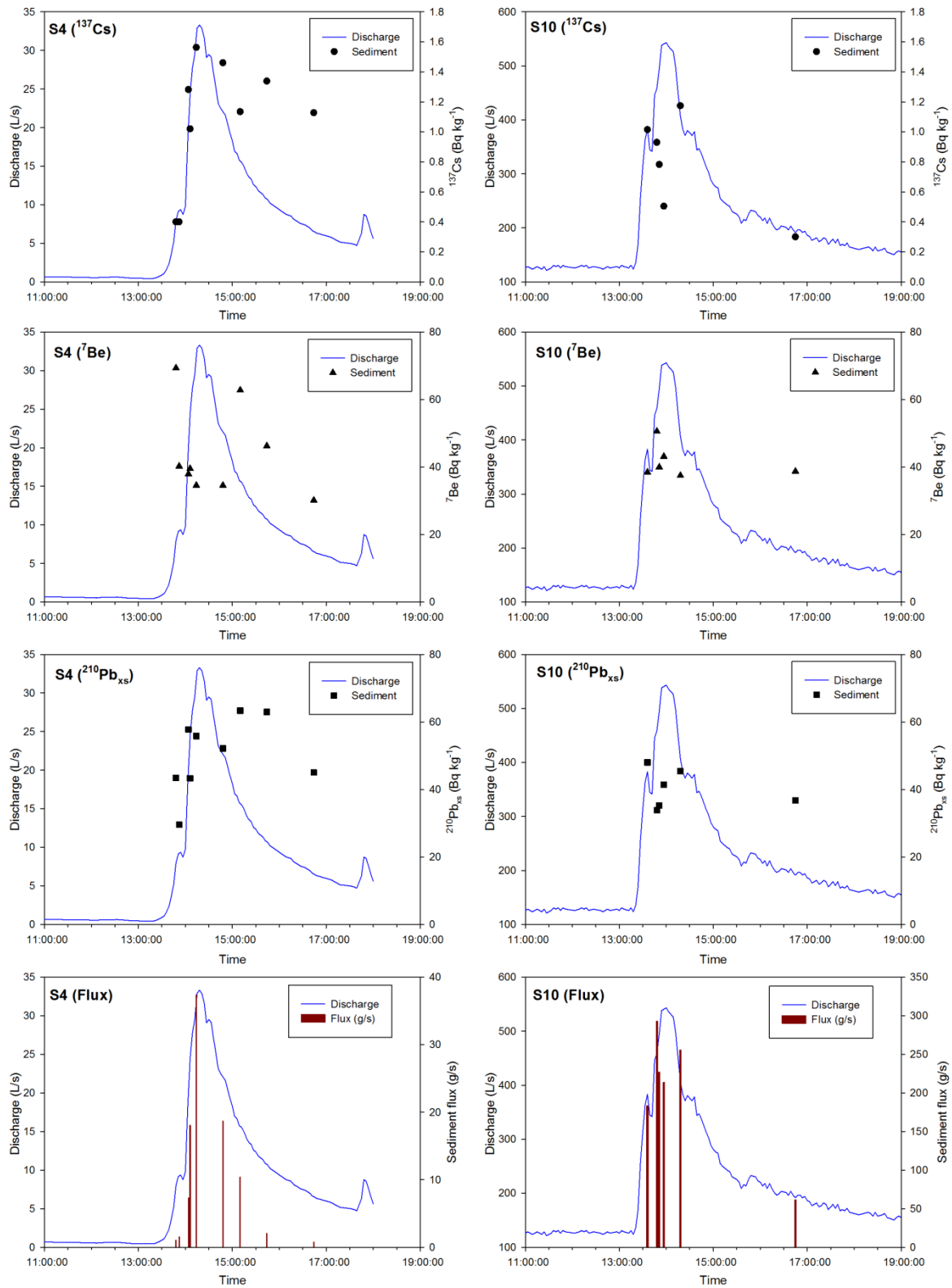


Fig. 5 Evolution of the weighted contributions of the four potential sources to the sediment flux sampled at S4 (Top) and S10 (Bottom) station during the June 16, 2014 flood

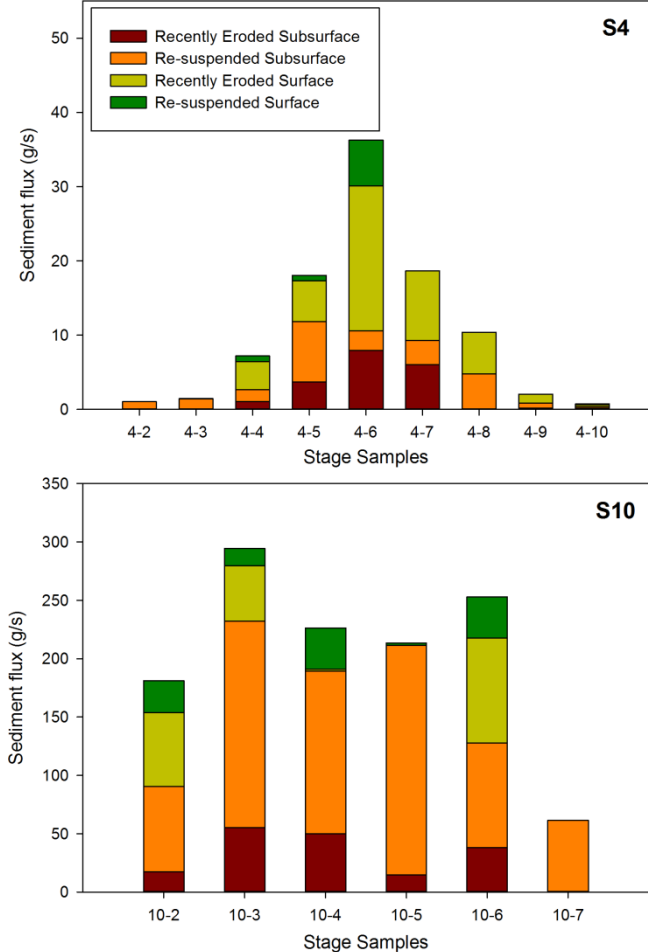


Fig. 6 Rainfall and discharge at the S10 monitoring station demonstrating the potential for ^7Be labelling of subsurface material transiting the stream network

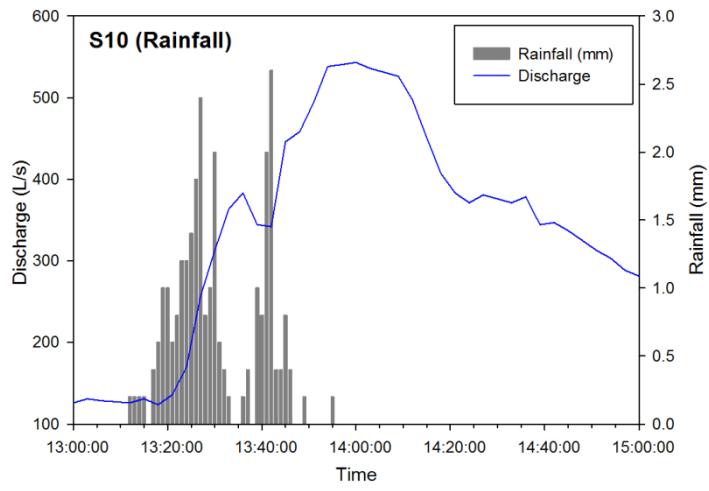


Table 1 Mean and standard deviations of radionuclide activities (Bq kg^{-1}) measured in the four sediment types (all measurements are provided in the Electronic Supplementary Material, Table S2)

Radionuclide activities (Bq kg^{-1})

Source	⁷ Be		¹³⁷ Cs		²¹⁰ Pb _{xs}	
	Mean	SD	Mean	SD	Mean	SD
Re-suspended Surface (<i>n</i> =35)	5.6	3.7	1.4	0.6	50	20.1
Recently Eroded Surface (<i>n</i> =9)	45.5	13.2	1.8	0.6	77	11.0
Re-suspended Subsurface (<i>n</i> =3)	37.3	26.1	0.4	0.1	35	11.4
Recently Eroded Subsurface (<i>n</i> =36)	3.3	2.1	0.6	0.5	20	18.5

Table 2 Mann-Whitney U-test results (p-values) for all possible source combinations with a * indicating non-significant discrimination between sources

Sources	Radionuclide	Re-suspended Surface (n=35)	Recently Eroded Surface (n=9)	Re-suspended Subsurface (n=3)	Recently Eroded Subsurface (n=36)
Re-suspended Surface	⁷ Be	--	--	--	--
	¹³⁷ Cs	--	--	--	--
	²¹⁰ Pb _{xs}	--	--	--	--
Recently Eroded Surface	⁷ Be	<0.001	--	--	--
	¹³⁷ Cs	0.130*	--	--	--
	²¹⁰ Pb _{xs}	<0.001	--	--	--
Re-suspended Subsurface	⁷ Be	0.005	0.579*	--	--
	¹³⁷ Cs	0.015	0.016	--	--
	²¹⁰ Pb _{xs}	0.176*	0.016	--	--
Recently Eroded Subsurface	⁷ Be	0.004	<0.001	0.005	--
	¹³⁷ Cs	<0.001	<0.001	0.813*	--
	²¹⁰ Pb _{xs}	<0.001	<0.001	0.097*	--

Table 3 Distribution modelling results for the groupings (ALL) and individual stage samples for both the four end-member and tow end-member mixing models including the % contribution and the compiled model error (CE)

Model	Recently Eroded Subsurface		Re-suspended Subsurface		Recently Eroded Surface		Re-suspended Surface		Elevated ⁷ Be		Subsurface	
	%	CE	%	CE	%	CE	%	CE	%	CE	%	CE
4-ALL	11	22	48	19	28	17	12	14	85	6	59	8
10-ALL	24	13	53	15	24	12	0	7	79	5	75	7
S4-2	0	14	100	13	0	9	0	4	100	7	95	6
S4-3	0	25	92	45	0	7	2	7	76	7	95	6
S4-4	14	20	22	30	52	23	10	10	71	5	48	8
S4-5	20	11	45	14	30	12	4	9	73	5	58	8
S4-6	21	18	7	24	52	20	16	12	67	8	22	9
S4-7	32	13	17	12	50	16	0	12	63	4	27	7
S4-8	0	7	46	14	54	15	0	10	100	6	52	9
S4-9	10	17	32	12	59	15	0	15	98	12	36	9
S4-10	24	12	29	13	30	13	17	13	55	7	50	9
S10-2	10	14	40	20	35	16	15	15	72	6	59	8
S10-3	19	14	60	17	16	11	5	11	100	6	65	8
S10-4	22	12	61	15	1	14	16	12	77	6	75	9
S10-5	7	11	92	25	0	13	1	8	84	7	90	9
S10-6	15	19	35	16	35	17	14	18	72	7	56	6
S10-7	0	13	100	32	0	8	0	5	76	7	99	6

Table 4 Weighted modelling results for the individual stage samples listing the contribution in grams per second (g/s) and the compiled error (CE) converted to g s^{-1}

Model	Recently Eroded Subsurface		Re-suspended Subsurface		Recently Eroded Surface		Re-suspended Surface	
	g s^{-1}	CE	g s^{-1}	CE	g s^{-1}	CE	g s^{-1}	CE
S4-2	0.0	0.0	1.1	0.1	0.0	0.0	0.0	0.0
S4-3	0.0	0.0	1.4	0.6	0.0	0.0	0.0	0.0
S4-4	1.0	0.2	1.6	0.5	3.8	0.9	0.8	0.1
S4-5	3.7	0.4	8.1	1.1	5.5	0.6	0.8	0.1
S4-6	7.9	1.4	2.6	0.6	19.5	3.9	6.1	0.8
S4-7	6.0	0.8	3.2	0.4	9.4	1.5	0.0	0.0
S4-8	0.0	0.0	4.8	0.7	5.6	0.8	0.0	0.0
S4-9	0.2	0.0	0.7	0.1	1.2	0.2	0.0	0.0
S4-10	0.2	0.0	0.2	0.0	0.2	0.0	0.1	0.0
S10-2	17.4	2.4	73.1	14.8	63.4	10.0	27.1	4.0
S10-3	55.2	7.7	176.8	30.4	47.7	5.4	14.8	1.6
S10-4	50.1	6.2	139.2	20.3	1.6	0.2	35.4	4.3
S10-5	15.0	1.7	196.4	48.5	0.0	0.0	2.1	0.2
S10-6	38.1	7.2	89.5	14.5	90.0	15.2	35.3	6.4
S10-7	0.0	0.0	61.5	20.0	0.0	0.0	0.0	0.0

ELECTRONIC SUPPLEMENTARY MATERIAL

SEDIMENTS, SEC 3 • HILLSLOPE AND RIVER BASIN SEDIMENT DYNAMICS • RESEARCH ARTICLE

Combining multiple fallout radionuclides (^{137}Cs , ^7Be , $^{210}\text{Pb}_{\text{xs}}$) to investigate temporal sediment source dynamics in tropical, ephemeral riverine systems

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Table S1 Pearson's correlation coefficients that were incorporated directly into the distribution modelling framework in Oracle's Crystal Ball software

Recently Eroded			
Surface	²¹⁰ Pb _{xs}	¹³⁷ Cs	⁷ Be
²¹⁰ Pb _{xs}	1		
¹³⁷ Cs	0.100	1	
⁷ Be	0.681	-0.026	1
Re-suspended			
Subsurface	²¹⁰ Pb _{xs}	¹³⁷ Cs	⁷ Be
²¹⁰ Pb _{xs}	1		
¹³⁷ Cs	0.510	1	
⁷ Be	0.436	0.074	1
Recently Eroded			
Subsurface	²¹⁰ Pb _{xs}	¹³⁷ Cs	⁷ Be
²¹⁰ Pb _{xs}	1		
¹³⁷ Cs	0.087	1	
⁷ Be	0.917	-0.118	1
Re-suspended			
Surface	²¹⁰ Pb _{xs}	¹³⁷ Cs	⁷ Be
²¹⁰ Pb _{xs}	1		
¹³⁷ Cs	0.642	1	
⁷ Be	0.398	0.115	1

Table S2 Dataset used for modelling and analyses of sources with radionuclides expressed in Bq kg⁻¹

Sample ID	Type	Sub-Type 2	Sub-Type 3	Sub-Type 4	²¹⁰ Pb _{xs}	¹³⁷ Cs	⁷ Be	error Pb	error Cs	error Be
LS-001	Sediment	S10	2	S10-2	48	1	38	4	0.4	6
LS-002	Sediment	S10	3	S10-3	34	0.9	51	4	0.3	4
LS-003	Sediment	S10	4	S10-4	35	0.8	40	3	0.2	4
LS-004	Sediment	S10	5	S10-5	41	0.5	43	3	0.2	4
LS-005	Sediment	S10	6	S10-6	45	1.2	37	3	0.2	4
LS-006	Sediment	S10	7	S10-7	37	0.3	39	5	0.1	7
LS-007	Sediment	S4	1	S4-1	10	0.6	22	5	0.3	9
LS-008	Sediment	S4	2	S4-2	43	0.4	69	6	0.2	8
LS-009	Sediment	S4	3	S4-3	30	0.4	40	5	0.2	8
LS-010	Sediment	S4	4	S4-4	58	1.3	38	3	0.3	4
LS-011	Sediment	S4	5	S4-5	43	1	40	7	0.4	7
LS-012	Sediment	S4	6	S4-6	56	1.6	35	5	0.4	6
LS-013	Sediment	S4	7	S4-7	52	1.5	35	3	0.3	5
LS-014	Sediment	S4	8	S4-8	63	1.1	63	6	0.4	7
LS-015	Sediment	S4	9	S4-9	63	1.3	46	6	0.4	6
LS-016	Sediment	S4	10	S4-10	45	1.1	30	4	0.3	6
LS-017	Source	Subsurface	Low ⁷ Be	Sub-Recent	21	1.7	1	2	0.1	1
LS-018	Source	Subsurface	Low ⁷ Be	Sub-Recent	4	0.1	1	1	0.1	1
LS-019	Source	Subsurface	Low ⁷ Be	Sub-Recent	5	0.1	6	2	0.1	6
LS-020	Source	Subsurface	Low ⁷ Be	Sub-Recent	43	1.2	6	3	0.2	6
LS-021	Source	Subsurface	Low ⁷ Be	Sub-Recent	8	0.7	6	2	0.1	6
LS-022	Source	Subsurface	Low ⁷ Be	Sub-Recent	58	1.2	7	4	0.2	7
LS-023	Source	Subsurface	Low ⁷ Be	Sub-Recent	31	0.7	2	2	0.1	2
LS-024	Source	Subsurface	Low ⁷ Be	Sub-Recent	14	0.6	2	1	0.1	2
LS-025	Source	Subsurface	Low ⁷ Be	Sub-Recent	23	1.1	2	1	0.1	2
LS-026	Source	Subsurface	Low ⁷ Be	Sub-Recent	9	0.2	1	1	0.1	1
LS-027	Source	Subsurface	Low ⁷ Be	Sub-Recent	9	0.6	2	1	0.1	2
LS-028	Source	Subsurface	Low ⁷ Be	Sub-Recent	23	0.1	2	2	0.1	2
LS-029	Source	Subsurface	Low ⁷ Be	Sub-Recent	0	0.1	5	4	0.1	2
LS-030	Source	Subsurface	Low ⁷ Be	Sub-Recent	3	0.1	3	1	0.1	3
LS-031	Source	Subsurface	Low ⁷ Be	Sub-Recent	3	0.1	2	1	0.1	2
LS-032	Source	Subsurface	Low ⁷ Be	Sub-Recent	5	0.1	2	1	0.1	2
LS-033	Source	Subsurface	Low ⁷ Be	Sub-Recent	24	0.5	2	2	0.1	2
LS-034	Source	Subsurface	Low ⁷ Be	Sub-Recent	35	0.6	2	2	0.1	2
LS-035	Source	Subsurface	Low ⁷ Be	Sub-Recent	40	1.8	2	2	0.1	1
LS-036	Source	Subsurface	Low ⁷ Be	Sub-Recent	26	1.5	1	2	0.1	1
LS-037	Source	Subsurface	Low ⁷ Be	Sub-Recent	24	0.5	2	2	0.1	2

LS-038	Source	Subsurface	Low ⁷ Be	Sub-Recent	9	0.1	4	9	0.1	1
LS-039	Source	Subsurface	Low ⁷ Be	Sub-Recent	5	0.1	1	1	0.1	1
LS-040	Source	Subsurface	Low ⁷ Be	Sub-Recent	4	0	1	1	0.1	1
LS-041	Source	Subsurface	Low ⁷ Be	Sub-Recent	32	1.3	7	3	0.2	7
LS-042	Source	Subsurface	Low ⁷ Be	Sub-Recent	12	0.3	6	1	0.0	1
LS-043	Source	Subsurface	Low ⁷ Be	Sub-Recent	75	0.7	5	3	0.1	5
LS-044	Source	Subsurface	Low ⁷ Be	Sub-Recent	46	0.7	9	2	0.1	1
LS-045	Source	Subsurface	Low ⁷ Be	Sub-Recent	0	0.2	1	2	0.1	1
LS-046	Source	Subsurface	Low ⁷ Be	Sub-Recent	9	0.4	5	2	0.1	5
LS-047	Source	Subsurface	Low ⁷ Be	Sub-Recent	11	0.1	5	2	0.1	5
LS-048	Source	Subsurface	Low ⁷ Be	Sub-Recent	19	0.6	4	1	0.1	4
LS-049	Source	Subsurface	Low ⁷ Be	Sub-Recent	60	1.3	5	1	0.1	1
LS-050	Source	Surface	Low ⁷ Be	Surf-Re-Sus	28	0.6	1	2	0.1	1
LS-051	Source	Surface	Low ⁷ Be	Surf-Re-Sus	56	0.1	11	2	0.1	3
LS-052	Source	Surface	Low ⁷ Be	Surf-Re-Sus	71	1.8	13	2	0.1	4
LS-053	Source	Surface	Low ⁷ Be	Surf-Re-Sus	57	1.6	2	1	0.1	2
LS-054	Source	Surface	Low ⁷ Be	Surf-Re-Sus	43	1.7	7	4	0.2	7
LS-055	Source	Surface	Low ⁷ Be	Surf-Re-Sus	54	2.2	5	3	0.1	5
LS-056	Source	Surface	Low ⁷ Be	Surf-Re-Sus	48	1.1	6	2	0.1	6
LS-057	Source	Surface	Low ⁷ Be	Surf-Re-Sus	72	1.9	8	2	0.1	2
LS-058	Source	Surface	Low ⁷ Be	Surf-Re-Sus	41	0.8	2	2	0.1	2
LS-059	Source	Surface	Low ⁷ Be	Surf-Re-Sus	37	1.3	1	1	0.1	1
LS-060	Source	Surface	Low ⁷ Be	Surf-Re-Sus	4	0.1	2	1	0.1	2
LS-061	Source	Surface	Low ⁷ Be	Surf-Re-Sus	81	2.2	2	2	0.1	1
LS-062	Source	Surface	Low ⁷ Be	Surf-Re-Sus	73	1.3	16	4	0.2	1
LS-063	Source	Surface	Low ⁷ Be	Surf-Re-Sus	52	1.4	15	3	0.1	1
LS-064	Source	Surface	Low ⁷ Be	Surf-Re-Sus	63	1.8	6	1	0.1	1
LS-065	Source	Surface	Low ⁷ Be	Surf-Re-Sus	58	1.6	5	2	0.1	1
LS-066	Source	Surface	Low ⁷ Be	Surf-Re-Sus	52	0.9	5	2	0.1	1
LS-067	Source	Surface	Low ⁷ Be	Surf-Re-Sus	44	0.4	6	2	0.1	1
LS-068	Source	Surface	Low ⁷ Be	Surf-Re-Sus	58	2.2	2	2	0.1	4
LS-069	Source	Surface	Low ⁷ Be	Surf-Re-Sus	75	2.2	6	3	0.2	6
LS-070	Source	Surface	Low ⁷ Be	Surf-Re-Sus	41	1.4	3	1	0.1	1
LS-071	Source	Surface	Low ⁷ Be	Surf-Re-Sus	35	0.7	5	1	0.1	1
LS-072	Source	Surface	Low ⁷ Be	Surf-Re-Sus	30	2	2	2	0.1	2
LS-073	Source	Surface	Low ⁷ Be	Surf-Re-Sus	61	1.3	7	2	0.1	1

LS-074	Source	Surface	Low ⁷ Be	Surf-Re-Sus	86	1	3	2	0.1	1
LS-075	Source	Surface	Low ⁷ Be	Surf-Re-Sus	69	2.7	8	5	0.2	8
LS-076	Source	Surface	Low ⁷ Be	Surf-Re-Sus	51	1.8	6	4	0.2	6
LS-077	Source	Surface	Low ⁷ Be	Surf-Re-Sus	35	1.8	3	2	0.1	3
LS-078	Source	Surface	Low ⁷ Be	Surf-Re-Sus	29	0.7	6	3	0.1	6
LS-079	Source	Surface	Low ⁷ Be	Surf-Re-Sus	17	0.7	4	2	0.1	4
LS-080	Source	Surface	Low ⁷ Be	Surf-Re-Sus	88	1.9	9	2	0.1	1
LS-081	Source-in-situ	Subsurface	High ⁷ Be	Surf-Re-Sus	47	0.4	67	5	0.4	6
LS-082	Source-in-situ	Subsurface	High ⁷ Be	Surf-Re-Sus	33	0.5	27	5	0.5	6
LS-083	Source-in-situ	Subsurface	High ⁷ Be	Surf-Re-Sus	24	0.3	18	4	0.3	5
LS-084	Source-in-situ	Subsurface	Low ⁷ Be	Sub-Re-Sus	19	0.9	2	3	0.3	2
LS-085	Source-in-situ	Subsurface	Low ⁷ Be	Sub-Re-Sus	5	0.1	4	2	0.1	1
LS-086	Source-in-situ	Subsurface	Low ⁷ Be	Sub-Re-Sus	7	0.3	3	3	0.3	3
LS-087	Source-in-situ	Surface	High ⁷ Be	Surf-Recent	65	1.3	35	9	0.5	5
LS-088	Source-in-situ	Surface	High ⁷ Be	Surf-Recent	88	2.3	66	5	0.5	7
LS-089	Source-in-situ	Surface	High ⁷ Be	Surf-Recent	91	1.6	47	8	0.4	5
LS-090	Source-in-situ	Surface	High ⁷ Be	Surf-Recent	82	2	50	4	0.3	4
LS-091	Source-in-situ	Surface	High ⁷ Be	Surf-Recent	65	2.1	41	2	0.2	2
LS-092	Source-in-situ	Surface	High ⁷ Be	Surf-Recent	79	2.7	39	5	0.5	6
LS-093	Source-in-situ	Surface	High ⁷ Be	Surf-Recent	90	1	61	5	0.5	7
LS-094	Source-in-situ	Surface	High ⁷ Be	Surf-Recent	67	1.2	49	5	0.4	7
LS-095	Source-in-situ	Surface	High ⁷ Be	Surf-Recent	68	1.6	22	5	0.4	3
LS-096	Source-in-situ	Surface	Low ⁷ Be	Surf-Re-Sus	32	1.2	7	2	0.1	1
LS-097	Source-in-situ	Surface	Low ⁷ Be	Surf-Re-Sus	32	1.9	5	2	0.2	1
LS-098	Source-in-situ	Surface	Low ⁷ Be	Surf-Re-Sus	46	1.2	5	3	0.1	1
LS-099	Source-in-situ	Surface	Low ⁷ Be	Surf-Re-Sus	17	1.1	3	1	0.1	1
