TOXAPHENE IN BELUGA WHALES (*Delphinapterus leucas*) FROM THE ST LAWRENCE, CANADA: LEVELS AND TRENDS

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Introduction

Toxaphene was first produced in the United States in 1947. Historically, the U.S. appeared to be the main producers and users of the toxaphene mixture in the world[1]. Concerns about the toxicity of this insecticide led the U.S. Environmental Protection Agency to progressively ban this product during the 1982-1986 period. In Canada, toxaphene has never been licensed for use, except for special minor applications[2]. Nevertheless, toxaphene is one of the most abundant organochlorinated pesticide in biota from Canada[3,4,5]. Aerial transport appears to be the most likely pathway for the introduction of toxaphene to Canada[6]. Soils from southern and southeastern U.S., where toxaphene have been extensively used, represent presumably the main source of this product to Canada[2].

After the U.S. toxaphene ban, concentrations in biota were expected to decrease in Canada. However, there were no significant reduction of toxaphene concentrations in marine mammals from the Arctic environment between the early 80s and the mid 90s[4]. In the Great Lakes, no clear temporal trend was observed for lake trout (*Salvelinus namaycush*) in Lake Superior between 1982 and 1992 [7]. However, a significant decrease of toxaphene concentrations was observed in lake trout from the Great Lakes, Michigan, Huron and Ontario, during the same time period[7]. In contrast, an increasing toxaphene contamination was suggested in beluga whales (*Delphinapterus leucas*) from the St Lawrence Estuary (SLE) between the late 80s and the early 90s[8]. These studies illustrate that temporal trends of toxaphene contamination differ between studied regions in Canada, which likely depend on their distinct contamination history. In addition, temporal trends of toxaphene contamination are complex to interpret mainly suffering from a relatively short time period since the toxaphene ban in the U.S.

The objective of this research was to measure levels of some of the most environmentally relevant toxaphene congeners (CHBs) in beluga whales from the SLE and to assess trends of toxaphene contamination during the 1988-1999 time period.

Material and methods

Samples. CHBs were measured in blubber samples of stranded beluga whales (26 males and 26 females) found on the shores of the SLE during the 1988-1999 period. The blubber samples, once received at the Maurice-Lamontagne Institute, were stored at –20 ºC until analysis.

Chemical analysis. Six CHBs (P26, P40, P41, P44, P50 and P62) were analyzed following the method described in detail in Gouteux et al. (2002), except that the internal quantification standard

used was D_8-4,4'-DDD instead of D_8-4,4'-DDT[9]. Since P40 and P41 were not chromatographicaly separated, they are presented as P-40/41.

QA/QC. One procedural blank and one pilot whale (*Glopbicephala* sp.) blubber sample (SRM 1945) were included in every batch of ten samples. No signal was detected in blanks. Repeated analysis of SRM 1945 (n=12) resulted in coefficients of variation within 15 % of the average values. Our laboratory performances on toxaphene analysis were validated through the NCP Interlaboratory study (NCP II-4) [9]. The mean recovery of internal standard was 77% (CV = 29%).

Results and Discussion

Levels and relative abundance of CHBs in belugas from the SLE. P26 and P50 were detected in all samples, contrary to P40/41, P44 and P62, which were below the detection limits in a few samples. Undetectable concentrations were removed from all statistical analyses (Table 1).

Table 1: Sample sizes and CHB concentration means, medians and ranges in male and female belugas from the SLE between 1988 and 1999. Concentrations are expressed in ng/g wet weight.

CHBs	Male				Female			
	n[a]	Mean	Median	Range	n[a]	Mean	Median	Range
P26	26	710	710	320-1240	26	280	220	42-1110
P40/41	24	75	68	30-140	23	48	42	15-120
P44	25	39	32	11-79	24	27	25	8-67
P50	26	1510	1420	560-3060	26	520	350	140-1690
P62	26	32	24	6.4-77	24	30	25	2.5-82

[a] number of samples where CHB congeners were detectable

Male belugas were generally more contaminated than females. CHB levels were roughly 1.5-3 times higher in males than in females. Such a difference of CHB contamination between males and females has been previously reported for P26, P50 and total toxaphene in SLE belugas[5,8].

Throughout the 1988-1999 time period, congeners P50 and P26 were predominant in blubber samples of both male and female belugas, representing in average 61 ± 8 % and 31 ± 5 % of the sum of the six CHBs (ΣCHBs), respectively. Other CHBs contributed individually for less than 5 % of ΣCHBs for both sexes. The predominance of P26 and P50 is well recognized in biota, mainly in fish and marine mammals, but also in human milk and blood[9,10]. In some marine mammals, the sum of these two congeners represents up to 90 % of total toxaphene levels[10].

P50 mean concentrations reported in this study were in the lower range of those previously reported for SLE belugas over similar time periods whereas P26 concentrations were very similar (Table 2). Moreover, P26 and P50 mean concentrations in belugas measured in the present study were in the same order of magnitude as those reported for animals living in the Arctic environment suggesting that atmospheric transport represents the main input of CHBs to these two distinct environments (Table 2).

Table 2: Average concentrations ± SD (ng /g wet weight) of two major toxaphene congeners, P-26 and P-50, in blubber of belugas from the SLE and Arctic environments.

Region	Sex	Period	n	Age	P26	P50	Reference
SLE	M	88-90	6	22.2 ± 3.8	830 ± 230	1520 ± 430	This study
SLE	M	87-90	15	20.1 ± 7.7	1250 ± 745	4120 ± 2130	Muir et al. (*5*)
SLE	M	93-94	6	21.8 ± 7.7	804 250	1530 ± 530	This study
SLE	M	93-94	9	19.2 ± 9.3[b]	749 ± 137	1510 ± 350	Muir et al. (*8*)
Arctic	M	before 92[a]	8	n.a.	1300 ± 325[b]	2350 ± 380[b]	Stern et al. (*13*)
Arctic	M	before 97[a]	20	n.a.[c]	468	840	Muir et al. (*14*)
SLE	F	88-90	6	20.2 ± 7.1	460 ± 360	820 ± 640	This study
SLE	F	87-90	21	18.6 ± 6.6	493 ± 435	1770 ± 1460	Muir et al. (*5*)
SLE	F	93-94	2[d]	11.3 ± 1.3	210 ± 50	380 ± 110	This study
SLE	F	93-94	7	19.5 ± 8.6	574 ± 293	1440 ± 783	Muir et al. (*8*)
Arctic	F	before 92[a]	8	n.a.	600 ± 310[b]	1050 ± 690[b]	Stern et al. (*13*)
Arctic	F	before 97[a]	5	n.a.	304	630	Muir et al. (*14*)

[a] Sampling periods were not indicated in the two references considered which were published in 1992 and 1997, respectively [b] Concentrations are expressed in lipid weight [c] n.a. refers to not available [d] Deviation from the mean was considered instead of SD.

Temporal trends of CHBs. Preliminary statistical analyses revealed that CHB concentrations in belugas were not related to the age of the animals examined. A simple linear regression model followed by an analysis of variance was used to determine time trends of CHB concentrations in SLE beluga whales[11]. P40/41, P44 and P62 concentrations were significantly ($p < 0.020$) related to the sampling year in both females and males (Figure 1). For P26, distinctive trends were observed between males and females (Figure 1). For females, there was a significant weak effect of time ($p = 0.041$; $r^2 = 0.16$) while there was no effect for males ($p = 0.447$). For P50, no significant time trend was observed for both males ($p = 0.240$) and females ($p = 0.112$) (Figure 1).

All regressions showed a negative slope suggesting a general decline of CHB levels in beluga whales from the SLE during the 1988-1999 time period. However, significant temporal trends were weak reflecting the general large inter-individual and between-year variations of contaminant concentrations in marine mammals[12]. Moreover, P50, the most predominant CHB congener, showed no significant reduction of its levels between 1988 and 1999 in SLE belugas.

The significant declines in CHB concentrations allowed us to evaluate $t_{1/2}$, the time required to reduce CHB levels in beluga by a factor of two. Values of $t_{1/2}$ varied between 3.1 ± 0.5 years for P62 in females and 11.5 ± 8.6 years for P40/41 in males. In average, CHB concentrations declined by a factor two in 8.5 years during the 1988-1999 time period.

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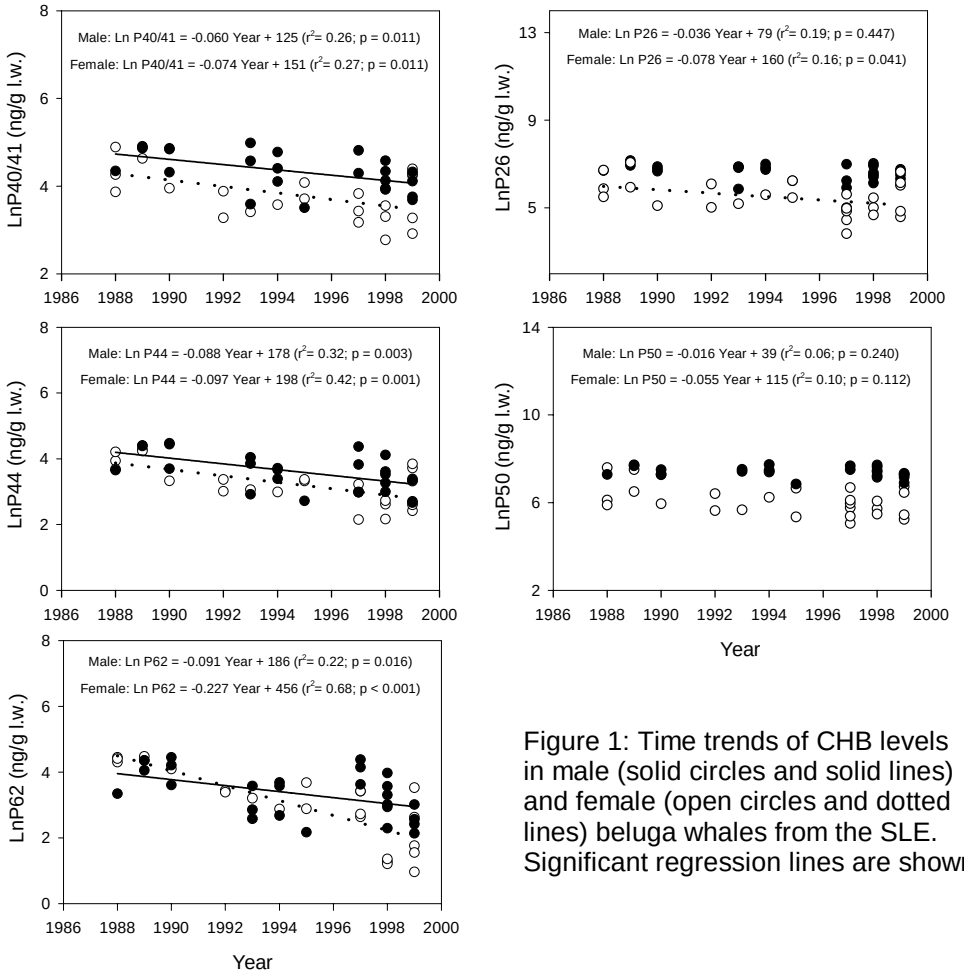

Figure 1: Time trends of CHB levels in male (solid circles and solid lines) and female (open circles and dotted lines) beluga whales from the SLE. Significant regression lines are shown.

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