FRESHWATER MUSSELS AS BIOMONITORS FOR ORGANIC INDUSTRIAL CONTAMINANTS AND PESTICIDES IN THE ST. LAWRENCE RIVER

by

J.L. Metcalfe[1] and M.N. Charlton[2]

[1]Rivers Research Branch
[2]Lakes Research Branch
National Water Research Institute
Canada Centre for Inland Waters
Burlington, Ontario L7R 4A6

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ABSTRACT

Native mussels (Elliptio complanata and Lampsilis radiata radiata) were collected from 17 stations in the St. Lawrence River between Lake Ontario and Trois Rivieres and from 3 stations in a major tributary, the Ottawa River, in October, 1985. Mussels were solvent-extracted and analyzed individually by dual capillary column gas chromatography for 7 organochlorine pesticides, 11 chlorobenzenes, octachlorostyrene and 63 PCB congeners. Bioconcentration patterns for contaminants in mussel tissues implicated Lake Ontario as the source of Mirex and DDT derivatives to the system and the Grass River as the major source of PCBs. Numbers of PCB congeners in mussels increased from 21-27 in the upper river to 56-59 in the Cornwall/Massena industrial core, mainly due to the appearance of di-, tri- and tetrachlorobiphenyls; an average of 43 congeners persisted as far downstream as Lac Saint-Pierre. Concentrations of most contaminants in mussels from the Ottawa River were 50-75% lower than the lowest values reported for the St. Lawrence River. This study provides information on the origin, bioavailability and persistence of organic contaminants in the St. Lawrence River ecosystem.

RÉSUMÉ

Des moules de la région (Elliptio complanata et Lampsilis radiata radiata) ont été prélevées en octobre 1985 à 17 sites dans le fleuve St-Laurent entre le lac Ontario et Trois-Rivières et à 3 sites dans un important tributaire, la rivière des Outaouais. Les moules ont été extraites par solvant et un dosage individuel de 7 pesticides organochlorés, 11 chlorobenzènes, un octachlorostyrène et 63 congénères des BPC a été réalisé par chromatographie en phase gazeuse sur colonne capillaire double. D'après les bioconcentrations des contaminants dans les tissus des moules, le lac Ontario représenterait une source de dérivés du DDT et de Mirex et la rivière Grass, la principale source de BPC. Le nombre des congénères de BPC dans les moules a augmenté de 21-27, dans le cours supérieur du fleuve, à 56-59, dans la région industrielle de Cornwall-Massena, surtout à cause de l'apparition des di-, tri- et tétrachlorobiphényles. En moyenne, 43 congénères ont persisté en aval jusqu'au lac St-Pierre. Les concentrations de la majorité des contaminants présents dans les moules provenant de la rivière des Outaouais étaient de 50-75 % inférieures aux valeurs les plus faibles rapportées pour le fleuve St-Laurent. La présente étude fournit des renseignements sur l'origine, la biodisponibilité et la persistance des contaminants organiques dans l'écosystème du fleuve St-Laurent.

MANAGEMENT PERSPECTIVE

The purpose of this study was to evaluate native unionid mussels as biomonitors for organic contaminants in the St. Lawrence River. Because mussels are large, abundant, sedentary, non-selective in terms of habitat and easily standardized for sources of biological variability, they are ideal candidates for biomonitoring. We found that native mussels (Elliptio complanata and Lampsilis radiata radiata) accumulated the major organic industrial contaminants and pesticides of concern in the system. Spatial distribution patterns for contaminants in mussel tissues generally reflected the known distribution of these compounds in bottom sediment and clearly identified the major sources of several important contaminants, including DDT derivatives, Mirex and PCBs. Congener-specific PCB analysis provided information on the persistence of various PCB compounds in mussels living downstream of the point source. Biomonitoring with mussels has several potential applications. It may be used to complement or supplement traditional ambient water and sediment monitoring programs, primarily to provide information on bioavailability. It can also be used to define the impact zone of point source pollution and to serve as a feedback mechanism for determining the effectiveness of pollution control measures as they are implemented.

PERSPECTIVE - GESTION

L'objet de la présente étude est d'évaluer l'utilité des moules de la famille des unionidés que l'on trouve dans la région pour la biosurveillance des contaminants organiques dans le fleuve St-Laurent. Étant donné que les moules sont grosses, abondantes, sédentaires, non sélectives en terme d'habitat et faciles à normaliser en fonction des sources de variabilité biologique, elles consistuent des candidates idéales à la biosurveillance. D'après les auteurs, les moules de la région (Elliptio complanata et Lampsilis radiata radiata) absorbent les principaux pesticides et contaminants industriels organiques dont on se préoccupe dans le système. Les configurations spatiales de distribution des contaminants dans les tissus des moules réflètent généralement la distribution connue de ces composés dans les sédiments de fond et permettent d'identifier de façon claire les principales sources de plusieurs contaminants importants, notamment les dérivés du DDT, le mirex et les BPC. Le dosage des BPC spécifiques aux congénères permet d'obtenir des renseignements sur la persistance de divers composés de BPC dans les moules situés en aval de la source. L'utilisation des moules pour la biosurveillance présente plusieurs applications potentielles. En effet, l'utilisation des moules peut servir à compléter ou à remplacer les programmes traditionnels de surveillance des sédiments et de l'eau, surtout pour fournir des renseignements sur la biodisponibilité. Cette technique peut également être utilisée pour définir la zone d'impact de la pollution ponctuelle et servir de mécanisme de feedback pour la détermination de l'efficacité des mesures de contrôle des polluants, à mesure que celles-ci sont mises en application.

INTRODUCTION

The upper St. Lawrence River receives toxic chemicals from the Great Lakes, from tributaries draining a highly industrialized area of New York State and from municipal and industrial effluents discharged directly into the river from both the Canadian and American shores. Among the organic contaminants of particular concern, because they have been detected most frequently and in the highest concentrations in water, sediment and/or aquatic organisms, are PCBs, chlorobenzenes and organochlorine pesticides.

The purpose of this study was to determine the distribution of organic contaminants in native mussels throughout the St. Lawrence River, in order to provide a measure of their bioavailability and persistence in organisms occupying a low trophic level in the system. This information, in turn, gives a general indication of the potential for food chain biomagnification of these contaminants and early warning of changes in environmental pollution which will ultimately affect organisms at higher trophic levels.

Mussels are ideal biomonitors for high energy systems such as the St. Lawrence River not only because of their well-known ability to bioconcentrate organic contaminants (Miller et al., 1966; Bedford et al., 1968; Elder and Mattraw, 1984; Kauss and Hamdy, 1985), but because they integrate contaminant uptake from the water, sediment and suspended particulates by virtue of their benthic, filter-feeding lifestyle. Furthermore, mussels are sedentary animals which are representative of well-defined areas. They are generally abundant in large rivers, occupy a wide range of habitats, and are large enough to be analyzed individually. Finally, mussels may contribute to the diets of many wildlife species including a bottom-feeding fish, the freshwater drum (Bur, 1982).

MATERIALS AND METHODS

Sample Collection and Preparation

Native mussels (F. Unionidae) were collected by Ponar dredge from 20 stations in the Ottawa and St. Lawrence Rivers in October, 1985 (Fig.1). These stations included #s 28, BP and 29 in the upper St. Lawrence River, #30 at the mouth of the Grass River, #37 located 15 km downstream of #30, #s 42, 44 and 47 in Lac Saint-Francois, #s 55 and 59 in Lac Saint-Louis, #s 95, 103, 107, and 112-115 in Lac Saint-Pierre and #s 166, 165 and 65 in the Ottawa River, which is a major tributary to the St. Lawrence. At the time of collection, mussels were rinsed with river water to remove adhering sediment, then wrapped in pre-fired aluminum foil and frozen. They were later identified, thawed and shucked in the laboratory and their reproductive condition noted. Soft tissues were placed individually in solvent-washed glass jars and weighed wet, then refrozen for storage prior to analysis. Mussels were aged by preparing thin sections of the shells, according to the methods of Clark (1980) and Kennish et al. (1980), and counting the internal growth rings.

Analytical Methods

All mussels were analyzed for the 7 organochlorine pesticides, 11 chlorobenzenes (CBs), octachlorostyrene and 63 polychlorinated biphenyl (PCB) congeners listed in Table 1. The analytical methods have been described in detail elsewhere (Oliver and Niimi, 1988). The individual mussels were thawed and ground with anhydrous Na_2SO_4, then soxhlet-extracted for 16 hours with 59:41 acetone:hexane. The extracts were back-extracted with water to remove the acetone and the hexane layer was then concentrated to a volume of 1 mL using a Snyder, followed by a Kuderna-Danish, condenser. This extract was cleaned up on an 8 mm I.D. pasteur pipette column packed with 1 cm Na_2SO_4 (top), 4 cm 40% H_2SO_4 on silica gel and 2 cm of Florisil (deactivated with 5% water). Prior to extraction, a series of surrogate spikes (1,3-dibromobenzene, 1,3,5-tribromobenzene, 1,2,4,5-tetrabromobenzene, 2,3,5,6-tetrachlorobiphenyl and octachloronaphthalene) were added to the sample. Recoveries of the surrogates indicated that the procedures efficiently recovered the analytes. A solvent blank was run and none of the study chemicals were detected. To determine analytical reproducibility, a duplicate analysis was performed on one mussel from station #59.

The contaminants were identified and quantified by dual capillary column (30 meter HP5 and HP17) on a Hewlett Packard 5890 gas chromatograph. The program rate was 50°C (2 minute hold), 2°C/min. to 250°C (30 minute hold). The injector temperature was 250°C and the detector was at 350°C. Hydrogen was used as the carrier gas. For congener-specific determination of PCBs, a mixture of Aroclors 1221, 1016, 1254 and 1262 was used for instrument calibration. To confirm peak identities and quantities, a series of 50 commercially available PCB congeners and 51 PCB congeners obtained from the National Research Council of Canada were run. Excellent agreement between secondary and primary standards was found. Detection limits for PCBs, pesticides, octachlorostyrene and higher chlorinated benzenes were 0.01 ng/g on a wet weight basis. Detection limits for di- and trichlorobenzenes were 0.50 and 0.05 ng/g, respectively.

RESULTS AND DISCUSSION

Detailed Description of Mussels Collected

Thirty-eight mussels were collected from the 20 sampling stations, including 26 *Elliptio complanata*, Subf. Ambleminae, and 12 *Lampsilis radiata radiata*, Subf. Lampsilinae (Table 2). All were successfully aged, with the exception of one *E. complanata* from station #55 which had a heavily eroded shell. *E. complanata* specimens

ranged in age from 6 to 16 years and L. r. radiata from 3 to 10 years; the mean age of E. complanata (10 years) was significantly greater, at $p < 0.05$, than that of L. r. radiata (7 years). However, as L. r. radiata tended to be heavier at a given age than E. complanata, the mean soft tissue wet weights for the two species (13.88 and 15.90 g, respectively) did not differ significantly. Seven specimens, including 6 L. r. radiata and 1 E. complanata, were gravid. This was not surprising as E. complanata, which is hermaphroditic, is a short-term breeder with its reproductive season occurring in late spring and early summer, while L. r. radiata, which has separate sexes, breeds throughout the year (Clarke, 1981). Species, weight, age and reproductive condition of mussels may influence their bioaccumulation of organic contaminants. The importance of biological variability in the interpretation of station-to-station distribution patterns of these contaminants in native mussel populations will be discussed in a later section.

Distribution of Organochlorine Pesticides

Most of the mussels (97%) contained detectable levels of p,p'-DDE. It was the dominant DDT derivative and the dominant organochlorine pesticide in mussels, accounting for over 50% of total organochlorines in specimens from the St. Lawrence River and over 80% in specimens from the Ottawa River. Tissue concentrations (Fig. 2) were highest at the inlet from Lake Ontario, at the mouth of the Grass River, and at one site in Lac Saint-Francois. Concentrations dropped off to consistently low levels in Lac Saint-Louis and Lac Saint-Pierre and were lower again in the Ottawa River. p,p'-DDE is also the major DDT derivative present in sediment. For example, Wilkins (1988) did not detect p,p'-DDD or p,p'-DDT in sediment from any of 75 sites sampled in the Maitland area (near station #29) in 1984; however, p,p'-DDE was present at 1-17 ng/g at 34 sites. In the international section of the river between Kingston and Cornwall, concentrations of p,p'-DDE were found to be highest at the mouth of the Grass River (90 ng/g) in 1975 (Kuntz, 1988) and in Kingston basin (45 ng/g) in 1981 (Merriman, 1987). Kauss et al. (1988) surveyed the Cornwall/Massena area in 1979 and observed that concentrations were generally highest along the American shore, with a maximum of 52 ng/g occurring in the deposition zone between the Grass and Raquette Rivers. Concentrations of total DDT averaging 1.5 but as high as 16.7 ng/g have been reported for Lac Saint-Francois (Sloterdijk, 1985). According to Suns et al. (1985), young-of-the-year spottail shiners (Notropis hudsonius) collected from the upper river in 1983 contained higher residues (16-25 ng/g wet wgt.) than those taken from the mouth of the Grass River (6 ng/g). In 1987, however, concentrations in the Cornwall/Massena area were higher in the Grass River (15 ng/g) than along the north shore (5-12 ng/g), and were highest (24-26 ng/g) along the south shore of Cornwall Island across from several American industries. These data suggest that Lake Ontario and the Grass River both contribute p,p'-DDE to the St. Lawrence River. The distribution of p,p'-DDD in mussels

(Table 3) was nearly identical to that of p,p'-DDE, but concentrations were much lower. Residues of both DDT derivatives in mussels dropped off quickly with distance from the sources.

DDT was banned from major use in Ontario and the United States in the early 1970s (Kauss et al., 1988); therefore, its degradation products p,p'-DDE and p,p'-DDD would be expected to occur more frequently and in higher concentrations than the parent compound in recent environmental samples. Interestingly, p,p'-DDT was detected in 5 of the 7 mussels collected from Lac Saint-Louis and in 2 specimens from Lac Saint-Pierre (Table 3). When present, p,p'-DDT accounted for a substantial proportion (20-40%) of the total DDT residue, suggesting possible recent use or improper disposal of DDT in these areas.

Ninety percent of mussels collected from St. Lawrence River stations contained Mirex. The distribution pattern implicated Lake Ontario as the source, as concentrations decreased gradually in a downstream direction and Mirex was never detected in mussels from the Ottawa River (Fig.3). This pesticide, which is highly lipid soluble and cannot be metabolized by aquatic organisms (Metcalf et al., 1973), appeared to be more persistent than p,p'-DDE as concentrations of Mirex declined much more slowly with distance from the source.

The only source of Mirex to the St. Lawrence River is Lake Ontario (Scrudato and DelPrete, 1982). Although the United States banned the manufacture and use of Mirex in the late 1970s (Newell et al., 1987), Mirex will continue to enter the St. Lawrence River for at least the next 100 years due to the remobilization of contaminated sediment from Lake Ontario (Lum et al., 1987). Despite this continuous input Mirex has never been detected in bottom sediments from the international section of the river (Merriman, 1987; Kauss et al., 1988; Kuntz, 1988; Wilkins, 1988), although concentrations ranging from <1.0-3.3 ng/g have been reported for Lac Saint-Francois (Sloterdijk, 1985). Merriman (1987) and Lum et al., 1987) observed concentrations of Mirex ranging from 2-6 ng/g in suspended sediment samples from the upper river. Particle-bound Mirex may, therefore, be the primary source of this contaminant to the food web. Relatively uniform concentrations of Mirex (6-8 ng/g wet wgt.) have been found in spottail shiners throughout the international section of the river (Suns et al., 1985), and this is evidence of continuous exposure.

G-chlordane was detected in 60% of the mussels. Its distribution pattern differed from that of Mirex and DDT derivatives in that concentrations were low in mussels from the upper river and the Ottawa River, and were not detected at all in the Cornwall/Massena area (Fig. 4); rather, they were highest in mussels from the three riverine lakes. G-chlordane has been detected at trace levels only (1-5 ng/g) in bottom sediment between Kingston and Cornwall, and has never been

detected in the Grass River (Merriman, 1987; Kauss et al., 1988; Kuntz, 1988). Chlordane is a lipid-soluble and highly persistent pesticide used for controlling termites and soil insect pests in Ontario (Kauss et al., 1988). Its use in the United States has been recently restricted (Newell et al., 1987). Although the environmental occurrence of G-chlordane in the lower St. Lawrence River is not known, mussel data suggest local sources or deposition in the three lakes.

Lindane was detected in only 2 mussels - one E. complanata from station #BP and one from station #29 - at trace concentrations. A-BHC, however, was measurable in 79% of the mussels from the St. Lawrence River at nearly uniform concentrations averaging 0.13 ng/g and in 50% of the mussels from the Ottawa River at slightly lower concentrations (Table 3). Neither hexachlorocyclohexane isomer has been detected, except rarely at trace levels, in sediment or suspended sediment from the international section of the river (Merriman, 1987; Kuntz, 1988; Wilkins, 1988). In the Cornwall area, Kauss et al. (1988) found A-BHC to be almost entirely absent from bottom sediment taken from both sides of the river, while Lindane was elevated along the south shore only with a maximum concentration of 62 ng/g observed at the mouth of the Grass River. Suns et al. (1985) did not observe detectable concentrations of total BHC in spottail shiners collected from sites in the area in 1983. Yamato et al. (1983) reported that uptake of HCH isomers from water by the marine short-necked clam (Venerupis japonica) was higher for A-BHC than for Lindane, and that Lindane was eliminated more rapidly. This may explain the more frequent occurrence of A-BHC in St. Lawrence River mussels.

Distribution of Chlorinated Aromatics

Mussels were analyzed for 11 chlorobenzenes (CBs), but only 4 were detected. 1,2,4-TCB was found in only one E. complanata from station #55 and QCB in only 4 specimens from Lac Saint-Louis and one from Lac Saint-Pierre, all at very low concentrations. Hexachlorobenzene (HCB), a fungicide, was present in 65% of mussels and concentrations did not vary greatly from station to station (Fig.5). Unfortunately, analytical reproducibility for this compound was poor.

Chlorobenzenes are not major contaminants of the St. Lawrence system. Although they have been reported in industrial and municipal effluents at Maitland and Cornwall, concentrations of HCB in sediment at Maitland are low (1-7 ng/g; Wilkins, 1988) and concentrations in the Cornwall/Massena area (Kauss et al, 1988) range from 2-28 ng/g along the north shore and are detectable along the south shore only in

the deposition zone downstream of the Grass River (3-15 ng/g). Merriman (1987) found that di-, tri- and tetrachlorobenzenes accounted for 71-100% of the total CBs in bottom and suspended sediment from the upper river. In contrast, most mussels contained primarily HCB and spottail shiners collected in 1983 from locations near stations 28, 29, 30 and 37 contained traces of HCB only (Suns et al, 1985). Oliver and Niimi (1988) reported a similar partitioning pattern for CBs in the Lake Ontario ecosystem, with HCB accounting for 100% of the CB residue in fish, 47-72% in organisms from lower trophic levels, and only 40% in bottom and suspended sediment.

1,2,3-TCB occurred in 30% of the mussels and, when present, accounted for a higher proportion of the total CBs than HCB (Table 3). There was no apparent pattern to the spatial distribution of mussels with detectable concentrations of this TCB, but all eleven individuals were of the species E. complanata. At the five stations where both species were collected, 5/10 E. complanata accumulated 1,2,3-TCB, while 7/7 L. r. radiata did not, indicating possible interspecific differences in the accumulation of CBs.

Octachlorostyrene (OCS) was present at low concentrations in 11 mussels from the St. Lawrence River only. OCS is associated with the production and improper disposal of wastes from the electrolytic production of chlorine in the Great Lakes region up to the early 1970s (Kaminsky and Hites, 1984). The major sources to Lake Ontario are the Niagara and Oswego Rivers, and OCS is transported to the St. Lawrence River via contaminated sediment from Lake Ontario. However, OCS is not a major contaminant of concern in the St. Lawrence River. Kaminsky and Hites (1984) observed lower concentrations of OCS in sediment at the inlet to the St. Lawrence River (<15 ng/g) than in the eastern basin of Lake Ontario (15-50 ng/g) in 1981-82. Also, while residues as high as 560 ng/g wet wgt. have been observed in spottail shiners collected near industrial point sources in the St. Clair River, concentrations in spottails from Maitland, Cornwall and the Grass River were much lower at 1-2 ng/g (Suns et al, 1985).

Distribution of PCBs

PCBs were detected in all 38 mussels collected. The highest concentration (492 ng/g) was observed in a mussel from the mouth of the Grass River; this concentration was at least 10X higher than in mussels from the three riverine lakes and about 100X higher than in the upper river or Ottawa River (Fig.6). PCBs were obviously major contaminants of mussels living downstream of Cornwall, as they accounted for 95% and 98% of the total organic contaminant residues in mussels from stations 30 and 37, respectively, and an average of 90% (72-100%) in mussels from the lakes. In contrast, only 50% (17-72%)

of the total residue in mussels from the upper river and 83% (69-91%) in the Ottawa River could be attributed to PCBs.

PCBs are highly stable synthetic organic compounds which have been extensively used as plasticizers, lubricants, flame retardants and heat transfer, dialectric and water proofing agents. They have been banned from manufacture and use in the United States for 10 years (Newell et al, 1987), but are still used in closed system electrical and heat transfer operations in Ontario (Kauss et al, 1988). It has been well-documented that American industries situated along the Grass River and the south shore of the St. Lawrence near Massena, New York, are the primary sources of PCBs to the system. Four sediment surveys conducted between 1975 and 1985 identified this area as by far the most contaminated with PCBs (Table 4). Concentrations of PCBs in sediment from Lac Saint- Francois in 1979-81 were highest (max. 1900 ng/g) along the south shore (Sloterdijk, 1985), implicating the American industries as the source of transboundary pollution. In 1983 (Suns et al, 1985) concentrations of PCBs in spottail shiners were 5 - 10 X higher at the mouth of the Grass River (950 ng/g wet wgt.) and in the deposition zone between the Grass and Raquette Rivers (1262 ng/g) than in the upper river or north shore Cornwall area. The pattern of tissue PCB concentrations in native mussels clearly reflects the known distribution of these contaminants throughout the St. Lawrence system.

In the only other data available on the occurrence of PCBs in mussels from the St. Lawrence River, Kauss et al (1988) reported a concentration of 68 ng/g wet wgt. for E. complanata collected near an industrial outfall known to contain PCBs. Concentrations in sediment were approximately 30 ng/g. Using a wet-dry wgt. conversion factor of 10 for mussels (J.L. Metcalfe, unpublished data), a bioconcentration factor (BCF) of 20X for mussels vs. sediment may be calculated. Elder and Mattraw (1984) also observed a BCF of 20X for PCBs in the Asiatic clam, Corbicula manilensis exposed to 1 ng/g PCBs in sediment of the Apalachicola River in Florida. In the present study, a BCF of 3X was determined for the mussel from station #30 by comparing its tissue concentration on a dry weight basis (4920 ng/g) with the value for sediment (1650 ng/g) collected concurrently from the same location (R.A.P., 1988). This decrease in BCF with increasing exposure concentration is a common phenomenon which indicates the compound is approaching saturation levels in the organism.

Oliver and Niimi (1988) found that concentrations of PCBs in various components of the Lake Ontario ecosystem, as well as the proportion of the total PCB residue made up of the more highly chlorinated isomers, increased with increasing trophic level and lipid content of the organism. Mussels occupy a low trophic level and have a lipid content of approximately 0.5% (J.L. Metcalfe, unpublished data) which is similar to that reported by Oliver and Niimi (1988) for macrozooplankton. However, mussels from the upper St. Lawrence River

(stations 28, BP and 29), which should reflect Lake Ontario exposure conditions, were closest in terms of their isomeric composition to slimy sculpins (Cottus cognatus) - a bottom-feeding fish with a lipid content of 8%. This implies that the extent of contact with contaminated sediment may be as important as, or more important than, lipid content or trophic level in determining the relative bioaccumulation of various PCB homologues. In support of this, Paarivirta et al (1985) found no evidence of food chain biomagnification of chlorophenols in a contaminated lake chain in Finland. However, sediment-associated organisms tended to accumulate the less soluble, material-bound compounds while filter-feeders accumulated the more water-soluble isomers. Mussels are difficult to categorize. Because of their burrowing habits, they are in direct contact with the sediment. Many species, including those in the present study are also deposit feeders; i.e., they feed on resuspended bottom sediment. However, mussels also pass large quantities of water through their gills during respiration and their siphons during feeding.

The 63 PCB congeners for which mussels were analyzed belong to 10 homologues, increasing in degree of chlorination from the mono- to the decachlorobiphenyls (Table 1). The isomeric composition of residues in mussels from various locations in the St. Lawrence system differed considerably (Fig. 7). In the upper river, an average of 63% of the total PCB residue was due to penta- and hexa- isomers and only 8% to tetra- isomers. No di- or trichlorobiphenyls were present. Mussels from the Ottawa River had a similar profile, with 70% penta- and hexa- isomers and only 12% di-, tri- and tetra- isomers. At stations 30 and 37 (latter not shown) in the American industrial core, however, 52% and 58% (respectively) of total PCBs were di-, tri- and tetra- isomers while only 35% and 34% were penta- and hexachlorobiphenyls. In Lac Saint-Francois, the profile began to shift back to the right as di- , tri- and tetra- isomers were reduced to 22% of the total and penta- and hexa- isomers increased to 51%. This was due to the disappearance of all but one of the 7 dichlorobiphenyls as well as one tri- and one tetra- congener which were introduced at Massena. The isomeric composition of residues in mussels from Lac Saint- Louis and Lac Saint-Pierre did not differ significantly from that of Lac Saint-Francois specimens. Averages of 27% and 30% di-, tri- and tetra- isomers and 52% and 54% penta- and hexa- isomers, respectively, were measured.

These findings suggest that, with the exception of some of the lower chlorinated congeners, the majority of the PCB compounds entering the river at Massena persist in resident mussels for several hundred kilometers downstream of the point source in a characteristic accumulation pattern which differs from that occurring in locations such as the upper river and the Ottawa River which are remote from point sources. Table 5 shows that only 25-27 individual PCB congeners were identified in mussels from the upper river. At the mouth of the

Grass River, this number doubled to 59. At station #37, 15 km downstream of the Grass River, 56 congeners were still present and in Lac Saint- Francois there were approximately 50. There was no decrease in this number in Lac Saint-Louis, but there was a slight reduction to an average of 43 (33-48) in Lac Saint-Pierre. (Station #95, which was in the mouth of the Yamaska River and therefore not representative of Lac Saint-Pierre, was excluded from this average). Only 13-20 congeners were detected in the Ottawa River. Concentrations of total PCBs were also lower here than in the upper river, which suggests that Lake Ontario is a significant, if secondary, source of PCBs to the St. Lawrence River.

Influence of Biological Variability on Spatial Distribution Patterns for Contaminants in Mussels

Specimens of both species were obtained from only six stations and replicates of one or the other species from only eight stations, thus, it was not possible to adequately assess the relative contribution of biological factors to variability in contaminant residue data. Furthermore, the effects of age and reproductive status could not be separated from species because E. complanata were older than L. r. r.radiata and the latter species was more frequently gravid. A careful examination of the data, however, revealed apparent interspecific differences in the accumulation of several contaminants. Concentrations of Mirex (Fig.3) were generally higher in L. radiata, while HCB residues (Fig.5) were frequently higher in E. complanata. As noted earlier, 1,2,3-TCB occurred in 42% of E. complanata but never in L. r. radiata. There was no evidence of differences in total PCB concentrations between species (Fig.6); however, E. complanata usually accumulated a larger number of congeners. For example, at station #28, twenty- four congeners were identified in this species vs. only 18 in L. r. radiata, at station #166 the ratio was 23 to 13 and at station #65 it was 25 to 22, although numbers were similar (50 vs. 51) at station #55. Comparisons were not possible for stations #BP and #59 because E. complanata from these locations were not analyzed for individual PCB congeners.

Differences in the pollution status of various zones in the St. Lawrence system were clearly evident despite considerable variation among individual mussels from two different families in different stages of the reproductive cycle, with wet weights 2.2-26.2g and ages 3-16 years. Leard et al (1980) were also able to discriminate among five river basins in Mississippi in terms of relative contamination with DDT derivatives, Chlordane, Endrin and Toxaphene using pesticide residues in seven difference species of freshwater mussels. With the exception of the imported Asiatic clam, Corbicula manilensis, interspecific differences in the bioaccumulation of pesticides among native unionid species were minor. Similarly, Bedford et al (1968)

found no significant differences in DDT residues among seven species of unionids from a relatively clean location in the Red Cedar River, Michigan. After exposing Anodonta grandis and L. r. siliquoidea to contaminated downstream sites, they reported small differences in uptake rates and equilibrium concentrations between the species which did not affect site-to-site comparisons of relative contamination.

Relative Contamination of Mussels from Various Locations in the Great Lakes System

Organic contaminant concentrations in mussels have been determined for caged E. complanata exposed in the Niagara River in 1980 (Kauss et al 1981) and in the St. Clair and Detroit Rivers in 1982-83 (Kauss and Hamdy 1985) and for native L. r. siliquoidea from the St. Clair River, Lake St. Clair and Detroit River in 1983 (Pugsley et al 1985). The data are summarized and compared with the results of the present study in Table 6. The values of Pugsley et al (1985) were recalculated on a wet weight basis, using their own conversion factor. Maximum concentrations of DDT derivatives were highest in mussels from the St. Lawrence River. Maximum Chlordane residues were highest in the Niagara and Detroit Rivers, while A-BHC concentrations were highest in the Niagara River and lowest in the St. Lawrence River. HCB residues were also lowest in mussels from the St. Lawrence River, but were highest - along with OCS concentrations - in caged or native mussels from the St. Clair River. Maximum concentrations of these chlorinated aromatics were found in mussels from the Sarnia waterfront, in an area downstream of a petrochemical complex (Kauss and Hamdy 1985). Maximum PCB concentrations were reported in mussels living or exposed near known point sources of these contaminants, including the mouth of the Grass River in the St. Lawrence system and along the heavily industrialized Michigan shoreline of the Detroit River. These residues (492 ng/g wet wgt. in the Grass River and 543 ng/g in the Detroit River) were similar to the 560 ng/g concentration reported for the marine green-lipped mussel, Perna viridis, from a bay in Hong Kong known to be heavily polluted by industrial sources of PCBs (Kannan et al 1988). The United States Environmental Protection Agency recommends 500 ng/g as the maximum allowable tissue concentration for the protection of aquatic biota (Elder and Mattraw 1984). New York State's Department of Environmental Conservation advises a limit of 110 ng/g PCBs in aquatic biota for the protection of wildlife which may consume aquatic organisms (Newell et al 1987). Concentrations of PCBs in mussels from several locations in the Great Lakes system appear to be approaching hazardous levels for both aspects of risk.

CONCLUSIONS

Native unionid mussels from the St. Lawrence River bioaccumulated the major organic industrial contaminants and pesticides of concern in the system. Spatial distribution patterns for contaminants in mussel tissues generally reflected the known distribution of these compounds in bottom sediment and clearly identified the major sources of several important contaminants, such as DDT derivatives, Mirex and PCBs.

E. complanata and L. r. radiata are particularly suitable biomonitors for the St. Lawrence River because they are ubiquitous throughout the system (Levasseur 1977; Langley et al 1980), occur on a wide variety of substrates including gravel, sand, mud and clay (Clarke 1981) and were found to occupy a wide range of depths (3.0-29.0 m) during the present study. As spatial biomonitors they are therefore superior to sportsfish, which are very mobile and in some cases migratory, and young-of-the-year forage fish, which are restricted to shallow near-shore zones. As a monitoring medium, they are also superior to sediment because they can be more easily standardized. While sources of biological variability in mussel samples can be controlled, factors such as particle size distribution and organic content, which profoundly affect the adsorption of contaminants to sediments, cannot.

Biomonitoring with native mussel populations has several potential applications. It may be used to complement or supplement traditional water and sediment ambient monitoring programs, primarily to provide information on bioavailability. Residues in mussels can also be used to define the impact zone of point source pollution and can serve as a feedback mechanism for determining the effectiveness of pollution control measures as they are implemented. Finally, models of the fate of organic contaminants in large rivers such as the St. Lawrence should include a mussel component. These organisms constitute the major portion of the benthic biomass in many areas and they significantly rework the sediment by means of their burrowing, respiratory and excretory activities (McCall et al 1979), thereby altering the profiles of contaminants in bottom sediment and contributing to their transformation.

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FIGURE CAPTIONS

Figure 1. Locations of the twenty mussel sampling stations in the St. Lawrence and Ottawa Rivers

Figure 2. Concentrations of p,p'-DDE (ng/g wet wgt.) in individual mussels from the twenty sampling stations (● = <DL in *E. complanata*; O = <DL in *L. r. radiata*)

Figure 3. Concentrations of Mirex (ng/g wet wgt.) in individual mussels from the twenty sampling stations (Legend for Figures 3-6 as per Figure 2)

Figure 4. Concentrations of G-chlordane (ng/g wet wgt.) in individual mussels from the twenty sampling stations

Figure 5. Concentrations of HCB (ng/g wet wgt.) in individual mussels from the twenty sampling stations

Figure 6. Concentrations of total PCBs (ng/g wet wgt.) in individual mussels from the twenty sampling stations

Figure 7. Isomeric composition of PCB residues, as percent total PCBs represented by each homologue, in mussels from three locations in the St. Lawrence River

Table 1. Organic contaminants investigated

Organochlorine pesticides:
- p,p'-DDT
- p,p'-DDE
- p,p'-DDD
- Mirex
- G-chlordane
- Lindane
- A-BHC

Chlorobenzenes:
- 1,3-DCB (dichlorobenzene)
- 1,4-DCB
- 1,2-DCB
- 1,3,5-TCB (trichlorobenzene)
- 1,2,4-TCB
- 1,2,3-TCB
- 1,2,3,5-TECB (tetrachlorobenzene)
- 1,2,4,5-TECB
- 1,2,3,4-TECB
- QCB (pentachlorobenzene)
- HCB (hexachlorobenzene)

Octachlorostyrene (OCS)

PCB (polychlorinated biphenyl) congeners*:

Mono-	1, 3.
Di-	4, 10, 7, 6, 5, 8, 12/13.
Tri-	19, 17/18, 24/27, 16, 32/26, 25, 31/28, 33, 22.
Tetra-	45, 53, 46, 52, 47/48, 44/42, 41/71, 64, 74, 70/76, 40, 66, 56/60/81.
Penta-	95, 92, 101, 84, 99, 87/97, 85, 110/151, 82, 118, 105.
Hexa-	136, 149, 132/146, 153, 141, 138, 129, 128.
Hepta-	178, 183, 174, 171, 180, 173, 170/190.
Octa-	203/196, 201, 195, 194/205.
Nona-	206.
Deca-	209.

*PCBs are listed as International Union of Pure and Applied Chemistry (IUPAC) numbers (Ballschmiter and Zell 1980).

Table 2. Detailed descriptions of mussels collected.

Station	Sampling depth (m)	Species	Reproductive condition	Wet wgt.(g)	Age (yrs.)
28	29.0	E		10.3	8
		E		11.0	6
		L		6.6	10
BP	14.0	E		21.7	8
		L		26.2	8
29	21.0	E		21.4	14+
		E		22.5	12
30	10.5	L	Gravid	13.4	9
37	7.0	L		8.7	4
42	3.3	E	Gravid	23.5	9
44	20.5	E		18.8	11
47	4.0	E		14.3	8
55	13.5	E		11.4	7
		E		14.9	9
		E		15.5	-
		L	Gravid	19.1	5
		L	Gravid	22.3	9
59	7.0	E		12.8	11
		L	Gravid	17.4	8
95	8.5	L		2.2	3
103	3.0	E		13.1	11
		E		19.0	6
		E		24.3	16
107	2.5	E		10.7	8
112	8.0	L	Gravid	17.9	8
113	3.0	E		8.6	7
114	4.0	E		12.5	8
115	8.0	E		16.2	6
166	5.0	E		17.2	16+
		E		21.0	12+
		L		13.1	6
165	10.0	E		15.0	9
		E		16.8	8
65	4.0	E		10.0	8
		E		13.7	8
		E		17.3	13+
		L		8.6	5
		L	Gravid	11.0	6

Table 3. Concentrations of minor organic contaminants in mussels (ng/g wet wgt.)

Station	Species	p,p'-DDD	p,p'-DDT	A-BHC	1,2,3-TCB	QCB	QCS
28	E	6.80	-	-	-	-	-
	E	2.12	-	0.20	-	-	-
	L	-	-	-	-	-	-
BP	E	0.34	-	0.13	0.30	-	-
	L	-	-	0.19	-	-	0.06
29	E	2.71	-	0.08	0.32	-	-
	E	-	-	0.12	-	-	0.05
30	L	0.58	-	-	-	-	-
37	L	-	-	-	-	-	-
42	E	1.00	-	0.12	-	-	0.07
44	E	0.17	-	0.21	0.37	-	-
47	E	-	-	-	0.67	-	-
55	E	-	0.54	0.12	-	0.05	0.04
	E	0.21	0.26	0.10	-	0.14	0.04
	E	-	-	0.14	0.49	-	-
	L	0.42	0.50	0.20	-	0.04	0.08
	L	0.20	0.21	0.10	-	-	0.03
59	E	-	-	0.11	0.64	-	-
	L*	0.39	0.80	0.20	-	0.06	0.21
	L*	0.36	0.75	0.12	-	-	0.17
95	L	-	-	-	-	-	-
103	E	-	-	0.07	-	-	-
	E	0.23	0.64	0.10	-	-	0.05
	E	0.29	-	0.15	0.28	-	-
107	E	-	-	0.18	0.80	-	0.05
112	L	0.39	-	0.11	-	-	0.16
113	E	0.51	-	0.13	-	-	-
114	E	0.34	0.54	0.11	-	-	-
115	E	0.22	-	0.10	-	-	-
166	E	-	-	0.05	-	0.03	-
	E	-	-	-	0.40	-	-
	L	-	-	-	-	-	-
165	E	-	-	0.04	-	-	-
	E	-	-	0.07	0.56	-	-
65	E	-	-	-	-	-	-
	E	-	-	0.06	-	-	-
	E	-	-	-	0.46	-	-
	L	-	-	-	-	-	-
	L	-	-	0.04	-	-	-

* Duplicate analysis.

Table 4. Summary of data on PCB contamination of sediments from the international section of the St. Lawrence River (ng/g dry wgt.)

	Reference and Year Sampled			
	1	2	3	4
Location	1975	1981	1979-82	1985
Kingston basin	200	310	-	-
Upper river	<50	80	<10	-
Cornwall, north shore	<50	-	ND-140	60
Grass River	1510	8740	1000	1650
Downstream Grass River	1590	-	1980	2300-2700
Raquette River	240	-	500	105- 155
St. Regis River	150	-	25	ND
Downstream St. Regis River	250	80	90	1105

(1) Kuntz 1988; (2) Merriman 1987; (3) Kauss et al. 1988; (4) R.A.P (1988).

Table 5. Numbers of PCB congeners identified in one or more mussels from each station*.

Station	#s Congeners in Mussels	Station	#s Congeners in Mussels
28	26	95	10
BP	25	103	46
29	27	112	46
30	59	113	33
37	56	114	43
42	50	115	48
55	52	166	22
59	50	165	13
		65	27

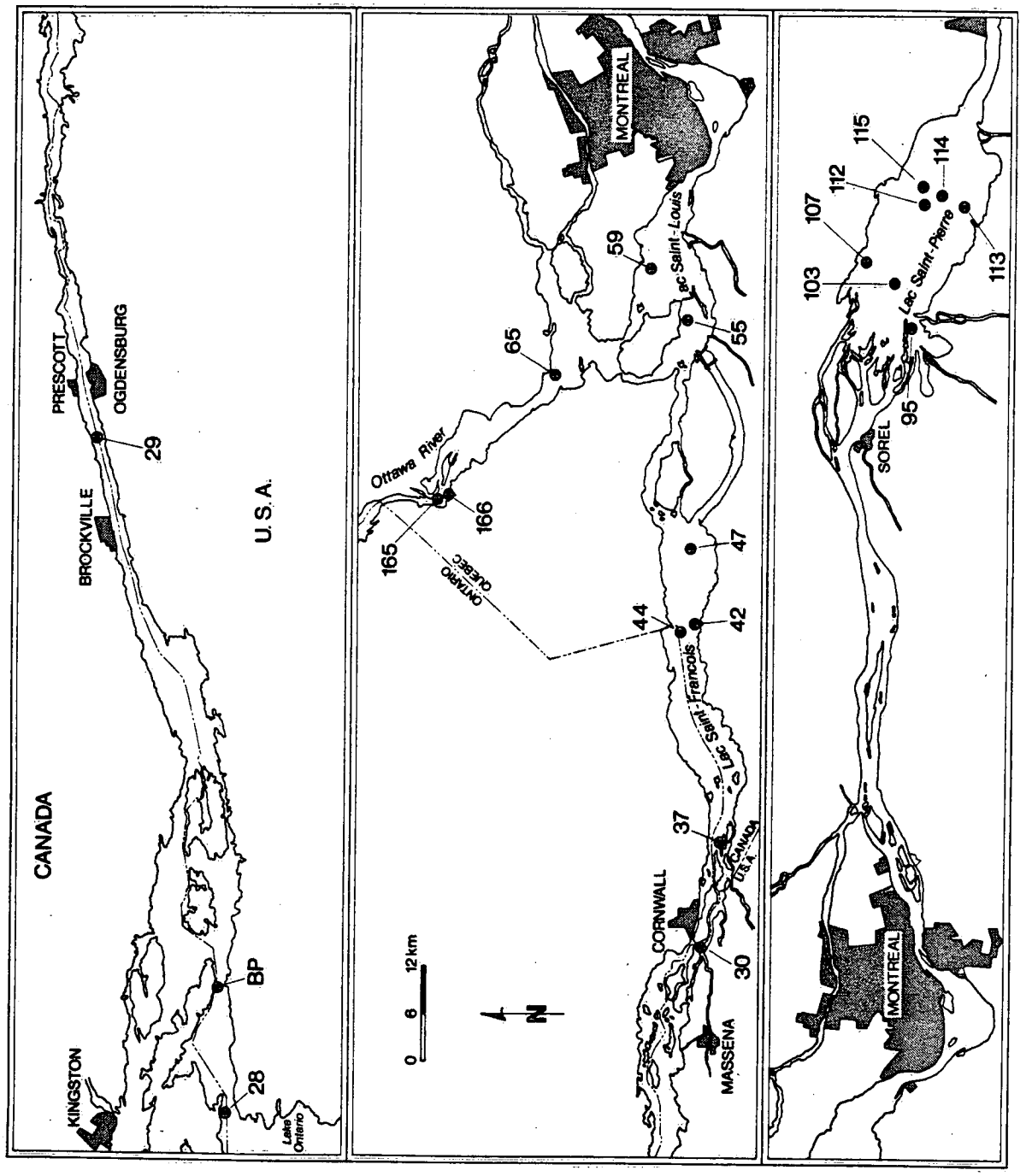
* Mussels from stations 44, 47 and 107 were analyzed only for total PCBs in each homologue, not for individual congeners.

Table 6. Maximum concentrations of organic contaminants in caged and native mussels (ng/g wet wgt.) from the Great Lakes system. (E.c. = Elliptio complanata; L.r.r. = Lampsilis radiata radiata; L.r.s. = Lampsilis radiata siliquoidea; C = caged; N = native).

	St. Lawrence River E.c.&L.r.r.(N) Ref 1**	Niagara River E.c. (C) Ref 2	St. Clair River E.c.(C) Ref 3	Lake St. Clair L.r.s.(N) Ref 4	Detroit River L.r.s.(N) Ref 4	E.c.(C) Ref 3	L.r.s.(N) Ref 4
Contaminant							
Total DDT	NA*	11	NA	NA	NA	NA	NA
p,p'-DDT	0.80	NA	<5	NA	NA	<5	NA
p,p'-DDD	7	NA	<5	NA	NA	5	NA
p,p'-DDE	24	NA	4	NA	NA	12	NA
Total Chlordane	NA	8	NA	NA	NA	NA	NA
G-Chlordane	0.30	NA	2	NA	NA	6	NA
A-BHC	0.20	12	4	NA	NA	<1	NA
HCB	0.50	4	24	NA	NA	3	NA
OCS	0.20	NA	55	29	23	4	9
Total PCBs	492	95	44	NA	NA	543	NA
Aroclor 1254	NA	NA	NA	16	105	NA	29

*NA = not analyzed.
**(1) This Study, (2) Kauss et al. 1981, (3) Kauss & Hamdy 1985 (4) Pugsley et al. 1985.

Figure 1


CANADA
U.S.A.
KINGSTON
BROCKVILLE
PRESCOTT
OGDENSBURG
BP
28
29
Lake Ontario
MONTREAL
Ottawa River
ONTARIO
QUEBEC
65
166
165
59
Lac Saint-Louis
55
47
44
42
Lac Saint-François
37
CORNWALL
MASSENA
30
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N
107
112
115
114
103
Lac Saint-Pierre
113
95
SOREL
MONTREAL

Figure 2

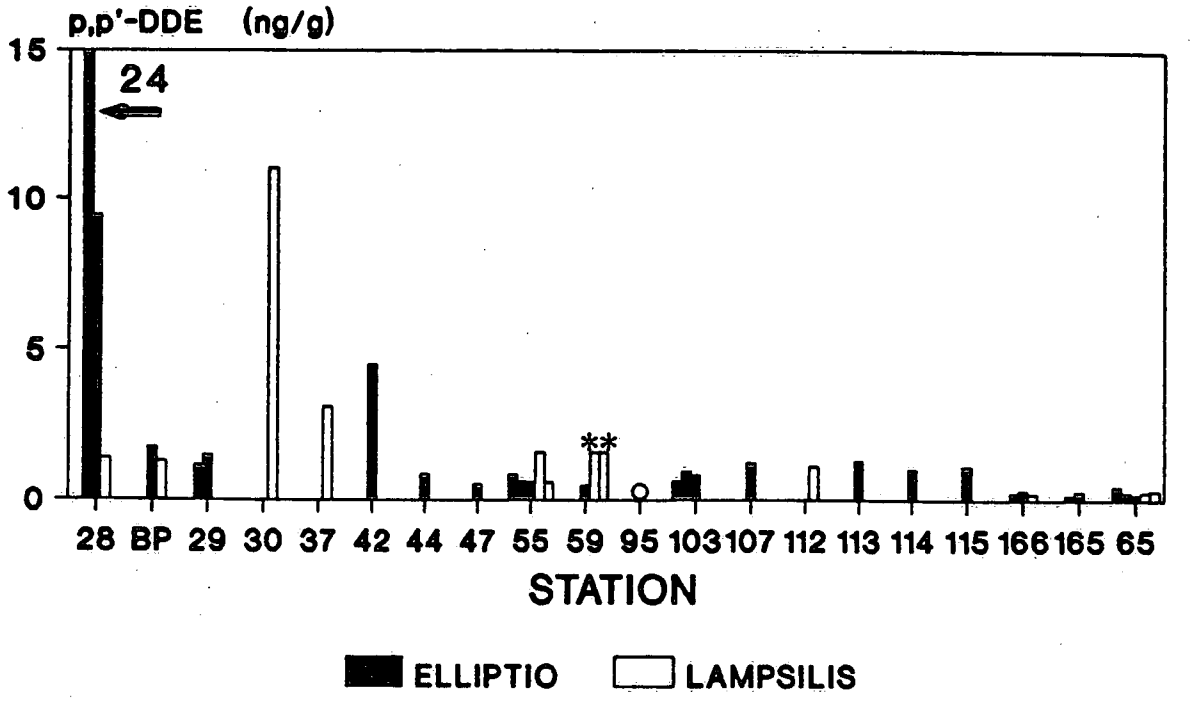
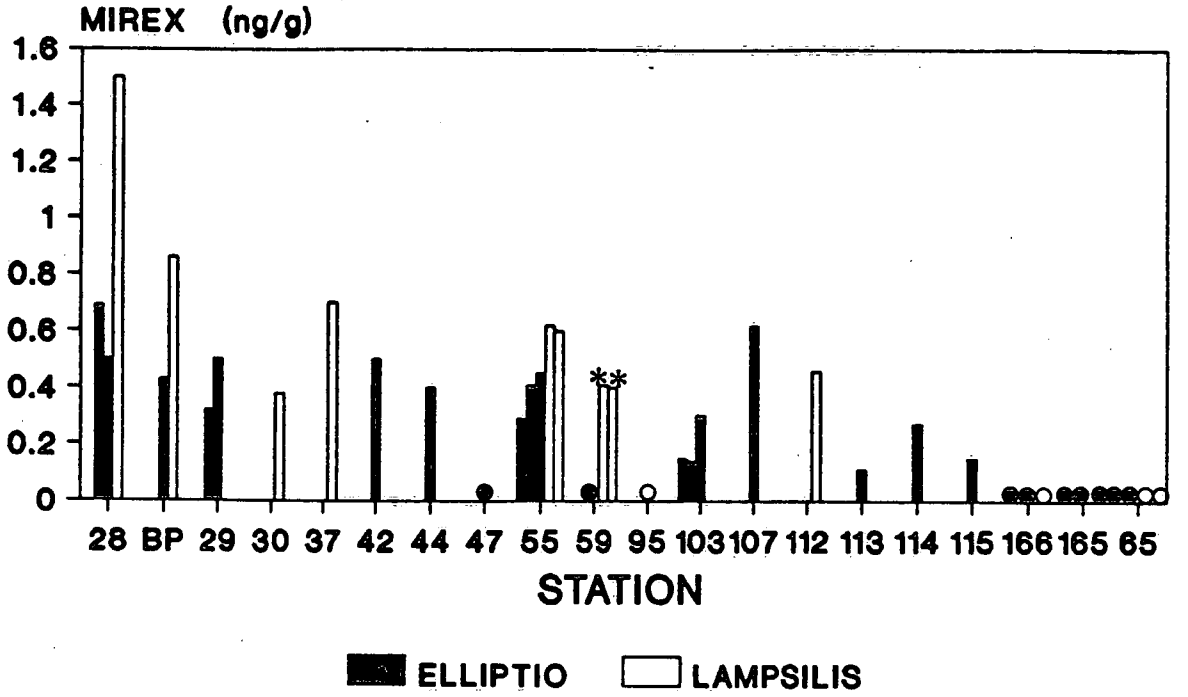

p,p'-DDE (ng/g)
15
10
5
0
24
**
28 BP 29 30 37 42 44 47 55 59 95 103 107 112 113 114 115 166 165 65
STATION
ELLIPTIO
LAMPSILIS

Figure 3


MIREX (ng/g)
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1.4
1.2
1
0.8
0.6
0.4
0.2
0
**
28 BP 29 30 37 42 44 47 55 59 95 103 107 112 113 114 115 166 165 65
STATION
ELLIPTIO
LAMPSILIS

Figure 4

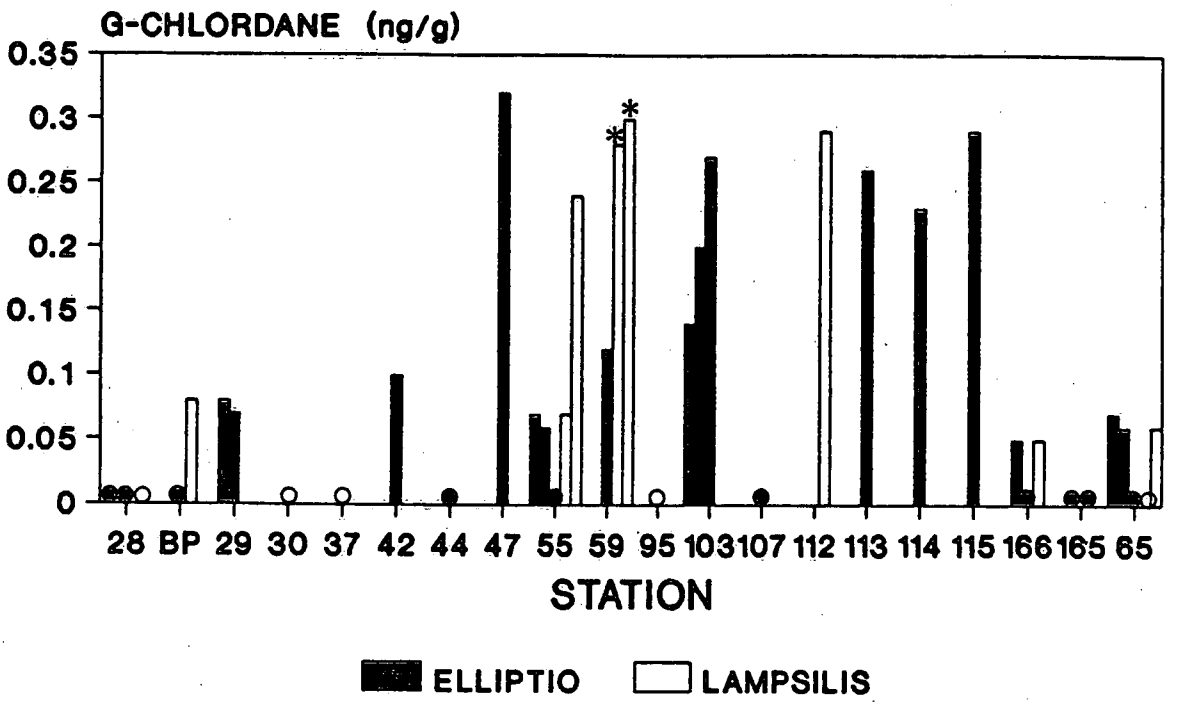
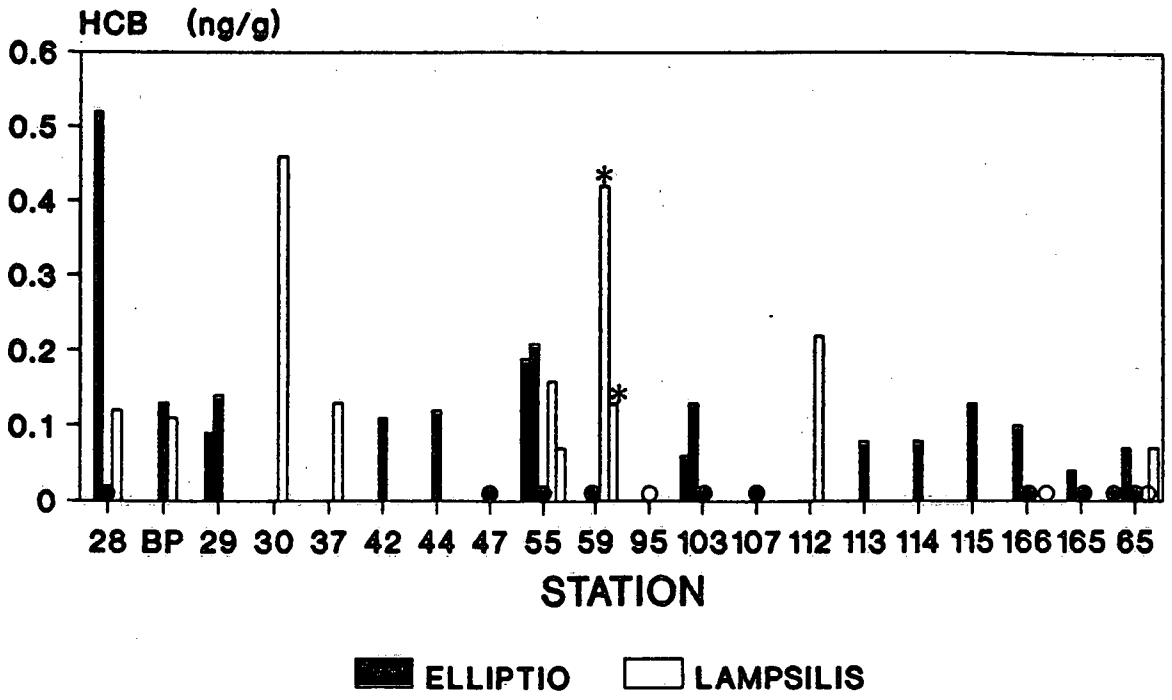

G-CHLORDANE (ng/g)
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
28 BP 29 30 37 42 44 47 55 59 95 103 107 112 113 114 115 166 165 65
STATION
ELLIPTIO
LAMPSILIS

Figure 5


HCB (ng/g)
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0.5
0.4
0.3
0.2
0.1
0
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STATION
ELLIPTIO
LAMPSILIS

Figure 6

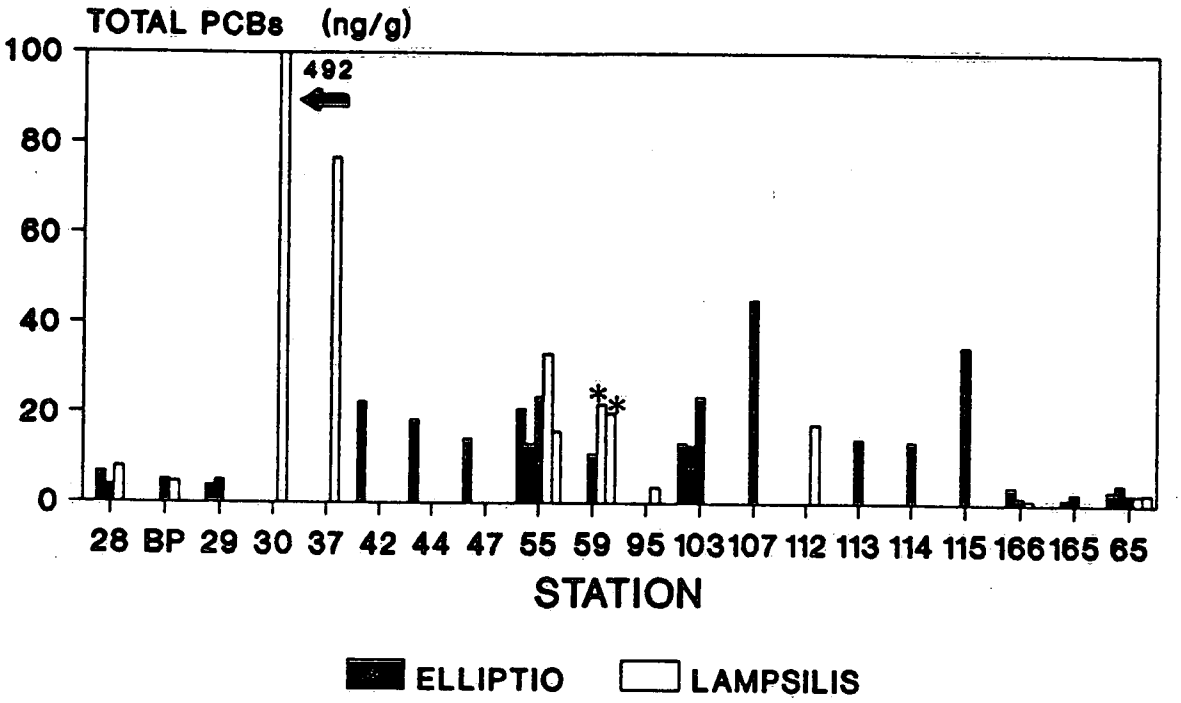
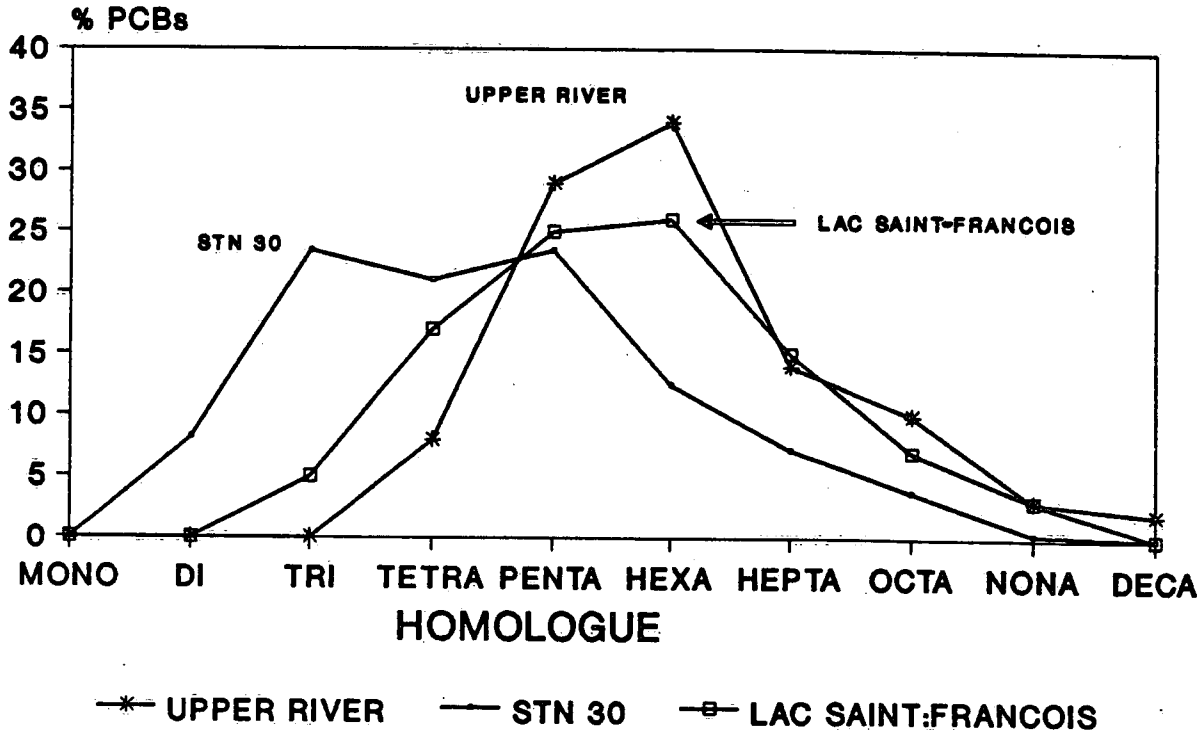

TOTAL PCBs (ng/g)
100
80
60
40
20
0
492
**
28 BP 29 30 37 42 44 47 55 59 95 103 107 112 113 114 115 166 165 65
STATION
ELLIPTIO
LAMPSILIS

Figure 7


% PCBs
40
35
30
25
20
15
10
5
0
UPPER RIVER
STN 30
LAC SAINT-FRANCOIS
MONO
DI
TRI
TETRA
PENTA
HEXA
HEPTA
OCTA
NONA
DECA
HOMOLOGUE
UPPER RIVER
STN 30
LAC SAINT:FRANCOIS