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

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

Mathilde Zebracki • Claire Alary • Irène Lefèvre • Vasilica Nan-Hammade • Olivier Evrard • Philippe Bonté

M. Zebracki (✉) • C. Alary • V. Nan-Hammade

Centre National de Recherches sur les Sites et Sols Pollués (CNRSSP), Douai, France

M. Zebracki • C. Alary

Département Génie Civil et Environnemental de l'Ecole des Mines de Douai (GCE/LGCgE), Douai, France

Irène Lefèvre • O. Evrard • P. Bonté

Laboratoire des Sciences du Climat et de l'Environnement (LSCE/IPSL), Unité Mixte de Recherche 8212 (CEA/CNRS/UVSQ), Gif sur Yvette Cedex, France

Vasilica Nan-Hammade

Institut Scientifique de Service Public (ISSeP), Colfontaine, Belgique

(✉) **Corresponding author:**

Mathilde Zebracki

e-mail: zebracki@free.fr

Abstract

Purpose: Contamination of river sediment reaches problematic levels in industrialized regions. This study investigates the relevance of using Beryllium-7 (^7Be), 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 ^7Be 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 ^7Be inventory between two successive samplings. Total inventory of ^7Be consists of two components, i.e., the Residual inventory and the New inventory.

Results and discussion: Inventories of ^7Be 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 $\mu\text{g cm}^{-2} \text{mo}^{-1}$ 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 (^7Be , 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, ^7Be 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 ^7Be -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 ^7Be 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 ^7Be 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 ^7Be 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 ^7Be in collected material rapidly after sampling (before ^7Be has decayed too much). In this context, the present paper investigates the potential of using ^7Be 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 ($1 \text{ m}^3 \text{ s}^{-1}$). 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 $500 \mu\text{g g}^{-1}$ 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 Eijkelpomp® 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 ^7Be 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 ^7Be 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), ^7Be 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 ^7Be gamma energy level (477 keV). Activity in $^{210}\text{Pb}_{\text{xs}}$ (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 $\pm 2 \sigma$.

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 ^7Be 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 ^7Be 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; $\text{HNO}_3\text{:HCl}$ 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 $\pm 1 \sigma$.

3.2. Short-term sediment dynamics

3.2.1 Inventory of ^7Be in bed sediment

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

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

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

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

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 ^7Be inventory reflects deposition of sediment, whereas a lower Total ^7Be inventory corresponds to the occurrence of sediment resuspension from the riverbed.

Total ^7Be inventory in surface bed sediment [$I(^7\text{Be})_{\text{TOTAL}}$, in mBq cm^{-2}] is calculated by multiplying 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}), and by summing ^7Be inventory of all individual subsections ($\sum_i A_i$, i corresponding to the sediment layer, i.e., 0–3 or 3–5 cm):

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

When calculating Total ^7Be inventory in sediment of the Lower Scarpe River, ^7Be activities lower than the limits of quantification were estimated to be equal to half of the limits of quantification. Total ^7Be inventories correspond to the mean of ^7Be 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 ^7Be activities, the ^7Be 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 ^7Be activities, and thus Total ^7Be 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 ^7Be inventory in sediment [$I(^7\text{Be})_{\text{NEW}}$, in mBq cm^{-2}] by the mean ^7Be activity [$A(^7\text{Be})_{\text{MEAN}}$, in Bq kg^{-1}] of newly deposited or resuspended sediment (Canuel et al. 1990):

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

In the present study, the mean ^7Be 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 $\text{g cm}^{-2} \text{ mo}^{-1}$:

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

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 $\mu\text{g cm}^{-2} \text{ mo}^{-1}$) is calculated by multiplying short-term sediment rate (R_{SEDIMENT} , in $\text{g cm}^{-2} \text{ mo}^{-1}$) by metal content (C_{METAL} , in $\mu\text{g g}^{-1}$) 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:

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

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 ⁷Be-labelled sediment in suspended sediment

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

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

The sediment source term is defined by sediment particles that have a ⁷Be/²¹⁰Pb activity ratio equal to that of wet fallout. The dry deposition of ⁷Be and the transfer time required for ⁷Be-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 ⁷Be and ²¹⁰Pb 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 ⁷Be/²¹⁰Pb_{xs} source term is calculated using areal activities in ⁷Be and ²¹⁰Pb 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 m³ 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 %).

¹³⁷Cs 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 ²³²Th was used as a proxy of fine-grained sediment content (Sakaguchi et al. 2006; Foucher et al. 2015). The positive trend found between ¹³⁷Cs activities and (a) fractions of fine material, or (b) ²³²Th 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 ¹³⁷Cs activities also reflect variations in sediment origin (Wallbrink et al. 1996).

4.3 Rainwater ⁷Be and ²¹⁰Pb activities

Rainwater ⁷Be and ²¹⁰Pb 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 ⁷Be washout from the atmosphere, origin of air masses, and atmospheric production rate of ⁷Be (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 ⁷Be in sediment

The vertical distribution of ⁷Be in sediment was investigated during the first sampling campaign (April 19, 2005). Activities in ⁷Be 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 ^7Be associated with sediments in depth. As expected, ^7Be 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 ^7Be inventory of bed sediment

In sediment layers collected from April 2005 to August 2006 ($n=258$), ^7Be inventories ranged from 1 to 135 mBq cm^{-2} (Table 2). Total ^7Be 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 ^7Be inventories between sediment replicates are then rather influenced by the spatial heterogeneity in the deposition of ^7Be -labelled particles on the riverbed, as sediment activity in ^7Be 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 ^7Be inventories in top and bottom sediment layers (0–3 cm vs. 3–5 cm), 4 types of sediment deposition patterns were found. The highest ^7Be activities were generally found in the top sediment layer (91 % of observations) indicating the recent deposition of ^7Be -labelled sediment. In some cases, both top and bottom sediments displayed low ^7Be inventories (< 5 % of cases), indicating either the resuspension of previously deposited sediment or the absence of sediment deposition. Rarely (< 3 % of cases), ^7Be inventory remained relatively constant with depth, indicating the occurrence of local physical disturbances, such as bioturbation that contributed to homogenize the ^7Be records in depth (Santschi et al. 1990, Ciutat et al. 2005). Although the potential loss of ^7Be 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 ^7Be inventories in sediment (Taylor et al. 2012, Taylor et al. 2013).

4.5 Beryllium-7 in suspended sediment

Activities in ^7Be of suspended sediment ($n=35$) ranged from 81 to 582 Bq kg $^{-1}$ (Table 3). Variations in ^7Be 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 ^7Be 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 ^7Be activities measured in suspended sediment (Fig. 3): in July 2005 and May 2006, increase in ^7Be activities (up to 582 Bq kg $^{-1}$) was associated with increasing monthly cumulated rainfall (up to 137 mm). In contrast, low ^7Be 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 ^7Be activities of suspended sediment coincided with flood occurrence (Fig. 3), suggesting a significant delivery of recently ^7Be -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 ^7Be 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 ^7Be activities observed between sites in July 2005, July 2006 and August 2006 suggest the occurrence of mixing processes in the river between “recent” (^7Be -enriched) and “older” (^7Be -depleted) sediment particles (Olsen et al. 1985; Wallbrink and Murray 1996; Matisoff et al. 2005). In July and August 2006, decreasing ^7Be activities in downstream direction was related to decreasing fractions of recent sediment (Table S3). In November 2005 and April 2006, ^7Be 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 ^7Be records in top sediment layers (0–3 cm) and suspended sediments indicates that both material types are affected by the supply of recent ^7Be -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 ^7Be records in suspended sediment to normalize variations in ^7Be inventories of sediment (Equation 3), as this material integrates the variability of ^7Be 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 $-0.5 \text{ g cm}^{-2} \text{ mo}^{-1}$ to $0.3 \text{ g cm}^{-2} \text{ mo}^{-1}$. These values are comprised within the range of short-term sediment dynamics found in other rivers and coastal systems [-0.7 to $1.4 \text{ g cm}^{-2} \text{ mo}^{-1}$ (Canuel et al. 1990); -0.08 to $0.3 \text{ g cm}^{-2} \text{ mo}^{-1}$ (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 $0.8 \text{ g cm}^{-2} \text{ y}^{-1}$, 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 $0.8 \text{ g cm}^{-2} \text{ y}^{-1}$ 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 $-0.06 \text{ g cm}^{-2} \text{ mo}^{-1}$ supplied fluxes to the water column of 12, 13, 23 and $145 \mu\text{g cm}^{-2} \text{ mo}^{-1}$ for Cd, Cu, Pb and Zn, respectively (Table S5, Electronic Supplementary Material). As a comparison, a sediment resuspension of $0.08 \text{ g cm}^{-2} \text{ mo}^{-1}$ in the Fox River corresponded to a $\sum \text{PCB}$ flux of $0.24 \mu\text{g cm}^{-2} \text{ mo}^{-1}$ (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 ^7Be records in riverbed and suspended sediments to calculate sediment deposition and resuspension rates over the short term. It is demonstrated that ^7Be 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 $-0.46 \pm 0.05 \text{ g cm}^{-2} \text{ mo}^{-1}$ to $0.3 \pm 0.2 \text{ g cm}^{-2} \text{ mo}^{-1}$, 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 $-0.06 \text{ g cm}^{-2} \text{ mo}^{-1}$ generated a flux of metals to the river water column of 12, 13, 23 and $145 \mu\text{g cm}^{-2} \text{ mo}^{-1}$ for Cd, Cu, Pb and Zn, respectively. Over the 15-months monitoring, 28 to 50 % of 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.

Acknowledgements This study was financially supported by the European Union (FEDER) and French authorities (DIREN). Water discharge data were provided by “Voies Navigables de France”. We gratefully acknowledge Vincent Ledoux (Société des Eaux de Douai), William Guerin (Agence de l’Eau Artois-Picardie), and Olivier Masson (Institut de Radioprotection et de Sûreté Nucléaire) for providing rainfall records, maps and ^7Be and ^{210}Pb activities in rainfall, respectively. We also gratefully acknowledge Manon Heysck, Florent Mourier, Bertrand Girondelot, Terence Daumeries and Philippe Bataillard (CNRSSP) for their assistance during fieldwork.

The authors also gratefully thank the two anonymous reviewers whose suggestions greatly improved the quality of the manuscript.

References

- Argese E, Ramieri E, Bettiol C, Pavoni B, Chiozzotto E, Sfriso A (1997) Pollutant exchange at the water/sediment interface in the Venice canals. *Water Soil Air Pollut* 99:255-263
- Audry S, Schäfer J, Blanc G, Jouanneau J-M (2004) Fifty-year sedimentary record of heavy metal pollution (Cd, Zn, Cu, Pb) in the Lot River reservoirs (France). *Environ Pollut* 132:413-426.
- Ayrault S, Roy-Barman M, Le Cloarec M-F, Rianti Priadi C, Bonté P, Göpel C (2012) Lead contamination of the Seine River, France: Geochemical implications of a historical perspective. *Chemosphere* 87:902-910
- Baskaran M, Santschi PH (1993) The role of particles and colloids in the transport of radionuclides in coastal environments of Texas. *Mar Chem* 43:95-114
- Benoit G, Rozan TF (1999) Influences of size distributions on the particle concentration effect and trace metal partitioning in rivers. *Geochim Cosmochim Acta* 63:113–127
- Ben Slimane A, Raclot D, Evrard O, Sanaa M, Lefèvre I, Ahmadi M, Tounsi M, Rumpel C, Ben Mammou A, Le Bissonnais Y (2013) Fingerprinting sediment sources in the outlet reservoir of a hilly cultivated catchment in Tunisia. *J Soils Sediments* 13:801-815
- Blake WH, Wallbrink PJ, Wilkinson SN, Humphreys GS, Doerr SH, Shakesby RA, Tomkins KM (2009) Deriving hillslope sediment budgets in wildfire-affected forests using fallout radionuclide tracers. *Geomorphol* 104:105-116.

Bonté P, Mouchel JM, Thomas AJ, Le Cloarec MF, Dumoulin JP, Sogon S, Tessier L (2000) Buffering of suspended sediment transport in lowland river during low water stages: quantification in river Seine using environmental radionuclides. *Acta Geologica Hispanica* 35(3-4):339-355

Bourcier L, Masson O, Laj P, Pichon JM, Paulat P, Freney E, Sellegri K (2011) Comparative trends and seasonal variation of ⁷Be, ²¹⁰Pb and ¹³⁷Cs at two altitude sites in the central part of France. *J Environ Radioact* 102:294-301

Calmano W, Hong J, Förstner U (1993) Binding and mobilization of heavy metals in contaminated sediments affected by pH and redox potential. *Water Sci Technol* 28:223-235

Canuel EA, Martens CS, Benninger LK (1990) Seasonal variations in ⁷Be activity in the sediments of Cape Lookout Bight, North Carolina. *Geochim Cosmochim Acta* 54:237-245

Ciutat A, Anschutz P, Gerino M, Boudou A (2005) The effects of bioturbation on Cadmium transfer and distribution into freshwater sediments. *Environ Toxicol Chem* 24:1048-1058

Conaway CH, Storlazzi CD, Draut AE, Swarzenski PW (2013) Short-term variability of ⁷Be atmospheric deposition and watershed response in a Pacific coastal stream, Monterey Bay, California, USA. *J Environ Radioact* 120:94-103

Dibb JE and Rice DL (1989) Temporal and spatial distribution of Beryllium-7 in the sediments of Chesapeake Bay. *Estuar Coast Shelf Sci* 28:395-406

Dominik J, Burrus D, Vernet J-P (1987) Transport of the environmental radionuclides in an alpine watershed. *Earth Planet Sci Lett* 93:165-180

Dominik J, Schuler C, Santschi PH (1989) Residence times of ²³⁴Th and ⁷Be in Lake Geneva. *Earth Planet Sci Lett* 93:345-358

Du J, Wu Y, Huang D, Zhang J (2010) Use of ⁷Be, ²¹⁰Pb and ¹³⁷Cs tracers to the transport of surface sediments of the Changjiang Estuary, China. *J Mar Syst* 82:286–294

Eggleton J and Thomas KV (2004) A review of factors affecting the release and bioavailability of contaminants during sediment disturbance events. *Environ Int* 30:973-980

Evrard O, Némery J, Gratiot N, Duvert C, Ayrault S, Lefèvre I, Poulenard J, Prat C, Bonté P, Esteves M (2010) Sediment dynamics during the rainy season in tropical highland catchments of central Mexico using fallout radionuclides. *Geomorphol* 124:42-54

Evrard O, Navratil O, Ayrault S, Ahmadi M, Némery J, Legout C, et al. (2011) Combining suspended
 sediment monitoring and fingerprinting to determine the spatial origin of fine sediment in a
 mountainous river catchment. *Earth Surf Proc Land* 36:1072–89
 Feng H, Cochran JK, Hirschberg DJ (1999) ²³⁴Th and ⁷Be as tracers for transport and sources of
 particle-associated contaminants in the Hudson River Estuary. *Sci Total Environ* 237/238:401-
 418
 Fitzgerald SA, Val Klump J, Swarzenski PW, Mackenzie RA, Richards KD (2001) Beryllium-7 as a
 tracer of short-term sediment deposition and resuspension in the Fox River, Wisconsin. *Environ*
Sci Technol 35:300-305
 Förstner U (2004) Sediment dynamics and pollutant mobility in rivers: An interdisciplinary approach.
Lakes & reserv: Res Manag 9:25-40
 Foucher A, Laceby JP, Salvador Blanes S, Evrard O, Le Gall M, Lefèvre I, Cerdan O, Rajkumar V,
 Desmet M (2015) Quantifying the dominant sources of sediment in a drained lowland agricultural
 catchment: the application of a thorium based particle size correction in sediment fingerprinting.
Geomorphol, in revision
 Gardner WD (1980) Field assessment of sediment traps, *J Mar Res* 38(1):41-52
 Gateuille D, Evrard O, Lefèvre I, Moreau-Guigon E, Alliot F, Chevreuil M, Mouchel J-M (2014) Mass
 balance and decontamination times of Polycyclic Aromatic Hydrocarbons in rural nested
 catchments of an early industrialized region (Seine River basin, France). *Sci Total Environ* 470-
 471:608-617
 Gibbs RJ (1973) Mechanisms of trace metal transport in rivers. *Sci* 180:71-73
 Giffin D, Reide Corbett D (2003) Evaluation of sediment dynamics in coastal systems via short-lived
 radioisotopes. *J Mar Syst* 42:83-96
 Gourdin E, Evrard O, Huon S, Reyss J-L, Ribolzi O, Bariac T, Sengtaheuanghoung O, Ayrault S
 (2014) Spatial and temporal variability of ⁷Be and ²¹⁰Pb wet deposition during four successive
 monsoon storms in a catchment of northern Laos. *J Environ Radioact* 136:195-205
 Haag I, Kern U, Westrich B (2001) Erosion investigation and sediment quality measurements for a
 comprehensive risk assessment of contaminated aquatic sediments. *Sci Total Environ* 266:249-
 257

- Hawley N, Robbins JA, Eadie BJ (1986) The partitioning of ^7Be in fresh water. *Geochim Cosmochim Acta* 50:1127-1131
- He Q and Walling DE (1996) Interpreting particle size effects in the adsorption of ^{137}Cs and unsupported ^{210}Pb by mineral soils and sediments. *J Environ Radioact* 30:117-137
- Horowitz AJ (2000) NASQAN: the design and implementation of a large-river suspended sediment and trace element flux programme. In: *The role of erosion and sediment transport in nutrient and contaminant transfer*, IAHS Publ 263:3-18
- Huang D, Du J, Zhang J (2011) Particle dynamics of ^7Be , ^{210}Pb and the implications of sedimentation of heavy metals in the Wenjiao/Wenchang and Wanquan River estuaries, Hainan, China. *Estuar Coast Shelf Sci* 93:431-437
- Hwang K-Y, Kim H-S, Hwang I (2011) Effect of Resuspension on the Release of Heavy Metals and Water Chemistry in Anoxic and Oxidic Sediments. *CLEAN – Soil Air Water* 39(10):908–915
- Jagercikova M, Cornu S, Le Bas C, Evrard O (2015) Vertical distributions of ^{137}Cs in soils: a meta-analysis. *J Soils Sediments* 15:81-95
- Jweda J, Baskaran M, van Hees E, Schweitzer L (2008) Short-lived radionuclides (^7Be and ^{210}Pb) as tracers of particle dynamics in a river system in southeast Michigan. *Limnol Oceanogr* 53(5):1934-1944
- Kaste JM, Baskaran M (2011) Meteoric ^7Be and ^{10}Be as process tracers in the environment. In: Baskaran, M. (Ed.), *Handbook of environmental isotope geochemistry*. *Adv Isot Geochem* 1:61–85
- Krishnaswami S, Benninger LK, Aller RC, von Damm KL (1980) Atmospherically-derived radionuclides as tracers of sediment mixing and accumulation in near-shore marine and lake sediments: evidence from ^7Be , ^{210}Pb , and $^{239,240}\text{Pu}$. *Earth Planet Sci Lett* 47:307-318
- Lacal J, da Silva MP, García R, Sevilla MT, Procopio JR, Hernández L (2003) Study of fractionation and potential mobility of metal in sludge from pyrite mining and affected river sediments: changes in mobility over time and use of artificial ageing as a tool in environmental impact assessment. *Environ Pollut* 124:291-305
- Lal D, Malhotra PK, Peters B (1958) On the production of radioisotopes in the atmosphere by cosmic radiation and their application to meteorology. *J Atmos Terr Phys* 12:306-328

- Le Cloarec M-F, Bonté P, Lefèvre I, Mouchel J-M, Colbert S (2007) Distribution of ^7Be , ^{210}Pb and ^{137}Cs in watersheds of different scales in the Seine River basin: Inventories and residence times. *Sci Total Environ* 375:125-139
- Le Cloarec M-F, Bonté PH, Lestel L, Lefèvre I, Ayrault S (2011) Sedimentary record of metal contamination in the Seine River during the last century. *Phys Chem Earth* 36:515-529
- Lee S, Moon JW, Moon HS (2003) Heavy metals in the bed and suspended sediments of Anyang River, Korea: implications for water quality. *Environ Geochem Health* 25(4):433-452
- Mabit L, Benmansour M, Walling DE (2008) Comparative advantages and limitations of the fallout radionuclides ^{137}Cs , ^{210}Pb and ^7Be for assessing soil erosion and sedimentation. *J Environ Radioact* 99:1799–1807
- Martin JM, Mouchel JM, Thomas AJ (1986) Time concepts in hydrodynamic systems with an application to ^7Be in the Gironde Estuary. *Mar Chem* 18:369-392
- Matisoff G, Wilson CG, Whiting PJ (2005) The $^7\text{Be}/^{210}\text{Pb}$ ratio as an indicator of suspended sediment age or fraction new sediment in suspension. *Earth Surf Proc Land* 30:1191–1201
- Meybeck M, Lestel L, Bonté P, Moilleron R, Colin J-L, Rousselot O, Hervé D, De Pontevès C, Grosbois C, Thévenot DR (2007) Historical perspective of heavy metals contamination (Cd, Cr, Cu, Hg, Pb, Zn) in the Seine River basin (France) following a DPSIR approach (1950–2005). *Sci Total Environ* 375:204–231
- Morillo J, Usero J, Gracia I (2002) Partitioning of metals in sediments from the Odiel River (Spain). *Environ Int* 28:263-71
- Olley JM, Murray AS, Mackenzie DH, Edwards K (1993) Identifying sediment sources in a gullied catchment using natural and anthropogenic radioactivity. *Water Resour Res* 29:1037–1043
- Olsen CR, Larsen IL, Lowry PD, Cutshall H, Todd JF, Wong GTF, Casey WH (1985) Atmospheric fluxes and marsh-soil inventories of ^7Be and ^{210}Pb . *J Geophys Res* 90(D6):10,487-10,495.
- Olsen CR, Larsen IL, Lowry PD, Cutshall H (1986) Geochemistry and deposition of ^7Be in river-estuarine and coastal waters. *J Geophys Res* 91(C1):896-908.
- Owens PN, Batalla RJ, Collins AJ, Gomez B, Hicks DM, Horowitz AJ, Kondolf GM, Marden M, Page MJ, Peacock DH, Petticrew EL, Salomons W, Trustrum NA (2005) Fine-grained sediment in river systems: environmental significance and management issues. *River Res Appl* 21:693-717

- Palinkas CM, Nittrouer CA, Wheatcroft RA, Langone L (2005) The use of ^7Be to identify event and seasonal sedimentation near the Po River delta, Adriatic Sea. *Mar Geol* 222-223:95-112
- Pham MK, Povinec PP, Nies H, Betti M (2013) Dry and wet deposition of ^7Be , ^{210}Pb and ^{137}Cs in Monaco air during 1998-2010: Seasonal variations of deposition fluxes. *J Environ Radioact* 120:45-57
- Pinto VM, Pires LF, Bacchi OOS, Arthur RCJ, Bruno IP, Reichardt K (2013) Spatial variability of ^7Be fallout for erosion evaluation. *Radiat Phys Chem* 83:1–7
- Prygiel E, Superville P-J, Dumoulin D, Lizon F, Prygiel J, Billon G (2015) On biogeochemistry and water quality of river canals in Northern France subject to daily sediment resuspension due to intense boating activities. *Environ Pollut* 197:295-308
- Roche PA, Billen G, Bravard JP, Decamps H, Pennequin D, Vindimian E, Wasson JG (2005) Research needs for the implementation of the water European Framework Directive. *C R Geosci* 337:243-267
- Roos P, Valeur JR (2006) A sediment trap and radioisotope study to determine resuspension of particle reactive substances in the sound between Sweden and Denmark. *Cont Shelf Res* 26:474–487
- Sakaguchi A, Yamamoto M, Sasaki K, Kashiwaya K (2006) Uranium and Thorium Isotope Distribution in an Offshore Bottom Sediment Core of the Selenga Delta, Lake Baikal, Siberia. *J Paleolimnol*, 35:807-818
- Salomons W, Förstner U (1984) *Metals in the Hydrocycle*. Springer-Verlag, New York
- Santschi P, Höhener P, Benoit G, Buchholtz-ten Brink M (1990) Chemical processes at the sediment-water interface. *Mar Chem* 30:269-315
- Smith HG, Blake WH, Taylor A (2014). Modelling particle residence times in agricultural river basins using a sediment budget model and fallout radionuclide tracers. *Earth Surf Proc Land* 39:1944-1959
- Sobanska S (1999) Etude de la spéciation du plomb et du zinc dans des poussières industrielles et dans un sol contaminé. Approches par méthodes spectroscopiques. PhD Thesis, University of Lille 1, France
- Sommerfield CK, Nittrouer CA, Alexander CR (1999) ^7Be as a tracer of flood sedimentation on the northern California continental margin. *Cont Shelf Res* 19:335-361

Superville P-J, Prygiel E, Mykkelsen O, Billon G (2015) Dynamic behaviour of trace metals in the Deûle River impacted by recurrent polluted sediment resuspensions: From diel to seasonal evolutions. *Sci Total Environ* 506-570:585-593

Tack FM, Callewaert OWJJ, Verloo MG (1995) Metal solubility as a function of pH in a contaminated, dredged sediment affected by oxidation. *Environ Pollut* 91:199-208

Taylor A, Blake WH, Couldrick L, Keith-Roach MJ (2012) Sorption behaviour of beryllium-7 and implications for its use as a sediment tracer. *Geoderma* 187-188:16-23

Taylor A, Blake WH, Smith HG, Mabit L, Keith-Roach MJ (2013) Assumptions and challenges in the use of fallout beryllium-7 as a soil and sediment tracer in river basins. *Earth Sci Rev* 126:85-95

Wallbrink PJ, Olley JM, Murray AS, Olive LJ (1996) The contribution of subsoil to sediment yield in the Murrumbidgee River basin, New South Wales, Australia. In: *Erosion and Sediment Yield: Global and Regional Perspectives*. Proceedings of the Exeter Symposium. IAHS Publ 236

Wallbrink PJ and Murray AS (1996) Distribution and Variability of ⁷Be in Soils Under Different Surface Cover Conditions and its Potential for Describing Soil Redistribution Processes. *Water Resour Res* 32:467–476

Walling DE, Owens PN, Carter J, Leeks GJL, Lewis S, Meharg AA, Wright J (2003) Storage of sediment-associated nutrients and contaminants in river channel and floodplain systems. *Appl Geochem* 18:195-220

Walling DE (2013) Beryllium-7: The Cinderella of fallout radionuclide sediment tracers? *Hydrol Proc* 27:830-844

Westrich B and Förstner U (2007) *Sediment Dynamics and Pollutant Mobility in Rivers*. Springer book

Zebracki M, Bonté P, Alary C (2005) Particle-associated metallic contaminants transfer in a river displaying polluted sediment. Use of environmental radionuclides. In: *Proceedings of the 10th International Symposium on the Interactions Between Sediments and Water*, Bled, Slovenia

Zebracki M, Bonté P, Alary C (2007) Beryllium-7 as a tracer for short-term sediment dynamics in the Scarpe River, France. In: *Proceedings of the Tenth International Symposium on River Sedimentation*. Effects of river sediments and channel processes on social, economic and environmental safety V:349-358, Moscow State University, Russia

671 Zebracki M (2008) Devenir des polluants métalliques associés aux sédiments contaminés dans un
1 cours d'eau en relation avec la dynamique sédimentaire. Ph.D. Thesis, University of Paris-Sud
2 672
3
4 673 XI, France
5
6 674 Zoumis T, Schmidt A, Grigorova L, Calmano W (2001) Contaminants in sediments: remobilisation and
7
8 675 demobilisation. Sci Total Environ 266:195-202
9

10 676

677 **Table captions**

1 678 Table 1: Depth distribution of ^7Be activity (Bq kg^{-1}) in sediment collected in April 2005 (sampling
2
3 679 campaign n°1 on April 19, 2005).
4 680

5 681 Table 2: From April 2005 to August 2006: Inventory of ^7Be (mBq cm^{-2}) in top and bottom sediment
6
7 682 layers (0–3 and 0–5 cm layers) collected in triplicate (a, b and c). Three sediment layers were not
8
9 683 analysed due to the loss of samples during subsampling procedure (“n.d.” is mentioned). Activities in
10 684 ^7Be lower than the limits of quantification were estimated to be equal to half of the limits of
11
12 685 quantification.
13 686

14
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
18 689 Deposition/Resuspension ($=D/RS$, in $\text{g cm}^{-2} \text{ mo}^{-1}$). When suspended sediment was not available (due
19
20 690 to the loss of sampling device), activity in ^7Be was assessed by calculating the mean activity
21 691 measured in suspended sediments collected during the same period at the other sites (mentioned in
22
23 692 grey).
24 693

25 694 Table 4: Metal contents in suspended sediments ($\mu\text{g g}^{-1}$) collected from April 2005 to August 2006
26
27 695 (standard analytical deviation did not exceed 5 %). When suspended sediment was not available (due
28
29 696 to the loss of sampling device), metal content was assessed by calculating the mean content
30 697 measured in suspended sediments collected at the same site during the other sampling periods
31
32 698 (mentioned in grey).
33 699

34
35 700 Table 5: From April 2005 to August 2006: Cumulated fluxes of contaminant deposition and
36 701 resuspension, and Net fluxes of contaminant ($\mu\text{g cm}^{-2}$). Proportions of Total resuspension regarding
37
38 702 Total deposition (%).
39 703
40
41 704

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" ($\text{m}^3 \text{s}^{-1}$) 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 ^7Be activity of suspended sediment (Bq kg^{-1}) and monthly-cumulated rainfall (mm) between successive sampling campaigns.

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

Fig. 5: Monthly sediment Deposition/Resuspension ($=D/RS$) rates ($\text{g cm}^{-2} \text{mo}^{-1}$) 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 ($\mu\text{g cm}^{-2}$) for the 3 sampling sites.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5a

Fig. 5b

Fig. 5c

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			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			
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 ($\mu\text{g cm}^{-2}$)		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

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 , $^{210}\text{Pb}_{\text{xs}}$ and ^{232}Th (Bq kg^{-1}), and $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$ activity ratio in suspended sediment. Areal activities in ^7Be 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 ($<63 \mu\text{m}$, in %); ^7Be activities (Bq kg^{-1}); metal contents (Cu and Pb in $\mu\text{g g}^{-1}$, Cd and Zn in mg g^{-1}).

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

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

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
		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	06/16/05	0–3 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
		3–5 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
		0–3 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
4	07/06/05	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
		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
12	04/26/06	0–3 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
		3–5 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
		0–3 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

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	%					16					19	32	86	41
	k16	sediment						18		18						

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