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1 **A global review of sediment source fingerprinting research incorporating fallout radiocesium ( $^{137}\text{Cs}$ )**

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9 **Abstract**

10 Information on the main sources supplying deleterious sediment loads to river systems is needed to  
11 improve our understanding of soil erosion processes. In particular, it is important to quantify the  
12 respective contributions of surface and subsurface sources to material degrading waterbodies.  
13 Radiocesium ( $^{137}\text{Cs}$ ), emitted during thermonuclear bomb testing (~1950–1980) and nuclear  
14 accidents provides significant discrimination between surface material exposed to atmospheric  
15 fallout and subsurface material sheltered from it. A systematic worldwide review of research articles  
16 (n=123) that used  $^{137}\text{Cs}$  to trace sediment sources showed that the United Kingdom (n=24), Australia  
17 (n=23) and the United States (n=20) had the highest number of publications utilizing  $^{137}\text{Cs}$  in a  
18 sediment tracing framework. In contrast, few studies were published for catchments in Africa (n=9)  
19 or South America (n=2). In the northern hemisphere, positive relationships were evident between  
20  $^{137}\text{Cs}$  activities in surface and subsurface sources and the proportion of thermonuclear bomb fallout.  
21 However, given the low proportions of fallout received in regions between 0–20°N and 0–20°S, the  
22 potential application of  $^{137}\text{Cs}$  tracing techniques may be limited in this area of the world as well as in  
23 agricultural regions with severe soil erosion (i.e. Chinese Loess Plateau and South Africa). In total,  
24 94% of the studies researching surface and subsurface sources that analyzed  $^{137}\text{Cs}$  as a potential  
25 tracing property included this parameter in end-member mixing models. The main challenges for the  
26 future of this technique are mainly related to the access to ultra-low background gamma  
27 spectrometry facilities and the development of surrogate tracers. Future research should ensure that  
28 basic catchment information and details on the sampling design are properly documented to ensure  
29 studies are reproducible. Moreover, researchers should provide raw measurement data to help  
30 improve our global understanding of  $^{137}\text{Cs}$  dynamics in soil erosion research.

31

32 **Keywords:** watersheds; soil erosion; sediment tracing; fallout radionuclides.

33

34 **Highlights**

- 35
- 36 • The meta-analysis compiled 123 articles considering  $^{137}\text{Cs}$  for sediment tracing
  - 37 • 70% of these studies were conducted in Europe, North America and Australia
  - 38 • Fallout levels are too low in regions near the Equator and with extensive erosion
  - 39 •  $^{137}\text{Cs}$  was included in 71 to 94% of studies modelling source contributions
  - 40 • Future research should systematically include details on sampling design

41 **1. Introduction**

42 Exponential population growth and the concomitant expansion and intensification of our global  
43 anthropogenic footprint has resulted in an unsustainable and non-renewable loss of soil resources  
44 threatening United Nations Sustainable Goals (e.g. food security) (Keesstra et al., 2016). To limit soil  
45 loss, best management practices need to be applied to landscapes degraded by accelerated soil  
46 erosion processes generating significant sediment loads that impair downstream aquatic systems  
47 (Owens et al., 2005). Accordingly, identifying the main areas of soil erosion is a prerequisite to  
48 improve our knowledge of the main factors driving soil erosion processes and guide the  
49 implementation of best management practices.

50 One approach that effectively identifies areas with elevated soil erosion rates is the sediment tracing  
51 or sediment source fingerprinting technique (Haddadchi et al., 2013; Owens et al., 2016; Walling,  
52 2013). This technique is increasingly used around the world to quantify sediment source  
53 contributions based on the measurement of biogeochemical and/or physical properties in both soils  
54 and sediment through the use of statistical procedures and un-mixing models (Haddadchi et al.,  
55 2013; Koiter et al., 2013b; Walling, 2005). Effective sediment tracer properties, or fingerprints, must  
56 significantly differentiate between potential sediment sources and behave conservatively during  
57 erosion, transport and deposition processes, or vary in a predictable and measurable way (Koiter et  
58 al., 2013b; Laceby et al., 2017; Smith and Blake, 2014). Although a variety of tracing parameters have  
59 been utilized in sediment source fingerprinting research (e.g. element geochemistry, colour  
60 parameters, magnetic properties, organic matter parameters), fallout radionuclides that originate  
61 from the atmosphere and are quickly and strongly bound to fine particles provide a notably powerful  
62 tool to investigate soil erosion sources and sediment transfer processes (Mabit et al., 2008).

63 In particular, caesium-137 ( $^{137}\text{Cs}$ ;  $T_{1/2} = 30 \text{ y}$ ) is often used to identify whether sediment has been  
64 generated by surface or subsurface erosion processes. This fallout radionuclide was mainly supplied  
65 to exposed soils during atmospheric thermonuclear bomb testing between the 1950s and the 1970s,  
66 with a peak in 1963 and 1965 in the northern and southern hemispheres, respectively (Arnaud et al.,  
67 2006; Cambray et al., 1989; Turney et al., 2018). Accordingly, surface soils exposed to atmospheric  
68 fallout are typically enriched in  $^{137}\text{Cs}$ , while subsurface soils (e.g. gullies and channel banks) that were  
69 sheltered from direct fallout are typically depleted in  $^{137}\text{Cs}$  (Campbell et al., 1985; Loughran et al.,  
70 1982; Peart and Walling, 1988).

71 In the 1980s, researchers investigated the utility of using  $^{137}\text{Cs}$  to discriminate between potential  
72 sediment sources in Australia (Campbell et al., 1985; Loughran et al., 1982) and in the UK (Peart and  
73 Walling, 1988). As  $^{137}\text{Cs}$  is concentrated near the soil surface, sediments eroded from rill or sheet  
74 erosion processes typically have  $^{137}\text{Cs}$  concentrations that are higher than sediments eroded from  
75 subsurface erosion processes, with erosion on cultivated soils plotting between these two potential

76 source end-members (He and Walling, 1995; Wallbrink and Murray, 1993). Walling and Woodward  
77 (1992) were the first to use un-mixing models demonstrating the potential to capitalize on the  
78 consistent differences of  $^{137}\text{Cs}$  in these potential source soils and quantify their contributions to  
79 sediment sampled in waterways.

80 Several review papers have recently been published on sediment tracing techniques in general  
81 (Haddadchi et al., 2013; Koiter et al., 2013b; Owens et al., 2016; Walling, 2013), or on fallout  
82 radionuclides in particular (Mabit et al., 2014; Matisoff, 2014; Taylor et al., 2013). However, these  
83 reviews have not provided a structured and comprehensive worldwide synthesis of  $^{137}\text{Cs}$  utilized as a  
84 tracer in sediment source fingerprinting research. Accordingly, this literature review was based on a  
85 Web of Science search to provide structure to our meta-analysis of articles incorporating  $^{137}\text{Cs}$  in  
86 sediment fingerprinting research to help facilitate the future application of this tracer worldwide. In  
87 particular, the advantages and drawbacks associated with the use of  $^{137}\text{Cs}$  in sediment source  
88 fingerprinting research are highlighted and perspectives for future research investigating sediment  
89 source contributions with  $^{137}\text{Cs}$  are outlined. Overall, this fallout radionuclide has significant potential  
90 to help guide targeting best management practices to reduce the effects of accelerated soil erosion  
91 worldwide.

## 92 **2. Literature overview**

93 Journal articles using cesium-137 published in the English language were pulled from the Web of  
94 Science up to 30 August 2019. The search keywords ‘cesium-137’, ‘ $^{137}\text{Cs}$ ’, ‘fallout radionuclides’,  
95 ‘sediment’ and ‘tracing’ or ‘fingerprinting’ were used in isolation and/or in combination.

### 96 *2.1 Study areas*

97 In total, the database comprised investigations conducted in 215 catchments from 123 journal  
98 articles. Studies conducted in the UK represented almost one fifth (19%) of the articles in the  
99 database. The large contribution of the British studies is likely explained by the pioneer work initiated  
100 by the research team of Professor Desmond Walling at the University of Exeter, and his former PhD  
101 students who continued in research after their theses.

102 Overall, studies conducted either in Europe or in North America constituted 52% of the articles. In  
103 North America, the work conducted in the US clearly dominates (representing 80% of the articles in  
104 this region). In Europe, outside of the UK, the countries with the largest number of articles were  
105 Spain (n=8) and France (n=6). Research carried out in Asia contributed to 20% of the articles (n=25),  
106 with the highest number of articles published in Japan (n=5) and China (n=5). A significant number of  
107 articles were published in Australia (n=23; 19% of the world total), including several papers that  
108 provided methodological background on the use of fallout radionuclides as fingerprints (Murray et  
109 al., 1993; Olley et al., 1993; Olley et al., 1996; Wallbrink et al., 1999). Although soil erosion is  
110 widespread in Africa and South America (Anache et al., 2017; Vanmaercke et al., 2014), very few

111 studies were conducted on these continents (Africa 7%, n=9; South America 2%, n=2). When  
112 mapping locations where  $^{137}\text{Cs}$  was measured (Fig. 1), a clear dominance of research conducted in  
113 the US, Europe and along the eastern coast of Australia is demonstrated.

## 114 *2.2. Research Design*

115 Most papers (93%) provided summary information on their catchments or study locations. For those  
116 catchments where the surface area was provided, the mean surface area was 3859 km<sup>2</sup> (standard  
117 deviation (SD) 17,630 km<sup>2</sup>), with this relatively high value being impacted by the large surface area of  
118 several studies in Australia (up to 130,000 km<sup>2</sup>).

119 The study design was strongly variable with the source numbers ranging between six and 893 (mean:  
120 123; SD: 150; median: 70), corresponding to a mean source density of 47 samples per km<sup>2</sup>. Although  
121 the most common sampling procedure was the collection of the uppermost 0–2 cm soil layer (n=39  
122 articles), multiple strategies were employed including sampling the 0–5 cm (n=25), 0–1 cm (n=9), 0–  
123 10 cm (n=4) or 0–2.5 cm (n=2) of surface soils. Sediment collected on hillslopes was used as a  
124 surrogate for source material in six studies. This technique may be useful to facilitate the direct  
125 comparison of source and sediment characteristics. Soil profiles were used to characterise sources in  
126 nine articles. Unfortunately, information on the source sampling depth was not provided in a large  
127 number of studies (n=34, 28%).

128 Sediment samples mainly consisted of suspended matter (n=31 articles), core(s) collected in lakes,  
129 reservoirs or floodplains (n=18), bed material (n=18), material collected in sediment traps or ‘time-  
130 integrated samplers’ (n=14) often based on the design proposed by Phillips et al. (2000), lag deposits  
131 (n=6) or a combination of a least two of the previous options (n=14). Of note, information on the  
132 type of sediment collected was not mentioned in 22 studies (18%). The number of sediment samples  
133 analysed strongly varied among studies, from four to 345 samples (mean: 48; SD: 61; median: 28). Of  
134 note, this information was not provided in 40 studies (33%).

135 Regarding particle size, the < 63 µm fraction was by far the most common (n=57 articles), followed by  
136 the < 2 mm fraction (n=28). A limited number of studies analysed the <10 µm fraction (n=9). Of note,  
137 among these nine studies, eight were carried out in Australian catchments. Only three studies  
138 investigated multiple particle size fractions. Occasionally, other particle size thresholds were used  
139 (<200 µm, n=2; <250 µm, n=2). Information on the particle size fraction analysed was not provided  
140 for 22 studies (18%).

141 Overall, the number of studies analysing  $^{137}\text{Cs}$  in tracing research in order to quantify sediment  
142 source contributions increased from only 1.2 articles per year between 1988–1998 to seven articles  
143 per year between 2009–2019 (Fig. 2) demonstrating the increasing popularity of this technique.

144

### 3. Spatial patterns of <sup>137</sup>Cs deposition across the world

The deposition of <sup>137</sup>Cs reached a peak in 1963 and 1965 in the northern and southern hemispheres, respectively (Fig. 3). Fallout became negligible after 1980 when China stopped testing nuclear weapons. There are localized exceptions, including most regions of Europe (Evangelidou et al., 2016) affected by the 1986 Chernobyl fallout and several Prefectures in Japan that received significant fallout following the Fukushima Dai-ichi Nuclear Power Plant accident in 2011 (Saito and Onda, 2015). However, even in areas affected by accidental fallout, this additional <sup>137</sup>Cs supply did not fundamentally affect the ability of this radionuclide to discriminate surface sources, enriched in <sup>137</sup>Cs, and subsurface sources, sheltered from the main fallout (Evrard et al., 2016).

Data available from the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) demonstrate that the majority of the fallout occurred in the Northern Hemisphere (~77%), relative to the Southern Hemisphere (~23%)(UNSCEAR, 2000). The results of long-term monitoring programmes of global fallout deposition were compiled to estimate the proportion of global fallout through worldwide networks operated by the Environmental Measurements Laboratory (EML) in the United States (Health and Safety Laboratory, 1977). Deposition densities at individual sites were averaged out for each 10-degree latitude band and multiplied by their area to obtain global deposition amounts. However, a significant proportion of data is lacking, and this spatial distribution should therefore be taken with caution (Fig. 4).

To compare surface and subsurface <sup>137</sup>Cs activities, data available from articles compiled in the current research were decay-corrected to 1 January 2019 (based on the sampling/reference date when it was provided or based on the 5-yr mean duration calculated in the current research between sampling and publication). To provide a worldwide basis for comparison of those activities, the mean <sup>137</sup>Cs activities reported for cultivated topsoil versus subsurface sources (i.e., gullies, badlands and channel banks) were grouped in 10° latitudinal bands and compared to the proportion of fallout received in each band after UNSCEAR (2000). Overall, a positive relationship is observed between the mean <sup>137</sup>Cs activities in surface (Fig. 5a) and subsurface (Fig. 5b) sources and the proportion of total fallout received in each latitudinal band of the Northern Hemisphere ( $0.62 < r^2 < 0.68$ ).

In the southern hemisphere, a positive relationship between the mean activities in the topsoil sources (Fig. 5c) or the subsurface sources (Fig. 5d) and the proportion of fallout received is also found from the Equator to the southern latitude. However, given the low quantity of data compiled, these relationships should be taken with caution. The higher uncertainties are likely due to the lower quantity of data available and their concentration between 0°-10°S and 30°-40°S in the southern hemisphere where 55 and 51% of missing data were calculated, respectively (Fig. 4).

In both hemispheres, a positive relationship was found between the mean <sup>137</sup>Cs activities in surface/subsurface sources and the increasing latitudes. However, the relatively large dispersion of

180 values found at similar latitudes may prevent the direct use of these relationships for characterizing  
181  $^{137}\text{Cs}$  activities in surface and subsurface sources (Fig. 6). The variations observed at similar latitudes  
182 may be explained by local factors (e.g. slope, orientation) and differences of land uses (cultivated vs.  
183 uncultivated) or farming practices (e.g. tillage vs. no-tillage) leading to varying levels of soil erosion.

184 This data clearly demonstrates that in 2019,  $^{137}\text{Cs}$  activities are sufficiently high in surface sources in  
185 areas located above  $20^\circ\text{N}$  and below  $20^\circ\text{S}$  to be relatively easily detectable with low-background  
186 gamma spectrometry equipment (Fig. 6). In contrast, for most areas located around the Equator (i.e.  
187 between  $20^\circ\text{N}$  and  $20^\circ\text{S}$ ),  $^{137}\text{Cs}$  activities in surface soils are likely to be very low (often  $< 2 \text{ Bq kg}^{-1}$ )  
188 and may therefore be increasingly difficult to differentiate from subsurface sources, which are  
189 generally lower than  $1.5 \text{ Bq kg}^{-1}$ .

#### 190 **4. Spatial applicability of $^{137}\text{Cs}$ based-tracing across the globe**

191 Although most of the studies using  $^{137}\text{Cs}$  were conducted in North America, Europe and along the  
192 eastern coast of Australia, evidence exists in the literature to outline areas where there is potential  
193 to use this radionuclide in sediment tracing studies and regions where  $^{137}\text{Cs}$  concentrations are not  
194 sufficient and/or will soon become too low to conduct this type of research. As  $^{137}\text{Cs}$  is preferentially  
195 bound to the finer particle size fractions (He and Walling, 1995), the method should be applied at  
196 locations where sediment contains significant amounts of clay and silt.

197 In the Northern Hemisphere, where 77% of the total fallout is estimated to have occurred (UNSCEAR,  
198 2000), this technique should be applicable to most regions of Europe and North America. In the  
199 studies reviewed, no problems were reported for the detection of  $^{137}\text{Cs}$  in these regions.  
200 Furthermore, the  $^{137}\text{Cs}$ -based technique to discriminate between surface and subsurface sources  
201 should be applicable in areas exposed to Chernobyl fallout, as subsoils were sheltered from  
202 atmospheric fallout (Belyaev et al., 2013). As the  $^{137}\text{Cs}$  technique has been extensively applied in  
203 Europe and North America, our review here will focus on contexts where there has been less  
204 application of this tracing technique.

205 In Asia, a variety of environmental contexts may complicate the utilization of  $^{137}\text{Cs}$  to trace surface  
206 and subsurface sources. On China's Loess Plateau, soil erosion is so extensive that Li et al. (2017)  
207 used  $^{137}\text{Cs}$  to discriminate between sediment derived from gully areas (activities  $\sim 1.5 \text{ Bq kg}^{-1}$ ) and  
208 inter-gully areas (activities  $\sim 0 \text{ Bq kg}^{-1}$ ). These results demonstrate that  $^{137}\text{Cs}$  will soon no longer be  
209 detectable in this type of environment exposed to very high erosion levels. In contrast, significantly  
210 higher  $^{137}\text{Cs}$  activities were detected in the black soil region of Northeast China, both in cultivated  
211 land (mean:  $2.0 \text{ Bq kg}^{-1}$ ) and in gully areas (mean  $0.3 \text{ Bq kg}^{-1}$ ), despite the fact that this region was  
212 also exposed to high soil degradation over the last several decades. Similar  $^{137}\text{Cs}$  activities ( $\sim 2 \text{ Bq kg}^{-1}$ )  
213 were detected in a catchment of the Sichuan Province (Tang et al., 2014) and the surface and

214 subsurface tracing method was also demonstrated to be applicable in the Upper Yangtze River basin  
215 (Long et al., 2012).

216 In Japan,  $^{137}\text{Cs}$  may still be used to discriminate between different sources of sediment, even after  
217 the Fukushima Dai-ichi Nuclear Power Plant accident in low-fallout areas (Ochiai et al., 2015). The  
218 shorter-lived  $^{134}\text{Cs}$  isotope ( $t_{1/2} = 2 \text{ y}$ ) is also detectable in this post-accidental context, which may  
219 provide discrimination between the global fallout  $^{137}\text{Cs}$  and the additional  $^{137}\text{Cs}$  Fukushima fallout, as  
220 the  $^{134}\text{Cs}$  deposited after the thermonuclear bomb tests conducted in the 1960s is no longer  
221 detectable. Background information on  $^{137}\text{Cs}$  activities prevailing in Japanese soils before the  
222 Fukushima Dai-ichi Nuclear Power Plant accident is available in the literature as sediment tracing  
223 techniques based on fallout radionuclides were conducted in forested (Fukuyama et al., 2010) and  
224 agricultural (Ahn et al., 2009) catchments in Japan before 2011. The only other region of the world  
225 where significant levels of  $^{134}\text{Cs}$  (and hence of extra  $^{137}\text{Cs}$ ) originating from Fukushima accident were  
226 reported is Hawaii, although this  $^{134}\text{Cs}$  fallout will rapidly decay to undetectable levels (McKenzie and  
227 Dulai, 2017).

228 In other Asian countries, sediment sources collected in a catchment in South Korea discriminated  
229 between channel banks (mean  $^{137}\text{Cs}$  activities of  $2.2 \text{ Bq kg}^{-1}$ ) and forest soil sources (mean  $13.5 \text{ Bq kg}^{-1}$ )  
230 (Lim et al., 2014). Another successful approach was conducted in a forested catchment of South  
231 Korea (Kim et al., 2013). The applicability of the approach in Taiwan was also demonstrated in a  
232 study conducted in a catchment covered with tea plantations (Zehetner et al., 2008). In contrast, in a  
233 catchment of Central Thailand,  $^{137}\text{Cs}$  activities were systematically  $< 1 \text{ Bq kg}^{-1}$  for all sources (i.e.,  
234 uncultivated, cultivated and pasture soils)(Srisuksawad et al., 2015). These results differed with those  
235 obtained in a study conducted in Eastern Indonesia, where surface (mean:  $1.9 \text{ Bq kg}^{-1}$ ) and  
236 subsurface ( $< 0.3 \text{ Bq kg}^{-1}$ ) source contributions could be discriminated based on their  $^{137}\text{Cs}$  fallout  
237 activities (Hobgen et al., 2014). Surprisingly, only two studies using  $^{137}\text{Cs}$  for tracing sediment source  
238 contributions were found in India (Froehlich, 2004; Froehlich and Walling, 2006).

239 In the Southern Hemisphere, where less fallout is estimated to have occurred (~23%) (UNSCEAR,  
240 2000), much less information is available regarding areas where the method is applicable. In Africa,  
241 this technique was shown to be applicable in Tunisia (Ben Slimane et al., 2013) as well as in Burkina  
242 Faso (Rode et al., 2018). This suggests that the technique can be used in the Maghreb and Sahel  
243 countries, at least in areas where soils are not too sandy. In addition, sufficient  $^{137}\text{Cs}$  levels were  
244 found in Zambia despite this country being located at a relatively low latitude (~16°S) (Walling et al.,  
245 2001).

246 Depending on the region, numerous areas in the Southern Hemisphere had high levels of soil  
247 degradation associated with the arrival of European settlers (i.e. from the early 19<sup>th</sup> century in South  
248 Africa), which likely led to the rapid erosion of the upper soil layers tagged with  $^{137}\text{Cs}$ . In South Africa,

249 Foster et al. (2017) measured low and highly variable  $^{137}\text{Cs}$  contents ( $2 \pm 2 \text{ Bq kg}^{-1}$ ) in topsoil,  
250 particularly in catchments having variable soil cover on steep rocky hillslopes. The difficulty in using  
251  $^{137}\text{Cs}$  as fingerprint was also demonstrated by van der Waal et al. (2015) who detected  $^{137}\text{Cs}$  in only  
252 50% of 28 surface soil samples collected from a small catchment in the Drakensberg mountain range  
253 region.

254 In contrast, in South America, higher levels of fallout radionuclides were found in an agricultural  
255 catchment of Southern Brazil (Minella et al., 2014) and in forested catchments of Chile (Schuller et  
256 al., 2013). The extremely low number of studies ( $n=2$ ) considering  $^{137}\text{Cs}$  as a potential source  
257 fingerprint in South America shows the large development potential existing in this region for the  
258 future. This is particularly true in those regions located below  $20^\circ\text{S}$  (i.e. Argentina, Chile, Paraguay,  
259 Uruguay and the southernmost part of Brazil) where  $^{137}\text{Cs}$  activities exceeding  $2 \text{ Bq kg}^{-1}$  should be  
260 widely found in surface sources.

261 In Oceania, this technique was mainly applied to catchments ( $n = 40$ ) located along the eastern coast  
262 of Australia (Krause et al., 2003; Olley et al., 2013b; Wilkinson et al., 2013). Significant and detectable  
263  $^{137}\text{Cs}$  activities were found in surface sources, including managed or unmanaged forests, grazed or  
264 unmanaged pastures, cropland and bushland, which were easily differentiated from subsurface  
265 sources including gullies and channel banks that were totally depleted in  $^{137}\text{Cs}$ . One exception was a  
266 mining catchment of New Caledonia, South Pacific Islands, where the very low  $^{137}\text{Cs}$  activities  
267 measured in mining and non-mining tributaries indicated the dominance of subsurface sources in the  
268 sediment contribution in this region. Given much higher activities were measured at similar latitudes  
269 in Australia, these low activities likely reflect the intensity of soil erosion processes and the  
270 dominance of subsoil erosion on this island (Sellier et al., 2019).

## 271 **5. Applications of $^{137}\text{Cs}$ in sediment source fingerprinting research**

272 Fifty studies (41%) used  $^{137}\text{Cs}$  to discriminate between surface and subsurface contributions (Table 1).  
273 In contrast, 49 studies (40%) used  $^{137}\text{Cs}$  to discriminate between land use contributions to sediment  
274 (Table 2). Other approaches investigated the contributions of sub-basins or tributaries ( $n=4$ ), areas  
275 with different geologies ( $n=3$ ), or different erosion processes (gully vs. rill or sheet erosion;  $n=2$ ).  
276 Finally, 13 studies (11%) examined multiple combinations of these approaches (Table 3).

### 277 *5.1 Surface versus subsurface erosion*

278 Subsurface sources generally included channel banks ( $n=27$  studies) and/or gullies ( $n=22$ ). Much less  
279 often, landslides ( $n=3$ ), unpaved roads ( $n=2$ ), or construction sites ( $n=2$ ) were also considered as a  
280 dominant subsurface source (Table 1). In addition to  $^{137}\text{Cs}$ , researchers often utilized other  
281 environmental radionuclides (ERNs) to discriminate between surface and subsurface sources,  
282 including:  $^{210}\text{Pb}_{\text{xs}}$  ( $n=32$  studies),  $^7\text{Be}$  ( $n=11$ ),  $^{226}\text{Ra}$  ( $n=5$ ),  $^{232}\text{Th}$  ( $n=4$ ),  $^{40}\text{K}$  ( $n=4$ ; Table 4). Plutonium

283 isotopes were also measured in three articles, with different isotopes or ratios analysed  
284 ( $^{238}\text{Pu}/^{239+240}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{239+240}\text{Pu}$ ). In Japan, one study also detected  $^{134}\text{Cs}$  emitted during the Fukushima  
285 Dai-ichi Nuclear Power Plant accident (Ochiai et al., 2015). Belmont et al. (2014) also examined  $^{10}\text{Be}$   
286 concentrations to take into account the long-term deposition and remobilisation of sediment  
287 between the floodplain and the main river channel.

288 Often, researchers only use  $^{137}\text{Cs}$  to discriminate between surface and subsoil sources. In many  
289 studies,  $^{137}\text{Cs}$  alone provided powerful discrimination between sediment originating from subsurface  
290 areas or from surface material. For instance, from a catchment in Oklahoma State (US), 12-fold  
291 higher levels of this radionuclide were found in 'overland' material compared to samples collected in  
292 gully areas (Zhang et al., 2016).

293 However, several small-scale studies conducted in the early 1990s in Australia showed that the  
294 combination of several radionuclide measurements (i.e.  $^7\text{Be}$ ,  $^{210}\text{Pb}_{\text{xs}}$  and  $^{137}\text{Cs}$ ) provided additional  
295 information to distinguish between different erosion processes (e.g. sheetwash vs. rill erosion) based  
296 on their different depths of penetration or their mixing in the soil (Wallbrink et al., 1999). This  
297 approach was further developed to be effective at the catchment scale. For instance, the  
298 combination of the three fallout radionuclides was used to compare the effects of tillage vs. no-  
299 tillage management of cropland on soil erosion and sediment yields in the US, because these  
300 different farming practices lead to different penetration/mixing depths of the radionuclides in the  
301 soil (Matisoff et al., 2002). In Australia, Hancock et al. (2014) showed that the use of  $^7\text{Be}$  provided a  
302 way to discriminate between vertical (i.e. channel banks, depleted in  $^7\text{Be}$ ) and horizontal (i.e., rills,  
303 scalds, gully floors, enriched in  $^7\text{Be}$ ) subsoil erosion sources.

304 In addition to ERNs, the most commonly used tracing properties were elemental geochemistry (n=6  
305 studies), total carbon and/or nitrogen (n=6), magnetic properties (n=5), total phosphorus (n=3),  $\delta^{15}\text{N}$   
306 and  $\delta^{13}\text{C}$  (n=2), and  $^{87}\text{Sr}/^{86}\text{Sr}$  (n=1). From the 50 articles examining the surface vs. subsurface  
307 contributions to sediment, 18 studies did not use mixing models. Among the 32 studies that did apply  
308 a source apportionment model, only two articles did not include  $^{137}\text{Cs}$  in the final tracer combination  
309 included in the mixing models.

## 310 *5.2 Soil erosion on different land uses*

311 Articles discriminating between land use contributions to sediment (n=49) mainly focused on the  
312 material supplied by cropland/cultivated land (n=40), channel banks (n=34), forests (n=24) and  
313 grassland/pasture (n=22). Other subsurface sources included gullies (n=11), roads (n=11) and  
314 badlands (n=4) along with other surface sources included shrubland (n=4) and rangeland (n=3) (Table  
315 2).

316 To discriminate this variety of sources, a number of potential tracing properties were examined. In  
317 addition to  $^{137}\text{Cs}$ , other ERNs considered were  $^{210}\text{Pb}_{\text{xs}}$  (n=30 articles),  $^{226}\text{Ra}$  (n=16),  $^7\text{Be}$  (n=7),  $^{40}\text{K}$  (n=8),  
318  $^{232}\text{Th}$  (n=6)(Table 2). Regarding the other types of properties examined, elemental geochemistry was  
319 the most frequent (n=33) followed by total carbon and nitrogen (n=22), total phosphorus (n=13),  
320 magnetic properties (n=10) and phosphorus fractions (n=3). Only one study analysed  $\delta^{15}\text{N}$   
321 (Mukundan et al., 2010) in conjunction with  $^{137}\text{Cs}$ . Only five studies remained descriptive and did not  
322 apply an un-mixing model to quantify the land use contributions to sediment. Among those that  
323 modelled the sediment source contributions, 35 articles (71%) included  $^{137}\text{Cs}$  in the optimal tracer  
324 combination in at least one of the investigated catchments.

325 Interestingly, the use of  $^{137}\text{Cs}$  was not restricted to the discrimination between surface and  
326 subsurface sources, and it sometimes provided further discrimination between other land use types  
327 such as scrubland enriched in this radioisotope compared to forest areas depleted in  $^{137}\text{Cs}$ ,  
328 depending on the magnitude of erosion processes occurring in these potential sources (Palazón et  
329 al., 2015). Of note, the  $^{137}\text{Cs}$  signature of different land uses may change with time in response to the  
330 soil erosion processes. In Laos, the replacement of slash-and-burn traditional cultivation with  
331 commercial teak plantations on steep hillslopes generated so much soil erosion, that the  
332 corresponding source signature switched from a typical surface soil  $^{137}\text{Cs}$  activity (mean:  $2.5 \text{ Bq kg}^{-1}$ )  
333 to that of a subsurface soil (mean:  $1.1 \text{ Bq kg}^{-1}$ ) in less than 20 years (Ribolzi et al., 2017).

334 The impact of changes in human practices on soil erosion may also be reflected by fallout  
335 radionuclide measurements. For instance, the reduction of cattle grazing pressure in Australian  
336 catchments was demonstrated to have led to a decline in both surface and subsurface soil erosion  
337 rates (Wilkinson et al., 2013). In another context, among other properties,  $^{137}\text{Cs}$  also contributed to  
338 estimate the impact of forestry operations including clearcutting, tree harvest and replantation on  
339 sediment source contributions in catchments of Chile (Schuller et al., 2013).

### 340 *5.3 Impacts of Roads on Soil Erosion*

341 The role of unsealed or unpaved roads has been investigated in 15 articles across several regions of  
342 the world, including Australia (n=3), Brazil (n=1), Burkina Faso (n=1), France (n=1), India (n=1), Iran  
343 (n=1), Japan (n=1), Mexico (n=1), South Korea (n=1), the UK (n=1) and the US (n=2) (Table 2). The  
344 road contribution to sediment can reach 20 to 500 times the areal contributions from forests (Motha  
345 et al., 2004; Motha et al., 2003) as road construction may rapidly accelerate soil erosion. For  
346 instance, the building of a highway in Oahu, Hawaii, was shown to significantly increase the fine  
347 sediment supply to the river (Hill et al., 1998). Soil erosion was also shown to be particularly intense  
348 on the truck trail network, depleted in fallout radionuclides, in Japanese cypress plantations  
349 (Mizugaki et al., 2008). In a catchment of Iran, it was shown that  $^{137}\text{Cs}$  provided discrimination  
350 between channel bank (mean:  $2.7 \text{ Bq kg}^{-1}$ ) and road cuttings (mean:  $1.0 \text{ Bq kg}^{-1}$ ). This finding was

351 mainly attributed to the fact that channel banks likely consisted of a mixture of surface and  
352 subsurface material originating from upstream catchment locations (Nosrati et al., 2018).

### 353 *5.5 Impacts of Wildfires*

354 The use of  $^{137}\text{Cs}$  (n=6) and plutonium isotopes (n=1) was particularly helpful in catchments exposed  
355 to wildfires, as other potential tracers including magnetic and geochemical properties were affected  
356 by variable wildfire modifications in burned soils (Blake et al., 2006) (Table 1). Furthermore,  $^{210}\text{Pb}_{\text{xs}}$   
357 was excluded from the list of the potential tracers because of its high apparent sensitivity to the  
358 reduction in ash content in channel deposits compared to hillslope material (Smith et al., 2012). In  
359 burned soils, increases in the concentration of  $^{210}\text{Pb}_{\text{xs}}$  and  $^{137}\text{Cs}$  were widely found in response to the  
360 combustion of organic matter and the redistribution of radionuclides to surface soil and ash  
361 (Wilkinson et al., 2009), with a stronger effect for  $^{210}\text{Pb}_{\text{xs}}$  for which ~30% of the total inventory may  
362 be found in the surface litter of eucalyptus forests. In contrast, in unburned soils,  $^{137}\text{Cs}$  is mainly  
363 associated with mineral soil due to bioturbation transferring decayed litter into the soil following the  
364 cessation of  $^{137}\text{Cs}$  fallout in the mid-1970s (Wilkinson et al., 2009).

365 However, in the framework of a rainfall experiment conducted after a major forest fire in Los Alamos,  
366 New Mexico (US), Johansen et al. (2003) found  $^{137}\text{Cs}$  concentrations that were 40 times higher in ash  
367 deposits and three times higher in the topmost 5 cm of soil compared to pre-burned soils.  
368 Concentration of  $^{137}\text{Cs}$  in sediment was also observed when soil erosion occurred, although the levels  
369 declined rapidly with time and returned to the pre-fire levels after ~250 mm of cumulative rainfall.  
370 Smith et al. (2013) provided a comprehensive review showing the techniques that may be used to  
371 discriminate fine sediment sources in burned catchments, with numerous examples taken in North  
372 America and in Australia. Additional sediment tracing approaches have since been conducted in  
373 catchments exposed to fires in other regions of the world including the Mediterranean region (e.g.,  
374 the Balearic Islands, Spain). The occurrence of higher fallout radionuclide levels in burned material  
375 compared to unburned sources was also observed in these regions (Estrany et al., 2016; García-  
376 Comendador et al., 2017).

### 377 *5.6 Use of fallout radionuclides in tile-drained catchments*

378 Another type of environment where particularly high  $^{137}\text{Cs}$  activities were detected in source material  
379 corresponds to drained catchments (n= 3) (Table 2). In a catchment located in the centre of France,  
380 Foucher et al. (2015) used  $^{137}\text{Cs}$  measurements to demonstrate that sediment transferred in these  
381 drains originated predominantly from surface sources. They also outlined a substantial enrichment of  
382 very fine particles (~2–4  $\mu\text{m}$ ) in the drains and proposed an original correction factor based on Th  
383 concentrations estimated based on the  $^{228}\text{Th}$  activities, which are analysed along with  $^{137}\text{Cs}$  by  
384 gamma spectrometry. Similar substantial particle size differences were observed by Russell et al.  
385 (2001) in drained catchments of the UK, where they also found significant proportions of sediment

386 originating from surface sources, although they underlined that their results were constrained by the  
387 representativeness of the study periods.

### 388 *5.7 Use of fallout radionuclides in urban catchments*

389 Relatively few studies (n=6) investigated sediment source contributions in urban catchments (Table  
390 2). Among the few studies that used  $^{137}\text{Cs}$  measurements, Ormerod (1998) calculated an increase in  
391 subsurface supply to sediment in downstream direction in a study conducted in New South Wales,  
392 Australia. In the UK, Carter et al. (2003) included  $^{137}\text{Cs}$  among a suite of properties to discriminate  
393 between channel banks, cultivated or uncultivated topsoils and two urban sources (i.e road dust and  
394 solids from sewage treatment works). Devereux et al. (2010) illustrated the difficulty to define an  
395 urban signature except for the road surfaces or what was referred to as 'street residue' in a US  
396 catchment. In Laos, an increase in the contribution of subsurface material supplied by the river was  
397 also attributed to subsoil exposed to rainfall in constructions sites in suburban areas (Huon et al.,  
398 2017). Finally, in a tributary of the Seine River (France), Froger et al. (2018) quantified the  
399 contribution of 'road deposited sediment' to the material transiting this river. Relatively depleted in  
400  $^{137}\text{Cs}$  (median activity:  $0.5 \text{ Bq kg}^{-1}$ ), this source was strongly enriched in  $^{210}\text{Pb}_{\text{xs}}$  (median:  $232 \text{ Bq kg}^{-1}$ )  
401 and  $^7\text{Be}$  (median:  $402 \text{ Bq kg}^{-1}$ ) and was shown to supply significant quantities of metal contaminants  
402 originating from urban runoff to the river. These studies illustrate that there is potentially an  
403 untapped potential of  $^{137}\text{Cs}$  research in urban catchments.

## 404 **6. Main challenges associated with the use of $^{137}\text{Cs}$ as a tracer**

### 405 *6.1 Particle size*

406 In the early 1990s, Walling and Woodward (1992) already recommended the community to take into  
407 account the particle size enrichment of  $^{137}\text{Cs}$  when comparing the signatures of sources and sediment  
408 material, and suggested to conduct particle size corrections (He and Walling, 1996). Indeed, the  
409 universal use of these corrections has received critiques (Smith and Blake, 2014) and an extensive  
410 review on the challenges and the opportunities associated with the tracer particle size effect have  
411 been discussed in details (Lacey et al., 2017). In summary, it is well known that  $^{137}\text{Cs}$  is rapidly bound  
412 to fine particle size fractions and that this enrichment must be carefully taken into account when  
413 designing sediment source tracing studies.

### 414 *6.2 Heterogeneity of source signatures*

415 Often, research has demonstrated that there may be some local scale heterogeneity caused by  
416 potential fallout deposition patterns or erosion patterns. On the one hand,  $^{137}\text{Cs}$  activities were  
417 shown to vary significantly depending on the hillslope position based on a study conducted in Iran.  
418 Accordingly, significantly higher  $^{137}\text{Cs}$  activities were found on the hillslope summit compared to the  
419 shoulder position, as well as on the toeslope compared to the backslope and the shoulder. These

420 results demonstrate that when  $^{137}\text{Cs}$  is associated with other environmental radionuclides (i.e.,  $^{40}\text{K}$ ,  
421  $^{232}\text{Th}$ ) and TOC, it may provide further detailed discrimination between different hillslope  
422 compartments (Nosrati, 2017). On the other hand,  $^{137}\text{Cs}$  was used in a study investigating the spatial  
423 variability of source properties (i.e. metals, trace elements, organic matter parameters) at the field  
424 scale (Devon, UK) and provided information on the magnitude and the spatial patterns of soil  
425 redistribution within the field that may cause in turn spatial variability in the fingerprint properties  
426 (Du and Walling, 2017). This result corroborates the research of Wilkinson et al. (2015) in Australia  
427 that demonstrated how sediment contributions of surface erosion in areas affected by high erosion  
428 rates as estimated based on fallout radionuclide measurements may underestimate the topsoil  
429 supply to sediment. Accordingly, these authors recommended to conduct source sampling schemes  
430 stratified by erosion rate across the catchment, with the collection of a larger number of source  
431 samples in highly eroded areas.

### 432 *6.3 Situations where $^{137}\text{Cs}$ was not selected as a relevant tracing property*

433 Although  $^{137}\text{Cs}$  was successfully used in numerous studies, in some catchments including the suburbs  
434 of Nanjing in China where extensive soil erosion occurs and several potential subsoil material sources  
435 are found (i.e. roads, mines), the fallout radionuclide was not selected as a discriminant property  
436 (Zhou et al., 2016). In an agricultural catchment of the Canadian Prairies, high  $^{137}\text{Cs}$  concentrations  
437 were found in the top 0-20 cm layer of channel banks likely because this location corresponds to the  
438 edge of the riparian soil that contains both original atmospheric fallout and also potentially  
439 deposited material derived from upper parts of the catchment (Koiter et al., 2013a). Similar  
440 difficulties were experienced by Boudreault et al. (2018) who measured similar  $^{137}\text{Cs}$  activities in  
441 agricultural topsoil (mean:  $7.2 \text{ Bq kg}^{-1}$ ) and agricultural subsurface material (mean:  $6.2 \text{ Bq kg}^{-1}$ ) in a  
442 catchment of New Brunswick (Eastern Canada). They also attributed this unexpected result to the  
443 sampling protocol which consisted in collecting surface samples in depositional areas near the  
444 stream edge or collecting channel bank aggregate samples from the top to the bottom of the profile  
445 into a single sample.

### 446 *6.4 Finding surrogate properties for $^{137}\text{Cs}$*

447 Given the decay of  $^{137}\text{Cs}$  since the main fallout period during the last century and the difficulties to  
448 detect this radionuclide in an increasing number of regions across the globe, surrogate properties  
449 can provide discrimination of surface and subsurface sources to sediment in these environments.

#### 450 *Excess $^{210}\text{Pb}$*

451 In catchments of South Africa, Foster et al. (2007) demonstrated the potential for  $^{210}\text{Pb}_{\text{xs}}$  to provide  
452 an alternative to  $^{137}\text{Cs}$  as the latter will soon become undetectable because of the lack of inputs and  
453 its 30-yr half-life. A good relationship between both radionuclides was also found in source and

454 sediment samples collected in a catchment of Burkina Faso (Rode et al., 2018). A similar finding was  
455 also demonstrated in a catchment of Southern England by comparing  $^{137}\text{Cs}$  and  $^{210}\text{Pb}_{\text{xs}}$  activities in  
456 surface, subsurface material and sediment (Walling and Amos, 1999), although the authors outlined  
457 a greater enrichment in  $^{210}\text{Pb}_{\text{xs}}$  than in  $^{137}\text{Cs}$  in suspended sediment and channel bank material  
458 compared to subsoil material. This was attributed to a higher content of suspended sediment in  
459 organic matter and the known preferential association of  $^{210}\text{Pb}_{\text{xs}}$  with the organic fraction. Other  
460 authors attributed this enrichment in  $^{210}\text{Pb}_{\text{xs}}$  in sediment compared to sources to the direct supply of  
461 additional  $^{210}\text{Pb}_{\text{xs}}$  fallout associated with rainfall in the channel (Wallbrink et al., 2002). Olley et al.  
462 (2013b) indicated that the correlation that is often observed between  $^{137}\text{Cs}$  and  $^{210}\text{Pb}_{\text{xs}}$  activities in  
463 source samples implies that they should not be used to provide independent estimates of the  
464 relative contribution of the sources to the river sediment. The comparison of the respective  
465 proportions of  $^{137}\text{Cs}$  and  $^{210}\text{Pb}_{\text{xs}}$  fallout per latitudinal band across the globe interestingly shows that  
466 in low-latitude regions (0–20° latitude) where  $^{137}\text{Cs}$  fallout levels are very low, atmospheric  $^{210}\text{Pb}$  may  
467 provide a surrogate tracer as it is continuously deposited at significantly higher levels than the initial  
468 levels of  $^{137}\text{Cs}$  fallout (Fig. 7).

#### 469 *Other radionuclides*

470 In early sediment tracing studies,  $^{226}\text{Ra}$  was measured along with  $^{137}\text{Cs}$  to discriminate between  
471 topsoil and subsoil sources (Murray et al., 1993). Unlike most radionuclides, geogenic  $^{226}\text{Ra}$  was  
472 found to be enriched in subsoil compared to topsoil in a catchment of Southeast Australia (Wallbrink,  
473 2004). However, because of large variations in its concentrations, the use of this geogenic  
474 radionuclide in tracing has never become extensive, despite early studies outlined its potential to  
475 discriminate sediment sources at the plot scale in Southeastern Australia (Murray et al., 1993; Olley  
476 et al., 1993). In a recent review article, Alewell et al. (2014) suggested that plutonium in general and  
477  $^{239+240}\text{Pu}$  in particular may provide the next generation tracer of soil redistribution due it is longer  
478 half-life compared to  $^{137}\text{Cs}$  (Table 4) and its high measurement precision.

#### 479 *Organic matter*

480 In catchments of South Africa where alternative tracers are actively sought given  $^{137}\text{Cs}$  is often below  
481 the detection limits in topsoils across several regions, statistically significant correlations, with  $r^2$   
482 values exceeding generally 90 %, were found between  $^{137}\text{Cs}$  and Loss-on-Ignition (LOI), which was  
483 considered as a true reflection of organic matter and not C derived from bedrock or other sources  
484 (Foster et al., 2017). The good relationship found between  $^{137}\text{Cs}$  and organic matter content in  
485 potential source samples was also confirmed in a more recent study conducted in Iran (Nosrati et al.,  
486 2018). In Laos, Huon et al. (2013) showed that  $^{137}\text{Cs}$  activities and Total Organic Carbon  
487 concentrations were better correlated in the topsoil (0–10 cm) compared to the deeper soil layers  
488 (10–20 cm, 20–30 cm), illustrating that a common process – soil erosion – controlled their  
489 progressive depletion in the surface layer. Based on a similar finding, surface and subsurface sources

490 were very well discriminated by both Total Organic Carbon and  $^{137}\text{Cs}$  in several catchments of the  
491 Tunisian Ridge (Ben Slimane et al., 2016). Similarly,  $^{210}\text{Pb}_{\text{xs}}$  activities were also shown to be well  
492 correlated ( $r^2 = 0.79$ ) with LOI measurements conducted in Australia (Olley et al., 2013a). However, in  
493 another study conducted in Iran, uncertainties associated with sediment source contributions were  
494 higher when using organic matter properties than when including  $^{137}\text{Cs}$ , likely because of the low  
495 organic matter contents (mean: 0.3 – 0.4%) found in this arid region (Nazari Samani et al., 2011),  
496 which illustrates that the validity of using this surrogate property should always be verified in the  
497 local conditions prior to its application.

498

#### 499 **7. Limitations of the published $^{137}\text{Cs}$ research**

500 Only one paper provided its full dataset of raw measurement data (Le Gall et al., 2016) in the article  
501 or in their Supplementary Material. For 13 articles, no data was given at all, and only those results of  
502 statistical tests used for sediment tracing were provided. Often, a combination of graphs and tables  
503 (n=31 studies) or only graphs (n=10) was provided. In a significant number of articles (n=54), only  
504 statistics of the measurements were summarized in tables or simply referred to in the text. Fifteen  
505 studies (7%) did not even provide information on the catchment area. Interestingly, information on  
506 the source sampling depth was also not provided in a large number of articles (n=34). Of note,  
507 information on the type of sediment collected was not mentioned in 22 studies and the number of  
508 sediment samples was not even provided in 16 articles. Information on the particle size fraction of  
509 interest was not provided for 22 articles published between 1996 and 2017, with 15 of those  
510 published since 2010. As fallout radionuclides decay with time, the availability of the reference date  
511 and/or the sampling date used for calculating their activities is of particular importance. However,  
512 this information was lacking for 44 studies (36 % of the total), which may prevent the decay-  
513 correction of radionuclide activities at a later date or data comparison between studies. In the future,  
514 researchers must include enough information in their articles to make them reproducible and  
515 reviewers should ensure that this minimal standard is met. Furthermore, the provision of raw data  
516 should also be encouraged in order to facilitate the incorporation of previous results when designing  
517 new research projects and help improve our global understanding of  $^{137}\text{Cs}$  dynamics in soil erosion  
518 research.

519

#### 520 **8. Perspectives for future research**

521 With the decrease of  $^{137}\text{Cs}$  activities due to radioactive decay, the surface source activities will  
522 increasingly become closer to those found in subsurface sources. Although this situation was already  
523 observed in China's Loess Plateau, in South Africa or in mining catchments of New Caledonia (Fig. 8),

524 its more frequent occurrence in the future will increase the uncertainties associated with this  
525 method. The use of 'well' hyperpure germanium detectors installed in underground laboratories  
526 sheltered from the cosmic rays could provide a temporary solution, but the use of these facilities will  
527 not be accessible to a variety of researchers. Furthermore, the relatively high cost of gamma  
528 spectrometry analyses and the need to collect sufficient material to conduct the analyses (e.g. using  
529 a continuous flow mobile centrifuge) were outlined as potential difficulties associated with the use of  
530  $^{137}\text{Cs}$  (Mukundan et al., 2010). The usual analysis time is 24 h per sample, although when low  
531 quantities of material are available or for very low activity samples, counting times of 48 h per  
532 sample are usual, which significantly decreases the analytical capacities and increases the unit cost  
533 per analysis.

534 Based on the data compiled in the current research,  $^{137}\text{Cs}$  should theoretically provide powerful  
535 discrimination between surface and subsurface sources of sediment in a large number of countries  
536 across the world where this technique has not been tested yet (Fig. 8). In Europe,  $^{137}\text{Cs}$  has been  
537 widely used for calculating sediment budgets based on the calculation of  $^{137}\text{Cs}$  inventories in soil  
538 profiles (Mabit et al., 2013; Parsons and Foster, 2011) although it has been less often included in  
539 sediment tracing research in Mediterranean countries as well as in the eastern and northern parts of  
540 the continent. In Northern Africa, there should be significant potential for application in other  
541 Maghreb countries as this technique was successfully implemented in Tunisia (Ben Slimane et al.,  
542 2016). In South America, the technique could be usefully tested in countries with a large agricultural  
543 surface area and located  $> 20^\circ\text{S}$  such as Argentina, Paraguay and Uruguay. Finally, in Oceania,  $^{137}\text{Cs}$   
544 could be tested as a fingerprint in New Zealand, in regions located in western and central Australia,  
545 as well in other Pacific Islands (including French Polynesia). The applicability of the technique in  
546 regions exposed to a colder climate could also be explored to investigate the impact of glacier retreat  
547 on sediment source dynamics among other potential research topics. For example, the feasibility of  
548  $^{137}\text{Cs}$  detection has been recently demonstrated in Maritime Antarctica (Navas et al., 2017).

549 Although there has been a tendency in the literature to argue in favour of the development of  
550 universal techniques and models that could be applied uniformly in catchments around the world,  
551 the current research demonstrates that this goal will likely never be achieved with  $^{137}\text{Cs}$  owing to the  
552 multiple environmental conditions, land use changes and the extent and variations of the  
553 anthropogenic impacts observed in different catchments worldwide. Although universal principles  
554 should be respected when implementing the sediment source tracing methods (i.e, comparing  
555 similar particle size fractions of soils and sediment), there is no universal rule regarding the types of  
556 the sources that can be discriminated based on the  $^{137}\text{Cs}$  method, whether this technique will work in  
557 all the catchments or achieve the source differentiation that was initially sought. There will always be  
558 a significant number of site-specific constraints that the researchers will have to identify and deal  
559 with when deriving their sample design and when interpreting the data, which may lead to

560 unexpected results. For example, owing to its independence of lithology,  $^{137}\text{Cs}$  was considered a  
561 useful property for distinguishing potential source types in catchments with heterogeneous geology  
562 (Walling and Woodward, 1992). This characteristic was thought to be universal. However, the  
563 diversity of applications and source type discriminations illustrated in the current research  
564 demonstrate that the universality of one method will never reasonably be achieved owing to the  
565 diversity of environmental contexts, anthropogenic activities and initial fallout levels found around  
566 the world.

567

## 568 **9. Conclusions**

569 The current meta-analysis of research incorporating  $^{137}\text{Cs}$  in sediment source fingerprinting research  
570 included 123 articles. Fifty-two percent of these articles provides results obtained in catchments  
571 located in Europe or in Northern America. The three countries in the world with the highest numbers  
572 of publications are the UK (n=24), Australia (n=23) and the US (n=20). In contrast, very few articles  
573 reported research from Africa (n=9) and South America (n=2). The research design of these studies  
574 was strongly variable, regarding the number and the size of the catchments investigated, or the  
575 amount and the type of source and sediment samples analysed. The increasing number of studies  
576 observed throughout time demonstrates the growing popularity of this technique. The atmospheric  
577 thermonuclear bomb testing that reached a peak between 1963 (Northern Hemisphere) and 1965  
578 (Southern Hemisphere) provided the main source of  $^{137}\text{Cs}$  fallout except in those few regions of the  
579 world where nuclear accidents occurred (Chernobyl, Fukushima). However, this accidental fallout  
580 does not prevent the use of  $^{137}\text{Cs}$  for discriminating surface vs. subsurface source contributions to  
581 sediment in these regions. Overall, positive relationships were found between the  $^{137}\text{Cs}$  activities  
582 found in surface/subsurface sources and the proportion of global fallout received. Given the low  
583 proportions of global fallout received in those latitudinal regions located between 0-20°N and 0-20°S,  
584 the applicability of the method may be increasingly limited in this part of the world. Furthermore, the  
585 very low  $^{137}\text{Cs}$  activities found in surface soils analysed in China's Loess Plateau and in South Africa  
586 may prevent the use of this sediment fingerprinting technique in these regions and reflect the  
587 severity of soil erosion processes occurring in these environments.

588 Forty-one percent of the reviewed studies included  $^{137}\text{Cs}$  for discriminating surface vs. subsurface  
589 erosion, while 40% of the articles investigated land use contributions to sediment. The power of  $^{137}\text{Cs}$   
590 to distinguish between surface vs. subsurface sources was demonstrated by the fact that among  
591 those studies that use un-mixing models, 94% included  $^{137}\text{Cs}$  in the optimal fingerprints included in  
592 the models. For land-use based studies, 71% of those that modelled sediment source contributions  
593 included  $^{137}\text{Cs}$  in the optimal tracer combination. The usefulness of this tracing property was also  
594 demonstrated in catchments exposed to wildfires, urban environments and tile-drains.

595 Among the main challenges associated with this method, the ongoing decay of  $^{137}\text{Cs}$  to undetectable  
596 levels raises the need to develop the use of detectors installed in underground laboratories and that  
597 of alternative tracers that may include  $^{210}\text{Pb}_{\text{xs}}$ , plutonium or organic matter properties. In the future,  
598 researchers need to systematically include basic catchment information in their articles as well as  
599 reproducible details on the sampling design including the number and the type of the source and  
600 sediment samples collected, the sampling depth, the sampling/reference dates and the particle size  
601 fraction considered. Furthermore, researchers should provide access to the raw data in order to  
602 facilitate the incorporation of previous research results. Despite previous suggestions regarding the  
603 development of universal techniques and models that could be applied uniformly in catchments  
604 across the globe, the current meta-analysis demonstrates that this goal will likely never be achieved  
605 as a result of the diversity of the environmental conditions and fallout levels found in catchments  
606 around the world.

607

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614

## 615 **Supplementary Material**

616 SM1. Characteristics of studies using  $^{137}\text{Cs}$  as a potential fingerprint to quantify sediment source  
617 contributions (literature review).

618 SM2. Tables synthesizing the  $^{137}\text{Cs}$  activity values found in surface versus subsurface sources of  
619 sediment (literature review).

620

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Table 1

Table 1. Studies including  $^{137}\text{Cs}$  to trace surface vs. subsurface source contributions to sediment around the world.

Reference	Country	Study site(s)	Surface area (km <sup>2</sup> )	Subsurface sources	Surface sources	Tracing properties considered	Environmental radionuclides considered	Use of an un-mixing model	Inclusion of $^{137}\text{Cs}$ in the model	Use of a model considering $^{137}\text{Cs}$ alone
Peart and Walling (1988)	UK	2	9.8; 46	Channel bank	Surface soil	ERNs, Elemental Geochemistry, C+N, total P	$^{137}\text{Cs}$	Yes	Yes	Yes
Murray et al. (1993)	Australia	1	n/a	Subsoil	Surface	ERNs	$^{137}\text{Cs}$ , $^7\text{Be}$ , $^{226}\text{Ra}$ , $^{232}\text{Th}$	No	No	No
Olley et al. (1993)	Australia	1	0.5	Gully, subsoil	Topsoil	ERNs	$^{137}\text{Cs}$ , $^7\text{Be}$ , $^{226}\text{Ra}$ , $^{232}\text{Th}$	No	No	No

Ormerod (1998)	Australia	1	10	Subsurface, urban gutters and drains, construction sites	Forest	ERNs	<sup>137</sup> Cs	Yes	Yes	Yes
Wallbrink et al. (1999)	Australia	1	0.01	Rills, gullies	Surface		<sup>137</sup> Cs, <sup>210</sup> Pb, <sup>7</sup> Be	No	No	No
Matisoff et al. (2002)	USA	1	69.5	Channel	Till, non-tilled, undisturbed	ERNs	<sup>137</sup> Cs, <sup>210</sup> Pb, <sup>7</sup> Be	No	No	No
Froehlich (2004)	India	1	0.2	Channel bank, gully bank, path material	Grassland, forest	ERNs	<sup>137</sup> Cs	No	No	No
Nagle and Ritchie (2004)	USA	1	492	Bank material	Cultivated soil	ERNs, C, N	<sup>137</sup> Cs	Yes	Yes	No

Whiting et al. (2005)	USA	1	96260	Channel	Surface	ERNs	<b><sup>137</sup>Cs, <sup>210</sup>Pb, <sup>7</sup>Be</b>	No	No	No
Froehlich and Walling (2006)	India	1	12540	Channel, gully, landslide, earthflow	Cultivated, tea garden, jungle	ERNs	<b><sup>137</sup>Cs, <sup>210</sup>Pb, <sup>226</sup>Ra</b>	No	No	No
Foster et al. (2007)	South Africa	2	1.5; 6.3	Non-topsoil	Topsoil	ERNs, Elemental Geochemistry, magnetic properties	<b><sup>210</sup>Pb, <sup>234</sup>Th, <sup>235</sup>U, <sup>7</sup>Be, <sup>137</sup>Cs, <sup>228</sup>Ac, 40K</b>	No	No	No
Nagle et al. (2007)	USA	1	2041	Channel bank	Cultivated, pasture, forest	ERNs	<b><sup>137</sup>Cs</b>	Yes	Yes	Yes
Mizugaki et al. (2008)	Japan	2	0.3, 0.6	Landslide scar, truck trail	Forest floor	ERNs	<b><sup>137</sup>Cs, <sup>210</sup>Pb</b>	Yes	Yes	No
Smith (2008)	Australia	1	1.6	Gully and channel walls	Slopes	ERNs	<b><sup>137</sup>Cs, <sup>210</sup>Pb</b>	Yes	Yes	Yes

Zehetner et al. (2008)	Taiwan	1	303	Not sampled	Sampled	ERNs	<sup>137</sup> Cs	No	No	No
Ahn et al. (2009)	Japan	1	1.4	Channel bank	Cultivated land, swamp	ERNs	<sup>137</sup> Cs	No	No	No
Ritchie et al. (2009)	USA	1	150 (7 sub-catchments)	Rills, gullies, channel bank	Shrubland, grassland	ERNs	<sup>137</sup> Cs	Yes	Yes	Yes
Wilkinson et al. (2009)	Australia	1	183	Gully walls	Burnt vs. unburnt surface	ERNs, total P, Elemental Geochemistry, <sup>238</sup> Pu/ <sup>239</sup> + <sup>240</sup> Pu	<sup>137</sup> Cs, <sup>210</sup> Pb	Yes	Yes	No
Evrard et al. (2010)	Mexico	3	3, 9, 12	Gullies, retreat scarp	Cropland, grassland, woodland	ERNs	<sup>7</sup> Be, <sup>210</sup> Pb, <sup>137</sup> Cs	No	No	No
Fukuyama et al. (2010)	Japan	1	0.05	Channel banks, gully walls	Forest plantation	ERNs	<sup>137</sup> Cs, <sup>210</sup> Pb	Yes	Yes	Yes

Smith et al. (2011)	Australia	2	1.36, 2.44			ERNs	<b>137Cs, 210Pb</b>	Yes	Yes	No
Caitcheon et al. (2012)	Australia	3	53000, 72000, 109000	<b>Channel banks, gullies</b>	<b>Alluvium on hillslopes</b>	ERNs	<b>137Cs, 210Pb</b>	Yes	Yes	Yes
Manjoro et al. (2012)	South Africa	1	n/a	<b>Gully, badlands</b>	<b>Original soil</b>	<b>ERNs, Elemental Geochemistry, magnetic properties</b>	<b>137Cs</b>	Yes	No	No
Owens et al. (2012)	Canada	2	158, 215	<b>Channel banks</b>	<b>Active slopes</b>	ERNs	<b>137Cs, 210Pb</b>	Yes	Yes	No
Smith et al. (2012)	Australia	2	0.233; 0.073	<b>Channel banks</b>	<b>Hillslope surface deposits</b>	ERNs	<b>137Cs, 210Pb, 239+240Pu</b>	Yes	Yes	Yes

Ben Slimane et al. (2013)	Tunisia	1	2.63	<b>Gully, channel bank</b>	<b>Topsoil</b>	<b>ERNs, C+N, total P</b>	<b>210Pb, 234Th, 226Ra, 228Ra, 228Th, 40K, 137Cs</b>	<b>Yes</b>	<b>Yes</b>	<b>No</b>
Olley et al. (2013a)	Australia	1	24350	<b>Channel banks, gullies, in-channel benches</b>	<b>Surface wash deposits, surface runoff traps</b>	<b>ERNs</b>	<b>137Cs, 210Pb</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>
Olley et al. (2013b)	Australia	7	188, 47, 247, 3842, 483, 911, 2942	<b>Channel bank</b>	<b>Cropping, grazing, forest including natural bush and managed forest</b>	<b>ERNs</b>	<b>137Cs, 210Pb</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>
Theuring et al. (2013)	Mongolia	1	15000	<b>Channel banks, gullies</b>	<b>Surface</b>	<b>ERNs</b>	<b>137Cs, 210Pb, 7Be</b>	<b>Yes</b>	<b>Yes</b>	<b>No</b>

Belmont et al. (2014)	USA	2	n/a	<b>Bluffs, channel banks</b>	<b>Upland</b>	<b>ERNs</b>	<b>7Be, 210Pb, 137Cs, 10Be</b>	<b>Yes</b>	<b>Yes (in some)</b>	<b>No</b>
Hobgen et al. (2014)	Indonesia	1	1227	<b>Subsurface soil</b>	<b>Surface soil</b>	<b>ERNs</b>	<b>137Cs, 210Pb, 239Pu</b>	<b>No</b>	<b>No</b>	<b>No</b>
Lim et al. (2014)	South Korea	1	3470	<b>Channel bank</b>	<b>Forest floor</b>	<b>ERNs</b>	<b>137Cs</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>
Foucher et al. (2015)	France	1	24	<b>Channel bank</b>	<b>Surface soil, hillslope sediment</b>	<b>ERNs</b>	<b>137Cs, 210Pb, 232Th</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>
Haiyan (2015)	China	1	9.9	<b>Gully, channel bank</b>	<b>Cultivated, uncultivated</b>	<b>ERNs, TOC</b>	<b>137Cs, 210Pb</b>	<b>Yes</b>	<b>Yes</b>	<b>No</b>
Ochiai et al. (2015)	Japan	1	6.5	<b>Inferred but not sampled</b>	<b>Surface soil</b>	<b>ERNs</b>	<b>137Cs, 210Pb, 134Cs</b>	<b>No</b>	<b>No</b>	<b>No</b>

Pulley et al. (2015a)	UK	1	1060	Channel banks	Surface agricultural land	ERNs, Elemental Geochemistry, magnetic properties	137Cs, 210Pb, 226Ra, 234Th, 235U, 214Pb, 228Ac, 212Pb, 40K	Yes	Yes	No
Pulley et al. (2015b)	UK	1	1634	Channel banks	Surface agricultural land	ERNs, Elemental Geochemistry, magnetic properties	226Ra, 234Th, 235U, 214Pb, 228Ac, 212Pb, 40K	Yes	Yes (in some catchments)	No
van der Waal et al. (2015)	South Africa	1	65	Gully, landslide	Sheet erosion areas	ERNs, magnetic properties	137Cs, 210Pb	Yes	No (too low 137Cs activities)	No
Wethered et al. (2015)	Australia	1	1000	Channel bed, deep gully	Agricultural land, forest land	ERNs	137Cs, 210Pb	Yes	Yes	No

Wilkinson et al. (2015)	Australia	1	129000	Gullies, channel banks, in-channel benches, subsoil scalds	Surface soil on hillslopes	ERNs	<b>137Cs, 210Pb</b>	Yes	Yes	No
Ben Slimane et al. (2016)	Tunisia	5	3.7, 0.6, 2.4, 2.6, 3.6	Gully, channel	Rill, interrill	ERNs, TOC	<b>137Cs</b>	Yes	Yes	Yes
Estrany et al. (2016)	Spain	2	9.8, 319	Stream channel	Surface soils	ERNs	<b>137Cs, 210Pb</b>	Yes	Yes	No
Evrard et al. (2016)	Laos	1	11.6	Resuspended or recently eroded subsurface	Resuspended or recently eroded surface	ERNs	<b>7Be, 137Cs, 210Pb</b>	Yes	Yes	Yes
Le Gall et al. (2016)	France	1	24	Channel banks	Cropland topsoil	ERNs, Elemental Geochemistry, Sr isotopes ( <b>87Sr/86Sr</b> )	<b>137Cs</b>	Yes	Yes	Yes

Zhang et al. (2016)	USA	1	15.6	<b>Gully</b>	<b>Overland</b>	<b>ERNs</b>	<b>137Cs</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>
Huon et al. (2017)	Laos	1	22	<b>Channel banks, gullies</b>	<b>Topsoil</b>	<b>ERNs, C+N, d13C, d15N</b>	<b>7Be/210Pb/137Cs</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>
García-Comendador et al. (2017)	Spain	1	4.8	<b>Burned vs. unburned channel bank</b>	<b>Burned vs. unburned surface material</b>	<b>ERNs</b>	<b>137Cs, 210Pb, 226Ra</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>
Jalowska et al. (2017)	USA	1	25123				<b>137Cs, 210Pb</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>
Li et al. (2017)	China	1	5.97	<b>Gully</b>	<b>Inter-gully (!)</b>	<b>ERNs</b>	<b>137Cs</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>

Reference	Country	Study site(s)	Surface area (km <sup>2</sup> )	Subsurface sources	Surface sources	Tracing properties considered	Environmental radionuclides considered	Use of an un-mixing model	Inclusion of <sup>137</sup> Cs in the model	Use of a model considering <sup>137</sup> Cs alone
Peart and Walling (1988)	UK	2	9.8; 46	Channel bank	Surface soil	ERNs, Elemental Geochemistry, C+N, total P	<sup>137</sup> Cs	Yes	Yes	Yes
Murray et al. (1993)	Australia	1	n/a	Subsoil	Surface	ERNs	<sup>137</sup> Cs, <sup>7</sup> Be, <sup>226</sup> Ra, <sup>232</sup> Th	No	No	No
Olley et al. (1993)	Australia	1	0.5	Gully, subsoil	Topsoil	ERNs	<sup>137</sup> Cs, <sup>7</sup> Be, <sup>226</sup> Ra, <sup>232</sup> Th	No	No	No

Ormerod (1998)	Australia	1	10	Subsurface, urban gutters and drains, construction sites	Forest	ERNs	<sup>137</sup> Cs	Yes	Yes	Yes
Wallbrink et al. (1999)	Australia	1	0.01	Rills, gullies	Surface		<sup>137</sup> Cs, <sup>210</sup> Pb, <sup>7</sup> Be	No	No	No
Matisoff et al. (2002)	USA	1	69.5	Channel	Till, non-tilled, undisturbed	ERNs	<sup>137</sup> Cs, <sup>210</sup> Pb, <sup>7</sup> Be	No	No	No
Froehlich (2004)	India	1	0.2	Channel bank, gully bank, path material	Grassland, forest	ERNs	<sup>137</sup> Cs	No	No	No
Nagle and Ritchie (2004)	USA	1	492	Bank material	Cultivated soil	ERNs, C, N	<sup>137</sup> Cs	Yes	Yes	No

Whiting et al. (2005)	USA	1	96260	Channel	Surface	ERNs	<b><sup>137</sup>Cs, <sup>210</sup>Pb, <sup>7</sup>Be</b>	No	No	No
Froehlich and Walling (2006)	India	1	12540	Channel, gully, landslide, earthflow	Cultivated, tea garden, jungle	ERNs	<b><sup>137</sup>Cs, <sup>210</sup>Pb, <sup>226</sup>Ra</b>	No	No	No
Foster et al. (2007)	South Africa	2	1.5; 6.3	Non-topsoil	Topsoil	ERNs, Elemental Geochemistry, magnetic properties	<b><sup>210</sup>Pb, <sup>234</sup>Th, <sup>235</sup>U, <sup>7</sup>Be, <sup>137</sup>Cs, <sup>228</sup>Ac, 40K</b>	No	No	No
Nagle et al. (2007)	USA	1	2041	Channel bank	Cultivated, pasture, forest	ERNs	<b><sup>137</sup>Cs</b>	Yes	Yes	Yes
Mizugaki et al. (2008)	Japan	2	0.3, 0.6	Landslide scar, truck trail	Forest floor	ERNs	<b><sup>137</sup>Cs, <sup>210</sup>Pb</b>	Yes	Yes	No
Smith (2008)	Australia	1	1.6	Gully and channel walls	Slopes	ERNs	<b><sup>137</sup>Cs, <sup>210</sup>Pb</b>	Yes	Yes	Yes

Zehetner et al. (2008)	Taiwan	1	303	Not sampled	Sampled	ERNs	<sup>137</sup> Cs	No	No	No
Ahn et al. (2009)	Japan	1	1.4	Channel bank	Cultivated land, swamp	ERNs	<sup>137</sup> Cs	No	No	No
Ritchie et al. (2009)	USA	1	150 (7 sub-catchments)	Rills, gullies, channel bank	Shrubland, grassland	ERNs	<sup>137</sup> Cs	Yes	Yes	Yes
Wilkinson et al. (2009)	Australia	1	183	Gully walls	Burnt vs. unburnt surface	ERNs, total P, Elemental Geochemistry, <sup>238</sup> Pu/ <sup>239</sup> + <sup>240</sup> Pu	<sup>137</sup> Cs, <sup>210</sup> Pb	Yes	Yes	No
Evrard et al. (2010)	Mexico	3	3, 9, 12	Gullies, retreat scarp	Cropland, grassland, woodland	ERNs	<sup>7</sup> Be, <sup>210</sup> Pb, <sup>137</sup> Cs	No	No	No
Fukuyama et al. (2010)	Japan	1	0.05	Channel banks, gully walls	Forest plantation	ERNs	<sup>137</sup> Cs, <sup>210</sup> Pb	Yes	Yes	Yes

Smith et al. (2011)	Australia	2	1.36, 2.44			ERNs	<b>137Cs, 210Pb</b>	Yes	Yes	No
Caitcheon et al. (2012)	Australia	3	53000, 72000, 109000	<b>Channel banks, gullies</b>	<b>Alluvium on hillslopes</b>	ERNs	<b>137Cs, 210Pb</b>	Yes	Yes	Yes
Manjoro et al. (2012)	South Africa	1	n/a	<b>Gully, badlands</b>	<b>Original soil</b>	<b>ERNs, Elemental Geochemistry, magnetic properties</b>	<b>137Cs</b>	Yes	No	No
Owens et al. (2012)	Canada	2	158, 215	<b>Channel banks</b>	<b>Active slopes</b>	ERNs	<b>137Cs, 210Pb</b>	Yes	Yes	No
Smith et al. (2012)	Australia	2	0.233; 0.073	<b>Channel banks</b>	<b>Hillslope surface deposits</b>	ERNs	<b>137Cs, 210Pb, 239+240Pu</b>	Yes	Yes	Yes

Ben Slimane et al. (2013)	Tunisia	1	2.63	<b>Gully, channel bank</b>	<b>Topsoil</b>	<b>ERNs, C+N, total P</b>	<b>210Pb, 234Th, 226Ra, 228Ra, 228Th, 40K, 137Cs</b>	<b>Yes</b>	<b>Yes</b>	<b>No</b>
Olley et al. (2013a)	Australia	1	24350	<b>Channel banks, gullies, in-channel benches</b>	<b>Surface wash deposits, surface runoff traps</b>	<b>ERNs</b>	<b>137Cs, 210Pb</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>
Olley et al. (2013b)	Australia	7	188, 47, 247, 3842, 483, 911, 2942	<b>Channel bank</b>	<b>Cropping, grazing, forest including natural bush and managed forest</b>	<b>ERNs</b>	<b>137Cs, 210Pb</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>
Theuring et al. (2013)	Mongolia	1	15000	<b>Channel banks, gullies</b>	<b>Surface</b>	<b>ERNs</b>	<b>137Cs, 210Pb, 7Be</b>	<b>Yes</b>	<b>Yes</b>	<b>No</b>

Belmont et al. (2014)	USA	2	n/a	<b>Bluffs, channel banks</b>	<b>Upland</b>	<b>ERNs</b>	<b>7Be, 210Pb, 137Cs, 10Be</b>	<b>Yes</b>	<b>Yes (in some)</b>	<b>No</b>
Hobgen et al. (2014)	Indonesia	1	1227	<b>Subsurface soil</b>	<b>Surface soil</b>	<b>ERNs</b>	<b>137Cs, 210Pb, 239Pu</b>	<b>No</b>	<b>No</b>	<b>No</b>
Lim et al. (2014)	South Korea	1	3470	<b>Channel bank</b>	<b>Forest floor</b>	<b>ERNs</b>	<b>137Cs</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>
Foucher et al. (2015)	France	1	24	<b>Channel bank</b>	<b>Surface soil, hillslope sediment</b>	<b>ERNs</b>	<b>137Cs, 210Pb, 232Th</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>
Haiyan (2015)	China	1	9.9	<b>Gully, channel bank</b>	<b>Cultivated, uncultivated</b>	<b>ERNs, TOC</b>	<b>137Cs, 210Pb</b>	<b>Yes</b>	<b>Yes</b>	<b>No</b>
Ochiai et al. (2015)	Japan	1	6.5	<b>Inferred but not sampled</b>	<b>Surface soil</b>	<b>ERNs</b>	<b>137Cs, 210Pb, 134Cs</b>	<b>No</b>	<b>No</b>	<b>No</b>

Pulley et al. (2015a)	UK	1	1060	Channel banks	Surface agricultural land	ERNs, Elemental Geochemistry, magnetic properties	137Cs, 210Pb, 226Ra, 234Th, 235U, 214Pb, 228Ac, 212Pb, 40K	Yes	Yes	No
Pulley et al. (2015b)	UK	1	1634	Channel banks	Surface agricultural land	ERNs, Elemental Geochemistry, magnetic properties	226Ra, 234Th, 235U, 214Pb, 228Ac, 212Pb, 40K	Yes	Yes (in some catchments)	No
van der Waal et al. (2015)	South Africa	1	65	Gully, landslide	Sheet erosion areas	ERNs, magnetic properties	137Cs, 210Pb	Yes	No (too low 137Cs activities)	No
Wethered et al. (2015)	Australia	1	1000	Channel bed, deep gully	Agricultural land, forest land	ERNs	137Cs, 210Pb	Yes	Yes	No

Wilkinson et al. (2015)	Australia	1	129000	Gullies, channel banks, in-channel benches, subsoil scalds	Surface soil on hillslopes	ERNs	<b>137Cs, 210Pb</b>	Yes	Yes	No
Ben Slimane et al. (2016)	Tunisia	5	3.7, 0.6, 2.4, 2.6, 3.6	Gully, channel	Rill, interrill	ERNs, TOC	<b>137Cs</b>	Yes	Yes	Yes
Estrany et al. (2016)	Spain	2	9.8, 319	Stream channel	Surface soils	ERNs	<b>137Cs, 210Pb</b>	Yes	Yes	No
Evrard et al. (2016)	Laos	1	11.6	Resuspended or recently eroded subsurface	Resuspended or recently eroded surface	ERNs	<b>7Be, 137Cs, 210Pb</b>	Yes	Yes	Yes
Le Gall et al. (2016)	France	1	24	Channel banks	Cropland topsoil	ERNs, Elemental Geochemistry, Sr isotopes ( <b>87Sr/86Sr</b> )	<b>137Cs</b>	Yes	Yes	Yes

Zhang et al. (2016)	USA	1	15.6	<b>Gully</b>	<b>Overland</b>	<b>ERNs</b>	<b>137Cs</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>
Huon et al. (2017)	Laos	1	22	<b>Channel banks, gullies</b>	<b>Topsoil</b>	<b>ERNs, C+N, d13C, d15N</b>	<b>7Be/210Pb/137Cs</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>
García-Comendador et al. (2017)	Spain	1	4.8	<b>Burned vs. unburned channel bank</b>	<b>Burned vs. unburned surface material</b>	<b>ERNs</b>	<b>137Cs, 210Pb, 226Ra</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>
Jalowska et al. (2017)	USA	1	25123				<b>137Cs, 210Pb</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>
Li et al. (2017)	China	1	5.97	<b>Gully</b>	<b>Inter-gully</b>	<b>ERNs</b>	<b>137Cs</b>	<b>Yes</b>	<b>Yes</b>	<b>Yes</b>

Table 2

Table 2. Studies including  $^{137}\text{Cs}$  to trace land use-based source contributions to sediment around the world.

Reference	Country	Study site(s)	Surface area (km <sup>2</sup> )	Subsurface sources	Surface sources	Tracing properties considered	Environmental radionuclides considered	Use of an un-mixing model	Inclusion of $^{137}\text{Cs}$ in the model	Use of a model considering $^{137}\text{Cs}$ alone	Remark
Walling and Woodward (1992)	UK	2	46, N/D	Channel banks	Cultivated, pasture	FRNSs	$^{137}\text{Cs}$ , $^{210}\text{Pb}$ , $^7\text{Be}$	Yes	Yes	No	n/a
Walling et al. (1993)	UK	2	46, N/D	Channel banks	Cultivated, pasture	FRNs, Magnetism, C+N	$^{137}\text{Cs}$ , $^{210}\text{Pb}$ ,	Yes	Yes	No	n/a
Collins et al. (1996)	UK	2	601; 4325	Channel banks	Cultivated, pasture	FRNs, Elemental Geochemistry, C+N, total P, particle size	$^{137}\text{Cs}$	Yes	No	No	n/a

Collins et al. (1997a)	UK	2	580; 87	Channel banks	Cultivated, pasture	FRNs, Elemental Geochemistry, C+N, total P	<sup>137</sup> Cs, <sup>210</sup> Pb	Yes	Yes	No	n/a
Collins et al. (1997b)	UK	2	46; 8.7	Channel banks	Cultivated, pasture, forest	FRNs, Elemental Geochemistry, C+N, total P, particle size	<sup>137</sup> Cs, <sup>210</sup> Pb	Yes	Yes (in 1 of the 2 catchments)	No	n/a
Wallbrink et al. (1998)	Australia	1	13500	Channel banks, gullies	Cultivated, pasture	FRNs	<sup>137</sup> Cs, <sup>210</sup> Pb	Yes	Yes	Yes	n/a

Walling and Amos (1999)	UK	1	63.5	Channel banks and margins (farm tracks)	Cropland, grassland	FRNs	<sup>137</sup> Cs, <sup>210</sup> Pb	No	No	No	
Collins et al. (2001)	Zambia	1	63	Channel banks, gullies	Grazing, communal cultivation, commercial cultivation	FRNs, Elemental Geochemistry, C+N, total P	<sup>137</sup> Cs, <sup>210</sup> Pb, <sup>226</sup> Ra	Yes	Yes	No	Cultivation type in Zambia (commercial, communal, grazing)

Walling et al. (2001)	Zambia	1	63	Channel banks, gullies	Bush grazing, commercial cultivation, communal cultivation	FRNs, Elemental Geochemistry, extractable metals, C+N, total P	<sup>137</sup> Cs, <sup>210</sup> Pb, <sup>226</sup> Ra	Yes	Yes	No	Cultivation types
Collins and Walling (2002)	Zambia; UK	5	63; 128; 8.7; 778; 852	Channel banks, gullies	Cultivated, uncultivated, forest	FRNs, Elemental Geochemistry, C+N, total P	<sup>137</sup> Cs, <sup>210</sup> Pb, <sup>226</sup> Ra	Yes	Yes (in 1 of the 5 catchments)	No	Cultivation type in Zambia (commercial, communal, grazing)
Foster et al. (2002)	UK	1	2.7	Subsoil (arable and grassland), channel bank	Arable topsoil, grassland topsoil, forest topsoil	FRNs, magnetic properties	<sup>137</sup> Cs	No	No	No	Only descriptive

Carter et al. (2003)	UK	1	1932	Channel banks	Cultivated, uncultivated, forest	FRNs, Elemental Geochemistry, C+N, P fractions	137Cs, 210Pb, 226Ra	Yes	Yes	No	Urban (road dust, solids from sewage treatment plants)
Krause et al. (2003)	Australia	6	0.05; 0.10; 0.13; 0.15; 1.19; 1.21	Gully wall; unsealed road	Ungrazed pasture	FRNs, Elemental Geochemistry	137Cs	Yes	Yes	No	
Motha et al. (2003)	Australia	1	65	Gravelled or ungravelled road	Harvest vs. undisturbed forest	FRNs, Elemental Geochemistry, organic content	137Cs, 210Pb	Yes	Yes	No	Forest management
Walling et al. (2003)	UK	2	4133, 4390	Subsoil, channel banks	Woodland, pasture, cropland, moorland (=topsoil)	FRNs, Elemental Geochemistry, C+N, magnetic properties	137Cs, 210Pb, 226Ra,	Yes	Yes	Yes	

Wallbrink (2004)	Australia	2	10000, 2400	Subsoil	Cultivated, uncultivated	FRNs	<sup>137</sup> Cs, <sup>226</sup> Ra	Yes	Yes	Yes	
Walling et al. (2006)	UK	2	166; 234	Channel banks	Cropland, grassland, forest	FRNs, Elemental Geochemistry, C+N, P fractions	<sup>137</sup> Cs, <sup>210</sup> Pb, <sup>226</sup> Ra	Yes	Yes (in 1 catchment)	No	
Martinez-Carreras et al. (2008)	Luxembourg	1	4.4	Channel banks	Forest topsoil, grassland topsoil, cropland topsoil	FRNs, Elemental Geochemistry, C+N, total P	<sup>137</sup> Cs, <sup>210</sup> Pb	Yes	Yes	No	

Pittam et al. (2008)	UK	1	59	Roads, channel banks, subsoil	Pasture, arable, woodland topsoil	FRNs, magnetic properties	<sup>137</sup> Cs, <sup>210</sup> Pb	Yes	No	No	
Walling et al. (2008)	UK	12	16; 21; 55; 42; 81; 109; 77; 93; 109; 55; 89; 69	Channel banks	Cropland, grassland, forest	FRNs, Elemental Geochemistry, C+N, P fractions	<sup>137</sup> Cs, <sup>210</sup> Pb, <sup>226</sup> Ra	Yes	Yes	No	
Juracek and Ziegler (2009)	USA	6	2920; 24; 49; 31; 22; 38	Channel banks	Cropland, grassland	FRNs, Elemental Geochemistry, C+N, total P	<sup>137</sup> Cs	Yes	Yes	No	
Devereux et al. (2010)	USA	1	188	Channel banks	Urban, rural, agriculture (upland, floodplain)	FRNs, Elemental Geochemistry	<sup>137</sup> Cs, <sup>40</sup> K	Yes	No	No	Urban (street residue)

Mukundan et al. (2010)	USA	1	182	Channel bank, construction roads	Forest, pastures, row crops	FRNs, Elemental Geochemistry, C, N, S, d15N	137Cs, 210Pb, 7Be	Yes	Yes	No	
Nazari Samani et al. (2011)	Iran	3	0.265; 0.051; 1.757	Gully wall	Topsoil, rangeland, dryland farming	FRNs, C+N, total P	137Cs	Yes	Yes	No	
Huisman and Karthikeyan (2012)	USA	1	12.4	Channel bank	Upland, stream bed	FRNs	137Cs, 7Be, 210Pb	Yes	Yes	No	
Huisman et al. (2013)	USA	1	12.4	Channel bank	Upland, stream bed	FRNs, total P	137Cs, 7Be, 210Pb	Yes	Yes	No	
Kim et al. (2013)	South Korea	1	0.3	Gully bank, forest road	Forest floor, paddy plain	FRNs	137Cs	Yes	Yes	Yes	

Hancock et al. (2014)	Australia	3	4700, 3700, 9400	Channel bank, subsoil	Pasture, forest	FRNs	238U, 226Ra, 210Pb, 228Ra, 228Th, 137Cs, 7Be, 40K	Yes	Yes	Yes	
Minella et al. (2014)	Brazil	1	1.2	Channel bank, unpaved roads	Cultivated	FRNs, Elemental Geochemistry, TOC, total P	137Cs	Yes	No	No	137Cs not included in tracing
Smith and Blake (2014)	UK	7	920; 238; 67; 97; 38; 219; 124	Channel banks	Pasture, cultivated	FRNs, Elemental Geochemistry, TOC	137Cs, 210Pb	Yes	Yes (in some catchments)	No	

Trajkovska et al. (2014)	USA	3	39, 288, 289	River channel, channel bank	Upland	FRNs	<sup>137</sup> Cs, <sup>210</sup> Pb	No	No	No	Comparison of an urban and a rural catchment
Palazón et al. (2015a)	Spain	1	1509	Subsoil (eroded areas+ badlands)	Forest/cropland/scrubland	FRNs, Elemental Geochemistry, magnetic properties	<sup>238</sup> U, <sup>226</sup> Ra, <sup>232</sup> Th, <sup>40</sup> K, <sup>137</sup> Cs	Yes	Yes	No	
Palazón et al. (2015b)	Spain	1	445	Subsoil (eroded areas+ badlands)	Forest/cropland/scrubland	FRNs, Elemental Geochemistry, magnetic properties	<sup>238</sup> U, <sup>226</sup> Ra, <sup>232</sup> Th, <sup>40</sup> K, <sup>210</sup> Pb, <sup>137</sup> Cs	Yes	Yes	No	

Srisuksawad et al. (2015)	Thailand	3	20.5	Channel bank	Cultivated, uncultivated, pasture	FRNs	<sup>137</sup> Cs	No	No	No	
Liu et al. (2016)	USA	1 (and subcategories)	15.6	Gully	Rangeland, cultivated land	FRNs, Elemental Geochemistry, C+N+H+S	<sup>137</sup> Cs, <sup>238</sup> U,	Yes	No	No	
Palazón et al. (2016)	Spain	1	1479	Subsoil (eroded areas+ badlands)	Forest/cropland/scrubland	FRNs, Elemental Geochemistry, magnetic properties	<sup>238</sup> U, <sup>226</sup> Ra, <sup>232</sup> Th, <sup>40</sup> K, <sup>210</sup> Pb, <sup>137</sup> Cs	Yes	Yes	No	

Zhou et al. (2016)	China	1	30	Channel bank	Cultivated, woodland	FRNs, Elemental Geochemistry	$^{137}\text{Cs}$	Yes	No	No	Mines and roads
Palazón and Navas (2017)	Spain	1	1509	Subsoil (eroded areas+ badlands)	Forest/cropland/scrubland	FRNs, Elemental Geochemistry, magnetic properties	$^{238}\text{U}$ , $^{226}\text{Ra}$ , $^{232}\text{Th}$ , $^{40}\text{K}$ , $^{210}\text{Pb}$ , $^{137}\text{Cs}$	Yes	Yes	No	
Boudreault et al. (2018)	Canada	1	14.5	Forest subsoil, agricultural subsoil	Forest topsoil, agricultural topsoil	FRNs, colour, particle size and shape	$^{137}\text{Cs}$	Yes	Yes	No	
Froger et al. (2018)	France	1	900	Urban, road deposited sediment	Forest, cropland	FRNs	$^{137}\text{Cs}$ , $^{210}\text{Pb}$ , $^7\text{Be}$	No	No	No	Descriptive part

Nosrati et al. (2018)	Iran	1	78	Road cuttings, channel banks	Hillslopes	FRNs, Elemental Geochemistry, TOC, TN	<sup>137</sup> Cs (and geochem)	Yes	Yes	No	
Rode et al. (2018)	Burkina Faso	1	580	Gully, channel bank, road	Agriculture, badlands	FRNs, Elemental Geochemistry	<sup>7</sup> Be/ <sup>210</sup> Pb/ <sup>137</sup> Cs	Yes	Yes	No	
Gaspar et al. (2019)	Spain	2	1.8, 43	Subsoil, channel banks	Agricultural, rangeland	FRNs, Elemental Geochemistry, TOC, magnetic properties, total P	<sup>238</sup> U, <sup>226</sup> Ra, <sup>232</sup> Th, <sup>40</sup> K, <sup>137</sup> Cs	Yes	Yes	No	After an exceptional flood

Huang et al. (2019a)	China	1	27.6	Gully	Cultivated topsoil, uncultivated topsoil	FRNs, Elemental Geochemistry	<sup>137</sup> Cs, <sup>210</sup> Pb	Yes	No	No	
Huang et al. (2019b)	China	1	27.6	Gully	Cultivated topsoil, uncultivated topsoil	FRNs, Elemental Geochemistry	<sup>137</sup> Cs	Yes	No	No	
Lim et al. (2019)	South Korea	1	2365	Channel banks	Cropland topsoils, forest floor topsoils	FRNs, Elemental Geochemistry, magnetic properties	<sup>137</sup> Cs, <sup>226</sup> Ra, <sup>210</sup> Pb	Yes	No	No	

Lizaga et al. (2019)	Spain	1	23	Eroded subsoil, channel bank	Cropland, open forest, pine afforestation	FRNs, Elemental Geochemistry, magnetic properties	<sup>238</sup> U, <sup>226</sup> Ra, <sup>232</sup> Th, <sup>40</sup> K, <sup>137</sup> Cs	Yes	Yes	No	Before/ after a major flood (flood stages)
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Table 3

Table 3. Studies including  $^{137}\text{Cs}$  to trace a combination of surface vs. subsurface and/or land use-based source contributions to sediment, or based on tributary based approaches or investigating the contributions of particular source types (i.e. mining vs. non-mining, contaminated vs. non-contaminated).

Reference	Country	Study site(s)	Surface area (km <sup>2</sup> )	Type of sources considered	Subsurface sources	Surface sources	Tracing properties considered	Environmental radionuclides considered	Use of an unmixing model	Inclusion of $^{137}\text{Cs}$ in the model	Use of a model considering $^{137}\text{Cs}$ alone	Remark
Walling and Woodward (1995)	UK	1	276	Land use + Geology	Channel banks	Cultivated, pasture	FRNs, Magnetism, C+N	$^{137}\text{Cs}$ , $^{210}\text{Pb}$ , $^{226}\text{Ra}$	Yes	Yes	No	Land uses were traced in different geologies; $^{226}\text{Ra}$ indicative of rock type
Collins et al. (1997)	UK	2	601; 4325	Land use + sub-basin	Channel banks	Cultivated, pasture, forest	FRNs, Elemental Geochemistry, C+N, total P	$^{137}\text{Cs}$	Yes	No	No	Tributary approach

Walling et al. (1999)	UK	1	4133	Land use, geology	Channel banks	Uncultivated topsoil, cultivated topsoil, forest	FRNs, Elemental Geochemistry, C+N, total P, magnetic properties	<sup>137</sup> Cs, <sup>210</sup> Pb, <sup>226</sup> Ra	Yes	Yes (for land use not geology)	No	
Russell et al. (2001)	UK	4	1.5; 0.31; 2.3; 0.96	Mix of land use, soil type and geology	Channel banks	Arable, hopyard, pasture	FRNs, Elemental Geochemistry, C+N, free oxide forms of Fe, Mn and Al, magnetic properties	<sup>137</sup> Cs, <sup>210</sup> Pb	Yes	Yes	No	Field drains

Motha et al. (2004)	Australia	1	11.1	Mix of land use and geology	Gravelled or ungravelled road	Cropland, grassland, undisturbed forest	FRNs, Elemental Geochemistry, magnetic properties	137Cs, 210Pb	Yes	Yes	No	Geology: cropland on basalt or granite material
Collins and Walling (2007a)	UK	2	437; 183	Land use + sub-basin	Channel banks	Cultivated, uncultivated, forest	FRNs, Elemental Geochemistry, C+N, total P	137Cs, 210Pb, 226Ra	Yes	Yes (in some catchments)	No	Tributary approach
Collins and Walling (2007b)	UK	3	234; 166; 231	Land use + sub-basin	Channel banks	Cultivated, pasture	FRNs, Elemental Geochemistry, C+N, P fractions	137Cs, 210Pb, 226Ra	Yes	n/a (!!!)	No	Tributary approach

Rhoton et al. (2008)	USA	1	150	Sub-basins (7)			FRNs, TOC+TIC, exchangeable cations, d13C, quantified colour, magnetic properties	137Cs, 40K, 226Ra,	Yes	Yes	No	Tracing the sub-basin contributions at the outlet
Simms et al. (2008)	Australia	1	91.5	Erosion processes	Gully floor, gully collapse	Rills, sheet erosion	FRNs	137Cs, 210Pb	No	No	No	Only descriptive
Hughes et al. (2009)	Australia	1	6000	Geologies and surface vs. subsurface	Gully sidewalls and headcuts, exposed river banks	Cultivated land, uncultivated pasture	FRNs, Geochemical Elements	137Cs, 210Pb, 226Ra	Yes	Yes (for erosion type)	No	

Ritchie et al. (2009)	USA	1	150	Sub-catchment/land use	Not informed	Shrub-dominated, grass-dominated	FRNs, TOC, d13C	137Cs	Yes	Yes	No	
Evrard et al. (2011)	France	1	907	Geologies	n/a	n/a	FRNs, Elemental Geochemistry	241Am, 7Be, 137Cs, 210Pb, 40K, 226Ra, 228Th, 234Th	Yes	No	No	Description of FRNs
Navratil et al. (2012)	France	1	907	Geologies	n/a	n/a	FRNs, Elemental Geochemistry	7Be, 137Cs, 210Pb, 40K, 226Ra, 228Ra, 234Th, 228Th	Yes	No	Yes	Description of FRNs

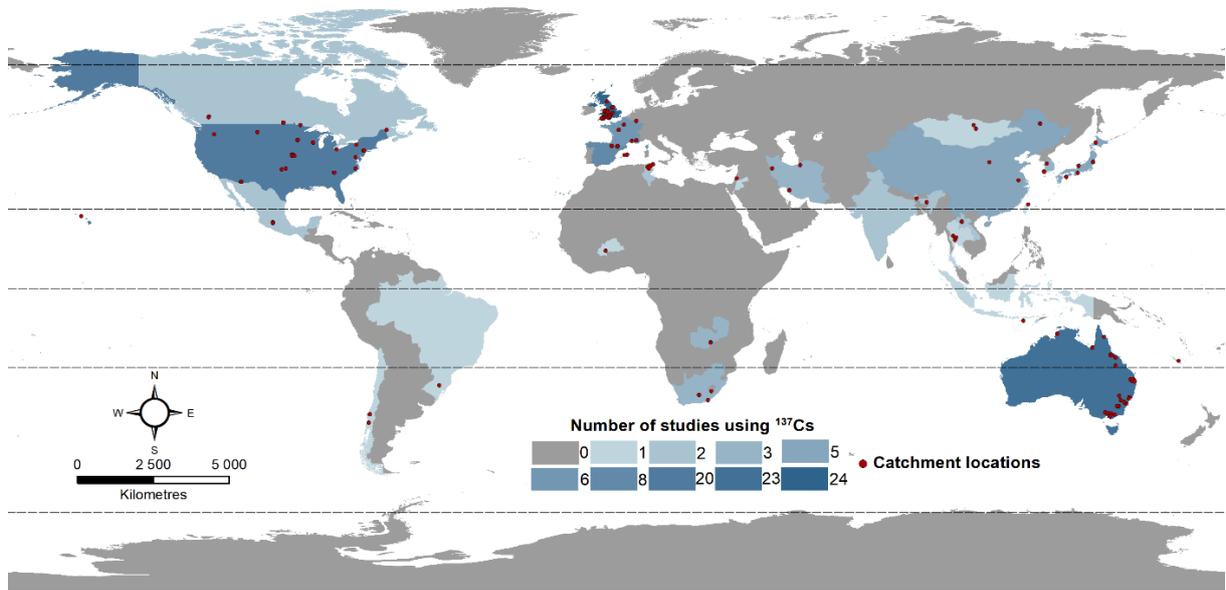
Evrard et al. (2013)	Mexico	4	12; 3; 9; 630	Land use + soil types	Gullies, forest roads;	Cropland	FRNs, C+N, spectroscopy (DRIFTS)	241Am, 7Be, 137Cs, 210Pb, 40K, 226Ra, 228Ra, 228Th, 234Th,	Yes	Yes (in 2 catchments)	No	
Koiter et al. (2013)	Canada	1	74.41	Mix of land use and geology	Channel banks; shale bedrock material	Topsoil from cropland and riparian zones	FRNs, Elemental Geochemistry	137Cs,	Yes	Yes	No	
Schuller et al. (2013)	Chile	4	0.198 ; 0.076 ; 0.127 ; 0.077	Pre-harvest, post-harvest, post-replanting, control			FRNs	137Cs, 210Pb, 40K, 226Ra	Yes	Yes	No	Forest management

Stout et al. (2013)	USA	1	4300	Tributary approach	Unclear	Unclear	FRNs	7Be, 210Pb, 137Cs, 10Be	No	No	No	Floodplain exchange
Wilkinson et al. (2013)	Australia	4	14, 1200; 9400, 130000	Mix of surface vs. subsurface, geologies, river confluences	Gully wall, gully floor deposits, rilled hillslope	Hillslope soil	FRNs, Elemental Geochemistry	137Cs, 210Pb	Yes	Yes (for surface vs. subsurface)	Yes (in some catchments)	
Kraushaar et al. (2015)	Jordan	1	263.5	Mix of land use and geology	Gullies; C-horizons	Cultivated with olives; cultivated with wheat and vegetables; grazed	FRNs, Elemental Geochemistry, C (TOC/TIC)	137Cs, 40K, 228Th, 232Th	Yes	No	No	Cultivation type

Zebracki et al. (2015)	France	1	96500	Sub-basin (geology/location)			FRNs	40K, 232Th, 238U, 137Cs, 239+240Pu	Yes	No	No	Upstream/Cevenol/Pre-Alpine
Du and Walling (2017)	UK	1	0.07	Eroding vs. deposition locations			FRNs, C+N, P fractions, Elemental Geochemistry	137Cs,	Yes	Yes	No	Field scale
Nosrati (2017)	Iran	1	4.47	Mix of land use and hillslope components	n/a	Forest/crop land (summit, shoulder, backslope, toeslope)	FRNs, C	40K, 238U, 226Ra, 232Th, 137Cs,	Yes	Yes	No	Hillslope components
Evrard et al. (2019)	Japan	2	77, 275	Contaminated vs. uncontaminated	Subsoil	Topsoil exposed to fallout	FRNs	137Cs, 134Cs	Yes	Yes	Yes	Following Fukushima (tributary approach)

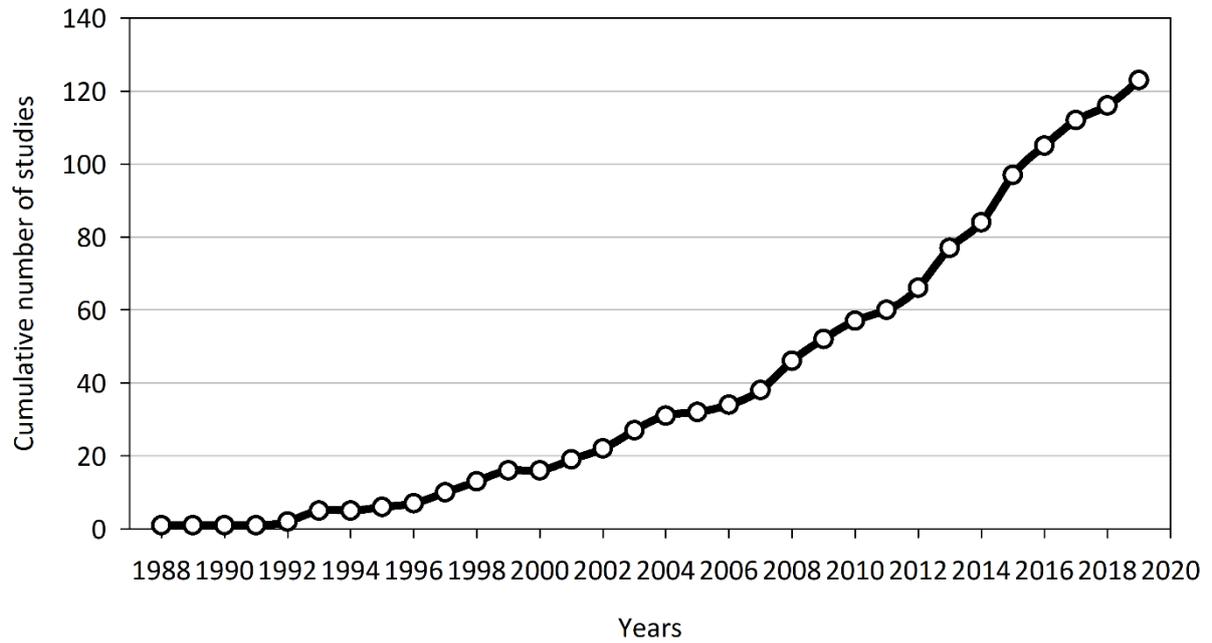
Sellier et al. (2019)	New Caledonia	1	397	<b>Mining vs. non-mining</b>	<b>Tributaries draining mines</b>	<b>Tributaries devoid of mines</b>	<b>FRNs, Elemental Geochemistry</b>	<b>137Cs, 210Pb, 228Th, 238U, 40K</b>	<b>Yes</b>	<b>No</b>	<b>No</b>	<b>Mining</b>
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1 Figure 1. Location of catchments and number of studies where  $^{137}\text{Cs}$  was used to discriminate  
2 sediment source contributions across the world.



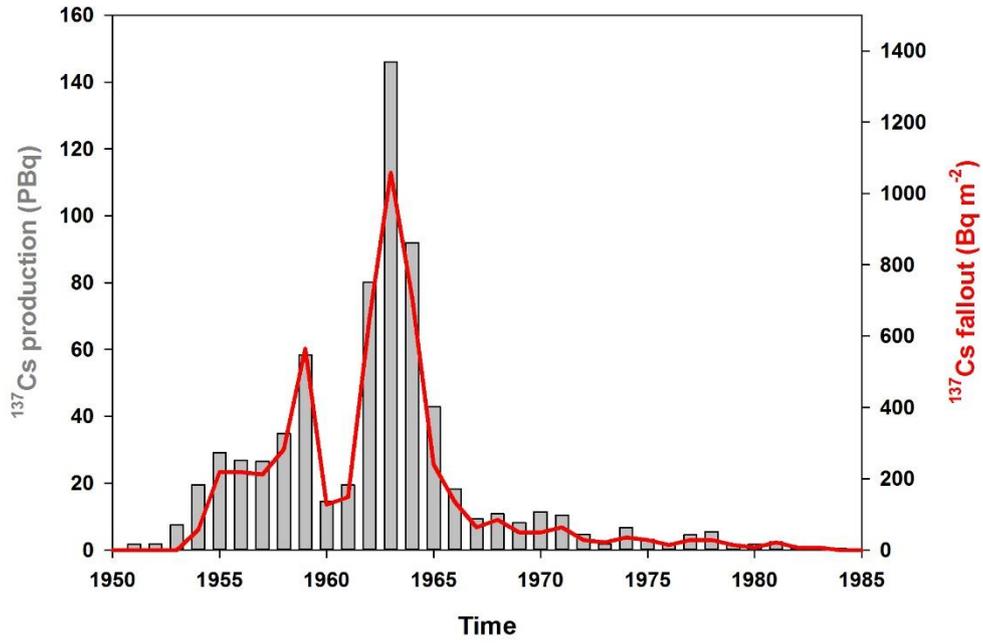
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23 Figure 2. Evolution of the cumulative number of studies including  $^{137}\text{Cs}$  measurements for quantifying  
24 sediment source contributions between 1988 and 2019.



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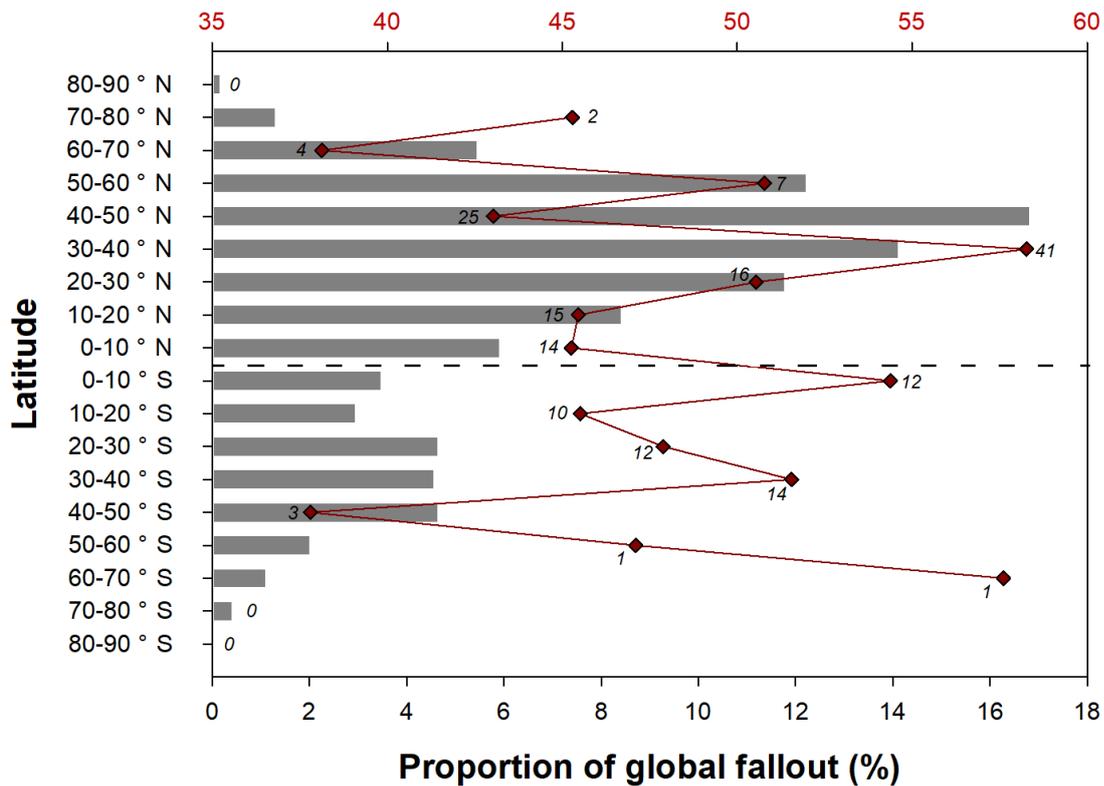
44 Figure 3. Temporal deposition of  $^{137}\text{Cs}$  across the globe after UNSCEAR data (UNSCEAR, 2000).



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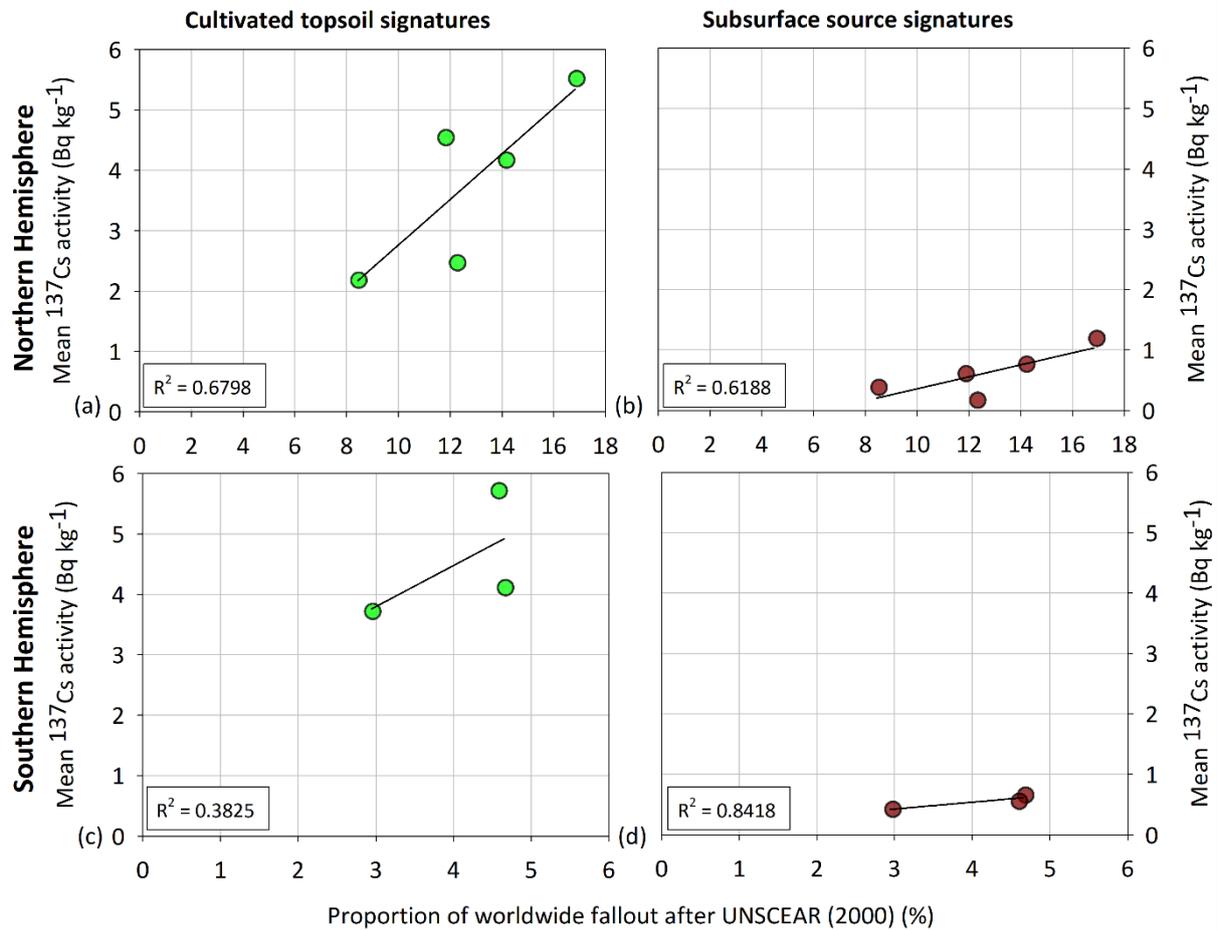
64 Figure 4. Distribution of radionuclide fallout from thermonuclear bomb testing with latitude in the  
 65 world, after UNSCEAR (2000), and the proportion of monthly bomb fallout data missing between  
 66 1954-1976 for each 10-degree latitude based on records from those available monitoring stations  
 67 (Health and Safety Laboratory, 1977). Overall, 50% of data were missing for this period, as most of  
 68 the monitoring stations did not show continuous records. Of note, the global fallout period (1963–  
 69 1967) is that with the lowest proportion of missing data, ranging from 20 to 30% in both  
 70 hemispheres, compared to higher proportions between 1954-1959 (~89%) and 1967-1976 (~40%) .

Proportion of EML <sup>90</sup>Sr data missing (%) between 1954-1976



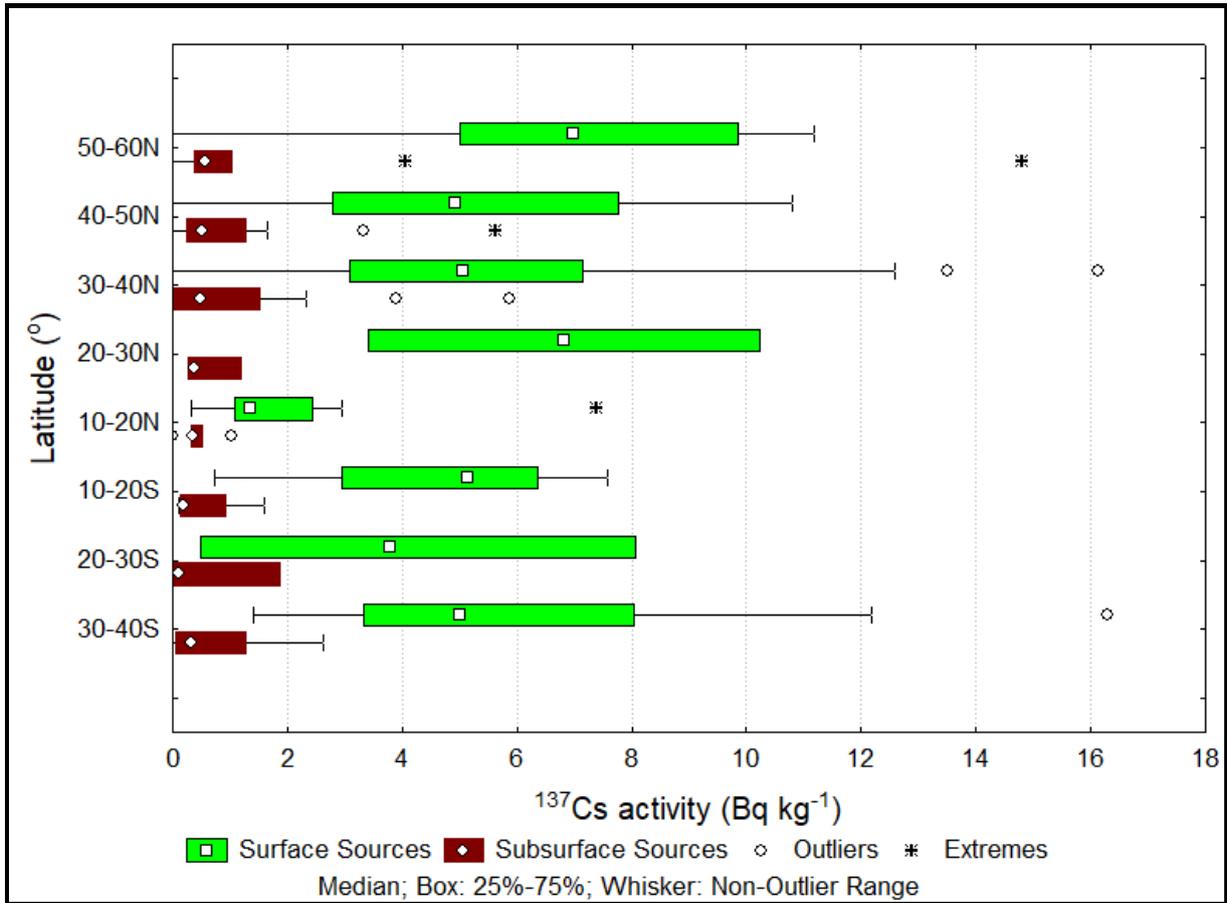
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83 Figure 5. Evolution of the  $^{137}\text{Cs}$  activities in (a) cultivated topsoil and (b) subsurface soils of the  
 84 northern hemisphere and (c) in cultivated topsoil and (d) subsurface soils of the southern  
 85 hemisphere depending on the proportion of  $^{137}\text{Cs}$  worldwide fallout after UNSCEAR (2000). Data  
 86 decay-corrected to January 1, 2019.



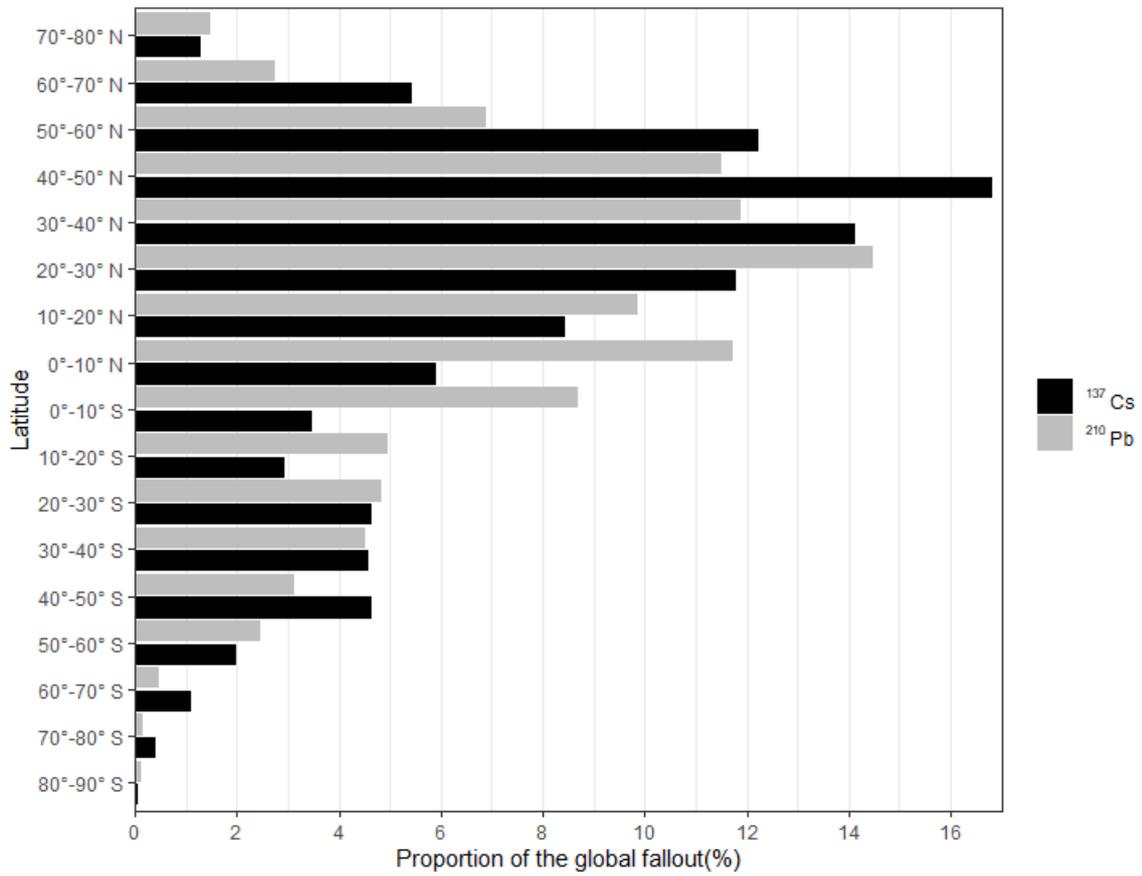
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96 Figure 6. Box-plots of  $^{137}\text{Cs}$  activities (decay-corrected to January 1, 2019) in surface and subsurface  
 97 sources across the globe.



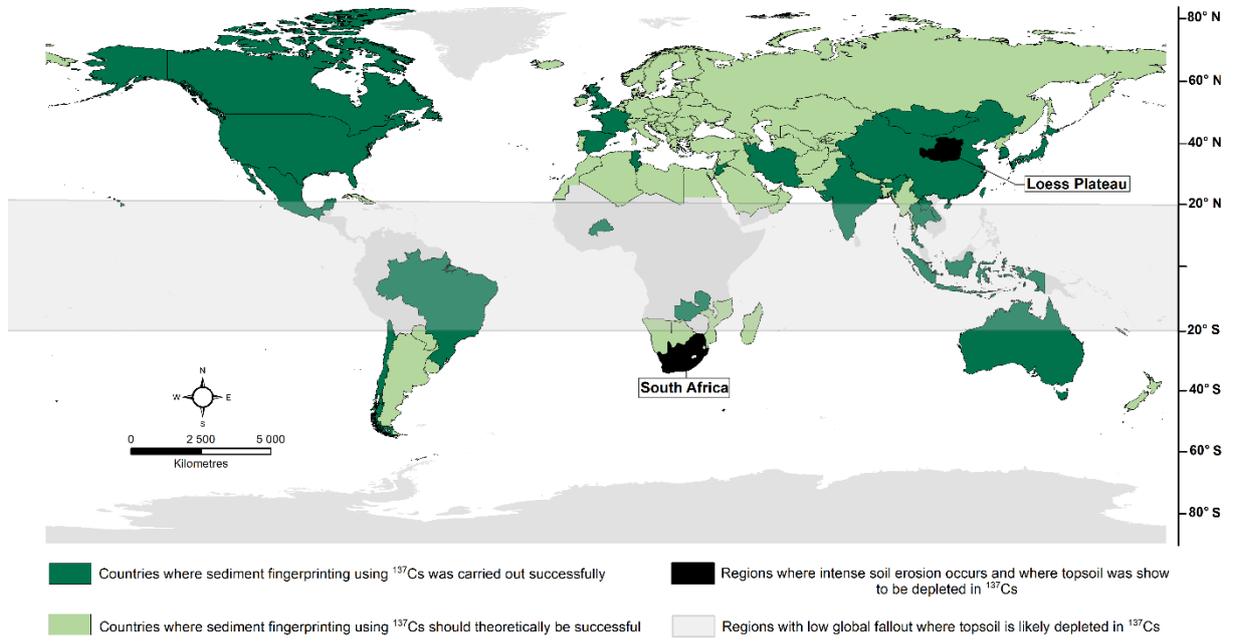
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110 Figure 7. Comparison of the proportion of the global fallout of atmospheric  $^{210}\text{Pb}$  (Baskaran, 2011)  
 111 and  $^{137}\text{Cs}$  (after UNSCEAR, 2000).



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126 Figure 8. Applicability of sediment fingerprinting techniques including  $^{137}\text{Cs}$  for discriminating surface  
127 and subsurface sources around the world.



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**SM1**

[Click here to download Supplementary material for online publication only: SM1\\_literature-review-data.xlsx](#)

SM2

[Click here to download Supplementary material for online publication only: SM2\\_137Cs\\_literature-values-Final.xlsx](#)