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

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Abstract

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

Materials and methods: Activities in ^{7}Be were measured in bed and suspended sediments collected each month over an entire hydrological year. Suspended sediments were analysed for metal contents (Pb, Zn, Cd, Cu). The determination of short-term sediment dynamics is based on the variation of Total ^{7}Be inventory between two successive samplings. Total inventory of ^{7}Be consists of two components, i.e., the Residual inventory and the New inventory.

Results and discussion: Inventories of ^{7}Be in sediment varied from 1 to 135 mBq cm^{-2}. Observed spatial and temporal variations reflected the dynamical behaviour of sediment in the studied channel section. The succession of sediment deposition and resuspension periods was demonstrated during the study, with maximum deposition and erosion rates of 0.3±0.2 and −0.46±0.05 g cm^{-2} mo^{-1}, respectively. A sediment resuspension rate of 0.06 g cm^{-2} mo^{-1} supplied to the water column a flux of 12, 13, 23 and 145 µg cm^{-2} mo^{-1} of Cd, Cu, Pb and Zn, respectively. During this 15-months study, 28 to 50 % of Pb that deposited in the riverbed sediment was resuspended, contributing thereby to the (short term) degradation of water quality.

Conclusions: The present work provides a useful tool for examining the role of sediment as a sink or source of contamination for the water column. It produces knowledge of how sediment processes affect the fate and the transport of contaminants and the implications for future and downstream outflows.

Keywords Beryllium-7 • Sediment deposition/resuspension rate • Metallic contaminant • Water-sediment interface

1 Introduction

Contamination of river sediment reaches problematic levels in industrialized and urbanized areas. Accumulation of contaminants in soils and their subsequent supply to rivers constitute a major environmental and public health problem in industrialized countries (Gateuille et al. 2014). Anthropogenic emissions largely contributed to the supply of metallic contaminants to river systems through point and diffuse sources (Ayrault et al. 2012). In river systems, metallic contaminants are preferentially associated with the finest fractions of particles (<63 µm) that are also the most easily transported (Salomons and Förstner 1984; Owens et al. 2005), and sediment constitutes therefore a significant pollution reservoir (Gibbs 1973; Lee et al. 2003; Westrich and Förstner 2007). A better understanding of riverine dynamics is therefore needed to improve our knowledge on the sources of the current contamination in rivers and to meet the targets required by the law [European Water Framework Directive (Roche et al. 2005)].

Physical disturbances induced by natural and/or anthropogenic events at the water-sediment interface can result in the resuspension of contaminated sediments and in the remobilization of metals from sediment in the water column (Haag et al. 2001; Zoumis et al. 2001; Prygiel et al. 2015; Superville et al. 2015). The fate of metallic contaminants is therefore closely linked with the evolution of sediment dynamics (Calmano et al. 1993; Horowitz 2000; Walling et al. 2003). Storage of sediment and associated contaminants can occur over the short or the long term depending on temporal variations of the river flow and its transport capacity.

Several fallout radionuclides were shown to be particle-reactive and to provide appropriate tools to quantify sediment dynamics in rivers. In particular, naturally occurring and short-lived Beryllium-7 (^{7}Be, 53 d half-life) was extensively used in different types of aquatic systems (Krishnaswami et al. 1980; Mabit et al. 2008, Walling 2013, Taylor et al. 2013), as in marine and coastal environments (Palinkas et al. 2005), lakes (Dominik et al. 1989) and rivers (Dominik et al. 1987; Fitzgerald et al. 2001; Smith et al. 2014). This radionuclide is produced in the atmosphere by the cosmic ray spallation of oxygen and nitrogen atoms, and is rapidly bound to aerosols and supplied to the Earth's surface by wet deposition (Lal et al. 1958; Olsen et al. 1985; Bourcier et al. 2011). Once in terrestrial and aquatic environments, ^{7}Be quickly sorbs onto particles, as shown by its high distribution coefficient [K_d values comprised between 10^4–10^5 (Olsen et al. 1986; Hawley et al. 1986; Jewda et al. 2008; Taylor et al. 2012)], and ^{7}Be-labelled sediment particles are mixed and transported in rivers (Baskaran and

Santschi 1993; Sommerfield et al. 1999). Beryllium-7 provides therefore a relevant tracer for quantifying sediment deposition and resuspension. Tracing is improved when combining ^{7}Be with Cesium-137 (^{137}Cs, 30 y half-life) and Lead-210 (^{210}Pb, 22 y half-life) measurements, i.e., two longer-lived fallout and particle-bound radionuclides (Du et al. 2010; Evrard et al. 2010; Huang et al. 2011).

Sediment residence times were quantified using ^{7}Be radionuclide mass balance models at the catchment scale (Dominik et al. 1987; Bonté et al. 2000; Jweda et al. 2008; Evrard et al. 2010). Furthermore, shorter-term sediment dynamics were quantified through the variation of ^{7}Be inventory in surface bed sediment (Dibb and Rice 1989; Canuel et al. 1990; Giffin and Reide Corbett 2003). However, to our knowledge, these techniques have been rarely applied to other particle-bound contaminants, such as metals (Feng et al. 1999; Huang et al. 2011) and PCBs (Fitzgerald et al. 2001). This situation can probably be attributed to the difficulty of conducting frequent sediment sampling in rivers and to analyse the ^{7}Be in collected material rapidly after sampling (before ^{7}Be has decayed too much).

In this context, the present paper investigates the potential of using ^{7}Be as a tracer of sediment and particle-bound contaminant dynamics in a heavily polluted river section. We therefore propose to quantify the resuspension of particle-bound metals in a river catchment strongly impacted by industrialization and urbanization. To this end, we selected a catchment of Northern France (Lower Scarpe River) where the metal pollution levels were high and representative of the ones found in similar regions of Northwestern Europe, and where the feasibility of radionuclide tracing was demonstrated (Zebracki et al. 2007; Zebracki 2008).

2 Study site

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

With a 37-km length, the Lower Scarpe River is one of the six main tributaries of the Scheldt River, which flows across France, Belgium and the Netherlands before reaching the North Sea. The river has been channelized during the 17th century. Intense coal mining exploitation that took place in the catchment between the 18th and the 20th centuries led to the subsidence of the land, so that the river level is higher than the nearby land.

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

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

3 Materials and methods

3.1 Field sampling and laboratory analyses

3.1.1 Sampling sites

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

3.1.2 Sampling procedures

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

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

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

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

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

3.1.3 Sample analyses

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

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

Specific activities were expressed in Bq kg^{-1} dry weight and were systematically decay-corrected to the date of sampling. Uncertainties on activities were expressed at ±2 σ.

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

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

Contents in metals were quantified in suspended sediments after *aqua regia* micro-wave (CEM µ-waves MARS 5) assisted dissolution (0.5 g of air-dried and crushed sediment; HNO_3:HCl 1:3). Solutions were analysed using Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES; Jobin-Yvon 138 ULTRACE). A certified reference material (TH2 Toronto Harbour Sediment for Trace Elements, National Water Research Institute, Canada) was used for quality control. Analysis was systematically conducted on 3 subsamples. Uncertainties on metal contents were derived from the standard analytical deviations and were expressed at ±1 σ.

3.2. Short-term sediment dynamics

3.2.1 Inventory of ^{7}Be in bed sediment

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

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

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

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

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

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

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

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

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

When calculating ^{7}Be activities, the ^{7}Be decay correction did not depend on the potential variability of sediment discharge throughout time in the river. In this context, the lack of information regarding the temporal variability of sediment discharge may lead to an underestimation of ^{7}Be activities, and thus Total ^{7}Be inventories (for instance, when the majority of sediment discharge occurs early in the period of time between the sampling events).

3.2.2 Short term sediment deposition/resuspension rate

Rate of sediment deposition/resuspension ($R_{SEDIMENT}$, in g cm^{-2}) is determined between two successive samplings by dividing the New ^{7}Be inventory in sediment [$I(^{7}Be)_{NEW}$, in mBq cm^{-2}] by the mean ^{7}Be activity [$A(^{7}Be)_{MEAN}$, in Bq kg^{-1}] of newly deposited or resuspended sediment (Canuel et al. 1990):

(3) $$R_{SEDIMENT} = {I(^{7}Be)_{NEW}}/{A(^{7}Be)_{MEAN}} \times 10^{-3}.$$

In the present study, the mean ^{7}Be activity is determined from suspended sediment (Fitzgerald et al. 2001; Zebracki et al. 2007; Zebracki 2008) and not in surface sediment investigated by other authors (Canuel et al. 1990; Giffin and Reide Corbett 2003), assuming that characteristics of sediment depositing on the bed or being resuspended are representative of the characteristics of suspended sediment.

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

(4) $$R_{SEDIMENT} = {I(^{7}Be)_{NEW}}/{A(^{7}Be)_{MEAN}} \times {30}/{N_{DAYS}}.$$

Positive values of sediment rate indicate deposition whereas negative values correspond to resuspension. For a given period, Net sediment rate corresponds to the sum of the monthly short-term rates of sediment deposition and resuspension.

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

Flux of contaminants associated with sediment deposition/resuspension (R_{METAL}, expressed in µg cm^{-2} mo^{-1}) is calculated by multiplying short-term sediment rate ($R_{SEDIMENT}$, in g cm^{-2} mo^{-1}) by metal content (C_{METAL}, in µg g^{-1}) in suspended sediment, assuming that characteristics of sediment

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

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

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

3.2.4 Fraction of recent ^{7}Be-labelled sediment in suspended sediment

To distinguish sediment recently labelled with ^{7}Be from older (^{7}Be-depleted) sediment, an indicator of the fraction of the suspended sediment that has been recently eroded from soils is given by the ^{7}Be/^{210}Pb$_{xs}$ activity ratio (Matisoff et al. 2005; Evrard et al. 2010). The fraction of recent ^{7}Be-labelled sediment (i.e., recent sediment, expressed in %) is given by dividing the ^{7}Be/^{210}Pb$_{xs}$ activity ratio in suspended sediment [(^{7}Be/^{210}Pb$_{xs}$)$_{\text{Suspended sed.}}$] by the ^{7}Be/^{210}Pb$_{xs}$ activity ratio in rainfall [(^{7}Be/^{210}Pb$_{xs}$)$_{\text{Source}}$]:

(6) $$Recent\ sediment, \% = 100 \times \left(\frac{^{7}Be}{^{210}Pb_{xs}}\right)_{\text{Suspended sed.}} \Bigg/ \left(\frac{^{7}Be}{^{210}Pb_{xs}}\right)_{\text{Source}}.$$

The sediment source term is defined by sediment particles that have a ^{7}Be/^{210}Pb activity ratio equal to that of wet fallout. The dry deposition of ^{7}Be and the transfer time required for ^{7}Be-labelled particles to be supplied from catchment soils to the river through erosion processes were neglected, as both phenomena were supposed to be of minor importance (Matisoff et al. 2005; Conaway et al. 2013; Pham et al. 2013). Fallout ^{7}Be and ^{210}Pb were monthly measured in the framework of the Permanent observatory of air radioactivity (OPERA-Air sampling network, operated by the French Institute of Radioprotection and Nuclear Safety). The closest station to the study area is situated at 180 km (Charleville-Mezieres). The ^{7}Be/^{210}Pb$_{xs}$ source term is calculated using areal activities in ^{7}Be and ^{210}Pb that were derived from activities in rainwater samples (Bq L^{-1}) collected each month and available rainfall data (L m^{-2}).

4 Results and discussion

4.1 Hydrological conditions

Between April 2005 and August 2006, the mean water discharge measured at the upstream lock gauging station was 1.0±0.2 m^{3} s^{-1}. One extreme rainfall event occurred on July 7, 2005 (137 mm of

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

4.2 Grain size of sediment

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

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

4.3 Rainwater ^{7}Be and ^{210}Pb activities

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

4.4 Beryllium-7 in bed sediment

4.4.1 Depth occurrence of ^{7}Be in sediment

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

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

4.4.2 Total ^{7}Be inventory of bed sediment

In sediment layers collected from April 2005 to August 2006 (n=258), ^{7}Be inventories ranged from 1 to 135 mBq cm^{-2} (Table 2). Total ^{7}Be inventories in sediment ranged from 5 to 151 mBq cm^{-2} (Table 3). Mean coefficients of variation between sediment replicates ranged from 28 % (k16) to 37 % (k13) and exceeded the ones obtained for grain size distribution (< 7 %). Variations in ^{7}Be inventories between sediment replicates are then rather influenced by the spatial heterogeneity in the deposition of ^{7}Be-labelled particles on the riverbed, as sediment activity in ^{7}Be depends on both its age and its origin. These mean uncertainties were consistent with the values previously reported from other aquatic environments [25−33 % in coastal systems (Canuel et al. 1990, Giffin and Reide Corbett 2003)].

By comparing ^{7}Be inventories in top and bottom sediment layers (0−3 cm vs. 3−5 cm), 4 types of sediment deposition patterns were found. The highest ^{7}Be activities were generally found in the top sediment layer (91 % of observations) indicating the recent deposition of ^{7}Be-labelled sediment. In some cases, both top and bottom sediments displayed low ^{7}Be inventories (< 5 % of cases), indicating either the resuspension of previously deposited sediment or the absence of sediment deposition. Rarely (<3 % of cases), ^{7}Be inventory remained relatively constant with depth, indicating the occurrence of local physical disturbances, such as bioturbation that contributed to homogeneize the ^{7}Be records in depth (Santschi et al. 1990, Ciutat et al. 2005).

Although the potential loss of ^{7}Be through remobilisation under changing environmental conditions (i.e., variations in pH, redox potential, age of sediment) during sediment deposition and resuspension stages is unlikely to have occurred in our study area given the rapid cycling of material through the system, this process, if confirmed, could lead to underestimations of ^{7}Be inventories in sediment (Taylor et al. 2012, Taylor et al. 2013).

4.5 Beryllium-7 in suspended sediment

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

During the study period, rainfall variations had an impact on ^{7}Be activities measured in suspended sediment (Fig. 3): in July 2005 and May 2006, increase in ^{7}Be activities (up to 582 Bq kg^{-1}) was associated with increasing monthly cumulated rainfall (up to 137 mm). In contrast, low ^{7}Be activities (< 223 Bq kg^{-1}) and low variations recorded from September 2005 to April 2006 were associated with low cumulated rainfall (< 52 mm).

Direct deposition of fallout radionuclides in river water is generally considered to be of minor importance (Jweda et al. 2008).

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

Activities in ^{137}Cs measured in suspended sediment (5.3−9.9 Bq kg^{-1}; Table S3) indicate that overall sediment supply in the Scarpe River derives from the erosion of surface soils (Ben Slimane et al. 2013; Jagercikova et al. 2015). The preferential erosion of top vs. subsurface soils is consistent with the low relief of the catchment and the overall occurrence of low-intensity continuous rainfall. In contrast, in mountainous river catchments affected by heavy storms, decreasing in ^{7}Be and ^{137}Cs activities of suspended sediment is observed due to the erosion of particles originating from deeper soils that are depleted in fallout radionuclides (Olley et al. 1993; Evrard et al. 2011; Conaway et al. 2013).

The differences in ^{7}Be activities observed between sites in July 2005, July 2006 and August 2006 suggest the occurrence of mixing processes in the river between “recent” (^{7}Be-enriched) and “older” (^{7}Be-depleted) sediment particles (Olsen et al. 1985; Wallbrink and Murray 1996; Matisoff et al. 2005). In July and August 2006, decreasing ^{7}Be activities in downstream direction was related to decreasing fractions of recent sediment (Table S3). In November 2005 and April 2006, ^{7}Be activities displayed low variations between sites (<23 %), suggesting a relative homogeneity of suspended material, which was confirmed by low variations in estimated fractions of recent sediment (13−19 %).

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

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

4.6 Sediment deposition/resuspension rate

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

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

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

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

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

4.7 Contaminant fluxes at water-sediment interface

4.7.1 Metal contents in suspended sediment

Suspended sediment displayed high contents of Cd, Cu, Pb and Zn (Table 4). These contents remain in the range of sediment contamination levels measured in other European rivers that were strongly impacted by mining exploitation, industrialization and urbanization (Morillo et al. 2002; Lacal et al. 2003; Walling et al. 2003; Audry et al. 2004; Meybeck et al. 2007; Le Cloarec et al. 2011; Prygiel et al. 2015). Variations in metal contents did not reflect variations in grain size (Table S2, Electronic Supplementary Material). Increasing metal content of suspended sediment in downstream direction reflects the supply of contaminants from various point and diffuse sources of contamination, including the lift station situated upstream of k16 (Fig. 1), at which suspended sediments were shown to display high levels of metallic contamination, especially in Cd (Table S4, Electronic Supplementary Material).

4.7.2 Flux of contaminants at water-sediment interface

The monthly particulate fluxes of metal deposition and resuspension were calculated (Table S5, Electronic Supplementary Material). Over the 15-months monitoring, the Net fluxes of contaminants were positive for all sites (Table 5), demonstrating the role of sediment as a sink of contaminants, except for Cd at k16 site, due to the combination of a high resuspension rate and a high Cd content. Maximum Net fluxes of contaminants were found at k36 site, in association with locally high sediment accumulation rate.

Although sediment of the Lower Scarpe River is shown to act as a sink for contaminants over years and decades, resuspension periods contribute to the supply of contaminants back into the water. In September 2005, sediment resuspension of −0.06 g cm^{-2} mo^{-1} supplied fluxes to the water column of 12, 13, 23 and 145 µg cm^{-2} mo^{-1} for Cd, Cu, Pb and Zn, respectively (Table S5, Electronic Supplementary Material). As a comparison, a sediment resuspension of 0.08 g cm^{-2} mo^{-1} in the Fox River corresponded to a $\sum PCB$ flux of 0.24 µg cm^{-2} mo^{-1} (Fitzgerald et al. 2001). During the 15−months monitoring, 28 to 50 % of Pb that deposited at surface bed sediment was resuspended

(Fig. 6). Even in such river systems (non-navigated, with low and regulated flow), minor resuspension periods may significantly contribute to the temporal degradation of the water quality.

Futhermore, it is well known that resuspension of anoxic sediment in oxic water columns affects the mobility of labile contaminants resulting in metal remobilization and enhancing their biodisponibility (Argese et al. 1997; Zoumis et al. 2001; Eggleton et al. 2004; Westrich and Förstner 2007; Hwang et al. 2011). Increasing solubility of particle-bound Zn, Cd, Cu and Pb is therefore expected during sediment resuspension stages (Tack et al. 1995; Förstner 2004; Superville et al. 2015).

5 Conclusions

This study provides to our knowledge one of the first attempts to combine quantifications of short-term sediment dynamics and particle-bound fluxes of metallic contaminants in a channelized river draining an industrialized catchment.

We show the relevance of using ^{7}Be records in riverbed and suspended sediments to calculate sediment deposition and resuspension rates over the short term. It is demonstrated that ^{7}Be is found in measurable quantities and continuously delivered to the river (i.e., through sediment supply and resuspension).

Rates of short-term sediment dynamics ranged from −0.46±0.05 g cm^{-2} mo^{-1} to 0.3±0.2 g cm^{-2} mo^{-1}, and Net annual sediment rates reflected large variations in sediment accumulation due to changes in channel morphology. Sediment of the Lower Scarpe River acts as a sink for contaminants due to the long-term accumulation of particle-bound metals in the riverbed. However, sediment acts as a source of contamination during short resuspension periods. Sediment resuspended at a rate of −0.06 g cm^{-2} mo^{-1} generated a flux of metals to the river water column of 12, 13, 23 and 145 µg cm^{-2} mo^{-1} for Cd, Cu, Pb and Zn, respectively. Over the 15-months monitoring, 28 to 50 % of Pb that deposited on the riverbed was resuspended and contributed to the concomitant degradation of water quality.

The use of fallout radionuclides to quantify dynamics of sediment and particle-bound contaminants proved to be relevant to demonstrate the role of sediment as a sink or source of contamination for water. Further investigations are required to evaluate decontamination times in order to guide the implementation of efficient management strategies aimed at improving water quality in these river systems.

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Table captions

Table 1: Depth distribution of ^{7}Be activity (Bq kg^{-1}) in sediment collected in April 2005 (sampling campaign n°1 on April 19, 2005).

Table 2: From April 2005 to August 2006: Inventory of ^{7}Be (mBq cm^{-2}) in top and bottom sediment layers (0−3 and 0−5 cm layers) collected in triplicate (a, b and c). Three sediment layers were not analysed due to the loss of samples during subsampling procedure ("n.d." is mentioned). Activities in ^{7}Be lower than the limits of quantification were estimated to be equal to half of the limits of quantification.

Table 3: From April 2005 to August 2006: Total, Residual and New inventories in ^{7}Be of sediment (mBq cm^{-2}); Activity in ^{7}Be of suspended sediment (Bq kg^{-1}); Monthly rate of sediment Deposition/Resuspension (=D/RS, in g cm^{-2} mo^{-1}). When suspended sediment was not available (due to the loss of sampling device), activity in ^{7}Be was assessed by calculating the mean activity measured in suspended sediments collected during the same period at the other sites (mentioned in grey).

Table 4: Metal contents in suspended sediments (µg g^{-1}) collected from April 2005 to August 2006 (standard analytical deviation did not exceed 5 %). When suspended sediment was not available (due to the loss of sampling device), metal content was assessed by calculating the mean content measured in suspended sediments collected at the same site during the other sampling periods (mentioned in grey).

Table 5: From April 2005 to August 2006: Cumulated fluxes of contaminant deposition and resuspension, and Net fluxes of contaminant (µg cm^{-2}). Proportions of Total resuspension regarding Total deposition (%).

Figure captions

Fig. 1: The Lower Scarpe River catchment. Location of the 3 sampling sites (k13, k16 and k36), and the upstream lock ("Fort de Scarpe"). The river receives water discharges from one tributary (T) and 2 lift stations (LS1, LS2), and liquid effluents originating from one wastewater treatment plant (WW) and two industrial plants (I1: gas production, I2: surface finishing).

Fig. 2: Evolution of daily rainfall (mm) and water discharge measured at the lock-station "Fort de Scarpe" ($m^3\ s^{-1}$) from April 2005 to August 2006 (source: "Voies Navigables de France"). Vertical lines indicate the sampling dates (for details see Table 2).

Fig. 3: Evolution in ^{7}Be activity of suspended sediment (Bq kg^{-1}) and monthly-cumulated rainfall (mm) between successive sampling campaigns.

Fig. 4: Plot of ^{7}Be inventories in the top (0−3 cm) sediment layer (mBq cm^{-2}) *versus* ^{7}Be activity in suspended sediment (Bq kg^{-1}).

Fig. 5: Monthly sediment Deposition/Resuspension (=D/RS) rates (g cm^{-2} mo^{-1}) calculated for (a) k13, (b) k16 and (c) k36 sites.

Fig. 6: From April 2005 to August 2006: Cumulated fluxes of deposition and resuspension and Net accumulation fluxes of Pb (µg cm^{-2}) for the 3 sampling sites.

Figures

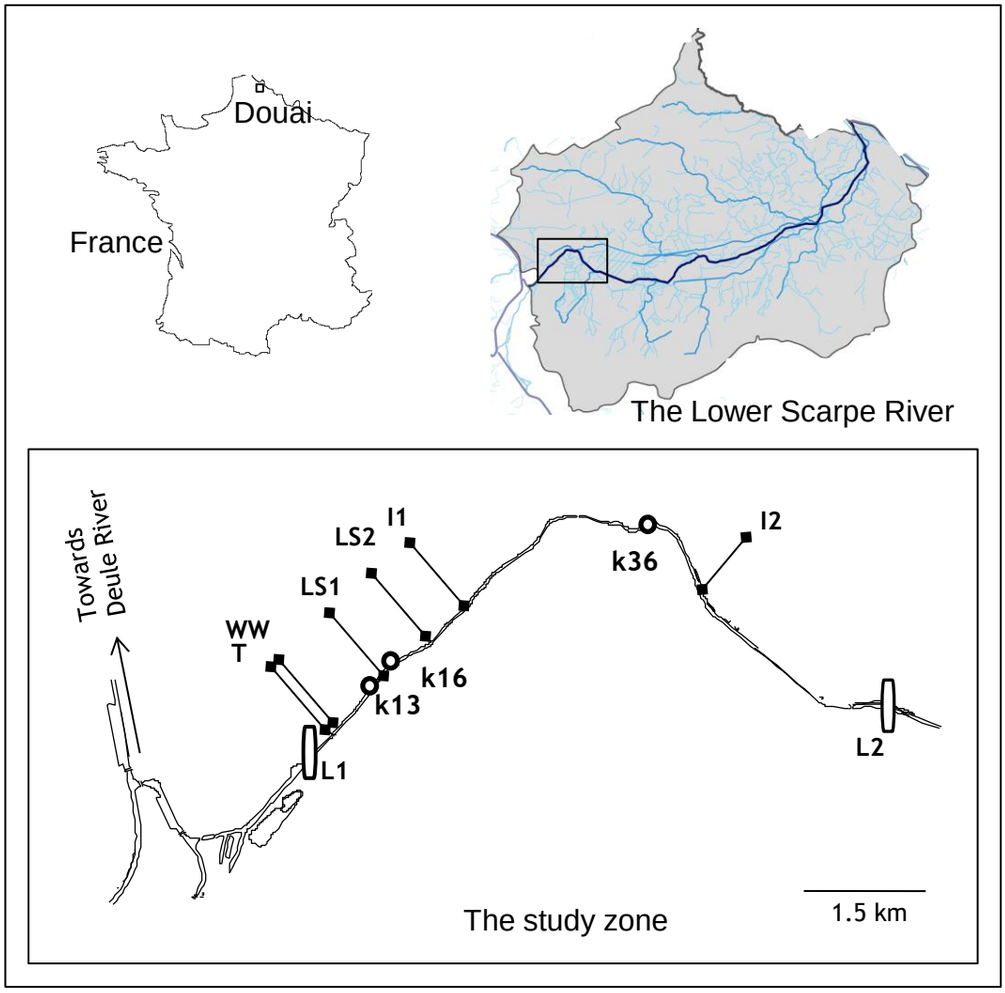

Fig. 1

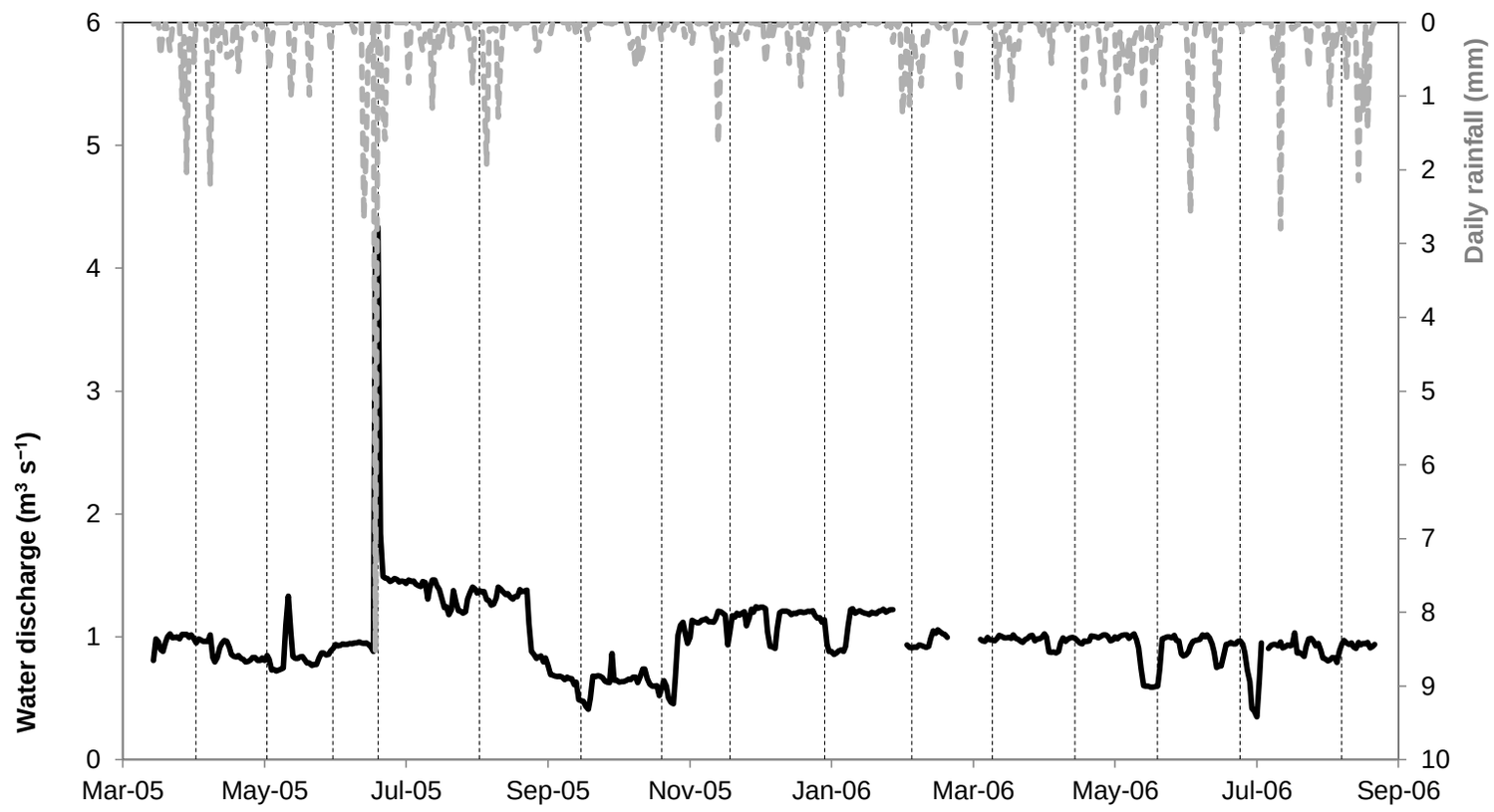
Water discharge (m³ s⁻¹)
Daily rainfall (mm)
Mar-05
May-05
Jul-05
Sep-05
Nov-05
Jan-06
Mar-06
May-06
Jul-06
Sep-06

Fig. 2

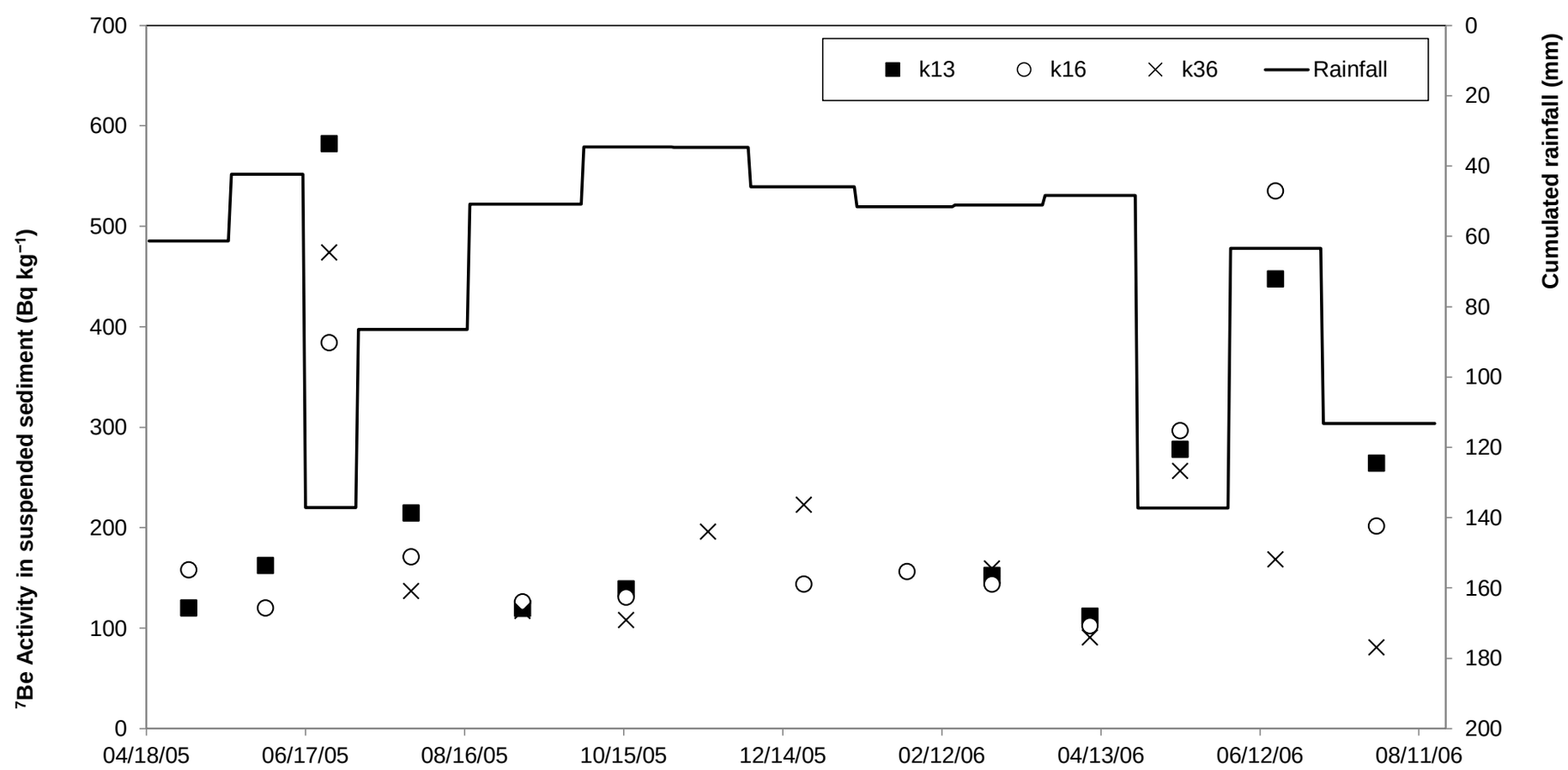
k13
k16
k36
Rainfall
⁷Be Activity in suspended sediment (Bq kg⁻¹)
Cumulated rainfall (mm)
04/18/05
06/17/05
08/16/05
10/15/05
12/14/05
02/12/06
04/13/06
06/12/06
08/11/06

Fig. 3

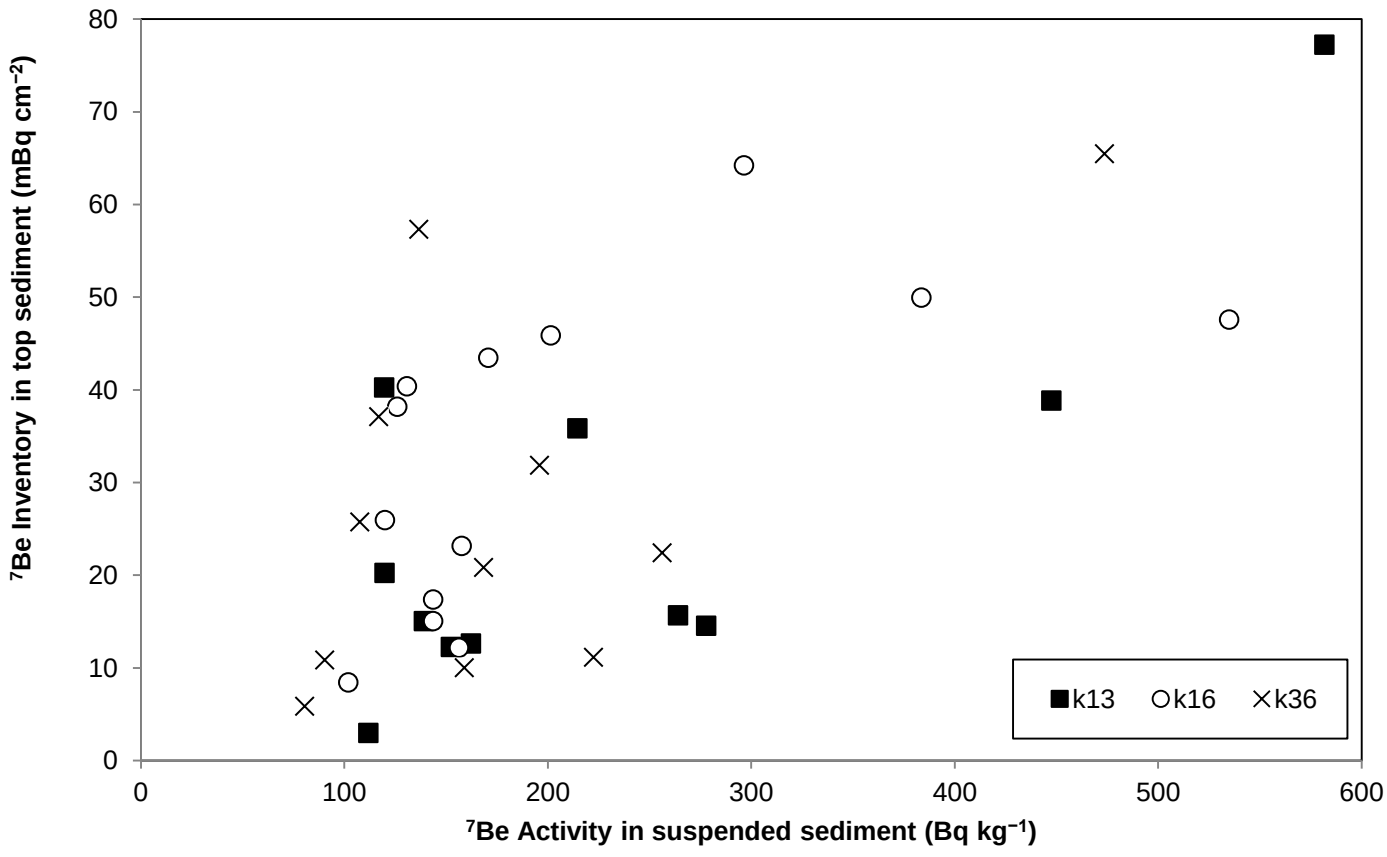
80
70
60
50
40
30
20
10
0
0
100
200
300
400
500
600
^{7}Be Inventory in top sediment (mBq cm^{-2})
^{7}Be Activity in suspended sediment (Bq kg^{-1})
k13
k16
k36

Fig. 4

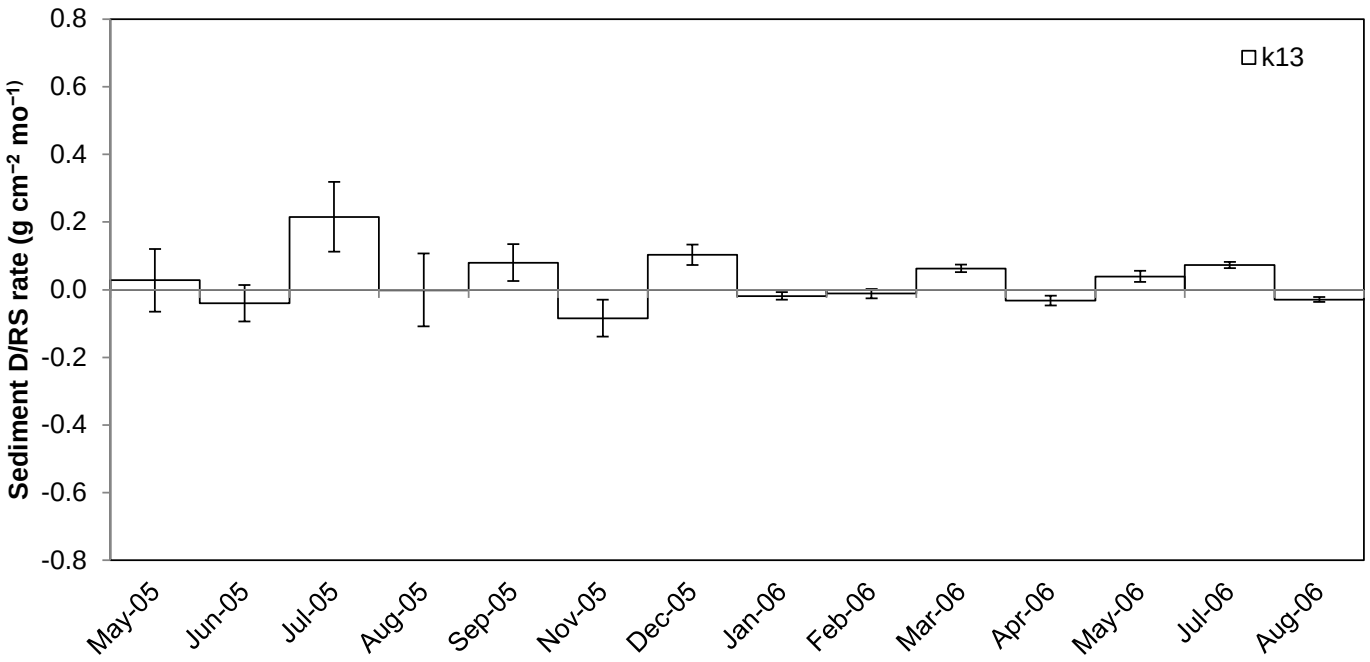

Fig. 5a

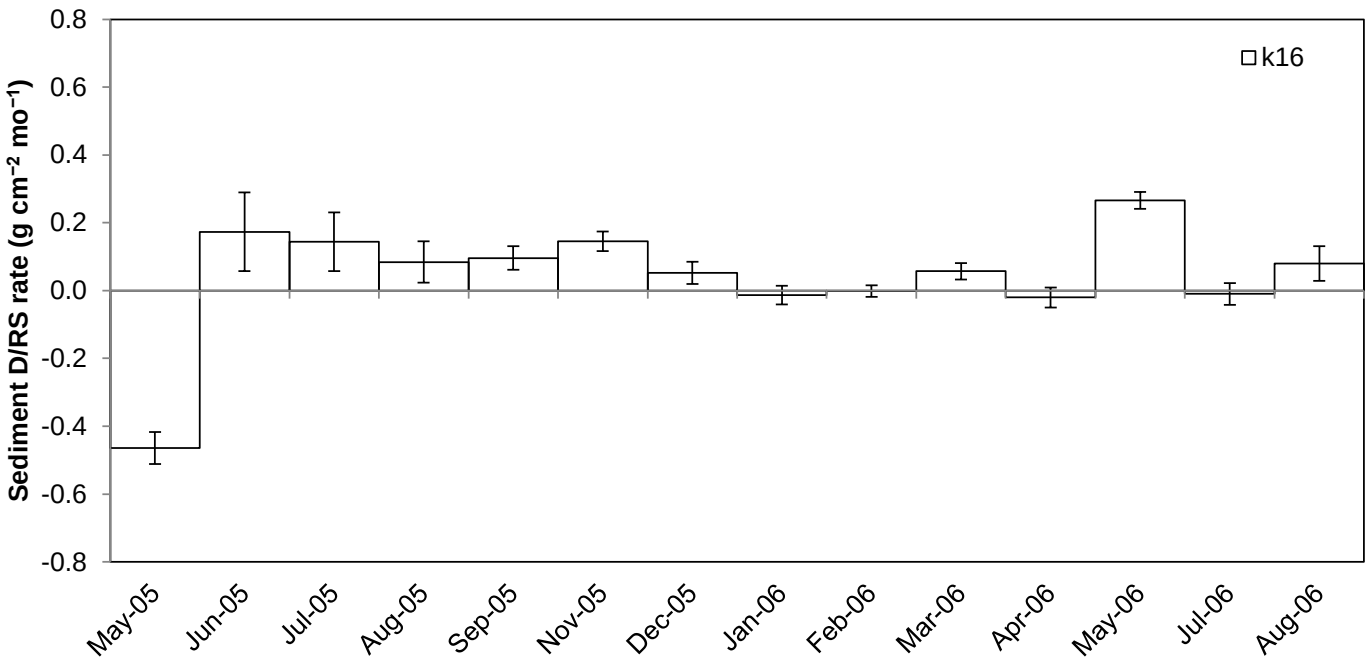

Fig. 5b

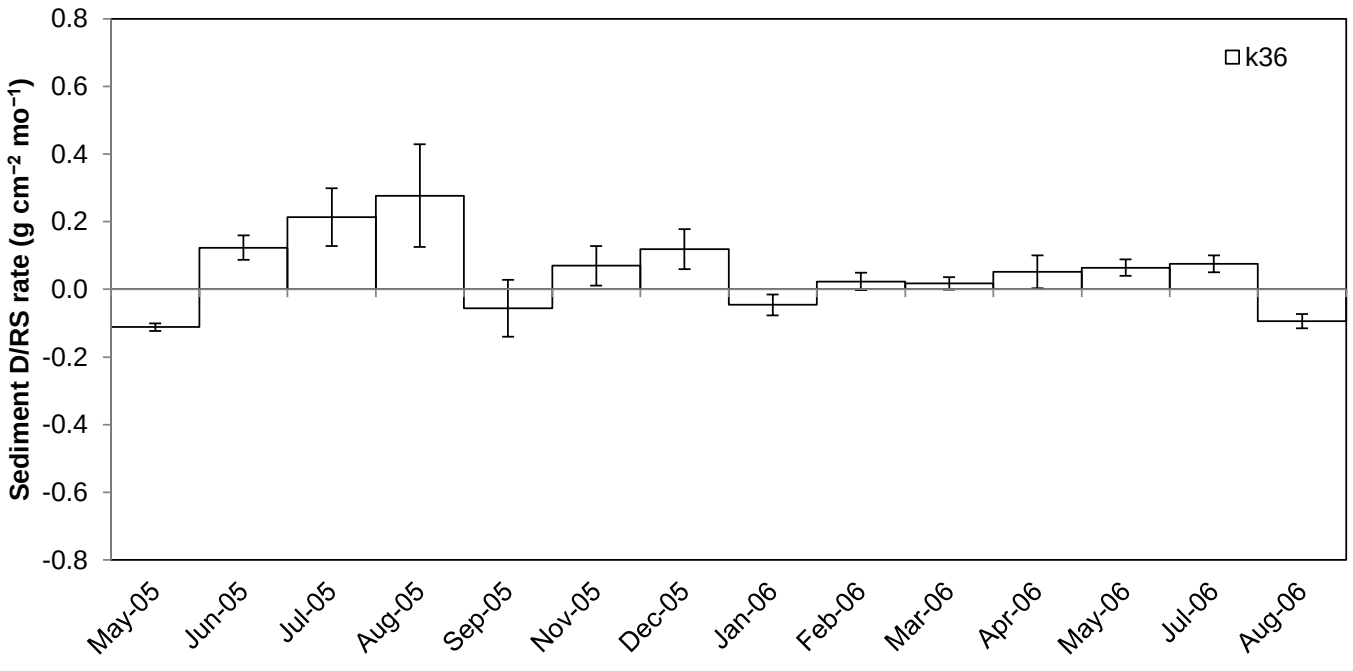

Fig. 5c

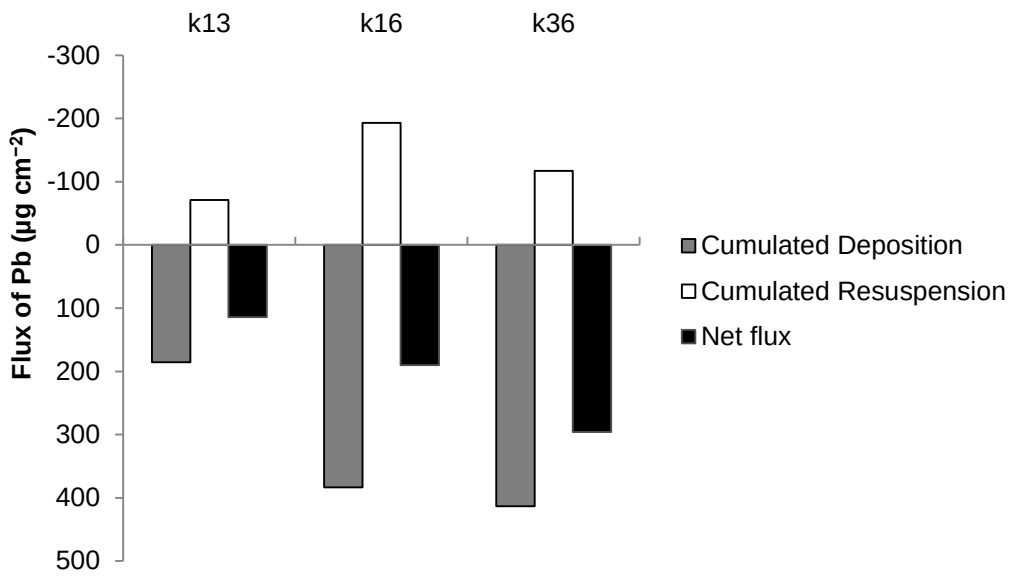
k13
k16
k36
-300
-200
-100
0
100
200
300
400
500
Flux of Pb (µg cm⁻²)
Cumulated Deposition
Cumulated Resuspension
Net flux

Fig. 6

Table 1

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

Supplementary Material
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Supplementary material: Table and figure captions

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

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

Table S3: Activities in ^{137}Cs, $^{210}Pb_{xs}$ and ^{232}Th (Bq kg^{-1}), and $^{7}Be/^{210}Pb_{xs}$ activity ratio in suspended sediment. Areal activities in ^{7}Be and ^{210}Pb (Bq m^{-2}) in rainwater samples monthly collected from August 2005 to September 2006 (data available were from the French Institute of Radioprotection and Nuclear Safety, OPERA-Air network). Fractions of recent sediment in suspended sediments (%).

Table S4: Characteristics of suspended sediment collected at the lift station “Vallée de Scarpe”: fraction of fine material (<63 µm, in %); ^{7}Be activities (Bq kg^{-1}); metal contents (Cu and Pb in µg g^{-1}, Cd and Zn in mg g^{-1}).

Table S5: Monthly fluxes of contaminants associated with sediment dynamics (µg cm^{-2} mo^{-1}).

Figure S1: (a) Plot of ^{137}Cs activities versus fractions of fine material in bed and suspended sediments (=SS); (b) Plot of ^{137}Cs activities versus ^{232}Th activities in bed and suspended sediments (=SS).

Figure S2: (a) Plot of ^{7}Be activities versus fractions of fine material in suspended sediments (=SS); (b) Plot of ^{7}Be activities versus organic carbon contents in suspended sediments (=SS); (c) Plot of ^{7}Be activities versus ^{232}Th activities in suspended sediments (=SS).

Supplementary Material

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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
	±	Bq m^{-2}						*3*	*6*	*4*	*2*	*0.3*	*4*	*9*	*0.8*	*12*
	^{210}Pb							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 %**	**^{7}Be Bq kg^{-1}**	**±**	**Cu µg g^{-1}**	**±**	**Pb µg g^{-1}**	**±**	**Cd mg g^{-1}**	**±**	**Zn mg g^{-1}**	**±**
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 ($\mu g\ cm^{-2}\ mo^{-1}$)									**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

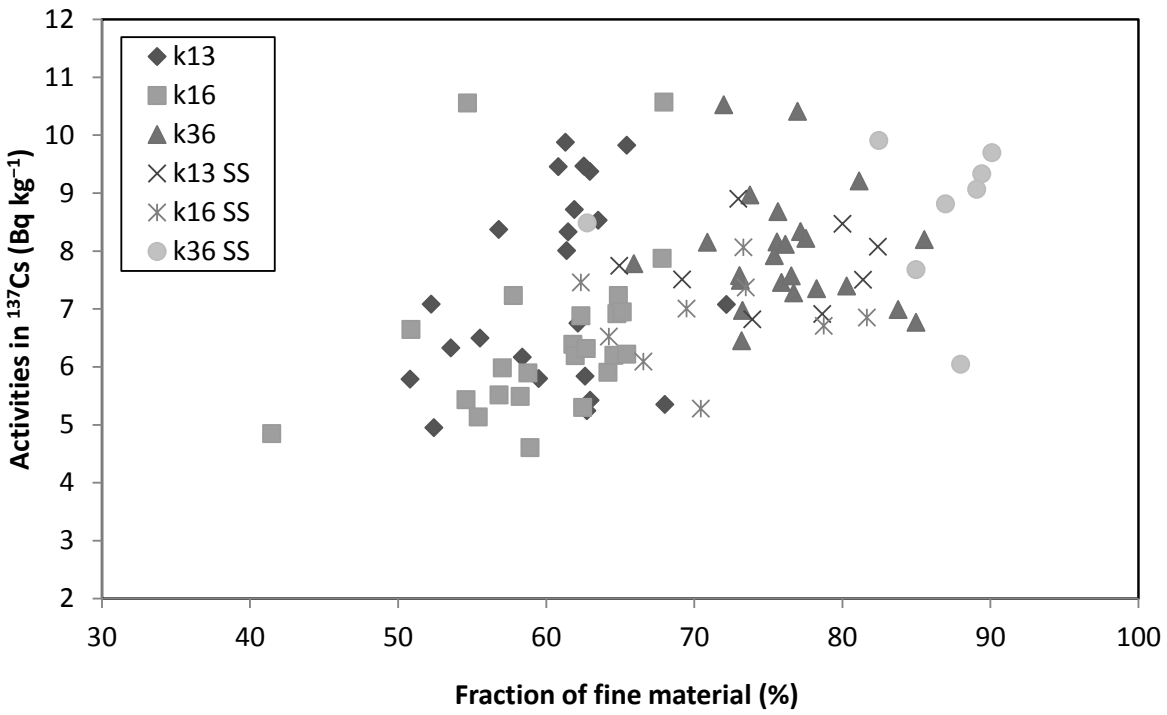

Fig. S1a

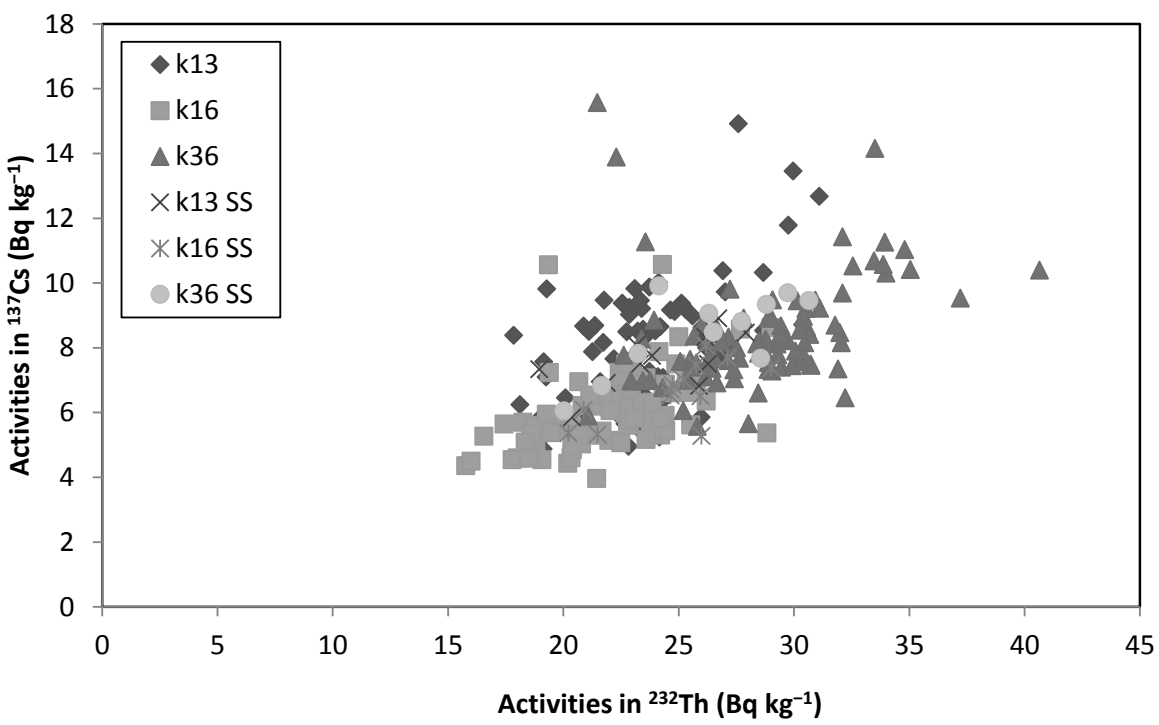

Fig. S1b

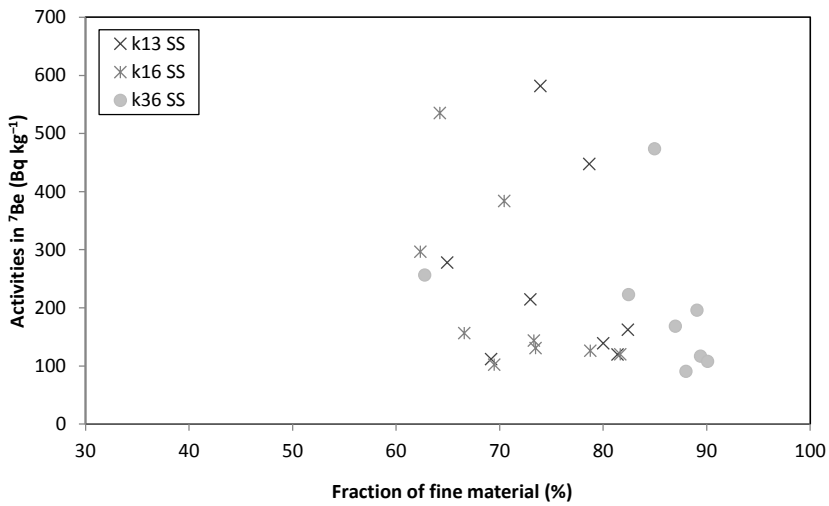

Fig. S2a

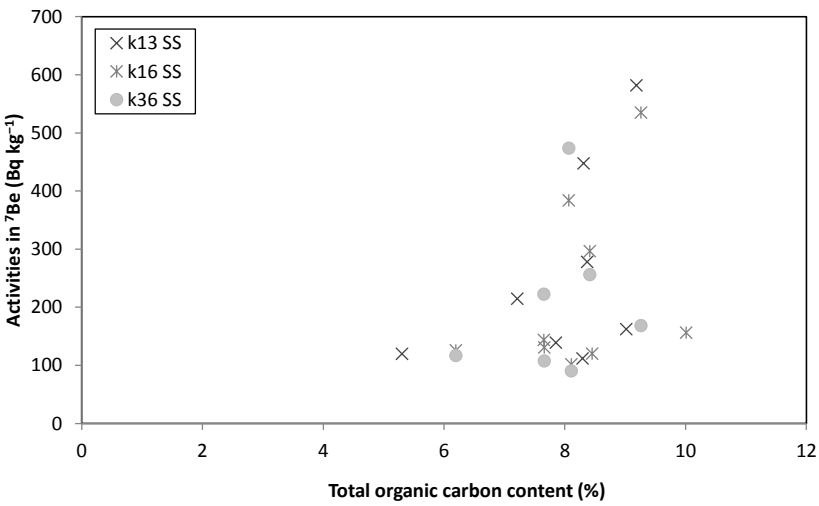

Fig. S2b

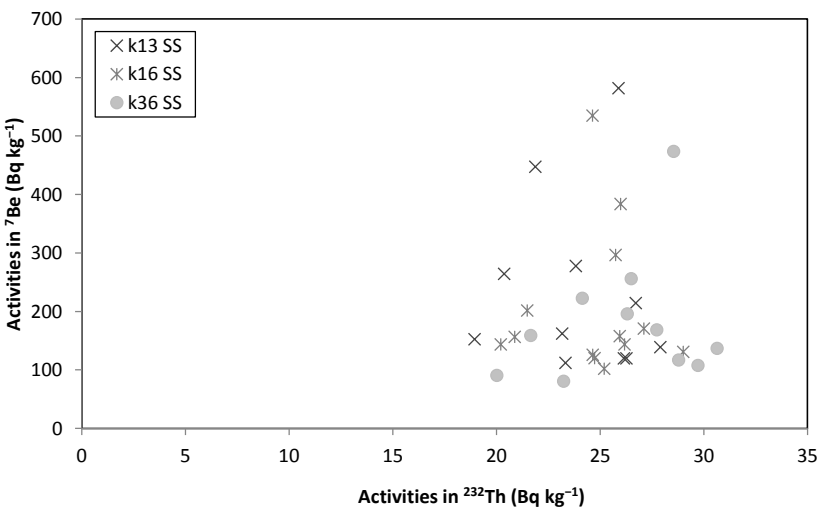

Fig. S2c