POLYCHLORINATED BIPHENYLS IN ST. LAWRENCE RIVER SEDIMENTS

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Management Perspective

Title: Polychlorinated biphenyls in St. Lawrence River sediments

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Perspective:

This manuscript reports new research results on settling and bottom sediments in the St. Lawrence River. Polychlorinated biphenyls (PCB) make up approximately 80% of the total organochlorine residue burden in both sediments and settling particulates, while DDT group insecticides, several chlorobenzene congeners and gamma-chlordane comprise the rest.

Significant differences in PCB levels between the various sites suggest nearby sources of PCB contaminants. However, upstream sources (Lake Ontario) also contribute strongly to the total contaminant burden in the river.

Seasonal resuspension and downstream movement of contaminated bottom sediments limit accumulation of these contaminants in the St. Lawrence River and its riverine lakes. According to our calculations, more than 90% of the incoming PCB load move through the system, at least over a full seasonal cycle.

Perspectives:

Ce document manuscrit présente les derniers résultats de travaux sur les sédiments qui se déposent et les dépôts de fond dans le fleuve Saint-Laurent. Les polychlorobiphényles (PCB) représentent environ 80 % de la charge totale de résidus organochlorés dans les sédiments et les particules qui se déposent, tandis que les insecticides du groupe DDT, plusieurs congénères de chlorobenzène et le gamma-chlordane constituent le reste.

Des écarts significatifs dans les concentrations de PCB entre les différents points de prélèvement indiquent la présence de sources proches de PCB. Cependant, des sources en amont (lac Ontario) contribuent aussi, dans une large part, à la charge de contaminants du fleuve.

La remise en suspension saisonnière et le mouvement vers l'aval des sédiments de fond contaminés limitent l'accumulation de ces substances dans le Saint-Laurent et ses lacs fluviaux. D'après nos calculs, plus de 90% de la charge de PCB qui pénètre dans le réseau, est charriée dans ce dernier pendant au moins un cycle saisonnier complet.

RÉSUMÉ

On a analysé des carottes de sédiments, prélevées à six stations, et des échantillons de pièges à sédiments, installés à sept stations, dans les lacs fluviaux du Saint-Laurent, afin de mesurer les concentrations de chlorobenzènes, de pesticides organochlorés et de polychlorobiphényles (PCB). On a procédé à l'analyse individuelle de trois sections d'un centimètre prélevées dans les trois centimètres supérieurs des carottes de sédiments. Trois des carottes de sédiments provenaient du même endroit que les pièges à sédiments.

Les résultats indiquent la présence d'isomères de di-, tri-, tétra-, penta- et hexachlorobenzène, de gamma-chlordane, de DDE et de DDT, ainsi que de résidus de PCB dans la plupart des sédiments en suspension et de fond. Dans les sédiments

de fond, les PCB représentaient en moyenne 81+/-11% (n=19) de la charge organochlorée totale. La concentration moyenne des PCB était de 135+/-138 ng/g (plage de 10 à 530 ng/g; n=19) dans les carottes, les valeurs les plus élevées ayant été relevées dans les lacs Saint-François et Saint-Louis.

Dans les échantillons des pièges à sédiments, les PCB constituaient en moyenne 78+/-13% (n=7) de la charge organochlorée totale. La concentration des PCB était en moyenne de 53+/-40 ng/g (variation de 20 à 110 ng/g; n=7) dans les particules en suspension des pièges, les valeurs les plus élevées ayant été relevées en aval de l'île Saint-Régis et dans le lac Saint-Pierre.

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ABSTRACT

Sediment cores from six stations and sediment trap samples from seven stations in the riverine lakes of the St. Lawrence River have been analyzed for chlorobenzenes, organochlorine pesticides and polychlorinated biphenyls (PCBs). The sediment cores were sectioned into the top three 1 cm sections which were analyzed individually. Three of the sediment cores were from the same sites as the sediment traps.

The results indicate the presence of di-, tri-, tetra-, penta- and hexachlorobenzene isomers, gamma-chlordane, DDE and DDT, and PCB residues in most of the suspended and bottom sediments. PCB in the bottom sediments made up on average 81+/-11% (n=19) of the total organochlorine burden. PCBs had a mean concentration of 135+/-138 ng/g (range 10 to 530 ng/g; n=19) in the core samples and were highest in Lakes St. Francis and St. Louis.

In sediment trap samples, PCBs made up on average 78+/-13% (n=7) of the total organochlorine burden. PCB concentrations had a mean of 53+/-40 ng/g (range 20 to 110 ng/g; n=7) in the trapped suspended particulates and were highest below Ile St. Regis and in Lake St. Pierre.

INTRODUCTION

With an average flow of 7700 m^3/s at the head and 12000 m^3/s at its mouth, the St. Lawrence River is Canada's largest river. Toxic organic contaminants enter its water primarily from three sources: (i) Lake Ontario, the source of the river; (ii) point sources along the river including inflowing tributaries, and (iii) by desorption and solubilization from particulates and sediments. Figure 1 shows the St. Lawrence River between Kingston, Ontario and Quebec City, Quebec.

Most of the St. Lawrence River is a fast flowing deep river (current velocity 0.5 to 1.5 m/s) with little deposition of suspended particulate matter. Consequently, both dissolved and particulate adsorbed contaminants are quickly flushed through the system with little opportunity for desorption and bioaccumulation via benthic organisms in these areas. In contrast, the three riverine lakes in the St. Lawrence River system (Figure 2), namely Lake St. Francis, Lake St. Louis and Lake St. Pierre, are large shallow basins with bathymetric and current conditions conducive to particle settling, burial, resuspension and chemical and biological contaminant transfer. Figure 3 gives a cross section of Lake St. Francis near station 44, between Pointe Beaudette on the north shore and Pointe Genier on the south shore. It shows the typical bathymetry of the St. Lawrence riverine lakes with shallow basins into which deep river channels are cut. Current measurements indicate that approximately one quarter of the

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total river flow in Lake St. Louis passes through the deep channel. However, the flows in the riverine lake basins vary strongly with the prevailing wind conditions and counter currents setting in a westerly direction have been noted in Lake St. Pierre (Kaiser, unpublished results, 1987). In the upper Great Lakes system, a very similar bathymetry is found for Lake St. Clair, a subject of intensive studies in recent years (UGLCC, 1988).

This paper describes results of PCB and selected organochlorine (OC) analyses of river sediments and settling particulates collected from several sites in the St. Lawrence River system riverine lakes. It explores the dynamics of contaminant transport in and through this system.

EXPERIMENTAL

Bottom Sediments

Sediment samples were collected with a benthos corer in October 1985 (stn. 62) and in June 1986 (stations 44, 48, 54, 59 and 95). The cores were sectioned under nitrogen atmosphere on board ship into the top three 1 cm sections. The sections were kept frozen until freeze drying followed by extraction as described below.

Suspended Sediments

Sediment traps consisting of plexiglas tubes 100 cm long with a 7 cm diameter (Charlton, 1983; Charlton and Oliver, 1986) were installed for two weeks in October 1985 at eight sites in Lakes St. Francis, St. Louis and St. Pierre. The sediment traps were secured so that their tops were 1.5 m above the lake bottom. Several grams of material were collected at each station, but due to the currents in the system, no attempt is made to equate deposition rates with sediment accumulation in the lakes.

Analysis

The analytical procedures were described in detail by Oliver and Nicol (1982). In brief, the samples were soxhlet extracted with acetone/hexane, the acetone removed with water and the hexane concentrated to the appropriate volume in Snyder and Kuderna-Danish condensors. Extract cleanup included passage through a small Pasteur pipette packed with 1 cm of sodium sulfate, 4 cm of acid treated silica gel and 2 cm of deactivated Florisil. Quantification of contaminants was carried out with dual capillary columns (30 m DB5 and 30 m DB17) and electron capture detection in a Varian 4600 gas chromatograph. Carrier gas was helium and a temperature program rate of 1 deg C/min was used between 50 and 250 C. Aroclor standards 1221, 1016, 1254 and 1262 were used in a ratio of 10:5:3.5:3 for calibration and quantification. Selected PCB congeners were also obtained from various sources and checked against the peak assignments made on the standard mixture.

RESULTS AND DISCUSSION

Table 1 gives the exact station locations and sample descriptions. Table 2 shows the observed concentrations for each sediment core subsection. The following contaminants were observed: two dichlorobenzene (DCB) isomers, three trichlorobenzene (TRICB) isomers, three tetrachlorobenzene (TECB) isomers, penta- (QCB) and hexachlorobenzene (HCB), pentachlorotoluene (PCT), hexachlorobutadiene (HCBD), octachlorostyrene (OCS), gamma-chlordane (CLD), the DDT group pesticides (DDE, DDD and DDT), mirex and polychlorinated biphenyls (PCB). Absent were 1,3-DCB, 2,4,5- and 2,3,6-TRICB, hexachloroethane, and alpha- and gamma-hexachlorocyclohexane (lindane).

Bottom Sediments

As apparent from the data in Table 2, the concentrations of the chlorobenzene congeners, HCBD, PCT, OCS, chlordane, the DDT group compounds and mirex were in the low ng/g (ppb) range in most samples. PCBs were the major constituents of the organochlorine contaminants with a mean of 81+/-11% of such residues for the 18 core sections and ranged from a low of 10ng/g (stn. 62, top section) to a high of 530 ng/g (stn. 48, bottom section). For all 18 sections, a mean of 139+/-140 ng/g PCB was found. However, if one removes the data for the Ottawa River/ Lac des deux Montagnes area (stn. 62) and that of the only high current velocity area at the eastern end of Lake St. Pierre (stn. 95), the mean PCB concentration increases to 193+/-144 ng/g PCB (n=15), avalue which is considered to be more representative of the riverine lake sediments.

The PCB levels also varied considerably with depth. For stations 44 and 48 in Lake St. Francis, PCB concentrations increased threefold from the surface sections (0 to 1 cm) to the bottom sections (2 to 3 cm). Little variation was noted in Lake St. Louis at the downstream station (stn. 59), while the upstream station (stn. 54), which is in the Ottawa River channel below Ile des Cascades, had the highest PCB level in the surface section. As noted, the sediment cores were taken in the summer when water turbulence is relatively low and comparatively little resuspension due to wave action is experienced.

Settling Particulates

Levels of the determined contaminants in settling particulates are given in Table 3. In general, the concentrations reflect those found in the nearby sediment cores. Except for some of the chlorobenzene congeners, PCT, OCS and mirex, the substances found in the cores were also present in the material collected in the sediment traps. Again, PCBs were the dominant contaminant group with 65 to 94% of the total organochlorine weight.

The sediment trap data generally confirm the information from the sediment cores. The somewhat lower levels of PCB in the settling particulates (53+/-40 ng/g PCB; n=7) may indicate a decline in recent PCB input to the system, relative to earlier and still in-place contamination, but the number of samples taken is insufficient to substantiate such a trend. A more detailed analysis of the bottom and suspended sediments for each lake is given below.

Lake St. Francis

Of the three stations in the Lake St. Francis area (stn. 40A, 44, 48), only stn 48 is in the open lake where most of the fine particulates are expected to settle. As shown in Table 2, the contaminant levels in this core were higher than in those from most other sites, particularly for the 2 to 3 cm bottom subsection. PCB levels of 530 ng/g were recorded for this section and a mean and standard deviation of 320+/-190 ng/g PCB for the entire core (0 to 3 cm). Very high concentrations of PCBs have previously been found in St. Lawrence River bottom sediments near Massena and Cornwall (Kauss et al., 1988; Merriman, 1987; Sloterdijk, 1985), in the depositional areas in Lake St. Francis (Sloterdijk, 1985) and in suspended sediments below Massena (Merriman, 1987). In biota, high PCB levels also occurred in that area as shown by data on spottail shiners and yellow perch (Sloterdijk, 1987) and native clams (Metcalfe and Charlton, 1988). The bottom section of the core at stn. 48 was also the only section with a measurable level (0.6 ng/g) of mirex. As known, mirex is essentially originating in the Lake Ontario basin (Kaiser, 1978) and has been found in Lake Ontario bottom sediments at levels up to 43 ng/g in the depositional zones (Holdrinet et al., 1978). Radiological dating of some of our sediment cores showed that these sediments are well mixed to a depth of at least 20 cm (S.R. Joshi, personal communication, 1988). Benthic samples taken with an Ekman dredge had few invertebrates including low numbers of oligochaetes and caddis fly larvae, but clams and particularly snails were present in large numbers at most sites (Charlton, unpublished data). Apart from some bioturbation by the benthic organisms, the shallow lake is subject to considerable wind stress leading to resuspension of the bottom sediments. Thus, the lake provides only a temporary retention of incoming particulates and contaminants associated with them.

Lake St. Louis

Lake St. Louis is located at the confluence of the Ottawa River tributary just above Montreal. Of the two stations, stn. 54 is in the Ottawa River channel near Ile des Cascades and stn. 59 in the depositional area off Pointe Claire. Station 62 in Lac des deux Montagnes is strictly in Ottawa River water, while both stn. 54 and 59 are significantly influenced by the Ottawa River water. For example, north of stn. 59, a distinct visual separation of the Ottawa River water (dark brown due to the high levels in humic substances) from the clearer St. Lawrence River water was evident during the sampling in the summer of 1986.

While PCB levels in the sediments of stn. 62 (35+/-38 ng/g) and stn. 54 (50+/-23 ng/g) were low in comparison with those in Lake St. Francis, they were high at stn. 59 (283+/-32 ng/g). This distribution pattern would be expected if the majority of PCBs were coming from sources upstream along the St. Lawrence River and possibly some local sources along the south shore of Lake St. Louis. Due to the comparatively small amount of particulates collected in the sediment trap at stn. 62, there was insufficient material (<1.0 g) for chemical analysis. However, levels of 50 ng/g PCB and 3.3 ng/g DDE in trapped sediments at stn. 54 indicate some input of these contaminants from the Ottawa River as well.

Recent investigations on the geochemistry and distribution of trace metals in Lake St. Louis' sediment cores concluded that sediments in the southern part of the lake are accumulated at a rapid rate of up to 20 cm in 5 years and that the lake provides only a temporary storage for metals associated with the clay-size sediment particles (Mudroch and Joshi, 1988).

Lake St. Pierre

In Lake St. Pierre, one station (95) at the western end of the lake was sampled for both bottom and settling sediments and three additional stations (112, 115 and 120) with sediment traps only. Of the latter, stn. 112 abd 115 are in relatively close proximity to each other (distance approximately 2.8 Km) in the main basin of the lake, just south of the shipping channel in water of approximately 7 m depth. In contrast, stn. 120 is just off Ile aux Sternes, where the river narrows and has a depth of approximately 12 m.

The amounts of particulates collected in the sediment traps decline with passage through the lake from 22 g (stn. 95) to 15 g (stn. 112) to 6.5 g (stn. 115), with a slight increase to 8 g at stn. 120. As mentioned above, stn. 120 is again in a high current zone and is more typical of a "river" station than a "lake" station.

The concentrations of PCBs in the particulates increase from stn. 95 (20 ng/g) to stn. 115 (110 ng/g). A similar increase of total PCB between station 95 (3.4 ng/g), stn. 112 (17.5 ng/g) and stn. 115 (34.5 ng/g) was observed in clams (Metcalfe and Charlton, 1988). This increase of PCB levels in downstream direction can be expected from the faster settling of the coarser and therefore more sandy material at the western end of the lake and the known association of these hydrophobic contaminants with the finer clay particles (Frank et al., 1977; Thomas, 1972).

The only sediment core sample from Lake St. Pierre (Table 1) confirms the data obtained from the particle traps (Table 2). Both the contaminant occurrences and concentrations are very similar in these samples. Three of the four Lake St. Pierre trap stations showed low concentrations of gamma-chlordane. This pesticide was also observed in sediment cores from stn. 62 (Ottawa River/ Lac des deux Montagnes) and stn. 95. The gamma-chlordane distribution therefore indicates its origin to be in the Ottawa River watershed with possible additions in the St. Lawrence River watershed below Lake St. Louis.

Contaminant Storage and Flux

It is of interest to compare the total PCB burden stored in the riverine lake sediments with the annual flux of PCB through the river. Unfortunately, the sediment trap data are not suitable to estimate the flux of settling particles. However, accurate sedimentation rates are available from radioactivity dated cores. Recent measurements by Mudroch and Joshi (1988) indicate a deposition rate of 20 cm in 30 years in the eastern, depositional zone of Lake St. Louis.

If one assumes that approximately 5% (12.5 km^2) of the total area (IWD, 1973) of Lake St. Francis is a true depositional zone, as indicated from the study by Sloterdijk (1985), and a total of 20 cm sediment accumulation with a mean PCB concentration of 200 ng/g (stations 44 and 48), the amount of PCB stored in Lake St. Francis sediments is calculated to approximately 1,250 kg. This compares with an annual PCB flux through Lake St. Francis of 468 kg (mean of two stations), of which 210 kg is found in particulates collected by sequential centrifugation with both a Westphalia and a Sorvall centrifuge (Kaiser et al., 1988). Given the general decline of PCB levels in fish and waterfowl in the Great Lakes basin since the ban on the use of PCB in 1977, it is reasonable to assume that also the PCB levels in particulates in the St. Lawrence River were higher, perhaps by a factor of two or three during the major deposition period of these sediments. Consideration of this assumption would bring the ratio of PCB load in the lake sediments to PCB flux on suspended particles to somewhere around 1250:600, or, approximately 2:1. This means that Lake St. Francis provides a storage of approximately two years flux only, in other words, approximately 93% of the incoming particulate-bound PCB load continues to move downstream. An analogous calculation for Lake St. Pierre, using a PCB concentration of 28 ng/g (stn. 95), a lake surface area of 350 km^2 (IWD, 1973) and, because of its more even bathymetry, a 10% depositional area, results in a lake sediment burden of 490 kg of PCB. This compares with an annual flux of 470 kg PCB on suspended particulates (Kaiser et al., 1988), indicating a similarly small net retention of PCB in Lake St. Pierre sediments.

CONCLUSIONS

Bottom sediment core and suspended sediment trap samples from the riverine lakes in the St. Lawrence River indicate the presence of a variety of organochlorine contaminants at most sites. Polychlorinated biphenyls average approximately 190 ng/g in the bottom sediments, representing some 85% of the total organochlorine residues. The remaining 15% are mainly composed of several chlorobenzene congeners and DDT group pesticides. Low levels of other compounds, including gamma-chlordane, hexachlorobutadiene, pentachlorotoluene, octachlorostyrene and mirex were also observed at several stations.

The distribution of PCBs and the other observed susbstances indicates sources to be both upstream (Lake Ontario) as well as within the St. Lawrence River system. Low levels of gamma-chlordane appear to be contributed by the Ottawa River.

The results are interpreted to indicate some settling of fine suspended particulates and adsorbed contaminants in the riverine lakes of the St. Lawrence River. However, seasonal resuspension and downstream movement of contaminated bottom sediments limit accumulation of these contaminants in the river and riverine lakes. Our calculations indicate that approximately 93% of the PCB load entering Lake St. Francis and an even higher percentage of that entering Lake St. Pierre pass through these lakes.

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TABLE 1

Station locations and descriptions.

Station No.	Latitude (N)	Longitude (W)	Sample[a]	Location
40-A	45 02'02"	74 36'01"	T	Lake St. Francis
44	45 02'19"	74 19'27"	S,T	Lake St. Francis
48	45 13'15"	74 12'16"	S	Lake St. Francis
54	45 20'19"	73 55'30"	S,T	Lake St. Louis
59	45 24'43"	73 49'39"	S	Lake St. Louis
62	45 26'52"	74 03'02"	S,T	Lac des deux Montagnes
95	46 07'36"	72 56'59"	S,T	Lake St. Pierre
112	46 13'59"	72 45'38"	T	Lake St. Pierre
115	46 14'53"	72 44'03"	T	Lake St. Pierre
120	46 16'42"	72 38'06"	T	Lake St. Pierre

[a] S: bottom sediment; T: suspended sediment trap.

TABLE 2

Contaminant concentrations in bottom sediment samples, values in ng/g dry weight; for abbreviations see text.

AREA	Lake St. Francis			Lake St. Francis			Lake St. Louis		
STATION	44	44	44	48	48	48	54	54	54
DEPTH (cm)	0-1	1-2	2-3	0-1	1-2	2-3	0-1	1-2	2-3
1,4-DCB		4.20	4.60						
1,2-DCB									
1,3,5-TRCB	2.00	6.10	8.40		0.88	1.30	2.40	0.96	
1,2,4-TRCB	3.20	5.30	7.00	2.20	4.30	4.80	5.90	4.10	4.00
1,2,3-TRCB			0.24			0.32			
HCBD	0.06	0.15	0.19	0.34	0.31	0.27		0.27	
1,2,3,5-TECB	0.47	1.20	1.60		0.39	0.54			
1,2,4,5-TECB	0.86	1.70	2.20	0.75	1.20	1.40	1.30	0.80	
1,2,3,4-TECB	0.62	0.92	1.20	0.50	0.85	0.92	0.90	0.67	
QCB	1.10	2.20	3.10	0.57	0.90	1.20	1.90	1.00	1.00
PCT		0.22	0.28						
HCB	1.70	2.80	3.70	2.30	1.20	1.50	2.60	1.90	1.50
OCS		0.11	0.12			0.12			
CLD	0.40					0.14	0.91	0.95	
DDE	3.10	4.80	5.60	2.20	3.10	5.20	5.30	3.10	3.40
DDD	2.30	4.60	6.10	1.40	1.30	2.40	6.40		
DDT							1.80		
Mirex						0.58			
PCB	46.00	91.00	130.00	160.00	270.00	530.00	140.00	66.00	33.00

AREA	Lake St. Louis			Lac des deux Mont.			Lake St. Pierre		
STATION	59	59	59	62	62	62	95	95	95
DEPTH (cm)	0-1	1-2	2-3	0-1	1-2	2-3	0-1	1-2	2-3
1,4-DCB					6.00	6.30	2.90	2.80	2.60
1,2-DCB								1.30	1.50
1,3,5-TRCB			2.70						
1,2,4-TRCB	6.70	6.40	6.90	1.50	1.70	1.80	1.60	1.50	1.30
1,2,3-TRCB								0.23	
HCBD						1.20	0.22	0.14	
1,2,3,5-TECB		1.10	1.10				0.14		
1,2,4,5-TECB		2.00	2.10				0.18	0.18	0.15
1,2,3,4-TECB	1.40	1.40	1.30		0.27	0.52	0.25	0.39	0.27
QCB	3.10	2.80	3.10			0.46	0.34	0.31	0.28
PCT									
HCB	3.70	3.40	3.50			6.70	0.57	0.49	0.43
OCS									
CLD					0.40	0.41	0.15		
DDE	7.70	6.00	6.00	1.60	1.90	3.40	0.51	0.41	0.40
DDD	9.20	2.70	3.90		2.00	3.30	1.40	3.80	1.50
DDT									
Mirex									
PCB	320.00	260.00	270.00	10.00	17.00	79.00	34.00	20.00	30.00

TABLE 3

Contaminant concentrations in trapped suspended sediments, values in ng/g dry weight; for abbreviations see text.

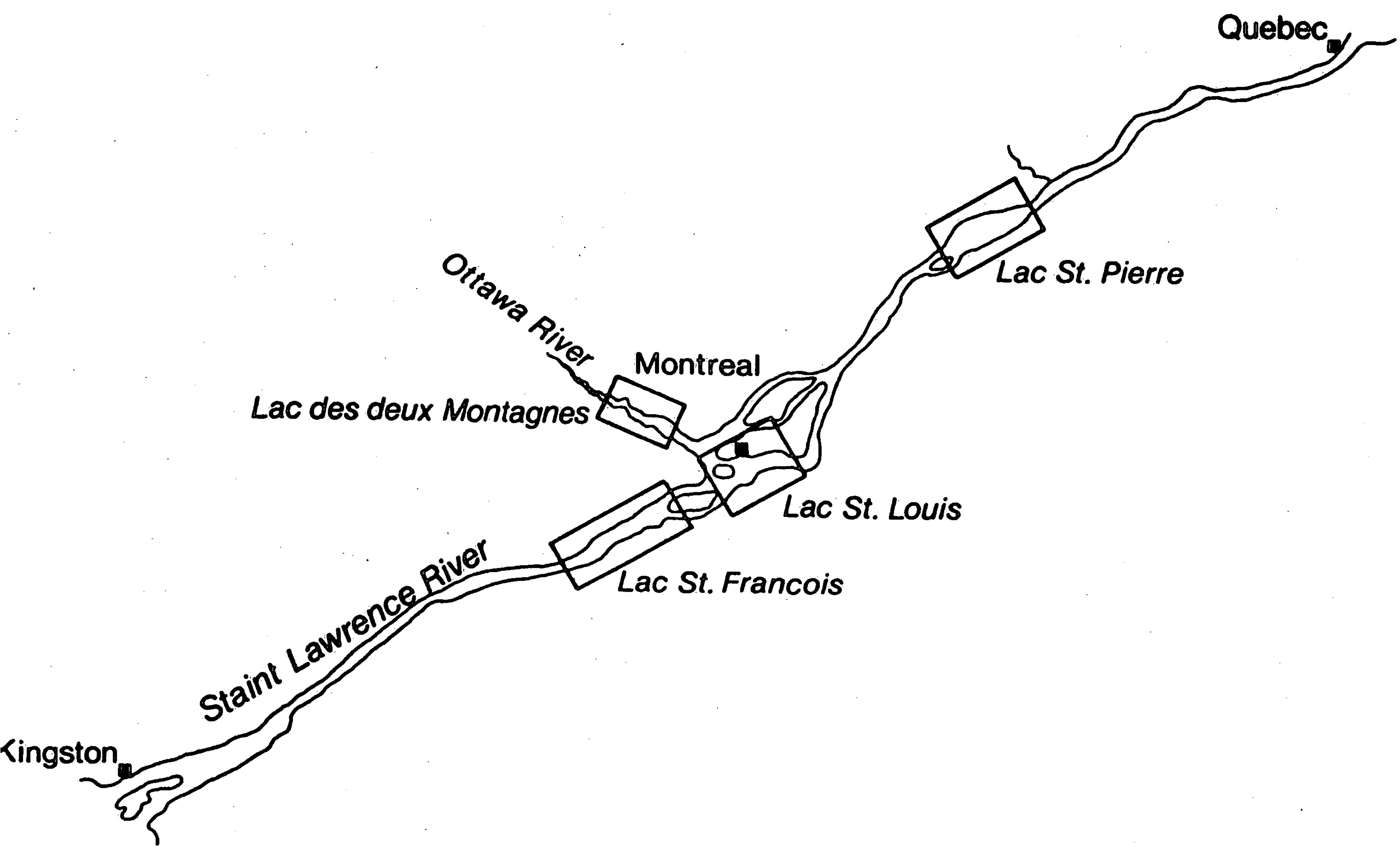
STATION WEIGHT [g]	40-A 5.7	44 1.8	54 1.1	95 22.0	112 15.0	115 6.5	120 8.0
1,4-DCB				2.60	4.40		7.00
1,2-DCB							
1,3,5-TRCB							
1,2,4-TRCB	3.30	6.50		1.60	1.90	1.90	2.80
1,2,3-TRCB				0.22	0.22		0.42
HCBD	0.27	0.35					
1,2,3,5-TECB							
1,2,4,5-TECB	0.58						
1,2,3,4-TECB	0.70			0.17	0.22	0.70	0.22
QCB	0.64	2.40	1.20	0.70	0.22	0.73	0.43
PCT							
HCB	1.10	3.40	2.70	0.24	0.35	1.10	0.40
OCS							
CLD				0.14	0.18		0.96
DDE	2.40	3.10	3.30	0.52	0.78	0.80	1.50
DDD	1.80			0.71	1.40	1.30	2.00
DDT					0.55		1.50
Mirex							
PCB	110.00	29.00	50.00	20.00	23.00	110.00	32.00

Fig. 1. The St. Lawrence River system between Kingston, Ontario and Quebec City, Quebec and sampling sites; rectangles indicate riverine lake areas which are detailed in Fig. 2.

Fig. 2. The St. Lawrence River system riverine lakes and sampling sites.

Fig. 3. Cross section of Lake St. Francis near station 44.

Fig. 4. Cross section of Lake St. Pierre, near station 115.



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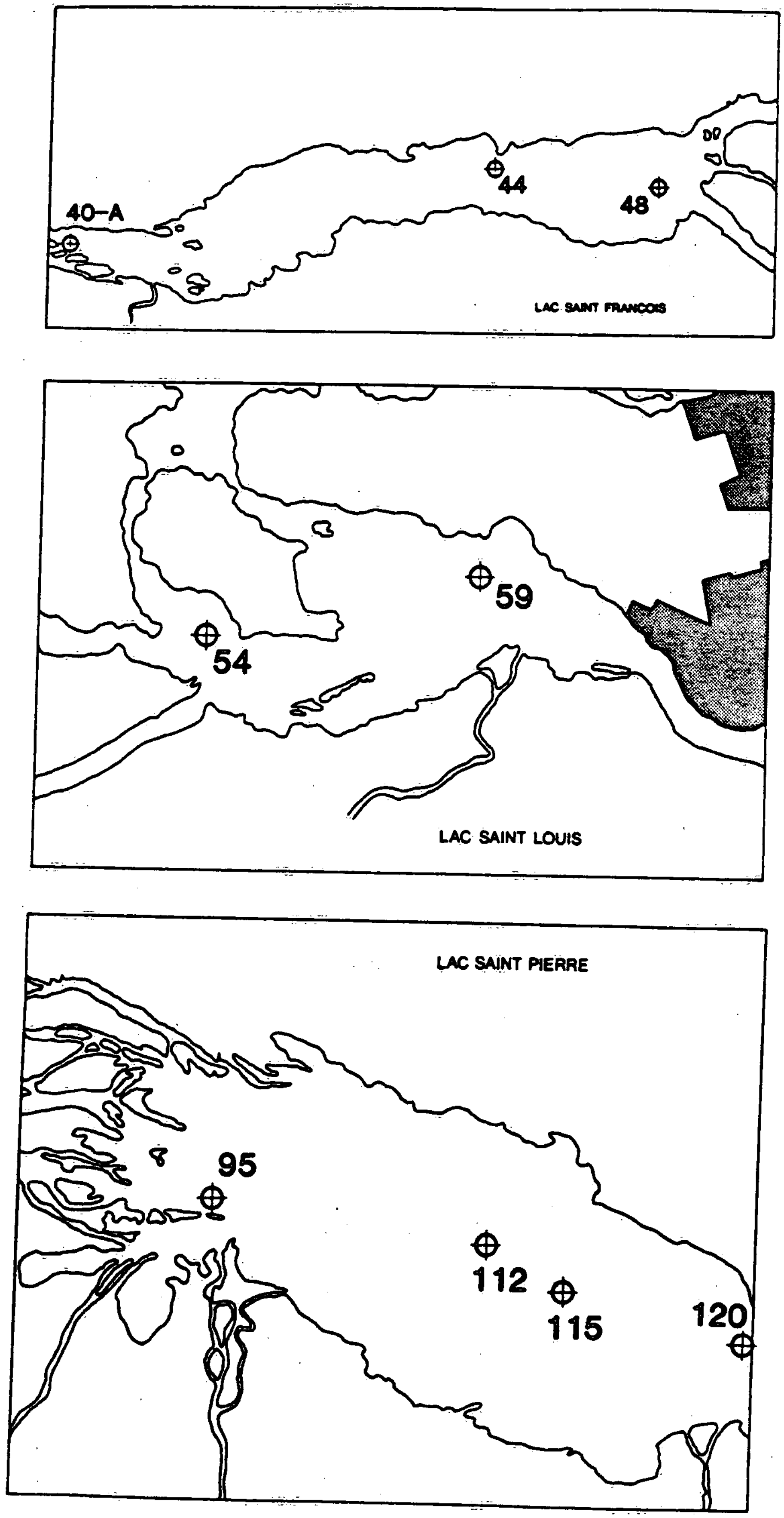

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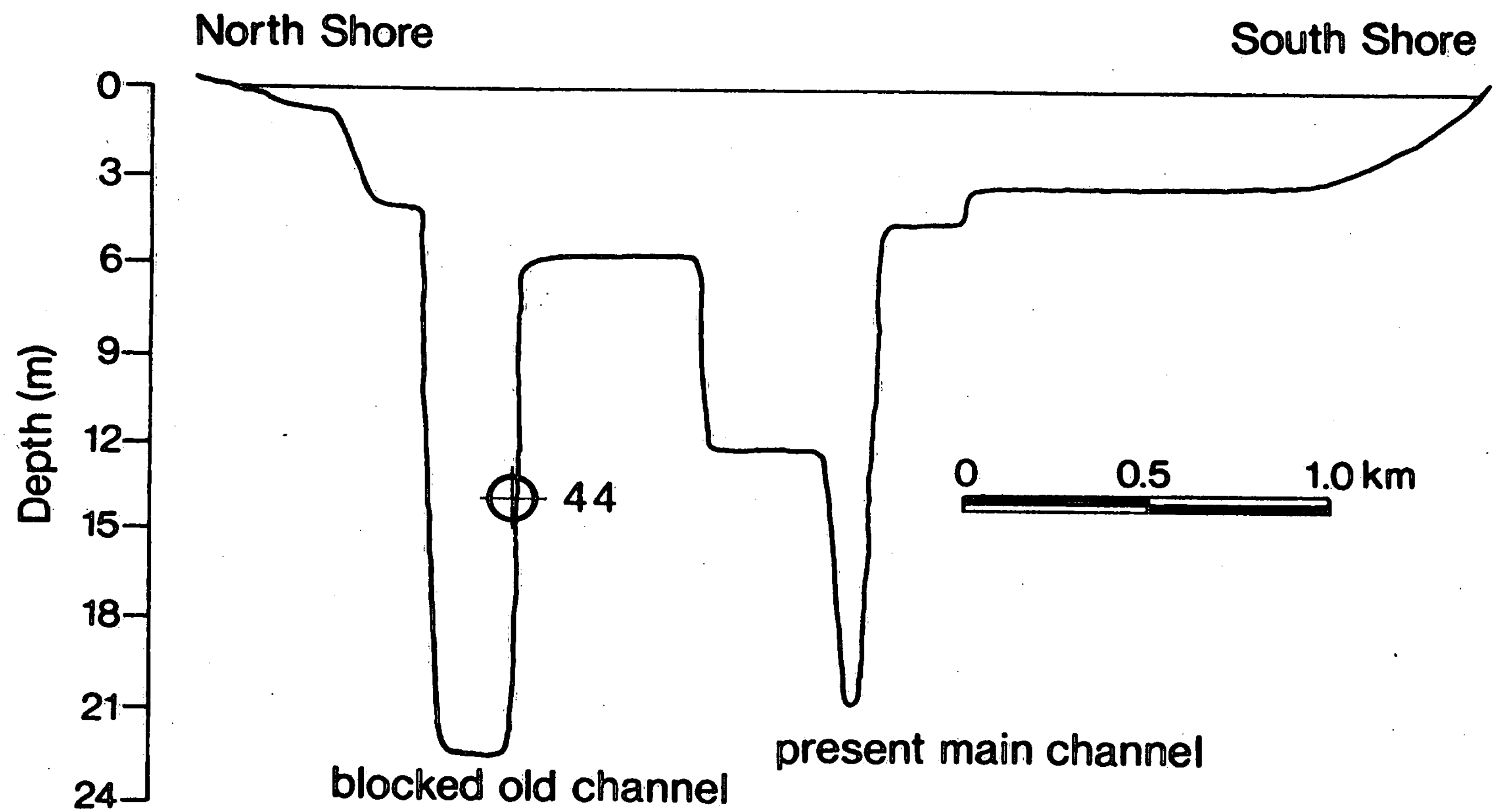

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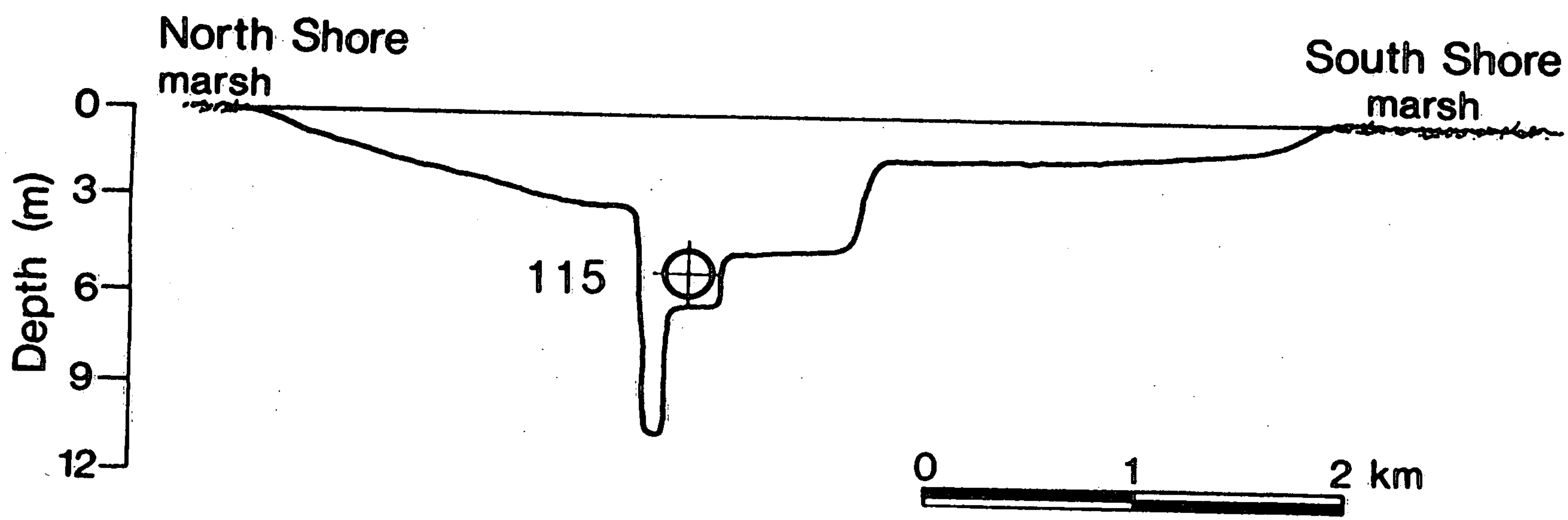

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