

Use of fallout radionuclides (7Be, 210Pb) to estimate resuspension of Escherichia coli from streambed sediments during floods in a tropical montane catchment

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1	Use of fallout radionuclides (⁷ Be, ²¹⁰ Pb) to estimate
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Abstract Consumption of water polluted by faecal contaminants is responsible for 2 million deaths annually, most of which occur in developing countries without adequate sanitation. In tropical aquatic systems, streambeds can be reservoirs of persistent pathogenic bacteria and high rainfall can lead to contaminated soils entering streams and to the resuspension of sediment-bound microbes in the streambed. Here we present a novel method using fallout radionuclides (⁷Be and $^{210}Pb_{xx}$) to estimate the proportions of *Escherichia coli*, an indicator of faecal contamination, associated with recently eroded soil particles and with the resuspension of streambed sediments (Fig. 1). We show that using these radionuclides and hydrograph separations we are able to characterize the proportion of particles originating from highly contaminated soils and that from the resuspension of particle-attached bacteria within the streambed. We also found that although overland flow represented just over one-tenth of the total flood volume, it was responsible for more than two-thirds of the downstream transfer of E. coli. We propose that data obtained using this method can be used to understand the dynamics of FIB (Faecal Indicator Bacteria) in streams thereby providing information for adapted management plans that reduce the health risks to local populations.

Keywords: FIB, South East Asia, Hydrological modelling, erosion, land use, overland flow, beryllium-7, lead-210.

Introduction

It has been estimated that almost 2 million people die annually as a consequence of waterborne diarrheal diseases caused by the use of untreated water. The majority of the victims are children between 0 to 5yr old living in developing countries (WHO 2012). The rural areas of South East Asia (SEA) are particularly affected because many of the poorest households living in the montane agro-ecosystems use contaminated river water for basic domestic needs such as washing, bathing and cooking (Ribolzi et al. 2011a, Rochelle-Newall et al. 2015, Saqalli et al. 2015). Moreover, in areas where proper sanitation infrastructures are lacking, open area human defecation and the use of shallow latrines is frequent (Guiteras et al. 2015, Sturma and Becerra 2012). Furthermore, in the absence of animal husbandry practices that protect streams through the installation of fences, the soil surface in rural areas of SEA is often contaminated with microbial faecal pollutants of human and animal origins (WorldBank 2009). Indeed, in tropical and temperate systems, land use is known to play an important

role in determining the degree of faecal contamination in a watershed (Ekklesia et al. 2015, Goto and Yan 2011).

The presence of faecal contamination in soil, sediment and water samples is generally determined using indicator bacteria. These faecal indicator bacteria (FIB) are used because they are much easier to culture and are considered to have a lower health risk than the pathogens for which they are an indicator (Ishii and Sadowsky 2008). Of the FIB generally used, Escherichia coli is often cited as being a good indicator of faecal contamination although some authors have questioned the relevance of its use in tropical systems (Byappanahalli and Fujioka 1998, Byappanahalli et al. 2012). Nevertheless, most of the methods used to determine the presence of *E. coli* are easy to employ, relatively cheap, and do not require complex analytical equipment and are therefore adapted for use in sites without access to high grade laboratory facilities.

Bacteria in aquatic systems can be either free-living or attached to particles and the relative percentages of each fraction vary as a function of turbidity and particle quality (Lemee et al. 2002, Suter et al. 2011). Probably as a consequence of their intestinal origin where they are attached to digested food particles, FIB are mostly particle-bound although the actual percentage associated with particles does vary with TSS concentrations (Byappanahalli et al. 2012, Nguyen et al. in press, Oliver et al. 2007). During storm events characterised by high solid loads (stormflow loading), FIB numbers are often higher than during the inter-storm, low flow periods characterised by lower particle loads (Causse et al. 2015, Chan et al. 2015, Hrdinka et al. 2012, Stumpf et al. 2010). During periods of low flow, the fraction of particle-bound FIB can be deposited and stored in the river channel and stream sediments to form an in-channel reservoir of FIB that can be mobilized during floods (e.g. Pachepsky et al. 2011, Pandey et al. 2012).

There is a paucity of knowledge on the distribution and survival of FIB in tropical freshwater environments (Rochelle-Newall et al. 2015). In these ecosystems higher and more stable temperatures, greater reserves of nutrients and organic carbon and more stable humidity conditions may lead to higher bacterial survival rates, particularly in sediments where FIB are protected from light and predators (Garzio-Hadzick et al. 2010, Pachepsky and Shelton 2011). Given the risks for human health associated with the consumption of contaminated water, quantifying the proportion of FIB resuspended with sediments during floods is crucial in tropical catchments where informal water sources often

supply the majority of domestic requirements. Providing solid estimates of this resuspension is essential for the validation of model simulations on FIB transport and survival (Cho et al. 2012, Kim et al. 2010, Pandey et al. 2012).

Particle-bound FIB that are transported in streams during floods come from two main sources: (1) recently eroded particles from the surface layer of soils on hillslopes and the riparian zone that are supplied to the river by overland flow and (2) resuspension of streambed sediments. However, estimating the relative proportions of these two sources to sediment transport during floods is a difficult challenge. To discriminate recently eroded particles and resuspended material, previous studies have tried to directly quantify resuspension by generating artificial floods in the stream channel during dry weather conditions (Muirhead et al. 2004, Yakirevich et al. 2013). However, these experiments are generally conducted on an isolated reach of the stream and the maximum intensity of these artificial floods remains limited. Moreover, they do not reproduce the dilution effect of pre-event groundwater, which is often a major contributor to natural streamflow (Kirchner 2003). Therefore they are unlikely to recreate the real conditions observed during a natural flood event.

The objective of this short communication is to estimate the fraction of particle attached E. coli resuspended from streambed sediments during a natural flood event in a tropical montane system in SEA. An original method based on previous applications using fallout radionuclide measurements (i.e. ⁷Be, ²¹⁰Pb_{xs}) is proposed (Evrard et al. 2010, Matisoff et al. 2005). These radionuclides, characterised by different half-lives (53 days and 22 years, respectively), are supplied to the soils by rainfall. Upon soil contact they are quickly and almost irreversibly bound to fine soil particles (Taylor et al. 2013). These two radionuclides were recently used to quantify the proportions of recently eroded sediment (sediment labelled by short-lived ⁷Be) and particles remobilised from the riverbed/channel (depleted in ⁷Be) in the study area (Gourdin et al. 2014b). In addition to the quantification of the contribution of both sources (soil erosion and sediment resuspension), this method can be used to estimate the age of resuspended sediments to which particle-bound bacteria are attached.

Material and methods

2 Study site

The study site is a catchment of 60 ha located in North Lao PDR (Fig. 2). It is a sub-basin of the Houay
Xon system, which is a tributary of the Mekong River. The geological substrate consists of argillites,
siltstones and fine-grained sandstones from Permian to Upper Carboniferous (Department of Geology
and Mines, 1990/1991). The soils developed on these bedrocks are mainly Entisol, Ultisol and Alfisol
(Soil Survey Staff 1999).

8 The terrain is steep with a mean slope gradient of 54%, ranging from 0% to more than 171%. 9 The climate is a wet-dry monsoon tropical climate with an average annual rainfall of ~1300 mm.year⁻¹. 10 The rainfall pattern is highly seasonal with 77% of the rainfall occurring during the rainy season from 11 mid-May to mid-October, whereas the November to March period is cooler and mostly dry.

The mean monthly temperature measured at the local meteorological station ranges between 12 to 35 °C, with the highest temperatures observed in April just before the onset of the wet season. 14 Mean monthly relative humidity varies between 17 and 100%. The watershed is characterised by the 15 occurrence of about 30 storms during the wet season of which around 20 generate a flood in the stream. 16 During these storms, rainfall reaches intensities that are commonly observed in tropical systems (up to 100 mmh⁻¹ over a time step of 6 mins).

Land use in the catchment is dominated by cropland (with a short fallow cycle) and teak plantations on the hillslopes (Huon et al. 2013). In the stream zone, small domestic gardens and fish ponds are found. These areas are generally uninhabited during the dry season. However, during the wet season, these zones are more frequently used by farmers and their livestock. During this time the inhabitants construct small semi-permanent camps where open air defecation is practiced. Livestock is of relatively low density and includes pigs, chickens, goats and dogs. Wild animals also live in the fallow areas and can contribute to faecal contamination. There are three swamp zones along the stream length.

26 This catchment can be considered as representative of the montane agro-ecosystems of SEA.
27 Monitored since 1998, this site is part of MSEC (Multi-scale Environmental Changes, <u>http://msec.obs-</u>

mip.fr/) South East Asian agro-hydrological observatory. It belongs to the French network of critical

Hydrological measurements and water sampling

zone observatories (RBV, Réseau de Bassins Versants, http://rnbv.ipgp.fr/).

Rainwater depth and intensity was measured (with a 6 minute time step) using an automatic rain gauge (Cimel). Steam discharge was measured at the catchment outlet using an automated recording station that consisted of a water level recorder (OTT, Thalimedes/SE200) and a V-notch weir. The control-rating curve was previously determined using cross-sectional metering and slug tests. Stream water was collected in plastic bottles by an automatic sampler (Automatic Pumping Type Sediment Sampler, ICRISAT) installed at the catchment outlet (Fig. 2). During this study, 12 1-liter samples were collected over the flood. Electrical conductivity normalized to 25°C (EC) of each sample was determined with an YSI® 556 probe. The concentration of total suspended solid (TSS) was determined for each sample after evaporation in an oven at 105°C for 48 h. Cumulated TSS exports were calculated by integrating the flux of TSS over the flood.

14 Escherichia coli determinations

The standardized microplate method (ISO 9308-3) was used for E. coli load determinations. This method is based on serial dilutions and an evaluation of the most probable number (MPN) using a culture medium specific for *E. coli*. Each sample was incubated at four dilution rates (i.e. 1:2, 1:20, 1:200 and 1:2000) in a 96-well microplate (MUG/EC, BIOKAR DIAGNOSTICS) and incubated for 36 to 48h at 44°C. Ringers' Lactate solution was used for the dilutions and one plate was used per sample. The number of positive wells for each microplate was noted and the MPN was determined using the Poisson distribution. Each sample was incubated within a few hours after the flood. The E. coli load at the sediment surface was also determined at four sites along the stream (Fig. 2). At each site, 10 g of sediment were collected from the upper 2 cm of the sediment surface and were resuspended in 11 of Ringers' Lactate solution. The resulting solution was agitated and the E. coli load was determined using the microplate method. This microplate method has previously been used with success in the studied ecosystem (Causse et al. 2015).

1 Radionuclide analyses

Radionuclide activities were measured in a total of five suspended sediment samples. Samples were packed into 15-60-ml (depending on the quantity available) polyethylene containers and sealed airtight. The ⁷Be and ²¹⁰Pb activities were determined by gamma spectrometry using the very low-background coaxial N- and P-type GeHP detectors at the Laboratoire des Sciences du Climat et de l'Environnement (LSCE; Gif-sur-Yvette, France; (Gourdin et al. 2014b). All results were expressed in Bq kg⁻¹. Activities were also decay-corrected for the sampling date. Uncertainties on radionuclide activities were ca. 10% for ²¹⁰Pb_{xs} and 20% for ⁷Be. All samples were analysed within less than 53 days (⁷Be half-life) following the rainfall event.

10 EC-based hydrograph separation

A tracer-based approach similar to the one described by Ribolzi et al. (1997) and Collins and Neal (1998) was used to separate storm hydrographs into event water, which mainly includes infiltration excess overland flow (Patin et al. 2012, Ribolzi et al. 2011b), and pre-event water, i.e. ground water pre-stored in the catchment (Vigiak et al. 2008). This approach uses a simple mixing model with two reservoirs and the electrical conductivity (EC) of water as a tracer. It is described by the following equations:

$$17 \qquad Q_{\rm sw} = Q_{\rm of} + Q_{\rm gw} \tag{1}$$

18
$$Q_{sw} EC_{sw} = Q_{of} EC_{of} + Q_{gw} EC_{gw}$$
(2)

Where Q_{sw} represents the instantaneous stream water discharge at the catchment outlet, Q_{of}, the instantaneous discharge of overland flow and Qgw, the instantaneous discharge of groundwater (expressed in 1 s⁻¹). EC_{sw} is the instantaneous EC measured in the stream. EC_{of} and EC_{gw} are the average values respectively for overland flow and groundwater during the storm event. ECof was measured on samples of overland flow that were collected from the soil surface on hillslopes draining to the stream during the rainfall event (Patin et al. 2012). Since groundwater feeds the stream during inter-stormflow periods (Ribolzi et al. 1997), ECgw was approximated from stream measurements carried out prior to storm event.

Solidograph separation and estimate of E. coli resuspension

Due to their different half-lives (53 days and 22 years respectively), ⁷Be and ²¹⁰Pb_{xs} are particularly relevant for quantifying the respective contributions of (1) particles detached from the soil surface along hillslopes and the riparian zone (i.e. recently tagged with ⁷Be) and (2) resuspension of stream bed sediments (i.e. isolated from recent fallout and depleted in ⁷Be). The percentage of "new" suspended particles (Pnew, recently labelled particles detached from the soil surface) in the total stream load at a given sampling time was estimated following the method of Matisoff et al. (2005): $P_{\text{new}} = 100 [(A/B) / (A_0/B_0)]$ (3)

where A and B are the ⁷Be and $^{210}Pb_{xs}$ activities (Bq kg⁻¹) of the suspended particles collected from the stream and A_0 and B_0 are the ⁷Be and ²¹⁰Pb_{xs} activities in rainfall (Bq l⁻¹). The value 5.4 was used to estimate the ratio A_0/B_0 . It was obtained by averaging signature of 4 rain showers that occurred immediately prior to the flood (i.e. 6-April, 23-May, 31-May and 1-Jun 2012). This value is consistent with other work from the same study area (Gourdin et al. 2014a, Gourdin et al. 2014b).

The proportion of "old" suspended particles (Pold, resuspended streambed sediments isolated from recent fallout) was estimated by the following equations:

16
$$P_{old} = 100 - P_{new}$$
 (4)

17
$$F_{old} = F_{sw} P_{old} / 100$$
 (5)

Where F_{sw} and F_{old} are, respectively, the total and old suspended solid particle flows (kg s⁻¹) at time t. The flow of *E. coli* from streambed sediment resuspension (F_{E.coli} old; expressed in MPN s⁻¹), mainly composed of old sediment, was then estimated from:

$$21 \qquad F_{E.coli_old} = F_{old} SB_{E.coli} \tag{6}$$

Where SB_{Ecoli} is the total load of *E. coli* in streambed sediment (MPN kg⁻¹). The flow of *E. coli* due to recently labelled particles detached from the soil surface ($F_{E,coli}$ new; expressed in MPN s⁻¹) was then estimated from:

$$25 F_{E.coli_new} = F_{E.coli_sw} - F_{E.coli_old} (7)$$

 $F_{E.coli_sw}$ is the flow of *E. coli* in the stream at time t (MPN s⁻¹) and SW_{E.coli} is the concentration of *E. coli* in the stream water sample (MPN l⁻¹) at time t.

Results

Description of the rainfall-runoff event

The studied flood was triggered by a storm that occurred on May 15th, 2012. It was the first significant erosive event of the rainy season. During the storm, maximum rainfall intensity occurred between 10:30 am and 10:36 am and reached 75 mm h⁻¹ (Fig. 3). The cumulated rainfall over the 48 min rain event was 33.5 mm. This corresponds to approximately a one-year return period (34.7 mm daily rainfall) according to Bricquet et al. (2003) for the 1950-2000 period. The lag time between the peak of rainfall and peak discharge was 12 minutes. This storm generated a stormflow of approximately 273 min during which discharge peaked at 90 L s⁻¹ at 10:48. This corresponds to a maximum specific discharge of 1.5 L s⁻¹ha⁻¹. The runoff coefficient for this event (depth of streamflow/depth of rain) was 3%.

15 Variations in the stream flow composition

The initial EC was 533 μ S cm⁻¹, characteristic of groundwater (Fig. 3). This then rapidly decreased to 358 μ S cm⁻¹ during the peak of storm flow, indicating the increasing contribution of overland flow that has a lower EC (40 μ S cm⁻¹). EC then progressively increased as the flood receded. TSS concentration reached a peak of 24g L⁻¹ and TSS loss during the 425min event was 4.3 Mg.

The activity of 210 Pb_{xs} remained relatively stable throughout the flood event at 80 Bq kg⁻¹). In contrast, ⁷Be activity was high at the start of the rain event (398 Bq kg⁻¹) it then further increased slightly to 438 Bq kg⁻¹ at beginning of the flood recession. Activity then dropped drastically to 62 Bq kg⁻¹ during the flood recession to thereafter increase to more enriched values towards the end of the flood event. *E. coli* concentrations in the floodwaters were 2100 MPN 100 ml⁻¹ during the early stage of the rising flood. This then increased a maximum value of 49000 MPN 100 ml⁻¹ at the start of the

(8)

4 Separation of suspended particles and *E. coli* fluxes

5 The separation of TSS flux into recently eroded particles ("new") and resuspended sediments flux 6 ("old") from the streambed is shown in Fig. 4. At the beginning of the flood the major proportion of 7 particles are "new" with an apparent age of 8 days indicating that they were recently marked by ⁷Be at 8 the bare soil surface. This peak corresponds to the peak in overland flow. As the flood progressed, the 9 proportion of "old" particles reached a maximum apparent age of 158 days at 140 minutes after the 10 start of the flood and then declined.

The flux of *E. coli* associated with particles recently labelled with ⁷Be was always higher than that associated with the older particles and this during the entire flood. The maximum flux of E. coli associated with recently labelled particles $(17.9 \times 10^6 \text{ MPN x s}^{-1})$ occurred at peak solid flux (77 mins after the flood peak). Flux then declined steadily until the end of the flood. In contrast to the peak flux of recent particles, the peak in E. coli associated with older particles occurred after a lag of approximately 1 hour. We conducted an uncertainty analysis (Monte Carlo type) following the method of Ribolzi et al (2000) to estimate the confidence levels on the proportions of E. coli attached to "old" particles at the point of maximum contribution in the hydrograph. We found that the associated errors lead to a coefficient of variation of 12% and 14% of the total fluxes of E. coli attached to "old" and "new" particles, respectively. Over the course of the flood a total of 125 x 10⁹ MPN E. coli were exported, of which about 89 % were associated with recently eroded particles and 11 % with particles resuspended from the riverbed.

Discussion

Here, we present a novel method using fallout radionuclides (^{7}Be and $^{210}Pb_{xs}$) to estimate the

25 proportions of *E. coli* associated with recently eroded soil particles and with the resuspension of

- 26 streambed sediments. This method allows the separation of the total flux of particles into two
- 27 components: "new "and "old" following the model of Matisoff et al. (2005). A key assumption for the

use of ⁷Be as a tracer is that radionuclides sorption to particles is rapid and irreversible over the timescales of interest (Taylor et al. 2012). Gourdin et al. (2014a) recently validated this assumption by implementing laboratory experiments and field observations during a storm event in the Houay Pano catchment. These authors also used this method to quantify the sources and dynamics of suspended sediment and particulate organic matter (Gourdin et al. 2014b, 2015). Here, we use an extension of this work to estimate the transfer times and sources of *E. coli* in an upland stream (Fig. 4).

Overland flow and groundwater

As observed by Gourdin et al (2014a) we found higher contributions of recently eroded sediment at the beginning of the rising flood stage and then a progressive dilution of recently eroded sediment by particles remobilised from the riverbed (Fig. 4). Overland flow represented just over one-tenth of the total flood volume but was responsible for more than two-thirds of the downstream transfer of E. coli (i.e. fraction attached to recently labeled particles). This result confirms the observations of Causse et al. (2015) in the same study area who concluded that the proportion of soil surface washoff was critical in determining the numbers of E. coli in the stream during storms. It is also in agreement with Kim et al. (2010) who simulated E. coli loads in the Little Cove Creek watershed (Pennsylvania) with a version of SWAT (Soil and Water Assessment Tool) that took into account streambed bacteria release. They found that surface runoff carried large numbers of E. coli to the stream and that sediment resuspension contributed to the baseline concentration of E. coli in stream water.

Another point that should also be emphasized is the importance of the groundwater contribution to flood runoff. During the studied flood event, groundwater largely dominated streamflow with an estimated contribution of up to 89%. This hydrological behavior is often observed in rural catchments with dense vegetation cover (e.g. Kirchner 2003). Genereux and Hooper (1998) estimated in a review paper on tracer-based hydrograph separations that the pre-event groundwater contribution to storms is on average $80 \pm 20\%$. Although this is well known to hydrologists and is often quantified using geochemical and isotopic tracer techniques, this phenomenon is rarely discussed in the literature with a view to better understanding the dynamics of FIB and environmental bacteria. In this context, groundwater that is little contaminated as compared to overland flow will tend to dilute streamwater during stormflow. At the same time, the contribution of groundwater adds to that of overland flow to increase discharge and the resuspension of sediments in the streambed.

Methodological considerations

The method presented here is adapted for tropical systems with contrasted dry and wet seasons. Moreover, it is particularly appropriate for events occurring at the start of the rainy season when the stock of ⁷Be supplied by rainfall to soils during the previous wet season has completely decayed. It is also important to note that the first floods are generally the most contaminated as the soils have accumulated contaminants throughout the dry season (Causse et al. 2015).

One of the central assumptions of this work is that E. coli are attached to soil particles and that this does not change during the erosion, resuspension and transport processes along hillslopes and downstream toward the creek. We hypothesized that enteric bacteria are more frequently associated with particles both within the host and in the external environment (soils and water) and that this remains relatively constant over short timescales. High proportions of environmental bacteria and enteric bacteria such as *E.coli* are associated with particles in soils (Oliver et al. 2007) and in aquatic systems (Lemee et al. 2002, Unanue et al. 1992). Our results, obtained during a flash flood with low E. *coli* numbers $(1 - 230 \text{ MPN } 100 \text{ ml}^{-1} \text{ E. coli}; \text{ Causse et al. 2015})$ and low TSS (< 20 mg L⁻¹) in the stream prior to the storm support this assumption. Although it is likely valid in this case study, this hypothesis should nevertheless be verified when applying this method to other study sites. Our method allows the determination of the age of different particles to which E. coli are bound. Therefore, although it does not provide an exact age for E. coli or of the particle, one can postulate that

the bacteria associated with the particles in the stream are older than those associated with the particles recently marked with ⁷Be (i.e. those at the soil surface on the slopes). We conducted an analysis of

- 21 error propagation and by far the largest proportion of the total error is by far associated with the
- 22 calculation of the "old" E. coli fraction (up to 75%). Most of this error probably arises from the

23 uncertainty associated with the SB_{E.coli} determination. Therefore, given the importance of the values of

- streambed *E. coli* in the calculation, it is critical to obtain a robust estimate of this concentration as the
- calculations of "old" and "new" *E. coli* fractions are strongly dependent on this value.

26 Land use

In these tropical agro-ecosystems there is an intensification of activities linked to the preparation of the land for annual crops at the start of the rainy season. This intensification also brings with it a sharp increase in the potential for soil surface contamination due to the use of informal latrines and open-air defecation by humans and livestock. This contamination is then mobilized by the erosive nature of rain drop impacts (Ribolzi et al. 2011b) and is transported to the stream by overland flow (Patin et al. 2012). Bare soils in teak plantations that lack an understory and in young fallow zones where soils have low infiltration rates (Patin et al. 2012) have particularly high overland flows. Moreover teak plantations often encroach on the riparian zones. Therefore any overland flow from the upslope teak plantation may also wash off any FIB available downslope in the riparian zone located between the plantation and stream proper. In the riparian zone, contaminated particles available at the soil surface can also be detached and disseminated downstream within local seepage zones by groundwater exfiltration in association with overland flow (Vigiak et al. 2008). Domestic gardens, temporary housing and fishponds where contamination levels can be high are often located in the riparian zones and the small paths that interconnect these areas can increase runoff (Ziegler et al. 2000) and enhance E. coli transport to the stream. Moreover, riparian zones in tropical systems are often characterised by swamp areas that are both a source and a trap of solid particles and FIB. They therefore play an important role in controlling the dynamic of "old" and "new" E. coli in these systems. Quantifying the contribution of these reservoirs to the total flux of E. coli during flood events allows a better understanding of the redistribution processes and of the dynamic of particle-bound FIB within catchments. This is critical if we wish to fully understand the dynamic of E. coli in watersheds. **Conclusion and perspectives** Here we present a way of estimating both the relative proportion of E. coli resuspended from

streambed sediments and those transferred from the soil surface to the stream by overland flow during a flood event (Fig. 1). At present, because of its analytical cost and level of expertise required for its implementation, the ${}^{7}\text{Be}/{}^{210}\text{Pb}_{xs}$ method is still limited to a small number of laboratories worldwide. Even though the systematic use of this method in a wide range of watersheds is limited, this method remains useful for distinguishing the source of *E. coli* (resuspension of streambed sediments *vs* soil

surface washoff). Furthermore, values obtained from this method can then be applied in appropriate models to understand the dynamics of FIB in streams, and then provide information for adapted

3 management plans that aim to reduce the health risks to the local populations.

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 Figure 1 - Graphical abstract showing: (1) the main water flow processes (i.e. overland flow, groundwater return flow, blue arrows) and sediment flow components (i.e. resuspension and soil erosion, black arrows) during floods in the Houay Pano catchment; (2) the general principle of the method using fallout radionuclide markers (i.e. ⁷Be and ²¹⁰Pb_{xs}) to estimate *Escherichia coli* load from the two main sources (i.e. streambed resuspension *vs* soil surface washoff); and 3) the main results obtained during the 15 May 2012 storm event (i.e. relative percentage contribution of each process to the total streamflow, values in parentheses).

9 Figure 2 - Geographical location of the Houay Pano catchment, hydro-meteorological equipment,
10 water and streamed sediment sampling points, and land use in 2012.

Figure 3 – Measurements during the 15 may 2012 stormflow event: rainfall intensity (6-mins time step); stream discharge; electrical conductivity (EC) of the stream water; total suspended solids (TSS) concentration; fallout radionuclides (⁷Be and ²¹⁰Pb_{xs}) activity; and *Escherichia coli* (*E. coli*) load of the stream.

Figure 4 – Total suspended solid (TSS) flow of "new" (i.e. recently eroded soil surface aggregates)

16 and "old" (i.e. resuspended sediment) particles estimated using the Matisoff et al. (2005) model;

17 overland flow (OLF) and groundwater (GW) discharge estimated using a tracer-based model (e.g.

18 Ribolzi et al., 1997) estimate of *Escherichia coli* (*E. coli*) flow attached to recently labelled and eroded

19 soil surface particles (new) and to resuspended particles from the streambed (old).

Bricquet J-P, Boonsaner A, Phommassack T, Toan TD (2003): Statistical analysis of long series rainfall data: A regional study in Southeast Asia. In: IWMI (Hrsg.), IWMI Seminar on Scientific Cooperation. IWMI South East Asia, Kasetsart University, Bangkok, Thailand, pp. 83-89 Byappanahalli MN, Fujioka RS (1998) Evidence that tropical soil environment can support the growth of Escherichia coli. Water Sci Technol 38: 171-174 Byappanahalli MN, Nevers MB, Korajkic A, Staley ZR, Harwood VJ (2012) Enterococci in the environment. Microbiol Mol Biol Rev 76: 685-706 Causse J, Billen G, Garnier J, Henri-des-Tureaux T, Olasa X, Thammahacksa C, Latsachak KO, Soulileuth B, Sengtaheuanghoung O, Rochelle-Newall E, Ribolzi O (2015) Field and modelling studies of *Escherichia coli* loads in tropical streams of montane agro-ecosystems Journal of Hydro-environment Research 10.1016/j.jher.2015.03.003 Chan YM, Thoe W, Lee JHW (2015) Field and laboratory studies of Escherichia coli decay rate in subtropical coastal water. Journal of Hydro-environment Research 9: 1-14 Cho KH, Pachepsky YA, Kim JH, Kim JW, Park MH (2012) The modified SWAT model for predicting fecal coliforms in the Wachusett Reservoir Watershed, USA. Water Res 46: 4750-4760 Collins RU, Neal C (1998) The hydrochemical impacts of terraced agriculture, Nepal. Sci Total Environ 212: 233-243 Ekklesia E, Shanahan P, Chua LHC, Eikaas HS (2015) Temporal variation of faecal indicator bacteria in tropical urban storm drains. Water Res 68: 171-181

References

1	Evrard O, Nemery J, Gratiot N, Duvert C, Ayrault S, Lefevre I, Poulenard J, Prat C,
2	Bonte P, Esteves M (2010) Sediment dynamics during the rainy season in
3	tropical highland catchments of central Mexico using fallout radionuclides.
4	Geomorphology 124: 42-54
5	Garzio-Hadzick A, Shelton DR, Hill RL, Pachepsky YA, Guber AK, Rowland R
6	(2010) Survival of manure-borne <i>E. coil</i> in streambed sediment: Effects of
7	temperature and sediment properties. Water Res 44: 2753-2762
8	Genereux DP, Hooper RP (1998): Oxygen and hydrogen isotopes in rainfall-
9	runoff-studies. In: Kendall C , McDonnell JJ (Editors), In Isotopes tracers in
10	catchment hydrology Elsevier Science B.V. , Amsterdam, pp. 319-346
11	Goto DK, Yan T (2011) Effects of land uses on fecal indicator bacteria in the
12	water and soil of a tropical watershed. Microbes and Environments 26: 254-260
13	Gourdin E, Evrard O, Huon S, Lefèvre I, Ribolzi O, Reyss J-L, Sengtaheuanghoung
14	O, Ayrault S (2014a) Suspended sediment dynamics in a Southeast Asian
15	mountainous catchment: Combining river monitoring and fallout radionuclide
16	tracers. Journal of Hydrology 519, Part B: 1811-1823
17	Gourdin E, Evrard O, Huon S, Reyss JL, Ribolzi O, Bariac T, Sengtaheuanghoung O,
18	Ayrault S (2014b) Spatial and temporal variability of Be-7 and Pb-210 wet
19	deposition during four successive monsoon storms in a catchment of northern
20	Laos. Journal of Environmental Radioactivity 136: 195-205
21	Gourdin E, Huon S, Evrard O, Ribolzi O, Bariac T, Sengtaheuanghoung O, Ayrault S
22	(2015) Sources and export of particle-borne organic matter during a monsoon
23	flood in a catchment of northern Laos. Biogeosciences 12: 1073-1089
24	Guiteras R, Levinsohn J, Mobarak AM (2015) Encouraging sanitation investment
25	in the developing world: A cluster-randomized trial. Science: aaa0491
	17

1	Hrdinka T, Novický O, Hanslík E, Rieder M (2012) Possible impacts of floods and
2	droughts on water quality. Journal of Hydro-environment Research 6: 145-150
3	Huon S, de Rouw A, Bonté P, Robain H, Valentin C, Lefèvre I, Girardin C, Le
4	Troquer Y, Podwojewski P, Sengtaheuanghoung O (2013) Long-term soil carbon
5	loss and accumulation in a catchment following the conversion of forest to arable
6	land in northern Laos. Agriculture, Ecosystems & Environment 169: 43-57
7	Ishii S, Sadowsky MJ (2008) Escherichia coli in the environment: Implications for
8	water quality and human health. Microbes and Environments 23: 101-108
9	Kim JW, Pachepsky YA, Shelton DR, Coppock C (2010) Effect of streambed
10	bacteria release on <i>E. coli</i> concentrations: Monitoring and modeling with the
11	modified SWAT. Ecol Model 221: 1592-1604
12	Kirchner JW (2003) A double paradox in catchment hydrology and geochemistry.
13	Hydrological Processes 17: 871-874
14	Lemee R, Rochelle-Newall E, Van Wambeke F, Pizay MD, Rinaldi P, Gattuso JP
15	(2002) Seasonal variation of bacterial production, respiration and growth
16	efficiency in the open NW Mediterranean Sea. Aquat Microb Ecol 29: 227-237
17	Matisoff G, Wilson CG, Whiting PJ (2005) The 7Be/210Pbxs ratio as an indicator
18	of suspended sediment age or fraction new sediment in suspension. Earth
19	Surface Processes and Landforms 30: 1191-1201
20	Muirhead RW, Davies-Colley RJ, Donnison AM, Nagels JW (2004) Faecal bacterial
21	yields in artificial flood events: quantifying in-stream stores. Water Res 38:
22	1215-1224
23	Nguyen TMH, Le TPQ, Garnier J, Janeau J-L, Rochelle-Newall E (in press)
24	Variability of faecal indicator bacteria (FIB) and die-off rates in the Red River,
25	North Vietnam Journal of Hydro-Environment Research
	18

1	Oliver DM, Clegg CD, Heathwaite AL, Haygarth PM (2007) Preferential
2	attachment of Escherichia coli to different particle size fractions of an
3	agricultural grassland soil. Water, Air, and Soil Pollution 185: 369-375
4	Pachepsky YA, Garzio-Hadzick A, Shelton DR, Hadzick ZZ, Hill RL (2011) Survival
5	of E. coli 0157:H12 in creek sediments after inoculation and re-inoculation.
6	International Journal of Environment and Pollution 46: 234-245
7	Pachepsky YA, Shelton DR (2011) Escherichia coli and Fecal Coliforms in
8	freshwater and estuarine Sediments. Critical Reviews in Environmental Science
9	and Technology 41: 1067-1110
10	Pandey PK, Soupir ML, Haddad M, Rothwell JJ (2012) Assessing the impacts of
11	watershed indexes and precipitation on spatial in-stream <i>E. coli</i> concentrations.
12	Ecological Indicators 23: 641-652
13	Patin J, Mouche E, Ribolzi O, Chaplot V, Sengtahevanghoung O, Latsachak KO,
14	Soulileuth B, Valentin C (2012) Analysis of runoff production at the plot scale
15	during a long-term survey of a small agricultural catchment in Lao PDR. Journal
16	of Hydrology 426: 79-92
17	Ribolzi O, Moussa R, Gaudu JC, Valles V, Voltz M (1997) Stream water regime
18	change at autumn recharge on a Mediterranean farmed catchment using a
19	natural tracer. Comptes Rendus De L Academie Des Sciences Serie Ii Fascicule a-
20	Sciences De La Terre Et Des Planetes 324: 985-992
21	Ribolzi O, Andrieux P, Valles V, Bouzigues R, Bariac T, Voltz M (2000)
22	Contribution of groundwater and overland flows to storm flow generation in a
23	cultivated Mediterranean catchment. Quantification by natural chemical tracing.
24	Journal of Hydrology 233: 241-257
	19

1	Ribolzi O, Cuny J, Sengsoulichanh P, Mousques C, Soulileuth B, Pierret A, Huon S,
2	Sengtaheuanghoung O (2011a) Land use and water quality along a mekong
3	tributary in Northern Lao PDR. Environ Manage 47: 291-302
4	Ribolzi O, Patin J, Bresson LM, Latsachack KO, Mouche E, Sengtaheuanghoung O,
5	Silvera N, Thiebaux JP, Valentin C (2011b) Impact of slope gradient on soil
6	surface features and infiltration on steep slopes in northern Laos.
7	Geomorphology 127: 53-63
8	Rochelle-Newall EJ, Nguyen TMH, Le TPQ, Sengtaheuanghoung O, Ribolzi O
9	(2015) A short review of faecal indicator bacteria in tropical aquatic ecosystems:
10	knowledge gaps and future directions. Frontiers in Microbiology: Aquatic
11	microbiology 6: 308
12	Saqalli M, Jourdren M, Maestripieri N, Guillerme S, Maire E, Soulileuth B,
13	Latsachach K, Sounyafong P, Tammahuxsa L, Sengtaheuanghoung O, Ribolzi O,
14	Becerra S (2015) Backward waters, modern waters: Perception-Based Regional
15	Mapping territory uses and water-related sanitary stakes in Luang Phabang area
16	(Lao PDR). Applied Geography 60: 184-193
17	Soil Survey Staff (1999) Soil Taxonomy. A Basic System of Soil Classification for
18	Making and Interpreting Soil Surveys. USDA-NRCS
19	Stumpf CH, Piehler MF, Thompson S, Noble RT (2010) Loading of fecal indicator
20	bacteria in North Carolina tidal creek headwaters: Hydrographic patterns and
21	terrestrial runoff relationships. Water Res 44: 4704-4715
22	Sturma A, Becerra S (2012): L'assainissement à Mayotte à quel prix ? Entre
23	vulnérabilité institutionnelle et sociale. In: Raynaud L , Poirot-Delpech S
24	(Editors), Par-delà le local et le global: Pour une socio-anthropologie de
25	l'environnement. L'Harmattan, Paris, pp. 207-223
	20

	1	Suter E, Juhl A, O'Mullan G (2011) Particle association of Enterococcus and tota	ıl
	2	Bacteria in the Lower Hudson River Estuary, USA. Journal of Water Resource a	nd
	3	Protection 3: 715-725	
	4	Taylor A, Blake WH, Couldrick L, Keith-Roach MJ (2012) Sorption behaviour of	1
	5	beryllium-7 and implications for its use as a sediment tracer. Geoderma 187-	
	6	188: 16-23	
	7	Taylor A, Blake WH, Smith HG, Mabit L, Keith-Roach MJ (2013) Assumptions ar	ıd
	8	challenges in the use of fallout beryllium-7 as a soil and sediment tracer in rive	r
	9	basins. Earth-Science Reviews 126: 85-95	
1	0	Unanue M, Ayo B, Azúa I, Barcina I, Iriberri J (1992) Temporal variability of	
1	1	attached and free-living bacteria in coastal waters. Microb Ecol 23: 27-39	
1	2	Vigiak O, Ribolzi O, Pierret A, Sengtaheuanghoung O, Valentin C (2008) Trappin	١g
1	3	efficiencies of cultivated and natural riparian vegetation of northern Laos.	
1	4	Journal of Environmental Quality 37: 889-897	
1	5	WHO 2012: World Health Organisation global data repository, World Health	
1	6	Organisation, <u>http://apps.who.int/ghodata/</u>	
1	7	WorldBank 2009: Economic impacts of sanitation in Lao PDR	
1	8	Yakirevich A, Pachepsky YA, Guber AK, Gish TJ, Shelton DR, Cho KH (2013)	
1	9	Modeling transport of Escherichia coli in a creek during and after artificial high	1-
2	20	flow events: Three-year study and analysis. Water Res 47: 2676-2688	
2	21	Ziegler AD, Sutherland RA, Giambelluca TW (2000) Partitioning total erosion o	n
2	22	unpaved roads into splash and hydraulic components: The roles of interstorm	
2	23	surface preparation and dynamic erodibility. Water Resour Res 36: 2787-2791	
2	24		
			21









