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

2

3 **Quantifying the resuspension of sediment and associated metallic contaminants with fallout**
4 **radionuclide measurements in a channelized river draining an industrial catchment**

5

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29 **Abstract**

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2 30 *Purpose:* Contamination of river sediment reaches problematic levels in industrialized regions. This
3
4 31 study investigates the relevance of using Beryllium-7 (^7Be), a short-lived environmental radionuclide
5
6 32 (53 d half life), for quantifying the deposition and resuspension of sediment and associated metallic
7
8 33 contaminants in a heavily polluted channelized river of Northern France (The Lower Scarpe River).

9
10 34 *Materials and methods:* Activities in ^7Be were measured in bed and suspended sediments collected
11
12 35 each month over an entire hydrological year. Suspended sediments were analysed for metal contents
13
14 36 (Pb, Zn, Cd, Cu). The determination of short-term sediment dynamics is based on the variation of
15
16 37 Total ^7Be inventory between two successive samplings. Total inventory of ^7Be consists of two
17
18 38 components, i.e., the Residual inventory and the New inventory.

19
20 39 *Results and discussion:* Inventories of ^7Be in sediment varied from 1 to 135 mBq cm^{-2} . Observed
21
22 40 spatial and temporal variations reflected the dynamical behaviour of sediment in the studied channel
23
24 41 section. The succession of sediment deposition and resuspension periods was demonstrated during
25
26 42 the study, with maximum deposition and erosion rates of 0.3 ± 0.2 and -0.46 ± 0.05 $\text{g cm}^{-2} \text{mo}^{-1}$,
27
28 43 respectively. A sediment resuspension rate of 0.06 $\text{g cm}^{-2} \text{mo}^{-1}$ supplied to the water column a flux of
29
30 44 12, 13, 23 and 145 $\mu\text{g cm}^{-2} \text{mo}^{-1}$ of Cd, Cu, Pb and Zn, respectively. During this 15-months study, 28
31
32 45 to 50 % of Pb that deposited in the riverbed sediment was resuspended, contributing thereby to the
33
34 46 (short term) degradation of water quality.

35
36 47 *Conclusions:* The present work provides a useful tool for examining the role of sediment as a sink or
37
38 48 source of contamination for the water column. It produces knowledge of how sediment processes
39
40 49 affect the fate and the transport of contaminants and the implications for future and downstream
41
42 50 outflows.

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44 51
45
46 52 **Keywords** Beryllium-7 • Sediment deposition/resuspension rate • Metallic contaminant • Water-
47
48 53 sediment interface

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50 54

55 1 Introduction

1
2 56 Contamination of river sediment reaches problematic levels in industrialized and urbanized areas.
3
4 57 Accumulation of contaminants in soils and their subsequent supply to rivers constitute a major
5
6 58 environmental and public health problem in industrialized countries (Gateuille et al. 2014).
7
8 59 Anthropogenic emissions largely contributed to the supply of metallic contaminants to river systems
9
10 60 through point and diffuse sources (Ayrault et al. 2012). In river systems, metallic contaminants are
11
12 61 preferentially associated with the finest fractions of particles (<63 µm) that are also the most easily
13
14 62 transported (Salomons and Förstner 1984; Owens et al. 2005), and sediment constitutes therefore a
15
16 63 significant pollution reservoir (Gibbs 1973; Lee et al. 2003; Westrich and Förstner 2007). A better
17
18 64 understanding of riverine dynamics is therefore needed to improve our knowledge on the sources of
19
20 65 the current contamination in rivers and to meet the targets required by the law [European Water
21
22 66 Framework Directive (Roche et al. 2005)].

23
24 67 Physical disturbances induced by natural and/or anthropogenic events at the water-sediment interface
25
26 68 can result in the resuspension of contaminated sediments and in the remobilization of metals from
27
28 69 sediment in the water column (Haag et al. 2001; Zoumis et al. 2001; Prygiel et al. 2015; Superville et
29
30 70 al. 2015). The fate of metallic contaminants is therefore closely linked with the evolution of sediment
31
32 71 dynamics (Calmano et al. 1993; Horowitz 2000; Walling et al. 2003). Storage of sediment and
33
34 72 associated contaminants can occur over the short or the long term depending on temporal variations
35
36 73 of the river flow and its transport capacity.

37
38 74 Several fallout radionuclides were shown to be particle-reactive and to provide appropriate tools to
39
40 75 quantify sediment dynamics in rivers. In particular, naturally occurring and short-lived Beryllium-7 (⁷Be,
41
42 76 53 d half-life) was extensively used in different types of aquatic systems (Krishnaswami et al. 1980;
43
44 77 Mabit et al. 2008, Walling 2013, Taylor et al. 2013), as in marine and coastal environments (Palinkas
45
46 78 et al. 2005), lakes (Dominik et al. 1989) and rivers (Dominik et al. 1987; Fitzgerald et al. 2001; Smith
47
48 79 et al. 2014). This radionuclide is produced in the atmosphere by the cosmic ray spallation of oxygen
49
50 80 and nitrogen atoms, and is rapidly bound to aerosols and supplied to the Earth's surface by wet
51
52 81 deposition (Lal et al. 1958; Olsen et al. 1985; Bourcier et al. 2011). Once in terrestrial and aquatic
53
54 82 environments, ⁷Be quickly sorbs onto particles, as shown by its high distribution coefficient [K_d values
55
56 83 comprised between 10^4 – 10^5 (Olsen et al. 1986; Hawley et al. 1986; Jewda et al. 2008; Taylor et
57
58 84 al. 2012)], and ⁷Be-labelled sediment particles are mixed and transported in rivers (Baskaran and
59
60

85 Santschi 1993; Sommerfield et al. 1999). Beryllium-7 provides therefore a relevant tracer for
1 86 quantifying sediment deposition and resuspension. Tracing is improved when combining ^7Be with
2
3 87 Cesium-137 (^{137}Cs , 30 y half-life) and Lead-210 (^{210}Pb , 22 y half-life) measurements, i.e., two longer-
4
5 88 lived fallout and particle-bound radionuclides (Du et al. 2010; Evrard et al. 2010; Huang et al. 2011).
6
7 89 Sediment residence times were quantified using ^7Be radionuclide mass balance models at the
8
9 90 catchment scale (Dominik et al. 1987; Bonté et al. 2000; Jweda et al. 2008; Evrard et al. 2010).
10
11 91 Furthermore, shorter-term sediment dynamics were quantified through the variation of ^7Be inventory in
12
13 92 surface bed sediment (Dibb and Rice 1989; Canuel et al. 1990; Giffin and Reide Corbett 2003).
14
15 93 However, to our knowledge, these techniques have been rarely applied to other particle-bound
16
17 94 contaminants, such as metals (Feng et al. 1999; Huang et al. 2011) and PCBs (Fitzgerald et al. 2001).
18
19 95 This situation can probably be attributed to the difficulty of conducting frequent sediment sampling in
20
21 96 rivers and to analyse the ^7Be in collected material rapidly after sampling (before ^7Be has decayed too
22
23 97 much).
24
25 98 In this context, the present paper investigates the potential of using ^7Be as a tracer of sediment and
26
27 99 particle-bound contaminant dynamics in a heavily polluted river section. We therefore propose to
28
29 100 quantify the resuspension of particle-bound metals in a river catchment strongly impacted by
30
31 101 industrialization and urbanization. To this end, we selected a catchment of Northern France (Lower
32
33 102 Scarpe River) where the metal pollution levels were high and representative of the ones found in
34
35 103 similar regions of Northwestern Europe, and where the feasibility of radionuclide tracing was
36
37 104 demonstrated (Zebracki et al. 2007; Zebracki 2008).
38
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41

42 106 **2 Study site**

43
44 107 The Lower Scarpe River flows in Northern France, where it drains a 624-km² catchment (Fig. 1). It
45
46 108 consists in a vast alluvial plain with a levelled topography (16–20 m altitude, 0.2 % mean slope). The
47
48 109 catchment surface is mainly covered with agricultural land (60 %) and urban areas (17 %).
49

50 110 With a 37-km length, the Lower Scarpe River is one of the six main tributaries of the Scheldt River,
51
52 111 which flows across France, Belgium and the Netherlands before reaching the North Sea. The river has
53
54 112 been channelized during the 17th century. Intense coal mining exploitation that took place in the
55
56 113 catchment between the 18th and the 20th centuries led to the subsidence of the land, so that the river
57
58 114 level is higher than the nearby land.
59
60
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65

115 The study area corresponds to a 7-km long and shallow river section located between two locks (25 m
116 channel width, 1–3 m water depth). The water discharge is measured with a 15-min time step at the
117 upstream lock (“Fort de Scarpe” gauging station operated by “Voies Navigables de France”). Flow is
118 low and regulated ($1 \text{ m}^3 \text{ s}^{-1}$). During flood periods, water overflow is mainly discharged into the Deule
119 River and partially into the Lower Scarpe River. Navigation in this river section has stopped in 2003. In
120 the study area, rainfall is measured using two tipping bucket rain gauges (operated by “Société des
121 Eaux de Douai”).

122 Sediment of the Lower Scarpe River is heavily contaminated with metals (Zebracki et al. 2005;
123 Zebracki 2008; Prygiel et al. 2015): metal contents in riverbed sediment reach up to 13,000, 2,400,
124 600 and $500 \mu\text{g g}^{-1}$ for Zn, Pb, Cd and Cu, respectively. Metallic contamination in sediment originates
125 from runoff on contaminated land and direct wastewater discharges. Past and current metallurgical
126 activities are responsible for soil contamination (Sobanska 1999).

127 **3 Materials and methods**

128 **3.1 Field sampling and laboratory analyses**

129 **3.1.1 Sampling sites**

130 Based on previous research that investigated the long-term accumulation of sediment and particle-
131 bound metals in this catchment (Zebracki et al. 2005; Zebracki 2008), 3 sampling sites representative
132 of the range of conditions found along the river were selected (Fig. 1). Sites k13 and k16 were situated
133 along a linear section of the channel at respective distances of 1.3 and 1.6 km from the upstream lock,
134 and k16 site was located just downstream of a lift station (where water from nearby land is pumped
135 and occasionally discharged into the river). The third sampling site (k36) is situated in a meander 2 km
136 further downstream where sediment accumulated at a larger rate and displayed higher contents in
137 metals than at k13 and k16 sites (Zebracki et al. 2005; Zebracki 2008).

138 **3.1.2 Sampling procedures**

139 The surface layer of riverbed sediment and suspended matter were collected from a light boat each
140 month at the 3 sites from April 2005 to August 2006 (15 campaigns separated by 20 to 43 d). Surface
141 bed sediment was collected using an Eijkelkamp® manual corer (Beeker type) that did not disturb the
142 water-sediment interface. Tube corer (57 mm internal diameter) was gently pushed into bed sediment

145 to collect a vertical column of sediment. Cores length did not exceed 20 cm. Sediment cores were
146 extruded and sectioned into subsamples 12 h maximum after sampling. Maximal depth of ^7Be
147 occurrence in sediment was investigated during the first sampling campaign (04/19/2005): for each
148 site, sediment cores were sectioned into 5 subsamples with 1-cm increments and subsamples were
149 mixed for further analyses. Sediment cores were systematically collected in 3 replicates within a 4-m²
150 area around each site.

151 Suspended matter was collected for measurement of time-integrated ^7Be activities in suspended
152 sediment at the same sites using vertical traps, which were immersed at mid-height of water depth
153 during the period of time between two successive samplings (i.e., 20–43 d). As previously reported by
154 Fitzgerald et al. (2001), ^7Be activities were expected not to vary significantly with water depth in
155 suspended material (i.e., < 30 %). Traps were built with PVC cylinders (60 cm height, 15 cm diameter)
156 and closed with a cap at the lower extremity. Aspect ratio (height vs. diameter) of traps relied within
157 the range of values suggested for effective particle trapping [i.e., 4:1 (Gardner 1980)]. Each trap was
158 ballasted and held in a vertical position using a floater. Additional suspended sediment was collected
159 at the lift station “Vallée de Scarpe” by immersing vertically a cylindrical 1.5 L mineral water PET bottle
160 with two holes of 5 cm diameter. On-site, material stored in the traps was collected with HDPE
161 Nalgene® jerricans and the remaining solids were recovered by centrifugation at the CNRSSP
162 laboratory.

163 Samples were oven-dried at 40°C for 2 d, crushed and homogenized in a ceramic mortar with a
164 pestle, and finally packed into tightly closed plastic containers for further analysis.

165 The location of the CNRSSP laboratory (Douai, France) in the close vicinity of the study area (2–7 km)
166 facilitated sampling procedures and helped to avoid degradation of samples during transport.

167

168 **3.1.3 Sample analyses**

169 Bed and suspended sediments were analysed for fallout radionuclides by gamma spectrometry at the
170 LSCE laboratory, Gif-sur-Yvette, France. Sample quantities varied between 1 and 55 g. Samples were
171 analysed for 24 h using very low-background and high-resolution, coaxial N-type GeHP detectors
172 (Canberra/Eurisys). Efficiencies and background were periodically calibrated with international
173 standards and extrapolated for ^7Be gamma energy level (477 keV). Activity in $^{210}\text{Pb}_{\text{xs}}$ (excess ^{210}Pb)
174 was calculated by subtracting the supported activity from the total ^{210}Pb activity that is in secular

175 equilibrium with the ^{238}U decay products. Due to its long-term radioactive decay, the activity in ^{232}Th
176 was estimated by measuring the content of its short-lived filiation product, i.e., ^{228}Ac , and assuming a
177 secular equilibrium within the radioactive decay series. The use of ^{228}Ac as a proxy to estimate ^{232}Th
178 content in sediment relies on the assumption that ^{228}Ra is not preferentially remobilized in the riverine
179 system.

180 Specific activities were expressed in Bq kg^{-1} dry weight and were systematically decay-corrected to
181 the date of sampling. Uncertainties on activities were expressed at $\pm 2 \sigma$.

182 As size distribution in sediment particles may influence the content of particle-bound fallout
183 radionuclides and metals (Benoit and Rozan 1999; Jweda et al. 2008; Kaste and Baskaran 2011), the
184 potential particle size effect was evaluated through grain size analysis. It was performed on a selection
185 of suspended and riverbed sediment samples with a Beckman Coulter LS-230 laser connected to a
186 hydro small-volume dispersion unit available at GCE laboratory, Douai, France. Bulk solid samples
187 were mixed and resuspended in a solution of sodium hexametaphosphate 5 %. Analysis was
188 systematically repeated for 3 subsamples. In addition, the potential particle size effect was evaluated
189 through the measurement of ^{232}Th content in sediment (i.e., ^{232}Th activity), as it was shown to provide
190 a proxy of fine-grained sediment content in sedimentary environments (Sakaguchi et al. 2006;
191 Foucher et al. 2015).

192 Since ^7Be sorption in sediment may depend on the nature of geochemical phases [i.e., Fe/Mn oxides
193 (Taylor et al. 2012) and organic phases (Blake et al. 2009)], the potential influence of organic carbon
194 content on ^7Be activities was evaluated by conducting total organic carbon analyses. Total carbon and
195 inorganic carbon contents were determined using a Shimadzu Total Organic Carbon analyser.
196 Analysis was systematically repeated for 3 subsamples.

197 Contents in metals were quantified in suspended sediments after *aqua regia* micro-wave (CEM μ -
198 waves MARS 5) assisted dissolution (0.5 g of air-dried and crushed sediment; $\text{HNO}_3:\text{HCl}$ 1:3).
199 Solutions were analysed using Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES;
200 Jobin-Yvon 138 ULTRACE). A certified reference material (TH2 Toronto Harbour Sediment for Trace
201 Elements, National Water Research Institute, Canada) was used for quality control. Analysis was
202 systematically conducted on 3 subsamples. Uncertainties on metal contents were derived from the
203 standard analytical deviations and were expressed at $\pm 1 \sigma$.

204

205 3.2. Short-term sediment dynamics

206 3.2.1 Inventory of ⁷Be in bed sediment

207 The use of ⁷Be to characterize short-term sediment dynamics relies on several assumptions:

- 208 - occurrence of continuous inputs of ⁷Be in the river,
- 209 - recently eroded sediment particles that settled down on the riverbed are labelled by ⁷Be
- 210 inputs,
- 211 - ⁷Be delivery and removal processes occur only in association with particulate matter,
- 212 - and fixation of ⁷Be to sediment particles is irreversible.

213 The determination of short-term sediment dynamics is based on the variation of Total ⁷Be inventory
214 between two successive samplings (Dibb and Rice 1989; Canuel et al. 1990). Total inventory of ⁷Be
215 [$I(^{7}\text{Be})_{\text{TOTAL}}$] consists of two components, i.e., the Residual inventory [$I(^{7}\text{Be})_{\text{RESIDUAL}}$] and the New
216 inventory [$I(^{7}\text{Be})_{\text{NEW}}$]:

$$217 \quad (1) \quad I(^{7}\text{Be})_{\text{TOTAL}} = I(^{7}\text{Be})_{\text{RESIDUAL}} + I(^{7}\text{Be})_{\text{NEW}}.$$

218 Residual inventory corresponds to the inventory calculated during the previous sampling period and
219 decay-corrected to the date of the next campaign. New inventory is calculated by subtracting this
220 residual inventory from the Total inventory for this campaign. In this context and for a given campaign,
221 a higher Total ⁷Be inventory reflects deposition of sediment, whereas a lower Total ⁷Be inventory
222 corresponds to the occurrence of sediment resuspension from the riverbed.

223 Total ⁷Be inventory in surface bed sediment [$I(^{7}\text{Be})_{\text{TOTAL}}$, in mBq cm⁻²] is calculated by multiplying
224 specific ⁷Be activity (A_i , in Bq kg⁻¹) by the areal mass of sediment in each core section (M_i , in g cm⁻²),
225 and by summing ⁷Be inventory of all individual subsections ($\sum_i A_i$, i corresponding to the sediment
226 layer, i.e., 0–3 or 3–5 cm):

$$227 \quad (2) \quad I(^{7}\text{Be})_{\text{TOTAL}} = \sum_i A_i \times M_i \times 10^{-3}.$$

228 When calculating Total ⁷Be inventory in sediment of the Lower Scarpe River, ⁷Be activities lower than
229 the limits of quantification were estimated to be equal to half of the limits of quantification. Total ⁷Be
230 inventories correspond to the mean of ⁷Be inventories obtained for sediment replicates.

231 It should be noted that this method provides results on sediment deposition or resuspension between
232 the sampling events. However, although they may not be reflected in the global results, these
233 processes may have occurred locally and temporarily between the sampling campaigns (Giffin and
234 Reide Corbett 2003).

235 When calculating ${}^7\text{Be}$ activities, the ${}^7\text{Be}$ decay correction did not depend on the potential variability of
1 sediment discharge throughout time in the river. In this context, the lack of information regarding the
2
3
4 237 temporal variability of sediment discharge may lead to an underestimation of ${}^7\text{Be}$ activities, and thus
5
6 238 Total ${}^7\text{Be}$ inventories (for instance, when the majority of sediment discharge occurs early in the period
7
8 239 of time between the sampling events).

9
10 240

11 241 **3.2.2 Short term sediment deposition/resuspension rate**

12 242 Rate of sediment deposition/resuspension (R_{SEDIMENT} , in g cm^{-2}) is determined between two
13
14 243 successive samplings by dividing the New ${}^7\text{Be}$ inventory in sediment [$I({}^7\text{Be})_{\text{NEW}}$, in mBq cm^{-2}] by the
15
16 244 mean ${}^7\text{Be}$ activity [$A({}^7\text{Be})_{\text{MEAN}}$, in Bq kg^{-1}] of newly deposited or resuspended sediment (Canuel et
17
18
19
20 245 al. 1990):

$$21 \quad 22 \quad 23 \quad 24 \quad 25 \quad 26 \quad 27 \quad 28 \quad 29 \quad 30 \quad 31 \quad 32 \quad 33 \quad 34 \quad 35 \quad 36 \quad 37 \quad 38 \quad 39 \quad 40 \quad 41 \quad 42 \quad 43 \quad 44 \quad 45 \quad 46 \quad 47 \quad 48 \quad 49 \quad 50 \quad 51 \quad 52 \quad 53 \quad 54 \quad 55 \quad 56 \quad 57 \quad 58 \quad 59 \quad 60 \quad 61 \quad 62 \quad 63 \quad 64 \quad 65$$
$$(3) \quad R_{\text{SEDIMENT}} = \frac{I({}^7\text{Be})_{\text{NEW}}}{A({}^7\text{Be})_{\text{MEAN}}} \times 10^{-3}.$$

25 247 In the present study, the mean ${}^7\text{Be}$ activity is determined from suspended sediment (Fitzgerald et
26
27 248 al. 2001; Zebracki et al. 2007; Zebracki 2008) and not in surface sediment investigated by other
28
29 249 authors (Canuel et al. 1990; Giffin and Reide Corbett 2003), assuming that characteristics of sediment
30
31 250 depositing on the bed or being resuspended are representative of the characteristics of suspended
32
33 251 sediment.

34
35 252 Sediment deposition/resuspension rate is normalised at the monthly scale (N_{DAYS} corresponds to the
36
37 253 time period between two successive sampling campaigns, in d) and is expressed in $\text{g cm}^{-2} \text{ mo}^{-1}$:

$$40 \quad 41 \quad 42 \quad 43 \quad 44 \quad 45 \quad 46 \quad 47 \quad 48 \quad 49 \quad 50 \quad 51 \quad 52 \quad 53 \quad 54 \quad 55 \quad 56 \quad 57 \quad 58 \quad 59 \quad 60 \quad 61 \quad 62 \quad 63 \quad 64 \quad 65$$
$$(4) \quad R_{\text{SEDIMENT}} = \frac{I({}^7\text{Be})_{\text{NEW}}}{A({}^7\text{Be})_{\text{MEAN}}} \times 30 / N_{\text{DAYS}}.$$

43 255 Positive values of sediment rate indicate deposition whereas negative values correspond to
44
45 256 resuspension. For a given period, Net sediment rate corresponds to the sum of the monthly short-term
46
47 257 rates of sediment deposition and resuspension.

48
49 258

51 259 **3.2.3 Application to the quantification of contaminants fluxes at water-sediment interface**

52 260 Flux of contaminants associated with sediment deposition/resuspension (R_{METAL} , expressed in
53
54 261 $\mu\text{g cm}^{-2} \text{ mo}^{-1}$) is calculated by multiplying short-term sediment rate (R_{SEDIMENT} , in $\text{g cm}^{-2} \text{ mo}^{-1}$) by
55
56 262 metal content (C_{METAL} , in $\mu\text{g g}^{-1}$) in suspended sediment, assuming that characteristics of sediment

263 depositing on the bed or being resuspended are representative of the characteristics of suspended
 1 264 sediment, and that metals associated with sediment remain fixed to particles during their transport:

3
 4 265 (5) $R_{METAL} = R_{SEDIMENT} \times C_{METAL}$.

5
 6 266 For a given period, Net flux of contaminants is obtained by summing the monthly short-term fluxes of
 7
 8 267 contaminant deposition and resuspension.

9
 10 268

11
 12 269 **3.2.4 Fraction of recent ⁷Be-labelled sediment in suspended sediment**

13
 14 270 To distinguish sediment recently labelled with ⁷Be from older (⁷Be-depleted) sediment, an indicator of
 15
 16 271 the fraction of the suspended sediment that has been recently eroded from soils is given by the
 17
 18 272 ⁷Be/²¹⁰Pb_{xs} activity ratio (Matisoff et al. 2005; Evrard et al. 2010). The fraction of recent ⁷Be-labelled
 19
 20 273 sediment (i.e., recent sediment, expressed in %) is given by dividing the ⁷Be/²¹⁰Pb_{xs} activity ratio in
 21
 22 274 suspended sediment [(⁷Be/²¹⁰Pb_{xs})_{Suspended sed.}] by the ⁷Be/²¹⁰Pb_{xs} activity ratio in rainfall
 23
 24 275 [(⁷Be/²¹⁰Pb_{xs})_{Source}]:

25
 26
 27
 28 276 (6) $Recent\ sediment, \% = 100 \times \frac{\left(\frac{{}^7Be}{{}^{210}Pb_{xs}} \right)_{Suspended\ sed.}}{\left(\frac{{}^7Be}{{}^{210}Pb_{xs}} \right)_{Source}}$.

29
 30
 31
 32 277 The sediment source term is defined by sediment particles that have a ⁷Be/²¹⁰Pb activity ratio equal to
 33
 34 278 that of wet fallout. The dry deposition of ⁷Be and the transfer time required for ⁷Be-labelled particles to
 35
 36 279 be supplied from catchment soils to the river through erosion processes were neglected, as both
 37
 38 280 phenomena were supposed to be of minor importance (Matisoff et al. 2005; Conaway et al. 2013;
 39
 40 281 Pham et al. 2013). Fallout ⁷Be and ²¹⁰Pb were monthly measured in the framework of the Permanent
 41
 42 282 observatory of air radioactivity (OPERA-Air sampling network, operated by the French Institute of
 43
 44 283 Radioprotection and Nuclear Safety). The closest station to the study area is situated at 180 km
 45
 46 284 (Charleville-Mezieres). The ⁷Be/²¹⁰Pb_{xs} source term is calculated using areal activities in ⁷Be and ²¹⁰Pb
 47
 48 285 that were derived from activities in rainwater samples (Bq L⁻¹) collected each month and available
 49
 50 286 rainfall data (L m⁻²).

51
 52 287

53
 54 288 **4 Results and discussion**

55
 56 289 **4.1 Hydrological conditions**

57
 58 290 Between April 2005 and August 2006, the mean water discharge measured at the upstream lock
 59
 60 291 gauging station was 1.0±0.2 m³ s⁻¹. One extreme rainfall event occurred on July 7, 2005 (137 mm of

292 rainfall during in 7 h; 100 yr recurrence interval) and triggered a flood recorded at the upstream lock
293 (Fig. 2). Overall, no relationship between rainfall and water discharge was found.

294

295 **4.2 Grain size of sediment**

296 Bed sediments were characterised by the dominance of the silt- and clay-sized material fractions
297 (51–86 % were <63 µm; Table S1, Electronic Supplementary Material). Considering the mean fraction
298 of fine material (<63 µm), bed sediment displayed finer material at k36 (76±5 %) than at k13 and k16
299 (60±6 %), reflecting the differences in sediment accumulation due to variations in channel morphology.
300 In suspended sediment (Table S2, Electronic Supplementary Material), the fraction of fine material
301 was dominant at each sampling site (62–90 %), and the mean fractions were comparable at the 3
302 sites (73–84 %).

303 ¹³⁷Cs was used to detect a potential grain size effect on the adsorption of fallout radionuclides to
304 sediment (He and Walling 1996; Bonté et al. 2000), and ²³²Th was used as a proxy of fine-grained
305 sediment content (Sakaguchi et al. 2006; Foucher et al. 2015). The positive trend found between ¹³⁷Cs
306 activities and (a) fractions of fine material, or (b) ²³²Th activities, confirms the potential occurrence of a
307 grain size effect (Fig. S1, Electronic Supplementary Material). However, data were poorly correlated
308 ($R^2 \leq 0.3$), suggesting that variations in ¹³⁷Cs activities also reflect variations in sediment origin
309 (Wallbrink et al. 1996).

310 **4.3 Rainwater ⁷Be and ²¹⁰Pb activities**

311 Rainwater ⁷Be and ²¹⁰Pb activities were available for the period between October 2005 and August
312 2006 (Table S3, Electronic Supplementary Material). Observed variations in radionuclide activities can
313 be explained by intra-event evolution (Gourdin et al. 2014), the efficiency of ⁷Be washout from the
314 atmosphere, origin of air masses, and atmospheric production rate of ⁷Be (Bourcier et al. 2011, Pinto
315 et al. 2013, Conaway et al. 2013).

316

317 **4.4 Beryllium-7 in bed sediment**

318 **4.4.1 Depth occurrence of ⁷Be in sediment**

319 The vertical distribution of ⁷Be in sediment was investigated during the first sampling campaign (April
320 19, 2005). Activities in ⁷Be varied from undetectable levels (< 3–7 Bq kg⁻¹) to 46 Bq kg⁻¹, and
321 decreased with depth at the 3 sampling sites (Table 1). This result reflects the progressive burial and

322 the associated radioactive decay of ^7Be associated with sediments in depth. As expected, ^7Be is only
1 detected in top layers of sediment (< 5 cm), and we therefore restricted sampling to the top 5-cm layer
2 323
3 of sediment during the next campaigns. These 5-cm long sediment cores were further sectioned into 2
4 324
5 subsamples (0–3 and 3–5 cm).
6 325

7 326

10 327 **4.4.2 Total ^7Be inventory of bed sediment**

11 328 In sediment layers collected from April 2005 to August 2006 (n=258), ^7Be inventories ranged from 1 to
12 328
13 135 mBq cm⁻² (Table 2). Total ^7Be inventories in sediment ranged from 5 to 151 mBq cm⁻² (Table 3).
14 329
15 Mean coefficients of variation between sediment replicates ranged from 28 % (k16) to 37 % (k13) and
16 330
17 exceeded the ones obtained for grain size distribution (< 7 %). Variations in ^7Be inventories between
18 331
19 sediment replicates are then rather influenced by the spatial heterogeneity in the deposition of ^7Be -
20 332
21 labelled particles on the riverbed, as sediment activity in ^7Be depends on both its age and its origin.
22 333
23 These mean uncertainties were consistent with the values previously reported from other aquatic
24 334
25 environments [25–33 % in coastal systems (Canuel et al. 1990, Giffin and Reide Corbett 2003)].
26 335

27 336 By comparing ^7Be inventories in top and bottom sediment layers (0–3 cm vs. 3–5 cm), 4 types of
28 336
29 sediment deposition patterns were found. The highest ^7Be activities were generally found in the top
30 337
31 sediment layer (91 % of observations) indicating the recent deposition of ^7Be -labelled sediment. In
32 338
33 some cases, both top and bottom sediments displayed low ^7Be inventories (< 5 % of cases), indicating
34 339
35 either the resuspension of previously deposited sediment or the absence of sediment deposition.
36 340
37 Rarely (<3 % of cases), ^7Be inventory remained relatively constant with depth, indicating the
38 341
39 occurrence of local physical disturbances, such as bioturbation that contributed to homogenize the
40 342
41 ^7Be records in depth (Santschi et al. 1990, Ciutat et al. 2005).
42 343

43 344 Although the potential loss of ^7Be through remobilisation under changing environmental conditions
44 344
45 (i.e., variations in pH, redox potential, age of sediment) during sediment deposition and resuspension
46 345
47 stages is unlikely to have occurred in our study area given the rapid cycling of material through the
48 346
49 system, this process, if confirmed, could lead to underestimations of ^7Be inventories in sediment
50 347
51 (Taylor et al. 2012, Taylor et al. 2013).
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350 4.5 Beryllium-7 in suspended sediment

1
2 351 Activities in ^7Be of suspended sediment ($n=35$) ranged from 81 to 582 Bq kg $^{-1}$ (Table 3). Variations in
3
4 352 ^7Be activities of suspended sediment are not explained by differences in fine-grained sediment nor
5
6 353 variations in organic carbon contents (Fig. S2, Electronic Supplementary Material), and they rather
7
8 354 reflect variations in the atmospheric supply of ^7Be and origin of sediment (Bonté et al. 2000; Le
9
10 355 Cloarec et al. 2007; Jweda et al. 2008).

11
12 356 During the study period, rainfall variations had an impact on ^7Be activities measured in suspended
13
14 357 sediment (Fig. 3): in July 2005 and May 2006, increase in ^7Be activities (up to 582 Bq kg $^{-1}$) was
15
16 358 associated with increasing monthly cumulated rainfall (up to 137 mm). In contrast, low ^7Be activities
17
18 359 (< 223 Bq kg $^{-1}$) and low variations recorded from September 2005 to April 2006 were associated with
19
20 360 low cumulated rainfall (< 52 mm).

21
22 361 Direct deposition of fallout radionuclides in river water is generally considered to be of minor
23
24 362 importance (Jweda et al. 2008).

25
26 363 In July 2005, increasing ^7Be activities of suspended sediment coincided with flood occurrence (Fig. 3),
27
28 364 suggesting a significant delivery of recently ^7Be -labelled sediment particles to the river.

29
30 365 Activities in ^{137}Cs measured in suspended sediment (5.3–9.9 Bq kg $^{-1}$; Table S3) indicate that overall
31
32 366 sediment supply in the Scarpe River derives from the erosion of surface soils (Ben Slimane et
33
34 367 al. 2013; Jagercikova et al. 2015). The preferential erosion of top vs. subsurface soils is consistent
35
36 368 with the low relief of the catchment and the overall occurrence of low-intensity continuous rainfall. In
37
38 369 contrast, in mountainous river catchments affected by heavy storms, decreasing in ^7Be and ^{137}Cs
39
40 370 activities of suspended sediment is observed due to the erosion of particles originating from deeper
41
42 371 soils that are depleted in fallout radionuclides (Olley et al. 1993; Evrard et al. 2011; Conaway et
43
44 372 al. 2013).

45
46 373 The differences in ^7Be activities observed between sites in July 2005, July 2006 and August 2006
47
48 374 suggest the occurrence of mixing processes in the river between “recent” (^7Be -enriched) and “older”
49
50 375 (^7Be -depleted) sediment particles (Olsen et al. 1985; Wallbrink and Murray 1996; Matisoff et al. 2005).
51
52 376 In July and August 2006, decreasing ^7Be activities in downstream direction was related to decreasing
53
54 377 fractions of recent sediment (Table S3). In November 2005 and April 2006, ^7Be activities displayed low
55
56 378 variations between sites (<23 %), suggesting a relative homogeneity of suspended material, which
57
58 379 was confirmed by low variations in estimated fractions of recent sediment (13–19 %).
59
60

380 The positive trend observed between ^7Be records in top sediment layers (0–3 cm) and suspended
381 sediments indicates that both material types are affected by the supply of recent ^7Be -labelled particles
382 (Fig. 4). This behaviour was observed in other coastal and riverine environments (Roos and
383 Valeur 2006; Jweda et al. 2008).

384 Our results confirm the relevance of using ^7Be records in suspended sediment to normalize variations
385 in ^7Be inventories of sediment (Equation 3), as this material integrates the variability of ^7Be delivery
386 over time.

387

388 **4.6 Sediment deposition/resuspension rate**

389 Results show an alternation of periods of sediment deposition and resuspension (Table 3, Fig. 5), with
390 rates ranging from $-0.5 \text{ g cm}^{-2} \text{ mo}^{-1}$ to $0.3 \text{ g cm}^{-2} \text{ mo}^{-1}$. These values are comprised within the range
391 of short-term sediment dynamics found in other rivers and coastal systems [-0.7 to $1.4 \text{ g cm}^{-2} \text{ mo}^{-1}$
392 (Canuel et al. 1990); -0.08 to $0.3 \text{ g cm}^{-2} \text{ mo}^{-1}$ (Fitzgerald et al. 2001)].

393 At k36 site, the increase of sediment deposition rate between July and August 2005 coincided with the
394 flood of July, suggesting the delivery of sediment during and after the flood occurrence. The
395 occurrence of sediment resuspension in September 2005 after three months of sediment deposition
396 (June, July and August 2005) may indicate that recently deposited sediment was not consolidated,
397 which facilitated its resuspension. Nonetheless, further investigations are required to examine the
398 characteristics of in-channel processes that result in sediment resuspension (as flow velocity, critical
399 shear-stress). The monitoring of turbidity alongside the river flow could for instance provide a better
400 characterization of suspended sediment hysteresis (e.g., Prygiel et al. 2015).

401 Frequency and magnitude of sediment deposition/resuspension differed between the 3 sites,
402 highlighting the spatial and temporal variations of short-term sediment dynamics.

403 During the 15-months monitoring, Net sediment rates were positive for all sites, indicating sediment
404 accumulation. These rates ranged from 0.3 to 0.7 g cm^{-2} , and proportions of resuspended sediment
405 reached 35–43 %. Over one year (May 2005 to May 2006), annual sedimentation rates were
406 estimated to 0.3 , 0.6 and $0.8 \text{ g cm}^{-2} \text{ y}^{-1}$, for k13, k16 and k36 sites, respectively. These results remain
407 in agreement with the annual sedimentation rates previously calculated with the longer-lived ^{137}Cs and
408 ^{210}Pb radionuclides over the two last decades of sediment accumulation [0.4 , 0.2 and $0.8 \text{ g cm}^{-2} \text{ y}^{-1}$
409 for k13, k16 and k36, respectively (Zebracki et al. 2005; Zebracki 2008)]. The close agreement

410 between annual sedimentation rates obtained with different methods and over different time scales
1 411 confirms the previous assumptions of a steady state of sediment accumulation.
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3

4 412

5 413 **4.7 Contaminant fluxes at water-sediment interface**

6 414 **4.7.1 Metal contents in suspended sediment**

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8
9
10 415 Suspended sediment displayed high contents of Cd, Cu, Pb and Zn (Table 4). These contents remain
11
12 416 in the range of sediment contamination levels measured in other European rivers that were strongly
13
14 417 impacted by mining exploitation, industrialization and urbanization (Morillo et al. 2002; Lacal et
15
16 418 al. 2003; Walling et al. 2003; Audry et al. 2004; Meybeck et al. 2007; Le Cloarec et al. 2011; Prygiel et
17
18 419 al. 2015). Variations in metal contents did not reflect variations in grain size (Table S2, Electronic
19
20 420 Supplementary Material). Increasing metal content of suspended sediment in downstream direction
21
22 421 reflects the supply of contaminants from various point and diffuse sources of contamination, including
23
24 422 the lift station situated upstream of k16 (Fig. 1), at which suspended sediments were shown to display
25
26 423 high levels of metallic contamination, especially in Cd (Table S4, Electronic Supplementary Material).
27

28 424

29 30 425 **4.7.2 Flux of contaminants at water-sediment interface**

31
32 426 The monthly particulate fluxes of metal deposition and resuspension were calculated (Table S5,
33
34 427 Electronic Supplementary Material). Over the 15-months monitoring, the Net fluxes of contaminants
35
36 428 were positive for all sites (Table 5), demonstrating the role of sediment as a sink of contaminants,
37
38 429 except for Cd at k16 site, due to the combination of a high resuspension rate and a high Cd content.
39
40 430 Maximum Net fluxes of contaminants were found at k36 site, in association with locally high sediment
41
42 431 accumulation rate.

43
44 432 Although sediment of the Lower Scarpe River is shown to act as a sink for contaminants over years
45
46 433 and decades, resuspension periods contribute to the supply of contaminants back into the water. In
47
48 434 September 2005, sediment resuspension of $-0.06 \text{ g cm}^{-2} \text{ mo}^{-1}$ supplied fluxes to the water column of
49
50 435 12, 13, 23 and $145 \mu\text{g cm}^{-2} \text{ mo}^{-1}$ for Cd, Cu, Pb and Zn, respectively (Table S5, Electronic
51
52 436 Supplementary Material). As a comparison, a sediment resuspension of $0.08 \text{ g cm}^{-2} \text{ mo}^{-1}$ in the Fox
53
54 437 River corresponded to a $\sum PCB$ flux of $0.24 \mu\text{g cm}^{-2} \text{ mo}^{-1}$ (Fitzgerald et al. 2001). During the
55
56 438 15-months monitoring, 28 to 50 % of Pb that deposited at surface bed sediment was resuspended
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439 (Fig. 6). Even in such river systems (non-navigated, with low and regulated flow), minor resuspension
1 440 periods may significantly contribute to the temporal degradation of the water quality.
2
3 441 Furthermore, it is well known that resuspension of anoxic sediment in oxic water columns affects the
4
5 442 mobility of labile contaminants resulting in metal remobilization and enhancing their bioavailability
6
7 443 (Argese et al. 1997; Zoumis et al. 2001; Eggleton et al. 2004; Westrich and Förstner 2007; Hwang et
8
9 444 al. 2011). Increasing solubility of particle-bound Zn, Cd, Cu and Pb is therefore expected during
10
11 445 sediment resuspension stages (Tack et al. 1995; Förstner 2004; Superville et al. 2015).
12
13

14 446

16 447 **5 Conclusions**

17
18 448 This study provides to our knowledge one of the first attempts to combine quantifications of short-term
19
20 449 sediment dynamics and particle-bound fluxes of metallic contaminants in a channelized river draining
21
22 450 an industrialized catchment.

23
24 451 We show the relevance of using ^7Be records in riverbed and suspended sediments to calculate
25
26 452 sediment deposition and resuspension rates over the short term. It is demonstrated that ^7Be is found
27
28 453 in measurable quantities and continuously delivered to the river (i.e., through sediment supply and
29
30 454 resuspension).

31
32 455 Rates of short-term sediment dynamics ranged from $-0.46 \pm 0.05 \text{ g cm}^{-2} \text{ mo}^{-1}$ to $0.3 \pm 0.2 \text{ g cm}^{-2} \text{ mo}^{-1}$,
33
34 456 and Net annual sediment rates reflected large variations in sediment accumulation due to changes in
35
36 457 channel morphology. Sediment of the Lower Scarpe River acts as a sink for contaminants due to the
37
38 458 long-term accumulation of particle-bound metals in the riverbed. However, sediment acts as a source
39
40 459 of contamination during short resuspension periods. Sediment resuspended at a rate of
41
42 460 $-0.06 \text{ g cm}^{-2} \text{ mo}^{-1}$ generated a flux of metals to the river water column of 12, 13, 23 and
43
44 461 $145 \mu\text{g cm}^{-2} \text{ mo}^{-1}$ for Cd, Cu, Pb and Zn, respectively. Over the 15-months monitoring, 28 to 50 % of
45
46 462 Pb that deposited on the riverbed was resuspended and contributed to the concomitant degradation of
47
48 463 water quality.

49
50 464 The use of fallout radionuclides to quantify dynamics of sediment and particle-bound contaminants
51
52 465 proved to be relevant to demonstrate the role of sediment as a sink or source of contamination for
53
54 466 water. Further investigations are required to evaluate decontamination times in order to guide the
55
56 467 implementation of efficient management strategies aimed at improving water quality in these river
57
58 468 systems.
59

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677 **Table captions**

1 678 Table 1: Depth distribution of ^7Be activity (Bq kg^{-1}) in sediment collected in April 2005 (sampling
2 campaign n°1 on April 19, 2005).
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4 680
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6 681 Table 2: From April 2005 to August 2006: Inventory of ^7Be (mBq cm^{-2}) in top and bottom sediment
7 682 layers (0–3 and 0–5 cm layers) collected in triplicate (a, b and c). Three sediment layers were not
8 analysed due to the loss of samples during subsampling procedure (“n.d.” is mentioned). Activities in
9 683 ^7Be lower than the limits of quantification were estimated to be equal to half of the limits of
10 684 quantification.
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15 687 Table 3: From April 2005 to August 2006: Total, Residual and New inventories in ^7Be of sediment
16 688 (mBq cm^{-2}); Activity in ^7Be of suspended sediment (Bq kg^{-1}); Monthly rate of sediment
17 689 Deposition/Resuspension ($=\text{D/RS}$, in $\text{g cm}^{-2} \text{ mo}^{-1}$). When suspended sediment was not available (due
18 690 to the loss of sampling device), activity in ^7Be was assessed by calculating the mean activity
19 691 measured in suspended sediments collected during the same period at the other sites (mentioned in
20 692 grey).
21 693

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24 694 Table 4: Metal contents in suspended sediments ($\mu\text{g g}^{-1}$) collected from April 2005 to August 2006
25 695 (standard analytical deviation did not exceed 5 %). When suspended sediment was not available (due
26 696 to the loss of sampling device), metal content was assessed by calculating the mean content
27 697 measured in suspended sediments collected at the same site during the other sampling periods
28 698 (mentioned in grey).
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34 700 Table 5: From April 2005 to August 2006: Cumulated fluxes of contaminant deposition and
35 701 resuspension, and Net fluxes of contaminant ($\mu\text{g cm}^{-2}$). Proportions of Total resuspension regarding
36 702 Total deposition (%).
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705 **Figure captions**

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3 707 Fig. 1: The Lower Scarpe River catchment. Location of the 3 sampling sites (k13, k16 and k36), and
4 708 the upstream lock ("Fort de Scarpe"). The river receives water discharges from one tributary (T) and 2
5 709 lift stations (LS1, LS2), and liquid effluents originating from one wastewater treatment plant (WW) and
6 710 two industrial plants (I1: gas production, I2: surface finishing).
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10 712 Fig. 2: Evolution of daily rainfall (mm) and water discharge measured at the
11 713 lock-station "Fort de Scarpe" ($\text{m}^3 \text{s}^{-1}$) from April 2005 to August 2006 (source: "Voies Navigables de
12 714 France"). Vertical lines indicate the sampling dates (for details see Table 2).
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16 716 Fig. 3: Evolution in ^7Be activity of suspended sediment (Bq kg^{-1}) and monthly-cumulated rainfall (mm)
17 717 between successive sampling campaigns.
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21 719 Fig. 4: Plot of ^7Be inventories in the top (0–3 cm) sediment layer (mBq cm^{-2}) *versus* ^7Be activity in
22 720 suspended sediment (Bq kg^{-1}).
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25 722 Fig. 5: Monthly sediment Deposition/Resuspension ($=D/RS$) rates ($\text{g cm}^{-2} \text{mo}^{-1}$) calculated for (a) k13,
26 723 (b) k16 and (c) k36 sites.
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30 725 Fig. 6: From April 2005 to August 2006: Cumulated fluxes of deposition and resuspension and Net
31 726 accumulation fluxes of Pb ($\mu\text{g cm}^{-2}$) for the 3 sampling sites.
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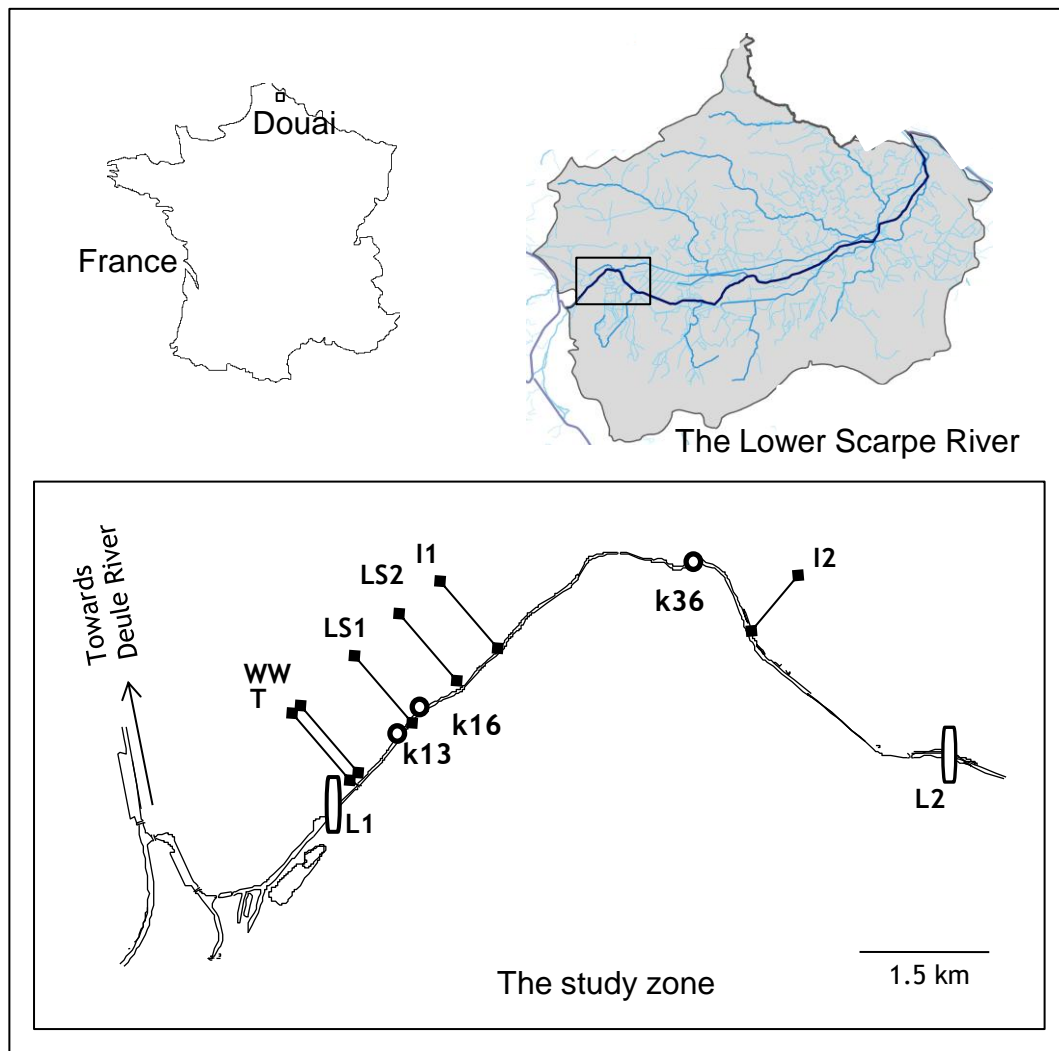


Fig. 1

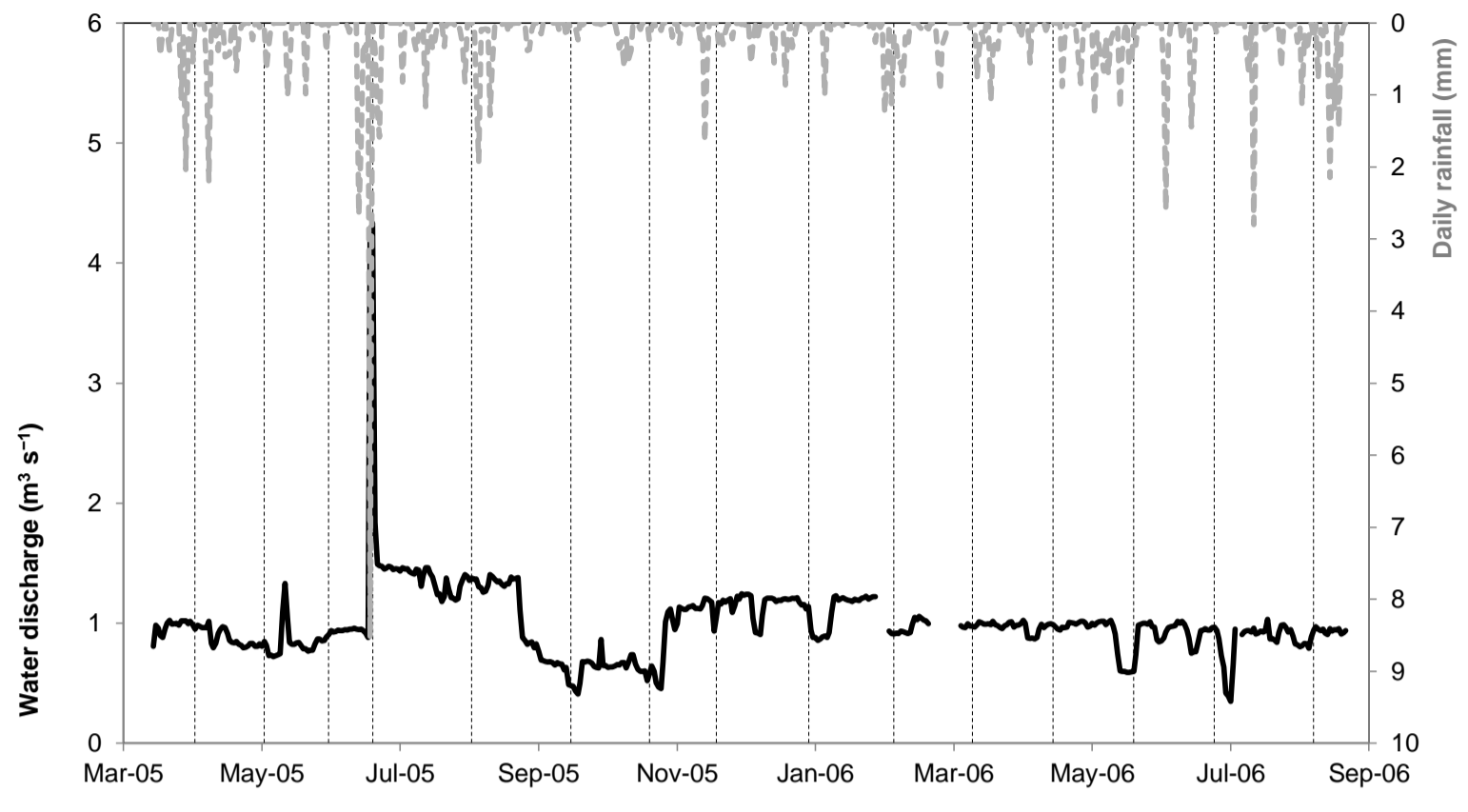


Fig. 2

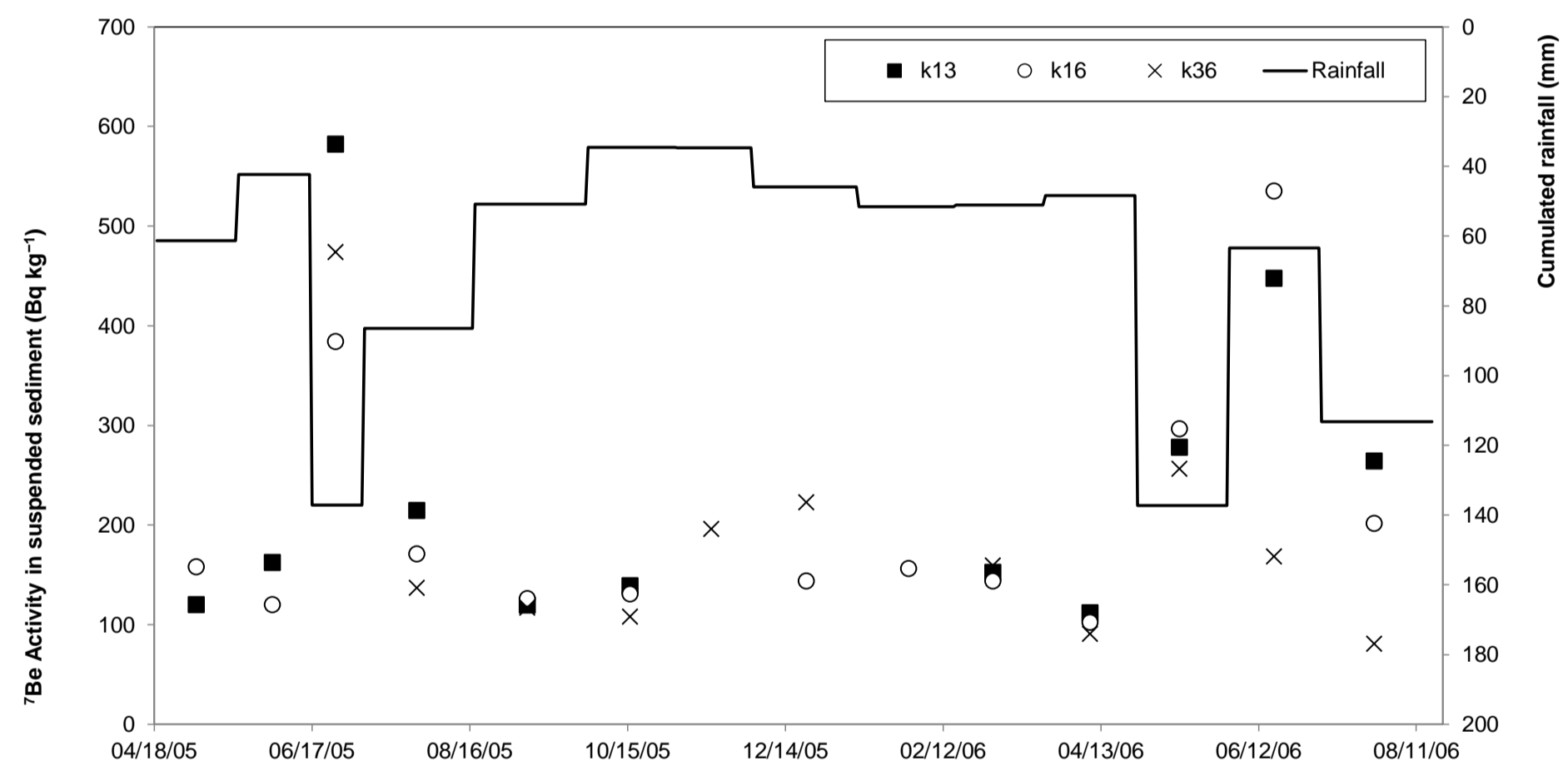


Fig. 3

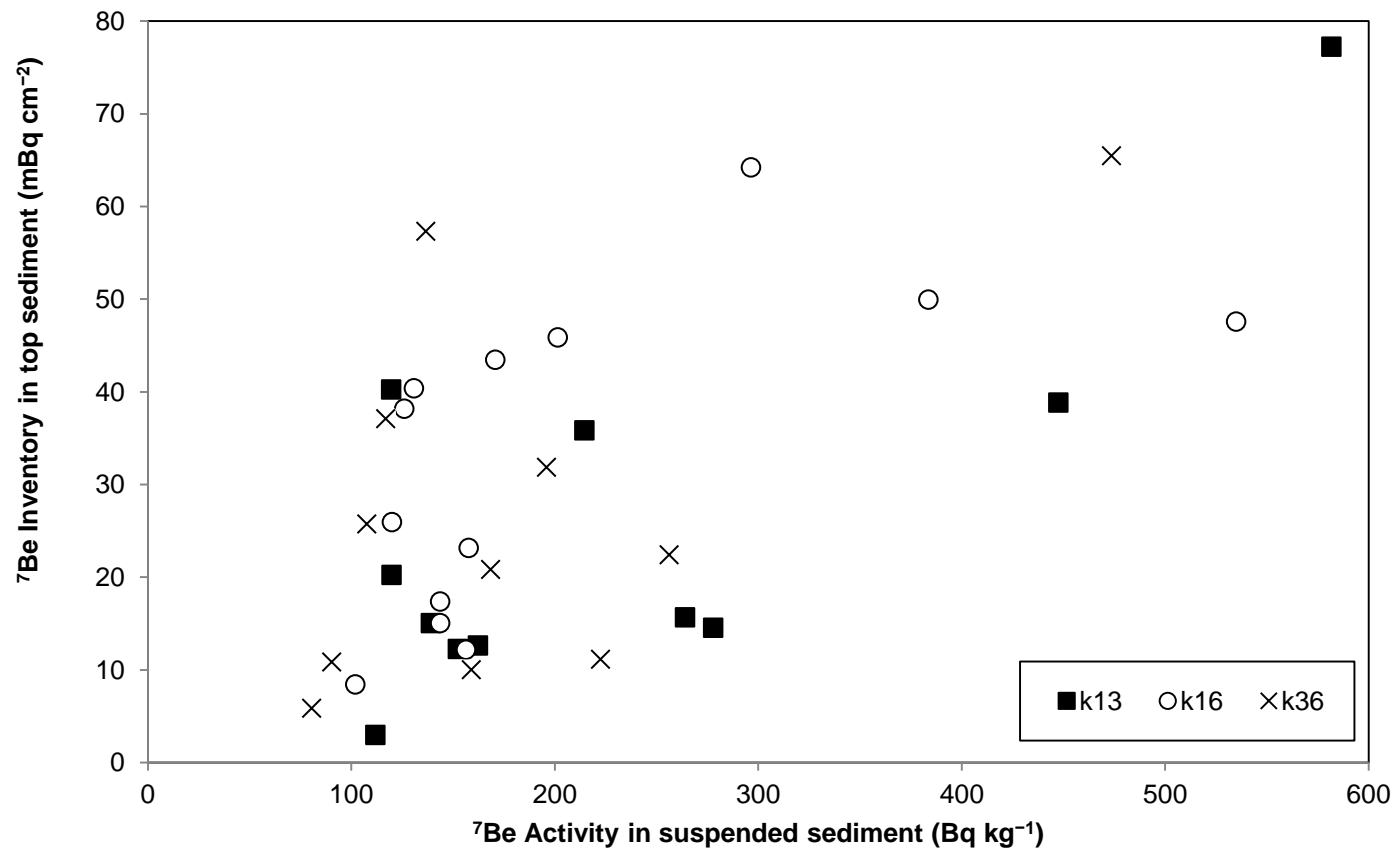


Fig. 4

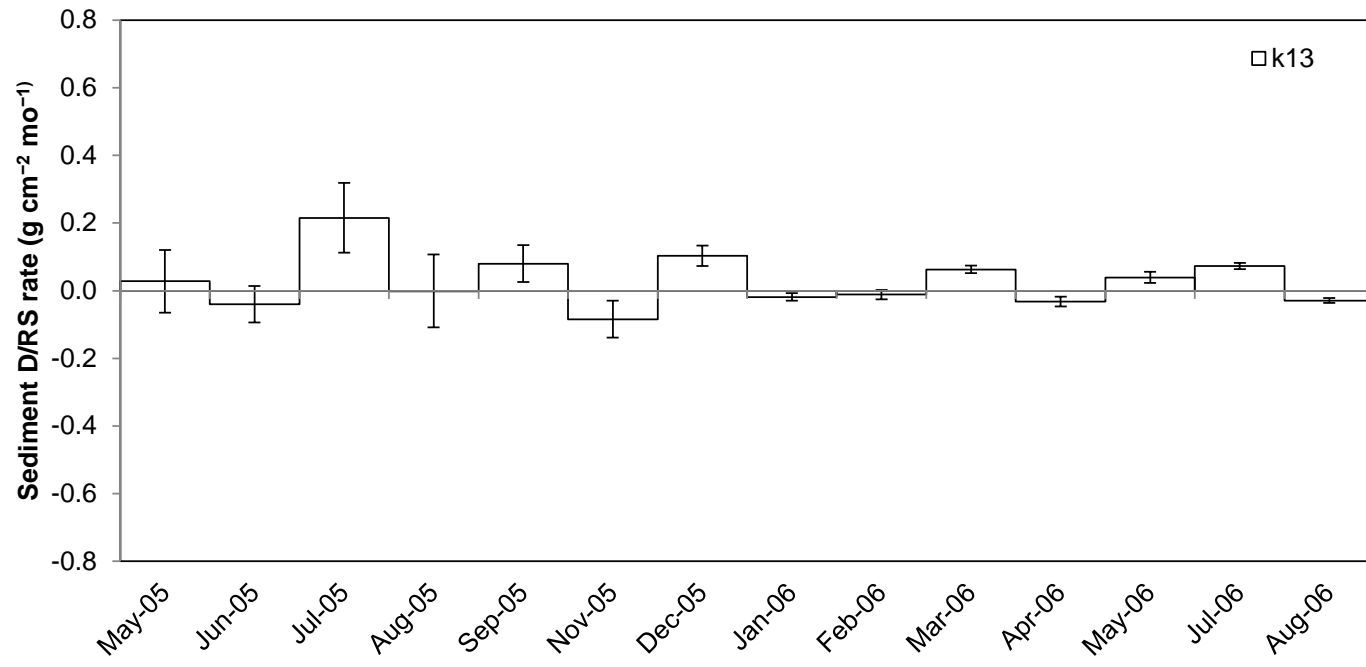


Fig. 5a

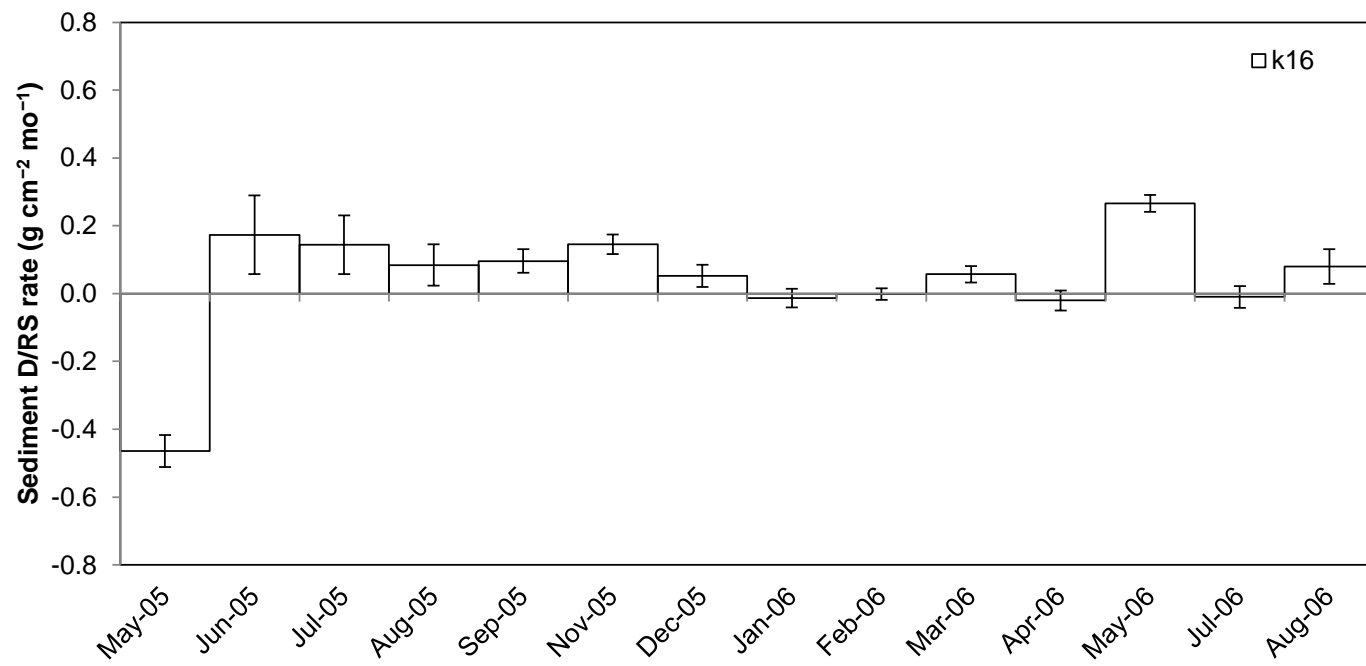


Fig. 5b

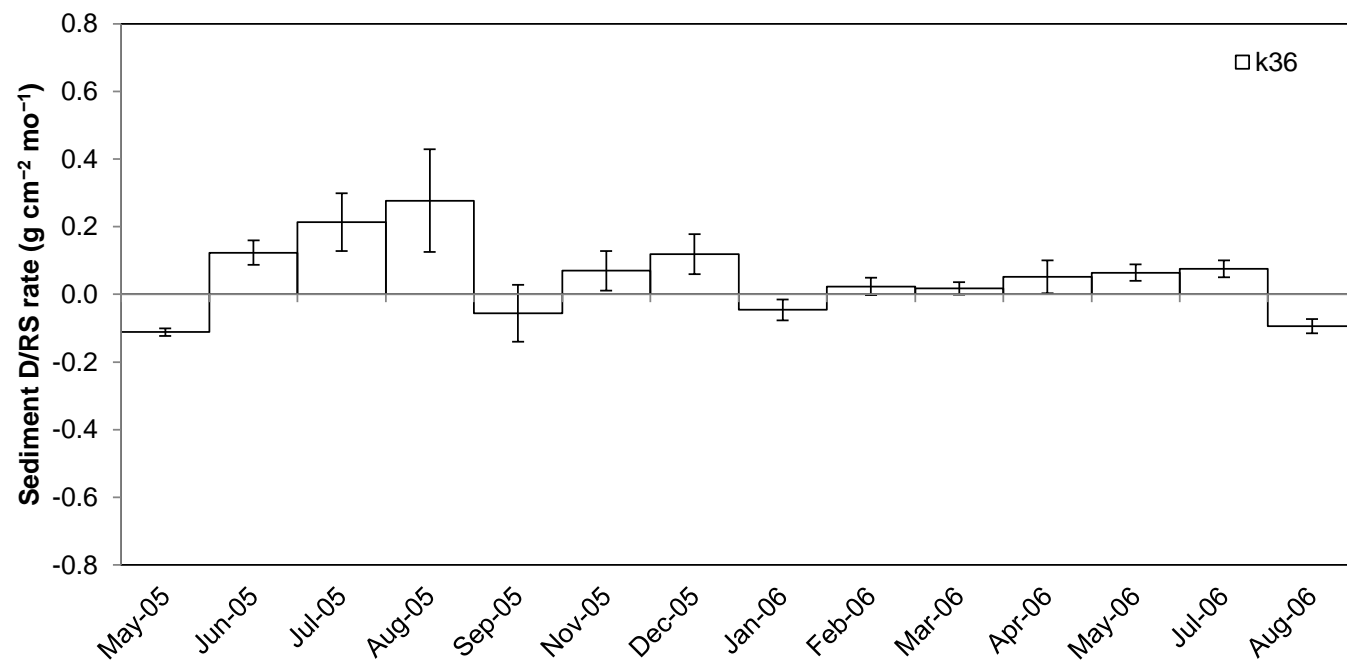


Fig. 5c

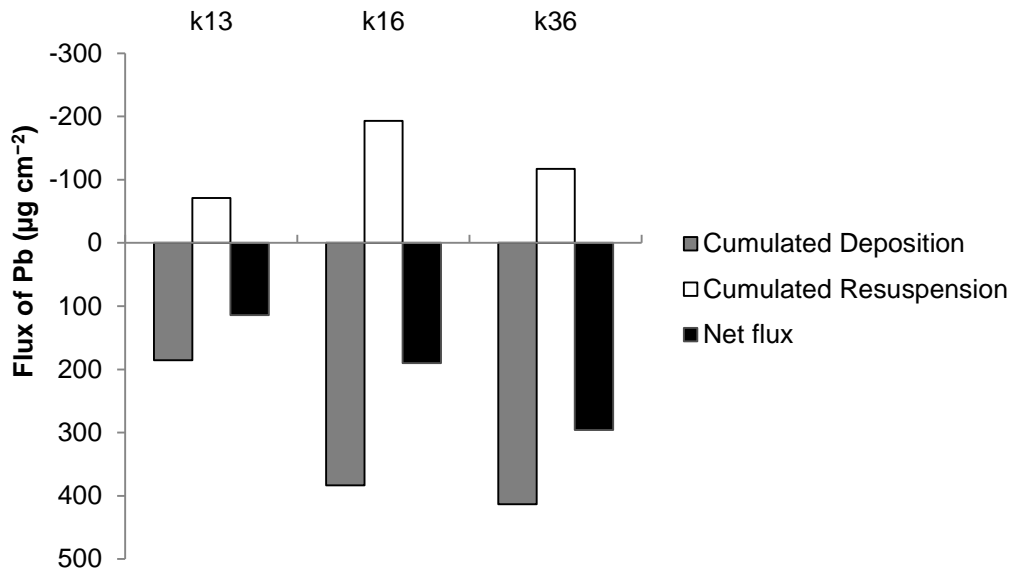


Fig. 6

Table 1

Site	k13	k16	k36
Sediment layer	^7Be Activity (Bq kg^{-1})		
0-1 cm	< 5	46 \pm 6	21 \pm 6
1-2 cm	16 \pm 5	31 \pm 6	6 \pm 5
2-3 cm	< 3	10 \pm 5	< 5
3-4 cm	< 3	5 \pm 3	< 4
4-5 cm	< 5	< 4	< 7

Table 2

Sampling n°		1			2			3			4			5			6			7			8		
Date		04/19/05			05/19/05			06/16/05			07/06/05			08/17/05			09/29/05			11/02/05			12/01/05		
Replicate					a	b	c	a	b	c	a	b	c	a	b	c	a	b	c	a	b	c	a	b	c
Site	Layer	⁷ Be inventory in sediment (mBq cm ⁻²)																							
k13	0-3 cm	34			30	7	24	12	14	12	49	30	152	30	32	46	39	49	33	18	1	26	30	28	20
	3-5 cm	7			23	30	7	4	2	3	15	6	34	15	11	31	6	4	5	1	1	n.d.	3	3	7
k16	0-3 cm	135			11	33	25	35	16	27	52	29	69	39	46	45	37	39	n.d.	43	38	39	40	36	22
	3-5 cm	16			2	10	4	21	9	11	17	6	29	9	16	23	9	17	n.d.	9	13	20	10	18	14
k36	0-3 cm	34			8	12	14	17	26	15	103	20	73	52	60	60	47	35	29	25	22	30	38	20	37
	3-5 cm	9			2	3	2	4	8	7	27	14	23	17	62	59	14	14	10	11	15	19	17	11	28

Sampling n°		9			10			11			12			13			14			15					
Date		01/10/06			02/16/06			03/22/06			04/26/06			05/31/06			07/05/06			08/17/06					
Replicate		a	b	c	a	b	c	a	b	c	a	b	c	a	b	c	a	b	c	a	b	c			
Site	Layer	⁷ Be inventory in sediment (mBq cm ⁻²)																							
k13	0-3 cm	10	10	16	8	2	1	14	15	7	3	4	1	5	17	22	41	40	36	19	17	12			
	3-5 cm	2	1	1	1	1	5	1	1	5	4	1	1	1	2	1	6	13	9	1	1	1			
k16	0-3 cm	22	16	14	14	8	14	11	20	14	5	9	11	62	59	72	23	51	68	40	42	55			
	3-5 cm	8	10	7	5	4	1	4	5	3	2	1	1	48	22	32	1	8	17	9	5	15			
k36	0-3 cm	17	15	2	13	8	10	9	9	12	8	17	7	23	30	14	21	35	6	8	8	1			
	3-5 cm	2	8	5	1	2	10	1	2	4	1	6	1	5	7	4	8	2	25	1	1	3			

Table 3

Sampling n°		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
Date		04/19/05	05/19/05	06/16/05	07/06/05	08/17/05	09/29/05	11/02/05	12/01/05	01/10/06	02/16/06	03/22/06	04/26/06	05/31/06	07/05/06	08/17/06
Duration (d)		t0	30	28	20	42	43	34	29	40	37	34	35	35	35	43
Site k13																
⁷ Be Inventory (mBq cm ⁻²)	Total	41	31	15.3	95	55	45	16	30	14	6	14.8	5	16	48	16
	±	-	11	0.4	40	9	4	6	2	2	2	0.9	1	5	2	2
	Residual	-	27	21	12	55	31	29	11	18	8	4	9	3	10	28
	New	-	3	-6	84	0	14	-13	20	-5	-2	11	-4	13	38	-11
⁷ Be Activity (Bq kg ⁻¹)	-	120	162	582	215	120	139	196	183	156	153	112	278	448	264	
Sediment D/RS rate (g cm ⁻² mo ⁻¹)	-	0.03	-0.04	0.22	0	0.08	-0.08	0.10	-0.02	-0.01	0.06	-0.03	0.04	0.07	-0.03	
±	-	0.09	0.05	0.10	0.1	0.05	0.05	0.03	0.01	0.01	0.01	0.01	0.02	0.01	0.01	
Site k16																
⁷ Be Inventory (mBq cm ⁻²)	Total	151	29	39	67	59	51	54	47	26	16	19	10	98	56	55
	±	-	7	8	16	5	3	2	5	3	2	3	2	8	15	6
	Residual	-	102	20	30	39	34	33	37	28	16	10	12	6	62	32
	New	-	-73	19	37	20	17	22	10	-2	0	9	-2	92	-6	23
⁷ Be Activity (Bq kg ⁻¹)	-	158	120	384	171	126	131	196	144	156	144	102	297	535	201	
Sediment D/RS rate (g cm ⁻² mo ⁻¹)	-	-0.46	0.17	0.14	0.08	0.10	0.15	0.05	-0.01	0	0.06	-0.02	0.27	-0.01	0.08	
±	-	0.05	0.12	0.09	0.06	0.03	0.03	0.03	0.03	0.02	0.02	0.03	0.02	0.03	0.05	
Site k36																
⁷ Be Inventory (mBq cm ⁻²)	Total	43	13	25	87	103	50	40	50	16	15	13	13	28	32	8
	±	-	2	4	24	15	6	4	9	4	3	2	4	5	2	1
	Residual	-	29	9	20	50	59	32	28	30	10	9	8	9	18	18
	New	-	-16	16	67	53	-9	8	23	-14	5	3	5	19	15	-11
⁷ Be Activity (Bq kg ⁻¹)	-	139	141	474	137	117	108	196	223	156	159	91	256	168	81	
Sediment D/RS rate (g cm ⁻² mo ⁻¹)	-	-0.11	0.12	0.21	0.3	-0.06	0.07	0.12	-0.05	0.02	0.02	0.05	0.06	0.08	-0.09	
±	-	0.01	0.04	0.09	0.2	0.08	0.06	0.06	0.03	0.03	0.02	0.05	0.02	0.02	0.02	

Table 4

Sampling n°		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
Date		04/19/05	05/19/05	06/16/05	07/06/05	08/17/05	09/29/05	11/02/05	12/01/05	01/10/06	02/16/06	03/22/06	04/26/06	05/31/06	07/05/06	08/17/06
Site k13																
Metal content (µg g⁻¹)	Cd	-	17	26	11	11	11	12	12	12	12	8	9	9	5	10
	Cu	-	222	216	186	196	157	164	171	171	171	155	150	154	139	140
	Pb	-	377	347	308	358	293	337	323	323	323	286	291	324	294	334
	Zn	-	1565	1451	1372	1360	1090	1084	1211	1211	1211	1037	1015	1228	970	1152
Site k16																
Metal content (µg g⁻¹)	Cd	-	321	244	319	157	71	64	130	90	110	72	37	73	104	24
	Cu	-	210	207	197	193	168	178	179	168	166	158	158	170	201	160
	Pb	-	387	362	393	358	315	339	339	337	311	298	288	348	335	337
	Zn	-	3332	2733	3785	2251	1667	1575	2245	1791	2156	1899	1384	2427	2679	1502
Site k36																
Metal content (µg g⁻¹)	Cd	-	188	188	316	271	207	217	211	149	188	119	132	146	158	139
	Cu	-	202	202	229	241	231	224	201	164	202	183	186	183	201	183
	Pb	-	379	379	417	433	409	419	376	330	379	323	325	388	364	390
	Zn	-	2650	2650	3992	3050	2588	2722	2565	1913	2650	2297	2175	2765	2503	2584

Table 5

Site	k13				k16				k36			
	Cd	Cu	Pb	Zn	Cd	Cu	Pb	Zn	Cd	Cu	Pb	Zn
Net flux	3	66	114	482	-2	94	190	964	188	164	296	2326
Cumulated Deposition ($\mu\text{g cm}^{-2}$)	6	102	186	735	150	199	383	2590	240	224	413	3098
Cumulated Resuspension	-3	-37	-71	-253	-152	-105	-193	-1626	-53	-60	-117	-772
Resuspension/Deposition (%)	50	36	38	34	101	53	50	63	22	27	28	25

1 **Supplementary material: Table and figure captions**

2

3 Table S1: Grain size distribution in bed sediment (%).

4 Table S2: Grain size distribution in suspended sediment (%).

5 Table S3: Activities in ^{137}Cs , $^{210}\text{Pb}_{\text{xs}}$ and ^{232}Th (Bq kg^{-1}), and $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$ activity ratio in suspended

6 sediment. Areal activities in ^7Be and ^{210}Pb (Bq m^{-2}) in rainwater samples monthly collected from

7 August 2005 to September 2006 (data available were from the French Institute of Radioprotection and

8 Nuclear Safety, OPERA-Air network). Fractions of recent sediment in suspended sediments (%).

9 Table S4: Characteristics of suspended sediment collected at the lift station "Vallée de Scarpe":

10 fraction of fine material ($<63 \mu\text{m}$, in %); ^7Be activities (Bq kg^{-1}); metal contents (Cu and Pb in $\mu\text{g g}^{-1}$,
11 Cd and Zn in mg g^{-1}).

12 Table S5: Monthly fluxes of contaminants associated with sediment dynamics ($\mu\text{g cm}^{-2} \text{mo}^{-1}$).

13 Figure S1: (a) Plot of ^{137}Cs activities versus fractions of fine material in bed and suspended sediments

14 (=SS); (b) Plot of ^{137}Cs activities versus ^{232}Th activities in bed and suspended sediments (=SS).

15 Figure S2: (a) Plot of ^7Be activities versus fractions of fine material in suspended sediments (=SS); (b)

16 Plot of ^7Be activities versus organic carbon contents in suspended sediments (=SS); (c) Plot of ^7Be

17 activities versus ^{232}Th activities in suspended sediments (=SS).

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23

Table S1

Sampling n°	Date	Sediment layer	Replicate	Grain size distribution, in %								
				k13			k16			k36		
				Clay	Silt	Sand	Clay	Silt	Sand	Clay	Silt	Sand
2	05/19/05	0-3 cm	a	2	60	38	2	62	36	3	73	24
			b	2	59	39	2	66	32	3	69	28
			c	2	57	42	2	60	38	3	74	23
		3-5 cm	a	2	52	46	2	56	42	3	75	22
			b	2	61	37	2	63	35	2	68	29
			c	2	61	37	2	66	32	3	70	27
3	06/16/05	0-3 cm	a	2	62	37	1	61	38	3	75	22
			b	2	60	39	1	58	41	2	79	19
			c	2	60	38	1	62	38	2	73	24
		3-5 cm	a	2	66	32	1	63	35	2	71	27
			b	2	61	37	2	61	38	3	74	23
			c	2	64	35	2	63	35	2	63	34
4	07/06/05	0-3 cm	a	2	51	48	2	54	45	2	71	26
			b	2	58	40	2	57	42	3	74	23
			c	-	-	-	2	53	45	2	73	25
		3-5 cm	a	1	51	48	1	40	59	3	73	24
			b	2	55	43	1	49	49	2	71	27
			c	1	49	49	2	55	43	2	74	24
12	04/26/06	0-3 cm	a	2	58	39	2	63	35	2	74	23
			b	2	61	37	2	57	41	2	81	16
			c	2	54	44	1	55	43	2	71	27
		3-5 cm	a	2	60	37	2	53	45	2	83	14
			b	3	69	28	2	61	37	2	78	20
			c	2	59	39	2	63	35	3	82	15

Table S2

Sampling n°	Date	Grain size distribution, in %								
		k13			k16			k36		
		Clays	Silts	Sands	Clays	Silts	Sands	Clays	Silts	Sands
3	06/16/05	2	80	18	2	80	18	-	-	-
4	07/06/05	2	72	26	2	68	30	3	82	15
5	08/17/05	2	71	27	-	-	-	-	-	-
6	09/29/05	2	79	19	2	77	21	2	87	11
7	11/02/05	2	78	20	2	72	27	3	87	10
8	12/01/05	-	-	-	4	87	10	3	86	11
9	01/10/06	-	-	-	2	71	27	2	80	18
10	02/16/06	-	-	-	2	65	33	-	-	-
12	4/26/2006	2	67	31	2	68	31	2	86	12
13	5/31/2006	3	62	35	3	60	38	3	59	37
14	7/5/2006	2	76	21	2	63	36	3	84	13

Table S3

Date			05/19/05	06/16/05	07/06/05	08/17/05	09/29/05	11/02/05	12/01/05	01/10/06	02/16/06	03/22/06	04/26/06	05/31/06	07/05/06	08/17/06
Sampling n°			2	3	4	5	6	7	8	9	10	11	12	13	14	15
Site k13	¹³⁷ Cs	Bq kg ⁻¹	8.3	8.1	6.8	8.90	7.5	8.5	-	-	-	7.3	7.5	7.7	6.9	5.8
	±		0.4	0.7	0.2	0.01	0.3	0.4				0.3	0.3	0.3	0.4	0.7
	²¹⁰ Pb _{xs}	Bq kg ⁻¹	67	56	137	81.4	64	83	-	-	-	55	58	70	94	66
	±		7	11	4	0.3	6	9				5	5	6	8	13
	²³² Th	Bq kg ⁻¹	26	23	26	27	26.3	27.9	-	-	-	18.9	23.3	23.8	21.9	20
	±		2	1	2	0.7	0.4	0.7				0.4	0.4	0.4	0.6	1
	⁷ Be/ ²¹⁰ Pb _{xs}		1.8	2.9	4.3	2.63	1.9	1.7	-	-	-	2.8	1.9	4.0	4.7	4
±		0.4	0.9	0.2	0.01	0.2	0.2				0.3	0.2	0.4	0.5	1	
Site k16	¹³⁷ Cs	Bq kg ⁻¹	6.5	6.9	5.3	7.7	6.7	7.4	-	8.1	6.1	5.4	7.0	7.5	6.5	5.3
	±		0.3	0.3	0.1	0.3	0.3	0.5		0.3	0.6	0.3	0.3	0.4	0.2	0.6
	²¹⁰ Pb _{xs}	Bq kg ⁻¹	58	53	86	69	53	72	-	57	41	52	57	83	107	79
	±		4	5	1	6	7	10		6	11	6	5	9	5	10
	²³² Th	Bq kg ⁻¹	26	24.7	26	27	24.6	29.0	-	26.2	20.9	20.2	25.2	25.7	24.6	21.5
	±		2	0.5	2	0.5	0.6	0.8		0.5	0.9	0.5	0.4	0.7	0.3	0.8
	⁷ Be/ ²¹⁰ Pb _{xs}		2.7	2.2	4.5	2.5	2.4	1.8	-	2.5	3.8	2.8	1.8	3.6	5.0	2.6
±		0.3	0.3	0.1	0.3	0.4	0.3		0.3	1.2	0.4	0.2	0.5	0.3	0.5	
Site k36	¹³⁷ Cs	Bq kg ⁻¹	-	-	7.7	9.5	9.3	9.7	9.1	9.9	-	6.8	6.0	8.5	8.8	7.8
	±				0.1	0.3	0.3	0.4	0.7	0.8		0.4	0.3	0.4	0.4	0.7
	²¹⁰ Pb _{xs}	Bq kg ⁻¹	-	-	111	71	70	80	67	61	-	57	54	68	68	62
	±				1	6	6	7	12	13		7	6	8	8	11
	²³² Th	Bq kg ⁻¹	-	-	29	30.6	28.8	29.7	26	24	-	21.7	20.0	26.5	27.7	23.2
	±				1	0.5	0.5	0.6	1	1		0.6	0.5	0.6	0.6	0.9
	⁷ Be/ ²¹⁰ Pb _{xs}		-	-	4.3	1.9	1.7	1.3	2.9	3.6	-	2.8	1.7	3.8	2.5	1.3
±				0.1	0.2	0.2	0.2	0.6	0.9		0.4	0.3	0.5	0.4	0.4	
Rainwater	Date		-	-	-	-	-	10/31/05	11/30/05	12/31/05	01/31/06	03/31/06	04/30/06	05/31/06	06/30/06	08/31/06
	⁷ Be							37	72	52	32	1.9	59	132	10.1	201
	±							3	6	4	2	0.3	4	9	0.8	12
	²¹⁰ Pb	Bq m ⁻²						3.6	4.8	3.8	4.4	<0,7	5.8	10.6	1.8	20.5
±							0.5	0.5	0.5	0.6		0.5	0.8	0.2	1.6	
k13	Recent sediment	%					16						19	32	86	41
k16						18		18	53		18	29	90	26		
k36						13	20	27				16	30	45	13	

Table S4

Suspended sediment from the lift station "LS1"												
Sampling n°	Date	< 63 μm %	^7Be Bq kg ⁻¹	\pm	Cu	\pm	Pb $\mu\text{g g}^{-1}$	\pm	Cd	\pm	Zn mg g ⁻¹	\pm
5	08/17/05	87	815	16	374	6	582	14	3.64	0.06	17.3	0.3
6	09/29/05	86	583	18	359	4	539	12	2.02	0.03	16.3	0.2
7	11/02/05	-	344	32	181	2	268	7	1.16	0.02	13.2	0.2
8	12/01/05	82	303	6	298	11	511	20	1.73	0.08	14.7	0.6
13	05/31/06	78	250	6	452	11	547	16	0.91	0.03	16.0	0.4
14	07/05/06	79	918	31	425	7	575	6	0.87	0.01	15.0	0.2

Table S5

Sampling n°	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	
Date	04/19/05	05/19/05	06/16/05	07/06/05	08/17/05	09/29/05	11/02/05	12/01/05	01/10/06	02/16/06	03/22/06	04/26/06	05/31/06	07/05/06	08/17/06	
Flux of contaminant deposition/resuspension ($\mu\text{g cm}^{-2} \text{mo}^{-1}$)	Site k13															
	Cd	-	0.5	-1	2	0	0.9	-1	1	-0.2	-0.1	0.5	-0.3	0.3	0.4	-0.3
	Cu	-	6	-9	40	0	13	-14	18	-3	-2	10	-5	6	10	-4
	Pb	-	10	-14	66	0	23	-28	33	-6	-4	18	-9	13	21	-10
	Zn	-	43	-58	296	-1	87	-91	125	-22	-14	65	-32	48	71	-34
	Site k16															
	Cd	-	-149	42	46	13	7	9	7	-1	0	4	-1	20	-1	2
	Cu	-	-97	36	28	16	16	26	9	-2	0	9	-3	45	-2	13
	Pb	-	-179	63	57	30	30	49	18	-4	0	17	-6	93	-3	27
	Zn	-	-1545	473	547	189	160	229	117	-23	-3	108	-28	647	-26	120
	Site k36															
	Cd	-	-21	23	67	75	-12	15	25	-7	4	2	7	9	12	-13
	Cu	-	-23	25	49	67	-13	16	24	-8	5	3	10	12	15	-17
Pb	-	-42	47	89	120	-23	29	45	-15	9	6	17	25	27	-37	
Zn	-	-296	326	851	845	-145	189	306	-88	62	41	113	177	188	-243	

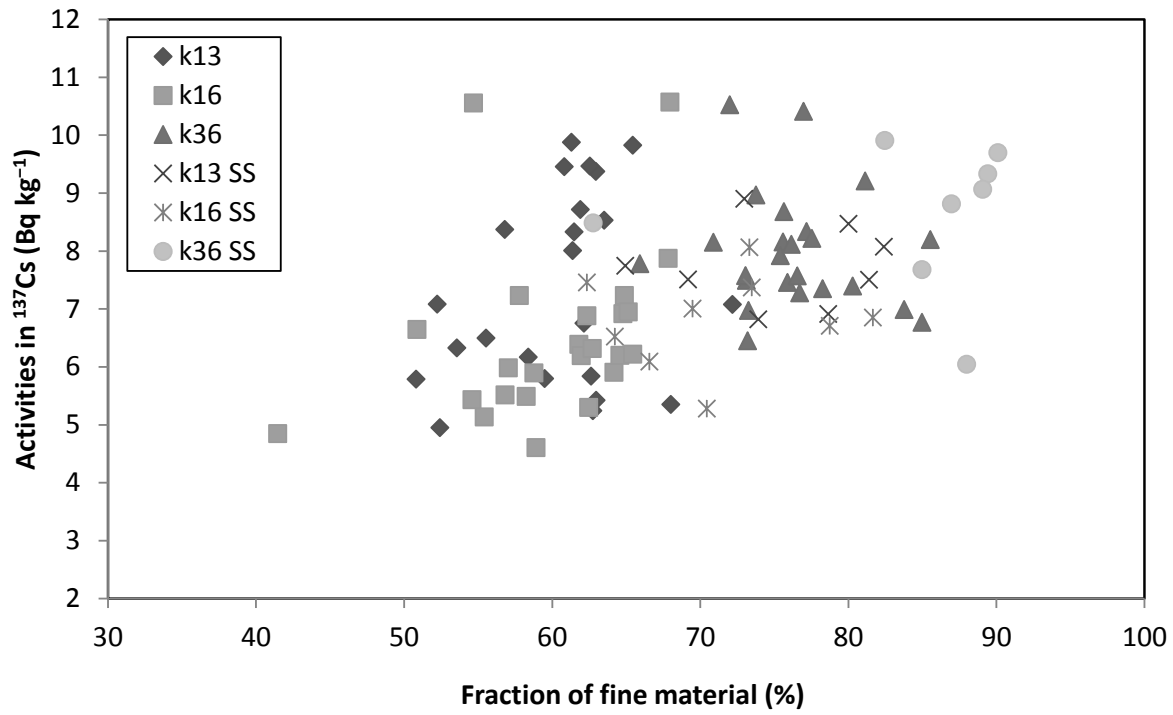


Fig. S1a

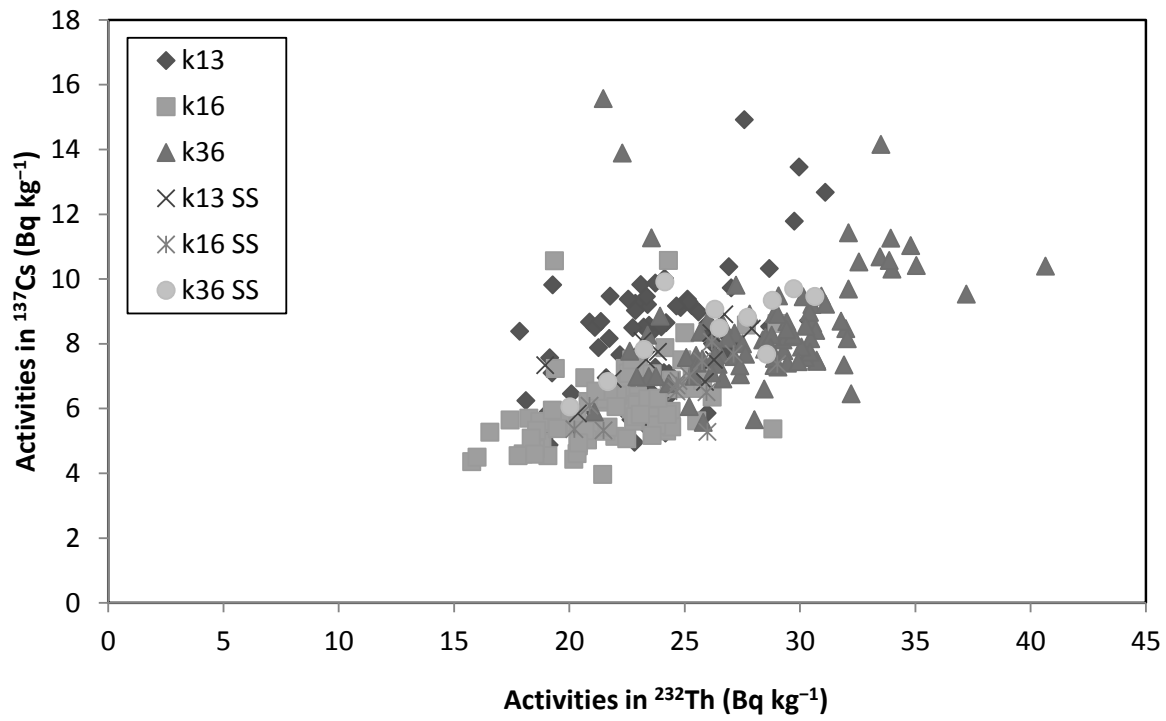


Fig. S1b

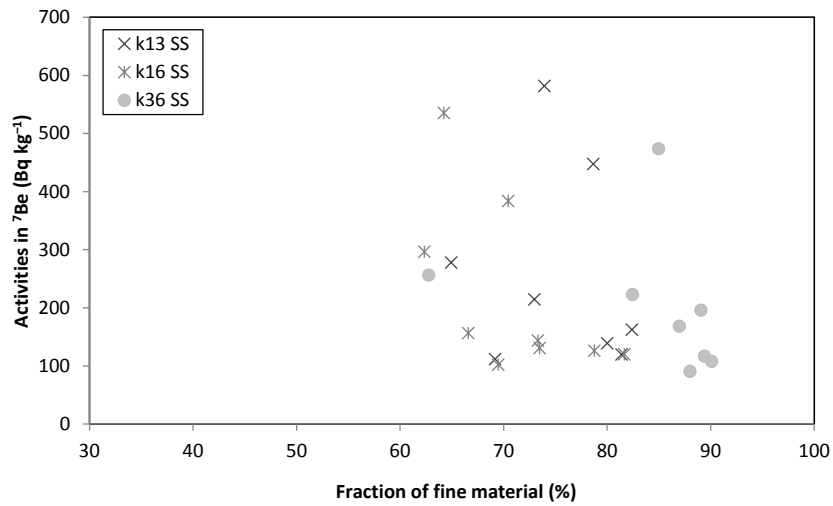


Fig. S2a

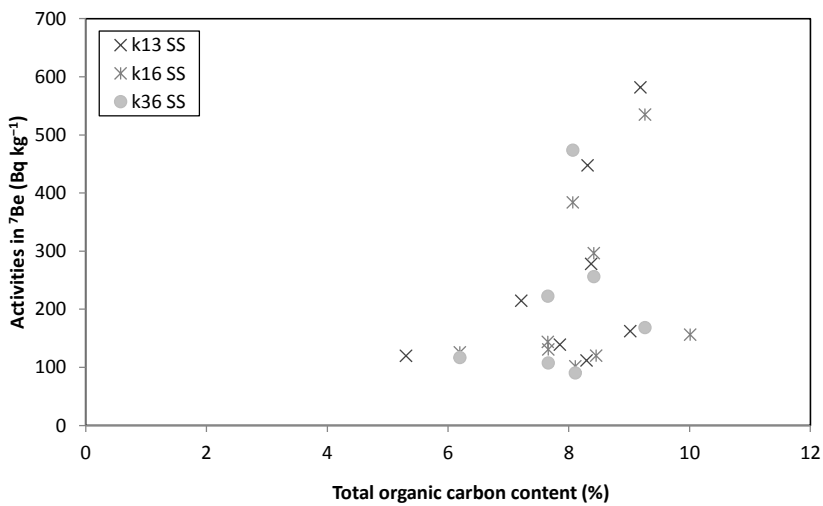


Fig. S2b

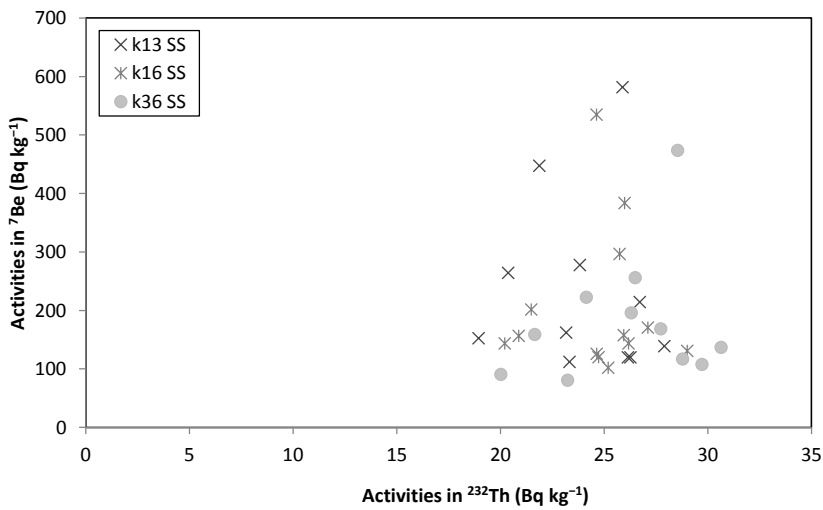


Fig. S2c