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Buffering of suspended sediment transport in lowland river during low water stages: quantification in river Seine using environmental radionuclides

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ABSTRACT

This study was undertaken to test the application of environmental radioactive tracers for estimating sediment mass and sediment residence time in rivers. A continuous sampling of the Seine river suspended matter (SM) using sediment traps was made during two months, between Paris and the estuary, along a 120 km long river section. The hydrological regime corresponded to the low water stage, where the SM transport is reduced. The measured tracers in the SM include short-lived natural (^{7}Be, ^{234}Th$_{xs}$) and artificial (^{131}I) radionuclides, as well as the longer-lived natural ^{210}Pb$_{xs}$ and its descendant the ^{210}Po. ^{137}Cs was used to check grain-size effects. A simple steady state model allowed us to estimate the total sediment mass, i.e. the SM, plus the resuspendable matter (RM), and the sediment residence time. Despite their different half-lives (8 to 53 days) and their different geochemical properties, consistent results were obtained with ^{131}I, ^{7}Be and ^{234}Th$_{xs}$. The best estimate of the sediment mass present in the river is (24-41)·10^{3} tons; it is essentially composed of the RM which is 10-17 times more abundant than the SM. In these hydrological conditions, the sediment residence time is quite long (1.6-2.8 months).

Key words: Suspended sediment. Residence time. Seine river. Radionuclides.

RESUMEN

En este estudio se trata de investigar la aplicación de los trazadores radioactivos ambientales para evaluar la masa y el tiempo de residencia de los sedimentos en el río Sena. Los sólidos en suspensión del río Sena han sido monitorizados en continuo utilizando trampas durante dos meses, entre la ciudad de París y el estuario (120 km). El régimen hidrológico corresponde a un período de aguas bajas, con reducida concentración de sedimentos en suspensión. Los trazadores medidos en los sedimentos incluyen radioisótopos con períodos radioactivos cortos, naturales (^{7}Be, ^{234}Th$_{xs}$), y artificiales (^{131}I), así como el ^{210}Pb natural y su descendiente el ^{210}Po. El ^{137}Cs fue utilizado para considerar el efecto de medida de las partículas. Un modelo simple nos permite evaluar la carga sedimentaria total, es decir los sedimentos en suspensión más los sedimentos resuspendidos, y sus tiempos de residencia. A pesar de diferencias de períodos radioactivos (entre 8 y 53 días) y de propiedades geoquímicas diferentes, se obtuvieron resultados válidos para el ^{131}I, el ^{7}Be y el ^{234}Th.

La carga sedimentaria del río Sena (24-41.10^3 t) está, en su mayoría, compuesta por sedimentos resuspendidos, que son 10-17 veces más abundantes que los sedimentos en suspensión. Para esas condiciones hidrológicas, el tiempo de residencia de los sedimentos en suspensión parece bastante largo (1.6-2.8 meses).

INTRODUCTION

River particles

River particulate fluxes to the sea include terrigenous, biogenic and anthropic particles. Since bottom load in rivers is mostly composed of sandy particles, the finer particles are mostly transported seaward as suspended matter (SM). However, they may be only temporarily suspended and spend significant amounts of time in layers of easily resuspendable matter (RM) which may accumulate over "permanent" sediment deposits. SM fluxes can be evaluated through direct sampling in the water column, but RM stocks, as well as exchange occurring between SM and RM, are much more difficult to quantify directly because of the questionable efficiency of current sampling procedures. In large rivers, resuspension obviously occurs during floods, but also during the low water stage where less strong but still significant erosional energy sources can be found in navigation and windy events in addition to the lower water shear over the bottom.

Both these particulate stocks usually content a large proportion of clay sized or organic material with a high specific surface area, so that a large solid surface is available in river water for trace element sorption, even with low SM concentrations (Millward et al., 1989; Garnier et al., 1991, 1993). It results that the river solid discharge can often be considered as a major pathway of the labile fraction of natural trace elements and of many particle - reactive pollutants. These chemical fluxes can be altered by "ageing" processes when particles remain a long time in depositional environments. "Ageing" processes affect very significantly both the distribution in time of chemicals and pollutants fluxes at the river basin outlet, and the extent of biological and chemical processes acting on the fate of pollutants. The mean age of a pool of particles leaving a given river section is a complex function of SM advection and RM deposition / resuspension processes, while individual particles generally have different ages inside the same pool.

The available tracers

Time sensitive tracers are required for assessing the global age of a particulate matter pool, and radionuclides can be used in this purpose. Environmental radionuclides have often been used to evaluate particulate transport processes in the environment (Walling and Woodward, 1992). Three basic methodologies have been proposed:

a) a well identified input (typically bomb-derived ^{137}Cs) can be used to follow the fate of particles presumably irreversibly marked by the tracer ;
b) the activity of a decaying substance on particles with an initially known concentration can be used to date the particles (typical application is ^{210}Pb in sediments under the constant surface concentration hypothesis) ;
c) the (hypothesised) balance of a decaying substance is used to identify a lacking input or output flux of particulate matter in the system (typically the use of short lived radionuclides inventory in sediments to evaluate sedimentation rates).

Few studies have so far considered the radioactivity of the Seine river (Jeandel et al., 1981a, 1981b; Thomas, 1988; Sogon, 1999), most of them were focussed of the Seine estuary where an important question was the tracing of marine sediment inputs by radionuclides issued from the La Hague nuclear fuel reprocessing plant (Thomas, 1988; Boust, 1998, 1999). In this study we explore the possibilities of using environmental radionuclides to study short term particles transports in the Seine river downstream of Paris, on time scales of weeks to months. This study is mostly based on methodology (c) where the unknown term in the radionuclide balance is the decay of nuclides bound to SM and RM. Evaluating the decay term allows to compute the total amount of SM and RM whenever a reasonable estimate of nuclide activity per mass of SM or RM can be proposed.

Radioactive tracers must then satisfy several conditions : (i) a half-life of the same order of magnitude, or lower, than the expected particle transit time, (ii) a measurable stock in the water column in the particles , and (iii) known inputs in the water system. Among the various radionuclides occurring in measurable concentrations in the Seine particles, the following ones present a potential interest for tracing sediment fluxes.

The natural cosmogenic ^{7}Be (half life 53 days) has been used in studies of watersheds and aquatic environments (Olsen et al., 1985, 1986, Martin et al., 1986, Dominik et al., 1987, Mouchel, 1988, Thomas, 1988; Walling and Woodward, 1992, Wallbrink and Murray, 1996). It is supplied to the water either by direct atmospheric deposition (wet plus dry) and by upstream river inputs (particulate and dissolved). Its production rate shows a 11 years cycle due to solar activity variations. In the near-ground aerosol, air concentrations show a seasonal cycle, with a spring maximum due to mixing with stratospheric air through the tropopause. Its subsequent atmospheric deposition is mainly governed by rainfall scavenging. Thus, over a given river section, part of the ^{7}Be is directly supplied to the water surface from the atmosphere. ^{7}Be is also delivered by upstream river inputs, which also essentially result from direct deposition at the water surface, since deposition on soils is probably rapidly lost by decay. In French rivers, its distribution between the particulate and dissolved phases may be characterised by an average distribution coefficient K_D (activity per kg of particle / activity per litre of water, after filtration at 0.4 µm) close to $5 \cdot 10^4$, showing its prevalent association to the particulate phase (Thomas, 1988).

Natural radionuclides from the U and Th decay series have long be used in aquatic environments. For instance, Olley et al. (1997) have used the ^{232}Th decay series to determine sediment residence times. Other useful tracers belong to the ^{238}U decay series; the radioactive decay products of this radionuclide which are cited in this study are:

$$^{238}\mathrm{U} \rightarrow {}^{234}\mathrm{Th} \rightarrow {}^{222}\mathrm{Rn} \rightarrow {}^{214}\mathrm{Bi} \rightarrow {}^{210}\mathrm{Pb} \rightarrow {}^{210}\mathrm{Po}$$

(the end-member is the stable ^{206}Pb).

^{210}Pb (half life 22 years) is in secular equilibrium with the ^{238}U decay products. It is first supplied to rivers by detrital particles; this constitutes the supported ^{210}Pb activity. An additional source of ^{210}Pb (the excess, or unsupported, activity, noted $^{210}\mathrm{Pb}_{xs}$) is the atmosphere. Following ^{222}Rn emanation from rocks containing ^{238}U, ^{210}Pb is formed in the atmosphere and rapidly adsorbed on aerosol particles, which reach soils and the aquatic system through wet and dry atmospheric deposition. The excess activities are calculated by subtracting the supported activity from the total activity. The supported activity may be determined by measuring directly the ^{238}U activity by alpha spectrometry, but a more convenient method is to use a gamma emitter among the ^{238}U descendants, assuming secular equilibrium for this isotope.

Upstream river inputs also result from atmospheric deposition, with probably some additional contribution from soil erosion. Its K_D in French rivers is unknown, but is likely to be much higher than for ^{7}Be according to the well known high particulate affinity of lead.

The long ^{210}Pb half life is not directly suitable for tracing short term processes, but ^{210}Pb decays to the short lived ^{210}Po (half life 138 days) and the excess $^{210}\mathrm{Po}_{xs}/^{210}\mathrm{Pb}_{xs}$ activity ratio increases from zero to 1.02 when equilibrium is reached between these isotopes (95% of the equilibrium is attained after 1.5 years), so that variations of the $^{210}\mathrm{Po}_{xs}/^{210}\mathrm{Pb}_{xs}$ ratio are a potential tool for tracing short term sediment transport.

Another radionuclide from the ^{238}U decay series is the ^{234}Th (half life 24 days) which is highly particle-reactive in waters, contrary to uranium. Following its production from dissolved uranium, it is rapidly scavenged by sediment particles. As in the case of ^{210}Pb, some ^{234}Th is also contained in detrital minerals, and it is its excess ($^{234}\mathrm{Th}_{xs}$) which may be used for tracing particles.

In addition to these natural tracers, an artificial iodine nuclide (^{131}I, half life 8 days) is detected in the Seine sediment particles downstream Paris. It most likely originates from hospitals where ^{131}I is used in radiotherapy. Its biogeochemical behaviour is certainly very different from the natural radionuclides mentioned above, in particular because its K_D is probably much lower (Anonymous, 1985). Thus, this radionuclide is supposed to be essentially present in the dissolved phase.

Another radionuclide used in this study in the long-lived ^{137}Cs (half life 30 y) produced by thermonuclear bomb testing during the 1960's (and the Chernobyl fallout) and now trapped in surface soils, where its stock essentially decreases by decay, and to a much lesser extend by the transfer of eroded fine-grained particles to the rivers (Thomas, 1988; Martin et al., 1994). Since its activities in the river SM strongly depends on the amount of clay minerals, which is also true for most of the above radioactive tracers, we shall test its usefulness to correct the tracers concentration variations for grain-size effects.

All these tracers except ^{210}Po can be measured by gamma spectrometry, which is a simple analytical technique.

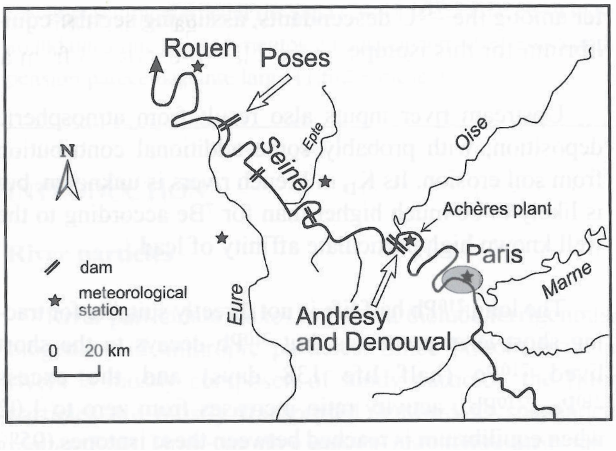

Figure 1: Location of sampling sites.

THE TRACER EXPERIMENT

Selection of an appropriate river section

The Seine river has a total drainage area of 78000 km^2 at Le Havre, an average flow of 435 m^3 s^{-1} and a solid discharge of 0.7 10^6 t y^{-1} (Meybeck et al., 1998), which gives a particle / water flux ratio of 51 mg l^{-1}, and a low sediment yield of about 10 t km^{-2} y^{-1}.

To evaluate particle transit time, a rather long river section is preferable to observe long transit times, and lateral inputs by tributaries, hardly measurable, should be as small as possible compared to upstream inputs. The lower course of the Seine river downstream Paris receives inputs at the confluence of the Oise river, and also by the Seine-Aval sewage treatment plant at Achères (Figure 1). Two upstream reference stations were chosen downstream these input sites, one on the left bank at Andrésy and the other on the right bank at Denouval. The downstream reference station was set upstream the dam at Poses, which is the physical boundary between the river and the estuary. In this 120 km long section, the river flows between embankments and receives only little tributaries, as shown by the drainage area at Andrésy which represents 93 % of the total drainage basin at Poses (68840 km^2) (Guerrini et al., 1998). This section can thus be viewed as a one-dimensional channel where the study of particle transport can be undertaken in favourable conditions.

Sampling

The sampling period (July to September) was chosen at the end of the 5 to 6 months long low water stage period occurring every year before the first flow increases in fall. In 1999, the sampling period was characterised by a typical summer low water stage, where particle transit time is expected to be the highest, with a small peak of discharge 2 weeks before the beginning of sampling, and a very small other one before collection of samples # 4. Just before collection of samples # 9, another small peak of discharge initiates the more rainy fall season. River discharge correlates the average rainfall over the river section, measured at 5 stations located between Paris and Rouen (Figure 1). For convenience, only the rainfall at Paris will be illustrated. The Seine river discharge at Poses is nearly equal to the sum of the flow from the Oise river and the Seine river at Paris, the latter representing about two thirds of the total discharge.

To ensure a continuous sampling and to obtain sample quantities allowing accurate spectrometric measurements, the SM was collected weekly by sediment traps, moored 2 to 3 metres away from unused wharves in order to ease the trap recovery process and avoid unrepresentative resuspension from banks. Trap locations have been carefully selected according to two criteria: a low velocity area

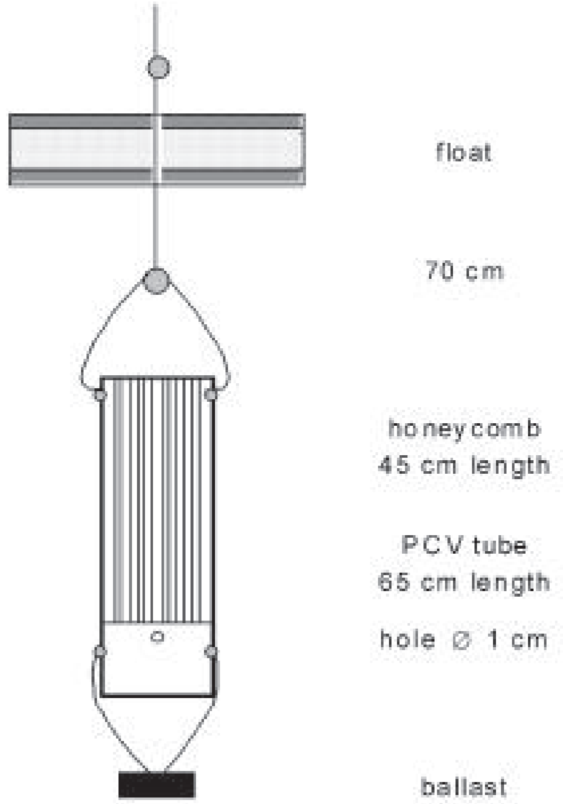

Figure 2. The suspended matter collector.

to improve particle settling in traps, and the best lateral homogeneity in the river.

The traps were built with PVC cylinders (20 cm diameter) closed at the lower extremity by a PVC cap (Figure 2). Inside the trap, a honeycomb structure allows particles retention and prevents losses by resuspension. A small hole communicates with the lower part of the cylinder; it is used to flush out the upper water column of trap content during retrieval. The traps are suspended at about 70 cm below the surface, between a ballast and a float which is maintained at about 1 m off the river bank. Three traps were lost during this collection period (Table 1). The SM content was determined by independent water sampling and filtration during trap retrieval.

In the laboratory, the trap contents were first sieved (200 µm nylon mesh) to remove coarse debris, and the remaining SM was recovered by centrifugation, freeze-dried and homogenised. Despite the low SM content (Table 1) , this technique allowed to obtain from each trap SM quantities of the order of 100-300 g dry weight.

Radionuclide concentration measurements

Dried sub-samples (60-80 g) were put in tightly closed plastic boxes for gamma counting. Coaxial HP Ge N-type detectors were used for gamma spectrometry (8000 channels, low background). Efficiencies and backgrounds are periodically controlled with an internal sediment standard, a mock-up of marine sediment with U and Th US standard from NBS at 1000 ppm (8.51 Bq g^{-1} for uranium series and 1.80 Bq g^{-1} for thorium series), pure KCl samples, and IAEA standards 135 and 375. Activities were decay corrected to the time of the collection period for which 50% of the counting was obtained, assuming a constant SM activity; this correction is important for short-lived isotopes such as ^{131}I.

In this study, the equilibrium activity of the ^{238}U series was determined by measuring a gamma emitter, the ^{214}Bi, which can easily be measured with a good accuracy inasmuch as ^{222}Rn does not escape from the sample. This has been ensured by allowing radon reequilibration in the sample container and making a second counting after 2 months. Then the $^{234}Th_{xs}$ and $^{210}Pb_{xs}$ were calculated by subtracting the ^{214}Bi activities.

For alpha spectrometry (^{210}Po), about 0.5 g of trapped solids were dissolved in a concentrated HNO_3-HF-$HClO_4$ mixture, in a sealed PTFE vessel, at 200°C (during about one week). Silica was evaporated as gaseous H_2SiF_6 at 105°C. Polonium isotopes were electro-deposited from a 0.5N HCl solution on a silver plate. Radiochemical yield was determined using a ^{208}Po standard solution provided by Amersham, UK. The homogeneity of our gamma and alpha measurements inside the ^{238}U series was checked using a Chinese loess-soil sample were the expected ^{210}Pb activity should be the same as the expected ^{210}Po activity. The result of 10 day counting of six ^{210}Po replicates using different detectors was 52 ± 1 Bq kg^{-1}, in good accordance with the value obtained by gamma spectrometry, 56 ± 1Bq kg^{-1}.

RESULTS

The radionuclide activities in the SM samples of the studied Seine river section are presented in Table 1.

The rather low ^{40}K concentrations (corresponding to 0.96-1.11 % K by weight) reflect the mixing of K-bearing detrital aluminosilicates with an abundant $CaCO_3$ fraction (10-30 %, Meybeck et al., 1998), and a rather high particulate organic matter content (11% COP).

In cultivated soils sampled in the Marne river watershed (Figure 1), the ^{137}Cs concentrations average 6 Bq kg^{-1} (Sogon et al., 1999). They are higher in the river SM collected in sediment traps in the Seine and Marne rivers. (3-17 Bq kg^{-1}) , probably because of a preferential transport with the finest soil fraction. In this study, the ^{137}Cs concentrations are comparable but more constant (Table 1, Figure 4). The plot in Figure 3 confirms that the concentrations vary with the 5-20 µm grain-size fraction, and that most of the ^{137}Cs variations may be ascribed to grain-size effects (i.e. differential settling of clay minerals) (Jeandel et al., 1981a, 1981b; He and Walling, 1996; Sogon, 1999). Since similar effects are expected for the other radioactive tracers, we shall normalise their concentrations to the ^{137}Cs content.

Comparison of ^{137}Cs in our 24 trap samples (9.5 ± 1.3 Bq kg^{-1}) with 12 SM samples collected in 1979-1985 near Paris and 20 km downstream Poses, in a large range of river flow, and separated by continuous flow centrifugation at 8000 rpm (9.0 ± 3.7 Bq kg^{-1}, corrected for decay to 1999), does not show evidence of a significant loss by our trap sampling technique of the fine-grained fraction responsible of the ^{137}Cs transport.

As compared to ^{137}Cs, the ^{7}Be concentrations are one order of magnitude higher, and more variable. Compari-

Table 1. Natural and artificial radionuclide concentrations in the SM trap samples in the Seine River.

	date of recovery	number of days	SM mg l^{-1}	^{40}K as % K	^{214}Bi	^{210}Pb	^{210}Pb$_{xs}$	^{234}Th	^{234}Th$_{xs}$	^{7}Be	^{131}I	^{137}Cs	^{210}Po$_{xs}$/^{210}Pb$_{xs}$
Andrésy													
1	19/07/99	6	12,3	1,00 ± 0,02	26 ± 0,5	124 ± 4	98 ± 4	41 ± 3	24 ± 3	275 ± 5	40 ± 1	9,3 ± 0,5	0,03 ± 0,03
2	26/07/99	7	11,8	1,00 ± 0,03	23 ± 0,6	118 ± 4	95 ± 4	41 ± 2	25 ± 3	240 ± 7	60 ± 1	9,3 ± 0,6	0,26 ± 0,06
3	04/08/99	9	9,0	1,06 ± 0,03	23 ± 0,4	108 ± 3	85 ± 3	44 ± 2	23 ± 2	195 ± 4	95 ± 1	9,5 ± 0,4	
4	11/08/99	7	10,1	0,97 ± 0,02	22 ± 0,4	132 ± 4	110 ± 4	32 ± 2	21 ± 3	440 ± 7	36 ± 2	9,3 ± 0,4	
5	19/08/99	8	10,0	0,99 ± 0,03	21 ± 0,6	108 ± 7	87 ± 7	37 ± 5	19 ± 6	348 ± 7	43 ± 1	10,0 ± 0,5	0,46 ± 0,07
6	27/08/99	8	17,2	1,03 ± 0,02	22 ± 0,4	119 ± 5	97 ± 5	42 ± 4	24 ± 5	280 ± 5	84 ± 1	9,8 ± 0,4	
7	02/09/99	6		0,98 ± 0,02	20 ± 0,4	114 ± 5	94 ± 5	43 ± 3	29 ± 4	213 ± 4	93 ± 1	9,4 ± 0,3	
8	10/09/99	8	3,6	0,99 ± 0,02	20 ± 0,4	115 ± 3	95 ± 3	42 ± 2	27 ± 3	215 ± 4	64 ± 1	9,1 ± 0,3	0,37 ± 0,04
9	21/09/99	11		0,91 ± 0,03	19 ± 0,5	167 ± 4	148 ± 4	33 ± 3	15 ± 3	450 ± 7	64 ± 1	9,5 ± 0,5	0,40 ± 0,02
Average			***10,6***	***0,99 ± 0,04***	***22 ± 2,1***	***123 ± 18***	***101 ± 19***	***39 ± 4***	***23 ± 4,2***	***295 ± 97***	***64 ± 22***	***9,5 ± 0,3***	***0,3 ± 0,2***
Denouval													
2	26/07/99	7	11,0	1,03 ± 0,03	23 ± 0,5	92 ± 3	69 ± 3	36 ± 3	16 ± 3	166 ± 4	73 ± 1	9,0 ± 0,4	
3	04/08/99	9	11,1	0,96 ± 0,05	19 ± 0,8	94 ± 2	75 ± 3	42 ± 4	27 ± 5	160 ± 2	88 ± 1	9,4 ± 0,2	
4	11/08/99	7	8,4	1,07 ± 0,04	24 ± 0,6	124 ± 4	100 ± 5	32 ± 3	14 ± 6	366 ± 8	41 ± 3	8,4 ± 0,5	
5	19/08/99	8	11,8	0,98 ± 0,03	21 ± 0,5	104 ± 3	83 ± 4	35 ± 2	21 ± 3	275 ± 3	30 ± 1	9,0 ± 0,2	
6	27/08/99	8	12,1	1,05 ± 0,04	24 ± 0,7	130 ± 5	106 ± 5	42 ± 4	18 ± 5	300 ± 7	70 ± 2	11,4 ± 0,6	
7	02/09/99	6		1,09 ± 0,04	23 ± 0,7	159 ± 9	136 ± 9	36 ± 4	27 ± 8	300 ± 7	120 ± 2	14,2 ± 0,8	
8	10/09/99	8	6,3	0,97 ± 0,04	24 ± 0,6	100 ± 4	76 ± 4	36 ± 3	14 ± 4	190 ± 6	66 ± 1	8,3 ± 0,5	0,65 ± 0,10
9	21/09/99	11		1,02 ± 0,03	27 ± 0,4	157 ± 3	130 ± 4			382 ± 9		11,3 ± 0,4	0,30 ± 0,02
Average			***10,1***	***1,02 ± 0,05***	***23 ± 2,4***	***120 ± 27***	***97 ± 26***	***37 ± 3,7***	***20 ± 5,6***	***267 ± 87***	***70 ± 30***	***10,1 ± 2,0***	***0,5 ± 0,2***
Poses													
1	19/07/99	6	28,7	1,01 ± 0,02	23 ± 0,3	80 ± 3	57 ± 3	42 ± 2	24 ± 3	91 ± 3	40 ± 1	8,3 ± 0,3	0,19 ± 0,07
2	26/07/99	7	26,1	0,96 ± 0,03	22 ± 1,0	88 ± 4	66 ± 5	45 ± 3	24 ± 4	76 ± 5	33 ± 3	8,2 ± 0,4	0,27 ± 0,07
5	19/08/99	8	21,0	0,97 ± 0,02	19 ± 0,4	92 ± 4	73 ± 5	35 ± 3	21 ± 4	145 ± 3	25 ± 1	8,6 ± 0,3	0,31 ± 0,06
6	27/08/99	8	24,3	1,04 ± 0,02	21 ± 0,5	110 ± 6	89 ± 4	45 ± 4	29 ± 5	148 ± 4	25 ± 1	9,8 ± 0,4	
7	02/09/99	6		1,11 ± 0,04	18 ± 0,7	106 ± 5	88 ± 5	45 ± 4	32 ± 5	140 ± 6	24 ± 1	10,5 ± 0,7	
8	10/09/99	8	23,3	1,10 ± 0,03	21 ± 0,4	88 ± 3	67 ± 3	42 ± 2	24 ± 3	99 ± 3	14 ± 1	8,2 ± 0,4	0,30 ± 0,04
9	21/09/99	11		0,99 ± 0,02	19 ± 0,4	99 ± 5	80 ± 5	39 ± 3	23 ± 4	160 ± 3	19 ± 1	8,0 ± 0,3	0,35 ± 0,06
Average			***24,7***	***1,03 ± 0,1***	***20 ± 1,8***	***95 ± 11***	***74 ± 12***	***42 ± 4***	***25 ± 3,8***	***123 ± 33***	***26 ± 9***	***8,8 ± 1,0***	***0,3 ± 0,1***

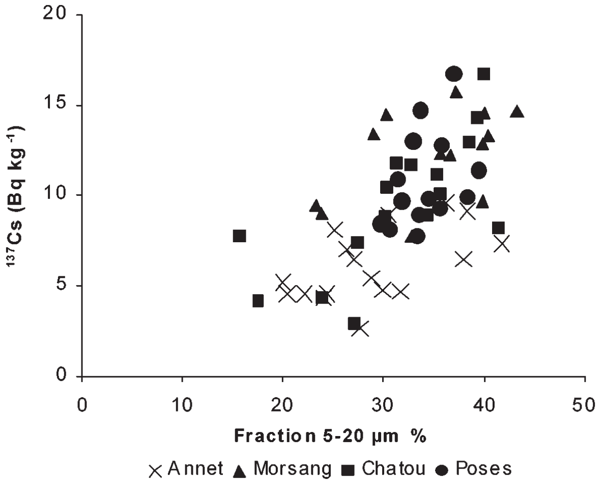

Figure 3. Variations of the ^{137}Cs concentrations with the abundance of the 5-20 µm fraction (trap samples). Marne river (Annet) and Seine river (Morsang, Chatou, Poses).

son of values at the upstream stations and at Poses clearly shows a downstream decrease (Figure 5). The average Andrésy/Poses and Denouval/Poses ratios are 2.4 and 2.1, respectively, ranging from 1.5 to 3.2.

^{131}I, which is present in all samples despite its short half-life, contributes significantly to the artificial radioactivity of the Seine SM. In each station, concentration variations are rather low. In most samples, the concentrations at Poses are lower than upstream since the average Andrésy/Poses and Denouval/Poses ratios, scattered between 1.0 and 4.7, average 2.8 and 3.2, respectively.

The concentrations of ^{214}Bi, total ^{234}Th, and to a lesser extent total ^{210}Pb, are quite constant in all samples. As compared to ^{214}Bi, the much higher total ^{210}Pb concentrations indicate for this radionuclide a large excess of atmospheric origin. The ^{210}Pb$_{xs}$ concentrations are similar in both upstream stations. On average they are slightly lower at Poses by a factor of about 0.7, but Figure 6 shows that this tendency suffers exceptions.

The distribution of the short-lived ^{210}Pb decay product, ^{210}Po$_{xs}$, is still incompletely documented, but the ^{210}Po$_{xs}$ / ^{210}Pb$_{xs}$ activity ratios are on the whole variable and quite low (Figure 7) and much lower than the equilibrium value (1.02). The evolution of these ratios with time is more regular at Poses than at the upstream stations, but no systematic changes between these two sets of data is visible. On the whole, despite some inconsistencies, the ratios seems to increase during the period of study by a factor of about 2.

The ^{234}Th$_{xs}$ concentrations are quite constant in all samples (19 ± 6 Bq kg^{-1}), without systematic differences between the three stations.

DISCUSSION

^{137}Cs

The ^{137}Cs concentrations (Figure 4) are quite constant with time at all stations. This indicates an overall homogeneity of the sediment stock downstream Paris during the low water stage. One exception is the high

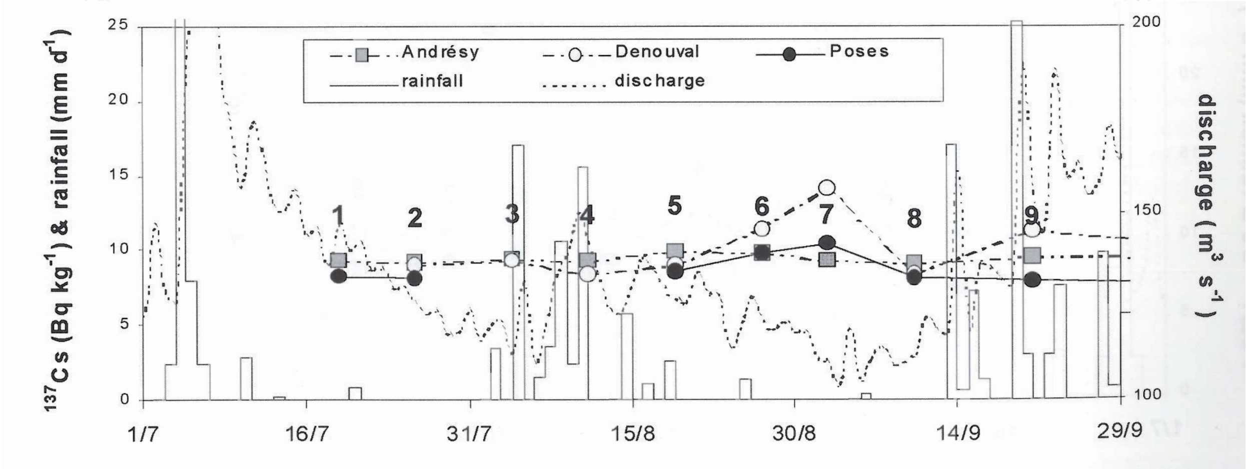

Figure 4. Distribution of ^{137}Cs concentrations in the SM trap samples. The Seine discharge is at Paris.

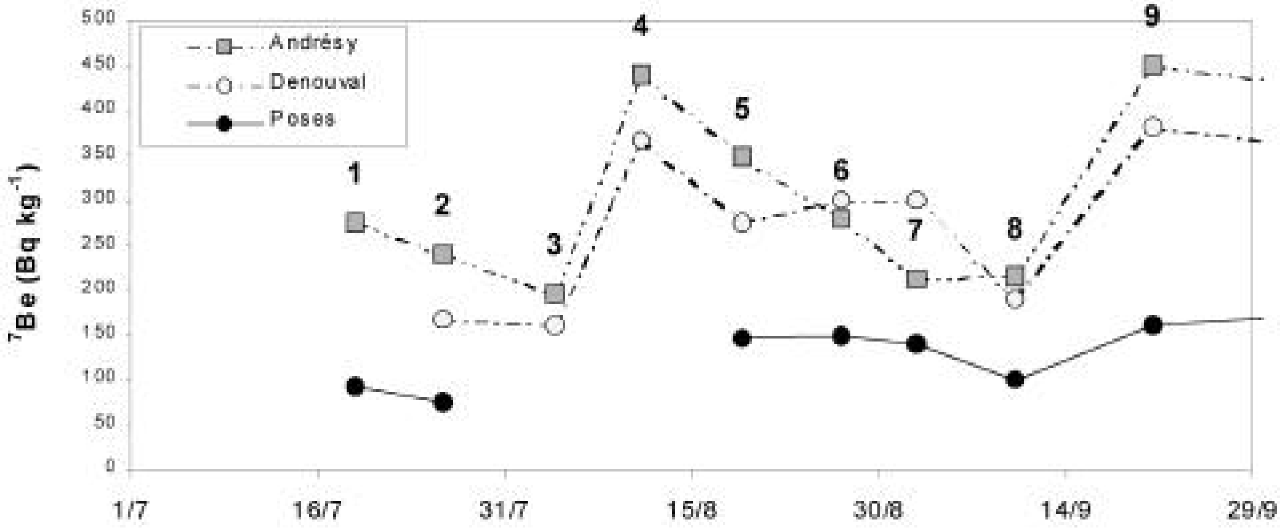

Figure 5. Distribution of ^{7}Be concentrations in the SM trap samples.

value (14.2 Bq kg^{-1}) in sample # 7 at Denouval, which also corresponds to high concentrations of other radionuclides, and is probably due to a more abundant clay fraction.

Short-lived tracers

A simple steady state model

The distribution of radionuclides in the river system is governed by inputs, outputs, and internal decay or production within the system. For the sake of simplicity, we model the river section as a simple system composed of three homogeneous compartments: the water volume between the upstream stations and Poses, the suspended particles, and the sediments stock. Exchanges include river inputs and outputs (particulate and dissolved), and atmospheric deposition. In a steady state approach, the tracer budget described in activity per unit time is: input = output + decay.

Several steps have to be followed in order to get an estimate of RM. Firstly, the decay term, which has to equilibrate the difference between inputs and outputs in a steady-state approach, allows to compute the total amount of radioisotope inside the system. Secondly, starting from

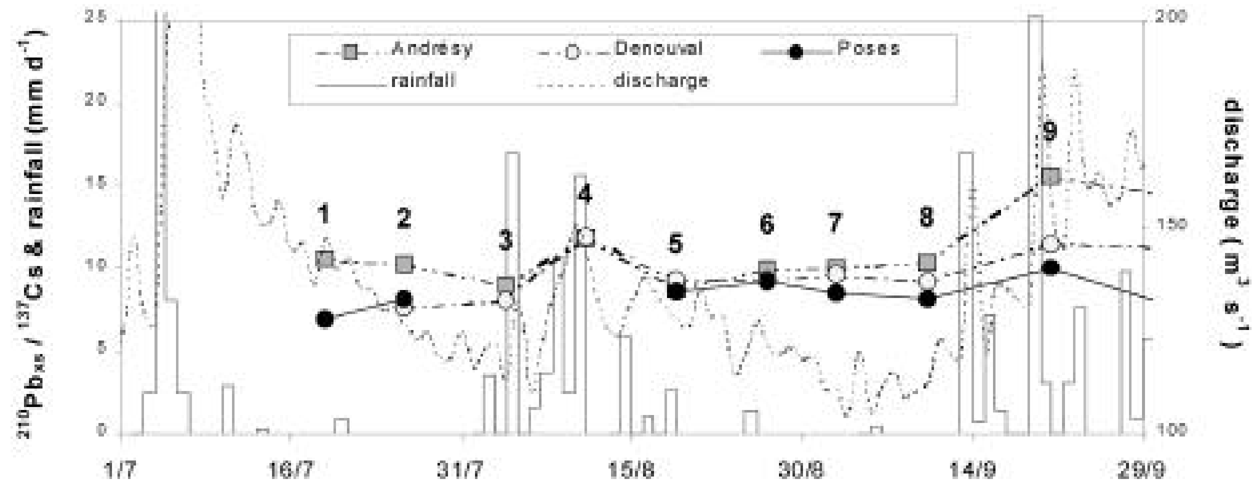

Figure 6. Distribution of $^{210}Pb_{xs}$ (normalised to ^{137}Cs content) in the SM trap samples. Discharge and rainfall data at Paris.

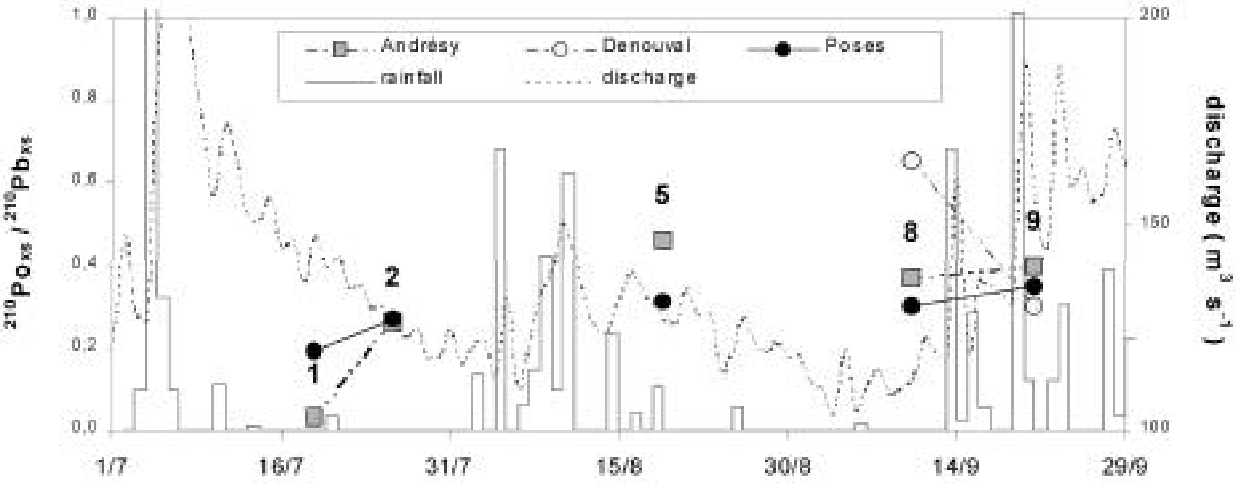

Figure 7. Distribution of $^{210}Po_{xs}/^{210}Pb_{xs}$ ratio in the SM trap samples.

the activity measured on trapped suspended solids and the elemental distribution coefficient (K_D), the dissolved tracer concentration can be computed and its total dissolved quantity in the river section is therefore estimated. Thirdly, once the dissolved quantity has been estimated, the resulting amount is attributed to particles, either suspended or deposited. If the average radioisotope activity per gram of SM and RM are known, the total mass of these two particulate stocks can be calculated. We made the assumption that the exchange kinetics between SM and RM are quick enough so that the activities in SM and RM are similar. An argument in favour of this hypothesis is the high settling velocities of suspended solids in river Seine (1 m h^{-1} on average), suggesting that the settling from SM into RM is a fast process. Accordingly, in order to compensate this loss of material out of the SM pool, the opposite resuspension flux from RM must also be fast.

However, this simple model, expected to function at steady-state, cannot cope with the difference between upstream and downstream suspended matter fluxes (Table 2). An additional source of solids is needed inside the sys-

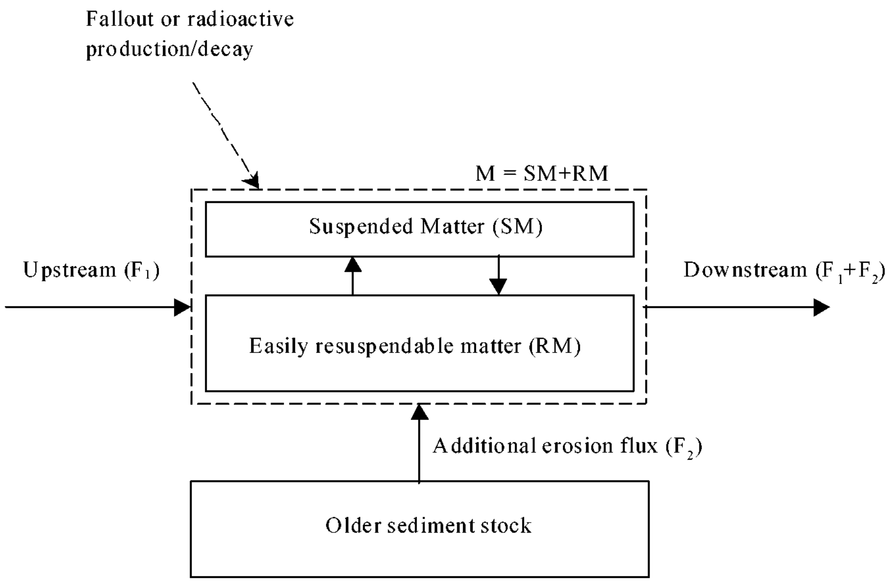

Figure 8. Simple steady-state fluxes and stocks model used to simulate suspended sediment transport in the lower river Seine.

tem, which can only be resuspension from an older sediment stock. We finally get to the still simple model structure sketched in figure 8, where the downward and upward exchanges between SM and RM are assumed fast enough compared to the overall residence time of solids and to the half-lives of the studied isotopes so that SM and RM can be considered homogeneous. Accordingly, we define M = SM+RM. The upstream flux (F_1) and the

Table 2. Model parameters and results for the July-September 1999 survey.

River section :
water surface 2.4 10^7 m^2 (1)
water volume 1.44 10^8 m^3 (1)
average discharge at Poses 228 m^3 s^{-1}
average SM content (10^{-6} kg l^{-1}) : Andrésy 10.7, Denouval 10.1, Poses 25 (box : 17)
SM content in the section : 2450 tons
solid discharge at the upstream stations 6200 t $month^{-1}$ (2)
solid discharge at Poses 15000 t $month^{-1}$ (2)

	^{7}Be	***^{131}I***	***$^{234}Th_{xs}$***	***$^{210}Pb_{xs}$***
Tracers :				
Half life (days)	53	8.05	24.1	8030
K_D (l kg^{-1})	50000	700	10^6	10^6
Average concentration in SM (Bq kg^{-1}) :				
upstream stations	282	67	24	99
Poses	123	26	25	74
Average concentration inside the section (Bqkg^{-1})(3)	203	46	24	87
Exchanges (GBq $month^{-1}$) :				
Atmospheric deposition	1.74	0	0	0.13
Upstream input :				
Particulate	1.75	0.42	0.15	0.61
Dissolved	3.37	57	0.01	0.06
Total input	*6.85*	*58*	*0.16*	*0.80*
Downstream output :				
Particulate	1.84	0.39	0.38	1.11
Dissolved	1.47	22	0.01	0.04
Total output	*3.31*	*23*	*0.39*	1.15
Input – output	*3.54*	*35*	*- 0.23*	*-0.35*
Box content (GBq) :				
Total activity (5)	8.98		0.58	
Dissolved activity	0.58	10.3	0.01	
Particulate activity	8.4	3.1	0.57	
Uranium :				
Dissolved uranium (mBq l^{-1}) (4)			5.9 ± 0.9	
Uranium activity in the box (GBq)			0.85	
Sediment mass (tons)(6)	***41000***	***91000***	***24000***	
Sediment residence time (months)	***2.8***	***6.1***	***1.6***	

(1) length 120 km, width 0.2 km, depth 6 m, (2) approximated by SM content × water discharge, (3) average of $((C_{Andrésy} + C_{Denouval})/2 + C_{Poses}) / 2$, for all samples, and standard deviation, (4) 5 measurements at stations from Nogent-sur-Seine to Rouen in 1998-1999 (OPRI), (5) the particulate + dissolved activities provide the decay term which is needed to equilibrate the isotope balance, (6) the total SM + RM stock

downstream flux ($F_1 + F_2$) are measured, with some uncertainty, and the erosion flux (F_2) from the older sediment box is computed by difference. The mean age of upstream particles at the outlet of the box is the residence time of upstream suspended solids in the system. If M is the amount of particles inside the system, the amount of particles in the system originating from upstream is given by $M.F_1/(F_1+F_2)$, and their mean age can be computed as the ratio stock over flux, i.e. $M/(F_1+F_2)$.

Since concentrations were not measured between the upstream and downstream stations defining the studied river section, concentrations inside the system are estimated by averaging for each sampling period the mean upstream concentration and the concentration at Poses.

The total particulate mass M is the only parameter directly provided by the model; as indicated in the introduction, this mass, which can be viewed as the total quantity of particles able to interact with dissolved compounds, cannot be determined by usual sampling techniques. This model is applied below to tracers with half-lives ranging from 1 week to 2 months (Table 2). A number of parameters are still poorly known, and the objective is a first test of feasibility, which will help identifying the critical points for further research.

^{7}Be

As shown by the overall ^{137}Cs constancy, the ^{7}Be downstream decrease in the SM cannot be imputed to an average coarser sediment fraction at Poses. Only a small part of the ^{7}Be activity variations originates from grain-size effects, as shown in Figure 9 where data from the upstream stations are more homogenous when ^{7}Be/^{137}Cs activity ratios are used instead of ^{7}Be activities. Thus, the upstream/downstream comparison indicates a loss of activity between the particles entering the system and those leaving it, due to the particle transit time and mixing processes.

The range of the concentration decrease between the upstream stations and Poses would correspond to a exponential decay time of 1 to 3 months. Thus a direct comparison of the samples collected at the same time would be meaningless. However, the persistent concentration difference by a factor of about 2, during the 2 months of the study, suggests that the SM transport and the mixing with the RM would result in a residence time of about 1.8 months of the particulate flux during this low water stage.

However, this simple interpretation may be biased if the atmosphere contributes significantly to the ^{7}Be budget. Figure 9 shows strong relations of ^{7}Be activity with water flow. The concentrations at Andrésy and Denouval reached a maximum in sample # 4, followed by a regular decrease. This maximum closely follows a small peak of both the river discharge and the rainfall at Paris (data at other meteorological stations downstream Paris show the same pattern), confirming significant atmospheric ^{7}Be inputs, either directly to the river section studied and upstream. Although no SM was sampled in early July, the same pattern seems to be valid for the samples # 1-3 after a small flood. This confirms that the atmospheric ^{7}Be deposition must be quantified to describe more accurately the budget of this tracer. Any significant direct atmospheric ^{7}Be deposition on the river section will increase the concentrations in the SM, and thus the above estimate of a residence time of 1.8 months is underestimated.

To get a comprehensive assessment of all inputs to the studied river section, the ^{7}Be atmospheric deposition was estimated from monthly measurements at Le Vésinet (SCPRI), a station located along the Seine at 10 km from Andrésy. The correlation between deposition (D, Bq m^{-2} month^{-1}) and rainfall (R, l m^{-2} month^{-1}) during a complete solar cycle (1976-1987) is: $D = 1.06\ R + 14$ ($r^2 = 0.62$). It has been applied to the period of study (during which solar activity was close to average values), where the average rainfall was 55 l m^{-2} month^{-1}. This gives an input of 1.74 10^9 Bq month^{-1}, which confirms that this term is not negligible (25% of inputs). Table 2 shows that nearly one half of the input is lost by internal decay. Thus the total ^{7}Be activity in the system is 9 GBq. A value of 203 Bq kg^{-1} and a K_D of 50000 l kg^{-1} are used to calculate the dissolved activity in the river section and the sediment mass (M = SM + RM), which is 41000 tons. Finally, the residence time of particles is estimated to 2.8 months. Such a time is significantly higher than the corresponding water residence time between Andrésy and Poses, which is estimated for the observed river discharge range to 3-12 days (M. Poulin, pers. comm.).

It is important to note that the sediment mass is much higher than the SM mass which is 17 10^{-6} kg l^{-1} x 1.44 10^{11} l = 2450 tons. A similar difference was obtained in the Seine inside Paris conurbation (Meybeck et al., 1998). This means that the RM plays an essential role in the ^{7}Be budget. But physically, once deposited on the river surface, a mass of 41000 tons of sediment with a density of 2 tons m^{-3} only represents a hardly detectable layer of 0.8 mm thickness.

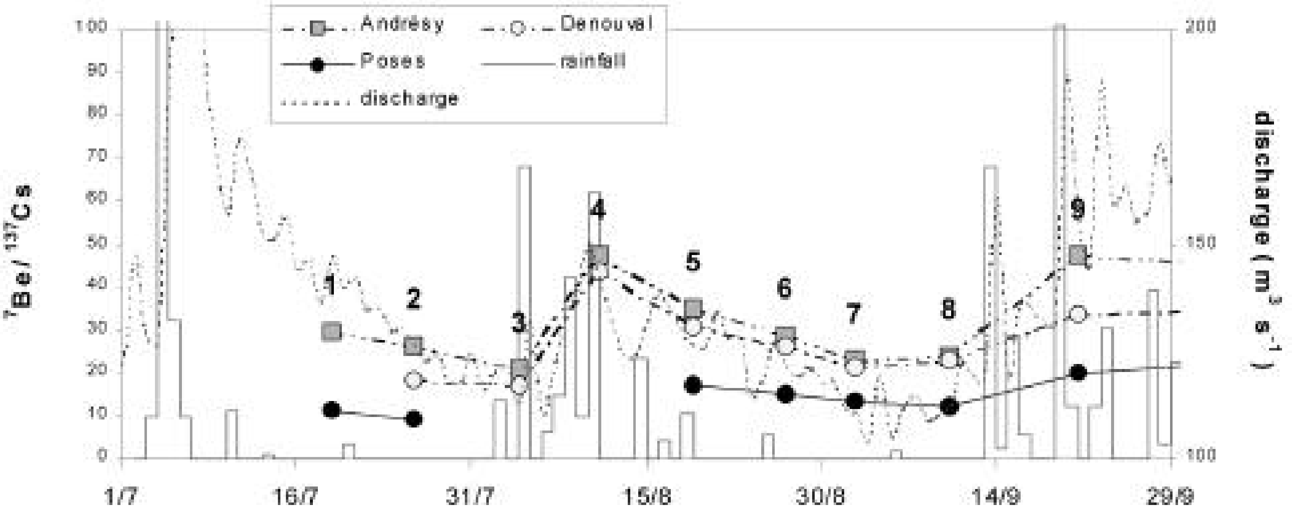

Figure 9. Distribution of ^{7}Be concentrations (normalised to ^{137}Cs content) in the SM trap samples. Discharge and rainfall data at Paris.

^{131}I

Its occurrence in the Seine river downstream Paris is not recent. In the 1979-1985 centrifuged SM samples mentioned above, this isotope was also present (22 ± 16 Bq kg^{-1}, n = 5). Several short ^{131}I fallout events were observed in this period after nuclear atmospheric tests, but the persistent occurrence of this nuclide in the effluents from the Seine-Aval waste water treatment plant (WWTP) confirms its likely origin from Paris or the suburban zone. In 1981-82, the input from the Seine-Aval plant was roughly estimated to 200-600 GBq y^{-1} (SCPRI).

This radionuclide is monitored at the Seine-Aval sewage treatment plant, on a continuous basis before water treatment, and about once a week in processed water. Average total concentrations in water average 1.2 Bq l^{-1} in both cases (January 1998-October 1999) without consistent variations with time (OPRI) ; some values however reach 2-8 Bq l^{-1}. The volume of water discharged to the Seine being about 2.1 10^6 m^3 day^{-1}, the ^{131}I input may be estimated to 920 GBq y^{-1}. During the period of study, average concentrations were 1.16 and 0.79 Bq l^{-1} before and after treatment respectively, from which an average input of 62 GBq month^{-1} was computed. After dilution in the average Seine flow during this period, the total ^{131}I concentration should be 0.105 Bq l^{-1}, and with a mean concentration of 67 Bq kg^{-1} in the SM at the upstream stations, this would give a very low K_D about 640, showing that the ^{131}I fraction associated to the particulate phase is very low as expected (about 1 %).

In the 1999 samples, ^{131}I concentrations at the upstream stations are 3 times higher than in the 1979-1985 samples, a difference which is compatible with the plant discharge estimates.

The downstream concentration decrease which is observed in most cases indicates that, as for ^{7}Be, some activity is lost due to particle transport and mixing. But contrary to ^{7}Be, concentrations in the upstream SM are inversely related to the river flow. Assuming a constant ^{131}I input to the river, and in the absence of atmospheric input, this would mean that dissolved ^{131}I concentrations tend to be diluted by increasing river flows, with a corresponding decrease of the fraction adsorbed on the SM.

The estimated concentration in the average SM is 46.5 Bq kg^{-1} , and the model (Table 2) gives a sediment mass of 91000 tons and a sediment residence time of 6.1 months. The reader may have expected a much stronger decay of ^{131}I, compared to ^{7}Be, during its transit from the upstream stations to Poses, because of its much shorter half-life. However, because the ^{131}I K_D is much lower than that of ^{7}Be , most of the ^{131}I is quickly transported in a dissolved form. The significant residence time to explain the observed decay of ^{131}I is that of water, not of particles.

^{234}Th$_{xs}$

Coupling the ^{234}Th$_{xs}$ measurements in the SM with measurements of dissolved uranium provides a tool analogous to ^{7}Be for estimating the sediment mass and residence time in the Seine river.

The few measurements available indicate that the dissolved uranium content in the filtrated Seine river water is rather constant at different seasons and over a wide distance inside the whole Seine river basin (0.48 ± 0.09 µg l^{-1}, or 5.9 ± 0.9 mBq 1^{-1}, Table 2). If the average ^{214}Bi content in the SM (22 Bq kg^{-1}, Table 1) is close to equilibrium with ^{238}U, this gives an uranium K_D of 3700. In a given volume of water where the suspended matter content is SMC (kg l^{-1}), and the elemental distribution coefficient is K_D (l kg^{-1}), the fraction of an element associated to the particulate phase is $F_P = K_D$ SMC / (K_D SMC + 1). For uranium, this gives $F_P = 0.06$. In Lake Geneva, where the SMC is very low (4 10^{-7} kg l^{-1}), the K_D of ^{234}Th averages 10^{7} (Dominik et al., 1989); considering the decrease of the K_D of ^{234}Th with an increasing SMC (Honeyman and Santschi, 1992), its K_D in our SM samples should be close to 10^{6}, which gives $F_P = 0.95$. Thus, nearly all the ^{238}U is dissolved, and nearly all the $^{234}Th_{xs}$ is fixed on the particles (Table 2).

If steady state is assumed, the $^{234}Th_{xs}$ budget in a given river section is:

upstream input with SM + production from dissolved U = downstream output with SM + disintegration

As shown in Table 2, the output is 2.4 times higher than the input. The imbalance of -0.23 GBq $month^{-1}$, which corresponds to the loss of 2.63 10^{8} atoms of ^{234}Th per second results from the combination of the production of particulate ^{234}Th by the disintegration of uranium and the radioactive decay of thorium.. With an uranium concentration of 5.9 mBq l^{-1}, the river section contains an activity of 0.85 GBq, which is the number of ^{238}U atoms which disintegrate per second to produce the same number of atoms of ^{234}Th. The budget is thus equilibrated with the disintegration of 0.85 10^{9} - 2.63 10^{8} = 5.87 10^{8} atoms of ^{234}Th per second (i.e. Bq). This exceeds the river input/output imbalance by one order of magnitude. Since this activity is rapidly fixed by the SM, which has a rather constant $^{234}Th_{xs}$ concentration of 24 ± 4 Bq kg^{-1}, the sediment mass is 5.87 10^{8} / 24, i.e. about 24000 tons, and the sediment residence time is 1.6 months.

It is important to note that all the above estimations of M (= SM+RM) are based on the assumption that the flux of short-lived radionuclides associated to the erosion flux F_2 from the older sediment stock is almost zero. It would be useful to gain information about the age of the older sediment stock, although it does not directly interfere with the residence time of upstream suspended solids previously defined. Longer-lived isotopes may be useful for that purpose.

How old is the older sediment stock ?

With its rather long half-life, ^{210}Pb cannot be used to trace particles in the same way as the short-lived tracers discussed above since the decay term becomes negligible due to its longer half-life. Table 2 shows the computed $^{210}Pb_{xs}$ budget. Using a correlation between the ^{210}Pb deposition and rainfall established by Zuo (1992) for the Netherlands, the direct fallout of $^{210}Pb_{xs}$ over the lower Seine was estimated to about 0.13 GBq $month^{-1}$.

The budget of $^{210}Pb_{xs}$ shows a net export downstream, mostly due to the net export of suspended solids (Table 2). An additional input is necessary to equilibrate the overall budget, and a contribution from the older sediments is the only possible explanation. The suspended solids balance shows that it represents an additional flux of 8800 tons of older sediments per month, with a $^{210}Pb_{xs}$ activity of 40 Bq kg^{-1}, which is about one half of its activity in the SM. This difference may result from grain size effects, radioactive decay, and inputs by soil erosion.

The ^{137}Cs budget established similarly leads to an activity of 8.1 Bq kg^{-1} in the older sediment, a value which is hardly lower than the measured activity in trapped particles (9.5 ± 1.3 Bq kg^{-1}). Accordingly, these old particles would be only slightly coarser than the trapped particles. Thus, the lower activity of $^{210}Pb_{xs}$ in the older sediments should not be due to a grain size effect.

A direct interpretation by the decay of $^{210}Pb_{xs}$ would indicate that the older sediments are about 22 years old (i.e. the half-life of ^{210}Pb), but this seems unlikely since it would imply a ^{137}Cs activity 1.7 times higher than measured in the SM. But we cannot exclude that the ^{210}Pb was inherited by the older sediments during a period of either a lower fallout, or a similar fallout but a higher suspended solid discharge, which would have in both cases resulted in lower $^{210}Pb_{xs}$ concentrations.

However, the above $^{210}Pb_{xs}$ activity is still much higher than the $^{210}Pb_{xs}$ activity in the soil samples so far studied (Sogon et al., 1999), and with similar ^{137}Cs activities. Therefore, it is obvious that most of the $^{210}Pb_{xs}$ in the older sediment stock originates from a direct fallout over

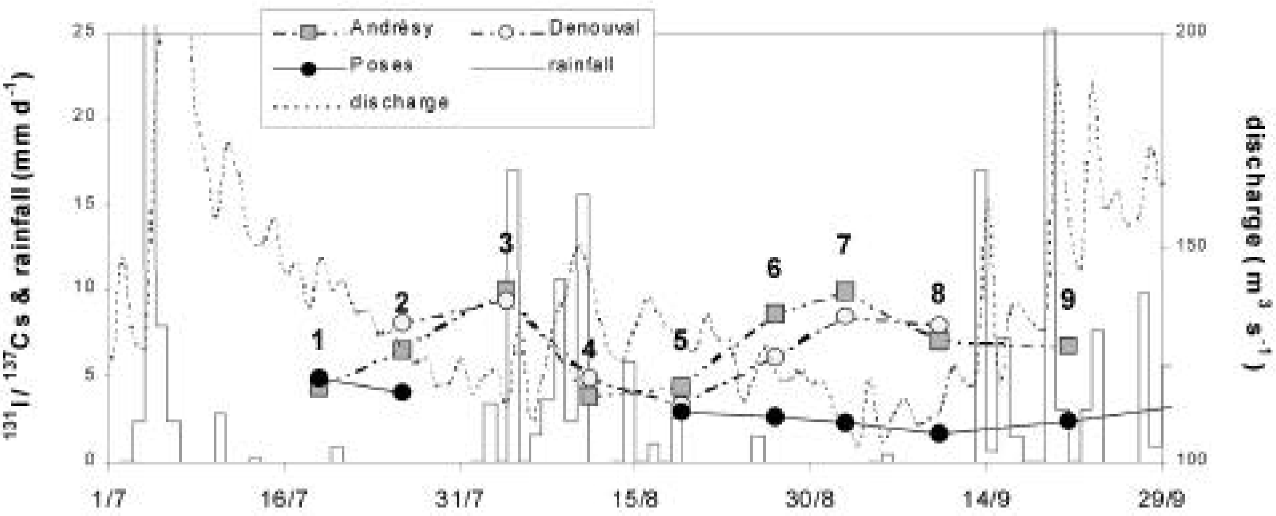

Figure 10. Distribution of ^{131}I concentrations (normalised to ^{137}Cs content) in the SM trap samples. Discharge and rainfall data at Paris.

the river which occurred prior to our sampling period and is not inherited from soils. This past fallout also brought some ^{7}Be which must have been partly adsorbed by the particles, while these particles, in contact with the large dissolved uranium pool, should also have been enriched with ^{234}Th$_{xs}$. This raises the question of the possible occurrence of residual ^{7}Be and ^{234}Th$_{xs}$ activities in the older sediment stock, which have been so far neglected in our approach, and might be a significant source of error in the previous estimates of the suspended and resuspendable sediment stocks.

With its 138 days half-life, ^{210}Po is an interesting candidate to further examine that question. The ^{210}Po$_{xs}$ budget is problematic because of more dispersed data. At a first approximation, an ill-defined average value of about 0.4 for the ^{210}Po$_{xs}$/^{210}Pb$_{xs}$ ratio can be tested for both the upstream and downstream stations. It is likely that the measured ratios are not directly inherited from the atmospheric deposition since the ^{210}Po$_{xs}$/^{210}Pb$_{xs}$ ratio in the rain is usually low (e.g. 0.095 in the atmospheric deposition near Lake Geneva; Dominik et al. 1987). Although no data are available in the Seine river basin, we shall use a fallout ratio of 0.1. Finally, the most problematic unknown is the ^{210}Po$_{xs}$/^{210}Pb$_{xs}$ ratio in the older sediments.

An important result is that, if the ratio is equal to 1 in the older sediment stock (a value which is reached after more than 1.5 years), the ^{210}Po$_{xs}$ budget can by no way be achieved, even if the suspended sediment stock (SM+RM) is zero. The lower the ratio in the older sediment stock, the higher the necessary suspended sediment stock must be, to equilibrate the ^{210}Po budget: the estimated sediment stock becomes meaningful (i.e. positive) only when the ratio is below 0.5. Although, these estimations are very rough because of the variability of the input data, they show that the older sediment stock with a rather low^{210}Po$_{xs}$/^{210}Pb$_{xs}$ ratio may not be that old, and thus we cannot discard the possibility of an additional input of ^{7}Be or ^{234}Th$_{xs}$ from the older sediments erosion . This source of uncertainty will therefore be included in the following sensitivity analysis.

Sensitivity analysis.

The sediment stock computations developed above are based on a number of data, either measured or estimated from literature data. In addition to previously considered uncertainties on the estimates of particulate activities the following sources of uncertainty have been tested:

(i) measurement errors, or measured variability of measured activities, which show that the system is not perfectly at steady-state. The variability was first tested simultaneously for upstream and downstream activity estimate. Then although the figures show rather parallel evolutions for upstream and downstream activities along the period, it was also tested separately for both estimates.

(ii) the average composition of particles inside the box by assuming its composition was 2/3 that of downstream particles and 1/3 that of upstream particles (or reverse) instead of half upstream and half downstream particles,

(iii) K_D's were multiplied by a factor three or divided by the same factor, given the high scatter on K_D's values given in the literature,

(iv) upstream and downstream suspended solids fluxes were modified by ± 20%,

(v) short-time tracers (^{7}Be, ^{234}Th and ^{131}I) in the older sediment stock were set to 25% and 50% of their activity in the sediment box, instead of 0%,

(vi) the upstream input of ^{131}I was set to the maximum and minimum values obtained from instantaneous monitoring activity measurements in the Seine-Aval waste water treatment plant,

(vii) the concentration of dissolved ^{238}U in water was set to its average measured value ± one standard deviation,

(viii) 20% error on the estimation of ^{7}Be atmospheric flux, and 50% error on the estimation of the ^{210}Pb flux which is much less well known.

Results of the uncertainty analysis are shown in Table 3, which allows to identify the more significant parameters which affect the sediment mass estimates using the various tracers. In particular, among the two isotopes which seem to give the more reliable results, ^{7}Be is much more sensitive to the estimate of its K_D than ^{234}Th. For both these isotopes the dissolved activity was only estimated in our study, but the uncertainty is much lower in the case of ^{234}Th because of its higher K_D. The estimates using these isotopes are similarly sensitive to their production terms (fallout in the case of ^{7}Be, and dissolved ^{238}U decay in the case of ^{234}Th). Although both isotopes do not lead to the same stock estimate, their discrepancy keeps within the uncertainties. The much higher stock estimate obtained with ^{131}I must be cautiously regarded because of the high level of uncertainty arising from the in-box evaluation of activity.

CONCLUSIONS

This first phase of our research on radioactive tracing of sediment transport in the Seine river downstream Paris during the low water stage has clarified the usefulness of the environmental radionuclides by applying a simple steady state model in which the suspended matter and the resuspendable matter are not distinguished. The parameters used in the model are sometimes only crude estimates.

This study confirms that cosmogenic ^{7}Be, which exhibits a measurable concentration decrease in the SM over the 120 km river course, is a well adapted tracer to study river sediment transport and mixing, if its atmospheric deposition can be determined. It allowed to assess a sediment residence time of 2.8 months. In order to improve this estimation, it is important to better assess its K_D in the Seine river. An alternative but similar methodology is based on the use of ^{234}Th$_{xs}$, for which the atmospheric deposition can be neglected, and which concentrations in the SM do not decrease in the river course, because of a strong in-situ production from dissolved uranium which contributes to balance its rapid decay. The assessment of a sediment residence time of 1.6 months is quite similar to the ^{7}Be result. Because ^{7}Be activities are significantly modified after short rain events, while ^{234}Th are not, future comparison of time series of both isotopes activities in suspended sediments should bring much important information with regards to the dynamics of sediments stocks.

The use of a less common short-lived tracer, ^{131}I, directly supplied to the water by a water treatment plant, gives a higher residence time (6 months), but with a much larger uncertainty. The source term (urban waste waters) in particular should be better assessed, and the observed activities are highly variable depending on the water discharge, which probably precludes the use of a steady-state model.

The sediment masses estimated from ^{7}Be and ^{234}Th are 10 to 17 times higher than the SM mass ; this confirms that the RM, which escapes direct observation and sampling, is the most significant particulate component of the river system with regard to sorption of particle-reactive natural and anthropic elements.

The ^{210}Pb-^{210}Po couple allows to give some insight on the characteristics, age and origin of the old sediment recycling by erosion of the river bottom. Although the limited ^{210}Po data set was insufficient to fully exploit its potential usefulness in this purpose, this tracer indicated that such erosion processes are to be considered in the future.

A limitation of the present approach is that the estimated residence times are larger than the duration of the survey during which the situation was characterised, and the steady state hypothesis may not describe properly the system. We are therefore developing a more realistic approach using a non steady state dynamic model, which should in the next future include the dynamics of the older sediment stock as an explicit variable in the system.

The long residence time of suspended solids, coupled with resuspension from an older sediment stock explains why the particulate pollution pulses from Paris area are so efficiently damped, typically after strong rain events creating severe sewer overflow situations (Estèbe et al., 1998). It also explains why all contaminant concentrations decrease on suspended solids from Paris to Poses during steady low water periods. It is most likely due to the input of older less contaminated sediments, deposited during the previous hydrological period with a higher solid discharge, and a higher dilution of urban contamination.

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