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# Understanding the complexity of sediment residence time in rivers: Application of Fallout Radionuclides (FRNs)

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1                    Understanding the Complexity of  
2                    Sediment Residence Time in Rivers:  
3                    Application of Fallout Radionuclides  
4                    (FRNs)  
5

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17

18 **Abstract**

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

39 **Keywords**

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

41 **CRediT author statement**

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

# 47 1 Introduction.

48 Riverine sediments, typically particles < 2 mm in diameter, play an important role in the healthy functioning of  
49 river ecosystems since they provide nutrients and contribute to habitat quality (Kemp et al. 2011; Jones et al.  
50 2012; Wharton et al. 2017). However, human activities such as reservoir and dam construction, land use  
51 changes, deforestation, mining activities and urbanisation have altered sediment fluxes in river systems resulting  
52 in enhanced, or reduced, sedimentation and contamination (Walling and Fang 2003; Owens et al. 2005; Syvitski  
53 and Kettner 2011; Wohl 2015). Excessive sediment supply into rivers has detrimental impacts on water quality  
54 (e.g. turbidity) which causes sedimentation in river channels, reservoirs and estuaries affecting aquatic habitats  
55 such as salmonids spawning gravels (Kemp et al. 2011). In addition, silts and clays are geochemically active and  
56 consequently are responsible for the transport of contaminants, including trace metals, phosphorus, Polycyclic  
57 Aromatic Hydrocarbons (PAHs), pesticides, radionuclides and Polychlorinated Biphenyls (PCBs) which have high  
58 sorptive affinity for fine-grained sediment particles (Owens et al. 2005; Wohl 2015; Alewell et al. 2017; Rügner  
59 et al. 2019; Owens 2020).

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

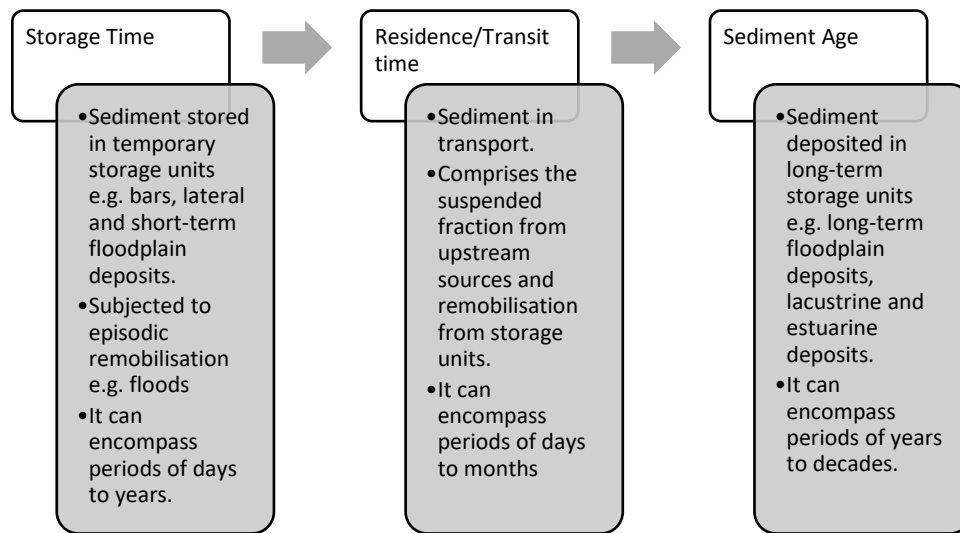
75 river basins, which can encompass days to months (Matisoff et al. 2005; Le Gall et al. 2017), decades (Wallbrink  
76 et al. 2002) and millennia or longer (Hoffmann 2015).

### 77 1.1 Definitions

78 Finding a consensus on the meaning of sediment residence times in rivers is problematic since definitions are  
79 often dependent on the methodology. For example, Whipp & Ehlers (2019) define sediment residence time as  
80 the “time sediments remain within the catchment as part of a population of minerals that could be dated in a  
81 random sample, assuming no long-term sediment storage in the catchment”. Alternatively, Skalak & Pizzuto  
82 (2010) define sediment residence time as “the time required to remove a mass of sediment equal to the total  
83 volume in storage”. Other authors make a distinction between terms such as sediment transit time and sediment  
84 residence time. For example, Gellis et al. (2019) define sediment transit time as “the time it takes for sediment  
85 to travel from a starting point in the watershed to an endpoint where sediment leaves the area of interest”,  
86 whereas residence time refers to “the mean time sediment particles spend in a storage reservoir”. Gellis et al.  
87 (2019) also noted that these terms can depend on the spatial scale used. Thus, transit time can encompass the  
88 time from when sediment is released to when it enters the river channel (e.g. Slattery et al. 2002) or the time  
89 that sediment spends in all storage reservoirs i.e. hillslopes, channel-bed and floodplains (e.g. Hoffmann 2015).  
90 On the other hand, residence time can span sediments deposited at a specific storage unit i.e. channel-bed (e.g.  
91 Skalak and Pizzuto 2010) or also include the suspended fraction (Evrard et al. 2010; Smith et al. 2014). In addition,  
92 Matisoff et al. (2005) used the term sediment age to refer to the time between particles receiving their  
93 radioactive tags to when they are sampled in the river as suspended, bed-stored, or estuarine sediment.

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

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



107

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

112

113 *1.2 Methods to determine sediment residence time*

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

121 sonar transducers and LiDAR to evaluate empirical sediment residence times. Radionuclides offer the possibility  
122 of determining both sediment age and residence times. For instance, Uranium-series isotopes have been used  
123 to document the time-dependence of weathering ages (Dosseto et al. 2014; Suresh et al. 2014) and  $^{14}\text{C}$  dating  
124 has been applied to estimate the mean sediment age of floodplain sediments (Sutfin and Wohl 2019). Short-  
125 lived radionuclides (e.g.  $^7\text{Be}$ ,  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$  and  $^{234}\text{Th}$ ) have also been used to assess sediment travel distances,  
126 sediment age and sediment residence time in a variety of landscapes (Olsen et al. 1986; Dominik et al. 1987;  
127 Wieland et al. 1991; Vogler et al. 1996; Bonniwell et al. 1999; Feng et al. 1999; Ciffroy et al. 2003; Forster et al.  
128 2009). In lakes, Wieland et al. (1991) modelled sediment residence times using  $^7\text{Be}$  and  $^{210}\text{Pb}$  fluxes in Lake  
129 Zurich, Switzerland, and Vogler et al. (1996) estimated sediment residence times in Lake Constance for the total  
130 radionuclide inventories of  $^{234}\text{Th}$  and  $^7\text{Be}$ . In estuaries, Olsen et al. (1986) assessed water column removal rates  
131 and residence time of  $^7\text{Be}$  in the James River Estuary, USA, and Ciffroy et al. (2003) determined sediment  
132 residence time of suspended particles using  $^7\text{Be}$  budgets in the turbidity maximum zone of the Loire estuary,  
133 France. The application of these radionuclides as sediment residence time tracers in lakes, coastal and estuarine  
134 zones has been discussed elsewhere (Dominik et al. 1989; Steinmann et al. 1999; Feng et al. 1999; Baskaran  
135 2001; Baskaran and Swarzenski 2007; Forster et al. 2009; Saari et al. 2010). For extensive reviews on the use of  
136 radionuclides in these environments the reader is directed to works from Du et al. (2011); Kaste et al. (2002);  
137 Kaste & Baskaran (2011); Waples et al. (2006).

### 138 *1.3 Rationale and objectives of this review*

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

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

## 153 **2 River sediment budgeting and its relevance to channel storage and** 154 **residence time.**

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

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



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

### 183 3 Suitability of medium and short-lived radionuclides as sediment 184 residence time tracers.

185 Medium and short-lived radionuclides (i.e.  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$ ,  $^7\text{Be}$  and  $^{234}\text{Th}$ ) have been extensively used as soil and  
186 sediment tracers to assess their redistribution, deposition rates and residence time (Feng et al. 1999; Blake et  
187 al. 2002; Waples et al. 2006; Mabit et al. 2008; Blake et al. 2009; Mabit et al. 2013; Taylor et al. 2013; Walling  
188 2013; Mabit et al. 2014; Taylor et al. 2019). However, it is not the aim of this review to examine the current  
189 knowledge of radionuclides as soil erosion and sediment tracers, for which the reader is directed to reviews by  
190 Mabit et al. (2014, 2013); Matisoff (2014); Matisoff and Whiting (2011); Parsons and Foster (2011); Taylor et al.  
191 (2013); and Walling (2013). Moreover, applications of  $^{234}\text{Th}$  as a tracer of sediment dynamics in freshwater  
192 systems (Waples et al. 2006) have mostly concerned lakes (Dominik et al. 1989; Vogler et al. 1996; Waples et al.  
193 2004). Some studies have determined  $^{234}\text{Th}$  in river sediments and water despite the typically low activity  
194 concentrations (Morris et al. 1994; Waples et al. 2003) but no sediment residence time studies using this  
195 radionuclide in river systems have been found. For this reason, we focus our discussion on Fallout Radionuclides  
196 (FRNs).

197  $^{137}\text{Cs}$  ( $t_{1/2} = 30.17$  years) is an anthropogenic radionuclide that was released to the stratosphere as a result of  
198 atmospheric thermonuclear weapon testing in the 1950s – 1980s, in addition there have been releases during  
199 nuclear accidents such as the 1986 Chernobyl and the 2011 Fukushima disasters. Before subsequent deposition  
200 on the Earth's surface,  $^{137}\text{Cs}$  circulated globally and was washed out by precipitation patterns (Ritchie and  
201 McHenry 1990).  $^{210}\text{Pb}$  ( $t_{1/2} = 22.2$  years) is of geogenic origin and is a natural decay product within the  $^{238}\text{U}$  decay

202 series derived from the decay of the inert gas  $^{222}\text{Rn}$  ( $t_{1/2} = 3.8$  days) which derives from its parent  $^{226}\text{Ra}$  ( $t_{1/2} =$   
203 1622 years). The  $^{210}\text{Pb}$  generated in situ by decay of  $^{226}\text{Ra}$  is termed supported  $^{210}\text{Pb}$  and is in equilibrium with  
204  $^{226}\text{Ra}$ . However,  $^{222}\text{Rn}$  diffuses to the atmosphere and undergoes a series of short-lived decays to  $^{210}\text{Pb}$  which  
205 may adsorb to aerosols and is delivered to the landscape by wet and dry fallout. Fallout  $^{210}\text{Pb}$  is termed  
206 unsupported, or excess  $^{210}\text{Pb}$  ( $^{210}\text{Pb}_{\text{ex}}$ ).  $^7\text{Be}$  ( $t_{1/2} = 53.3$  days) is a cosmogenic radionuclide produced by cosmic ray  
207 spallation of nitrogen and oxygen in the stratosphere and troposphere and it is delivered to the earth's surface  
208 through wet and dry deposition.

209 An advantage of using  $^{137}\text{Cs}$ ,  $^{210}\text{Pb}_{\text{ex}}$  and  $^7\text{Be}$  as sediment chronometers, is that the contrasting half-lives that  
210 these radionuclides possess can be used to model sediment residence time from days to decades in different  
211 catchment compartments (Wallbrink et al. 2002; Matisoff et al. 2005; Le Cloarec et al. 2007; Gellis et al. 2019).  
212 Additionally, naturally and continuously produced  $^7\text{Be}$  and  $^{210}\text{Pb}$  radioisotopes can be used at many sites in the  
213 world widening their application (Wallbrink et al. 2002; Matisoff et al. 2005; Le Cloarec et al. 2007; Jweda et al.  
214 2008; Evrard et al. 2010; Smith et al. 2014). The basis for the use of these radionuclides as tracers is their ability  
215 to rapidly and strongly adsorb onto soil and sediment particles following both wet and dry deposition (Welp and  
216 Brümmer 1999; Du et al. 2011; Matisoff and Whiting 2011; Taylor et al. 2012; Matisoff 2014; Singleton et al.  
217 2017; Ryken et al. 2018) with reported partition coefficients,  $K_d$ , of the order  $10^5$  (Olsen et al. 1986; Hawley et  
218 al. 1986; Dominik et al. 1987; Van Hoof and Andren 1989; You et al. 1989; Steinmann et al. 1999; Kaste et al.  
219 2002; Jweda et al. 2008). However, sorption behaviour of  $^{137}\text{Cs}$  and  $^7\text{Be}$  have been questioned, especially in  
220 changing environments through the catchment sediment cascade which can alter sediment residence time  
221 estimations (Parsons and Foster 2011; Taylor et al. 2012, 2013; Ryken et al. 2018). For example, partitioning of  
222  $^7\text{Be}$  and  $^{137}\text{Cs}$  have been reported to be strongly influenced by pH and salinity (You et al. 1989; Kaste et al. 2002;  
223 Giannakopoulou et al. 2007; Hong et al. 2011; Kaste and Baskaran 2011; Taylor et al. 2013), therefore, its  
224 application in estuarine environments as a sediment particle tracer is challenging and requires case-by-case  
225 validation.

## 226 4 Models to evaluate sediment residence time in river systems.

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

229 mass balances to assess sediment residence times began in late 1980s using data from the Rhone catchment,  
230 Switzerland (Dominik et al. 1987). However, it was not until late 1990s that other authors employed different  
231 methodologies to determine/model sediment residence times. For example, Wallbrink et al. (1998) determined  
232 the proportional contribution from three potential sources: cultivated and uncultivated land and subsoil  
233 material from gullies and channel banks. Then, applying a mixing model with various concentrations of  $^{210}\text{Pb}_{\text{ex}}$   
234 corrected as a function of radioactive decay in the channel, they found that the mean residence time of fine-  
235 grained material was  $10 \pm 5$  years (Table 1). However, evidence that sediment residence times in the  
236 Murrumbidgee river, Australia could be of the order of weeks to months was noticed from changes in the mean  
237  $^{137}\text{Cs}$  activity concentrations of suspended sediments between flood and low-flow conditions, and the presence  
238 of  $^7\text{Be}$  activity in sediments from flood water. Bonniwell et al. (1999) used  $^7\text{Be}/^{210}\text{Pb}$  and  $^{210}\text{Pb}/^{137}\text{Cs}$  ratios to  
239 assess the fraction of new sediment in suspension, residence times and transport distances. In their study, the  
240 basis for the use of  $^7\text{Be}/^{210}\text{Pb}$  ratios was established because it corrected for the relative sorption and enrichment  
241 effects resulting from variations in grain size and particulate matter composition, and by using  $^{210}\text{Pb}/^{137}\text{Cs}$  ratios  
242 they accounted for variations in mineralogy and source area activity concentrations. Residence times in  
243 sediments from the channel of River Gold Fork, USA, ranged from 1.6 to 103 days (Table 1) from the upper to  
244 the lower part of the catchment. In addition, Wallbrink et al. (2002) determined residence times using  $^{210}\text{Pb}_{\text{ex}}$   
245 activity concentrations in river bed sediments from the Brisbane and Logan rivers, Australia. They assumed that  
246 increases in  $^{210}\text{Pb}_{\text{ex}}/^{137}\text{Cs}$  ratios in sediments occurred because of primarily direct input of fresh flux of  $^{210}\text{Pb}_{\text{ex}}$  to  
247 the sediments in the river channel. This was supported because no such higher ratios were measured at potential  
248 soil erosion sources within the catchment. Using the initial inventory of  $^{210}\text{Pb}_{\text{ex}}$  within the mobile layer of river  
249 sediment derived from the catchment erosion,  $^{210}\text{Pb}_{\text{ex}}$  inventories at the time of sample collection and the  
250 inventory of  $^{210}\text{Pb}_{\text{ex}}$  which occurs when depositional flux decay within the sediment profile reach equilibrium,  
251 they calculated residence times for deposited sediments in rivers Brisbane and Logan of 0 – 21 and 0 – 9 years  
252 (Table 1), respectively. This model was also applied by Douglas et al. (2009) in the Maroochy River estuary,  
253 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 <sup>2</sup>	<sup>137</sup> Cs, <sup>210</sup> Pb and <sup>7</sup> Be	Two-box model.	800 – 1,400 y; 1 – 220 d	Dominik et al. (1987)
Murrumbidgee River, Australia.	13,500 km <sup>2</sup>	<sup>210</sup> Pb <sub>ex</sub>	<sup>210</sup> Pb <sub>ex</sub> source decay as a function of in-channel residence time	10 y	Wallbrink et al. (1998)
Gold Fork River, USA.	389 km <sup>2</sup>	<sup>137</sup> Cs, <sup>210</sup> Pb and <sup>7</sup> Be	Normalised activity ratios	1.6 – 103 d	Bonniwell et al. (1999)
Brisbane and Logan River catchments. Australia.	13,600 and 3,076 km <sup>2</sup>	<sup>137</sup> Cs and <sup>210</sup> 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 <sup>2</sup>	<sup>7</sup> Be and <sup>210</sup> Pb <sub>ex</sub>	<sup>7</sup> Be/ <sup>210</sup> Pb <sub>ex</sub> ratio	50 – 300 d	Matisoff et al. (2005)
River Seine basin, France.	7 to 65,700 km <sup>2</sup>	<sup>137</sup> Cs, <sup>210</sup> Pb and <sup>7</sup> 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 <sup>2</sup>	<sup>7</sup> Be and <sup>210</sup> Pb <sub>ex</sub>	Single-box model	0.2 – 2.1 d ( <sup>7</sup> Be), 0.5 – 8.6 d ( <sup>210</sup> Pb)	Jweda et al. (2008)
Maroochy river estuary, Australia.	630 km <sup>2</sup>	<sup>137</sup> Cs and <sup>210</sup> 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 <sup>2</sup>	<sup>137</sup> Cs, <sup>210</sup> Pb and <sup>7</sup> Be  <sup>7</sup> Be and <sup>210</sup> Pb <sub>ex</sub>	Two-box balance model.  <sup>7</sup> Be/ <sup>210</sup> Pb <sub>ex</sub> ratio	50 – 200 d (river box); 5,000 – 23,000 y (soil box)  101 – 163 d	Evrard et al. (2010)
White, West rivers and Mink Brook; USA.	29 to 319 km <sup>2</sup>	<sup>7</sup> Be and <sup>210</sup> Pb <sub>ex</sub>	Stacked reservoirs model.	4 to > 300 d	Gartner et al. (2012)
Pheasant sub-catchment, USA	12.4 km <sup>2</sup>	<sup>7</sup> Be and <sup>210</sup> Pb <sub>ex</sub>	<sup>7</sup> Be/ <sup>210</sup> Pb <sub>ex</sub> ratio	40 – 319 d	Huisman and Karthikeyan (2012)
Pheasant sub-catchment, USA.	12.4 km <sup>2</sup>	<sup>7</sup> Be and <sup>210</sup> Pb <sub>ex</sub>	<sup>7</sup> Be/ <sup>210</sup> Pb <sub>ex</sub> ratio	9 – 318 d	Huisman et al. (2013)
River Tamar basin, UK.	38 to 219 km <sup>2</sup>	<sup>137</sup> Cs, <sup>210</sup> Pb and <sup>7</sup> 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 <sup>2</sup>	<sup>7</sup> Be and <sup>210</sup> Pb <sub>ex</sub>	<sup>7</sup> Be/ <sup>210</sup> Pb <sub>ex</sub> ratio	123 – 322 d	Lamba et al. (2015)
Loroux catchment, France.	25 km <sup>2</sup>	<sup>7</sup> Be and <sup>210</sup> Pb <sub>ex</sub>	<sup>7</sup> Be/ <sup>210</sup> Pb <sub>ex</sub> ratio	20 – 200 d	Le Gall et al. (2017)
Midwestern USA rivers	6.8 to 5,893 km <sup>2</sup>	<sup>210</sup> Pb <sub>ex</sub>  <sup>7</sup> Be and <sup>210</sup> Pb <sub>ex</sub>	Age of surface derived sediments  <sup>7</sup> Be/ <sup>210</sup> Pb <sub>ex</sub> ratio	0 – 174 d  61 – 282 d	Gellis et al. (2017)
White clay creek, USA.	7.25 km <sup>2</sup>	<sup>7</sup> Be and <sup>210</sup> Pb <sub>ex</sub>	<sup>7</sup> Be/ <sup>210</sup> Pb <sub>ex</sub> ratio	22 – 110 d	Karwan et al. (2018)
Orge river catchment	900 km <sup>2</sup>	<sup>7</sup> Be and <sup>210</sup> Pb <sub>ex</sub>	<sup>7</sup> Be/ <sup>210</sup> Pb <sub>ex</sub> ratio	18 – 140 d	Froger et al. (2018)
Clinton River, Southeast Michigan, USA	1,946 km <sup>2</sup>	<sup>210</sup> Po <sub>ex</sub> and <sup>210</sup> Pb <sub>ex</sub>	Single-box model	0.3 – 3.9 d ( <sup>210</sup> Pb), 0.9 – 13.4 d ( <sup>210</sup> Po)	Baskaran et al. (2020)
Walnut creek	52.6 km <sup>2</sup>	<sup>7</sup> Be and <sup>210</sup> Pb <sub>ex</sub>	Age of surface derived sediments	44 – 205 d ( <sup>7</sup> Be), 1 – 58 y ( <sup>210</sup> Pb)	Gellis et al. (2019)
Ducktrap River, USA.	9 km reach	<sup>7</sup> Be	CIA (Constant Initial Activity) aging model	0 to > 160 d	Fisher et al. (2010)
South River, USA	37 km reach	<sup>14</sup> C, <sup>137</sup> Cs and <sup>210</sup> Pb <sub>ex</sub>	Reservoir theory model	1 – 60 y	Skalak and Pizzuto (2010)



256

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

258 A different approach was adopted by Skalak & Pizzuto (2010) who applied several radiometric dating methods  
 259 ( $^{14}\text{C}$ ,  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$ ) to infer the distribution of ages of sediment stored within Fine-Grained Channel Margin  
 260 (FGCM) deposits of the South River, USA. By applying the reservoir theory<sup>1</sup> to the population of ages dated on  
 261 the deposits, they found a sediment residence time of 1.75 years with a very small portion of sediments with  
 262 storage times of the order of decades (60 years). One of the main benefits of this approach is that it provides  
 263 both residence time and a distribution of ages. Similarly, Fisher et al. (2010) studied sediment storage times  
 264 using  $^7\text{Be}$  coupled with a Constant Initial Activity (CIA) sediment aging model to assess transitional bedload  
 265 storage times in bars associated with in-channel obstructions (large wood debris and boulders). They identified  
 266 two dominant transport regimes with differing storage times: 1) transport-limited reaches with storage times >  
 267 100 days associated with channel obstructions, and 2) supply-limited reaches associated with steeper gradients  
 268 and greater stream power capable of mobilising fine-grained sediments from channel obstructions with  
 269 generally < 100 days of sediment storage. Although they addressed successfully the bed-storage time problem

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<sup>1</sup> 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).

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

- 272 1)  $^7\text{Be}$  activity concentration dilution by sediment depleted in  $^7\text{Be}$  through long-term in-channel residence  
273 times (greater decay) and/or frequent landslides or bank collapses which may supply  $^7\text{Be}$ -depleted  
274 sediments diminishing initial activities used to feed the CIA model, and
- 275 2)  $^7\text{Be}$  sediment enrichment by fresh tagging from atmospheric inputs into submerged bars at low-flow  
276 conditions which may overprint the inherited  $^7\text{Be}$  signal and thus increase initial activities of  $^7\text{Be}$ .

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

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

#### 290 4.1 Single box model

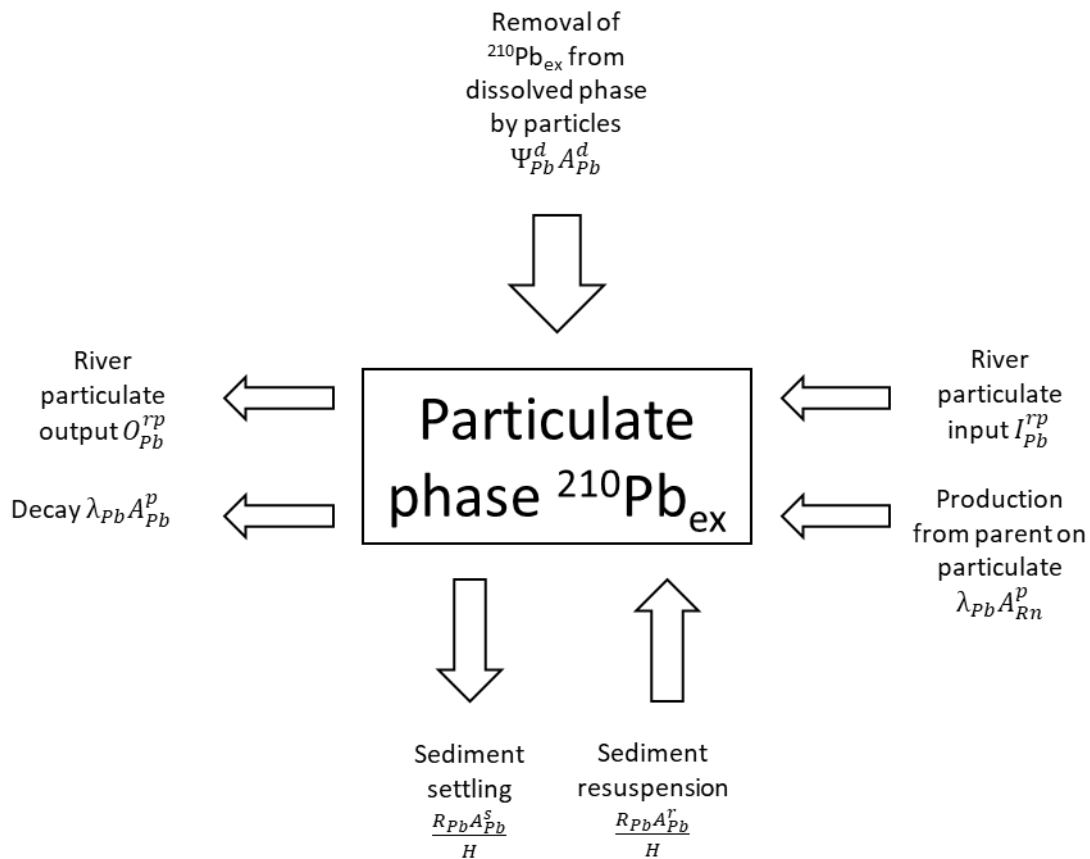
291 The single box model described here uses FRNs to assess sediment residence times in river systems (Jweda et  
292 al. 2008; Baskaran et al. 2020). Particle residence times are obtained by means of mass balances of particulate  
293  $^{210}\text{Pb}_{\text{ex}}$  (Eq. 1) and  $^7\text{Be}$  (Eq. 2) to obtain sediment resuspension rates as follows (Figure 3):

$$294 \Psi_{Pb}^d A_{Pb}^d + \lambda_{Pb} A_{Rn}^p + I_{Pb}^{rp} + \frac{R_{Pb} A_{Pb}^r}{H} = O_{Pb}^{rp} + \lambda_{Pb} A_{Pb}^p + \frac{R_{Pb} A_{Pb}^s}{H} \quad \text{Eq. 1}$$

295 and

296 
$$\Psi_{Be}^d A_{Be}^d + I_{Be}^{rp} + \frac{R_{Be} A_{Be}^r}{H} = O_{Be}^{rp} + \lambda_{Be} A_{Be}^p + \frac{R_{Be} A_{Be}^s}{H}$$
 Eq. 2

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



305

306 **Figure 3.** Diagram of the single box model illustrating the sources and sinks of particulate  $^{210}\text{Pb}$ . Modified from Jweda et al.  
 307 (2008). The same model concept applies to  $^7\text{Be}$  sources and sinks but without the input from the decay of a parent  
 308 radionuclide.

309

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

$$313 \quad R_{Pb} = \frac{H(\Psi_{Pb}^c A_{Pb}^d - \lambda_{Pb} A_{Pb}^p)}{A_{Pb}^s - A_{Pb}^r} \quad \text{Eq. 3}$$

314 and

$$315 \quad R_{Be} = \frac{H(\Psi_{Be}^c A_{Be}^d - \lambda_{Be} A_{Be}^p)}{A_{Be}^s - A_{Be}^r} \quad \text{Eq. 4}$$

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

$$318 \quad \tau_{Pb}^p = SPM \frac{H}{R_{Pb}^r} 365 \quad \text{Eq. 5}$$

$$319 \quad \tau_{Be}^p = SPM \frac{H}{R_{Be}^r} 365 \quad \text{Eq. 6}$$

320 where  $\tau_{Pb}^p$  and  $\tau_{Be}^p$  are the residence times of particulate  $^{210}\text{Pb}_{\text{ex}}$  and  $^7\text{Be}$  (d), respectively, SPM is the Suspended  
 321 Particulate Matter ( $\text{g cm}^{-3}$ ),  $H$  is the height of the water column, and  $R_{Pb}^p$  and  $R_{Be}^r$  are the resuspension rates for  
 322  $^{210}\text{Pb}_{\text{ex}}$  and  $^7\text{Be}$ , from Eq. 3 and 4, respectively.

323 Using the single box approach, Jweda et al. (2008) modelled sediment residence times in the Clinton river from  
 324 0.5 to 8.6 days using  $^{210}\text{Pb}_{\text{ex}}$  and from 0.2 to 2.1 days using  $^7\text{Be}$  (Table 1). They found that particulate radionuclide  
 325 residence times were significantly lower than the dissolved radionuclide residence time. This was attributed to  
 326 the influence of colloidal-bound radionuclides (water samples filtered to  $< 0.5 \mu\text{m}$  pore size) during resuspension  
 327 of bottom sediments which were accounted within the dissolved fraction. Moreover, an inverse strong  
 328 relationship between  $^{210}\text{Pb}$  and  $^7\text{Be}$  log  $K_{ds}$  and log SPM was observed ( $R > 0.90$ ) suggesting a particle-  
 329 concentration effect attributed to the influence of colloidal cycling of particle-reactive species in the river. In  
 330 another study in the same river calculated sediment residence times ranged from 0.32 to 3.86 days using  $^{210}\text{Pb}_{\text{ex}}$   
 331 and from 0.9 to 13.4 days using  $^{210}\text{Po}_{\text{ex}}$  (Baskaran et al. 2020).

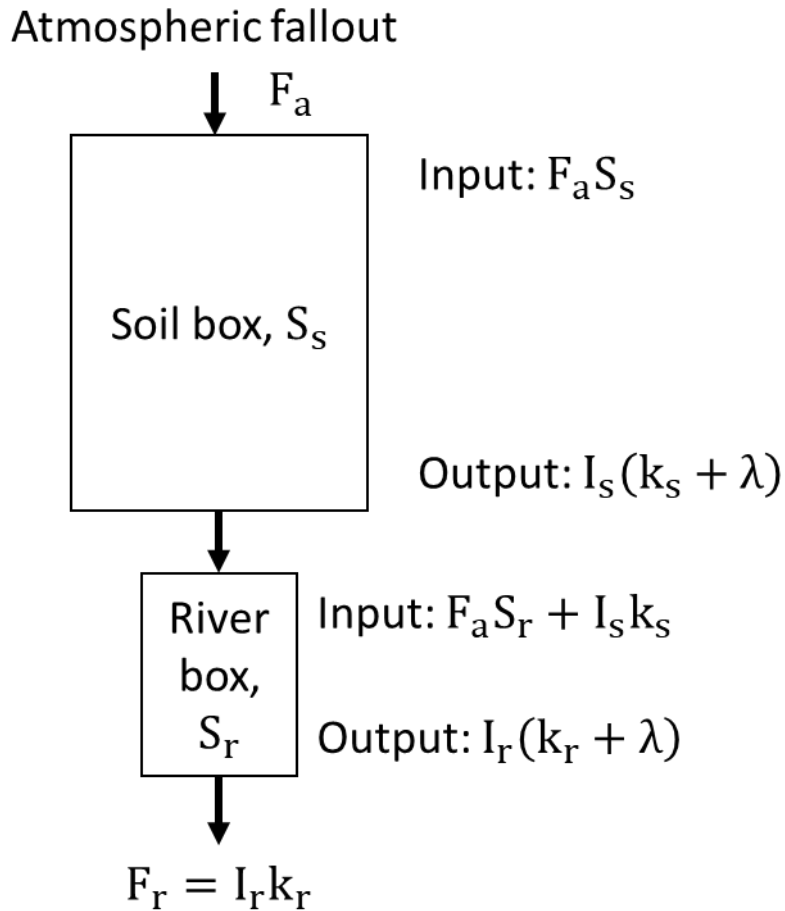
332 One of the advantages of this model is that it incorporates the dissolved fraction of the radionuclides (Eq. 1 and  
 333 2; Figure 3). Although it is difficult to sample and measure dissolved radionuclides, integration of this fraction



334 compensates for *in situ* particle scavenging from the available dissolved radionuclides, which in turn accounts  
335 for fresh atmospheric input and radionuclide desorption from particles. Another important advantage is the  
336 quantification of the sediment resuspension rate, which allows estimation of sediment reworking from the  
337 storage compartment i.e. bottom sediments as in Jweda et al. (2008). This term could have potential implications  
338 for elucidating the quantities of old sediment in storage that can be remobilised. This gap in knowledge has been  
339 identified as a major difficulty regarding the use of  $^7\text{Be}$  as tracer in sediment residence time studies (Fisher et al.  
340 2010; Gellis et al. 2017) unless this contribution (i.e. sediment depleted in  $^7\text{Be}$ ) can be quantified within the  
341 timeframes of study.

#### 342 4.2 Two-box balance model

343 The two-box balance model to determine sediment residence time was first developed by Dominik et al. (1987),  
344 later improved by Le Cloarec et al. (2007) and applied internationally (i.e. Mexico and UK) by Evrard et al. (2010)  
345 and Smith et al. (2014). Here the catchment is subdivided into two boxes (Figure 4) as follows: a) the soil box  
346 (given an s subscript in the text and equations) is characterised by an area  $S_s$  with low transport velocities and  
347 long residence times. It comprises the uppermost soil surface subjected to radionuclide fallout (Evrard et al.  
348 2010), and b) the river box (given an r subscript in the text and equations) is characterised by the river area,  $S_r$ ,  
349 and its immediate surroundings e.g. runoff and sediment-generating areas on hillslopes connected to the river  
350 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  $S_s + S_r = 1$  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  $F_a S_s = I_s(k_s + \lambda)$  Eq. 8

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

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

$$364 \quad F_a S_r + I_s k_s = I_r (k_r + \lambda) \quad \text{Eq. 9}$$

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

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

$$367 \quad F_r = I_r k_r \quad \text{Eq. 10}$$

368 These four equations are written for  $^{137}\text{Cs}$ ,  $^{210}\text{Pb}_{\text{ex}}$  and  $^7\text{Be}$  considering the partitioning coefficients of the  
 369 radionuclides between water and suspended matter, none of which were addressed in the former version of  
 370 Dominik et al. (1987). In order to solve these equations, the following assumptions based on the  $\lambda$  values are  
 371 made: 1) the radionuclide residence time in the soil compartment is expected to be of the order of decades to  
 372 centuries, therefore, most of the  $^7\text{Be}$  will be lost by decay implying its export from the soil compartment is  
 373 negligible that is:  $k_s \ll k_{\text{Be}}$  and  $I_{\text{sBe}} k_s \ll S_r F_{\text{aBe}}$ ; 2) the decay rate of  $^{210}\text{Pb}_{\text{ex}}$  is considered negligible compared  
 374 the export rate from the rapid compartment:  $\lambda_{\text{pb}} \ll k_r$ . It is also assumed that the duration of sediment storage  
 375 in the rapid compartment does not result in net decay of  $^7\text{Be}$  to levels below the limit of detection (Smith et al.  
 376 2014).

377 The rapid compartment area,  $S_r$ , can be determined by combining Eq. 9 and 10 based on the first assumption.

378 Then,  $S_r$  is a function of the ratio  $F_r/F_a$  for  $^7\text{Be}$  and the export rate ( $k_r = 1/\tau$ ):

$$379 \quad S_r = \left( \frac{F_r}{F_a} \right)_{\text{Be}} \left( \frac{\lambda_{\text{Be}}}{k_r} + 1 \right) \quad \text{Eq. 11}$$

380 The slow box residence time,  $\tau_s$ , is then computed by solving Eq. 7 to 9 for  $^{210}\text{Pb}_{\text{ex}}$  with the assumption that

381  $\lambda_{\text{pb}} \ll k_r$  and combining this with Eq. 11:

$$382 \quad \tau_s = \frac{\left( \frac{1}{\lambda_{\text{pb}}} \right) \left[ 1 - \left( \frac{F_a}{F_r} \right)_{\text{pb}} \right]}{\left( \frac{R_r}{R_a} \right) (1 + \lambda_{\text{Be}} \tau_r) - 1} \quad \text{Eq. 12}$$

383 where  $R_r = (F_r)_{\text{Be}} / (F_r)_{\text{pb}}$  and  $R_a = (F_a)_{\text{Be}} / (F_a)_{\text{pb}}$ .

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

$$387 \quad \tau_s = \frac{[M - (F_r)_{Cs} \tau_r]}{(F_r)_{Cs} (\lambda_{Cs} \tau_r + 1)} \quad \text{Eq. 13}$$

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

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

$$391 \quad a(\tau_r)^2 + b(\tau_r) + c = 0 \quad \text{Eq. 14}$$

392 where

$$393 \quad a = -(F_r)_{Cs} \left( \frac{R_r}{R_a} \right) \lambda_{Be} \lambda_{Pb},$$

$$394 \quad b = M \left( \frac{R_r}{R_a} \right) \lambda_{Be} \lambda_{Pb} - (F_r)_{Cs} \lambda_{Pb} \left( \frac{R_r}{R_a} - 1 \right) - (F_r)_{Cs} A \lambda_{Cs} \text{ and}$$

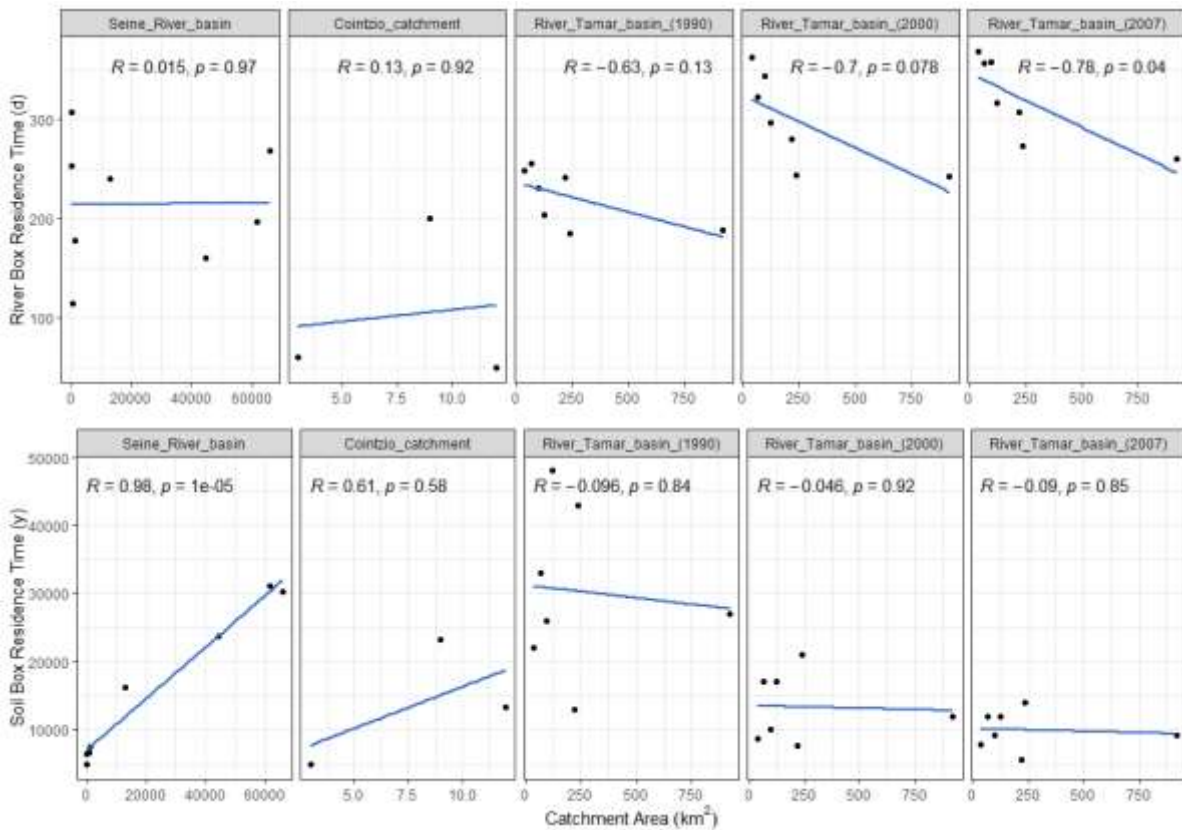
$$395 \quad c = \lambda_{Pb} M \left( \frac{R_r}{R_a} - 1 \right) - A (F_r)_{Cs}$$

396 in which

$$397 \quad A = 1 - \left( \frac{F_a}{F_r} \right)_{Pb}.$$

398 The two-box model has been applied in various catchments with catchment surface areas ranging from 7 to  
 399 65,000 km<sup>2</sup> (see Table 1) and sediment residence times ranging from 4,800 to 30,321 years in the soil box and  
 400 from 50 to 365 days in the river box. Le Cloarec et al. (2007) found a strong positive relationship between the  
 401 soil box residence time and the catchment area within the Seine basin (see Figure 5, where  $R = 0.98$ ,  $p < 0.001$ ),  
 402 while no relationship was found between the river box residence times and the catchment surface area. A  
 403 correlation analysis of residence times modelled using the two-box model in the available literature  
 404 demonstrates that this case is an exception, and we find no significant relationship between these two variables  
 405 in the literature (Figure 5). Interestingly, an inverse relationship ( $R < -0.6$ ) was found in the River Tamar basin,  
 406 UK, between river box residence time and catchment surface area. However, only the 2007 period for the Tamar

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



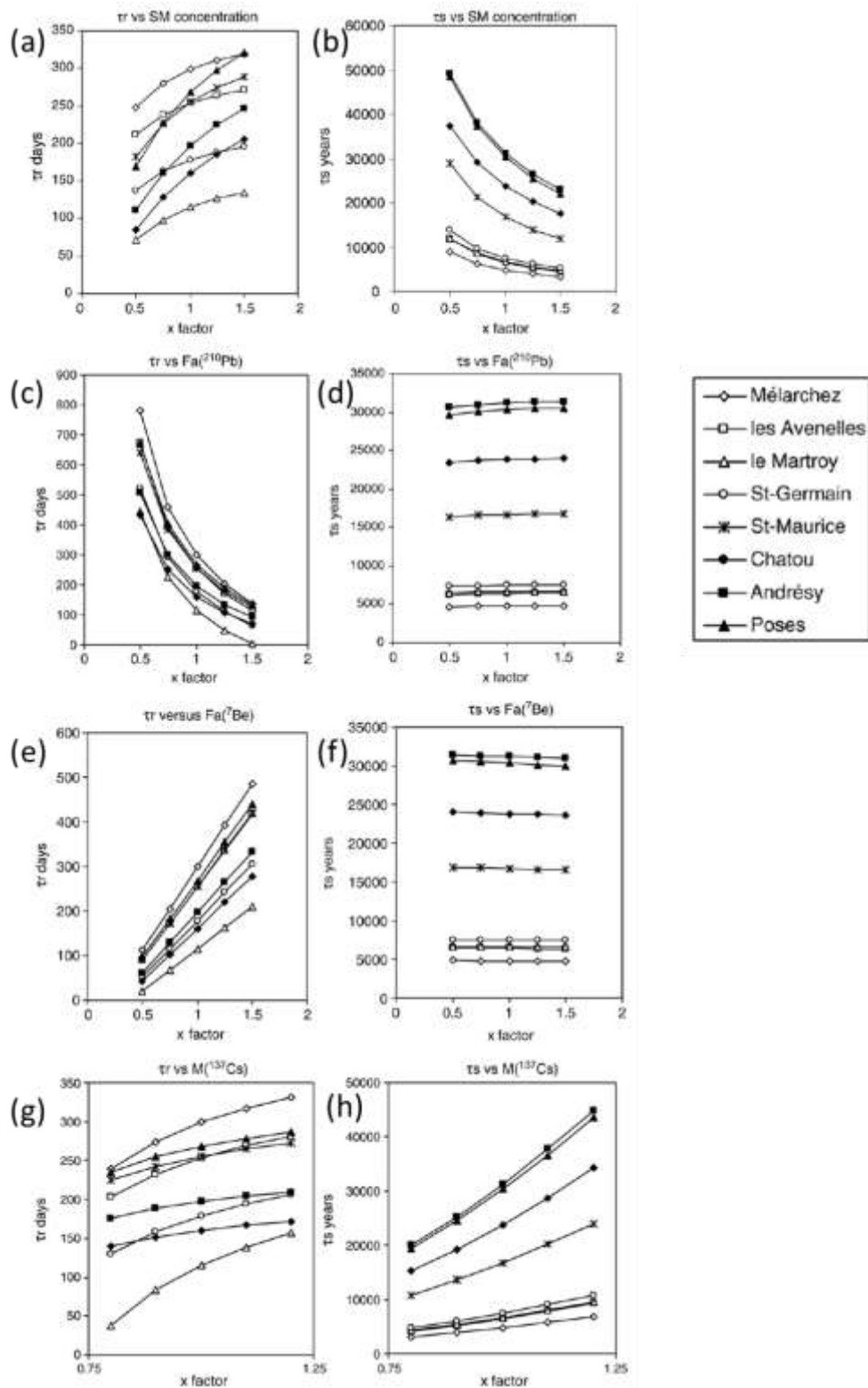
413

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

418

419 Sensitivity analysis was carried out by Le Cloarec et al. (2007) to assess the two-box model performance (Figure  
 420 6), and uncertainty simulations showed an important variation of sediment residence times for the soil and river  
 421 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  $^7\text{Be}$  and  $^{210}\text{Pb}$  (i.e.  $F_a$ ) did not significantly influence soil box residence times ( $\tau_s$ ) (Figure  
424 6d, f), whereas a notable variation was found in the river box residence times ( $\tau_r$ ) (Figure 6c, e). For example,  
425 variations in the atmospheric flux of  $^{210}\text{Pb}$  (about 1.5 times) decreased residence time by a factor of 2 in the river  
426 box. The influence of the  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  inventory, and atmospheric fluxes of  $^7\text{Be}$  and  $^{210}\text{Pb}$  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).



432

433 **Figure 6.** Sensitivity analysis of river and soil boxes residence times ( $\tau_r$  and  $\tau_s$ ), illustrating the response of modelled  
 434 residence times by changing parameters such as suspended matter (SM) concentration (a, b), atmospheric fluxes ( $F_a$ ) of  
 435  $^{210}\text{Pb}$  (c, d) and  $^7\text{Be}$  (e, f), and the  $^{137}\text{Cs}$  catchment inventory (g, h) in different sub-catchments of the Seine River basin.

436 Extracted and modified from Le Cloarec et al. (2007).

437 4.3 The  ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{ex}}$  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  ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{ex}}$  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:

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

443 where  $\lambda_{7\text{Be}}$  and  $\lambda_{210\text{Pb}}$  are the decay constants of  ${}^7\text{Be}$  and  ${}^{210}\text{Pb}$ , respectively, A and B are the activity  
444 concentrations of  ${}^7\text{Be}$  and  ${}^{210}\text{Pb}_{\text{ex}}$  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  ${}^7\text{Be}$  and  ${}^{210}\text{Pb}_{\text{ex}}$  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  ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{ex}}$  ratio in  
448 the source.

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

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

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

- 452 1) dry and wet fallout are included although  ${}^7\text{Be}$  and  ${}^{210}\text{Pb}_{\text{ex}}$  are delivered to the landscape primarily during  
453 precipitation events;
- 454 2)  ${}^7\text{Be}$  and  ${}^{210}\text{Pb}_{\text{ex}}$  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,  ${}^7\text{Be}$  and  ${}^{210}\text{Pb}_{\text{ex}}$  are assumed to be rapidly and irreversibly absorbed  
457 to particulate matter;
- 458 4)  ${}^7\text{Be}$  and  ${}^{210}\text{Pb}_{\text{ex}}$  are not partitioned differentially onto particulate matter; and
- 459 5) since radionuclide sorption behaviour of  ${}^7\text{Be}$  and  ${}^{210}\text{Pb}_{\text{ex}}$  is similar, differences along the flow path  
460 caused by particle size are eliminated by considering their ratio (including mineralogical variations).



461 The  ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{ex}}$  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  ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{ex}}$  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  ${}^7\text{Be}$  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  ${}^7\text{Be}$  and 4-fold for  ${}^{210}\text{Pb}$   
468 at different stations within the storm. Furthermore,  ${}^7\text{Be}/{}^{210}\text{Pb}$  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  ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{ex}}$  ratio, since freshly  
474 mobilised sediment will reflect the  ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{ex}}$  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  ${}^{210}\text{Pb}_{\text{ex}}$  as a  
477 result of an accumulated inventory and being exposed to fresh fallout of  ${}^7\text{Be}$ , 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  ${}^7\text{Be}/{}^{210}\text{Pb}_{\text{ex}}$  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).

487 4.4 Age of surface derived sediments

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

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

$$493 \quad ^7\text{Be}_{\text{corr}} = \frac{{}^7\text{Be}_{\text{target}} - \left[ {}^7\text{Be}_{s_1} \left( \frac{\%s_1}{100} \right) + \dots + {}^7\text{Be}_{s_n} \left( \frac{\%s_n}{100} \right) \right]}{\frac{\%surf}{100}} \quad \text{Eq. 17}$$

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

$$501 \quad ^7\text{Be}_{\text{age}} = \frac{\ln\left(\frac{{}^7\text{Be}_{\text{corr}}}{{}^7\text{Be}_{\text{surf}}}\right)}{-\lambda_{7\text{Be}}} \quad \text{Eq. 18}$$

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

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

$$506 \quad {}^{210}\text{Pb}_{\text{excorr}} = \frac{{}^{210}\text{Pb}_{\text{ex target}} - \left[ {}^{210}\text{Pb}_{\text{ex } s_1} \left( \frac{\%s_1}{100} \right) + \dots + {}^{210}\text{Pb}_{\text{ex } s_n} \left( \frac{\%s_n}{100} \right) \right]}{\frac{\%surf}{100}} \quad \text{Eq. 19}$$

507 where  ${}^{210}\text{Pb}_{\text{excorr}}$  is the estimated  $^{210}\text{Pb}_{\text{ex}}$  activity in the surface;  ${}^{210}\text{Pb}_{\text{ex target}}$  is the measured  $^{210}\text{Pb}$  activity in  
 508 the target sample (e.g. suspended sediment);  ${}^{210}\text{Pb}_{\text{ex } s_1}$  is the mean activity concentration in the non-surface  
 509 derived sediment source 1;  $\%s_1$  is the percentage of contribution of the sediment source 1; and  $\%surf$  is the

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

$$512 \quad {}^{210}\text{Pb}_{ex\ age} = \frac{\ln\left(\frac{{}^{210}\text{Pb}_{ex\ corr}}{{}^{210}\text{Pb}_{ex\ surf}}\right)}{-\lambda_{210\text{Pb}}} \quad \text{Eq. 20}$$

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

516 The age of the surface-derived sediment approach was first applied by Gellis et al. (2017) using  ${}^7\text{Be}$  in a study  
517 comprising samples from 99 catchments in the Midwestern region of USA where residence times ranged from 0  
518 to 174 days in bed sediments and from 0 to 84 in suspended sediments. They also found higher concentrations  
519 of pesticides (i.e. bifenthrin and DDE) in samples from the streambed with greater proportion of surface-derived  
520 sediments and relatively young residence times (< 100 days), which gives an indication of the role that sediment  
521 sources might play in the delivery of contaminated sediment and how quickly they move through the river  
522 system. In a subsequent study carried out in the agricultural Walnut Creek catchment (Iowa, USA), modelled  
523 sediment residence times ranged from 44 – 205 days using  ${}^7\text{Be}$  and from 1 – 58 years using  ${}^{210}\text{Pb}_{ex}$  (Gellis et al.  
524 2019) (Table 1). In this study, sediment transport and storage were depicted in three boxes with three types of  
525 ages: 1) a rapid box with less than a year (based on  ${}^7\text{Be}$  results), 2) a decadal box comprising from 10 to 100  
526 years (based on  ${}^{210}\text{Pb}_{ex}$  results) and 3) a geological box from 100 to > 1,000 years (based on the literature).

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

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

## 538 5 Challenges, opportunities and future research needs.

539 The development of sedimentary models has contributed to an improved understanding of the temporal  
540 dynamics of catchment sediment storage and transport, however there are still some pitfalls that require careful  
541 consideration and further research. Most of these constraints are closely related to assumptions surrounding  
542 the behaviour of  $^7\text{Be}$  when using it as soil and sediment tracer. For instance, there is no way at present of  
543 separating out the effects of decay and dilution of  $^7\text{Be}$  activity concentrations in sediments, which raises one of  
544 the biggest limitations of the sediment residence/transit time methods when using FRNs (Matisoff et al. 2005;  
545 Fisher et al. 2010; Walling 2013; Gellis et al. 2019). The mixing of newly tagged  $^7\text{Be}$  sediment with  $^7\text{Be}$ -dead  
546 sediment (e.g. sediment from channel banks and/or sediment stored in streambeds and floodplains) is thus an  
547 issue yet to be addressed. More research is needed in this regard to quantify the uncertainty, or otherwise  
548 apportion the amount, of  $^7\text{Be}$ -depleted sediments that are stored in riverine compartments. Furthermore,  
549 because most of the  $^7\text{Be}$  activity concentration in soils is found in the top centimetre of surface soil (Blake et al.  
550 1999; Schuller et al. 2006; Walling 2013), the catchment soil may be subjected to deeper erosion that will tend  
551 to lower activity concentrations of suspended sediment which can result in overestimations of sediment  
552 residence times (Gellis et al. 2019).

553 Equally important is the direct fallout of  $^7\text{Be}$  and  $^{210}\text{Pb}_{\text{ex}}$  in wetted areas of the stream channel which may  
554 increase activity concentrations of radionuclides in suspended sediments and sediments stored in the surface of  
555 the bed channel. It has been reported that direct fallout of radionuclides on stream channels is negligible  
556 compared to sediment inputs from erosion of the catchment soils (Hancock et al. 2014). Although this  
557 assumption has been supported in catchments with enhanced overland flow and surface runoff (Evrard et al.  
558 2016; Le Gall et al. 2017), studies in forested and forest-influenced catchments have shown that channel  
559 interception of FRNs can be important (Karwan et al. 2016, 2018). Future studies should thus acquire information  
560 regarding hydrological flow paths and hillslope connectivity to address possible problems when assessing  
561 sediment residence times, or otherwise, quantify the proportion of radionuclide inputs into channels from

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

564  $^7\text{Be}$  signatures of a specific source will continue changing through time in response to radioactive decay and  
565 input of fresh fallout. It is important to recognise, therefore, that  $^7\text{Be}/^{210}\text{Pb}_{\text{ex}}$  ratios could be expected to vary  
566 both between storm events and seasonally, as well as from year to year, due to various controls on  $^7\text{Be}$  and  
567  $^{210}\text{Pb}_{\text{ex}}$  fallout (Walling 2013; Gourdin et al. 2014a). Consequently, characterisation of signature of dominant  
568 sources should be undertaken as a core component of any sediment residence time study. This poses important  
569 challenges when planning sampling and its related logistics, particularly due to the short half-life of the  $^7\text{Be}$  and  
570 the need to encompass, as much as possible, the variability of  $^7\text{Be}$  in the sediment source signature (Gourdin et  
571 al. 2014a).

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

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

586 Finally, the influence of environmental and anthropogenic factors on sediment residence time in river systems  
587 are poorly understood. Only a few studies have attempted to link and quantify the relationship between  
588 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.

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