Science of the Total Environment 485–486 (2014) 377–386

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

# A twenty-one year temporal trend of persistent organic pollutants in St. Lawrence Estuary beluga, Canada

Michel Lebeuf *, Lena Measures, Michelle Noël, Meriem Raach, Steve Trottier

*Department of Fisheries and Oceans, Maurice Lamontagne Institute, 850 Route de la Mer, Mont-Joli, Québec, Canada*

## HIGHLIGHTS

- Most legacy POPs in beluga exhibited weak but significant decreasing linear trends.
- Trends of most legacy POPs were equivalently described by more than one model.
- Temporal trends of PBDEs were best described by a two-segment piecewise model.
- Biological variables did not significantly affect trends of POPs in beluga.
- Trends of POPs observed in beluga are in agreement with regulations.

## ARTICLE INFO

*Article history:*
Received 5 December 2013
Received in revised form 26 February 2014
Accepted 20 March 2014
Available online 16 April 2014

Editor: F. Riget

*Keywords:*
Persistent organic pollutants
Beluga whale
Temporal trends
Models
Stable isotope
St. Lawrence Estuary

## ABSTRACT

Persistent organic pollutants (POPs) were measured in blubber from 144 stranded adult belugas (*Delphinapterus leucas*) found on the shores of the St. Lawrence Estuary (SLE) between 1987 and 2007. Temporal trends of POP concentrations (ln transformed) in beluga were described by three models, zero slope (ZS), linear (L) and two-segment piecewise (PW). Often two but sometimes all three models were equivalent in describing temporal trends based on Akaike's Information Criterion for small sample sizes. Over this 21-year time period, concentrations of most legacy POPs, including PCBs, DDTs and HCHs, exhibited relatively weak ($\leq$11% per year) but significant decreasing trends in beluga. For PBDEs, temporal trends were best described by a PW model, characterizing a rapid increase until 1997–1998 followed by a slower increase for males and a steady-state for females. Potential cofactors such as blubber lipid content and carcass state of preservation did not show any significant temporal trends over the time period considered. Nitrogen stable isotope ratios ($\delta^{15}N$) in beluga liver, a proxy of trophic level, could not be associated to any effect on temporal trends of POP concentrations in beluga. Several POPs exhibited significant relationships with age of beluga and data were age-adjusted. Temporal trends of POP concentrations adjusted for age of beluga were reassessed but results were essentially identical as those obtained with the original POP data. Overall, POP temporal trends observed in SLE beluga are consistent with changes expected from regulations and restrictions in the use of these compounds in developed countries.



## 1. Introduction

The population of beluga (*Delphinapterus leucas*) from the St. Lawrence Estuary (SLE) is threatened according to the Committee on the Status of Endangered Wildlife in Canada (COSEWIC, 2004). The population is estimated at only about 10% of what it was at the beginning of the 20th century, due to intensive exploitation until 1979. Despite various actions to protect the SLE beluga population, abundance indices suggest that it is not recovering (Hammill et al., 2007). The SLE beluga population is concentrated at the mouth of the Saguenay River, where it occupies a relatively small area of 2790 km$^2$ (Lemieux Lefebvre et al., 2012). The current summer home range has changed very little in the last 20 years, although the beluga range outside of summer is not well known. The SLE beluga live downstream of the Great Lakes and the St. Lawrence fluvial section, a densely populated, highly industrialized region of Canada and the United States. Although no single factor has been directly linked to the lack of recovery, this population inhabits a polluted ecosystem. The most recent analysis in the Species at Risk Recovery Strategy indicates that chemical pollution is considered a serious threat to the SLE beluga population (DFO, 2012).

Since Sergeant (1980), several studies have reported high concentrations of persistent organic pollutants (POPs as defined by the Stockholm Convention, 2001) in blubber from the SLE beluga (Lebeuf, 2009). Only

* Corresponding author at: Maurice Lamontagne Institute, Fisheries and Oceans Canada, 850 Route de la Mer, Mont-Joli, (Québec) G5H 3Z4, Canada. Tel.: +1 418 775 0690; fax: +1 418 775 0718.
*E-mail address:* michel.lebeuf@dfo-mpo.gc.ca (M. Lebeuf).

http://dx.doi.org/10.1016/j.scitotenv.2014.03.097
0048-9697/

a few studies reported long term (>10 years) time trends of either persistent organochlorine or organobromine compounds (Muir et al., 1996a, 1996b; Gouteux et al., 2003; Lebeuf et al., 2004, 2007). Results showed that legacy POPs such as polychlorinated biphenyls (PCBs) or various organochlorine pesticides (OCPs) are decreasing or do not exhibit significant temporal trends in SLE beluga. However, there is a need to provide an update of the current trends of these compounds in SLE beluga, especially for emerging polybrominated diphenyl ethers (PBDEs) that showed a strong increase during the 1980–90s. Temporal trend studies of POPs are useful, among other things, to report current contamination trends and to document the effects of government regulations or changes in industrial applications (Lebeuf and Nunes, 2005; Hickey et al., 2006).

Several biological variables such as sex and age have been recognized to influence the contamination of beluga and other marine mammals with POPs. Females are generally less contaminated with POPs than males because of an efficient transfer of most of these chemicals to their offspring during gestation and lactation (e.g. Desforges et al., 2012). A relationship between age and concentration of POPs is reported in marine mammals, including beluga, and the relationship is often more frequently demonstrated in males than females (Stern et al., 2005). Martineau et al. (1987) and Muir et al. (1996b) reported age vs POP concentration relationships in stranded SLE beluga while Hobbs et al. (2003) observed the same relationship in biopsied SLE beluga. Consequently, age of beluga, or standard length sometimes used as a proxy, could bias temporal trends of POPs if not accounted for (Hoguet et al., 2013). Changes in the ecosystem could also affect the contamination of beluga with POPs without changing the overall contamination of the ecosystem. For instance, it has been suggested that changes in environmental factors such as climate variability or changes in trophic dynamics could drastically affect POP concentrations in fish (French et al., 2006). Several fish populations, including Atlantic cod (*Gadus morhua*), Rainbow smelt (*Osmerus mordax*), Atlantic tomcod (*Microgadus tomcod*), and American eel (*Anguilla rostrata*) have drastically declined in the SLE (Castonguay et al., 1994; Myers et al., 1997). These fish species are believed to be part of the beluga diet (Vladykov, 1946; Hickie et al., 2000). Changes in fish populations may result in a change in the diet of beluga which could have an important effect on the level of contaminants acquired by beluga. Consequently, changes in the ecosystem, in particular in the structure of the food web, influence the interpretation of temporal trends of contaminants (Hebert and Weseloh, 2006). One way to assess the degree of change in the diet of beluga is by measuring nitrogen stable isotopes ($^{15}N$ and $^{14}N$) in beluga tissues over time. It is recognized that nitrogen stable isotopes are transferred differently from prey to predators, and the ratio ($^{15}N/^{14}N$) increases up the food chain (Minagawa and Wada, 1984; Kelly, 2000). To the best of our knowledge, no attempts have been made to link temporal trends of POPs in beluga with temporal trends in trophic position.

The main objective of this study was to provide an update on temporal trends of POPs in SLE beluga over a 21-year time period, between 1987 and 2007, including PBDEs for which recent regulations came into force in North America (Ward et al., 2008). Different models including zero slope (ZS), linear (L) and two-segment piecewise (PW) were used to characterize POP temporal trends in SLE beluga. This study also examined the influence of biological variables, including sex, age and trophic position of beluga, on temporal trends of POPs in beluga.

## 2. Materials and methods

### 2.1. Previously published and new data

PBDE data for 28 male and 26 female belugas were previously reported for the 1987–1999 time period by Lebeuf et al. (2004). This study reports new PBDE data for 16 males and 14 females for the 1987–1999 time period and extends this time period to 2007 with new PBDE data for 34 males and 24 females. PCB and OCP data were reported previously by Lebeuf et al. (2007) for 44 males and 42 females for the 1987–2002 time period. This study reports new PCB and OCP data for an additional 14 males and 4 females for the 1987–2002 time period and an extension of this time period to 2007 with new data for 20 males and 20 females.

### 2.2. Sampling

Samples were obtained from 66 female and 78 male stranded belugas found on the shores of the SLE between 1987 and 2007. During that time period, about 300 beluga carcasses, mostly adults, were found on the shores of the SLE as part of the SLE beluga carcass monitoring program initiated in 1983. This study reports data on approximately half of the carcasses found on the shores of the SLE during the 21-year time period examined.

Stranded belugas were found between March and December and the day of stranding expressed using the Julian calendar was calculated from the date of stranding recorded for each animal. Four animals were collected outside of the SLE. Based on their similar levels of POPs and nitrogen stable isotope ratios compared to other SLE belugas, these four individuals were considered to be from the SLE population and not from populations in the Canadian Arctic. Standard length of each animal was measured from the rostrum to the notch of the tail fluke. Carcass state of preservation was classified as good, fair or poor (codes 2 to 4) according to the classification of Geraci and Lounsbury (2005), although intermediate coding was also used (e.g. 2.5). Beluga carcasses collected prior to 1997 were not systematically coded except for those that were subject to a necropsy at the veterinary laboratory of the University of Montreal (Saint-Hyacinthe, Quebec). However, the large majority of beluga carcasses examined in this study were subject to a necropsy, namely 74% and 77% for males and females, respectively. The age of beluga was determined by counting growth layer groups (GLGs) on longitudinal tooth sections for each animal. The GLGs of some belugas may have been underestimated due to difficulty in reading worn growth layers. According to Stewart et al. (2006), the age of beluga in years corresponds to the number of GLGs and age in years is used in this study. In addition, teeth (available for 84% of beluga) were systematically read again in order to standardize the reading method, validate or correct previous readings. Only adult animals of 10 years or older were included in this study.

A block of skin–blubber–muscle was collected at 60–70% of the 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 belugas collected prior to 1997 that did not undergo a necropsy. Most blubber samples collected prior to 1997 were initially separated into three layers identified as distal (adjacent to the skin), middle (mid-blubber) and proximal (adjacent to the muscle), placed in individual solvent-rinsed glass jars and stored at −20 °C. In order to characterize the full depth of the blubber, layers were combined in equal quantities and homogenized before POP chemical analysis. For belugas sampled in 1997 and after, a block of blubber extending from the skin to the muscle was collected, wrapped in solvent-rinsed aluminum foil and placed in a sealed plastic bag, and stored at −20 °C until analysis. A subsample of the full depth of blubber (i.e. from the skin to the muscle) was taken from the block of blubber, homogenized and analyzed for POPs.

Among belugas from which blubber was sampled for POP analysis, 31 females and 31 males also had their liver sampled for nitrogen stable isotope analysis. Liver samples were collected from carcasses, immediately wrapped in solvent-rinsed foil, placed in a sealed plastic bag and then stored at −20 °C (Raach et al., 2011). Subsamples of liver were taken, homogenized and analyzed for nitrogen stable isotopes. Liver samples from beluga carcasses stranded before 1993 were not available.