Using in-situ radionuclides to model sediment transfer at the flow event time scale

P. Laguionie[1], A. Crave[2], P. Bonté[1] & I. Lefèvre[1]
[1]*Laboratoire des Sciences du Climat et de l'Environnement, CEA-CNRS, France*
[2]*Géosciences Rennes, CNRS, France*

Abstract

Sediment flow modeling over geological time scales requires a good understanding of the sediment transport modes occurring in rivers. Mean sediment residence time within hydraulic networks in response to flow events needs to be determined. In this background, the fallout natural environment radionuclide beryllium-7 (^{7}Be, half-life of 53.3 days) and the in-situ artificial radionuclide iodine-131 (^{131}I, half-life of 8.05 days) provide reliable tools to better understand the sediment transfer at the flow event time scale. Our model is based on the radioactive decay of the ^{7}Be and ^{131}I associated with suspended sediments. The short half-lives of ^{7}Be and ^{131}I allow phenomena to be studied at the flow event time scale. In this context radionuclide inputs have to be well-identified. The artificial iodine ^{131}I source is well-defined as this radionuclide most likely originates from hospitals where it is used in radiotherapy. In the case of ^{7}Be, the possibility that cities are radionuclide traps as they may provide high radionuclide concentration to channels, at time t, compared with bank inputs, is discussed. Natural environment radionuclide potentiality to estimate mean sediment residence time in a river section is demonstrated from in-situ data.
Keywords: in-situ radionuclides, short-time scales, sediments, transfer model.

1 Introduction

Sediment transfer models in river usually consider a relief at the equilibrium state, whereas they are established from local erosion-sedimentation processes involved at short time scales. Thus, it is of primary importance to determine the

morphogenesis events which links the two time scales. In rivers, it is common that most of the annual suspended sediments are transported during a few large runoff events. These transported sediments can either rapidly pass through the river network to the catchment outlet, or settle in the channel before to be remobilized during following events. Several events may be required by sediments to reach the catchment outlet. Therefore, it is necessary to resort to in-situ measurement methods at the flow event time scale. Nowadays, these methods are confronted with three main metrological limits: (i) bedload measurement; (ii) sampling frequency which has to be representative of the transport phenomenon time scales; and (iii) location of data collection. Issued data uncertainties are at the origin of error propagations in spatial and temporal integrations of numerical models. That is why an in-situ, *i.e.* non-numerically, integration method is required when phenomena kinetics are both space- and time-scale dependent. There is no more need to distinguish bedload, saltation and suspended load, and sediment motionless periods are naturally taken into account. The use of sediment-associated time sensitive environmental tracers becomes necessary. Both natural and artificial environmental radionuclides afford a means of tracing sediment. Their radio-chronometric properties allow dealing with large space and time scale ranges, which agrees with sediment transport process scales. Environmental radionuclides have often been used to evaluate particulate transport processes in the environment Walling and Woodward [1] and they have the advantage to be worldwide present in environmental systems.

In theses environmental systems, radionuclides are shared between both the dissolved and particulate phases. At the equilibrium state, the radioactivity ratio between these two phases is named the partition coefficient K_d (activity per kilogram of particles over activity per liter of water after filtration at 0.45 µm). Theoretically, K_d model should be based on the thermodynamic chemical equilibrium Baik *et al.* [2]. Practically, it is used as a black box model and becomes an empirical parameter. In rivers, radionuclide adsorption and desorption kinetics are not negligible compared to the hydrodynamic process time scales. So, the partition coefficient has to be interpreted as a time-dependent variable $k_d(t)$. This is the main difference with the historically first use of radionuclides in estuarial, coastal and oceanic environments.

To measure sediment radioactivity, high-resolution gamma-spectrometry provides a non-destructive, multi-elemental and less time-consuming technique. There were recent advances in the use of the environmental radionuclides caesium-137 (half-life 30.07 years) and unsupported lead-210 (half-life 22.3 years) to quantify medium-term rates of erosion and sediment accumulation Blake *et al.* [3]. However, it still remains a need of exploring the potentiality of using other shorter-lived radionuclides to provide evidence of sediment mobilization, transport and storage over shorter time scales, particularly for individual events Blake *et al.* [3]. Beryllium-7 ([7]Be, half-life 53.3 days) and iodine-131 (^{131}I, half-life 8.05 days) with their shorter half-lives offer considerable potential as short-term tracers. The restrictive criteria to use these gamma-ray emitter radionuclides as sediment tracers are: (i) half-life sensitive to

the hydrosedimentary process time scale; (ii) presence in the aquatic environments; (iii) known inputs in the water system; and (iv) radioactivity superior to the gamma-spectrometer detection threshold.
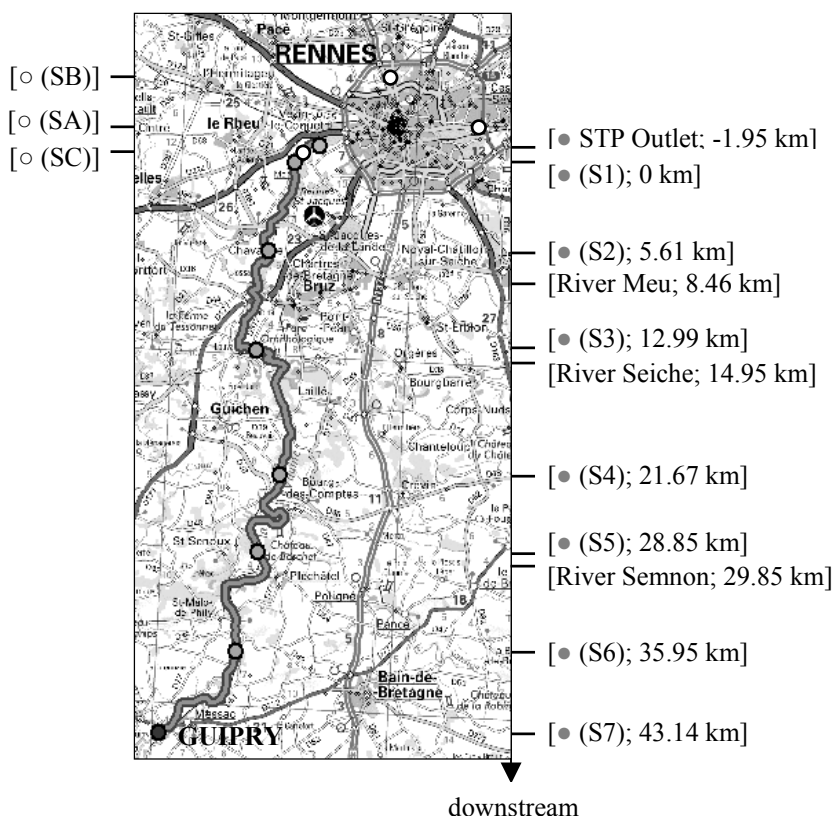

Figure 1:   Samplers, sewage treatment plant (STP) outlet and main tributaries locations in the River Vilaine section from Rennes to Guipry.

[7]Be was already successfully used in former studies to investigate the residence and settling time of particles in lakes, coastal waters Olsen *et al.* [4], and rivers Bonté *et al.* [5], Bonniwell *et al.* [6]. [7]Be is naturally produced in the upper atmosphere and diffuses through the atmosphere until it attaches to aerosols. [7]Be is supplied to a river section by both direct wet plus dry atmospheric deposition and particulate plus dissolved upstream cross-section inputs. The atmospheric deposition at the day-time scale is mainly governed by rainfall scavenging Wallbrink and Murray [7]. High K_d values show the [7]Be prevalent association to the particulate phase.

Concerning ^{131}I, it originates from hospitals where it is used in radiotherapy, research facilities, licensed manufacturers, and excreta from patients treated with

radioactive pharmaceuticals. Next, it enters municipal sewer systems through wastewater discharges. Originally in a dilute form, ^{131}I may become concentrated in the sludge solids during wastewater treatment. In rivers, ^{131}I is supposed to be essentially present in the dissolved phase Bonté *et al.* [5]. Due to its short half-life, over 90% of the ^{131}I decays away within 30 days. There is clearly a lack of data about ^{131}I radioactivity in river sediments.

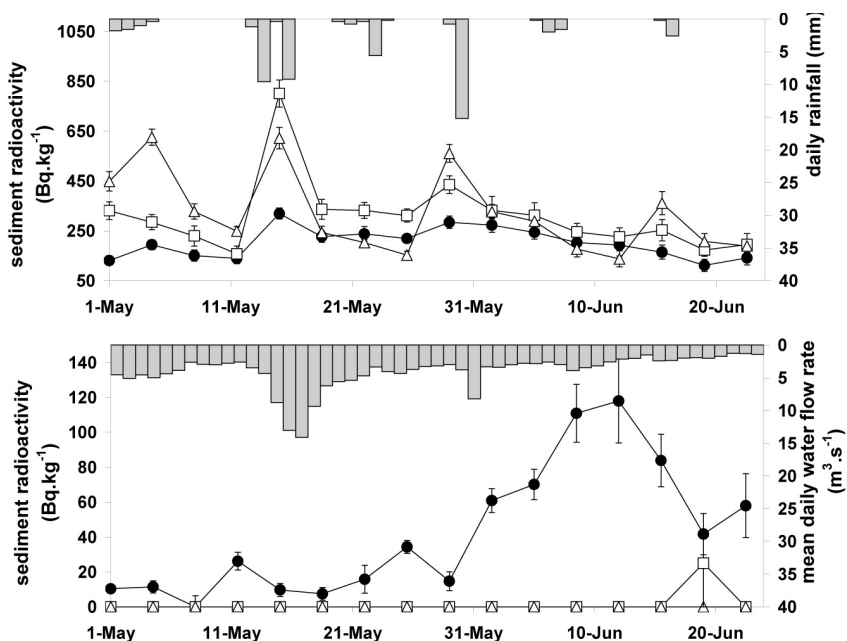Figure 2:   Results from sampling campaign 1; (up) [7]Be and (down) [131]I sediment radioactivity; ● in the river Vilaine downstream Rennes (SC), □ in the river Vilaine upstream Rennes (SA), î in the river Ille upstream Rennes (SB).

2   Study area

River Vilaine catchment is located in the Region of Brittany in the North West of France, and characterized by low gradients and predominantly agricultural land use. The drained 10 400 km² elevate from 0 to 300 m above the sea level and receive an average annual rainfall around 800 mm. The river Vilaine is schist-, sandstone- and mud-bed river. The river section under interest is located between Rennes which is an urban area of around 200 000 inhabitants, and Guipry, a small village located 45 km downstream Rennes, fig. 1. At Rennes and Guipry, the daily mean water flows are respectively 11 and 27 $m^3.s^{-1}$ in average and up to 183 and 491 $m^3.s^{-1}$ during flood events. The three main tributaries along this river section are rivers Meu, Seiche and Semnon.

3   Methods

Due to the rainfall deficiency since 2001, the mean water flow rate recorded at Rennes gauging station linearly decreased from 2001 to 2005. However, the river Vilaine high suspended sediment concentrations during low water flow rates allow setting up an experiment methodology with: (i) a permanent hydraulic regime; and (ii) a homogeneous material made of detritus silica and organic material. This is a great advantage since particle radioactivity is size and mineralogy dependent He and Walling [8]. The mean particle diameter d_{50} is 9.1±5 µm (average of 58 particle size analysis on samples collected from Rennes to Guipry between March and June 2005; the particle sizes were measured using a laser diffraction granulometer, after removal of the organic fraction, and chemical dispersion).

3.1 Sampling strategy

Two sampling campaigns (1 and 2) were carried out: (i) 04/29-06/24/2005 to follow in time sediment radioactivity inputs into Rennes-Guipry river section; and (ii) 06/21-06/25/2005 to follow in space, from (S1) to (S7), fig. 1, the sediment radioactivity linear distribution. During the sampling campaign 1, three automatic samplers of the Rennes sewage treatment plant were used to collect sediment samples (SC), (SA) and (SB), respectively at 500 m upstream (S1), and in the Rivers Vilaine and Ille both upstream Rennes (note that Ille and Vilaine confluence is located in Rennes city center), fig. 1. Each sample results of the addition of sub-samples collected every 15 minutes during alternatively 3 and 4 days (from Tuesday to Friday and from Friday to Tuesday). Dates reported in results correspond to each sampling period mean date, fig. 2. Sampling points of campaign 2 are shown in fig. 1. Given its location relative to the treatment plant outlet, the first sampling point (S1) can be taken as the background point. The other points are sequentially labeled (S2) to (S7) along the River Vilaine. Each sampler consists in 3 horizontal bottles fixed on a vertical support from 0.25 to 0.61 m from the river bottom. Supports were directly laid on the river bottom by a diver in cross-section areas with the highest water velocities. Time-integrative suspended sediment samplings occurred during 94±3 hours. More than 4 g (dried weight) of sample were collected at each sampling point. Turbidity profiles and maximum river cross-section heights were recorded (06/22-06/23/2005) by an in-situ multi-parameter Troll 9000 equipped with a pressure sensor. Samples of campaigns 1 and 2 were freeze-dried during 24 hours and then sub-sampled or directly put in tightly closed plastic boxes for gamma counting. Meteorological information was recorded from 04/29 to 05/25/2006 to check there was no major [7]Be atmospheric deposition onto the catchment during the sampling period, and water flow rates were retrieved from the French DIREN database to check the permanence of the hydraulic regime.

Adsorption kinetics of water radioactivity ([7]Be in experiment k_1 and ^{131}I in experiment k_2) by sediments was investigated in two different contexts. Experiment k_1 (10/19-10/28/2004): 9 samples of non-radioactive clay sediment (10

g) were resuspended in rain water volumes (30 l) for different periods of time ranging from 70 min to 9.4 days. Experiment k_2 (01/14-01/19/2006): 6 samples of non-radioactive mud sediment (10 g) and 6 samples of non-radioactive silica sediment (10 g) were resuspended in water volumes (20 l) collected at the sewage treatment plant outlet for different periods of time ranging from 135 min to 5 days. Samples of experiment k_1 and k_2 were periodically manually shaken during the required contact times. After a given settling time, sediments were collected back by siphoning and centrifugation, freeze-dried during 24 hours and then sub-sampled or directly put in tightly closed plastic boxes for gamma counting.

3.2 Gamma spectrometry

Coaxial N-type HPGe detectors were used for gamma-radioactivity measurements (8192 channels, low background). Efficiencies and backgrounds are periodically controlled with an internal sediment standard. A minimum data collection period of 50 000 s was sufficient to provide good statistics. Due to the short half-lives of the radionuclides used, sample radioactivities were decay corrected back from the collection period to the counting starting date.

4 Results and discussion

4.1 Inputs

Rainfall was particularly low over the sampling period and caused low water flow rates, fig. 2. Consequently, a steady-state hydraulic regime can be assumed, especially during the 25 last days with a mean water flow of 2.4 ± 1.5 m^3.s^{-1}. Fig. 2 clearly shows a correlation between ^{7}Be concentration fluctuations over the sampling period at the three sampling stations (SA), (SB) and (SC), and rainfall events. Whereas time-punctual high concentrations were expected downstream Rennes urban area (SC), the highest concentrations occur in Rivers Ille (SB) and Vilaine (SA) upstream Rennes city. This can be explained by the Rennes drainage system which stocks urban area runoff water consecutively to a rainfall event and rejects it to river Vilaine after treatment during wet periods. For the last 25 days, radioactivity ratio between the dissolved and particulate phases can be considered at the equilibrium state as there is no significant rainfall event during this long period of time.

In the case of ^{131}I, there is clearly no relation between its concentration and mean daily water flow even if sewage treatment plant discharge can represent up to a third of the total Vilaine water flow, fig. 2. The only ^{131}I source is confirmed to be the sewage treatment plant. However, one exception is the upstream Rennes station (SA) non-null value (06/19/2006), which shows that time-punctual uncontrolled rejects to river may also occur. Inputs to Rennes-Guipry river section are characterized by a large pick of ^{131}I concentrations spread over the last 25 days.

4.2 Adsorption kinetics of water radioactivity by sediment

Results of experiments k_1 and k_2 are plotted in figure 3. With ^{7}Be, a time-dependent enrichment ratio, r, can be defined as the ratio of the radionuclide

concentration in sediments at a t time to the radionuclide concentration in sediments at the infinity time, *i.e.* when the equilibrium between the solid and dissolved phase radioactivity is reached. From data plotted in figure 3, the enrichment ratio may be modeled, under the assumption of equilibrium reached, as follow:
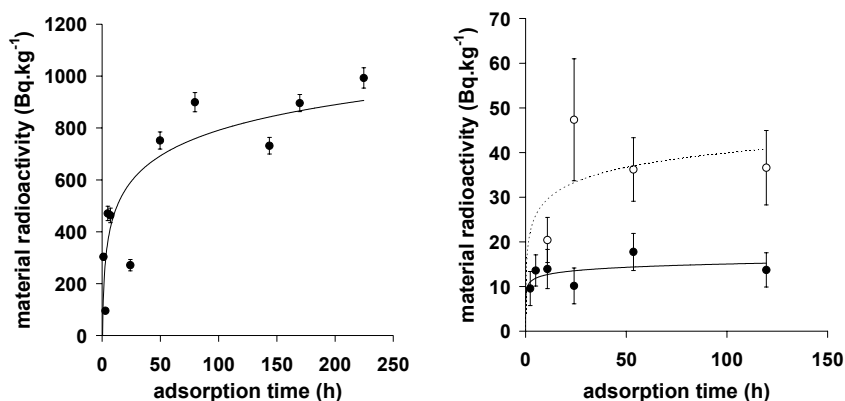
$$r = 142 \ln t + 137 \qquad (1)$$

Figure 3:   Results from experiment k_1 (left): adsorption of ^{7}Be by clay; results from experiment k_2 (right): adsorption of ^{131}I by ● silica and ○ mud; the smooth curves are least-squares fitted results for the logarithmic function; the precision on radioactivity values is provided at the 95% (left) and 68.3% (right) level of confidence.

With ^{131}I, adsorption kinetics is less obvious to analyze, fig. 3. The equilibrium state seems to be reached rapidly. However, the infinity time is difficult to experiment due to the ^{131}I short half-life of 8.05 days.

4.3 Mean sediment residence time

In a one-dimensional scheme, sediment radioactivity along river section is given by eqn. (2):

$$A_i(x) = \sum_j A_{i,0}^j \exp\left(-\frac{\lambda_i x^j}{V_s}\right) \qquad (2)$$

where λ_i (s^{-1}) is the i-radionuclide constant decay, V_s (m.s^{-1}) the mean sediment velocity, and $A_{i,0}^j$ and $A_i^j(x)$ (Bq.kg^{-1}) respectively the sediment radioactivity through a j-numbered source cross-section (tributaries or upstream cross-section) and at a x^j-distance (m) from it.

Table 1:   Physico-Geochemical parameters recorded from (S1) to (S7); suspended sediment concentrations are determined from multi-parameter Troll 9000 laboratory calibration with in-situ sediment; sediment concentrations are mean values calculated over sampler height (0.25 to 0.61 m from the river bottom).

Location	^{131}I concentration (Bq.kg^{-1})	^{7}Be concentration (Bq.kg^{-1})	Maximum water height (m)	Suspended sediment concentration (mg.l^{-1})
(S1)	58±5	118±18	2.21	20±11
(S2)	24±2	100±12	1.70	20±5
(S3)	Sampler turned upside down			
(S4)	13±3	93±15	1.76	11±4
(S5)	10±2	94±13	2.25	8±2
(S6)	Sampler turned upside down			
(S7)	3±3	97±21	2.08	8±2

In figure 4, the fit of eqn. (2) with (S1) and (S2) radioactivity values, table 1, gives almost the same mean sediment velocities: 5.4 mm.s^{-1} with ^{7}Be data and 6.8 mm.s^{-1} with ^{131}I data. Next, eqn. (2) fits well with (S4), (S5) and (S7) ^{131}I radioactivity data, which gives a mean sediment velocity of 18 mm.s^{-1} in the downstream part of the river section. This means that tributaries Meu and Seiche increase water velocities in the river Vilaine section, whereas tributary Semnon does not bring any transport energy to the system. This result agrees well with hydraulic data analysis. Indeed, over the 25 last days of the sampling period 1, the mean water discharge at Guipry can be shared as follow: 74% from the upstream cross-section, 11% from tributary Meu, 12% from tributary Seiche and 3% from tributary Semnon, whereas the maximum water height remains almost unchanged, table 1.

Whereas the only source of ^{131}I originates from the upstream cross-section, ^{7}Be can also comes from tributaries, which explains the ^{7}Be concentration increasing in samples (S4), (S5) and (S7). This can be modeled considering a null-sediment concentration in tributary water flow, *i.e.* the dissolved phase alone is radioactive. This hypothesis is valid from the suspended sediment concentration gap between (S2) and (S4), table 1. Then, downstream from tributaries, the total sediment radioactivity results from the addition of the sediment radioactivity expected under the hypothesis of no tributary with the time-dependant sediment enrichment due to the sudden dissolved phase radioactivity increase. The enrichment ratio is modeled by eqn. (1). The unknown parameter is the radionuclide concentration in the tributary dissolved phase. Considering the same radionuclide concentration in the different tributary dissolved phases, this parameter becomes proportional to the tributary water discharge. Tributaries Meu and Seiche were artificially added to form a single tributary at 14.95 km downstream from (S1). With the mean sediment velocity of 18 mm.s^{-1} previously obtained from ^{131}I data, ratio of Meu plus Seiche over

Semnon dissolved phase radioactivity is equal to 0.38. This value and the water flow ratio 0.13 are of the same order of magnitude in regard to assumptions made.
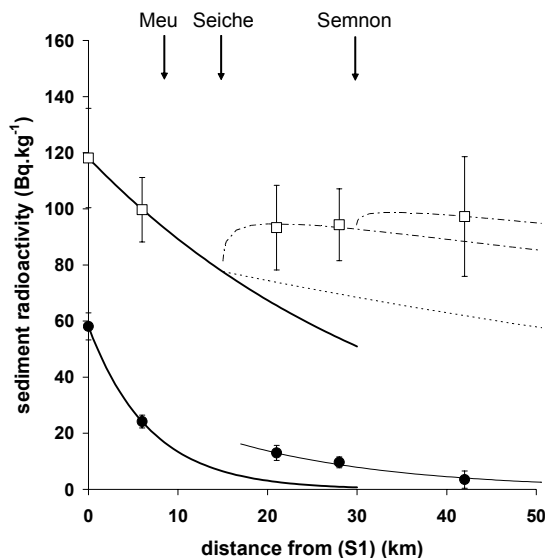

Figure 4:   Sediment concentration of samples (S1), (S2), (S4), (S5) and (S7) in □ ^{7}Be and ● ^{131}I; the full-line smooth curves are least-squares fitted results for the eqn. (2); the dotted-line curve is the expected sediment radioactivity in ^{7}Be without any tributary; the broken-line curves are fitted results taken into account the three main tributaries (Meu, Seiche and Semnon); the precision on radioactivity values is provided at the 68.3% level of confidence.

    As a result, sediments spend 32 days in the first 14.95 km of the river section and 9.6 days in the following 14.9 km. Over this time period, the steady-state hydraulic regime can be assumed, fig. 2. Due to radioactivity input variability, fig. 2, the mean sediment velocity is overestimated towards Guipry with ^{7}Be data, and in the first third of the section with ^{131}I data.

5   Conclusions

The results presented in this paper confirm the potential for using ^{131}I and ^{7}Be as tracers in river sediment transfer investigations at the flow event time scale. The use of in-situ ^{7}Be (half-life of 53.3 days) and artificial radionuclide ^{131}I (half-life of 8.05 days) was investigated in favourable conditions: (i) a permanent hydraulic regime; (ii) a homogeneous material in suspension. Whereas high time-punctual concentrations were expected downstream Rennes urban area, a sampling campaign showed that Rennes drainage system regulates ^{7}Be inputs in river section. Sewage treatment plant outlet was confirmed to provide a well-

identified source of ^{131}I into the system, which is helpful to discuss [7]Be data. A simple one-dimensional model bases on the ^{131}I and [7]Be sediment concentration analysis was proposed to estimate mean sediment velocity, and thus mean sediment residence time in a river section. Its main particularity is to take into account tributary [7]Be inputs and adsorption kinetics of dissolved [7]Be by sediments.

Acknowledgements

References

[1] Walling, D.E. and Woodward, J.C., Use of radiometric fingerprints to derive information on suspended sediment sources. *Proc. On the Oslo symposium on erosion and sediment transport monitoring programmes in river basins*, I.A.H.S. Publication, 210, pp. 153-164, 1992.

[2] Baik, M.H., Cho, W.J. and Hahn, P.S., Sorption of U(VI) onto granite surfaces: a kinetic approach. *Journal of Radioanalytical and Nuclear Chemistry*, **260(3)**, pp. 495-502, 2004.

[3] Blake, W.H., Walling, D.E. and He, Q., Using cosmogenic beryllium-7 as a tracer in sediment budget investigations. *Geografiska Annaler*, **84A(2)**, pp. 89-101, 2002.

[4] Olsen, C.R., Larsen, I.L., Lowry, P.D., Cutshall, N.H. and Nichols, M.M., Geochemistry and deposition of [7]Be in river estuarine and coastal waters. *Journal of Geophysical Research*, **91**, pp. 896-908, 1986.

[5] Bonté, P., Mouchel, J.M., Thomas, A.J., Le Cloarec, M.F., Dumoulin, J.P., Sogon, S. and Tessier, L., 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)**, pp. 339-355, 2000.

[6] Bonniwell, E.C., Matisoff, G. and Whiting, P.J., Determining the times and distances of particle transit in a mountain stream using fallout radionuclides. *Geomorphology*, **27**, pp. 75-92, 1999.

[7] Wallbrink, P.J. and Murray, A.S., Fallout of [7]Be in south Eastern Australia. *Journal of Environmental Radioactivity*, **25**, pp. 213-228, 1994.

[8] He, Q. and Walling, D.E., Interpreting particle size effects in the adsorption of ^{137}Cs and unsupported ^{210}Pb by mineral soils and sediments. *Journal of Environmental Radioactivity*, **30(2)**, pp. 117-137, 1996.