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# Temporal trends (1987–2002) of persistent, bioaccumulative and toxic (PBT) chemicals in beluga whales (*Delphinapterus leucas*) from the St. Lawrence Estuary, Canada

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**Abstract**

Temporal trends of persistent, bioaccumulative and toxic (PBT) chemicals were examined in beluga whales (*Delphinapterus leucas*) from the St. Lawrence Estuary (SLE), Canada. Blubber samples of 86 adult belugas were collected from animals stranded on the shore of the SLE between 1987 and 2002 and analyzed for several regulated PBTs, including polychlorinated biphenyls (PCBs), p,p′dichlorodiphenyltrichloroethane (DDT) and its metabolites, chlordane (CHL) and related compounds, hexachlorocyclohexane (HCH) isomers, hexachlorobenzene (HCB) and Mirex. In addition, time trends of *tris*(4-chlorophenyl) methane (TCPMe) and *tris*(4-chlorophenyl)methanol (TCPMOH), two compounds that may origin from DDT formulations, were also examined. Concentrations of most of the PBTs examined had exponentially decreased by at least a factor of two (half-life time ($t_{1/2}$)<15 years) in beluga between 1987 and 2002 while no increasing trends were observed for any of the PBTs measured. The decreasing trends of PBT concentrations in SLE beluga may be due to a decline in contamination of its diet following North American and international regulations on the use and production of these compounds or by a change in its diet itself or by a combination of both. Some PBTs did not exhibit any significant trends in beluga possibly because the most intense elimination phase subsequent to legislative regulations occurred prior to the 1987–2002 time period. Other chemicals, such γ-HCH, did not significantly decrease likely because they are still currently used in some restricted applications. Conversely, α-HCH showed a significant decreasing trend indicating that ΣHCHs is not representative of all HCHs. Both TCPMe and TCPMOH exhibited no trends in beluga during the time period examined. The metabolic capacity of SLE beluga has apparently accelerated the depletion of at least one PBT, namely CB-28/31. A significant relationship between the half-life of PBTs in beluga and log Kow was observed for most of the chemicals examined. Several factors are expected to have influenced the temporal changes of PBT concentrations in beluga which limit the usefulness of this species as a bioindicator of changes in PBT contamination in the SLE ecosystem.


*Keywords:* Organochlorine pesticides; Polychlorinated biphenyls; Beluga whale; Temporal trends; *tris*(4-chlorophenyl)methanol; St. Lawrence Estuary Canada

## 1. Introduction

The population of beluga (*Delphinapterus leucas)* in the St. Lawrence Estuary (SLE) is currently listed as

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threatened by the Committee on the Status of Endangered Wildlife in Canada (COSEWIC, 2006). Historically, the size of that population has been estimated at some 7800–10000 animals but currently about 1100 individuals reside in the SLE (DFO, 2005). The large decrease in the size of the SLE beluga population was the direct result of hunting. Protected since 1979, the SLE beluga population would have been expected to increase but no clear sign of recovery over the last 15 years has been shown (Hammill et al., in press).

Several causes have been suggested to explain the lack of recovery of the SLE beluga population, including the potential toxicological impacts due to the presence of elevated concentrations of persistent, bioaccumulative and toxic chemicals (PBTs) in their tissues (Béland et al., 1993; Martineau et al., 1994; Bailey and Zinger, 1995). Recently, concentrations of polybrominated diphenyl ethers, a class of persistent organobromine chemicals, were reported to double every 3 years in SLE beluga (Lebeuf et al., 2004). The SLE beluga population inhabits an area located downstream of the industrialized and human populated Great-Lakes and the St. Lawrence River. It is believed that the presence of PBTs in the SLE mainly results from fluvial transport of toxic chemicals released and/or atmospherically deposited upstream in the large drainage basin of the Great-Lakes and St. Lawrence River (Lebeuf and Nunes, 2005).

Over the years, Canada, United States of America (USA) and several other countries, have taken regulatory actions to restrict or prohibit the manufacturing, the use and the import of several PBTs in agreement with the Stockholm convention whose objective is to reduce and/or eliminate the release of persistent organic pollutants (UNEP, 2001). The expected decrease of PBT concentrations in ecosystems and biota resulting from regulation of toxic chemicals is commonly assessed through temporal trend studies. In eastern Canada, time trends of PBTs in biota are relatively scarce although some trends have been reported in organisms such as fish, bird eggs, and marine mammals (e.g. Elliott et al., 1988; Addison and Stobo, 2001; Hickey et al., 2006). Temporal changes of several PBTs in SLE beluga between 1982 and 1994 have been reported by Muir et al. (1996b) while Gouteux et al. (2003) focused specifically on toxaphene congeners in beluga collected between 1988 and 1999.

The main objective of this study was to report the temporal trends of several regulated PBTs and their related compounds, including polychlorinated biphenyls (PCBs), 4,4′-dichlorodiphenyltrichloroethane (DDT) and its metabolites, technical chlordane (CHL) and related compounds, hexachlorocyclohexane (HCH) isomers, hexachlorobenzene (HCB) and Mirex in SLE beluga over a 15-year time period between 1987 and 2002. In addition, time trends were also examined for *tris*(4-chlorophenyl)methane (TCPMe) and *tris*(4-chlorophenyl)methanol (TCPMOH), two persistent and bioaccumulative compounds that may originate from DDT technical formulations (Buser, 1995). The influence of several factors on the time trends of these chemicals in beluga, including regulations, physico-chemical properties and propensity of these chemicals to be transformed by beluga were also assessed.

## 2. Materials and methods

### 2.1. Samples

Blubber samples were obtained from 42 female and 44 male stranded beluga whales found on the shores of the SLE between 1987 and 2002 (Fig. 1). Stranded dead beluga were found between April and December and for each animal the month of stranding was recorded. Carcass condition was classified as good, fair or poor (codes 2 to 4) according to the classification of Geraci and Lounsbury (2005), although intermediate coding was used occasionally (i.e. 2.5). Beluga carcasses collected prior to 1997 were not systematically coded except for those that underwent a necropsy at the veterinary laboratory of Université de Montréal (Saint-Hyacinthe, Québec). Growth layer groups (GLGs), representing pairs of light and dark lines, were counted on longitudinal tooth sections of each animal. Until recently, the age of beluga was estimated based on the hypothesis that beluga deposit two growth layer groups per year (Brodie, 1982) but a recent study provided new evidence that only one growth layer group was deposited yearly by beluga (Stewart et al., 2006). In this study, GLGs instead of age in years are reported and only adult animals with more than 20 GLGs were considered. The GLGs of several animals may have been underestimated due to difficulty in reading worn growth layers. Total length of each animal was measured from the rostrum to the notch of the tail fluke. In general, a sample block of skin-blubber-muscle was collected at 60–70% of body length from the rostrum, approximately midway between the spinal column and the mid-lateral region of each individual. The thickness of the blubber layer was recorded except for some animals collected prior to 1997 that did not undergo a necropsy. Most blubber samples collected prior to 1997 were initially separated in three layers identified as upper (adjacent to the skin), middle (mid-