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Geographic distribution of selected contaminants in Great Blue Herons from the St. Lawrence River system, Quebec (1989–1994)

Jean-Luc DesGranges
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Abstract

From 1989 to 1994, we studied heavy metal and organochlorine levels in several tissues (eggs, liver, kidneys, blood, and feathers) of Great Blue Herons (*Ardea herodias*) from 31 heron colonies, 25 of them located in the St. Lawrence system (river, estuary, and gulf) and 6 located on inland lakes elsewhere in the province of Quebec (reference colonies). The fluvial corridor of the St. Lawrence (essentially the sections near Montréal) is considered a major source of contamination, and it accounted for the highest contaminant concentrations measured in the tissues (and body compartments) of heron nestlings. Contaminants in heron nestlings are clearly more representative of regional pollution than contaminants in eggs, where the levels probably reflect the situation on the migration and wintering grounds of the adult females. The ease with which contaminant levels can be measured in the feathers and blood of heron nestlings, combined with the close correlation that exists between these levels and those found in the liver, supports the routine use of non-lethal tissue sampling methods.

Key words: Great Blue Heron, *Ardea herodias*, bioaccumulation, Hg, OC-PCB, St. Lawrence River

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1. Introduction

In 1988, the Government of Canada and the Gouvernement du Québec agreed to harmonize their efforts to protect and clean up the St. Lawrence River, the most heavily polluted watershed in Quebec (St. Lawrence Centre 1996). From the inception of the St. Lawrence Plan, it was clear that a monitoring program would be required to determine the effects of environmental contamination on species living in the St. Lawrence system and to evaluate the effectiveness of the pollution clean-up measures. From the beginning, it was essential to provide researchers with spatiotemporal trends in contaminants, particularly the levels of heavy metals, polychlorinated biphenyls (PCBs), and organochlorine pesticides.

Because piscivorous birds are at the top of the food chain, assessing the contaminant levels in their tissues is a good way to monitor contaminants in the entire ecosystem (Diamond and Filion 1987; Furness and Greenwood 1993). The Great Blue Heron (*Ardea herodias*) was chosen as an indicator species for contaminant levels in the St. Lawrence and for associated toxicological effects (Champoux et al. 2006). This species was selected because colonies of this large bird (~2.5 kg), which has a long lifespan (~20 years), are found throughout the St. Lawrence River valley, from the river proper to the Gulf of St. Lawrence (DesGranges and Desrosiers 2006). In addition, adults stay close to their nesting colonies for nearly five months of the year while the young are growing, and the young, which are fed on fish caught locally, are believed to act as integrators of contamination at the regional scale (Benoit et al. 1993).

To assess the bioavailability of contaminants in the St. Lawrence system, we studied the concentrations of heavy metals and organochlorine compounds in the eggs, liver, kidneys, blood, and feathers of Great Blue Herons from 31 colonies from 1989 to 1994. Most of the colonies (25) were in the St. Lawrence system; a few colonies located on inland lakes elsewhere in Quebec (6) served as reference sites. As auxiliary research, we looked at the physiological effects of contaminants in Great Blue Herons (Champoux et al. 2000, 2002).

2. Study area and methods

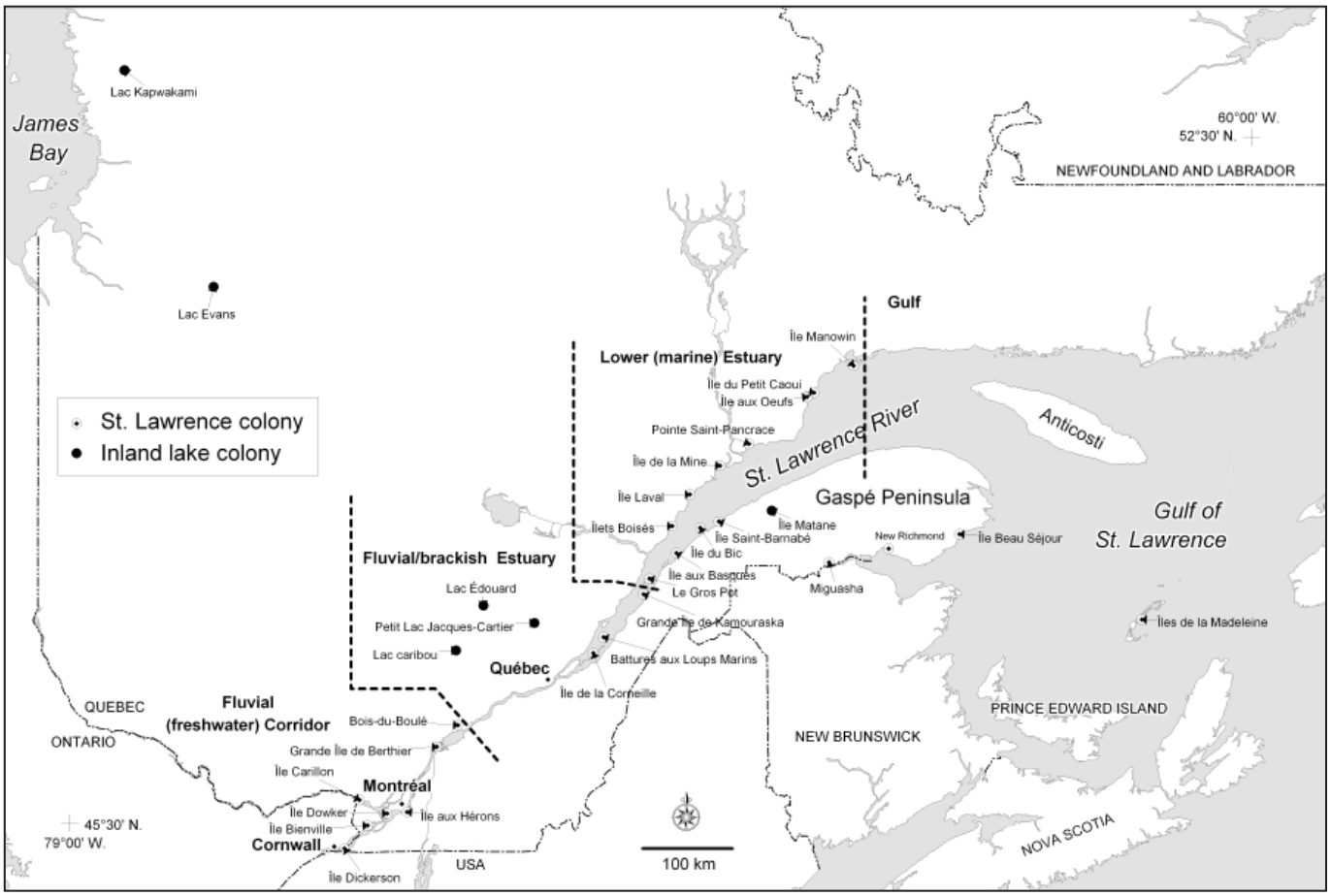
2.1 Samples and chemical analyses

Sampling was carried out at 25 colonies located along a 1350-km corridor of the St. Lawrence River and Gulf of St. Lawrence (from Cornwall, Ontario, to the Îles de la Madeleine) (Fig. 1). The fluvial portion of the river, which is freshwater (n = 7 colonies), and the fluvial/brackish estuary (n = 3) provide situations of high exposure to contaminants. The colonies of the lower estuary (n = 11) and the Gulf of St. Lawrence (n = 4) (both saltwater) are fairly distant from sources of industrial pollution. In addition, six reference colonies located on inland lakes distant from the St. Lawrence (as far as the James Bay region) were used to determine the baseline levels in freshwater situations less prone to contamination.

Biological tissues were sampled from a total of 217 nests between 1977 and 1994 (primarily from 1989 to 1994). At most colonies, we took one egg from each of five nests in the spring and, on a second visit, collected data from nestlings 35 to 45 days old from each of the same five nests ($\bar{x}$ = 36.6 days; SD = 8.4; n = 168 nestlings). A total of 201 eggs collected from 31 colonies was analyzed for metals and organochlorine compounds; some of the eggs were also used in a study of biomarkers (Boily et al. 1994; Champoux et al. 2000, 2002). A total of 168 nestlings from 21 colonies was sampled. Blood and feather samples were taken from 136 and 131 nestlings, respectively (some individuals were too young to allow a sufficient quantity of feathers or blood for the chemical analyses to be collected safely). Finally, a total of 127 heron nestlings from 31 colonies (over five years) was sacrificed while the remaining ones where banded and put back into their nest. Rodrigue et al. (2005) enumerate the sampling design (numbers of samples and pools per tissue, per age, per nest, and per colony), as well as the chemical analysis scheme (tissues analyzed for specific chemicals, with detection limits).

We based our assessment of nestling age on tarsus length (Quinney 1982). We plucked about 3 g of feathers from each nestling for mercury analysis. These feathers, most of which had reached three-quarters of their development, were collected symmetrically from different locations on the body so that the bird's ability to fly would not be impaired (fifth primary, fifth secondary, 2 coverts from each wing, 4 feathers from the lower and upper tail coverts, as well as a tail feather).

Figure 1
Locations of the 31 Quebec Great Blue Heron (*Ardea herodias*) colonies where eggs and adult and nestling tissues were collected between 1989 and 1994. Twenty-five colonies were located along the St. Lawrence River and 6 colonies were situated on inland lakes away from the St. Lawrence river system.



We collected 5 mL or less of blood from the brachial vein of one wing from each nestling, using a syringe containing ethylenediaminetetraacetic acid (EDTA). This is one-third of the volume of blood that can safely be taken without creating health risks for birds of this size (Campbell 1988). The blood sample was transferred to a Vacutainer tube containing EDTA, kept on ice, and frozen the same day.

Some nestlings were sacrificed by decapitation and the tissues were rapidly dissected in order to study the pathway followed by the contaminants in their internal organs (when sample sizes permitted).

Adult feathers (primarily flight feathers) were collected from nests or from the ground below nests during one of the visits. Adults killed accidentally at the nest in the process of developing a capture technique were also analyzed.

To get an estimate of the amount of mercury in a tissue, we multiplied its weight by the concentration of mercury found. The weight of liver and kidneys was measured, the weight of blood was estimated using the equation published in Campbell (1988), and the weight of feathers was obtained by plucking 5 nestlings with fully grown feathers.

Total mercury analysis was carried out using the method described in Adeloju and Mann (1987) (MET-CHEM-AA-03C). The samples (≈0.5 g) were digested in a mixture of nitric and sulphuric acids (ratio of 1 to 2) at 70°C. Potassium dichromate was added to the mixture to complete the oxidation of organic mercury compounds. Mercury concentrations were determined using a cold vapour technique (CVAAS) by an atomic absorption spectrometer, model 3030-AAS (Perkin-Elmer), equipped with a VGA-76 (Varian) hydride generator and a PSC-55 autosampler (Varian).

The chemical analysis used for organochlorine pesticides and polychlorinated biphenyls (PCBs) is described in Won et al. (2001). A total of 22 organochlorine compounds and 41 PCB congeners were sought. The PCB congener classification used here is that of the International Union of Pure and Applied Chemistry (IUPAC) (Ballschmiter and Zell 1980). The standard procedure used is described in the Laboratory Service Methods Manual as MET-CHEM-OC-04C. Briefly, the sample was extracted with a mixture of dichloromethane (DCM) and hexane (ratio of 1 to 1) and dehydrated with anhydrous sodium sulphate (Na_2SO_4). Lipids and biogenic materials were removed by gel permeation chromatography, and further cleanup was performed by Florisil column chromatography. Quantitative analyses of organochlorines and PCBs were performed by a capillary gas chromatograph coupled to a mass selective detector. Owing to the high cost of the organochlorine analyses, we pooled the samples for analysis. The concentration obtained from the analysis of a composite sample corresponds fairly closely to the mean of the individual concentrations (Turle and Collins 1992). Quality control of the chemical analyses was handled by the National Wildlife Research Centre (NWRC) at Environment Canada. Because the National Wildlife Research Centre performs analyses on the eggs of Herring Gulls (*Larus argentatus*) on

a regular basis, this material can be used as a reference for organochlorine analyses. As a rule, the National Wildlife Research Centre adds four samples with known values for mercury analyses. Two blanks are always used for each series of analyses. Analytical accuracy was tested by analyzing samples in duplicate and triplicate. The coefficient of variation for the analyses was less than 10% for total mercury.

Chemical concentrations in fresh tissues are expressed on a wet weight basis; concentrations in feathers are expressed on a dry weight basis.

2.2 Statistical analyses

Statistics on contaminant levels in biological tissues (including standard deviation (SD)) were calculated for eggs and the tissues of heron nestlings (liver, kidneys, blood, and feathers) from 1) individual broods, 2) the nests in each colony, and 3) the colonies in the different areas under study. Preliminary Shapiro-Wilks *W* tests (Zar 1984) showed that most of our contaminant concentrations were not normally distributed ($P > 0.05$). Furthermore, since sample sizes were small in several instances (colony/year/tissue/age), we choose not to log-transform the data and therefore used non-parametric analyses in the majority of cases. The raw data were processed directly, except for the proportions (p), which were transformed into arcsin $\sqrt{p}$, as recommended by Zar (1984). To compare two or more independent samples, we used two different tests, the Mann-Whitney *U* test and the Kruskal-Wallis *H* test. For comparisons of matched samples, the Mann-Whitney *U* test was replaced by the Wilcoxon signed-rank *Z* test and the Kruskal-Wallis *H* test was replaced by the Friedman test (χ^2 test) (Zar 1984).

In some cases, multiple comparison tests were performed after the Kruskal-Wallis tests had been done. The multiple comparison tests were performed using the *q* statistic, a non-parametric method analogous to Tukey's test (Zar 1984). The correlations between the variables were calculated using the Spearman rank correlation coefficient (r_s). For the regression analyses, linear models were used with the least squares method. The linearity of the relationships became evident when the distribution of the residuals was examined (Neter and Wasserman 1974).

Principal coordinate analyses (PCOORD) were used to position the colonies, tissues, and contaminants in ordination space so that associations among areas could be compared for adult and nestling tissues. Analyses were performed on the matrix of object similarities (calculated with Gower's general coefficient of similarity; Gower 1971) using method S15 in Legendre and Legendre (1998).

The results of the principal coordinate analyses are represented in two-dimensional ordination, accompanied by a graph illustrating the relative contribution of each variable to the dispersion of the objects. This contribution corresponds to the Pearson product moment correlation of the values and the positions of the objects on each of the two axes.

The descriptive statistics and the tests of normality were performed using JMP 2.0 software (SAS Institute 1991). All the other analyses were carried out using the R Package (Legendre and Vaudor 1991): SIMIL procedure for calculating similarity matrices and PCOORD for the principal coordinate analyses.

3. Results

3.1 Within- and between-colony variability

The coefficients of variation (all 31 colonies) for all contaminant levels in the eggs and in the tissues from each young heron from the same nest were low (generally less than 25%). The coefficients increased as the level of comparison moved from that of individual nests to that of colonies in different regions (see Rodrigue et al. [2005] for details).

For individual nests, the mean coefficient of variation for mercury levels was 13% for the feathers and 23% for the blood. The variability of concentrations of dichlorodiphenyldichloroethylene (*p,p′*-DDE = 1,1-bis-(4-chlorophenyl)-2,2-dichloroethene) in the tissues of young herons from the same nest was higher than the variability of concentrations of mercury and PCBs. For individual nests, the mean coefficients of variation for *p,p′*-DDE and PCBs were 25% and 24%, respectively, in the eggs; 14% and 17%, respectively, in the liver; and 46% and 26%, respectively, in the blood.

In general, the variability of contaminant concentrations for all the samples from a given colony was higher than that for each nest considered individually. The mean coefficient of variation for mercury in eggs was 64%; in nestling tissues, the mean coefficient of variation for mercury varied between 18% and 30%. Within a given colony, the mean coefficients of variation for *p,p′*-DDE and PCBs were higher: 129% and 102%, respectively, in the eggs; 93% and 80%, respectively, in the liver; and 91% and 108%, respectively, in the blood.

For all the colonies taken together, the mean coefficient of variation for mercury concentrations in the eggs was 24%; however, in nestling tissues it ranged from 34% to 69%. For all the colonies, the mean coefficients of variation for *p,p′*-DDE and PCBs were of the same order of magnitude as those for the colonies considered separately. They ranged from 61% to 91% for the eggs, from 64% to 92% for the liver, and 100% for the blood.

In general, lower coefficients of variation were obtained for internal organs than for the blood and eggs. By contrast, these coefficients were markedly higher (from 2 to 4 times; 50% to 100%) for organochlorines than for mercury, regardless of the tissue.

3.2 Mercury contamination

Analysis of mercury in adult tissues was limited to the liver and feathers. The mean mercury level in the liver tissues

of adults from the colonies along the St. Lawrence was 3.82 µg/g wet weight (maximum of 31 colonies = 5.13 µg/g wet weight, Île du Petit Caoui). The corresponding level in the feathers was 6.32 µg/g dry weight (maximum 14.45 µg/g dry weight, Île Laval in the lower estuary) (Table 1).

The concentration of mercury in adult feathers varied widely because the samples analyzed consisted of several types of feathers from different points in the annual lifecycle and because the feathers came from birds of different ages and genders (see Furness et al. 1986).

The mean mercury concentration in eggs from the St. Lawrence River was 0.26 µg/g wet weight (maximum 0.92 µg/g wet weight, Île de la Corneille in the fluvial/brackish estuary) (tables 1 and 2).

This concentration was much lower (<5%) than the concentration in the feathers of the adults from the same nests (Wilcoxon signed-rank test performed on the mean values per colony: $Z = -3.41$; $P = 0.007$; $n = 15$). However, the concentration of mercury in the feathers of adults was similar to (1.2 ×) the concentration of mercury in the feathers of their nestlings (Mann-Whitney $Z = -1.07$; $P = 0.3$; $n = 90$).

The mean mercury concentrations in the liver tissues of nestlings was 0.52 µg/g wet weight (maximum 1.27 µg/g wet weight, Île Dowker in the fluvial corridor). The mean mercury concentrations in the blood of nestlings was 0.48 µg/mL wet weight (maximum 1.76 µg/mL wet weight, Île aux Hérons, also located in the fluvial corridor) (tables 1 and 2).

Mercury levels in the blood and liver of nestlings were highly correlated (Fig. 2a).

When it was possible to do paired comparisons of nests and colonies, we found that the eggs contained 2.3 times less mercury on average than the blood from young herons (Wilcoxon signed-rank test $Z = -2.90$; $P = 0.004$; $n = 15$) and 21 times less mercury on average than the feathers from young herons (Wilcoxon signed-rank test $Z = -3.62$; $P = 0.0003$; $n = 17$; Fig. 3a).

The mean mercury concentrations in nestling feathers (5.75 µg/g dry weight; maximum 10.5 µg/g dry

Table 1
Mean total mercury concentrations (standard deviation, number of samples, and range of concentrations) in Great Blue Herons (*Ardea herodias*) from a total of 31 colonies in Quebec (25 in the St. Lawrence system and 6 reference sites on inland lakes) between 1989 and 1994

			Region									
		All colonies	St. Lawrence system				Inland lakes				Mann-Whitney	
Stage	Tissue	$\overline{X}$	$\overline{X}$	SD	*n* (pool)	Range	$\overline{X}$	SD	*n* (pool)	Range	*Z*	*P*[a]
Adult	Feathers (µg/g dry weight)	7.3	6.32	3.22	17	2.33–14.45	10.29	5.31	3	4.11–13.4	1.11	**0.01**
	Liver (µg/g wet weight)	6.9	3.82	1.39	5	1.59–5.13	24.75	-	1	-	-	-
Egg	Egg (µg/g wet weight)	0.27	0.26	0.15	44 (117)	0.33–0.92	0.31	0.08	12 (21)	0.15–0.43	2.23	**0.03**
Nestling	Blood (µg/mL wet weight)	0.45	0.48	0.34	38 (100)	0.11–1.76	0.32	0.23	10 (23)	0.14–0.84	1.93	**0.05**
	Feathers (µg/g dry weight)	5.68	5.75	2.08	54 (102)	1.1–10.5	5.44	1.67	16 (25)	3.7–8.8	0.69	0.49
	Liver (µg/g wet weight)	0.53	0.52	0.25	37 (69)	0.15–1.27	0.58	0.33	11 (15)	0.31–1.32	0.23	0.82
	Kidneys (µg/g wet weight)	0.44	0.42	0.16	36 (83)	0.11–0.75	0.48	0.09	11 (15)	0.35–0.63	1.44	0.15

[a] Numbers in bold are significant ($P \leq 0.05$)

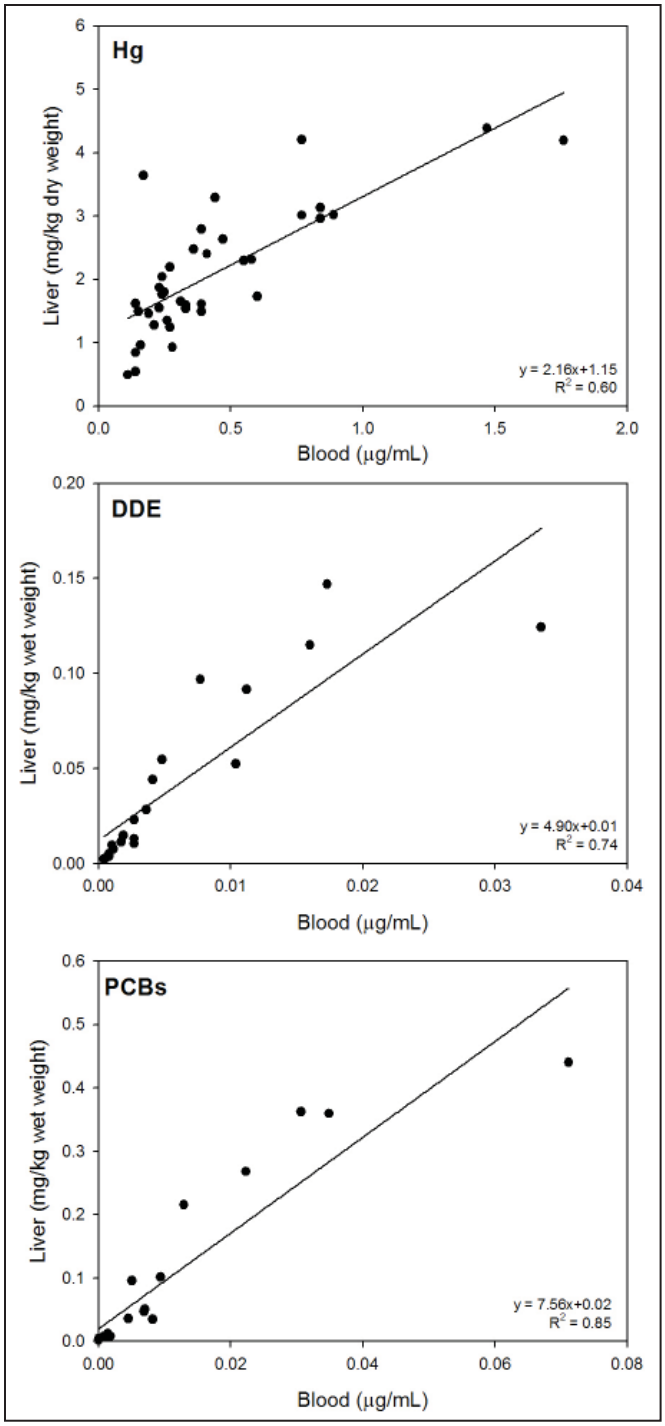
Table 2
Mean total mercury concentrations (standard deviation, number of samples, and range of concentrations) in Great Blue Heron (*Ardea herodias*) eggs and nestlings from a total of 31 colonies in Quebec (25 in the St. Lawrence system and 6 reference sites on inland lakes) between 1989 and 1994

	Egg (µg/g wet weight)				Feathers (µg/g dry weight)				Blood (µg/mL wet weight)				Liver (µg/g wet weight)				Kidneys (µg/g wet weight)			
Region	$\overline{X}$	SD	*n* (pool)	Range	$\overline{X}$	SD	*n* (pool)	Range	$\overline{X}$	SD	*n* (pool)	Range	$\overline{X}$	SD	*n* (pool)	Range	$\overline{X}$	SD	*n* (pool)	Range
Fluvial section (freshwater)	0.28	0.04	7 (31)	0.24–0.37	6.89	1.59	17 (46)	5.4–10.2	0.85[a]	0.56	7 (42)	0.30–1.76	0.81	0.36	6 (22)	0.40–1.27	0.48	0.15	25 (40)	0.20–0.75
Fluvial/brackish estuary	0.23	0.22	13 (21)	0.03–0.92	5.72	1.65	11 (15)	3.6–9.2	0.43	0.17	11 (15)	0.17–0.77	0.56	0.13	11 (12)	0.33–0.72	0.37	-	2 (11)	0.32–0.41
Lower estuary (saltwater)	0.28	0.13	21 (56)	0.08–0.55	5.50	1.88	23 (38)	2.6–10.5	0.42	0.21	17 (40)	0.16–0.96	0.45	0.17	17 (32)	0.24–0.82	0.31	0.10	8 (29)	0.15–0.42
Gulf of St. Laurence	0.24	0.06	3 (9)	0.18–0.28	1.27	0.27	3 (3)	1.1–1.6	0.13[b]	0.02	3 (3)	0.11–0.14	0.18	0.04	3 (3)	0.15–0.22	0.11	-	1 (3)	-
Total St. Lawrence system	0.26	0.15	44 (117)	0.03–0.92	5.75[a]	2.08	54 (102)	1.1–10.5	0.48[c]	0.34	38 (100)	0.11–1.76	0.52	0.25	37 (69)	0.15–1.27	0.42	0.16	36 (83)	0.11–0.75
Inland lakes	0.31	0.08	12 (21)	0.15–0.43	5.44[b]	1.67	16 (25)	3.7–8.8	0.32[d]	0.23	10 (23)	0.14–0.84	0.58	0.33	11 (15)	0.31–1.32	0.48	0.09	11 (15)	0.35–0.63
All colonies	0.27	0.14	56 (138)	0.03–0.92	5.68	1.98	70 (127)	1.1–10.5	0.45	0.32	48 (123)	0.11–1.76	0.53	0.27	48 (84)	0.15–1.32	0.44	0.15	47 (98)	0.11–0.75

[a] Values with the same letter or no letter are not significantly different ($P > 0.05$)

weight, Îles de la Mine in the lower estuary) were 17 times higher than those in the blood, 13 times higher than those in the kidneys, and 10 times higher than those in the liver. The feathers represent the main mercury reservoirs; while the young are growing, an estimated 90% of the mercury is stored in the feathers (see Rodrigue et al. [2005] for details).

Figure 2
Correlations in Great Blue Herons (*Ardea herodias*) between mercury in blood and mercury in the liver (A; 39 pools of 65 samples, same individuals for both); correlations between *p,p′*-DDE in blood and *p,p′*-DDE in the liver (B; 20 pools of 34 samples, same individuals for both); and correlations between PCBs in blood and PCBs in the liver (C; 20 pools of 34 samples, same individuals for both)

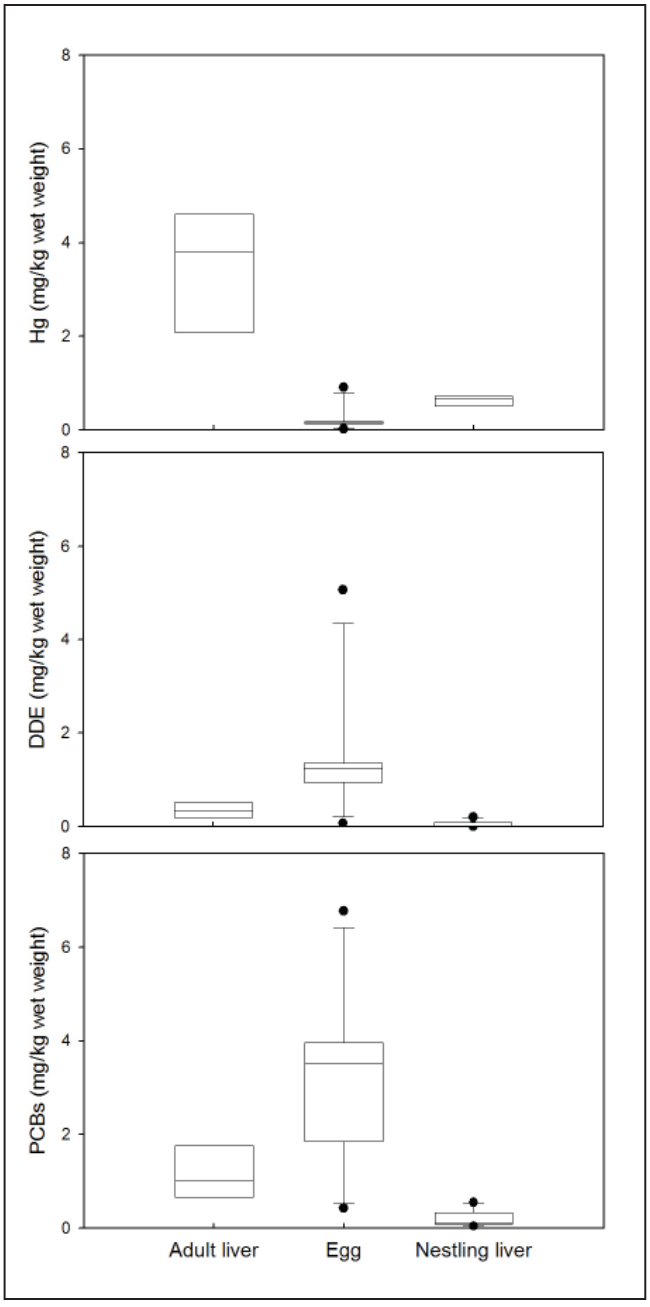


3.3 Contamination by organic compounds

3.3.1 Organochlorine pesticides

The 63 organochlorine compounds analyzed (22 organochlorines and 41 PCB congeners) were not detected in every sample. Egg samples contained variable combinations of 76% of them (SD 10; n = 55 pools [139 eggs]), liver samples had variable combinations of 62% of them (SD 25.3; n = 76 pools [105 nestlings]), and blood samples had variable combinations of 43.9% of them (SD 27; n = 30 pools [83 nestlings]) (Table 3).

Figure 3
Comparison of levels of mercury (Hg) (A), *p,p′*-DDE (B), and PCBs (C) in adult Great Blue Herons (*Ardea herodias*) (n = 4), Great Blue Heron eggs (n = 11), and Great Blue Heron nestlings (n = 10). Box plots show the median, upper and lower quartiles, and outliers.



p,p′-DDE, dieldrin, photomirex, mirex, *trans*- and *cis*-nonachlor, oxychlordane, and hexachlorobenzene were the organochlorine pesticides detected most frequently (>50%) in eggs and in the liver and blood of young herons (Rodrigue et al. 2005). Many of the organochlorine compounds of interest (e.g., 1,2,3,4-tetrachlorobenzene, 1,2,4,5-tetrachlorobenzene, pentachlorobenzene, and α-, β-, and γ-hexachlorocyclohexane) were not detected or were detected in very few tissues.

p,p′-DDE, a major metabolite of dichloro-diphenyl-trichloroethane (DDT = 4,4′-(2,2,2-trichloroethane-1,1-diyl)bis(chlorobenzene)), made up 98% of total DDTs in the eggs, 88% of total DDTs in the liver of young herons, and 84% of total DDTs in the blood of young herons (Rodrigue et al. 2005).

The mean *p,p′*-DDE concentration in the eggs was 3.85 µg/g wet weight (SD 5.1; n = 55 pools [139 eggs]; maximum 31 µg/g wet weight, Île du Petit Caoui in the lower estuary) (Table 4).

The mean *p,p′*-DDE level in the liver tissues of nestlings was 0.04 µg/g wet weight (SD 0.04; n = 76 pools [105 nestlings]; maximum 0.208 µg/g wet weight, Île de la Corneille in the fluvial/brackish estuary) (Table 4).

The mean level of *p,p′*-DDE in the blood was 0.006 µg/mL wet weight (SD 0.006; n = 30 pools [83 nestlings]; maximum 0.034 µg/mL wet weight, Île Manowin in the lower estuary) (Table 4).

The concentrations of *p,p′*-DDE in the liver and blood were also highly correlated with each other (Fig. 2b).

Note that 91% of the *p,p′*-DDE levels measured in the liver were below 0.1 µg/g wet weight; this likely reflects the nestlings' short exposure time (37 days on average) as well as a probable low level of exposure (Rodrigue et al. 2005).

The mean *p,p′*-DDE concentration in the liver tissues of adults captured in nests in a heron colony in the fluvial/brackish estuary was 0.32 µg/g wet weight (SD 0.15; n = 6; maximum 0.52 µg/g, Île de la Corneille). This value is five times lower than the concentration in the eggs they themselves had laid ($\bar{x}$ = 1.45 µg/g wet weight; SD = 1.26; n = 11; maximum 5.07 µg/g wet weight) but five times higher than the concentration in the liver tissues of young from the same colony ($\bar{x}$ = 0.06 µg/g wet weight; SD = 0.07; n = 6; maximum 0.21 µg/g wet weight) (Fig. 3b).

3.3.2 Polychlorinated biphenyls

The mean concentration of PCBs in the eggs of Great Blue Herons was 4.08 µg/g wet weight (maximum 29.67 µg/g wet weight, Île Matane in Lac Matapédia) (Fig. 3c, Table 5).

The mean total concentration of PCBs in the liver tissues of adults was 0.82 µg/g wet weight (n = 6) (Fig. 3c). The mean total concentration of PCBs in the liver tissues of

Table 3
Mean detection rate of organochlorine pesticides and PCBs in Great Blue Heron (*Ardea herodias*) eggs and nestlings from a total of 31 colonies in Quebec (25 colonies along the St. Lawrence system and 6 reference sites on inland lakes) between 1989 and 1994, expressed as a percentage (standard deviation, number of pools, and range of concentrations)

		Region									
	All colonies	St. Lawrence system				Inland lakes				Mann-Whitney	
Tissue	Mean percentage	Mean percentage	SD	*n*	Range (%)	Mean percentage	SD	*n*	Range (%)	*Z*	*P*[a]
Egg	76.3	77.2	10.6	44 (118 eggs)	57.8–94.9	72.8	6.4	11 (21 eggs)	59.4–84.4	–1.47	0.14
Nestling liver	62.7	65.0	25.6	56 (84 nestlings)	9.4–91.7	56.5	23.7	20 (21 nestlings)	15.9–80.0	–1.99	**0.05**
Nestling blood	43.9	48.4	28.5	23 (72 nestlings)	1.6–87.3	29.1	15.2	7 (11 nestlings)	18.8–59.4	–1.59	0.11

[a] Numbers in bold are significant ($P \leq 0.05$)

Table 4
Mean concentrations of *p,p′*-DDE (standard deviation, number of pools, and range of concentrations) in Great Blue Heron (*Ardea herodias*) eggs and nestlings from a total of 31 colonies in Quebec (25 colonies along the St. Lawrence system and 6 reference sites on inland lakes) between 1989 and 1994

	Egg (µg/g wet weight)				Nestling liver (µg/g wet weight)				Nestling blood (µg/ml wet weight)			
Region	$\overline{X}$	SD	*n* (eggs)	Range	$\overline{X}$	SD	*n* (nestlings)	Range	$\overline{X}$	SD	*n* (nestlings)	Range
Fluvial section (freshwater)	3.24	3.0	7 (31)	0.98–9.9	0.06[a]	0.04	25 (40)	0.015–0.140	0.006	0.002	6 (28)	0.004–0.010
Fluvial/brackish estuary	1.86	1.7	13 (21)	0.08–5.8	0.07	0.07	11 (12)	0.010–0.208	0.010	0.007	3 (7)	0.003–0.017
Lower estuary (saltwater)	5.08	7.1	21 (57)	0.04–31	0.03[b]	0.04	17 (29)	0.002–0.124	0.006	0.009	14 (37)	0.000–0.034
Gulf of St. Laurence	4.28	3.7	3 (9)	0.95–8.2	0.03	0.03	3 (3)	0.011–0.067				
Total St. Lawrence system	3.78	5.3	44 (118)	0.04–9.9	0.05	0.04	56 (84)	0.002–0.208	0.006	0.008	23 (72)	0.000–0.034
Inland lakes	4.11	4.5	11 (21)	1.61–17.3	0.03	0.03	20 (21)	0.005–0.097	0.005	0.006	7 (11)	0.001–0.011
All colonies	3.85	5.1	55 (139)	0.04–31	0.04	0.04	76 (105)	0.002–0.208	0.006	0.006	30 (83)	0.000–0.034

[a] Values with the same letter or no letter are not significantly different ($P > 0.05$)

nestlings was 0.15 µg/g wet weight (maximum 0.55 µg/g wet weight, Battures aux Loups Marins in the fluvial/brackish estuary). The corresponding concentration in the blood was 0.02 µg/mL wet weight (maximum 0.07 µg/mL wet weight, Île aux Hérons in the fluvial section) (Fig. 3c, Table 5).

The concentrations of PCBs in the liver and blood from young herons were highly correlated with each other (Fig. 2c). All of the levels obtained for both liver and blood (<0.6 µg/g wet weight in liver and <0.08 µg/mL wet weight in blood) were low compared to the levels in eggs; this finding undoubtedly reflects the nestlings' short exposure time (37 days on average).

The congeners that made up more than 50% of the total PCBs were numbers 118, 138, 153, and 180 for the eggs; numbers 118, 138, 153, and 182/187 for the liver; and numbers 118, 138, 149, and 153 for the blood.

3.4 Between-region comparisons

Mercury concentrations in the eggs collected from inland lakes (the reference colonies) were significantly higher than the concentrations in the eggs from the St. Lawrence River system (0.31 µg/g vs. 0.26 µg/g wet weight; $P < 0.05$) (Table 1).

Principal coordinate analysis (PCOORD) performed on the 13 main organochlorine compounds found in more than 50% of the eggs (*p,p′*-DDT, dichlorodiphenyldichloroethane [*p,p′*-DDD = 1-chloro-4-[2,2-dichloro-1-(4-chlorophenyl)ethyl]benzene], *p,p′*-DDE, *cis*-chlordane, *trans*- and *cis*-nonachlor, oxychlordane, heptachlor epoxide, dieldrin, penta- and hexachlorobenzene, octachlorostyrene, and PCBs) failed to reveal any regional patterns (Fig. 4).

This finding suggests that adult females are bioaccumulating the contaminants in part on their wintering and migration grounds, and this is masking any regional patterns. However, the females require a large amount of food during egg development and they lay their first egg about a month after they reach the colonies of the St. Lawrence (DesGranges 1995), so it is likely that the contamination in the eggs reflects in part the contaminant levels in food ingested on the breeding grounds (Lewis et al. 1992; Stewart et al. 1997; Evers et al. 2005).

A second principal coordinate analysis was carried out, this time on the 13 organochlorine compounds found in more than 50% of the livers of young herons (*p,p′*-DDD, *p,p′*-DDE, photomirex, mirex, *cis*-chlordane, *trans*- and *cis*-nonachlor, oxychlordane, hepatachlor epoxide, dieldrin, hexachlorobenzene, octachlorostyrene, and PCBs). This analysis revealed clear differences in contamination among the different geographical areas under study (Fig. 5).

Since young herons are more representative of regional pollution than heron eggs, young herons are more effective in documenting the spatial distribution of contaminants along the St. Lawrence. For example, a significant difference in the level of contamination was noted between the colonies of the river system and those located inland (percentage of *p,p′*-DDE and PCBs detected in liver tissues from young herons (wet weight): Mann-Whitney $U = -2.22$; $P \leq 0.03$). The concentrations of *p,p′*-DDE (Table 4) and of PCBs (Table 5) were also higher in the river proper than in the lower estuary, whereas levels of PCBs

Figure 4
Dispersion of the five study regions (the fluvial section, the fluvial/brackish estuary, and the lower estuary of the St. Lawrence River, the Gulf of St. Lawrence, and the inland lakes; see Figure 1) on the first two axes of a principal coordinate analysis (PCOORD) performed on the 13 organochlorine compounds detected in more than 50% of Great Blue Heron (*Ardea herodias*) eggs ($\bar{x}$ (SD); sample sizes in parentheses). See section 3.4 for the list of compounds.

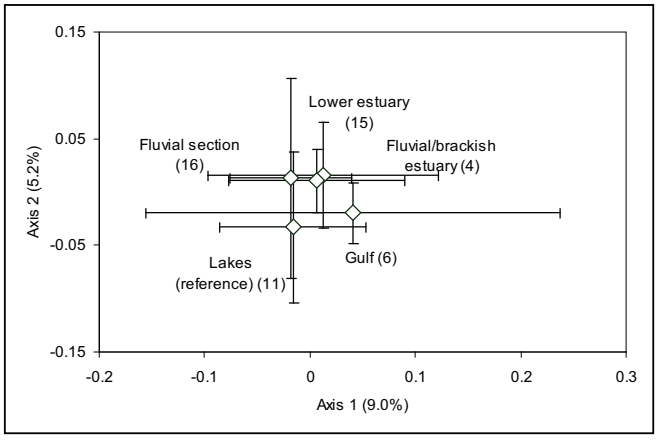


Table 5
Mean concentrations of PCBs (standard deviation, number of pools, and range of concentrations) in Great Blue Heron (*Ardea herodias*) eggs and nestlings from a total of 31 colonies in Quebec (25 colonies along the St. Lawrence system and 6 reference sites on inland lakes) between 1989 and 1994

	Egg (µg/g wet weight)				Liver (µg/g wet weight)				Blood (µg/mL wet weight)			
Region	$\bar{X}$	SD	*n* (eggs)	Range	$\bar{X}$	SD	*n* (nestlings)	Range	$\bar{X}$	SD	*n* (nestlings)	Range
Fluvial section (freshwater)	3.69	1.7	7 (31)	1.1–5.9	0.283[a]	0.140	25 (40)	0.078–0.505	0.040	0.021	6 (28)	0.022–0.071
Fluvial/brackish estuary	4.29	3.7	13 (21)	0.43–15.4	0.237[a]	0.182	11 (12)	0.047–0.551	0.037	0.029	3 (7)	0.009–0.067
Lower estuary (salt water)	3.42	4.5	21 (57)	0.28–18.7	0.047[b]	0.055	17 (29)	0.002–0.216	0.005	0.006	14 (37)	0.000–0.019
Gulf of St. Lawrence	8.72	9.9	3 (9)	2.76–20.1	0.094	0.056	3 (3)	0.045–0.155	-	-	-	-
Total St. Lawrence system	4.08	4.47	44 (118)	0.28–20.1	0.192	0.163	56 (84)	0.002–0.551	0.018	0.021	23 (72)	0.000–0.071
Inland lakes	4.23	8.5	11 (21)	0.32–29.7	0.041	0.059	20 (21)	0.006–0.268	0.006	0.007	7 (11)	0.001–0.022
All colonies	4.11	5.4	55 (139)	0.28–29.7	0.15	0.16	76 (105)	0.002–0.551	0.02	0.02	30 (83)	0.000–0.071

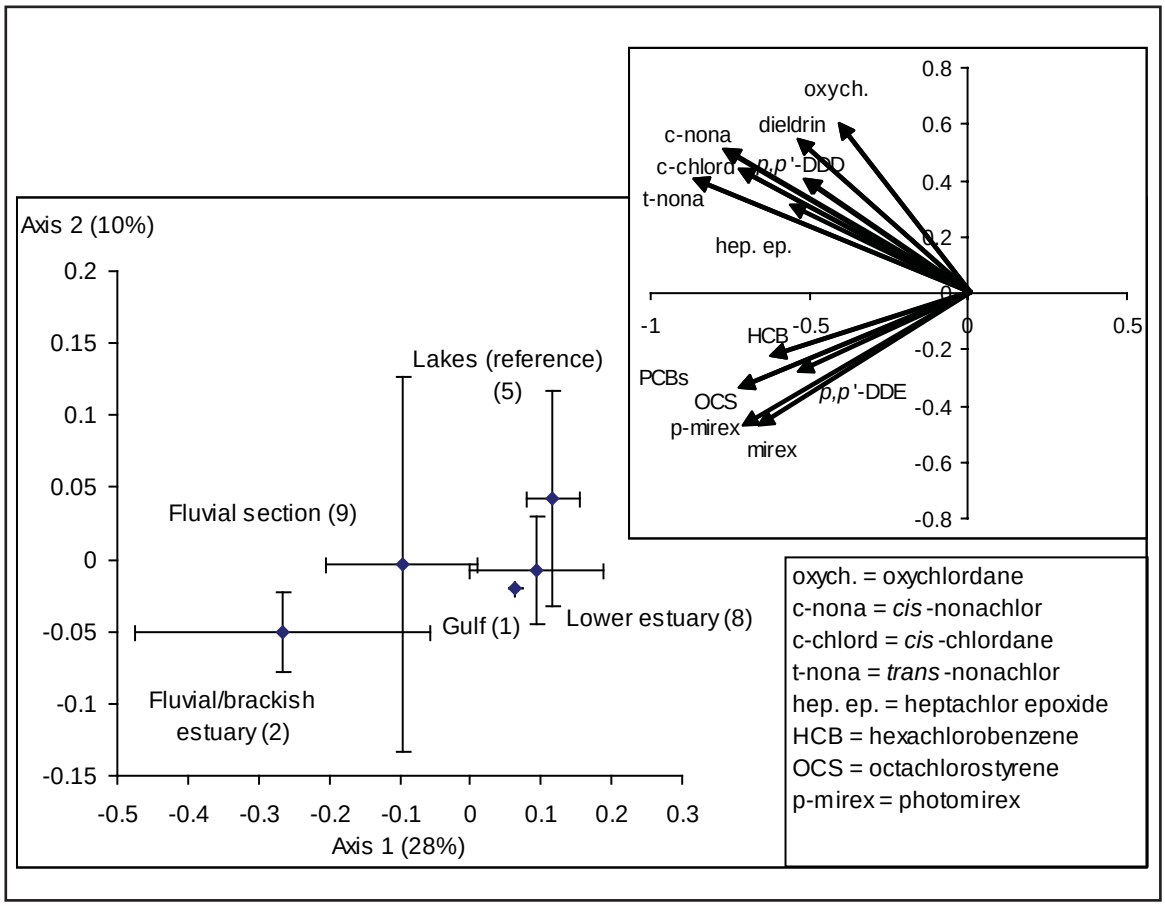
[a] Values with the same letter or no letter are not significantly different ($P > 0.05$)

were higher in the fluvial/brackish estuary than in the lower estuary ($q > 2.64$; $P < 0.05$). The proportions of homologues of PCBs in Great Blue Heron eggs did not differ between the St. Lawrence River system and the inland region (Mann-Whitney U; $P > 0.05$).

The proportion of homologues of PCBs with 3 or 4 chlorine atoms was higher in the liver tissues of nestlings from the St. Lawrence system than in the liver tissues of nestlings from inland lake colonies (Mann-Whitney $U = -2.84$ (St. Lawrence) and -4.34 (inland); $P < 0.005$), whereas the opposite was observed for homologues of PCBs with 7 or 8 chlorine atoms (Mann-Whitney $U = -3.91$ (St. Lawrence River) and -3.52 (inland lakes); $P < 0.0005$). Homologues of PCBs with 3, 4, or 8 chlorine atoms were present in larger quantities in the blood from nestlings from the St. Lawrence River system than in the blood from nestlings from the inland regions; the opposite was found for homologues with 6 chlorine atoms (Mann-Whitney $-2.91 < U < -2.27$).

Homologues of PCBs with 3 or 4 chlorine atoms were present in greater proportions in the liver tissues of nestlings from the river and the fluvial/brackish estuary than in the liver tissues of nestlings from the lower estuary and colonies located away from the St. Lawrence (Kruskal-Wallis $H > 24$; $P < 0.0001$). As we are dealing with proportions, it follows that the other homologues exhibited the opposite pattern. The homologues of PCBs with 3 or 4 chlorine atoms were found in greater proportions in the blood from nestlings from the river proper than from the areas away from the St. Lawrence, and the same result was found for homologues of PCBs with 4 atoms in the blood from nestlings from the fluvial/brackish estuary (Kruskal-Wallis $H > 10$; $P \leq 0.005$).

Figure 5
Dispersion of the five study regions (the fluvial section, the fluvial/brackish estuary, and the lower estuary of the St. Lawrence River, the Gulf of St. Lawrence, and the inland lakes; see Figure 1) on the first two axes of a principal coordinate analysis (PCOORD) performed on the 13 organochlorine compounds detected in more than 50% of the livers of young Great Blue Herons (*Ardea herodias*) ($\bar{x}$ (SD); sample sizes in parentheses). Vectors in the inset show the relative contribution of the 13 organochlorine compounds to the dispersion. See section 3.4 for the list of compounds.

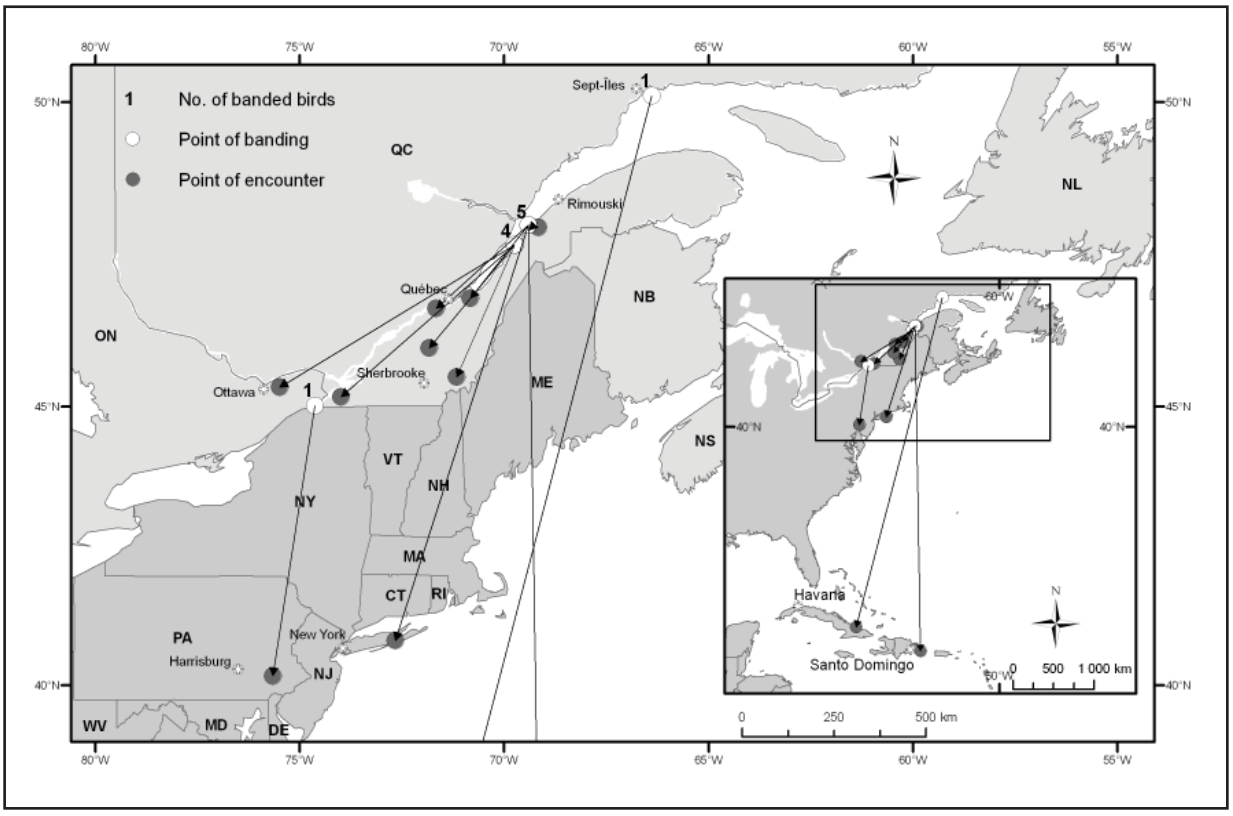


4. Discussion

This study shows that a number of toxic chemicals are bioaccumulating in Great Blue Herons of the St. Lawrence River system. Despite recent progress in decontamination made with the ongoing cleanup of the St. Lawrence River, contaminant levels measured in Great Blue Heron eggs during the 1990s are comparable (once adjusted for analytical differences) to those observed in 1979 (Laporte 1982). In addition, the mean concentration of mercury in the feathers of young Great Blue Herons hatched in Quebec (7.0 µg/g dry weight) is slightly above the concentration of mercury in chick feathers from wading birds (including Great Blue Herons) in Florida (3.5 µg/g) (Beyer et al. 1997). Moreover, the mean concentration of mercury in the liver tissues of adults from three colonies in Quebec was higher than the level found in the liver tissues of their own young (7.31 µg/g wet weight in 6 adults vs. 0.46 µg/g wet weight in 27 nestlings). This finding suggests that young may be more representative of regional pollution (i.e., pollution near their colonies) than their parents, whose contaminant levels at the beginning of the breeding season reflect, in part, the situation that prevailed on their migration and wintering grounds.

Figure 6 illustrates the banding return sites (encounters) for a total of 11 Great Blue Heron young banded at 4 colonies in Quebec. During the fall migration, Great Blue Herons in Quebec appear to travel up the St. Lawrence River as far as the central St. Lawrence lowlands (the region south of the St. Lawrence River extending from Cornwall to Quebec City) and then fly to the Atlantic coast and continue southward. At least some individuals fly as far as the Greater Antilles. This migratory route cuts through heavily

Figure 6
Banding and encounter locations of Great Blue Herons (*Ardea herodias*) banded in Quebec. The Canadian Bird Banding Office of the Canadian Wildlife Service, Environment Canada, graciously provided banding data.



industrialized regions and agricultural zones in which some pesticides and heavy metals are still used on a regular basis. These remote sources of contamination undoubtedly create the fuzziness in contaminant levels we detected in adult Great Blue Herons from Quebec and their eggs. A study of Osprey (*Pandion haliaetus*) contamination in northern Quebec (DesGranges et al. 1998) reached a similar conclusion.

With respect to *p,p′*-DDE, 36% of the eggs analyzed had a *p,p′*-DDE concentration higher than 3 µg/g wet weight, and 20% of the egg samples presented concentrations greater than 5 µg/g wet weight, which is the threshold for impairment of egg development in Ospreys (Noble and Elliott 1990; DesGranges et al. 1998). It should be noted that Vermeer and Reynolds (1970) found a DDE level of 78 µg/g in a Great Blue Heron egg in which the chick was pipping. A White-faced Ibis (*Plegadis chihi*) population in Utah where eggs showed DDE concentrations exceeding 3 µg/g produced 30% fewer chicks than another population with DDE levels below 1.5 µg/g (Steele 1984).

The average shell thickness of eggs collected in Quebec between 1989 and 1994 ranged from 0.29 mm to 0.60 mm, with a mean value of 0.41 mm (SD 0.04; $n = 208$) (Rodrigue et al. 2005). Mean eggshell thickness is therefore comparable to that measured during the period before *p,p′*-DDT was introduced in southern Canada in 1947 (0.39 mm; Anderson and Hickey 1972), and it is comparable to measurements from the United States (0.40 mm; Ohlendorf et al. 1979).

For PCBs, 78% of the eggs had concentrations lower than 5 µg/g wet weight, which is the threshold for impairment of embryo development (Eisler 1986). It should be noted that three composite samples of eggs (22%) had concentrations of PCBs exceeding 16 µg/g wet weight.

The fact that young herons reared in the freshwater part of the St. Lawrence River show greater levels of contamination than those reared near the lower estuary and those reared in inland colonies attests to the greater bioavailability of contaminants in the freshwater sector. It may even point to the ongoing discharge of bioaccumulative pollutants in this portion of the St. Lawrence, which receives inflows from the Great Lakes basin and from tributaries of the St. Lawrence that drain the most densely populated region of Quebec. Bonin et al. (1995) also found differences in contaminant levels in mudpuppies (*Necturus maculosus*) associated with different water masses of the freshwater portion of the St. Lawrence.

The findings of this research led to recommendations concerning long-term monitoring of contaminants in the Great Blue Heron and the use of this species as an indicator of the state of the waters of the St. Lawrence (DesGranges 1992; Champoux et al. 2000, 2002; Rodrigue et al. 2005). Although contaminant levels in Great Blue Herons are sometimes near or exceed the levels considered harmful to piscivorous birds, the Great Blue Herons of Quebec are considered to be in good health overall (Champoux et al. 2006). This probably explains why Quebec's Great Blue Heron population is currently stable or perhaps growing (DesGranges and Desrosiers 2006).

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Appendix

Colonies are presented in reverse order of longitude and increasing order of latitude.

St. Lawrence River and Gulf of St. Lawrence (25 colonies)

Freshwater section (7 colonies)

Île Dickerson	45°02′ N 74°35′ W
Île Carillon	45°31′ N 74°18′ W
Île Bienville	45°17′ N 74°10′ W
Île Dowker	45°24′ N 73°54′ W
Île aux Hérons	45°25′ N 73°35′ W
Grande île de Berthier	46°07′ N 73°01′ W
Bois-du-Boulé	46°15′ N 72°53′ W

Fluvial/brackish estuary (3 colonies)

Île de la Corneille	47°05′ N 70°36′ W
Battures aux Loups Marins	47°14′ N 70°26′ W
Grande île de Kamouraska	47°37′ N 69°52′ W

Lower estuary (11 colonies)

Île Le Gros Pot	47°52′ N 69°41′ W
Îlets Boisés	48°25′ N 69°19′ W
Île aux Basques	48°09′ N 69°15′ W
Île Laval	48°45′ N 69°02′ W
Île du Bic	48°24′ N 68°52′ W
Île Saint-Barnabé	48°28′ N 68°33′ W
Île de la Mine	49°03′ N 68°33′ W
Pointe Saint-Pancrace	49°16′ N 68°03′ W
Île aux Œufs	49°37′ N 67°11′ W
Île du Petit Caoui	49°49′ N 67°02′ W
Île Manowin	50°06′ N 66°24′ W

Gulf of St. Lawrence (4 colonies)

New Richmond	48°11′ N 65°50′ W
Île Beau Séjour	48°20′ N 64°42′ W
Miguasha	48°05′ N 66°19′ W
Îles de la Madeleine	47°38′ N 61°29′ W

Inland lakes (reference sites) (6 colonies)

James Bay (2 colonies)

Lac Kapwakami	53°04′ N 78°23′ W
Lac Evans	50°49′ N 76°58′ W

Laurentians (3 colonies)

Lac du Caribou	46°56′ N 72°50′ W
Lac Édouard	47°35′ N 72°22′ W
Petit lac Jacques-Cartier	47°24′ N 71°31′ W

Appalachians (1 colony)

Île Matane (Lac Matapédia)	48°35′ N 67°36′ W

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