2022-10

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10.1016/j.earscirev.2022.104188
Earth-Science Reviews
Elsevier

Understanding the Complexity of Sediment Residence Time in Rivers: Application of Fallout Radionuclides (FRNs)

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Abstract

Riverine sediments play an important role in the healthy functioning of river ecosystems as they provide nutrients and a connectivity signal throughout the catchment sediment cascade. However, excess sediment supply to rivers can have several detrimental impacts on water quality, availability and ecology. The application of catchment management practices requires a comprehensive understanding of both spatial and temporal sediment dynamics to tackle point and diffuse river pollution. While the sources, pathways and fate of eroded sediment in river systems have been widely studied, temporal dynamics have received less attention, mainly due to the complexity of the processes and the lack of methods available to assess these dynamics. This contribution reviews the application of Fallout Radionuclides (FRNs) as sediment residence time tracers. We explore their suitability as sediment chronometers in rivers, the relevance of sediment residence time to sediment budgeting, and discuss the current models that have been employed to determine sediment residence time in river systems. Our review also identifies the challenges, opportunities and the future research needs for a comprehensive application of FRNs to evaluate sediment residence time. In evaluating approaches to sediment residence time, we have summarised several pitfalls requiring consideration and identified avenues for further research. For instance, attention should be given to sorption behaviour when using ^{7}Be and ^{137}Cs as residence time tracers in rivers under changing environmental conditions; particle size effects; activity concentration dilution by mixing of newly tagged ^{7}Be sediment with ^{7}Be-poor sediment from older or different sources; source controls on ^{7}Be delivery to rivers; and the influence of direct fallout into stream channels. Finally, further research is needed to assess the influence of environmental and anthropogenic factors on sediment residence time such as land use, topography, flow regimes, soil type, soil erosion measures and climate change, all of which have important implications from a catchment management perspective.

Keywords

Sediments; Rivers; Residence Time; Fallout Radionuclides, Contaminants; Sediment Storage.

CRediT author statement

E. Munoz-Arcos: Conceptualisation, Formal Analysis, Investigation, Resources, Data Curation, Writing – Original Draft, Visualisation, Funding acquisition; G. E. Millward: Conceptualisation, Writing – Review and Editing, Supervision; C. C. Clason: Writing – Review and Editing, Supervision; C. Bravo-Linares: Writing – Review and Editing, Supervision; W. H. Blake: Conceptualisation, Writing – Review and Editing, Supervision, Project administration, Funding acquisition.

1 Introduction.

Riverine sediments, typically particles < 2 mm in diameter, play an important role in the healthy functioning of river ecosystems since they provide nutrients and contribute to habitat quality (Kemp et al. 2011; Jones et al. 2012; Wharton et al. 2017). However, human activities such as reservoir and dam construction, land use changes, deforestation, mining activities and urbanisation have altered sediment fluxes in river systems resulting in enhanced, or reduced, sedimentation and contamination (Walling and Fang 2003; Owens et al. 2005; Syvitski and Kettner 2011; Wohl 2015). Excessive sediment supply into rivers has detrimental impacts on water quality (e.g. turbidity) which causes sedimentation in river channels, reservoirs and estuaries affecting aquatic habitats such as salmonids spawning gravels (Kemp et al. 2011). In addition, silts and clays are geochemically active and consequently are responsible for the transport of contaminants, including trace metals, phosphorus, Polycyclic Aromatic Hydrocarbons (PAHs), pesticides, radionuclides and Polychlorinated Biphenyls (PCBs) which have high sorptive affinity for fine-grained sediment particles (Owens et al. 2005; Wohl 2015; Alewell et al. 2017; Rügner et al. 2019; Owens 2020).

Fluxes of sediments in fluvial systems transport materials between hillslopes and floodplains, riparian zones, the active channel, and the hyporheic and groundwater zone providing a connectivity signal within the river landscape. However, it is well known that rivers are efficient in trapping sediments, and consequently only a fraction of the eroded sediment may be transported to the basin outlet (Walling 1983; Fryirs 2013). Therefore, sedimentation in river corridors drives channel morphology, and enhanced sedimentation in river channels can have several implications including bed fining, channel narrowing allowing vegetation encroachment, altered bedform type or dimensions, bed aggradation, altered channel planform and enhanced floodplain sedimentation (Wohl 2015). Furthermore, sediments can have adverse impacts on channel-beds, such as the filling of interstitial spaces between coarser streambed sediment (also known as streambed colmation) which provides habitats for aquatic insects and fish eggs, and facilitates hyporheic exchange (Kemp et al. 2011; Jones et al. 2012; Wharton et al. 2017). Therefore, understanding when and how streambed colmation takes place and the quantity of sediment storage, and the dwell time, are fundamental to the assessment of the environmental impacts of upstream erosion and sediment pulses into rivers. Hence, quantifying the timescales of sediment transfer throughout a river system is critical for understanding both river basin sediment dynamics and the fate of adsorbed contaminants. This timescale is represented by the period that sediments spend within

river basins, which can encompass days to months (Matisoff et al. 2005; Le Gall et al. 2017), decades (Wallbrink et al. 2002) and millennia or longer (Hoffmann 2015).

1.1 Definitions

Finding a consensus on the meaning of sediment residence times in rivers is problematic since definitions are often dependent on the methodology. For example, Whipp & Ehlers (2019) define sediment residence time as the "time sediments remain within the catchment as part of a population of minerals that could be dated in a random sample, assuming no long-term sediment storage in the catchment". Alternatively, Skalak & Pizzuto (2010) define sediment residence time as "the time required to remove a mass of sediment equal to the total volume in storage". Other authors make a distinction between terms such as sediment transit time and sediment residence time. For example, Gellis et al. (2019) define sediment transit time as "the time it takes for sediment to travel from a starting point in the watershed to an endpoint where sediment leaves the area of interest", whereas residence time refers to "the mean time sediment particles spend in a storage reservoir". Gellis et al. (2019) also noted that these terms can depend on the spatial scale used. Thus, transit time can encompass the time from when sediment is released to when it enters the river channel (e.g. Slattery et al. 2002) or the time that sediment spends in all storage reservoirs i.e. hillslopes, channel-bed and floodplains (e.g. Hoffmann 2015). On the other hand, residence time can span sediments deposited at a specific storage unit i.e. channel-bed (e.g. Skalak and Pizzuto 2010) or also include the suspended fraction (Evrard et al. 2010; Smith et al. 2014). In addition, Matisoff et al. (2005) used the term sediment age to refer to the time between particles receiving their radioactive tags to when they are sampled in the river as suspended, bed-stored, or estuarine sediment.

For consistency throughout this text with regards to defining sediment temporal dynamics the terminology outlined in Figure 1 will be used. Once sediment enters a stream channel it may be transported either through, or stored temporarily in, different riverine units (e.g. river bed, bars, lateral deposits and short-term floodplain deposits). The time this sediment spends in these temporary units is referred to here as its storage time. Sediments from these storage units can be released during stormflow events contributing to the suspended load which also comprises sediments from upstream sources. Therefore, the time encompassing from upstream sediment release to the time spent in riverine temporary storage and in transport is referred to here as the residence or transit time. Finally, the time that a sediment particle has been resident in a long-term storage unit towards the outlet of a river is given by its age (e.g. long-term floodplain deposits, lacustrine and estuarine

deposits). Therefore, sediment age spans the time that a particle has been in temporary storage, transport and deposition in the long-term storage unit. Although it is recognised that particles may be reworked during storage in these long-term deposition units, they are rarely moved to upstream catchment areas (e.g. during a tidal surge).

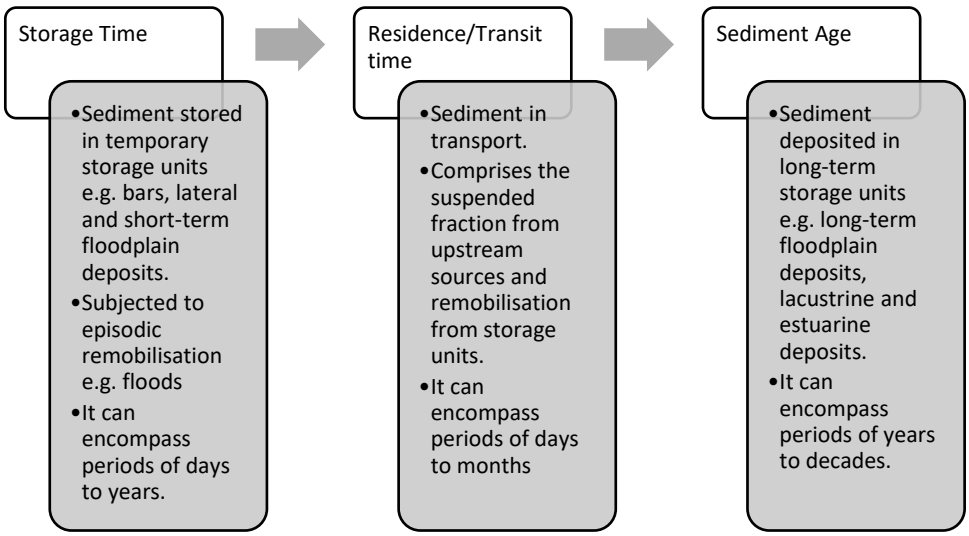


Figure 1. Relationships between the different concepts used here to describe the temporal dynamics of sediment storage and transport in rivers. The storage time refers to the time that sediment spends in a short-term deposition unit, residence/transit time refers to the time that sediment spends in temporary storage and in transport throughout the river system, and sediment age refers to the time that sediment has remained stored in long-term deposition units.

1.2 Methods to determine sediment residence time

Several methods have been used to assess sediment temporal dynamics in different catchment compartments (Voepel et al. 2013; Hoffmann 2015; Sutfin and Wohl 2019; Carretier et al. 2020). Here we focus our discussion on riverine processes that encompass temporary storage and transport i.e. sediment residence/transit time. To date, a variety of methods have been developed to determine sediment residence time. For example, Wang Jin et al. (2015) estimated fine-grained sediment residence time using the post-earthquake rate of sediment export (measured using daily suspended sediment discharges) triggered by landslide sediment mobilised by the Wenchuan earthquake, China. Furthermore, Voepel et al., (2013) determined a bed elevation time series using

sonar transducers and LiDAR to evaluate empirical sediment residence times. Radionuclides offer the possibility of determining both sediment age and residence times. For instance, Uranium-series isotopes have been used to document the time-dependence of weathering ages (Dosseto et al. 2014; Suresh et al. 2014) and ^{14}C dating has been applied to estimate the mean sediment age of floodplain sediments (Sutfin and Wohl 2019). Short-lived radionuclides (e.g. ^{7}Be, ^{210}Pb, ^{137}Cs and ^{234}Th) have also been used to assess sediment travel distances, sediment age and sediment residence time in a variety of landscapes (Olsen et al. 1986; Dominik et al. 1987; Wieland et al. 1991; Vogler et al. 1996; Bonniwell et al. 1999; Feng et al. 1999; Ciffroy et al. 2003; Forster et al. 2009). In lakes, Wieland et al. (1991) modelled sediment residence times using ^{7}Be and ^{210}Pb fluxes in Lake Zurich, Switzerland, and Vogler et al. (1996) estimated sediment residence times in Lake Constance for the total radionuclide inventories of ^{234}Th and ^{7}Be. In estuaries, Olsen et al. (1986) assessed water column removal rates and residence time of ^{7}Be in the James River Estuary, USA, and Ciffroy et al. (2003) determined sediment residence time of suspended particles using ^{7}Be budgets in the turbidity maximum zone of the Loire estuary, France. The application of these radionuclides as sediment residence time tracers in lakes, coastal and estuarine zones has been discussed elsewhere (Dominik et al. 1989; Steinmann et al. 1999; Feng et al. 1999; Baskaran 2001; Baskaran and Swarzenski 2007; Forster et al. 2009; Saari et al. 2010). For extensive reviews on the use of radionuclides in these environments the reader is directed to works from Du et al. (2011); Kaste et al. (2002); Kaste & Baskaran (2011); Waples et al. (2006).

1.3 Rationale and objectives of this review

Increasing attention given to the temporal dynamics of fine-grained sediment in river systems (Du et al. 2011; Walling 2013; Matisoff 2014; Collins et al. 2020) raises the need for a revision of the use of medium and short-lived radionuclides as chronometers, the methods applied, the identification of potential limitations, and implications from a catchment management perspective. If the temporal dynamics of suspended and channel-bed sediments are better understood, then the timeframe when catchment management practices become effective can be better constrained, or otherwise, to justify the difficulty to implement mitigation plans over short timescales. This is a key aspect to consider when developing mitigation actions to solve river basin point and diffuse pollution, and the subsequent ecological consequences that fine sediment storage and remobilisation can have on rivers. In this context, this review primarily addresses the use of short-lived radionuclides to assess sediment residence time in rivers to: 1) assess the existing literature regarding sediment

residence time with a focus on the use of Fallout Radionuclide (FRNs) tracers in river systems; 2) provide an assessment of the main sediment residence time models applied to date; 3) discuss the assumptions and challenges of these different methods; and 4) identify future research needs for a comprehensive evaluation of sediment residence time in river systems.

2 River sediment budgeting and its relevance to channel storage and residence time.

Sediment dynamics in river systems involve complex processes, and their quantitative assessment faces many uncertainties. River sediment budgeting has become increasingly used to overcome this issue because it provides an understanding of the sediment mobilisation, transport, storage and yield (Walling and Collins 2008). River sediment budgets can be understood, in simplistic terms, as the mass balance between the sediment sources, deposition areas and outputs. One of the most significant findings from budgetary studies is the importance, and magnitude, of sediment storage in rivers. From the total amount of sediment produced (i.e. eroded) in upland surfaces (i.e. sources) only a fraction makes its way to the basin outlet (i.e. the sediment yield). This discrepancy has been named the 'sediment delivery problem' by Walling (1983) and it has been argued that various sediment storage mechanisms operating within a catchment may explain this discrepancy (Trimble 1983; Walling 1983; Fryirs 2013). Since then, an expanding body of research has been carried out to explain and disentangle the mechanisms, pathways and fates of eroded sediment within a river basin, however, the temporal dynamics of these processes have received less attention due in part to the complexity of the problem and the lack of available methods.

An issue related to sediment budgeting concerns interpretation of the delivery ratio term (i.e. the ratio of the inputs to the output) (Walling 1983; Parsons 2012). For example, substantial variability at different temporal scales (e.g. between stormflow events, seasons or years) has been found in sediment delivery ratios within a catchment, the so-called 'temporal lumping' as described by Walling (1983). Therefore, it is necessary to accompany channel sediment budgeting with an improved understanding and evaluation of the sediment residence times within different storage units to avoid temporal biases and uncertainties in the interpretation of the sediment delivery ratio.

In many catchments, sediments spend significant time stored in riverine compartments, and delivery is therefore controlled by storage and sporadic remobilisations over various timescales. These storage units can be defined as transient, short-lived landforms, such as bars, lateral deposits and the streambed, that are frequently reworked during stormflow events where they play a key role in the (dis)connectivity of the catchment sediment cascades (Fryirs 2013). Thus, river channel sediment budgeting becomes a key tool in the assessment of sediment reworking/exchange magnitudes/quantities in rivers subjected to episodically driven sediment remobilisation and deposition for a comprehensive understanding of the sediment dynamics in these temporary storage units.

3 Suitability of medium and short-lived radionuclides as sediment residence time tracers.

Medium and short-lived radionuclides (i.e. ^{210}Pb, ^{137}Cs, ^{7}Be and ^{234}Th) have been extensively used as soil and sediment tracers to assess their redistribution, deposition rates and residence time (Feng et al. 1999; Blake et al. 2002; Waples et al. 2006; Mabit et al. 2008; Blake et al. 2009; Mabit et al. 2013; Taylor et al. 2013; Walling 2013; Mabit et al. 2014; Taylor et al. 2019). However, it is not the aim of this review to examine the current knowledge of radionuclides as soil erosion and sediment tracers, for which the reader is directed to reviews by Mabit et al. (2014, 2013); Matisoff (2014); Matisoff and Whiting (2011); Parsons and Foster (2011); Taylor et al. (2013); and Walling (2013). Moreover, applications of ^{234}Th as a tracer of sediment dynamics in freshwater systems (Waples et al. 2006) have mostly concerned lakes (Dominik et al. 1989; Vogler et al. 1996; Waples et al. 2004). Some studies have determined ^{234}Th in river sediments and water despite the typically low activity concentrations (Morris et al. 1994; Waples et al. 2003) but no sediment residence time studies using this radionuclide in river systems have been found. For this reason, we focus our discussion on Fallout Radionuclides (FRNs).

^{137}Cs ($t_{1/2}$ = 30.17 years) is an anthropogenic radionuclide that was released to the stratosphere as a result of atmospheric thermonuclear weapon testing in the 1950s – 1980s, in addition there have been releases during nuclear accidents such as the 1986 Chernobyl and the 2011 Fukushima disasters. Before subsequent deposition on the Earth's surface, ^{137}Cs circulated globally and was washed out by precipitation patterns (Ritchie and McHenry 1990). ^{210}Pb ($t_{1/2}$ = 22.2 years) is of geogenic origin and is a natural decay product within the ^{238}U decay

series derived from the decay of the inert gas ^{222}Rn ($t_{1/2}$ = 3.8 days) which derives from its parent ^{226}Ra ($t_{1/2}$ = 1622 years). The ^{210}Pb generated in situ by decay of ^{226}Ra is termed supported ^{210}Pb and is in equilibrium with ^{226}Ra. However, ^{222}Rn diffuses to the atmosphere and undergoes a series of short-lived decays to ^{210}Pb which may adsorb to aerosols and is delivered to the landscape by wet and dry fallout. Fallout ^{210}Pb is termed unsupported, or excess ^{210}Pb ($^{210}Pb_{ex}$). ^{7}Be ($t_{1/2}$ = 53.3 days) is a cosmogenic radionuclide produced by cosmic ray spallation of nitrogen and oxygen in the stratosphere and troposphere and it is delivered to the earth's surface through wet and dry deposition.

An advantage of using ^{137}Cs, $^{210}Pb_{ex}$ and ^{7}Be as sediment chronometers, is that the contrasting half-lives that these radionuclides possess can be used to model sediment residence time from days to decades in different catchment compartments (Wallbrink et al. 2002; Matisoff et al. 2005; Le Cloarec et al. 2007; Gellis et al. 2019). Additionally, naturally and continuously produced ^{7}Be and ^{210}Pb radioisotopes can be used at many sites in the world widening their application (Wallbrink et al. 2002; Matisoff et al. 2005; Le Cloarec et al. 2007; Jweda et al. 2008; Evrard et al. 2010; Smith et al. 2014). The basis for the use of these radionuclides as tracers is their ability to rapidly and strongly adsorb onto soil and sediment particles following both wet and dry deposition (Welp and Brümmer 1999; Du et al. 2011; Matisoff and Whiting 2011; Taylor et al. 2012; Matisoff 2014; Singleton et al. 2017; Ryken et al. 2018) with reported partition coefficients, K_d, of the order 10^5 (Olsen et al. 1986; Hawley et al. 1986; Dominik et al. 1987; Van Hoof and Andren 1989; You et al. 1989; Steinmann et al. 1999; Kaste et al. 2002; Jweda et al. 2008). However, sorption behaviour of ^{137}Cs and ^{7}Be have been questioned, especially in changing environments through the catchment sediment cascade which can alter sediment residence time estimations (Parsons and Foster 2011; Taylor et al. 2012, 2013; Ryken et al. 2018). For example, partitioning of ^{7}Be and ^{137}Cs have been reported to be strongly influenced by pH and salinity (You et al. 1989; Kaste et al. 2002; Giannakopoulou et al. 2007; Hong et al. 2011; Kaste and Baskaran 2011; Taylor et al. 2013), therefore, its application in estuarine environments as a sediment particle tracer is challenging and requires case-by-case validation.

4 Models to evaluate sediment residence time in river systems.

Models have been developed and tested to evaluate sediment residence time in rivers within a variety of landscapes, catchment sizes and regions across the world (see Table 1 and Figure 2). Application of radionuclide

mass balances to assess sediment residence times began in late 1980s using data from the Rhone catchment, Switzerland (Dominik et al. 1987). However, it was not until late 1990s that other authors employed different methodologies to determine/model sediment residence times. For example, Wallbrink et al. (1998) determined the proportional contribution from three potential sources: cultivated and uncultivated land and subsoil material from gullies and channel banks. Then, applying a mixing model with various concentrations of $^{210}Pb_{ex}$ corrected as a function of radioactive decay in the channel, they found that the mean residence time of fine-grained material was 10 ± 5 years (Table 1). However, evidence that sediment residence times in the Murrumbidgee river, Australia could be of the order of weeks to months was noticed from changes in the mean ^{137}Cs activity concentrations of suspended sediments between flood and low-flow conditions, and the presence of ^{7}Be activity in sediments from flood water. Bonniwell et al. (1999) used $^{7}Be/^{210}Pb$ and $^{210}Pb/^{137}Cs$ ratios to assess the fraction of new sediment in suspension, residence times and transport distances. In their study, the basis for the use of $^{7}Be/^{210}Pb$ ratios was stablished because it corrected for the relative sorption and enrichment effects resulting from variations in grain size and particulate matter composition, and by using $^{210}Pb/^{137}Cs$ ratios they accounted for variations in mineralogy and source area activity concentrations. Residence times in sediments from the channel of River Gold Fork, USA, ranged from 1.6 to 103 days (Table 1) from the upper to the lower part of the catchment. In addition, Wallbrink et al. (2002) determined residence times using $^{210}Pb_{ex}$ activity concentrations in river bed sediments from the Brisbane and Logan rivers, Australia. They assumed that increases in $^{210}Pb_{ex}/^{137}Cs$ ratios in sediments occurred because of primarily direct input of fresh flux of $^{210}Pb_{ex}$ to the sediments in the river channel. This was supported because no such higher ratios were measured at potential soil erosion sources within the catchment. Using the initial inventory of $^{210}Pb_{ex}$ within the mobile layer of river sediment derived from the catchment erosion, $^{210}Pb_{ex}$ inventories at the time of sample collection and the inventory of $^{210}Pb_{ex}$ which occurs when depositional flux decay within the sediment profile reach equilibrium, they calculated residence times for deposited sediments in rivers Brisbane and Logan of 0 – 21 and 0 – 9 years (Table 1), respectively. This model was also applied by Douglas et al. (2009) in the Maroochy River estuary, Australia. In their study, modelled sediment residence times averaged between 1 – 28 years.

254 **Table 1.** Studies that have used FRNs to model/determine sediment residence time in the literature.

Site location	Site area/length	Radionuclide(s)	Modelling approach	Sediment residence time	Reference
Rhone Watershed, Switzerland.	5,220 km^2	^{137}Cs, ^{210}Pb and ^{7}Be	Two-box model.	800 – 1,400 y; 1 – 220 d	Dominik et al. (1987)
Murrumbidgee River, Australia.	13,500 km^2	$^{210}Pb_{ex}$	$^{210}Pb_{ex}$ source decay as a function of in-channel residence time	10 y	Wallbrink et al. (1998)
Gold Fork River, USA.	389 km^2	^{137}Cs, ^{210}Pb and ^{7}Be	Normalised activity ratios	1.6 – 103 d	Bonniwell et al. (1999)
Brisbane and Logan River catchments. Australia.	13,600 and 3,076 km^2	^{137}Cs and ^{210}Pb	Comparison of catchment soil erosion inventories to riverbed sediment inventories	0 – 21 and 0 – 9 y	Wallbrink et al. (2002)
Old Woman Creek, Weeks bay and South Slough, USA.	69.5, 24.3 and 73 km^2	^{7}Be and $^{210}Pb_{ex}$	$^{7}Be/^{210}Pb_{ex}$ ratio	50 – 300 d	Matisoff et al. (2005)
River Seine basin, France.	7 to 65,700 km^2	^{137}Cs, ^{210}Pb and ^{7}Be	Two-box model.	115 – 307 d (river box); 4,859 – 31,192 y (soil box)	Le Cloarec et al. (2007)
Clinton River, Southeast Michigan, USA.	1,980 km^2	^{7}Be and $^{210}Pb_{ex}$	Single-box model	0.2 – 2.1 d (^{7}Be), 0.5 – 8.6 d (^{210}Pb)	Jweda et al. (2008)
Maroochy river estuary, Australia.	630 km^2	^{137}Cs and ^{210}Pb	Comparison of catchment soil erosion inventories to river bed sediment inventories	1 – 28 y	Douglas et al. (2009)
Cointzio catchment, México.	3 to 12 km^2	^{137}Cs, ^{210}Pb and ^{7}Be	Two-box balance model.	50 – 200 d (river box); 5,000 – 23,000 y (soil box)	Evrard et al. (2010)
		^{7}Be and $^{210}Pb_{ex}$	$^{7}Be/^{210}Pb_{ex}$ ratio	101 – 163 d	
White, West rivers and Mink Brook; USA.	29 to 319 km^2	^{7}Be and $^{210}Pb_{ex}$	Stacked reservoirs model.	4 to > 300 d	Gartner et al. (2012)
Pheasant sub-catchment, USA	12.4 km^2	^{7}Be and $^{210}Pb_{ex}$	$^{7}Be/^{210}Pb_{ex}$ ratio	40 – 319 d	Huisman and Karthikeyan (2012)
Pheasant sub-catchment, USA.	12.4 km^2	^{7}Be and $^{210}Pb_{ex}$	$^{7}Be/^{210}Pb_{ex}$ ratio	9 – 318 d	Huisman et al. (2013)
River Tamar basin, UK.	38 to 219 km^2	^{137}Cs, ^{210}Pb and ^{7}Be	Two-box model.	185 – 368 d (river box); 77,000 – 48,000 y (soil box)	Smith et al. (2014)
Pheasant Valley Catchment, USA.	50 km^2	^{7}Be and $^{210}Pb_{ex}$	$^{7}Be/^{210}Pb_{ex}$ ratio	123 – 322 d	Lamba et al. (2015)
Loroux catchment, France.	25 km^2	^{7}Be and $^{210}Pb_{ex}$	$^{7}Be/^{210}Pb_{ex}$ ratio	20 – 200 d	Le Gall et al. (2017)
Midwestern USA rivers	6.8 to 5,893 km^2	$^{210}Pb_{ex}$	Age of surface derived sediments	0 – 174 d	Gellis et al. (2017)
		^{7}Be and $^{210}Pb_{ex}$	$^{7}Be/^{210}Pb_{ex}$ ratio	61 – 282 d	
White clay creek, USA.	7.25 km^2	^{7}Be and $^{210}Pb_{ex}$	$^{7}Be/^{210}Pb_{ex}$ ratio	22 – 110 d	Karwan et al. (2018)
Orge river catchment	900 km^2	^{7}Be and $^{210}Pb_{ex}$	$^{7}Be/^{210}Pb_{ex}$ ratio	18 – 140 d	Froger et al. (2018)
Clinton River, Southeast Michigan, USA	1,946 km^2	$^{210}Po_{ex}$ and $^{210}Pb_{ex}$	Single-box model	0.3 – 3.9 d (^{210}Pb), 0.9 – 13.4 d (^{210}Po)	Baskaran et al. (2020)
Walnut creek	52.6 km^2	^{7}Be and $^{210}Pb_{ex}$	Age of surface derived sediments	44 – 205 d (^{7}Be), 1 – 58 y (^{210}Pb)	Gellis et al. (2019)
Ducktrap River, USA.	9 km reach	^{7}Be	CIA (Constant Initial Activity) aging model	0 to > 160 d	Fisher et al. (2010)
South River, USA	37 km reach	^{14}C, ^{137}Cs and $^{210}Pb_{ex}$	Reservoir theory model	1 – 60 y	Skalak and Pizzuto (2010)

255

Figure 2. Location of the study sites that have measured FRNs to derive sediment residence times.

A different approach was adopted by Skalak & Pizzuto (2010) who applied several radiometric dating methods (^{14}C, ^{210}Pb and ^{137}Cs) to infer the distribution of ages of sediment stored within Fine-Grained Channel Margin (FGCM) deposits of the South River, USA. By applying the reservoir theory[1] to the population of ages dated on the deposits, they found a sediment residence time of 1.75 years with a very small portion of sediments with storage times of the order of decades (60 years). One of the main benefits of this approach is that it provides both residence time and a distribution of ages. Similarly, Fisher et al. (2010) studied sediment storage times using ^{7}Be coupled with a Constant Initial Activity (CIA) sediment aging model to assess transitional bedload storage times in bars associated with in-channel obstructions (large wood debris and boulders). They identified two dominant transport regimes with differing storage times: 1) transport-limited reaches with storage times > 100 days associated with channel obstructions, and 2) supply-limited reaches associated with steeper gradients and greater stream power capable of mobilising fine-grained sediments from channel obstructions with generally < 100 days of sediment storage. Although they addressed successfully the bed-storage time problem

[1] Reservoir theory describes the change in abundance of a substance in a reservoir in terms of its inputs and outputs through the reservoir. This reservoir can be defined as any volume enclosed by a boundary such as a lake, an ecosystem or a soil (Eriksson 1971; Mudd and Yoo 2010).

in a study of a 9 km reach of the Ducktrap River (USA), certain precautions were noted with regards to the application of ^{7}Be when using CIA aging model:

1) ^{7}Be activity concentration dilution by sediment depleted in ^{7}Be through long-term in-channel residence times (greater decay) and/or frequent landslides or bank collapses which may supply ^{7}Be-depleted sediments diminishing initial activities used to feed the CIA model, and
2) ^{7}Be sediment enrichment by fresh tagging from atmospheric inputs into submerged bars at low-flow conditions which may overprint the inherited ^{7}Be signal and thus increase initial activities of ^{7}Be.

More recently, a novel technique to determine sediment residence time in riverbed sediment at various depths was developed by Gartner et al. (2012). In their study, the channel bed is divided into a vertically stacked series of reservoirs based on the assumption that residence times in stream beds would likely increase with depth, yielding not a single residence time but rather a distribution of residence times varying with depth. Applying this model in cores taken from the White and West Rivers (Vermont) and the Mink Brook (New Hampshire), USA, they found sediment residence times of about 2 months in unregulated rivers while residence times exceed 6 months in regulated rivers. In addition, based on ^{7}Be and $^{210}Pb_{ex}$ depth profiles, they suggested two mechanisms of bed material exchange: filtration; and scour and fill. For those core profiles that showed systematic decline in FRN activity a filtration mechanism was inferred, whereas scour and fill was linked to an inconsistent variation in activity concentrations with depth.

As shown in Table 1, different models have been developed to assess sediment residence time in rivers, but relatively few have been applied more than once and in different settings. These models are reviewed in more depth in the following sections.

4.1 Single box model

The single box model described here uses FRNs to assess sediment residence times in river systems (Jweda et al. 2008; Baskaran et al. 2020). Particle residence times are obtained by means of mass balances of particulate $^{210}Pb_{ex}$ (Eq. 1) and ^{7}Be (Eq. 2) to obtain sediment resuspension rates as follows (Figure 3):

$$\Psi_{Pb}^{d}A_{Pb}^{d} + \lambda_{Pb}A_{Rn}^{p} + I_{Pb}^{rp} + \frac{R_{Pb}A_{Pb}^{r}}{H} = O_{Pb}^{rp} + \lambda_{Pb}A_{Pb}^{p} + \frac{R_{Pb}A_{Pb}^{s}}{H} \quad \text{Eq. 1}$$

and

$$\Psi_{Be}^{d} A_{Be}^{d} + I_{Be}^{rp} + \frac{R_{Be} A_{Be}^{r}}{H} = O_{Be}^{rp} + \lambda_{Be} A_{Be}^{p} + \frac{R_{Be} A_{Be}^{s}}{H} \quad \text{Eq. 2}$$

Where I_{Pb}^{rp} and I_{Be}^{rp} are the input fluxes of particulate $^{210}Pb_{ex}$ and ^{7}Be (dpm cm^{-3} yr^{-1}), respectively, O_{Pb}^{rp} and O_{Be}^{rp} are the output fluxes of particulate $^{210}Pb_{ex}$ and ^{7}Be (dpm cm^{-3} yr^{-1}), respectively, H is the mean depth of the water column (cm), A_{Rn}^{p} is the activity of ^{222}Rn (dpm cm^{-3}) adsorbed onto particulate matter, A_{Pb}^{d} and A_{Be}^{d} are the activities of dissolved ^{210}Pb and ^{7}Be (dpm cm^{-3}), A_{Pb}^{p} and A_{Be}^{p} are the activities of particulate $^{210}Pb_{ex}$ and ^{7}Be (dpm cm^{-3}), A_{Pb}^{r} and A_{Be}^{r} are the activities of $^{210}Pb_{ex}$ and ^{7}Be in the resuspended material (dpm g^{-1}), A_{Pb}^{s} and A_{Be}^{s} are the activities of $^{210}Pb_{ex}$ and ^{7}Be in the settling particulate matter collected in sediment traps (dpm g^{-1}), Ψ_{Pb}^{d} and Ψ_{Be}^{d} are the first-order removal rate constants of $^{210}Pb_{ex}$ and ^{7}Be (y^{-1}) from the dissolved phase onto particles, and R_{Pb} and R_{Be} are sediment resuspension rates using $^{210}Pb_{ex}$ and ^{7}Be (g cm^{-2} yr^{-1}), respectively.

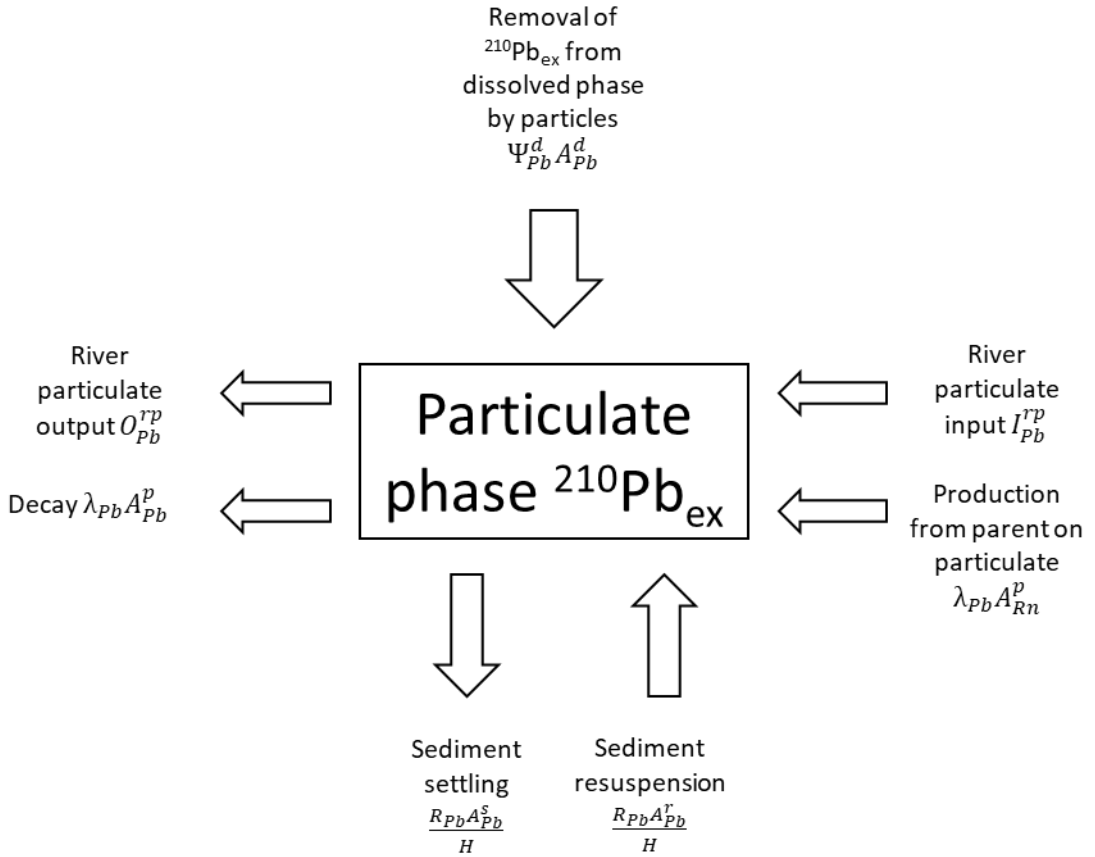


Figure 3. Diagram of the single box model illustrating the sources and sinks of particulate ^{210}Pb. Modified from Jweda et al. (2008). The same model concept applies to ^{7}Be sources and sinks but without the input from the decay of a parent radionuclide.

Assuming that $^{210}Pb_{ex}$ and ^{7}Be activities of the upper layer of bottom sediment are equal to those of resuspended sediment and that the production term from ^{222}Rn is negligible, then sediment resuspension rates can be obtained from mass balance equations of $^{210}Pb_{ex}$ (Eq. 3) and ^{7}Be (Eq. 4):

$$R_{Pb} = \frac{H\left(\Psi_{Pb}^{c} A_{Pb}^{d} - \lambda_{Pb} A_{Pb}^{p}\right)}{A_{Pb}^{s} - A_{Pb}^{r}} \quad \text{Eq. 3}$$

and

$$R_{Be} = \frac{H\left(\Psi_{Be}^{c} A_{Be}^{d} - \lambda_{Be} A_{Be}^{p}\right)}{A_{Be}^{s} - A_{Be}^{r}} \quad \text{Eq. 4}$$

Where Ψ_{Pb}^{c} and Ψ_{Be}^{c} are the rate constants corresponding to the scavenging of dissolved ^{210}Pb and ^{7}Be onto particles (d^{-1}), respectively. Then, particle residence times can be calculated as follows:

$$\tau_{Pb}^{p} = SPM \frac{H}{R_{Pb}^{r}} 365 \quad \text{Eq. 5}$$

$$\tau_{Be}^{p} = SPM \frac{H}{R_{Be}^{r}} 365 \quad \text{Eq. 6}$$

where τ_{Pb}^{p} and τ_{Be}^{p} are the residence times of particulate $^{210}Pb_{ex}$ and ^{7}Be (d), respectively, SPM is the Suspended Particulate Matter (g cm^{-3}), H is the height of the water column, and R_{Pb}^{p} and R_{Be}^{r} are the resuspension rates for $^{210}Pb_{ex}$ and ^{7}Be, from Eq. 3 and 4, respectively.

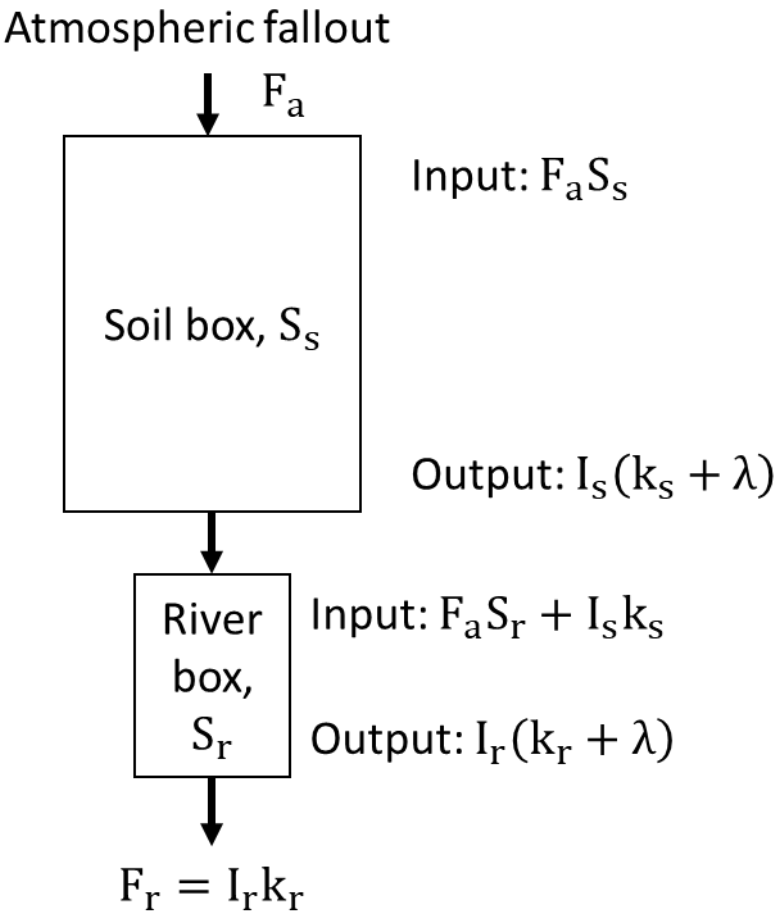
Using the single box approach, Jweda et al. (2008) modelled sediment residence times in the Clinton river from 0.5 to 8.6 days using $^{210}Pb_{ex}$ and from 0.2 to 2.1 days using ^{7}Be (Table 1). They found that particulate radionuclide residence times were significantly lower than the dissolved radionuclide residence time. This was attributed to the influence of colloidal-bound radionuclides (water samples filtered to < 0.5 µm pore size) during resuspension of bottom sediments which were accounted within the dissolved fraction. Moreover, an inverse strong relationship between ^{210}Pb and ^{7}Be log K_ds and log SPM was observed (R > 0.90) suggesting a particle-concentration effect attributed to the influence of colloidal cycling of particle-reactive species in the river. In another study in the same river calculated sediment residence times ranged from 0.32 to 3.86 days using $^{210}Pb_{ex}$ and from 0.9 to 13.4 days using $^{210}Po_{ex}$ (Baskaran et al. 2020).

One of the advantages of this model is that it incorporates the dissolved fraction of the radionuclides (Eq. 1 and 2; Figure 3). Although it is difficult to sample and measure dissolved radionuclides, integration of this fraction

compensates for *in situ* particle scavenging from the available dissolved radionuclides, which in turn accounts for fresh atmospheric input and radionuclide desorption from particles. Another important advantage is the quantification of the sediment resuspension rate, which allows estimation of sediment reworking from the storage compartment i.e. bottom sediments as in Jweda et al. (2008). This term could have potential implications for elucidating the quantities of old sediment in storage that can be remobilised. This gap in knowledge has been identified as a major difficulty regarding the use of ^{7}Be as tracer in sediment residence time studies (Fisher et al. 2010; Gellis et al. 2017) unless this contribution (i.e. sediment depleted in ^{7}Be) can be quantified within the timeframes of study.

4.2 Two-box balance model

The two-box balance model to determine sediment residence time was first developed by Dominik et al. (1987), later improved by Le Cloarec et al. (2007) and applied internationally (i.e. Mexico and UK) by Evrard et al. (2010) and Smith et al. (2014). Here the catchment is subdivided into two boxes (Figure 4) as follows: a) the soil box (given an s subscript in the text and equations) is characterised by an area S_s with low transport velocities and long residence times. It comprises the uppermost soil surface subjected to radionuclide fallout (Evrard et al. 2010), and b) the river box (given an r subscript in the text and equations) is characterised by the river area, S_r, and its immediate surroundings e.g. runoff and sediment-generating areas on hillslopes connected to the river network, faster exchanges and shorter residence times (Smith et al. 2014).



351

352 **Figure 4.** Conceptual diagram of the two box-model for sediment residence time estimations using FRNs. Boxes are
353 characterised by an area S_s (soil box) and S_r (river box). Modified from Evrard et al. (2010).

354 The two-box model requires solution to four equations:

355 Firstly, the fraction of atmospheric flux, F_a, present in each box and is given by:

356 $$S_s + S_r = 1 \qquad \text{Eq. 7}$$

357 Then the mass balance equations for each box are required. In the soil box, the loss of material is either
358 by transport into the river box or by radioactive decay:

359 $$F_a S_s = I_s(k_s + \lambda) \qquad \text{Eq. 8}$$

360 where k_s is the rate of transfer out of the soil box, related to the residence time τ_s, with $k_s = 1 / \tau_s$; I_s is the
361 radionuclide inventory in the soil box, and λ is the constant of radioactive decay.

The third equation represents the addition of atmospheric inputs to the river box flux coming from the soil box. Both fluxes are required to balance the output from the river and the radioactive decay:

$$F_a S_r + I_s k_s = I_r(k_r + \lambda) \quad \text{Eq. 9}$$

where I_r is the radionuclide inventory in the river box and k_r is the rate of output transfer from the river box.

Finally, the flux exported from the river box, F_r, is calculated:

$$F_r = I_r k_r \quad \text{Eq. 10}$$

These four equations are written for ^{137}Cs, ^{210}Pb$_{ex}$ and ^{7}Be considering the partitioning coefficients of the radionuclides between water and suspended matter, none of which were addressed in the former version of Dominik et al. (1987). In order to solve these equations, the following assumptions based on the λ values are made: 1) the radionuclide residence time in the soil compartment is expected to be of the order of decades to centuries, therefore, most of the ^{7}Be will be lost by decay implying its export from the soil compartment is negligible that is: $k_s \ll k_{Be}$ and $I_{s_{Be}} k_s \ll S_r F_{a_{Be}}$; 2) the decay rate of ^{210}Pb$_{ex}$ is considered negligible compared the export rate from the rapid compartment: $\lambda_{Pb} \ll k_r$. It is also assumed that the duration of sediment storage in the rapid compartment does not result in net decay of ^{7}Be to levels below the limit of detection (Smith et al. 2014).

The rapid compartment area, S_r, can be determined by combining Eq. 9 and 10 based on the first assumption. Then, S_r is a function of the ratio F_r/F_a for ^{7}Be and the export rate ($k_r = 1/\tau$):

$$S_r = \left(\frac{F_r}{F_a}\right)_{Be} \left(\frac{\lambda_{Be}}{k_r} + 1\right) \quad \text{Eq. 11}$$

The slow box residence time, τ_s, is then computed by solving Eq. 7 to 9 for ^{210}Pb$_{ex}$ with the assumption that $\lambda_{Pb} \ll k_r$ and combining this with Eq. 11:

$$\tau_s = \frac{\left(\frac{1}{\lambda_{Pb}}\right)\left[1-\left(\frac{F_a}{F_r}\right)_{Pb}\right]}{\left(\frac{R_r}{R_a}\right)(1+\lambda_{Be}\tau_r)-1} \quad \text{Eq. 12}$$

where $R_r = (F_r)_{Be}/(F_r)_{Pb}$ and $R_a = (F_a)_{Be}/(F_a)_{Pb}$.

Another relationship implicating τ_s and τ_r can be derived from the mass balance equations of ^{137}Cs. Given the atmospheric flux of ^{137}Cs is currently negligible i.e. $(F_a)_{Cs} = 0$, its inventory in soils can be determined. Based on Eq. 9, one can then determine τ_s as follows:

$$\tau_s = \frac{[M-(F_r)_{Cs}\tau_r]}{(F_r)_{Cs}(\lambda_{Cs}\tau_r+1)} \quad \text{Eq. 13}$$

where $M = (I_s)_{Cs} + (I_r)_{Cs}$.

The river box residence time, τ_r, may then be obtained by combining Eq. 11 and 12 and finding a solution to the equation:

$$a(\tau_r)^2 + b(\tau_r) + c = 0 \quad \text{Eq. 14}$$

where

$a = -(F_r)_{Cs}\left(\frac{R_r}{R_a}\right)\lambda_{Be}\lambda_{Pb}$,

$b = M\left(\frac{R_r}{R_a}\right)\lambda_{Be}\lambda_{Pb} - (F_r)_{Cs}\lambda_{Pb}\left(\frac{R_r}{R_a} - 1\right) - (F_r)_{Cs}A\lambda_{Cs}$ and

$c = \lambda_{Pb}M\left(\frac{R_r}{R_a} - 1\right) - A(F_r)_{Cs}$

in which

$A = 1 - \left(\frac{F_a}{F_r}\right)_{Pb}$.

The two-box model has been applied in various catchments with catchment surface areas ranging from 7 to 65,000 km^2 (see Table 1) and sediment residence times ranging from 4,800 to 30,321 years in the soil box and from 50 to 365 days in the river box. Le Cloarec et al. (2007) found a strong positive relationship between the soil box residence time and the catchment area within the Seine basin (see Figure 5, where R = 0.98, p < 0.001), while no relationship was found between the river box residence times and the catchment surface area. A correlation analysis of residence times modelled using the two-box model in the available literature demonstrates that this case is an exception, and we find no significant relationship between these two variables in the literature (Figure 5). Interestingly, an inverse relationship (R < -0.6) was found in the River Tamar basin, UK, between river box residence time and catchment surface area. However, only the 2007 period for the Tamar

was significant ($p < 0.05$). This finding suggests that residence times are, to some extent, independent of the catchment area, and that local factors such as topography, land use and climate may play a significant role in sediment release, transport and storage. Nevertheless, comparison between these studies is not straightforward as methods for data collection are different and some correlations are influenced by single extreme values. In this case, more studies (and data) are needed to better assess the influence of various catchment features on sediment residence time.

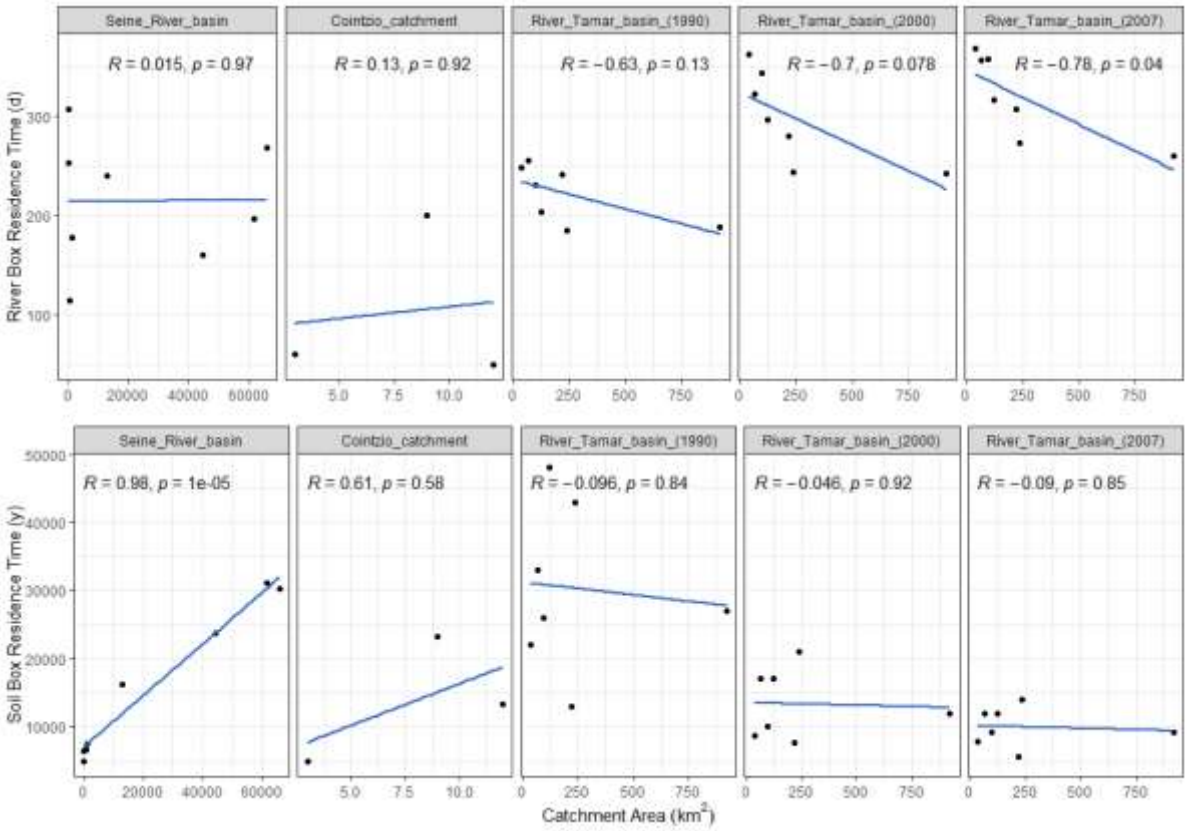


Figure 5. Correlation analysis between catchment area and soil box and river box residence times for data extracted from Evrard et al. (2010); Le Cloarec et al. (2007) and Smith et al. (2014) on the Cointzio, Seine, and Tamar river basins, respectively. Note that every data point (n = 8 for Seine basin, n = 3 Cointzio catchment, and n = 7 for River Tamar basin) represents a sub-catchment with a given area within the basin.

Sensitivity analysis was carried out by Le Cloarec et al. (2007) to assess the two-box model performance (Figure 6), and uncertainty simulations showed an important variation of sediment residence times for the soil and river boxes when suspended matter concentrations (i.e. suspended load) were modified (Figure 6a, b). The effect of

422 changes in this parameter on the estimated residence times were approximately a factor of 2. Variation in the
423 atmospheric fluxes of ^{7}Be and ^{210}Pb (i.e. F_a) did not significantly influence soil box residence times (τ_s) (Figure
424 6d, f), whereas a notable variation was found in the river box residence times (τ_r) (Figure 6c, e). For example,
425 variations in the atmospheric flux of ^{210}Pb (about 1.5 times) decreased residence time by a factor of 2 in the river
426 box. The influence of the ^{137}Cs catchment inventory was not as important as previous parameters when
427 modelling river box residence times (Figure 6g), but it exerts an important control when modelling soil box
428 residence times (Figure 6h). Overall, sensitivity analyses showed that estimation of suspended sediment fluxes,
429 ^{137}Cs inventory, and atmospheric fluxes of ^{7}Be and ^{210}Pb exert important controls on model outputs.
430 Furthermore, it was also found that ignoring the radionuclide K_d could influence residence time estimations by
431 a factor of 2 (Le Cloarec et al. 2007) and by factors of 1 - 1.3 (Smith et al. 2014).

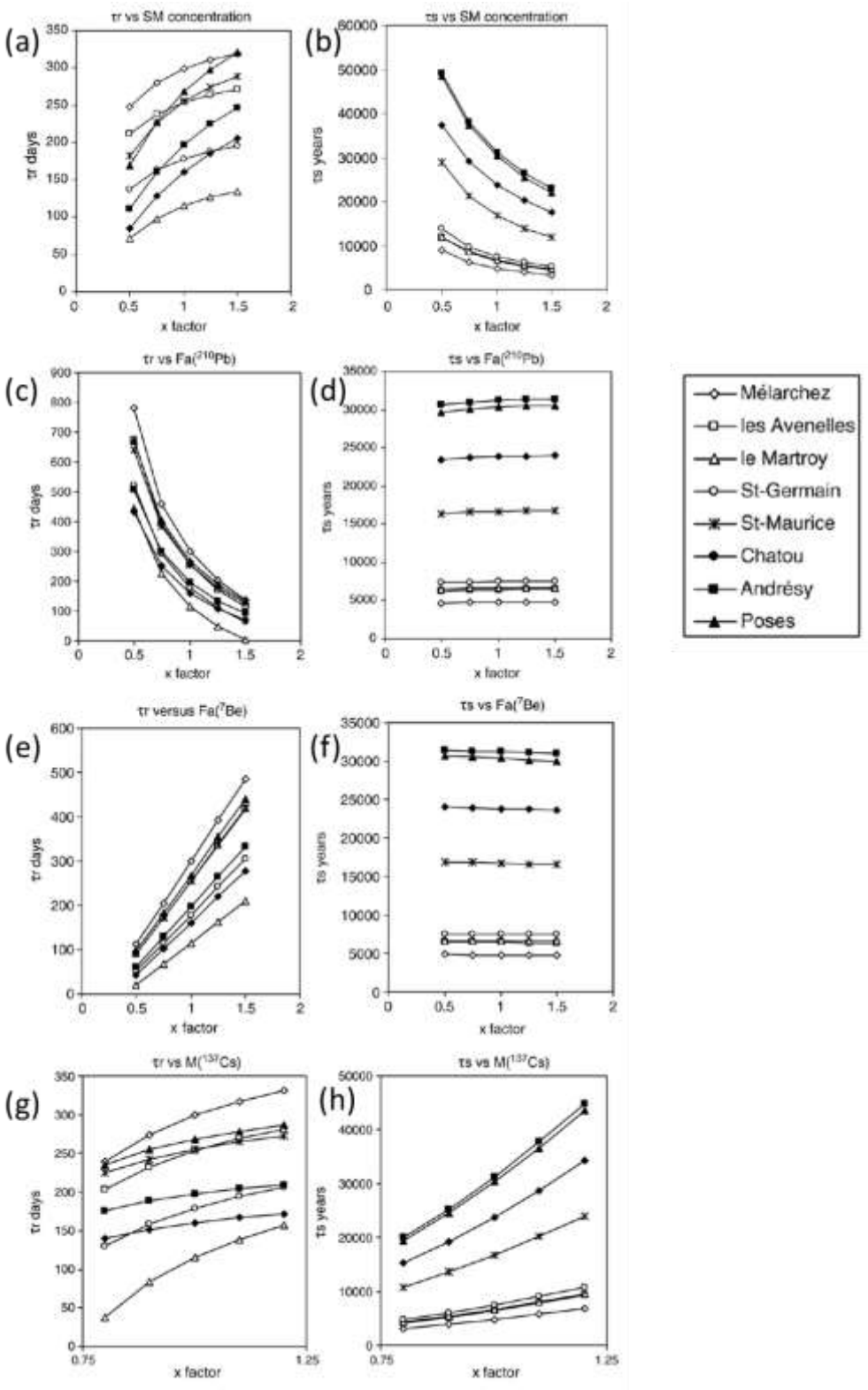


Figure 6. Sensitivity analysis of river and soil boxes residence times (τ_r and τ_s), illustrating the response of modelled residence times by changing parameters such as suspended matter (SM) concentration (a, b), atmospheric fluxes (F_a) of ^{210}Pb (c, d) and ^{7}Be (e, f), and the ^{137}Cs catchment inventory (g, h) in different sub-catchments of the Seine River basin. Extracted and modified from Le Cloarec et al. (2007).

437 *4.3 The $^{7}Be/^{210}Pb_{ex}$ ratio as an indicator of sediment age or the fraction of new sediment in suspension*

438 Another method used to determine sediment residence time includes the evaluation of the $^{7}Be/^{210}Pb_{ex}$ ratio as
439 an indicator of sediment age (hereafter sediment residence time as explained in section 1.1) or, alternatively,
440 the fraction of new sediment in suspension (Matisoff et al. 2005).

441 In this approach, sediment residence times are calculated as follows:

442 $$t = \frac{-1}{\left(\lambda_{^{7}Be} - \lambda_{^{210}Pb}\right)} \ln\left(\frac{A}{B}\right) + \frac{1}{\left(\lambda_{^{7}Be} - \lambda_{^{210}Pb}\right)} \ln\left(\frac{A_0}{B_0}\right) \quad \text{Eq. 15}$$

443 where $\lambda_{^{7}Be}$ and $\lambda_{^{210}Pb}$ are the decay constants of ^{7}Be and ^{210}Pb, respectively, A and B are the activity
444 concentrations of ^{7}Be and $^{210}Pb_{ex}$ in the sediment samples (e.g. suspended and/or channel bed sediments)
445 respectively, and A_0 and B_0 are the activity concentrations of ^{7}Be and $^{210}Pb_{ex}$ in the source, respectively. The
446 source term may refer to activity ratios from precipitation (Matisoff et al. 2005) or sediments from overland flow
447 (Le Gall et al. 2017). The second term in Eq. 15 is a constant with a value determined by the $^{7}Be/^{210}Pb_{ex}$ ratio in
448 the source.

449 Alternatively, the contribution of recently eroded particles can be calculated as follows:

450 $$\%\ \text{"new" sediment} = 100 \times \left(\frac{A/B}{A_0/B_0}\right) \quad \text{Eq. 16}$$

451 The $^{7}Be/^{210}Pb_{ex}$ chronometer application is carried out under several assumptions:

452 1) dry and wet fallout are included although ^{7}Be and $^{210}Pb_{ex}$ are delivered to the landscape primarily during
453 precipitation events;
454 2) ^{7}Be and $^{210}Pb_{ex}$ are delivered to the soil in a constant proportion regardless season, latitude or proximity
455 to the ocean and variations in the atmospheric flux is eliminated by using their ratio;
456 3) once deposited onto the landscape, ^{7}Be and $^{210}Pb_{ex}$ are assumed to be rapidly and irreversibly absorbed
457 to particulate matter;
458 4) ^{7}Be and $^{210}Pb_{ex}$ are not partitioned differentially onto particulate matter; and
459 5) since radionuclide sorption behaviour of ^{7}Be and $^{210}Pb_{ex}$ is similar, differences along the flow path
460 caused by particle size are eliminated by considering their ratio (including mineralogical variations).

461 The $^{7}Be/^{210}Pb_{ex}$ approach to determine sediment residence time has, however, been subject to critical appraisal
462 (Walling 2013). An aspect that has been challenged is the 'source' term (i.e. the initial activity ratio) required by
463 the method. The use of the $^{7}Be/^{210}Pb_{ex}$ ratio in rainfall as the initial activity ratio was used as a constant term in
464 several works (Matisoff et al. 2005; Evrard et al. 2010; Huisman et al. 2013). Nevertheless, it is known that
465 substantial variability can be found, both temporarily and spatially, in the atmospheric fluxes of ^{7}Be both
466 between and within storm events. For example, Gourdin et al. (2014a) showed that spatial variability in activity
467 concentrations were significant within the same storm with differences up to 6-fold for ^{7}Be and 4-fold for ^{210}Pb
468 at different stations within the storm. Furthermore, $^{7}Be/^{210}Pb$ activity ratios increased 2-fold during one storm
469 reflecting different controls on the fallout between these two radionuclides. Consequently, recommendations
470 of complete rainfall sampling, deposition separation of respective successive storms, spatially distributed
471 collection of rainwater, and the reduction of uncertainties related to rainfall collection were made (Gourdin et
472 al. 2014a). The assumption of constant activities derived from rainfall is thus poorly supported by the available
473 evidence. Also Walling (2013) criticised the sediment source controls on the $^{7}Be/^{210}Pb_{ex}$ ratio, since freshly
474 mobilised sediment will reflect the $^{7}Be/^{210}Pb_{ex}$ ratio of their sources and thus their relative contributions. In this
475 case, contributions from one source to another may change through time (i.e. within and between storm
476 events). For example, some sources have particulate matter with a given activity concentration of $^{210}Pb_{ex}$ as a
477 result of an accumulated inventory and being exposed to fresh fallout of ^{7}Be, therefore modifying their ratio.
478 One way to overcome this issue is the collection of sediments from overland flows to estimate source
479 radionuclide activity concentrations of fresh sediment inputs into the river (Gourdin et al. 2014b). This approach
480 is particularly helpful if sediment is originated primarily from catchment surface sources (Le Gall et al. 2017) but
481 the relative contribution of sources needs to be quantified.

482 The $^{7}Be/^{210}Pb_{ex}$ ratio is the most applied method to study the temporal dynamics of sediment within river
483 systems. Some studies have applied this method to assess the fine sediment dynamics during floods (Gourdin et
484 al. 2014b; Le Gall et al. 2017), while others have used it to investigate the sediment dynamics and sources of
485 sediment associated pollutants such as trace metals (Froger et al. 2018), pesticides (Gellis et al. 2017) and
486 phosphorus (Huisman and Karthikeyan 2012; Huisman et al. 2013; Lamba et al. 2015).

4.4 Age of surface derived sediments

Recently, a new approach linked a sediment fingerprinting method with sediment dating in order to determine the age of the surface-derived portion of sediments (Gellis et al. 2017, 2019). This method uses ^{7}Be and ^{210}Pb$_{ex}$ for two age classes: ^{7}Be up to ~ 1 year and ^{210}Pb$_{ex}$ up to ~ 85 years (Gellis et al. 2019). A generalised version of this model is as follows:

Firstly, ^{7}Be activity concentrations are corrected to the percent of surface-derived sediment:

$$^{7}Be_{corr} = \frac{^{7}Be_{target} - \left[^{7}Be_{s_1}\left(\frac{\%s_1}{100}\right) + \cdots + ^{7}Be_{s_n}\left(\frac{\%s_n}{100}\right)\right]}{\frac{\%surf}{100}} \quad \text{Eq. 17}$$

where $^{7}\mathrm{Be}_{\mathrm{corr}}$ is the estimated ^{7}Be activity in the surface; $^{7}Be_{target}$ is the measured activity in the target sample; $^{7}Be_{s_1}$ is the mean activity concentration of ^{7}Be in the non-surface derived sediment source 1, if it can be measured e.g. fresh tag of ^{7}Be on exposed sub-surfaces such as horizontally aligned sub-surfaces (i.e. rilled and scalded hillslopes) and/or gully areas during high rainfall and higher river flows (Wallbrink and Murray 1993; Hancock et al. 2014; Evrard et al. 2016); $\%\mathrm{s}_1$ is the % of contribution of the sediment source 1 to the target sediment sample obtained by the fingerprinting results; and $\%\mathrm{surf}$ is the surface derived percentage obtained from the sediment source apportionment. The age of the target sediment is then computed as follows:

$$^{7}Be_{age} = \frac{\ln\left(\frac{^{7}Be_{corr}}{^{7}Be_{surf}}\right)}{-\lambda_{^{7}Be}} \quad \text{Eq. 18}$$

where $^{7}\mathrm{Be}_{\mathrm{age}}$ is the age of the of the topsoil derived sediment; $^{7}\mathrm{Be}_{\mathrm{corr}}$ is the estimated surface ^{7}Be activity concentration; $^{7}\mathrm{Be}_{\mathrm{surf}}$ is the estimated ^{7}Be activity concentration of the source material (surface); and $\lambda_{^{7}\mathrm{Be}}$ is the decay constant for ^{7}Be.

Next, the estimated topsoil activity for ^{210}Pb is determined as follows:

$$^{210}Pb_{ex_{corr}} = \frac{^{210}Pb_{ex_{target}} - \left[^{210}Pb_{ex_{s_1}}\left(\frac{\%s_1}{100}\right) + \cdots + ^{210}Pb_{ex_{s_n}}\left(\frac{\%s_n}{100}\right)\right]}{\frac{\%surf}{100}} \quad \text{Eq. 19}$$

where $^{210}Pb_{ex_{corr}}$ is the estimated ^{210}Pb$_{ex}$ activity in the surface; $^{210}Pb_{ex_{target}}$ is the measured ^{210}Pb activity in the target sample (e.g. suspended sediment); $^{210}Pb_{ex_{s_1}}$ is the mean activity concentration in the non-surface derived sediment source 1; $\%\mathrm{s}_1$ is the percentage of contribution of the sediment source 1; and $\%\mathrm{surf}$ is the

surface derived percentage obtained from the sediment source apportionment. Finally, the age of the target sediment is computed as follows:

$$^{210}Pb_{ex_{age}} = \frac{\ln\left(\frac{^{210}Pb_{ex_{corr}}}{^{210}Pb_{ex_{surf}}}\right)}{-\lambda_{^{210}Pb}} \quad \text{Eq. 20}$$

Where $^{210}Pb_{ex_{age}}$ is the age of topsoil-derived sediment; $^{210}Pb_{ex_{corr}}$ is the estimated surface $^{210}Pb_{ex}$ activity concentration; $^{210}Pb_{ex_{surf}}$ is the weighted $^{210}Pb_{ex}$ activity concentration in the surface; and $\lambda_{^{210}Pb}$ is the decay constant of ^{210}Pb.

The age of the surface-derived sediment approach was first applied by Gellis et al. (2017) using ^{7}Be in a study comprising samples from 99 catchments in the Midwestern region of USA where residence times ranged from 0 to 174 days in bed sediments and from 0 to 84 in suspended sediments. They also found higher concentrations of pesticides (i.e. bifenthrin and DDE) in samples from the streambed with greater proportion of surface-derived sediments and relatively young residence times (< 100 days), which gives an indication of the role that sediment sources might play in the delivery of contaminated sediment and how quickly they move through the river system. In a subsequent study carried out in the agricultural Walnut Creek catchment (Iowa, USA), modelled sediment residence times ranged from 44 – 205 days using ^{7}Be and from 1 – 58 years using $^{210}Pb_{ex}$ (Gellis et al. 2019) (Table 1). In this study, sediment transport and storage were depicted in three boxes with three types of ages: 1) a rapid box with less than a year (based on ^{7}Be results), 2) a decadal box comprising from 10 to 100 years (based on $^{210}Pb_{ex}$ results) and 3) a geological box from 100 to > 1,000 years (based on the literature).

In their model, Gellis et al. (2019) defined age as the residence/transit time (see section 1.1) encompassing the time from when sediments enter a channel from a surface source to when the target sample is collected. As ^{7}Be is used as an indicator of the age of the surface derived sediment, an assumption of surficial erosion is made, and that erosion of deeper soil layers is neglected. Although this assumption was supported by a fingerprinting study, the effects of deeper erosion may be important and should be accounted for. In this regard, deeper surface erosion would tend to reduce ^{7}Be activity in sediments (Wallbrink and Murray 1993; Walling 2013) which, in turn, can increase sediment residence time estimations. Gellis et al. (2017) also acknowledge that fresh input of ^{7}Be on wetted channel areas of a stream channel can increase the activity of ^{7}Be. It has been estimated that direct contribution of ^{7}Be to large rivers could cause a 10 – 12% increase in activity concentrations (Hancock

et al. 2014). Hence, if atmospheric inputs on wet areas of the channel are important, then estimated ages of sediment might be younger.

5 Challenges, opportunities and future research needs.

The development of sedimentary models has contributed to an improved understanding of the temporal dynamics of catchment sediment storage and transport, however there are still some pitfalls that require careful consideration and further research. Most of these constraints are closely related to assumptions surrounding the behaviour of ^{7}Be when using it as soil and sediment tracer. For instance, there is no way at present of separating out the effects of decay and dilution of ^{7}Be activity concentrations in sediments, which raises one of the biggest limitations of the sediment residence/transit time methods when using FRNs (Matisoff et al. 2005; Fisher et al. 2010; Walling 2013; Gellis et al. 2019). The mixing of newly tagged ^{7}Be sediment with ^{7}Be-dead sediment (e.g. sediment from channel banks and/or sediment stored in streambeds and floodplains) is thus an issue yet to be addressed. More research is needed in this regard to quantify the uncertainty, or otherwise apportion the amount, of ^{7}Be-depleted sediments that are stored in riverine compartments. Furthermore, because most of the ^{7}Be activity concentration in soils is found in the top centimetre of surface soil (Blake et al. 1999; Schuller et al. 2006; Walling 2013), the catchment soil may be subjected to deeper erosion that will tend to lower activity concentrations of suspended sediment which can result in overestimations of sediment residence times (Gellis et al. 2019).

Equally important is the direct fallout of ^{7}Be and $^{210}Pb_{ex}$ in wetted areas of the stream channel which may increase activity concentrations of radionuclides in suspended sediments and sediments stored in the surface of the bed channel. It has been reported that direct fallout of radionuclides on stream channels is negligible compared to sediment inputs from erosion of the catchment soils (Hancock et al. 2014). Although this assumption has been supported in catchments with enhanced overland flow and surface runoff (Evrard et al. 2016; Le Gall et al. 2017), studies in forested and forest-influenced catchments have shown that channel interception of FRNs can be important (Karwan et al. 2016, 2018). Future studies should thus acquire information regarding hydrological flow paths and hillslope connectivity to address possible problems when assessing sediment residence times, or otherwise, quantify the proportion of radionuclide inputs into channels from

precipitation, especially at storm events. This would allow for a corrected sediment residence time that considers the influence of rainfall-delivered radionuclide tags onto sediment particles.

^{7}Be signatures of a specific source will continue changing through time in response to radioactive decay and input of fresh fallout. It is important to recognise, therefore, that $^{7}Be/^{210}Pb_{ex}$ ratios could be expected to vary both between storm events and seasonally, as well as from year to year, due to various controls on ^{7}Be and $^{210}Pb_{ex}$ fallout (Walling 2013; Gourdin et al. 2014a). Consequently, characterisation of signature of dominant sources should be undertaken as a core component of any sediment residence time study. This poses important challenges when planning sampling and its related logistics, particularly due to the short half-life of the ^{7}Be and the need to encompass, as much as possible, the variability of ^{7}Be in the sediment source signature (Gourdin et al. 2014a).

Radionuclide activity concentrations may correlate with grain size (He and Walling 1996), and their effects on tracer properties have been widely debated in the literature (Smith and Blake 2014; Laceby et al. 2017). Therefore, selective transport of particles, in terms of their size, can strongly influence sediment residence time calculations. Particle size analysis is, consequently, highly recommended in this regard, and when applicable correction factors should be applied to the radionuclide activity concentrations.

As discussed in section 3, the sorption behaviour of ^{137}Cs and ^{7}Be have been questioned, especially in changing environments (Parsons and Foster 2011; Taylor et al. 2012, 2013; Ryken et al. 2018). Rivers are dynamic systems that are subjected to rapidly changing conditions, influencing variables such as pH, redox, dissolved oxygen, temperature and conductivity. This has important implications when determining sediment residence times. For example, partitioning of ^{7}Be and ^{137}Cs between water and the particulate phase have been reported to be strongly influenced by pH and salinity (You et al. 1989; Kaste et al. 2002; Giannakopoulou et al. 2007; Hong et al. 2011; Kaste and Baskaran 2011; Taylor et al. 2013) suggesting the potential for these tracers to desorption under changing environments in the wider catchment. The potential for overestimation of residence times must, therefore, be considered.

Finally, the influence of environmental and anthropogenic factors on sediment residence time in river systems are poorly understood. Only a few studies have attempted to link and quantify the relationship between residence times and catchment processes under different scenarios such as changing land use (Smith et al. 2014)

589 and pollution (Huisman et al. 2013; Gellis et al. 2017; Froger et al. 2018). More research is thus needed to
590 understand the influence of various catchment characteristics such as land use, topography, flow regimes, and
591 soil type on sediment release, transport, and residence time in rivers. Moreover, assessment of the impact of
592 soil erosion measures, catchment management practices, and climate change on sediment residence times in
593 river channels is still lacking but has important implications from a catchment management perspective.

594 **Acknowledgements**

595 E. Munoz-Arcos would like to acknowledge the support by the Agencia Nacional de Investigacion y Desarrollo
596 (ANID) through the PhD scholarship ID 72210264. The work represents a contribution to the joint International
597 Atomic Energy and UN FAO Coordinated Research Programme D1.50.18 “Multiple Isotope Fingerprints to
598 Identify Sources and Transport of Agro-Contaminants”.

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