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Quality of dredged material in the river Seine basin (France). II. Micropollutants

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Abstract

Dredging rivers is needed to ensure safe navigable waters, rivers and waterways. To anticipate the management of dredged materials in the case of the river Seine basin, the quality of the sediments in the river is checked every 3 years before dredging operations. The river Seine Basin is heavily submitted to pollution pressure from nearby industrial activities and urban expansion of Paris and its region. Here, the micropollutant content of the sediment sampled in 1996, 1999 and 2000 before dredging is discussed compared to regulatory standards. The results indicate that most of the sediment samples from the river Seine basin are lightly to moderately contaminated with organic and inorganic micropollutants (heavy metals, PAH, PCB), which makes the management after dredging easier. This pollution is strongly correlated with the organic matter content and to the fine fraction (<50 μm) of the sediment. These results can lead to other management options than the ones already used in the river Seine basin: (1) dumping of lightly to moderately polluted sediments in quarries; and (2) physical treatment (sieving, hydrocycloning) of contaminated sediments issued from 'hot spots'.
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Keywords: Dredged material; Sediments; Quality; River Seine basin; Heavy metals; PAH

1. Introduction

The increasing urban environmental pollution and human impact leads to water quality deterioration. With the release of polluted water into the receiving water bodies, contaminants are absorbed onto suspended particles and subsequently accumulated in the underlying sediment (Hauge et al., 1998; Lau and Chu, 1999). Hence, dredging contaminated sediments means removing the pollution which is contained in the sediment.

Dredging techniques vary depending on the volume, texture, water content and grain size of the sediments to be removed. The method that causes the least environmental impact on the water

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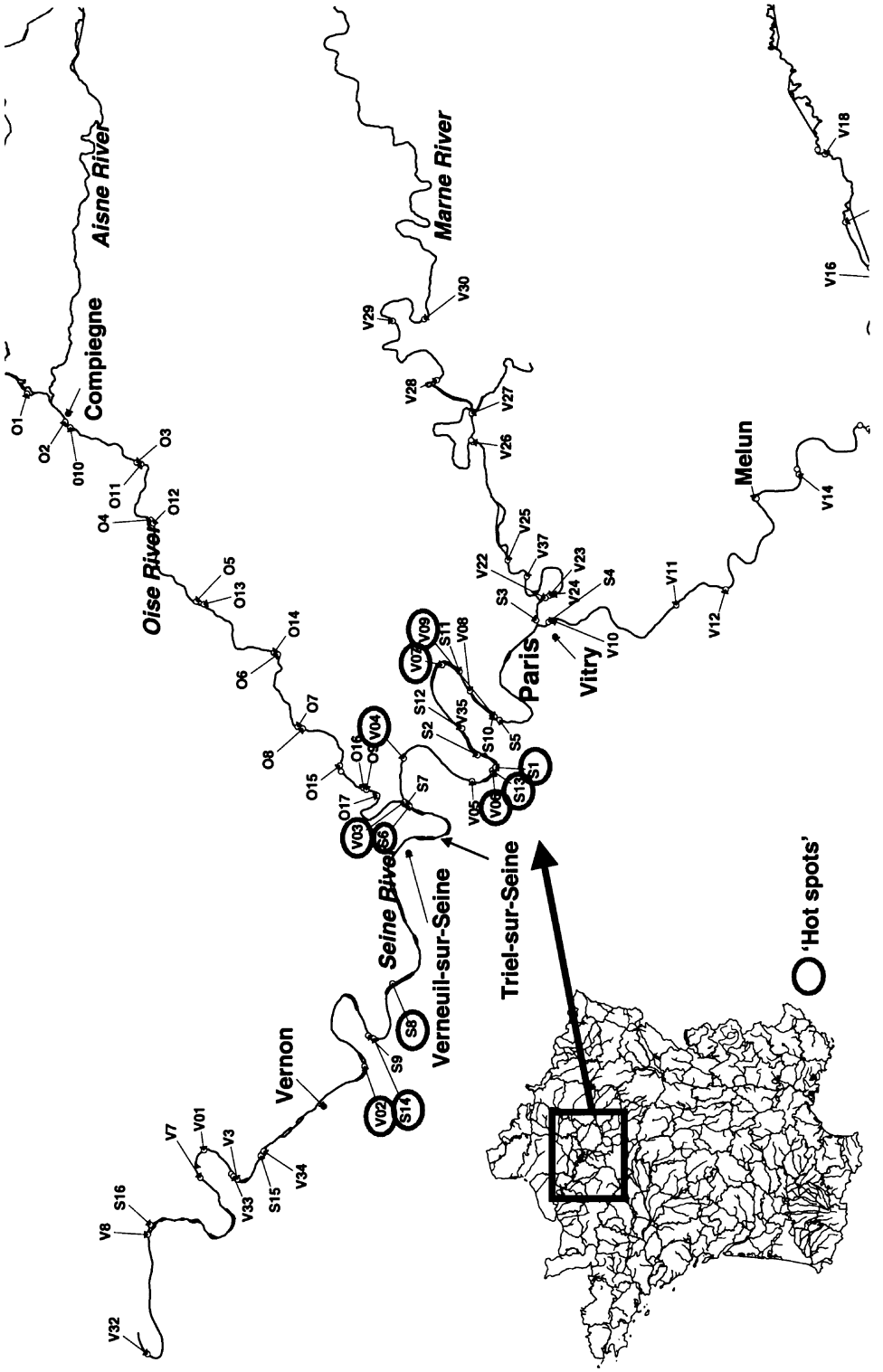

Fig. 1. Part of the Seine basin concerned in this study and site codes corresponding to the sediment samples collected during 1996, 1999 and 2000 campaigns: 'Hot spots' i.e. sites of significant sediment contamination appear within circles.

Table 1
List of micropollutants determined in the sediment (S) sampled in 1996, 1999 and 2000, in the leachates (L) and in the pore water (P), methods used to analyse each parameter and detection limits (dl) for the different methods

Analysis	Method	Analysis	Sample	dl (S) ($\mu g\ g^{-1}$)	dl (L) ($\mu g\ g^{-1}$)	dl (P) ($\mu g\ l^{-1}$)
Total hydrocarbons	X 31-410 ISO TR 11046	IR	S, L, P	20	1	50
Total PAH (16 EPA species)	NF T 90-115	HPLC	S, L, P	0.01	0.01[a]	0.01
Total aliphatic hydrocarbons		GC/FID	S, L, P	2	20[a]	20
Toluene	NF ISO 11423-1	GC/FID	S	0.1	nd	nd
Pesticides	NF EN ISO 6468 T90-120	GC/MS	S	0.15	nd	nd
Total PCB (6)	NF EN ISO 6468 T90-120	GC/ECD	S	0.005	nd	nd
As	NF EN ISO 11885	ICP/AES	S, L, P	5	0.05	5
Cd	NF EN ISO 11885	ICP/AES	S, L, P	0.8	0.02	2
Cu, Cr, Ni, Pb, Zn	NF EN ISO 11885	ICP/AES	S, L, P	5	0.1	10
Al	NF EN ISO 11885	ICP/AES	S, L, P	5	1	10
Fe, Mn	NF EN ISO 11885	ICP/AES	S, L, P	5	0.1	10
Hg	XP T 90-113-2	Atomic fluorescence	S, L, P	0.05	0.005	0.5
Phenolic compounds	Acidic extraction	GC/MS	S, L	2	nd	nd

[a] $\mu g/l$.

quality, during dredging operations, is the hydraulic method. However, mechanical dredging yields materials containing high solid levels: this is an important criteria given for better management of dredged material (DM), i.e. avoiding large water volumes management (Hauge et al., 1998).

Once dredged, DM can be treated, valorised, or disposed. Before any management decision, it is necessary to characterise the DM chemically, physically and mechanically (Boutouil et al., 1997). However, the economic, political context and management issues, such as the lack of disposal sites, can interfere with the decision provided by the characterisation of DM. In the specific case of the river Seine basin, mechanical dredging from a boat was chosen to remove approximately 150 000 m^3 of sediment per year from the Seine basin. Dredging in the Seine basin is mainly 'maintenance dredging' for navigation. DM is then dumped into wet disposal sites, i.e. disused quarries, downstream Paris. From 1992 to 1998, the site of Triel and from 1999 to 2001, the site of Rouillard have been filled with the DM extracted from the river Oise downstream Compiègne and from the river Seine downstream Vitry (Fig. 1). According to an estimate in the 1980s by the US Army Corps of Engineers (Combs et al., 1982) and in a recent study in the Netherlands (Cuypers et al., 1998), approximately one third of all dredged materials are contaminated by municipal sewage, industrial discharges and agricultural runoff.

In a previous paper (Carpentier et al., 2002), we characterised the physico-chemical properties and major contamination patterns of the sediments sampled from the river Seine basin before dredging. This work revealed the wide variety of the sediment quality, particularly the carbon content and the grain size, along the river Seine basin. The analysis of porewaters revealed high concentrations in ammonium that might lead to a management concern: in fact, the dumping of such sediment after dredging might induce an increase of ammonium concentrations in the receiving waters at the disposal sites (Carpentier et al., 2001). After correlation analysis and geographic distribution study, the impact of the great Parisian region (including Paris itself and close counties) was not clearly observed. This impact was only observed when the location of the sediment sample was far enough, i.e. approximately 100 km from the urban and/or industrial sources. For example, samples V30 or V18 on Marne and Seine river, respectively, upstream Paris showed very different organic matter contents compared to S1 or S6 on Seine river downstream Paris. In order to improve our knowledge on the quality of river Seine basin

Table 2
Metal content ($\mu g\ g^{-1}$) in the sediment collected in 1996, 1999 and 2000

Code	Y[a]	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn	Al	Fe	Mn	Code	Y[a]	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn	Al	Fe	Mn
V01	1996		<0.8	<10	12	0.76	<5	32	70				S6	1999	<5	1.8		62	0.64	10	103	220	2600	3300	38
V02	1996		<0.8	<10	28	15.5	<5	50	98				S7	1999	<5	3		83	0.92	19	83	293	3800	8200	50
V03	1996	6.93	<0.8	46	172	1.61	22	194	535				S8	1999	<5	3		90	0.64	16	69	250	1800	7200	43
V04	1996	3.33	<0.8	24	102	6.31	<5	132	240				S9	1999	<5	1.5		54	0.65	20	70	225	2200	15 100	39
V05	1996		<0.8	24	80	2.78	<5	174	290				V1	1999	<5	<0.8	44	91	0.8	17	42	278			
V06	1996		<0.8	22	92	5.55	<5	158	246				V2	1999	6	<0.8	48	96	0.8	18	9	295			
V07	1996	5.69	6	46	122	2.5	20	278	563				V3	1999	<5	<0.8	13	<5	<0.05	<5	<5	58			
V08	1996	4.08	<0.8	20	70	2.35	<5	152	236				V4	1999	<5	<0.8	25	29	<0.05	<5	12	88			
V09	1996		<0.8	<10	64	5.9	<5	136	283				V5	1999	<5	<0.8	10	<5	0.1	<5	15	46			
V10	1996		<0.8	20	68	<0.05	<5	74	168				V6	1999	<5	<0.8	16	26	0.192	<5	43	100			
V11	1996		<0.8	<10	42	<0.05	<5	82	118				V7	1999	<5	<0.8	13	8	0.109	<5	12	43			
V12	1996		<0.8	<10	16	<0.05	<5	48	52				V8	1999	6	<0.8	78	73	0.414	24	80	300			
V13	1996		<0.8	<10	22	<0.05	<5	118	94				O1	1999	<5	<0.8		9	0.29	14	42	81	6600	17 000	15
V14	1996		<0.8	<10	10	0.37	<5	36	32				O2	1999	<5	<0.8		<5	0.09	6	24	39	6100	9400	9
V15	1996		<0.8	<10	14	0.27	<5	44	26				O3	1999	<5	<0.8		24	0.3	20	33	141	5900	14 500	25
V16	1996		<0.8	<10	60	1.28	<5	<5	76				O4	1999	<5	<0.8		16	0.3	17	28	99	5100	12 300	18
V17	1996		<0.8	<10	16	0.36	<5	<5	30				O5	1999	<5	<0.8		27	0.39	22	51	183	3100	2300	32
V18	1996		<0.8	<10	44	<0.05	<5	<5	54				O6	1999	<5	<0.8		14	0.3	8	32	86	6800	8600	16
V19	1996		<0.8	<10	12	<0.05	<5	<5	36				O7	1999	<5	<0.8		26	0.37	20	50	173	6900	18 300	30
V20	1996		<0.8	<10	14	<0.05	<5	56	80				O8	1999	<5	<0.8		32	0.45	20	58	185	12 900	11 200	32
V21	1996		<0.8	<10	14	<0.05	<5	214	260				O9	1999	<5	<0.8		28	0.44	21	53	103	5300	23 000	19
V22	1996		<0.8	28	68	<0.05	<5	86	238				S10	2000	11	0.91	36	114	0.53	20	80	247	28 661	29 177	469
V23	1996	3.35	<0.8	<10	28	0.31	<5	38	82				S11	2000	6	<0.8	11	23	0.18	9	44	116	6652	6643	243
V24	1996		<0.8	20	50	0.28	<5	72	144				S12	2000	13	1.33	48	131	0.56	28	107	355	32 778	34 240	495
V25	1996		<0.8	24	48	<0.05	<5	70	132				S13	2000	<5	<0.8	11	27	0.25	<5	64	87	3119	4676	199
V26	1996		<0.8	<10	20	<0.05	<5	<5	78				S14	2000	13	2.06	73	136	0.76	30	102	450	36 148	36 960	509
V27	1996		<0.8	<10	18	<0.05	<5	42	60				S15	2000	10	1.65	63	127	0.64	25	88	400	34 464	33 711	456
V28	1996		<0.8	20	38	<0.05	<5	58	118				S16	2000	12	1.66	63	120	0.65	26	83	380	30276	36 787	484
V29	1996		<0.8	20	26	<0.05	<5	32	88				O10	2000	11	<0.8	40	28	0.15	23	24	144	24 488	39 859	343
V30	1996		<0.8	<10	30	<0.05	<5	42	90				O11	2000	6	<0.8	34	28	0.15	17	39	136	15 886	21 756	356
V31	1996		<0.8	24	56	<0.05	<5	32	132				O12	2000	10	<0.8	41	32	0.17	20	30	179	25 105	32 261	379
S1	1999	<5	3		94	24	24	144	286	1800	9700	49	O13	2000	9	<0.8	35	31	0.23	16	33	191	21 337	31 955	419
S2	1999	<5	<0.8		56	0.49	16	71	188	3000	5600	33	O14	2000	12	0.92	49	39	0.26	24	38	225	32 588	38 650	481
S3	1999	<5	<0.8		30	0.23	14	62	122	5900	5300	22	O15	2000	7	<0.8	26	23	0.21	11	28	125	1562	15 496	358
S4	1999	<5	<0.8		85	0.56	15	198	414	5000	5600	70	O16	2000	11	<0.8	40	42	0.28	19	44	220	23 764	35 368	406
S5	1999	<5	1.3		60	0.51	21	73	210	2600	10 900	37	O17	2000	7	<0.8	30	32	0.28	14	35	171	4017	20 921	391

See Fig. 1 for sampling site location.
[a] Year of collection.

Table 3
Metal content (μg g^{-1}) of the sediments collected before dredging in the Seine Basin in 1996, 1999 and 2000

Site		As	Cd	Cr	Cu	Hg	Ni	Pb	Zn	Al	Fe	Mn	Reference
Seine River, 1996–2000	n	41	42	32	50	50	50	50	50	33	33	33	This study
(sediment)	d_{10}	<5	<0.8	10.1	0	0.15	3.6	23.1	79.9	2280	5360	18.2	
	d_{50}	<5	0.8	32	31.5	0.45	17	52	189	6100	15 100	50	
	d_{90}	11	2.0	61.6	120.2	3.06	24	152.6	382	32 126	36 503	479	
	Min	<5	<0.8	4	<5	<0.05	<5	<5	39	1562	2300	9	
	Max	13	6	78	172	24	30	278	563	36 148	39 859	509	
	Mean	3.8	1.1	32.5	49.3	1.65	14.7	72.0	210	12 371	18 362	199	
	SD	4.9	1.1	20.0	44.4	4.1	8.4	57.2	123	11 780	12 291	195	
	CV	129	107	62	90	247	58	79	59	95	67	98	
Seine River, 1996–1998	Min		0.8	43				82	192		16 600		Meybeck et al., 1998
(TSS)	Max		7.9	380				300	825		25 300		
	Annual mean		2.7		88	0.9		109	401	52 700			
North of France River	Mean		2.8	133	220		42	360	1180				Ruban et al., 1998
Detroit River, USA	Mean		7.2	129	85.1		71.3	176.0	792				Besser et al., 1996
Elbe River, Germany (TSS)	Mean		11.5		150	6			1500				Netzband et al., 1998
Danube River, Hungary	Mean		0.8	70.7	42		36	47.8	159				Gruiz et al., 1998
Rio de Janeiro River, Brazil	Mean	3.4			28.6		13.6	44.2	110				Almeida et al., 2001
Canal, Belgium	Mean		12.9	267	128			721	3200	14 580	29 000	2480	Cauwenberg et al., 1998

Number of samples (n), median (d_{50}), first and last decile (d_{10}, d_{90}), minimum and maximum (min, max), standard deviation (SD) and coefficient of variation in % (CV) of the data given in Table 2.

sediments, we also analysed the micropollutants in the sediments. Here, we are not only interested in the sediment quality itself, but we are also concerned about the quality of the DM that it becomes, after extraction from the river bed, and its fate once moved to another receiving environment. For that reason, we also analysed micropollutants in porewaters of the sediments and leachates obtained with the same sediment samples. The study of leachates is, in the case of disposal, one of the techniques recommended by the French standard NF XP X 31-210 for assessing the impact of materials on their close environment, after dumping. The study of porewaters was also performed in order to assess the potential release of pollutants that might occur during the management of DM. The aim of this work is to translate these sediment and pore quality data into dredging and treatment strategies for the river Seine basin.

2. Material and methods

2.1. Sediment collection

The sampling procedure has been fully described previously. Briefly, samples were collected from the bottom of the rivers Seine, Oise and Marne by handcore sampling (20–50 cm depth depending on the DM quality) (Fig. 1). Samples were collected in May and June 1996, April 1999 and May 2000 in representative areas determined by the Navigation Services of the Seine basin (SNS), according to the dredging needs for the following year, resulting from bathymetric survey of the river. Once cored, sediment samples were transferred into fully filled and closed glass containers (1 l) and were kept in a store room (4 °C) for 48 h maximum before homogenisation and analysis.

2.2. Methods

Different parameters were analysed both on the sediment (Table 1) and in the leachates obtained with the same sediment samples. Leachates were resulting from mixing 200 g of bulk sediment for 24 h with 2 l of distilled water, according to the French AFNOR standard XP X31-210. Parameters were also measured on pore waters. The method to extract pore waters is described in a previous paper (Carpentier et al., 2002). Most parameters were measured following the French standard procedures i.e. AFNOR (Table 1). All samples were analysed by laboratories certified by the French Ministry of the Environment, i.e. certified by the COFRAC (Comité Français d'Accréditation). For international quality control, the COFRAC calibration certificates co-operates with other European calibration services (the European co-operation for Accreditation, EA). The harmonisation of the accreditation criteria and procedures are orientated towards the European standards for calibration and testing laboratories and their assessment and accreditation (EN ISO/IEC 17025 and EN 45 000 series). The detection limits for inorganic and organic micropollutant content of sediment samples were chosen in order to be significantly below regulatory standards (Table 1).

Table 4
Alert values (A) and Intervention values (I) for soil remediation ($\mu g\ g^{-1}$)

Pollutant	A. Alert value	I. Intervention value
As	29	55
Cd	0.8	12
Cr	100	380
Cu	36	190
Hg	0.3	10
Ni	35	210
Pb	85	530
Zn	140	720
Benzene	0.05	2
Toluene	0.05	130
PCB_{tot}[a]	0.02	1
PAH_{tot}[b]	1	40
DDT + DDD + DDE	0.0025	4
Alkanes	50	5000
$Phtalates_{tot}$	0.1	60
HCH	–	2

Dutch circular, 04/02/2000, Ministry of Housing, Spatial Planning and Environment, Holland.

[a] PCB_{tot} = sum of PCB 28, 52, 101, 118, 153, 180.

[b] HAP_{tot} = sum of 10 PAH [Anthracene, Fluoranthene, Benzo(*k*)fluoranthene, Benzo(*ghi*)perylene, Indeno(123-*cd*)pyrene, Benzo(*a*)anthracene, Benzo(*a*)fluoranthene, Chrysene, Naphtalene, Phenanthrene].

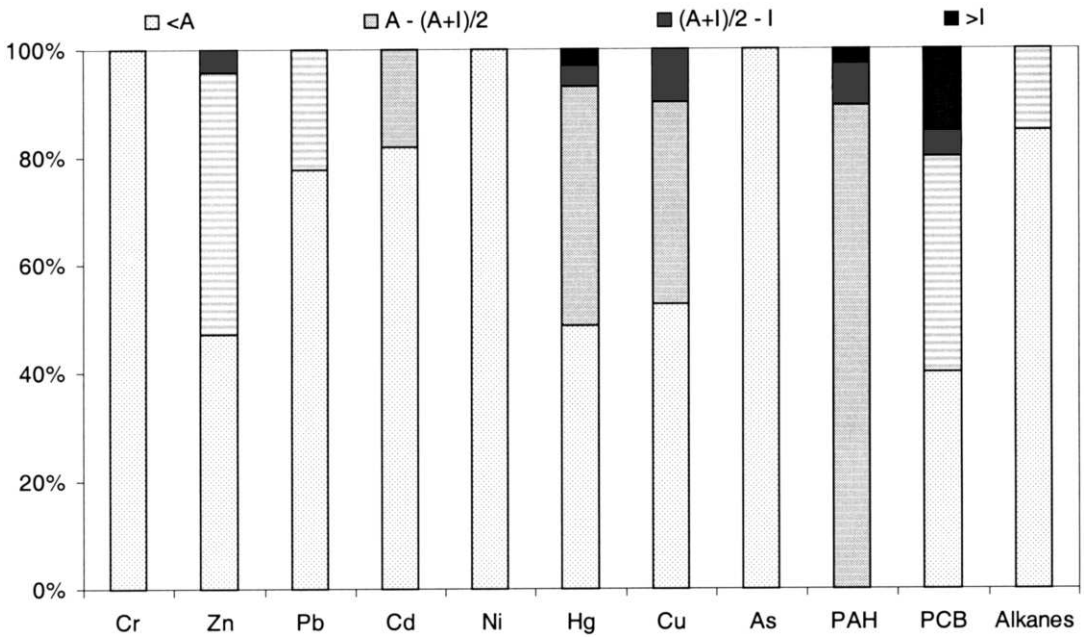

Fig. 2. Statistical distribution of the number of sediment samples presenting a contamination in heavy metals or organic micropollutants below (<A) the Alert value of the Dutch decree on the quality of sediments, above (>I) the Intervention value of this decree, or within these two values.

3. Results and discussion

3.1. Sediment quality

3.1.1. Heavy metals

The average metal content of the sediment collected in various dredged areas of the river Seine basin (Table 2 and Table 3) is generally slightly lower than the average values found in the total suspended solids (TSS) from the river Seine basin (Meybeck et al., 1998). In fact, in the case of Zn, levels vary from 190 to 700 $\mu g\ g^{-1}$ in the TSS, whereas in the sediment analysed in the present study, concentrations vary from 40 to 560 $\mu g\ g^{-1}$. The same difference is observed for Cd, Cu, Pb. This slight difference can be explained by the nature of the sediment itself. As we found out previously (Carpentier et al., 2002), grain size of sediments from the river Seine basin, and particularly the percentage of particles below 50 μm is on average 20%. In the study concerning TSS, this percentage was ranging from 40 to 45%. This may explain the higher heavy metal content in TSS samples, compared to sediment samples, because heavy metals are preferably adsorbed on fine particles (Evans et al., 1990; Warren and Zimmermann, 1994; Perrin and Zimmer, 1995). However, the difference observed between sediments and TSS from the river Seine basin remains small compared to the difference of heavy metals content between sediments sampled at different locations of the Seine basin.

Concentrations of metallic pollutants found in the sediment sampled in the Seine basin are low (210 $\mu g\ g^{-1}$ on average for Zn) compared to the values found in French Northern rivers 'Petite Marque' and 'Riez Simon' near Lille (59) (Ruban et al., 1998) which contain on average 1180 $\mu g\ g^{-1}$ for Zn. This difference might be due to the characteristics of the sediment (TOC for the North of France river = 83 $g\ kg^{-1}$ whereas average TOC for this study = 37 $g\ kg^{-1}$). In the case of sediments from canal Gent Terneuzen in Belgium (Cauwenberg et al., 1998), Zn concentrations can

Table 5
Organic micropollutant content ($\mu g\ g^{-1}$) in the sediment collected in 1996, 1999 and 2000

Code	Y[a]	HC_{tot}	PAH_{tot}	Ali_{tot}	PCB_{tot}	Pesticides	Toluene	Phenols	Code	Y[a]	HC_{tot}	PAH_{tot}	Ali_{tot}	PCB_{tot}	Pesticides	Toluene	Phenols
V01	1996	3							S3	1999		13.0	14.6				
V02	1996	118							S4	1999		11.3	33.1				
V03	1996	522	6.66		22.9			<2	S5	1999		5.7	33.4				
V04	1996	45	11.78		1.32			<2	S6	1999		83.8	55.2				
V05	1996	213							S7	1999		19.1	133.1				
V06	1996	243							S8	1999		22.6	24.3				
V07	1996	98	4.78		1.33			<2	S9	1999		10.3	66.1				
V08	1996	457	1.78		0.92			<2	V1	1999	388						
V09	1996	65							V2	1999	214						
V10	1996	427							V3	1999	21						
V11	1996	269							V4	1999	121						
V12	1996	66							V5	1999	24						
V13	1996	57							V6	1999	21						
V14	1996	357							V7	1999	63						
V15	1996	71							V8	1999	46						
V16	1996	111							O1	1999		1.4	12.4				
V17	1996	56							O2	1999		2.1	23.4				
V18	1996	24							O3	1999		3.5	22.7				
V19	1996	121							O4	1999		3.5	44.9				
V20	1996	117							O5	1999		5.9	51.9				
V21	1996	745							O6	1999		6.2	11.1				
V22	1996	440						<2	O7	1999		5.4	38.9				
V23	1996	60	1.4		0.06				O8	1999		10.4	60.5				
V24	1996	263							O9	1999		4.5	30.9				
V25	1996	543							S10	2000	459	7.5	<2	0.025	0.91	1	1.1
V26	1996	87							S11	2000	44	6.2	<2	0.049	<0.15	<0.1	<2
V27	1996	12							S12	2000	918	4.8	<2	0.022	<0.15	2.6	5
V28	1996	64							S13	2000	90	23.7	6.9	0.012	<0.15	<0.1	<2
V29	1996	84							S14	2000	1135	7.7	4.4	0.094	<0.15	0.9	<2
V30	1996	114							S15	2000	1252	13.1	2.9	0.129	<0.15	0.4	<2
V31	1996	129							S16	2000	712	7.7	4.5	0.114	<0.15	1.4	<2
V32	1996	6							O10	2000	163	4.7	<2	0.006	<0.15	<0.1	<2
V33	1996	12							O11	2000	180	4.9	<2	0.018	<0.15	<0.1	<2
V34	1996	1014							O12	2000	343	3.7	<2	<0.005	<0.15	0.3	<2
V35	1996	584							O13	2000	342	12.7	5.7	0.064	<0.15	0.2	<2
V36	1996	225							O14	2000	367	5.9	13	<0.005	<0.15	0.2	<2
V37	1996	266							O15	2000	96	3.2	7.3	0.011	<0.15	<0.1	<2
V38	1996	295							O16	2000	380	8.5	8.1	<0.005	<0.15	0.5	<2
S1	1999		29.8	25.5					O17	2000	194	7.9	6.7	<0.005	<0.15	0.6	<2
S2	1999		18.4	28													

See Fig. 1 for sampling site location.
[a] Year of collection.

even reach 3200 $\mu g\ g^{-1}$ due also to different characteristics (TOC for Belgian canal = 52 g kg^{-1} and fine particles $<50\ \mu m = 70\%$ whereas average fine particles for this study = 21%). This difference is also observed for other heavy metals such as Cu, Pb, Cd, Ni. Hence, we can assume that sediments from the river Seine basin are less polluted with heavy metals than other rivers or canals situated in industrialised and densely populated areas, this difference being mainly due to the characteristics of the sediment such as the grain size or the organic content which are linked to the heavy metals contents.

To give a general idea of the metal contamination of sediments in the river Seine basin, we compared metal levels to the limits given by the Dutch decree on the quality of DM (circular on target values and intervention values for soil remediation, Ministry of Housing, Spatial Planning and Environment, updated 04/02/2000) which is often used as a reference in Europe. This decree gives two kinds of values: 'A' for Alert value which is

Table 6
Organic pollutant content (μg g^{-1}) in the sediment collected before dredging in the Seine basin in 1996, 1999 and 2000

Site		HC_{tot}	PAH_{tot}	Ali_{tot}	PCB_{tot}	Pesticides	Toluene	Phenols	Reference
Seine River, 1996–2000	n	61	38	33	20	15	15	20	This study
(sediment)	d_{10}	21	3.4	<2	<0.005	0.91	0.2	1.5	
	d_{50}	194	6.7	13	0.03	0.91	0.5	3.0	
	d_{90}	794	20.5	54	1.3	0.91	1.5	4.6	
	Min	3	1.4	<2	<0.005	0.91	0.2	1.1	
	Max	1252	84	133	22.9	0.91	2.6	5	
	Mean	302	10.9	23	1.4	0.91	0.8	3.0	
	S.D.	330	13.9	27	5.2	–	0.7	2.8	
	CV	109	127	118	367	–	91	90	
Seine River, 1995	Min		6	61	0.5				Meybeck et al., 1998
(TSS)	Max		11	100	3				
North of France River	Mean		770						Ruban et al., 1998
Louisiana waterways, USA	Mean		7.3						Mowat and Bundy, 2001
New Jersey, USA	Mean		17.4		0.6				Pollice et al., 1996
Rhine River, Germany	Mean		2.6		5.5				Van Der Hurk et al., 1997
Detroit River, USA	Mean		76.8		2.5				Besser et al., 1996

Number of samples (n), median (d_{50}), first and last decile (d_{10}, d_{90}), minimum and maximum (min, max), standard deviation (SD), and coefficient of variation in % (CV) of the characteristics given in Table 5.

the target value for sediments; and 'I' for 'Intervention' value for which sediments are considered as contaminated (Table 4). Most samples present metal contents either inferior to the 'A' limit, or between 'A' and an intermediate value [(A+I)/2)] for which sediments show moderate contamination. Fig. 2 shows the distribution of the number of sites showing different quality classes considering the Dutch decree. A few samples present concentrations above this average value (V04, V06, V09 for Hg, V04, V07, S14 for Zn and V03, V07, S10, S12, S14, S15, S16 for Cu), and only two samples show concentrations above the 'I' level: S1 and V02 for Hg. As a result, this comparison with the Dutch decree limits and the comparison with other values found in natural environments tend to indicate that most of the sediment samples from the river Seine basin are lightly to moderately contaminated with heavy metals.

3.1.2. Organic micropollutants

Total PAH levels, i.e. sum of 16 species listed by USEPA, found in the river Seine basin sediments (Table 5 and Table 6) are similar to the values in TSS previously collected in the river Seine (Meybeck et al., 1998). The sediments sampled in the present study contain 6.7 μg g^{-1} (median) of total PAH with a maximum of 84 μg g^{-1} and a minimum of 1.4 μg g^{-1}. TSS containing from 6 to 11 μg g^{-1} (median) of total PAH from upstream Paris to the sampling site at Chatou, downstream Paris, correspond to the site S2 in the present study (Fig. 1). Unlike heavy metals, total PAH contents in sediments and in TSS present similar levels.

Total PAH in the sediments from the river Seine basin show average values of 10.9 μg g^{-1}. In the North of France, a very industrialised region, river sediments contain far higher total PAH contents that can reach 770 μg g^{-1} (Ruban et al., 1998). Compared to the alert and intervention values from the Dutch decree (04/02/2000), most samples issued from the river Seine basin present concentrations below 'A' value and/or between 'A' and [(A+I)/2] values (Fig. 2). Still, some samples show values superior to [(A+I)/2], i.e. S1, S8, S13 and one sample shows a concentration above the 'I' intervention value, S6. But out of 38 samples, we can assume that the majority of the sediment samples are lightly to moderately contaminated if we consider the Dutch decree, and by comparing the case of the river Seine basin to other urban river basins.

Table 7
Metal and organic micropollutant content ($\mu g\ g^{-1}$) in the leachates of the sediments sampled in 1996, 1999 and 2000

Code	Y[a]	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn	Al	Fe	Mn	HC_{tot}	PAH_{tot}[b]	Ali_{tot}[c]
V03	1996	0.126	<0.02	<0.1	2.5	<0.005	<0.1	<0.1	6.5				<1	0.016	
V07	1996		<0.02	<0.1	<0.1	<0.005	<0.1	<0.1	1.4				<1		
V08	1996		<0.02	<0.1	<0.1	<0.005	<0.1	<0.1	0.7						
V10	1996	0.09	<0.02	<0.1	1.6	<0.005	<0.1	<0.1	1.3						
V32	1996	0.01	<0.02	<0.1	<0.1	<0.005	<0.1	<0.1	0.3						
V33	1996	0.02	<0.02	<0.1	<0.1	<0.005	<0.1	<0.1	0.4						
V34	1996	0.35	<0.02	<0.1	<0.1	0.06	<0.1	<0.1	3.9						
V35	1996	0.15	<0.02	<0.1	<0.1	<0.005	<0.1	<0.1	1.4						
V36	1996	0.06	<0.02	<0.1	<0.1	<0.005	<0.1	<0.1	0.9						
V37	1996	0.02	<0.02	<0.1	<0.1	0.14	<0.1	<0.1	<0.1						
V38	1996	0.02	<0.02	<0.1	<0.1	0.02	<0.1	<0.1	0.6						
S1	1999	<0.05	<0.02		<0.1	<0.005	<0.1	<0.1	<0.1	<1	0.1	0.1		<0.01	<0.02
S2	1999	<0.05	<0.02		<0.1	<0.005	<0.1	<0.1	0.1	0.2	0.2	0.1		<0.01	<0.02
S3	1999	<0.05	<0.02		<0.1	0.01	<0.1	<0.1	<0.1	<1	<0.1	<0.1		<0.01	<0.02
S4	1999	<0.05	<0.02		<0.1	0.009	<0.1	<0.1	<0.1	<1	<0.1	<0.1		<0.01	<0.02
S5	1999	<0.05	<0.02		<0.1	0.009	<0.1	<0.1	<0.1	<1	0.2	0.1		<0.01	<0.02
S6	1999	<0.05	<0.02		<0.1	0.009	<0.1	<0.1	0.2	<1	0.4	<0.1		<0.01	<0.02
S7	1999	<0.05	<0.02		<0.1	0.009	<0.1	<0.1	0.2	<1	<0.1	0.7		<0.01	<0.02
S8	1999	<0.05	<0.02		<0.1	0.009	<0.1	<0.1	<0.1	<1	0.1	0.2		<0.01	<0.02
S9	1999	<0.05	<0.02		<0.1	0.009	<0.1	<0.1	<0.1	<1	<0.1	0.2		<0.01	<0.02
V1	1999	<0.05	<0.02	<0.1	<0.1	<0.005	<0.1	<0.1	<0.1				<1		
V2	1999	<0.05	<0.02	<0.1	<0.1	<0.005	<0.1	<0.1	<0.1				<1		
V3	1999	<0.05	<0.02	<0.1	<0.1	<0.005	<0.1	<0.1	<0.1				<1		
V4	1999	<0.05	<0.02	<0.1	<0.1	<0.005	<0.1	<0.1	<0.1				<1		
V5	1999	<0.05	<0.02	<0.1	<0.1	<0.005	<0.1	<0.1	<0.1				<1		
V6	1999	<0.05	<0.02	<0.1	0.2	<0.005	<0.1	<0.1	<0.1				<1		
V7	1999	<0.05	<0.02	<0.1	0.1	<0.005	<0.1	<0.1	0.2				<1		
V8	1999	<0.05	<0.02	<0.1	0.1	<0.005	<0.1	<0.1	<0.1				<1		
O1	1999	<0.05	<0.02		<0.1	0.009	<0.1	<0.1	<0.1	<1	<0.1	0.3		<0.01	<0.02
O2	1999	<0.05	<0.02		<0.1	0.01	<0.1	<0.1	<0.1	<1	0.5	<0.1		<0.01	<0.02
O3	1999	<0.05	<0.02		<0.1	0.009	<0.1	<0.1	<0.1	<1	0.3	0.2		<0.01	<0.02
O4	1999	<0.05	<0.02		<0.1	0.01	0.2	<0.1	0.2	<1	0.1	0.2		<0.01	<0.02
O5	1999	<0.05	<0.02		<0.1	0.01	<0.1	<0.1	<0.1	<1	<0.1	0.2		<0.01	<0.02
O6	1999	<0.05	<0.02		<0.1	<0.005	<0.1	<0.1	<0.1	0.3	0.3	<0.1		<0.01	<0.02
O7	1999	<0.05	<0.02		<0.1	<0.005	<0.1	<0.1	<0.1	<1	<0.1	0.6		<0.01	<0.02
O8	1999	<0.05	<0.02		<0.1	<0.005	<0.1	<0.1	<0.1	<1	0.2	0.6		<0.01	<0.02
O9	1999	<0.05	<0.02		<0.1	<0.005	<0.1	<0.1	<0.1	<1	<0.1	0.3		<0.01	<0.02
S10	2000	<0.05	<0.02	<0.1	<0.1	<0.005	0.1	<0.1	0.2	<1	1.1	<0.1	<1	<0.01	<0.02
S11	2000	0.05	<0.02	<0.1	<0.1	<0.005	0.1	<0.1	<0.1	1.5	0.8	<0.1	<1	<0.01	<0.02
S12	2000	<0.05	<0.02	<0.1	<0.1	<0.005	<0.1	<0.1	0.5	1.1	0.3	1.6	<1	<0.01	<0.02
S13	2000	<0.05	<0.02	0.6	<0.1	<0.005	0.3	<0.1	0.2	<1	2.4	0.1	<1	<0.01	<0.02
S14	2000	<0.05	<0.02	<0.1	<0.1	<0.005	<0.1	<0.1	0.2	1.6	0.4	<0.1	<1	<0.01	<0.02
S15	2000	<0.05	<0.02	0.2	<0.1	<0.005	0.1	<0.1	0.2	<1	0.7	<0.1	<1	<0.01	<0.02
S16	2000	<0.05	<0.02	<0.1	<0.1	<0.005	<0.1	<0.1	0.3	1.6	0.4	0.1	<1	<0.01	<0.02
O10	2000	<0.05	<0.02	0.3	<0.1	<0.005	0.2	<0.1	0.2	<1	1.5	<0.1	<1	<0.01	<0.02
O11	2000	<0.05	<0.02	0.1	<0.1	<0.005	<0.1	<0.1	0.2	1.4	1.5	0.1	<1	<0.01	<0.02
O12	2000	<0.05	<0.02	<0.1	<0.1	<0.005	<0.1	<0.1	0.2	1.5	1.1	<0.1	1.5	<0.01	<0.02
O13	2000	<0.05	<0.02	0.2	<0.1	<0.005	0.1	<0.1	0.2	<1	1.4	<0.1	<1	<0.01	<0.02
O14	2000	<0.05	<0.02	0.3	0.2	<0.005	0.2	<0.1	<0.1	<1	1.3	<0.1	<1	<0.01	<0.02

Table 7 *(Continued)*

Code	Y[a]	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn	Al	Fe	Mn	HC_{tot}	PAH_{tot}[b]	Ali_{tot}[c]
O15	2000	<0.05	<0.02	<0.1	<0.1	<0.005	<0.1	<0.1	0.2	1.7	2	<0.1	<1	<0.01	<0.02
O16	2000	<0.05	<0.02	0.3	0.2	<0.005	0.2	<0.1	0.2	<1	1.4	<0.1	<1	<0.01	<0.02
O17	2000	<0.05	<0.02	<0.1	<0.1	<0.005	<0.1	<0.1	0.2	1.2	1	<0.1	<1	<0.01	<0.02

See Fig. 1 for sampling site location.
[a] Year of collection.
[b] $\mu g\ l^{-1}$.
[c] $mg\ l^{-1}$.

PCBs were also measured in this survey. Total PCB contents obtained in Seine sediment samples collected in 1992 to 1995 (Meybeck et al., 1998) varied from 500 to 3000 ng g^{-1}. In the sediments analysed for the present survey, total PCB content varied between detection limit levels (5 ng g^{-1}) and 1300 ng g^{-1}, with the exceptional site V03 at the Seine–Oise river junction, showing a concentration of 22 900 ng g^{-1}. This exceptional value at site V03 may be due to the influence of the Oise river and its associated pollution which joins the Seine river at this site, and also the industries and combined sewer overflows upstream at this site on the Seine river. Thus, we can assume that this range of total PCB concentrations corresponds to the one previously observed on the sediments measured in the river Seine basin (Meybeck et al., 1998). Compared to other river basins, sediments from the river Seine basin show lower total PCB amounts (mean = 1.4 $\mu g\ g^{-1}$) than sediments from the Rhine river in the Netherlands which presents an average level of 5.5 $\mu g\ g^{-1}$ (Van Der Hurk et al., 1997). Again, in that Dutch study, sediments presented loss on ignition of 14% on average, whereas in this study, the mean organic matter content is of only 8%. This difference might explain the PCB contents since PCB contents vary as a function of organic matter contents (Fowler, 1990). Compared to the Dutch decree, most samples from the river Seine basin showed total PCB concentrations below the 'A' value and between 'A' and [(A+I)/2] intermediate value (Fig. 2). Nevertheless, the concentration of sample V08 on Oise river was above [(A+I)/2] and concentrations of samples V03, V04 and V07 on Seine river downstream Paris were above 'I' value. Again, concerning the total PCB level, as for heavy metals and PAH, we can state that most of the sediment samples from the river Seine basin are lightly to moderately contaminated compared to Dutch criteria.

We also measured pesticide, toluene and phenols contents in the sediments: we do not discuss these results in the present paper since most values were found below detection limits (Table 5 and Table 6).

3.1.3. Leachates and pore water quality

Concentrations of micropollutants in the leachates obtained from the river Seine basin sediments are mostly below detection limits (Table 7): exceptions are rare. We also measured micropollutants in porewaters. Concentrations were also frequently below detection limits (Table 8). From these results, we can assume that there is only a minor short term risk of micropollutants release by DM. Alternatively, we suggest that the leaching test performed by mixing a 100 g/l suspension of sediment, during 24 h, is unable to simulate sediment diagenesis under anoxic conditions. Thus, it is difficult to anticipate the behaviour of micropollutants once the sediment has been dredged, unless we monitor the evolution of dredged materials with time in situ (Carpentier, 2002).

3.2. Geographic distribution and correlation analysis

Correlation analysis was performed to assess the probable link between micropollutants and physico-chemical parameters presented and discussed in a previous paper (Carpentier et al., 2002), as well as between micropollutants themselves (Table 9). The correlation between the sediments organic matter content and some heavy metals is significant ($r > 0.7$, $n > 46$, $P < 0.01$): Cr; Ni; As; Fe; Al;

Table 8
Metal and organic micropollutant content ($\mu g\ l^{-1}$) in porewaters of sediments sampled in 1996, 1999 and 2000

Code	Y[a]	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn	Al	Fe	Mn	HC_{tot}	PAH_{tot}	Ali_{tot}
S1	1999	<5	<2		10	<0.5	<10	<10	40	<10	168	1910		0.18	<20
S2	1999	8.5	<2		<10	<0.5	<10	<10	40	20	310	6970		0.2	40
S3	1999	8.6	<2		<10	0.73	<10	<10	30	<10	186	2350		<0.01	<20
S4	1999	<5	<2		<10	0.79	<10	<10	20	<10	215	5350		<0.01	<20
S5	1999	<5	<2		<10	0.69	<10	<10	20	<10	299	5670		<0.01	20
S6	1999	<5	<2		<10	0.74	<10	<10	30	<10	300	560		0.35	<20
S7	1999	10.8	<2		<10	1.06	<10	<10	10	10	1010	552		0.22	40
S8	1999	<5	<2		<10	0.5	<10	<10	50	<10	665	2950		0.27	<20
S9	1999	5.6	<2		<10	0.83	<10	<10	<10	<10	282	5620		<0.01	<20
V1	1999	9.4	<2	<10	<10	<0.5	26	<10	20				<50		
V2	1999	5.8	<2	<10	<10	<0.5	25	<10	20				<50		
V3	1999	<5	<2	<10	<10	0.73	<10	<10	10				*x*		
V4	1999	6.6	<2	<10	20	<0.5	<10	<10	30				<50		
V5	1999	<5	<2	<10	<10	<0.5	14	<10	20				<50		
V6	1999	<5	<2	<10	<10	<0.5	17	<10	20				<50		
V7	1999	<5	<2	<10	<10	<0.5	13	<10	20				<50		
V8	1999	<5	<2	<10	<10	<0.5	26	<10	20				190		
O1	1999	15.1	<2		<10	0.76	11	<10	40	<10	309	1230		<0.01	<20
O2	1999	11.8	<2		<10	0.7	<10	<10	30	<10	524	477		<0.01	<20
O3	1999	<5	<2		<10	0.66	<10	<10	<10	<10	170	521		<0.01	<20
O4	1999	<5	<2		<10	0.65	211	<10	40	<10	634	610		<0.01	<20
O5	1999	<5	<2		<10	0.76	<10	<10	20	<10	309	935		<0.01	<20
O6	1999	<5	<2		<10	<0.5	<10	<10	40	<10	191	423		<0.01	<20
O7	1999	<5	<2		<10	<0.5	<10	<10	40	20	1600	358		0.1	<20
O8	1999	<5	<2		<10	<0.5	<10	<10	30	10	1500	283		0.1	<20
O9	1999	<5	<2		<10	<0.5	<10	<10	30	<10	204	214		<0.01	<20
S10	2000												<50		
S11	2000														
S12	2000												80		
S13	2000														
S14	2000												120		
S15	2000												<50		
S16	2000												60		
O10	2000												<50		
O11	2000												<50		
O12	2000												<50		
O13	2000												<50		
O14	2000												<50		
O15	2000												<50		
O16	2000												<50		
O17	2000												<50		

See Fig. 1 for sampling site location.
[a] Year of collection.

and Mn; as well as the correlation between organic matter and total hydrocarbons ($r=0.63$, $n=61$, $P<0.01$). This kind of correlation has often been demonstrated in literature (Irwin et al., 1997). Also, as expected, fine particles percentage was correlated significantly to some heavy metal contents: Cr; As; Fe; Al; and Mn ($r>0.7$, $n>46$, $P<0.01$); and to Ni ($r=0.47$, $n=71$, $P<0.01$). Fine particles were also significantly correlated to total hydrocarbons ($r=0.36$, $n=61$, $P<0.01$). Concerning organic micropollutants and heavy metals, we found out that total hydrocarbon concentrations

Table 9
Correlation matrix of analysed parameters for sediment samples of the Seine basin from 1996, 1999 and 2000

	TOC	Density	Humidity	OM	Fine particles	HC_{tot}	PAH_{tot}	Ali_{tot}	PCB_{tot}	Toluene	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn	Al	Fe	Mn
TOC	1																				
Density	**−0.70**	1																			
Humidity	0.44	**−0.86**	1																		
OM	0.36	−0.77	**0.69**	1																	
Fine particles	−0.03	**−0.56**	**0.53**	**0.51**	1																
HC_{tot}	**0.58**	**−0.60**	**0.68**	**0.63**	0.36	1															
PAH_{tot}	0.16	0.11	−0.10	−0.17	−0.27	0.07	1														
Ali_{tot}	0.20	0.29	0.26	−0.32	−0.38	−0.19	0.29	1													
PCB_{tot}	0.34	−0.17	0.22	−0.35	−0.11	0.07	−0.04	−0.24	1												
Toluene	0.54	−0.43	0.59	0.59	0.38	0.58	−0.18	−0.26	0.21	1											
As	0.22	**−0.68**	0.40	**0.81**	**0.71**	**0.70**	−0.17	**−0.52**	−0.14	0.60	1										
Cd	**0.59**	−0.13	0.17	0.20	0.00	0.20	0.24	0.39	−0.05	0.54	0.05	1									
Cr	**0.47**	**−0.66**	**0.77**	**0.73**	**0.66**	**0.60**	−0.05	−0.03	0.11	0.45	**0.64**	0.37	1								
Cu	**0.62**	**−0.51**	**0.51**	**0.47**	0.28	**0.62**	0.13	−0.02	0.50	**0.75**	0.39	**0.47**	**0.71**	1							
Hg	0.27	0.18	−0.02	−0.07	−0.17	−0.07	0.23	0.04	0.19	**0.67**	−0.13	0.27	−0.06	0.23	1						
Ni	**0.48**	−0.62	**0.77**	**0.77**	**0.47**	**0.61**	−0.13	0.00	0.11	0.58	**0.60**	0.38	**0.91**	**0.51**	0.07	1					
Pb	0.32	−0.20	0.21	0.14	−0.12	0.34	0.16	0.19	0.47	**0.75**	0.01	**0.48**	0.25	**0.61**	0.30	0.17	1				
Zn	**0.50**	−0.58	**0.59**	**0.54**	0.30	**0.63**	0.08	0.05	0.49	**0.65**	0.40	**0.56**	**0.78**	**0.89**	0.18	**0.63**	**0.75**	1			
Al	0.26	−0.62	0.37	**0.82**	**0.76**	**0.78**	−0.24	**−0.47**	0.45	0.48	**0.94**	−0.01	**0.86**	**0.51**	−0.16	**0.62**	0.02	**0.52**	1		
Fe	0.12	−0.40	0.37	**0.75**	**0.80**	**0.57**	−0.35	**−0.49**	0.24	0.36	**0.91**	−0.06	**0.83**	0.34	−0.13	**0.63**	−0.16	0.37	**0.89**	1	
Mn	0.13	−0.44	0.07	**0.83**	**0.68**	**0.74**	−0.21	**−0.59**	0.39	0.60	**0.92**	−0.05	**0.86**	0.43	−0.14	0.40	−0.05	0.43	**0.84**	**0.83**	1

$n > 15$. The highly significant ($P < 0.01$) correlations are indicated in bold.

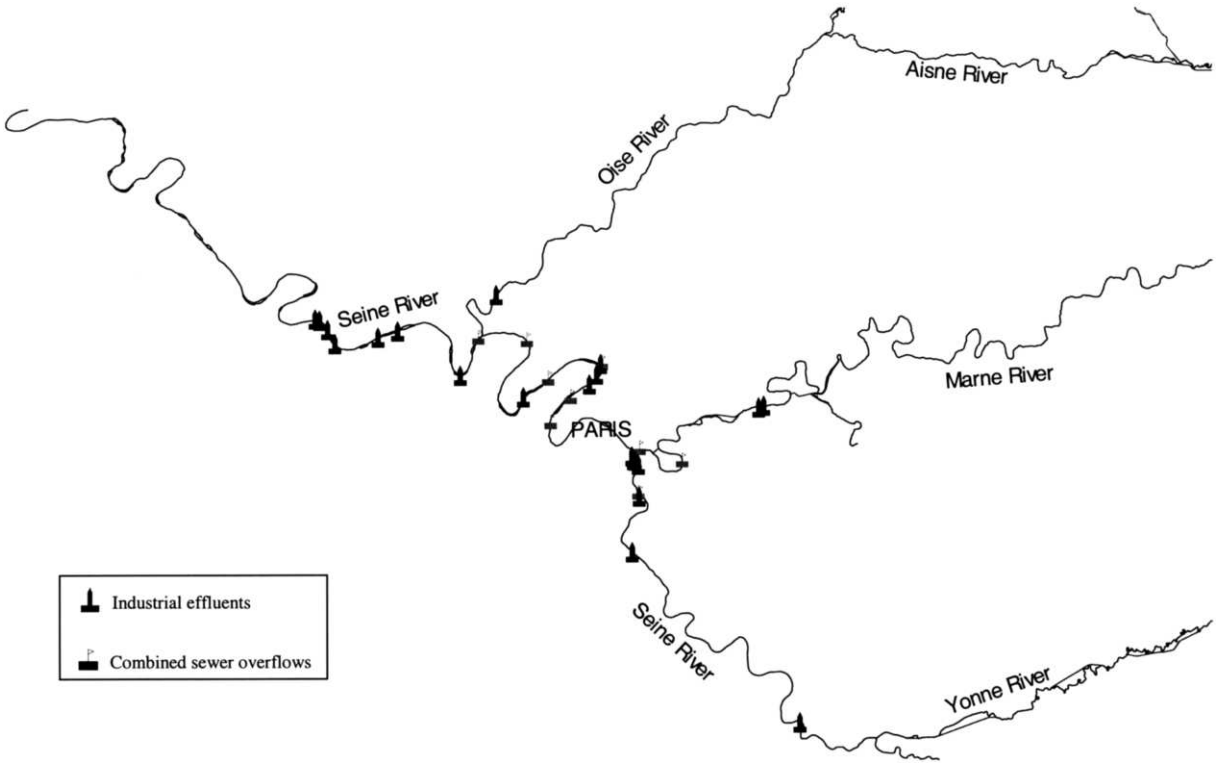

Fig. 3. Sites of major industrial effluents inventoried for this study and combined sewer overflows (listed in Meybeck et al., 1998) in the river Seine basin.

were significantly correlated ($r>0.7$, $n>33$, $P<0.01$) to some heavy metal concentrations: Cr; Zn; Ni; Cu; As; Al; and Mn, probably indicating the influence of industrial or urban runoff. But total PAH and total PCB levels were not correlated to one another or with heavy metals. A few heavy metals were correlated to one another: Cr with Zn; Ni; Cu; Fe; Al; Mn. The fact that Cr is correlated to Fe, Al and Mn, which are crustal metals, corresponds to the fact that part of Cr in the environment is natural. Its correlation with Zn, Ni and Cu could be related to the anthropogenic origin of Cr, since Ni, Cu and Zn are involved in many industrial processes (Rodier et al., 1996). Fe, Al and Mn are strongly correlated ($0.83<r<0.89$) because they are crustal metals and, therefore, belong to the matrix of the sediments (Rodier et al., 1996), but their content is also strongly linked to the As one ($0.91<r<0.94$), which is quite unexpected. These results suggest As is both adsorbed onto organic matter and charged surfaces of clays (Irwin et al., 1997).

We performed a geographic distribution analysis using Mapinfo® Geographic Information System in order to present a general overview of the micropollutant content of sediments along the whole river Seine basin, and to try and link the most polluted areas to the main point sources identified in the same region (combined sewer overflows were listed in Meybeck et al., 1998 and industrial effluents were inventoried for this study) (Fig. 3). Unlike physico-chemical parameters, micropollutants show a very clear pattern: highly contaminated sites are regularly situated in the same areas of the river Seine basin, whatever the micropollutant considered. Downstream all major source of pollutants, i.e. industrial effluent and/or combined sewer overflow sites, the sediment samples present higher micropollutant contents. Fig. 4 illustrates this relation with Zn: the eight 'hot

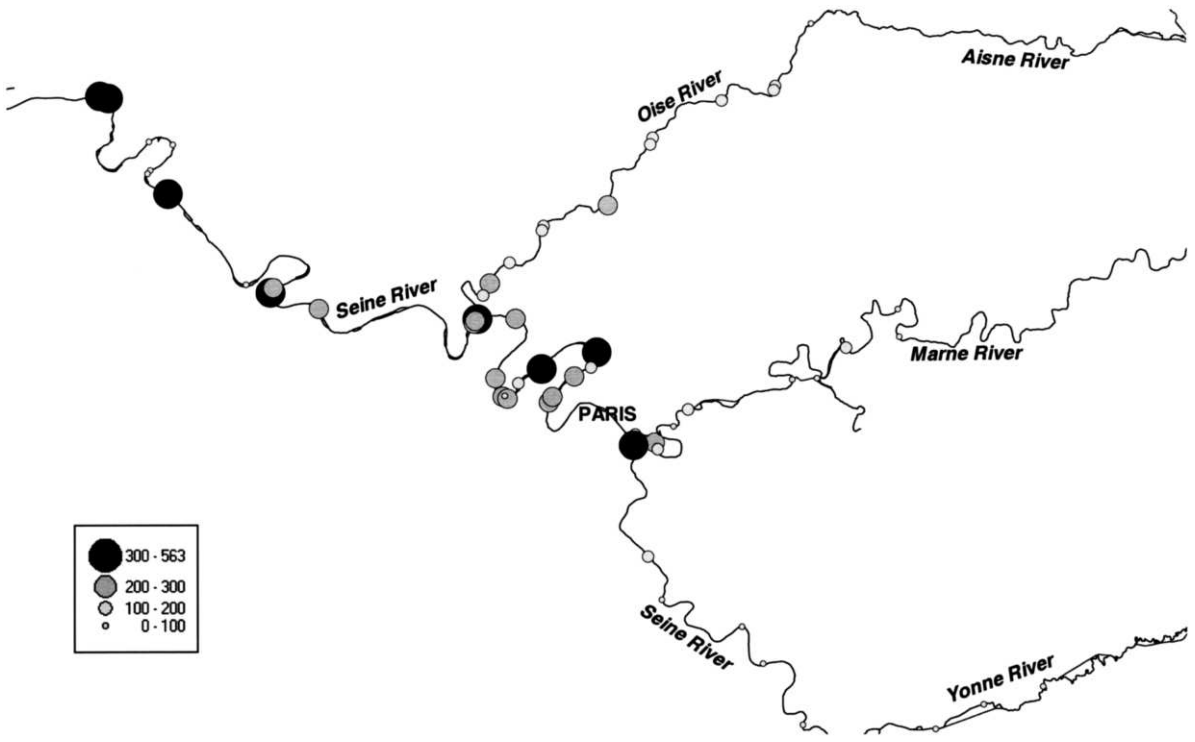

Fig. 4. Geographic distribution of Zn content ($\mu g\ g^{-1}$) in the sediment collected along the river Seine basin during 1996, 1999 and 2000 campaigns: highly contaminated sites correspond to the 'hot spots' (Fig. 1) and to the sites of possible pollution release (Fig. 3).

spots' correspond exactly to a cluster of industries and waste water treatment plants as well as combined sewer overflow sites. We can state that these contamination distribution maps represent a recent fingerprint of diverse pollution, since the specific areas sampled for this study are dredged frequently and, thus, the surface sediment that has been sampled was present and contaminated for less than 3 years. It is important to note that similar distribution maps were also found for Cu, Pb, Cr, As, Hg, Ni, Cd and total hydrocarbons, and to a lesser extent, for total PAH and PCB and Fe, Al and Mn.

4. Conclusions

Before dredging operations within the river Seine basin, the quality of the sediments to be dredged is regularly checked (once every 3 years) in order to determine the management strategy of dredged materials. In this survey, we present the results obtained for samples collected in 1996, 1999 and 2000 and suggest dredging and management strategies. Depending upon the sediment contamination, the following strategies are recommended: (1) dumping of light or moderately polluted sediments in quarries referring to the limits given by the Dutch decree on target values and intervention values for soil remediation, Ministry of Housing, Spatial Planning and Environment, updated 04/02/2000; (2) identifying some 'hot spot' sites and give the sediments dredged here a separate cleaning (sieving, hydrocycloning) in order to discriminate highly contaminated fine particles from sand or gravel which can be reused.

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