

Temporal Trends of Organochlorine Concentrations in Umbilical Cord Blood of Newborns from the Lower North Shore of the St. Lawrence River (Québec, Canada)

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This study describes the time trends of organochlorines [OCs; 14 polychlorinated biphenyls (PCBs) and 11 chlorinated pesticides] in umbilical cord plasma of newborns in a remote Canadian coastal population. We analyzed 408 cord blood samples collected between 1993 and 2000 for PCBs, chlordanes, dichlorodiphenyltrichloroethane (DDT), dichlorodiphenyltrichloroethylene (DDE), hexachlorobenzene (HCB), and n-3 fatty acids. We also gathered information on the mothers (age, past and present residence, ethnic group, use of tobacco during pregnancy, and breast-feeding during previous pregnancies). From 1993 to 2000, mean concentrations of PCBs, chlordanes, DDT/DDE, and HCB in cord blood decreased by 63%, 25%, 66%, and 69%, respectively ($p < 0.0001$). Multiple regression analysis with the year of birth as the main independent variable yielded a strong significant exponential decrease for all contaminants (in all age and ethnic groups). We detected no monthly or seasonal pattern. We used n-3 fatty acids concentration as a surrogate of maternal fish consumption. Fish consumption declined only slightly between 1993 and 2000, but this decrease did not contribute significantly to the reduction of OCs. These results show that prenatal exposure to persistent OCs has declined significantly between 1993 and 2000 in this population. **Key words:** aboriginal, chlorinated pesticides, coastal population, diet, environmental exposure, food contamination, human, newborn, polychlorinated biphenyls, pregnancy, seafood, time trend, umbilical cord blood. *Environ Health Perspect* 110:835–838 (2002). [Online 9 July 2002] <http://ehpnet1.niehs.nih.gov/docs/2002/110p835-838dallaire/abstract.html>

Persistent organochlorines (OCs) such as chlorinated pesticides and polychlorinated biphenyls (PCBs) have been emitted by industrialized countries for many years. Substantial regulatory actions have been taken since the late 1970s to limit their emission, but they are still released because of improper storage and ongoing use in certain parts of the world. These substances bioaccumulate in fat tissues, are biomagnified in the food chain, and reach high levels in top predator species (Muir et al. 1992, 1999; Braune et al. 1999). Human populations with seafood-rich diets have elevated concentrations of OCs in their blood (Dewailly et al. 1993; Ryan et al. 1997; Humphrey et al. 2000; Sjodin et al. 2000; Bjerregaard et al. 2001). Because OCs readily cross the placental barrier (Saxena et al. 1981; Jacobson et al. 1984; Ando et al. 1985), high concentrations in the blood of pregnant women usually cause significant prenatal exposure for the fetus (Schwartz et al. 1983; Moss 1997; Muckle et al. 2001).

A general downward trend of the concentrations of OCs in tissue of wild animals and humans has been observed in the last decade. Levels of dichlorodiphenyltrichloroethane (DDT) and PCBs decreased in tissues of freshwater fishes between 1976 and 1986 (Schmitt et al. 1999). In herring gull eggs from the Great Lakes, levels of PCBs and dichlorodiphenyltrichloroethylene (DDE) also declined until the mid 1980s,

after peaking in the mid 1970s (Hebert et al. 1994; Ryckman et al. 1994). A similar trend has been observed in the concentrations of PCBs in fat tissues of polar bears, ringed seals, and sea birds in the Canadian arctic (Muir et al. 1999; Muir and Norstrom 2000). In environmentally exposed humans, levels of PCBs in breast milk have dropped in the last 15 years in Germany (Schade and Heinzow 1998) and Sweden (Noren 1993; Noren and Meironyte 2000). In Michigan, PCB blood concentrations increased between 1973 and 1983 but declined between 1983 and 1993 in a control population but not in fish eaters (He et al. 2001). No recent study has been published on time trends of OCs environmental exposure for the fetus.

The lower north shore of the St. Lawrence River (Québec, Canada) is a remote coastal region of 15 small communities scattered along 400 km of marine coastline and inhabited by both Caucasians and natives (mainly Montagnais). Although consumption of fish is common in this region, the principal source of exposure to OCs comes from the consumption of seabird eggs (Dewailly et al. 1992). Since 1993, we have been collecting cord blood samples to monitor OCs prenatal exposure and temporal trends in this region. In light of the general downward trend of OCs environmental exposure of the last years, we report here the time trends in prenatal exposure to PCBs

and chlorinated pesticides for infants born between 1993 and 2000.

Materials and Methods

Subjects and blood sampling. Our target population consisted of pregnant women who have lived for at least 5 years in one of the 15 small communities of the lower north shore region of the St. Lawrence River (Québec, Canada; Figure 1). We invited mothers admitted for full-term delivery in the two regional health centers of the region to participate in the study. We conducted the first phase of this program between April 1993 and December 1997, and the second phase between November 1999 and January 2001. Of 432 women eligible for the study, 403 (93%) agreed to participate. We did not try to include women for whom the delivery was at risk or when special or urgent medical care was needed. Therefore, a slight bias toward healthier pregnancy may be present.

We asked the participants to answer a questionnaire on sociodemographic characteristics and to sign an informed consent form. The questionnaire included information about smoking, previous pregnancies, health problems, ethnic origin, and present and past residence. After the delivery, we collected 20 mL of umbilical cord blood, which we centrifuged and froze at -80°C . We sent the vials to the Institut National de Santé Publique du Québec (Québec City, Canada) every 3 months. We also reviewed the medical chart of the newborns to gather information on the newborns and the deliveries.

Determination of chlorinated compound levels. We determined concentrations of 14 PCB congeners (IUPAC nos. 28, 52, 99, 101, 105, 118, 128, 138, 153, 156, 170, 180, 183, and 187) and of 11 chlorinated

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pesticides [aldrine, α -chlordane, γ -chlordane, *cis*-nonachlor, *p,p'*-DDE, *p,p'*-DDT, hexachlorobenzene (HCB), mirex, oxychlordane, *trans*-nonachlor, and β -hexachlorocyclohexane] in blood samples by high-resolution

gas chromatography. We extracted plasma samples (2 mL), which we cleaned on florisil columns, took to a final volume of 100 μ L, and analyzed on an HP-5890 series II gas chromatograph equipped with dual-capillary

columns and dual Ni-63 electron-capture detectors (Hewlett-Packard, Palo Alto, CA, USA). We identified peaks by their relative retention times obtained on the two columns, using a computer program developed in-house. The limit of detection was 0.02 μ g/L for PCB congeners and pesticides except for dieldrin, for which the detection limit was 0.1 μ g/L. For the sake of simplicity, we have grouped together the 14 PCB congeners, the chlordanes (*trans*-nonachlor, oxychlordane, *cis*-nonachlor, α -chlordane, γ -chlordane), and DDT and DDE (termed Σ PCBs, Σ chlordanes, and Σ DDT/DDE, respectively). Because OCs are stored mainly in body fat, we express all contaminant results on a lipid basis. We assigned a value of half the detection limit of the method when we did not detect a contaminant in a sample. The laboratory methods were rigorously the same for the entire study period.

Determination of blood lipids. We measured total cholesterol, free cholesterol, and triglycerides in plasma samples by standard enzymatic procedures and determined phospholipids according to the enzymatic method of Takayama et al. (1977) using a commercial kit (Wako Pure Chemical Industries, Richmond, VA, USA). We estimated the concentration of total plasma lipids according to the formula developed by Phillips et al. (1989).

Determination of n-3 concentrations. To determine the fatty acid composition in plasma phospholipids, we extracted 200- μ L aliquots of plasma following the addition of chloroform:methanol (2:1, v/v) in the presence of a known amount of internal standard (diheptadecanoyl phospholipid). We isolated total phospholipids from the lipid extract by thin-layer chromatography using heptane/isopropyl ether/acetic acid (60:40:3, v/v/v) as the developing solvent. Following transmethylation, using BF₃/methanol, we determined the fatty acid profile by capillary gas-liquid chromatography. We express the fatty acid composition of plasma phospholipids as percentages of the total area of all fatty acid peaks from C14:0 to C24:1 (Holub et al. 1987). In this study, plasma phospholipid concentrations of fatty acids correspond to percentages of total fatty acids, by weight.

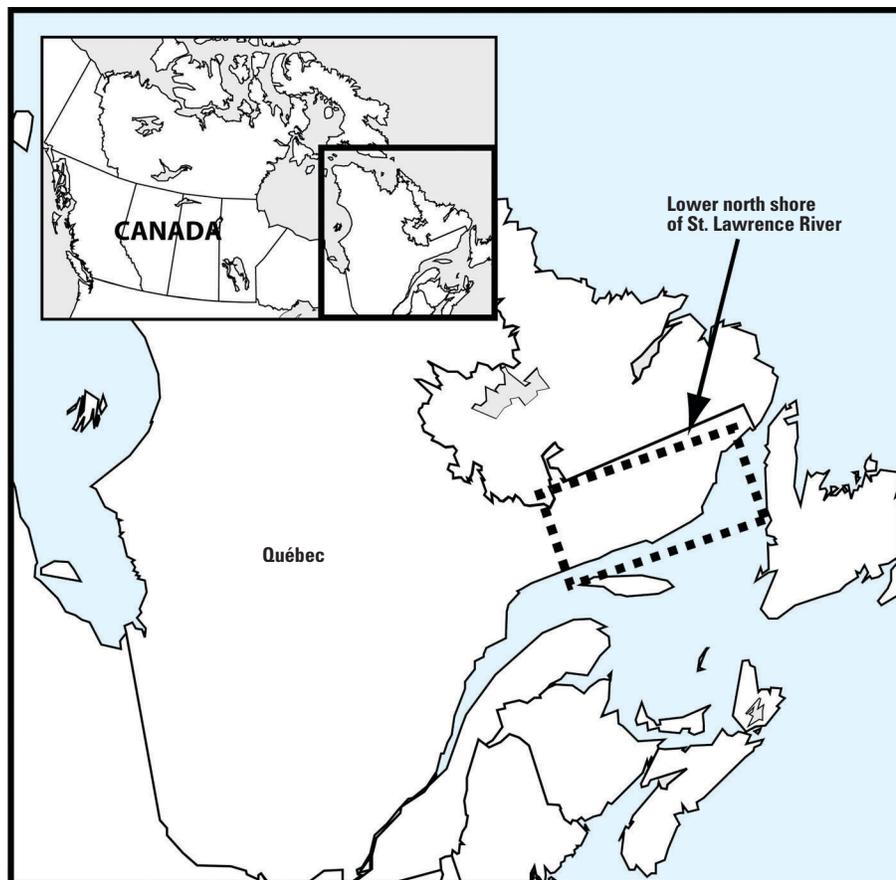


Figure 1. Location of the lower north shore region in Québec, Canada.

Table 1. Descriptive characteristics of mothers and newborns.

Characteristics	Total population	Caucasians	Natives
Mothers			
Number of participants (%)	392 (100)	224 (57.1)	168 (42.9)
Age (years) ^a	24.8 ± 5.6	26.0 ± 5.3	23.1 ± 5.7*
Number of previous pregnancies ^a	1.5 ± 1.4	1.1 ± 1.0	2.0 ± 1.8*
Proportion smoking during pregnancy (%)	57.3	41.6	77.6*
Newborns			
Sex (% proportion male)	50.2	51.8	48.2
Weight (kg) ^a	3.49 ± 0.50	3.40 ± 0.47	3.61 ± 0.52*
Length (cm) ^a	51.8 ± 2.3	51.5 ± 2.3	52.2 ± 2.1*

^aArithmetic means ± SD. *Significantly different compared to Caucasians (chi-square test for proportions and *t*-test for means, *p* < 0.05).

Table 2. Adjusted mean concentrations^a (and 95% confidence interval) of contaminants in cord blood by the year of birth.

Year of birth	Σ PCBs (μ g/kg)	Σ Chlordanes (μ g/kg)	Σ DDT/DDE (μ g/kg)	HCB (μ g/kg)
1993 (<i>n</i> = 62)	345.2 (301.5–395.2)	32.9 (30.7–35.3)	295.7 (264.0–331.3)	35.5 (32.0–39.2)
1994 (<i>n</i> = 62)	252.5 (220.5–289.2)	29.6 (27.6–31.7)	226.1 (201.7–253.3)	19.2 (17.3–21.2)*
1995 (<i>n</i> = 82)	243.5 (216.4–274.2)*	28.6 (26.9–30.4)	208.1 (188.5–229.9)*	17.9 (16.3–19.5)*
1996 (<i>n</i> = 71)	222.5 (195.9–252.8)*	28.1 (26.4–30.0)	182.4 (163.9–203.0)*	16.9 (15.4–18.6)*
1997 (<i>n</i> = 50)	188.5 (162.1–219.2)*	25.9 (24.0–28.0)*	154.6 (136.3–175.5)**	13.3 (11.9–14.9) [#]
2000 (<i>n</i> = 65)	153.5 (134.4–175.4) ^{##}	25.8 (24.1–27.6)*	119.1 (106.6–133.2) ^{##}	11.6 (10.5–12.8) ^{##}

^aGeometric means on a lipid basis adjusted for ethnic origin, region of residence, and age. *Concentration significantly different from that of 1993. **Concentration significantly different from that of 1993 and 1994. [#]Concentration significantly different from that of 1993, 1994, and 1995. ^{##}Concentration significantly different from that of 1993, 1994, 1995, and 1996. *t*-Test on adjusted means with Scheffe's correction for multiple comparisons.

Statistical analysis. Contaminant concentration variables had log-normal distributions, and we log-transformed them for all of the analyses. Therefore, we present all contaminant results as geometric means and confidence intervals. All the other variables had normal distributions, and we present them as arithmetic mean ± SD. Of the 403 infants, we included only 392 in the analysis because of missing values (usually caused by insufficient quantity of blood for all the chemical and biochemical analyses).

For the descriptive characteristics of the mothers and the newborns, we used the chi-square statistic to compare proportions and the *t*-test to compare unadjusted means. For temporal trends, we performed multiple regression modeling using the year of birth of the baby (treated as both category and continuous) as the main independent variable. We found a significant association between the region of residence (east or west) and the exposure. We decided to control for this variable because the recruitment of the participants was not equivalent in the two regions every year. We also controlled for the age of the mother (continuous) and the ethnic origin (native or Caucasian) because both of these characteristics are associated with exposure in this population (Dewailly et al. 1999). We used the *t*-statistic with Scheffe's correction for multiple comparisons to compare adjusted means. Only twelve births occurred late in 1999 and three early in 2001. We included these births in the year 2000 in all analyses. We performed the database management and all the statistical analyses with SAS software (SAS institute, Cary, NC, USA). By convention, we considered *p* < 0.05 significant.

Results

Table 1 presents the descriptive characteristics for the 392 mothers and newborns included in the analysis, stratified for ethnic origin. Caucasian women were significantly older and had lower parity than did native women. For native women, the proportion of smokers during pregnancy was almost twice that observed for Caucasian women (77.6% vs. 41.6%). Native women gave birth to significantly heavier and longer babies than did Caucasian women.

Table 2 presents the adjusted mean concentrations of contaminants in cord blood according to the year of birth of the baby. These results show a steady decrease of the mean concentrations for all groups of contaminants between 1993 and 2000. Between 1993 and 2000, the unadjusted mean levels of ΣPCBs, Σchlordanes, ΣDDT/DDE, and HCB in cord blood decreased by 63.1%, 25.2%, 65.7%, and 69.4% respectively (*p* < 0.0001). When adjusted for ethnicity, age of

the mother, and the region of residence, the decreases observed were slightly lower (50.6%, 21.0%, 55.4%, and 65.1% for ΣPCBs, Σchlordanes, ΣDDT/DDE, and HCB, respectively, *p* < 0.0001). We detected no significant seasonal or monthly pattern.

We calculated the adjusted annual decreases by multiple regression, treating the year of birth as a continuous variable. Table 3 presents these results stratified by ethnic origin. The trends for ΣPCBs, Σchlordanes, ΣDDT/DDE, and HCB for both ethnic origins were strongly significant (*p* < 0.0001). The only exception to this was the trend of the chlordanes concentrations in the Caucasian population, which was still significant but less than the other trends (*p* = 0.009). Because the calculated trends were linear using log-transformed dependent variables (contaminant concentrations), our results denote an exponential decrease of the concentrations of contaminants in cord blood between 1993 and 2000. When we analyzed PCB congeners separately, we observed the strongest decreases in the most abundant congeners. In fact, all the non-significant trends we observed were for congeners for which > 50% of the samples were

below the limit of detection. The annual decrease for the sum of the PCBs congeners increased from 10.0% to 12.2% when we excluded the least abundant congeners (detected in less than 50% of the samples). We observed little variation between trends according to the ethnic origin. Most of the differences were nonsignificant, but PCB congeners 99, 105, 156, and 183 showed a significantly steeper trend for natives than for Caucasians. When we stratified the data by the age of the mother, the downward trends were similar in every age group (data not shown).

We examined the proportion of n-3 fatty acids in the blood as a surrogate of long-term fish consumption (Silverman et al. 1990). We found a significant correlation between n-3 fatty acids and OC concentrations for natives but not for Caucasians (results not shown). We also found a slight nonsignificant decrease in n-3 proportions between 1993 and 2000 in natives. As shown in Table 4, the small variation in n-3 could not explain the diminution of OC concentrations. This indicates that a potential fish consumption diminution was only slightly responsible for the observed trends.

Table 3. Adjusted annual decreases (%)^a (and 95% confidence interval) of contaminants in cord blood between 1993 and 2000.

Contaminants	Total population (n = 392)	Caucasians (n = 224)	Natives (n = 168)
ΣChlordanes	3.1 (1.9–4.3)**	2.2 (0.6–3.8)*	4.1 (2.3–5.8)**
ΣDDT/DDE	11.6 (9.8–13.4)**	10.8 (8.2–13.4)**	12.4 (10.1–14.7)**
HCB	12.5 (10.8–14.2)**	12.3 (9.9–14.6)**	12.6 (10.0–15.0)**
ΣPCB congeners (%) ^b	10.0 (7.8–12.2)**	9.3 (6.2–12.3)**	10.6 (7.6–13.5)**
28 (4.9)	0.3 (–1.3–1.8)	–0.5 ^c (–2.3–1.3)	1.2 (–1.6–4.0)
52 (13.3)	0.2 (–1.4–1.7)	0.4 (–1.9–2.8)	–0.1 ^c (–1.8–1.3)
99 (73.3)	13.8 (11.6–16.0)**	12.0 (8.8–15.0)**	5.8 (12.7–18.8)**
101 (17.9)	6.5 (4.6–8.48)**	5.5 (3.1–7.8)**	8.0 (4.8–11.1)**
105 (29.1)	5.0 (3.0–6.9)**	1.3 (–0.9–3.5)	8.8 (5.8–11.8)**
118 (78.6)	13.1 (10.5–15.5)**	11.6 (8.0–15.0)**	14.3 (10.8–17.7)**
128 (2.0)	–0.6 ^c (–1.5–0.3)	–1.6 ^c (–2.6–0.5)*	0.8 (–0.8–2.3)
138 (96.4)	12.9 (10.2–15.4)**	13.3 (9.5–16.7)**	12.0 (8.6–15.3)**
153 (97.7)	12.2 (9.4–14.9)**	13.4 (9.3–17.2)**	10.5 (6.9–13.9)**
156 (46.7)	4.9 (2.7–7.0)**	2.7 (–0.2–5.5)	7.0 (4.0–9.9)**
170 (58.2)	9.6 (7.1–12.1)**	8.2 (4.7–11.6)**	10.9 (7.5–14.1)**
180 (88.8)	12.1 (9.3–14.8)**	12.0 (8.0–15.9)**	11.9 (8.3–15.5)**
183 (36.5)	3.6 (1.6–5.5)**	1.4 (–1.0–3.7)	5.8 (2.8–8.7)**
187 (75.0)	11.0 (8.5–13.4)**	10.5 (6.9–13.8)**	11.3 (7.9–14.6)**

^aPercentage of decrease per year calculated by multiple regression in which the year of birth is a continuous variable. The slope is adjusted for the region of residence, the age of the mother, and the ethnic origin (when applicable). Because the dependent variable (contaminants concentrations) of the regression was logarithmically transformed, each annual decrease and confidence interval were calculated from the slope (β) of the regression estimate (and its confidence interval) according to [100 × (1–e^β)]. ^bPercentage of the samples above the limit of detection. ^cA negative value denotes an increase, rather than a decrease. **p* < 0.01; ***p* < 0.001.

Table 4. Total decrease of OCs in cord blood between 1993 and 2000 attributed to n-3 fatty acids.

Contaminants	Adjusted ^a total decrease		Adjusted ^a decrease explained by n-3	
	Caucasians	Natives	Caucasians	Natives
ΣChlordanes	15.9%	23.9%	0.9%	1.4%
ΣDDT/DDE	53.3%	55.8%	1.6%	0.9%
HCB	66.8%	60.4%	2.0%	0.4%
ΣPCBs	47.7%	52.5%	0.7%	2.2%

^aThe differences between mean level of 1993 and 2000 were adjusted for the region of residence and the age of the mother.

Discussion

The results of this study underline a strong exponential downward trend of prenatal exposure to PCBs and chlorinated pesticides. We observed the trends for all the examined contaminants, in both ethnic groups and in all age groups. The substances considered in this study are all recognized persistent pollutants, and their use is prohibited or severely restricted in Canada. Despite a very long half-life, the reduction of the production and release of these OCs in the last 10–20 years is expected to result in a decrease of the burden in the environment, in wild animals and, ultimately, in humans.

The ubiquitous decrease in OC burden suggests that the generalized reduction in OCs in wild caught fish and animals might have played an important role. As noted in the introduction, a decrease of PCBs and DDT in wild animals and fish has already been observed (Muir et al. 1999; Schmitt et al. 1999; Muir and Norstrom 2000). The population targeted in this study is exposed to OCs mainly through seabird egg consumption (Dewailly et al. 1992). During the last 10 years, PCB, HCB, and DDE levels in herring gull eggs sampled in the lower north shore region have dropped by more than 85% (N. Burgess, unpublished results). This decrease has most likely played a key role in the trends observed in this study. A similar decrease in contaminants was also observed in herring gull eggs from the Great Lakes in the 1980s and early 1990s (Hebert et al. 1994; Ryckman et al. 1994).

We cannot, however, rule out the influence of a modification of dietary habits. The proportion of n-3 fatty acids in blood reflects the long-term fish consumption of an individual (Silverman et al. 1990). In our study, n-3 fatty acids did not show any significant time trend and could not explain the decrease observed in OCs. Reduction of fish consumption is thus unlikely to have contributed to the decrease of OCs in cord blood. However, a modification of seabird egg consumption could have greatly influenced the OC exposure of the mothers and, subsequently, of the newborns without affecting the proportion of n-3 in blood. Awareness of the level of contamination of specific species or types of tissues might have been involved. In 1996, a report from the public health board of the lower north shore region stated that a decrease of seabird egg consumption had been observed between 1990 and 1995 (Cartier 1996). Furthermore, many efforts have been made to inform this population of the contamination of seabird

eggs and of the potential effects of OCs of human health. A complete dietary study would be needed to better associate dietary changes with OC exposure.

The time trends of DDT/DDE and HCB concentrations observed in this study are similar to trends in breast milk found by Noren and Meironyte (2000). The trend for PCBs, however, was twice as strong in our study. Although the populations, the time intervals, and the analytical methods varied, the fact that we observed similar trends in two populations of mothers with different cultures, lifestyles, and diet supports the hypothesis that a generalized reduction of OC concentration in the environment has played a role in the diminution of OCs in our population.

To our knowledge, this is the first study evaluating the time trend of OC prenatal exposure. The fetus is particularly vulnerable to xenobiotics, and close monitoring of the populations at risk for high exposure is essential. This study shows that the situation has greatly improved in the last years. Prenatal exposure has diminished, and exposure through breast milk will most likely be decreased as well.

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