

# Legacy contaminants in the eastern Beaufort Sea beluga whales (*Delphinapterus leucas*): are temporal trends reflecting regulations?<sup>1</sup>

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**Abstract:** Once deposited onto Arctic ecosystems, persistent organic pollutants (POPs) biomagnify in foodwebs such that relatively high levels have been detected in predators like beluga whales (*Delphinapterus leucas*). Our study aimed at assessing temporal trends of legacy POPs in eastern Beaufort Sea beluga blubber collected during traditional harvests in the Mackenzie Estuary area, Northwest Territories. Concentrations of polychlorinated biphenyls (PCBs) and 14 pesticides were quantified in 185 blubber samples collected between 1989 and 2015. The majority of legacy POPs analyzed showed no significant changes during the study period ( $\Sigma$ PCBs,  $\Sigma$ chlordanes,  $\Sigma$ dichlorodiphenyltrichloroethane, hexachlorobenzene, dieldrin, and mirex) and, therefore, did not reflect regulations put into place over the past decades. Although  $\alpha$ - (82.6  $\pm$  12.6 and 15.9  $\pm$  1.9 ng/g in 1989 and 2015, respectively) and  $\gamma$ -hexachlorocyclohexane ( $\gamma$ -HCH) (126.6  $\pm$  23.9 and 13.9  $\pm$  0.9 ng/g, respectively) showed significant decrease between 1989 and 2015,  $\beta$ -HCH showed a more complex trend with concentrations increasing between 1989 (71.2  $\pm$  13.4 ng/g) and 2004 (276.8  $\pm$  13.5 ng/g) before decreasing until 2015 (174.7  $\pm$  8.3 ng/g). Differences in trends likely reflect physico-chemical properties affecting transport to the Arctic. With climate change and melting sea ice potentially affecting the transport and release of legacy POPs, continuous monitoring is necessary.

**Key words:** PCBs, DDT, HCH, Inuvialuit settlement region, beluga whale.

**Résumé :** Une fois déposés dans les écosystèmes arctiques, les polluants organiques persistants (POP) sont sujets à la bioamplification dans la chaîne alimentaire à un point tel que des niveaux relativement élevés ont été détectés chez des prédateurs comme les bélugas (*Delphinapterus leucas*). Notre étude a visé à évaluer les tendances temporelles des POP historiques retrouvés dans la graisse prélevée de bélugas dans l'est de la mer de Beaufort au cours des récoltes traditionnelles dans la région de l'estuaire du Mackenzie, dans les Territoires du Nord-Ouest. Les concentrations de biphényles polychlorés (BPC) et de 14 pesticides ont été quantifiées dans 185 échantillons de graisse recueillis entre 1989 et 2015. La majorité des POP historiques analysés n'ont montré aucun changement important pendant la période d'étude ( $\Sigma$ BPC,  $\Sigma$ chlordanes,  $\Sigma$ dichlorodiphényltrichloroéthane, hexachlorobenzène, dieldrine et mirex) et n'ont donc pas reflété l'adoption des règlements mis en place au cours

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des décennies passées. Tandis que  $\alpha$ - (82,6  $\pm$  12,6 et 15,9  $\pm$  1,9 ng/g en 1989 et 2015, respectivement) et  $\gamma$ -hexachlorocyclohexane ( $\gamma$ -HCH) (126,6  $\pm$  23,9 et 13,9  $\pm$  0,9 ng/g, respectivement) ont montré une diminution importante entre 1989 et 2015,  $\beta$ -HCH a montré une tendance plus complexe composée de concentrations augmentant entre 1989 (71,2  $\pm$  13,4 ng/g) et 2004 (276,8  $\pm$  13,5 ng/g) avant de diminuer jusqu'en 2015 (174,7  $\pm$  8,3 ng/g). Les différences au niveau des tendances reflètent probablement des propriétés physicochimiques ayant une incidence sur le transport vers l'Arctique. Avec l'avènement du changement climatique et la fonte des glaces de mer ayant potentiellement une incidence sur le transport et le rejet des POP historiques, une surveillance continue s'avère nécessaire. [Traduit par la Rédaction]

*Mots-clés* : PCB, DDT, HCH, région désignée des Inuvialuit, béluga.

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## Introduction

Persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs) and various organochlorine pesticides [e.g., dichlorodiphenyltrichloroethane (DDT), dieldrin, hexachlorobenzene (HCB)] have been of concern for the past several decades. They share similar physico-chemical properties such as low water solubility and low vapour pressure, which allow them to be transported over great distances and bioaccumulated in the fatty tissue of various species leading to potential adverse health effects (Wania and Mackay 1993; Hoekstra et al. 2003; Borgå et al. 2004; Stern et al. 2005; Routti et al. 2010; Brown et al. 2014; Noël et al. 2014). The remote Arctic is not immune to contamination and has been described as a substantial sink for anthropogenic POPs (Macdonald et al. 2000). Although there exist a few local contaminant sources such as distant early warning line sites and community settlement areas, the majority of POP contamination in the Arctic originates from southern latitudes, in particular the populated and industrialized areas of Europe, North America, and Asia. POPs can reach the Arctic via atmospheric transport, ocean currents or river discharges (Barrie et al. 1992; Wania and MacKay 1993). However, long range atmospheric transport has been described as the major pathway delivering contaminants used in the mid- and low-latitudes to the Arctic in a matter of a few days (Wania and MacKay 1993; Bailey et al. 2000).

“Legacy” POPs refer to chemicals that were used historically. To eliminate or reduce the emissions of POPs, several international, national, and regional conventions have been developed with one of the most important ones being the Stockholm Convention on persistent organic pollutants. It was ratified in 2004 with originally 12 chemicals listed due to their persistence, toxicity, and bioaccumulative properties as well as their potential for long-range transport. As of 2017, another 16 POPs had been added to the list with the goal of restricting, prohibiting, and/or eliminating their production and use to protect human health and the environment (<http://chm.pops.int>). The Arctic Monitoring and Assessment Program (AMAP) was established in 1991 to report on contaminant levels in the circumpolar Arctic environment and northern aboriginal communities based on ongoing national and international research (AMAP 1998). At a national level, the Northern Contaminants Program (NCP) supported the development and collections of samples for contaminant monitoring throughout Canada's Arctic (NCP 2002). Both AMAP and NCP assessments emphasized the importance of long-term contaminant monitoring to not only identify emerging contaminants of concern but also, in the case of legacy POPs, to evaluate the effectiveness of regulatory measures that have been put into place to reduce their levels in the environment (AMAP 1998, 2004; NCP 2002).

Marine predators such as beluga whales (*Delphinapterus leucas*) have been described as ideal sentinel species for contaminant monitoring. They have long life spans, feed at a high trophic level and have large blubber stores that can accumulate POPs (Ross 2000; Bossart 2011). The eastern Beaufort Sea beluga population is one of the largest estimated at 40 000

(Harwood et al. 1996; Hill and DeMaster 1999). With one of the largest home ranges among circumpolar belugas, they migrate from their wintering grounds in the Bering Sea to the Beaufort Sea in the spring where they summer (Richard et al. 2001; Hauser et al. 2014; Citta et al. 2015). They form one of the world's largest summering aggregations in the Mackenzie Estuary where they are harvested for subsistence by local Inuvialuits (Harwood et al. 2002). The beluga harvest has been monitored for over 40 yr (Harwood et al. 2002) and has resulted in the collection of data and tissues for long-term monitoring of various contaminants as well as health endpoints. The NCP has supported the monitoring of contaminants including legacy POPs, metals and new and emerging chemicals for this population (Loseto et al. 2015; Smythe et al. 2018 this issue). As such, we are afforded the unique opportunity to examine temporal trends of POPs in the eastern Beaufort Sea Beluga population. In the present paper, we summarize over 25 yr of legacy POP data in beluga whale blubber including PCBs, DDT, HCB,  $\alpha$ -,  $\beta$ -, and  $\gamma$ -hexachlorocyclohexane (HCH), chlordanes, dieldrin, and mirex. Together with the important variables such as age, blubber thickness, diet (via biotracers), that have been recorded for all 185 whales sampled, we investigate the potential factors affecting the long-term trends of legacy POPs.

## Material and methods

### Sample collection

In collaboration with the annual beluga harvest by Inuvialuit hunters, blubber samples were collected from the Mackenzie Estuary at a field station near or at Hendrickson Island close to the community of Tuktoyaktuk, NT, Canada. Between 1989 and 2015, a total of 185 beluga samples were collected for contaminant analyses. Over the years, blubber samples were consistently taken from the ventral side just posterior of the arm fin to limit the potential for variability that may occur within the blubber layer. Samples were stored on site in a portable  $-20\text{ }^{\circ}\text{C}$  and shipped to Fisheries and Oceans Canada in Winnipeg where they were stored in a  $-80\text{ }^{\circ}\text{C}$  freezer until analysis. Length, girth, and blubber thickness were recorded. In addition, the jaw was removed for age determination from the teeth (Stewart et al. 2006).

### PCB and organochlorine pesticide analyses

The blubber layer is known to be stratified in terms of lipid deposition and contaminant concentrations (Krahn et al. 2004; Waugh et al. 2014). For contaminant analyses, the outer layer of the blubber was removed and a cross section of the layer from the surface to the inner layer was analyzed.

Blubber samples collected between 1989 and 2012 were analyzed at the Freshwater Institute, Fisheries and Oceans Canada, Winnipeg, Manitoba (MB) following procedures described elsewhere (Muir et al. 1990; Stern et al. 2005). In brief, 2 g of partially thawed blubber were combined with anhydrous  $\text{Na}_2\text{SO}_4$  prior to being extracted twice with hexane in a small (50 mL) ball mill. Surrogate recovery standards of PCB30 and octachloronaphthalene were added prior to extraction. Lipid content was determined gravimetrically on a fraction of the extract. About 100 mg of lipid was then separated on Florisil into three fractions of increasing polarity: (1) fraction eluted with hexane and containing PCBs, *p,p'*-DDE, *trans*-nonachlor, and mirex; (2) fraction eluted with hexane:DCM (85:15) containing  $\alpha$ -,  $\beta$ -, and  $\gamma$ -HCH, *p,p'*-DDD, *p,p'*-DDT, and chlordanes, and; (3) fraction eluted with a 1:1 mixture of hexane:DCM and containing dieldrin. After addition of aldrin as a volume corrector, each fraction was analyzed by high-resolution gas chromatography (GC) with  $^{63}\text{Ni}$  electron capture detection,  $\text{H}_2$  as the carrier gas and  $\text{N}_2$  as the make-up gas. Details on quality assurance can be found elsewhere (Stern et al. 2005). In brief, 40 organochlorine (OC) pesticides and 103 PCB congeners (including co-eluting) were quantified using external

**Table 1.** Blubber biopsy samples were collected from a total of 185 male beluga whales harvested at Hendrickson Island located in the Mackenzie Estuary near the community of Tuktoyaktuk, Northwest Territories (NT).

| Year | n  | Age (years) | Length (cm)  | Lipid (%)  | <sup>13</sup> C (‰) | <sup>15</sup> N (‰) |
|------|----|-------------|--------------|------------|---------------------|---------------------|
| 1989 | 12 | 25.0 ± 14.9 | NA           | 90.4 ± 1.1 | NA                  | NA                  |
| 1994 | 10 | 31.4 ± 10.7 | 416.8 ± 8.7  | 94.3 ± 0.9 | -18.1 ± 0.3         | 17.8 ± 0.3          |
| 1995 | 15 | 30.1 ± 10.7 | 421.1 ± 7.7  | 81.2 ± 0.9 | -18.1 ± 0.4         | 17.5 ± 0.5          |
| 1996 | 10 | 26.8 ± 6.8  | 411.8 ± 7.8  | 92.3 ± 1.4 | -18.7 ± 0.3         | 17.7 ± 0.2          |
| 2001 | 17 | 30.9 ± 10.0 | 387.8 ± 8.1  | 82.1 ± 0.9 | -18.8 ± 0.3         | 17.3 ± 0.3          |
| 2002 | 9  | 29.4 ± 8.1  | 420.8 ± 5.4  | 90.9 ± 0.8 | -18.4 ± 0.4         | 17.3 ± 0.2          |
| 2003 | 9  | 25.9 ± 9.3  | 408.3 ± 11.6 | 91.2 ± 1.3 | NA                  | NA                  |
| 2004 | 10 | 24.7 ± 5.6  | 415.6 ± 9.3  | 84.9 ± 2.2 | -18.6 ± 0.5         | 17.1 ± 0.4          |
| 2005 | 10 | 25.5 ± 9.6  | 415.3 ± 8.2  | 92.3 ± 1.1 | -18.3 ± 0.4         | 15.8 ± 0.1          |
| 2006 | 14 | 22.7 ± 7.2  | 406.4 ± 6.8  | 90.5 ± 1.7 | NA                  | NA                  |
| 2007 | 10 | 24.8 ± 6.6  | 422.7 ± 9.8  | 85.7 ± 2.2 | -18.3 ± 0.4         | 16.9 ± 2.2          |
| 2008 | 10 | 31.4 ± 9.3  | 411.3 ± 9.2  | 79.9 ± 1.9 | -17.9 ± 1.1         | 16.6 ± 0.9          |
| 2009 | 9  | 27.1 ± 2.6  | 408.4 ± 7.4  | 93.9 ± 1.3 | -17.9 ± 0.8         | 12.9 ± 1.2          |
| 2010 | 10 | 23.4 ± 4.2  | 420.0 ± 13.4 | 92.1 ± 1.1 | -18.4 ± 1.2         | 17.6 ± 1.2          |
| 2011 | 10 | 25.1 ± 6.2  | 407.1 ± 6.9  | 92.3 ± 1.2 | -18.9 ± 0.5         | 16.9 ± 0.5          |
| 2012 | 10 | 25.8 ± 7.1  | 406.3 ± 8.3  | 93.5 ± 0.6 | -18.9 ± 0.3         | 17.3 ± 0.4          |
| 2013 | 11 | 33.7 ± 7.5  | 414.1 ± 2.5  | 94.7 ± 2.9 | -19.3 ± 0.2         | 17.2 ± 0.7          |
| 2014 | 3  | 32.6 ± 1.5  | 395.4 ± 21.8 | 96.7 ± 1.6 | -19.7 ± 0.3         | 17.3 ± 0.5          |
| 2015 | 5  | 23.1 ± 4.3  | 397.1 ± 5.5  | 91.5 ± 2.1 | -19.4 ± 0.2         | 16.9 ± 0.2          |

standard mixtures (Ultra Scientific, North Kingstown, RI, USA). Standard reference material (NIST cod liver oil 1588) and duplicates were run every 12 samples.

Blubber samples collected after 2012 were analyzed at ALS Environmental, Winnipeg, MB. In brief, samples were mixed with sodium sulphate and Soxhlet extracted for 16 h with dichloromethane after the addition of a suite of labelled standards. The extracts were reduced in volume and lipids were removed via gel permeation chromatography (GPC). The resultant extracts were then split. A portion of the extract was prepared for analysis of organochlorine pesticides by low-resolution GC/MS using a mixed-bed chromatography column. A portion of the extract was prepared for analysis of PCBs by low-resolution GC/MS using acidified silica and alumina column chromatography.

#### Stable isotope analyses

Freeze-dried homogenized beluga muscle samples were used for carbon and nitrogen isotope analyses. A chloroform/methanol extraction was performed to remove the lipids for the carbon isotope determination. Analyses were performed using continuous flow, ion-ratio, mass spectrometry (CF-IRMS) (University of Winnipeg Isotope Laboratory, Winnipeg, MB, Canada). Details on the procedures as well as QA/QC methods can be found elsewhere (Loseto et al. 2008). Carbon and nitrogen results were expressed as standard delta ( $\delta$ ) notation in units of per mil or per 1000 (‰). The standards used for carbon and nitrogen analyses were Vienna PeeDee Belemnite (VPDB) and IAEN-N1 (IAEA, Vienna, Austria), respectively.

#### Data analyses

Inuvialuit hunters typically select medium to larger sized males, such that the number of females sampled was limited. While males tend to accumulate POPs throughout their lifetime, POP concentrations in females increase until they reach sexual maturity when they start offloading some of their POP burden to their offspring during gestation and lactation (Ross et al. 2000). To limit potential confounding influence due to gender, we only included males in the present analyses. There was no significant yearly difference in the age of the beluga whales sampled between 1989 and 2015 (Table 1, ANOVA:  $p > 0.05$ ) so that no age-correction of the data was necessary.

PCB and OC pesticide results were expressed on a lipid weight (lw) basis. Because not all congeners were detectable in all samples of beluga whale blubber, substitution had to be applied. Concentrations below the limit of detection were treated as zero in calculations (Vorkamp et al. 2011). In the rest of the paper, the compounds were grouped as follows:  $\Sigma$ PCBs = sum of congeners 28, 31, 52, 101, 105, 118, 138, 156, and 180;  $\Sigma$ chlordanes = sum of *trans*-nonachlor, *cis*-nonachlor, *trans*-chlordanes, *cis*-chlordanes, and oxychlordanes;  $\Sigma$ DDT = sum of *p,p'*-DDD, *p,p'*-DDE, and *p,p'*-DDT.

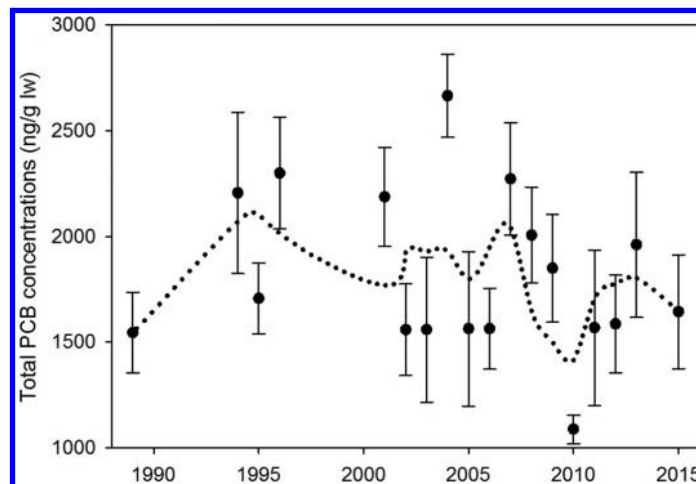
Unless otherwise stated, SPSS was used for statistical analyses (SPSS, version 20, IBM Corporation, Armonk, NY, USA). Normality and homogeneity of variances were tested for POPs, age, length, girth, blubber thickness,  $\delta^{15}\text{N}$ , and  $\delta^{13}\text{C}$  using the Kolmogorov–Smirnov and Levene's test, respectively. If the assumptions of normality and homogeneity of variances were not met, data were log transformed before further statistical analyses. Annual median concentrations were used for temporal trend analyses. This method has proven to be less sensitive to outliers and not influenced by how laboratories report below detection limit values than using geometric mean (Nicholson et al. 1998; Rigét et al. 2010). Outliers were detected by making boxplots and removed from further statistical analyses (Weijs et al. 2010). Simple regression analyses were performed to assess temporal changes of the legacy POPs analyzed and describe the linear component. Annual rates of change were calculated using the expression  $(1 - e^{-b}) \times 100$ , where  $b$  is the slope of the ln-linear regression. A three-point moving average was used to describe the non-linear component.

## Results and discussion

A total of 185 blubber samples were collected from beluga whales harvested between 1989 and 2015 in the Mackenzie delta, NT, Canada. The dataset was comprised of adult males of similar age between years ( $p > 0.05$ ) averaging  $28.1 \pm 9.1$  yr old (Table 1). Because diet is the main route of exposure to POPs, carbon and nitrogen stable isotope analyses were conducted and revealed significant changes over the study period ( $p < 0.001$  for carbon and nitrogen; Table 1). To evaluate potential temporal changes in diet that might affect contaminant accumulation, we examined temporal trends in  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  independently.  $\delta^{15}\text{N}$  decreased from 1990 until late 2000s ( $r = 0.53$ ,  $p < 0.001$ ) before remaining stable. In contrast,  $\delta^{13}\text{C}$  decreased over the study period ( $r = 0.30$ ,  $p < 0.001$ ). In their paper on mercury temporal trend in the same population of beluga whales, Loseto et al. (2015) discussed, in details, potential dietary shift between 1990 and 2012. They explained the challenges of using stable isotopes to infer diet for this particular population of beluga whales as they migrate every year from the Bering Sea to the Beaufort Sea and  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  baselines are known to differ between regions (Schell et al. 1998; Loseto et al. 2015). They proposed that the decline in  $\delta^{15}\text{N}$  may have resulted from feeding at lower trophic level and/or spatial variability in prey signals across their migratory path. While it could also explain variation in  $\delta^{13}\text{C}$ , the decrease of  $\delta^{13}\text{C}$  was suggested to be the result of a shift from feeding in a nearshore/benthic habitat potentially closer to the Bering/Chukchi area to feeding more pelagically in the Beaufort Sea food web (Loseto et al. 2015).

Legacy POPs, even though banned several decades ago, are still found in various environmental compartments including marine predators. In beluga whales sampled in 2015, the ranking of legacy POP concentrations from highest to lowest was  $\Sigma$ PCBs >  $\Sigma$ chlordanes >  $\Sigma$ DDTs > HCB >  $\beta$ -HCH > Mirex >  $\alpha$ -HCH >  $\gamma$ -HCH.  $\Sigma$ PCBs,  $\Sigma$ chlordanes, and  $\Sigma$ DDTs accounted between 77% and 89% of the total legacy POPs measured depending on the years. These trends are consistent with what has been reported in the Cook Inlet beluga whales (Hoguet et al. 2013). However, the pattern changed slightly over time with  $\Sigma$ DDTs dominating during two time periods: 1994–2001 ( $42.2\% \pm 5.7\%$  on average) and 2009–2012 ( $41.1\% \pm 6.5\%$  on average). While it remains unclear why DDT levels surpassed PCBs in some years,

**Fig. 1.** Total polychlorinated biphenyl (PCB) concentrations (median  $\pm$  standard deviation) in 185 beluga whale blubber samples collected on Hendrickson Island, NT, Canada, did not reveal any significant change between 1989 and 2015. A three-point moving average was used to smooth the trend (dotted line; [Nicholson et al. 1998](#)).



it might be related to the fact that there is likely a continuing minor input of “new” DDT by atmospheric transport into the eastern Beaufort Sea beluga food web as explained later in the paper.

#### Trends in PCB concentrations

There was no significant change in  $\Sigma$ PCB concentrations in beluga whale blubber between 1989 ( $1543.8 \pm 191.5$  ng/g lw) and 2015 ( $1643.4 \pm 267.4$  ng/g lw) ([Fig. 1](#)). After World War II, PCBs were widely used in dielectric fluids, elastic sealants, flame retardants as well as other applications that allowed for the expansion of safe electrical service in rapidly expanding cities. Historical PCB production has been estimated at 1300 kt with most of its use occurring in the Northern hemisphere ([Breivik et al. 2007](#)). Their manufacturing was banned by many countries in the 1970s including Canada who banned production and importation of PCBs in 1977 ([Diamond et al. 2010](#)) which resulted in significant decreases of their concentrations in various environmental matrices including arctic biota ([Rigét et al. 2006](#); [Lebeuf et al. 2007](#); [Lohmann et al. 2007](#)). The lack of significant change in PCB concentrations reported here between 1989 and 2015 is similar to what was observed previously in ringed seals (*Phoca hispida*) from Holman and Ulukhaktuk. In both studies, they reported that the bulk of PCB decline in seal blubber occurred until early 1980s ([Braune et al. 2005](#); [Addison et al. 2014](#)). However, while they reported an overall  $\Sigma$ PCB decline of 2.2% per year between 1972 and 2010 ([Addison et al. 2014](#)), the present results show an increase of 0.3% per year, although not significant. Similarly, East Greenland polar bears showed an insignificant increase in  $\Sigma$ PCBs between 2000 and 2010 while other subpopulations showed increases ranging from 2.5% to 15.3% per year. These trends were attributed to a combination of only slight declines of PCBs in Arctic air over the period of 1998 to 2006 and to a shift in diet resulting in slower rates of decrease of legacy POPs ([Dietz et al. 2013](#); [McKinney et al. 2015](#)).

The recalcitrant PCB-153 and 138 dominated the patterns throughout the period comprising on average  $27.7\% \pm 3.2\%$  and  $23.0\% \pm 5.9\%$  of  $\Sigma$ PCBs, respectively, over the study period. This is in accordance with previous studies reporting these two congeners as dominant in various cetacean species due to their resistance to metabolism ([Ross et al. 2000](#);

**Table 2.** Rate of annual change of all the legacy POPs analyzed in beluga whales between 1989 and 2015.

| Compound        | Annual change (%) |
|-----------------|-------------------|
| ΣPCBs           | 0.30              |
| PCB-52          | -0.30             |
| PCB-101         | 0.76              |
| PCB-118         | 0.30              |
| PCB-138         | 2.95              |
| PCB-153         | 0.57              |
| ΣDDTs           | -1.30             |
| DDT             | -3.7              |
| DDE             | -0.6              |
| DDD             | -3.1              |
| HCB             | -0.43             |
| α-HCH           | -4.77             |
| β-HCH*          | -1.22             |
| γ-HCH           | -5.35             |
| Σchlordanes     | 0.10              |
| Oxychlordanes   | 0.39              |
| Trans-chlordane | -4.03             |
| Cis-chlordane   | 3.60              |
| Trans-nonachlor | -0.19             |
| Cis-nonachlor   | -2.82             |
| Dieldrin        | 0.05              |
| Mirex           | -0.15             |

**Note:** \*represents the rate of decline between 2004 and 2015 as concentrations were increasing before then; bold represents significant trends ( $p < 0.05$ ).

Desforges et al. 2013a). Looking at temporal trends for the top five major individual congeners, Table 2 shows that, even though not significant, average annual changes range from a decline of 0.3% per year for PCB-52 to an increase of 2.9% per year for PCB-138. This is different from what was observed in ringed seals from Ulukhaktuk where all individual congeners showed a decline ranging from <1.5% to 4% per year (Addison et al. 2014). While the contributions to ΣPCBs of most of the congeners analyzed did not vary significantly over time, those of PCB-101, 138, and 180 did. The contribution of PCB-101 and 138 increased between 1989 and 2015 ( $r = 0.26$ ,  $p < 0.001$  and  $r = 0.22$ ,  $p < 0.001$ , respectively) as opposed to what was observed in ringed seals (Addison et al. 2014), maybe reflecting the limited ability of beluga whales to metabolize those specific congeners. In contrast, the contribution of PCB-180 decreased between 1989 and 2015 ( $r = 0.53$ ,  $p < 0.01$ ) which is opposite to what would be expected as this congener has a lower susceptibility to degradation due to the lack of unsubstituted vicinal positions (Addison et al. 2014). Together, these results suggest that patterns might be influenced by species-specific metabolism capabilities. In addition, it can be noted that PCB levels were associated with  $\delta^{13}\text{C}$  ( $r = 0.23$ ;  $p = 0.007$ ). While adjusting the data for  $\delta^{13}\text{C}$  (not shown) reduced the inter-year variation in PCB concentrations, the trend remained not significant over the study period. Overall, this suggests that a change in diet over time might also have contributed to change in PCB patterns. Similarly, McKinney et al. (2009, 2010) reported altered contaminant time trends in polar bears associated with feeding changes.

PCB levels reported here were in the same range as those reported in beluga whales from Cook Inlet and eastern Chuckchi Sea (1992–2005, Hogue et al. 2013), Nunavut (1995, Stern et al. 2005), and Svalbard, Norway (1995–1997, Andersen et al. 2001) but an order of magnitude lower than those reported in the highly contaminated St Lawrence

beluga whales (1987–1990, Muir et al. 1996). Even though levels might be considered moderate, 60% of the beluga whales sampled in 2015 surpassed the conservative 1.3 mg/kg lw threshold that has been developed for harbour seals which exhibited dose-related alterations in immune function, endocrine function (vitamin A and thyroid hormones), and gene expression (aryl hydrocarbon, retinoic acid, and thyroid hormone receptor) (Mos et al. 2010). PCB-related transcriptomic and endocrine disruption have been reported for this population of beluga whales (Desforges et al. 2013b; Noël et al. 2014) highlighting the importance of continuous monitoring of these “legacy” POPs that still pose a significant threat to the health of these beluga whales.

#### Trends in $\Sigma$ DDT concentrations

There was no significant change in  $\Sigma$ DDT concentrations in beluga whale blubber between 1989 ( $2341.4 \pm 370.1$  ng/g lw) and 2015 ( $1518.9 \pm 189.4$  ng/g lw) (Fig. 2). DDT was first developed in the 1940s and quickly became a broadly used pesticide to control malaria, typhus, various insect-borne human diseases as well as insects in crop and livestock production, institutions, homes, and gardens. Overall, DDT usage has been estimated at 4.5 Mt between 1950 and the mid-1990s (Li and Macdonald 2005). While DDT was banned by many countries including the United States and Western countries in the early 1970s, it is still produced and used for malaria control in other regions of the world (Li and Macdonald 2005). Nevertheless,  $\Sigma$ DDT concentrations have been decreasing in various biota including in the Arctic (Braune et al. 2005).  $\Sigma$ DDTs trend in our beluga whales was similar to the PCB trend and suggested that the majority of the decline happened before 1989. The average annual decline of 1.3% (Table 2) was lower than what was reported in ringed seals where the rates of decline of  $\Sigma$ DDTs ranged from 3% to 7.3% per year depending on location (Addison et al. 2014). Even though not significant for any of the three isomers, *p,p'*-DDT and *p,p'*-DDD declined at a rate of 3.7% and 3.1% per year, respectively, while *p,p'*-DDE only declined at a rate of 0.6% per year.

DDE is only found as impurities in commercial formulations of DDT so that most of its occurrence in the environment results from the dechlorination of DDT, a common process in all living organisms (Aguilar 1984). The conversion of DDT to DDE is a long process that continues for several years after input has stopped. DDE/ $\Sigma$ DDTs ratio is, therefore, a common indicator of the “age” of DDT in the environment. In the present study, this ratio increased at a rate of 0.5% per year (not significant) which is half what has been observed in Arctic ringed seals (1.2% per year; Addison et al. 2014). The difference might be the result of a combination of factors: (1) species difference in their ability to metabolize DDT; (2) difference in diet; and/or (3) continuing minor input of “new” DDT by atmospheric transport into the eastern Beaufort Sea beluga food web. Studies have shown that long-range transport continues to be a source of DDT to the Arctic (Bossi et al. 2013).

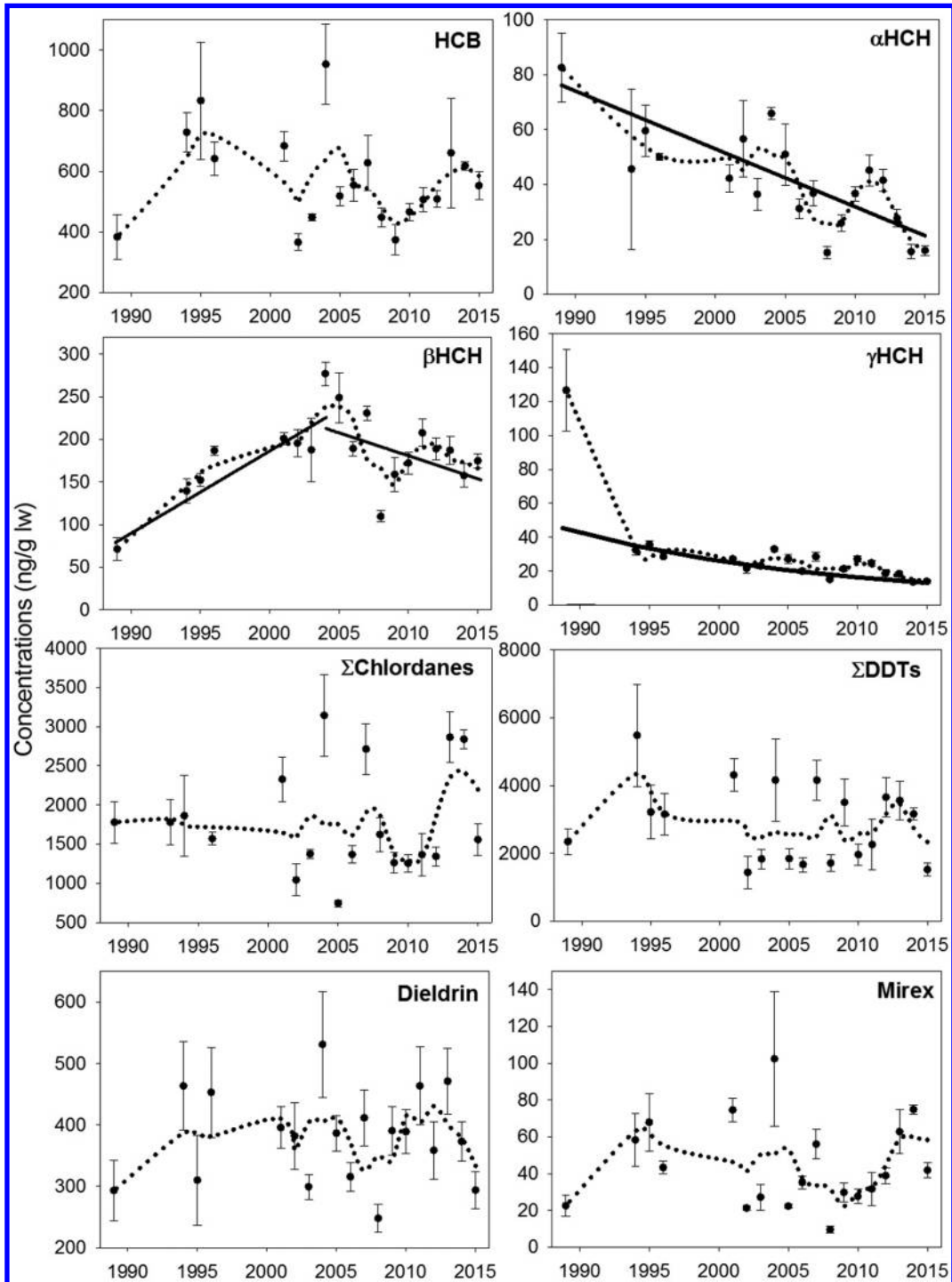
Levels reported in the present study are in the same range as those reported in beluga whales from Cook Inlet (1992–2005, Hogue et al. 2013) and Hudson Bay (1994, 1998; Stern et al. 2005) but approximately half what was observed in beluga whales from the eastern Chukchi Sea (1989–2000, Hogue et al. 2013) and Svalbard (Andersen et al. 2001). As expected,  $\Sigma$ DDT levels in the St Lawrence beluga whales were an order of magnitude higher than the present results (1987–1990, Muir et al. 1996).

#### Trends in HCH isomer concentrations

Three isomers of HCH are being reported:  $\alpha$ -HCH,  $\beta$ -HCH, and  $\gamma$ -HCH. HCH isomers were the only legacy POPs analyzed here showing significant changes between 1989 and 2015.  $\alpha$ - and  $\gamma$ -HCH both declined significantly between 1989 and 2015 ( $r = 0.56$ ,  $p < 0.001$ ;  $r = 0.91$ ,  $p < 0.001$ , respectively; Fig. 2). A breakpoint analysis using SegReg (<https://www.waterlog.info/segreg.htm>) identified 2004 as the breakpoint in the  $\beta$ -HCH dataset where  $\beta$ -HCH



**Fig. 2.**  $\alpha$ -,  $\beta$ -, and  $\gamma$ -HCH analyzed in 185 beluga whale blubber samples were the only legacy pesticides that showed significant changes between 1989 and 2015. A three-point moving average was used to smooth the trend (dotted line; Nicholson et al. 1998).



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increased between 1989 and 2004 ( $r = 0.81$ ,  $p < 0.001$ , Fig. 2) before showing a significant decline ( $r = 0.25$ ,  $p < 0.001$ , Fig. 2). HCH was a widely used insecticide between the 1940s and early 1990s. The technical formulation was comprised mainly of the  $\alpha$ -isomer (60%–70%) followed by  $\gamma$ - (10%–15%) and  $\beta$ -HCH (5%–12%), the latter being the only active ingredient in the formulation. Lindane consists almost entirely of  $\gamma$ -HCH (Iwata et al. 1993). Historical usage reached 1.5 Mt and 720 kt for technical HCH and lindane, respectively (Voldner and Li 1995; Li 1999). Although technical HCH is no longer used, applications of lindane continue in many countries (Li and Macdonald 2005). The decline of  $\alpha$ - and  $\gamma$ -HCH observed in the present study is in accordance with emission trends and general trends observed in Arctic air and water (Li and Macdonald 2005). Similar declines were observed in other biota such as beluga whales from the St Lawrence (1986–2002, Lebeuf et al. 2007) and Cumberland Sound (1982–2002, Braune et al. 2005), polar bears (*Ursus maritimus*) from Hudson Bay (1968–2002, Braune et al. 2005) and ringed seals from Greenland (Rigét et al. 2008). The rapid Arctic environmental response to change in global HCH emissions was attributed to the rapid delivery of those two isomers from southern latitudes to the Arctic via atmospheric transport (Li et al. 1998). Although the historical emission pattern for  $\beta$ -HCH is similar to that of  $\alpha$ -HCH, trends in the environment are usually very different just like what was observed here for the eastern Beaufort Sea beluga whales. Due to different physico-chemical properties,  $\alpha$ -HCH was directly transported to the Arctic via the atmosphere, whereas  $\beta$ -HCH partitioned into the waters of the Pacific Ocean through precipitation and air-sea exchange before the air masses could reach the Arctic.  $\beta$ -HCH was then transported northward through the Bering Sea to reach the Western Arctic (Li et al. 2002). Because transport via ocean currents is slower than via the atmosphere and because the Bering Sea acted as a “bottleneck” for the delivery of  $\beta$ -HCH to the Arctic,  $\beta$ -HCH concentrations peaked in Arctic water 10 yr after the  $\alpha$ -HCH peak (Li and Macdonald 2005). Similar to our results, levels of  $\beta$ -HCH increased in ringed seals from Ulukhaktok between 1978 and 2006 (Addison et al. 2009) and the authors suggested that it was due to a combination of delayed delivery of  $\beta$ -HCH to the Arctic and an additional lag for the incorporation of  $\beta$ -HCH into the food web.

$\beta$ -HCH was the dominant isomer in beluga whale blubber, especially in recent years. Since 2013, it has accounted for more than 80% of  $\Sigma$ HCHs. Its contribution increased dramatically between 1989 ( $26.5\% \pm 2.0\%$ ) and 2004 ( $74.0\% \pm 1.7\%$ ) reflecting the divergent temporal trends explained above. Patterns usually vary in biota with  $\alpha$ -HCH being dominant in ringed seals from Ulukhaktok (Addison et al. 2009) and fur seals from southern Alaska (Willett et al. 1998) but  $\beta$ -HCH dominating in harbour seals (*Phoca vitulina*) from the North Atlantic Ocean (Willett et al. 1998).  $\beta$ -HCH is known to be the most resistant to metabolism (Willett et al. 1998) so the current pattern observed in beluga whales may suggest a limited ability of this species to metabolize this compound compared to ringed seals. In addition, Fisk et al. (2003) reported a gradient in  $\beta$ -HCH contribution across the Arctic with ringed seals from the Bering-Chukchi region having higher  $\beta$ -HCH concentrations than further east. This is in accordance with seawater results where the Bering and Chukchi seas tend to have higher levels of  $\beta$ -HCH compared with the rest of the Arctic as a result of dilution of surface waters away from the prominent sources off India and Asia (Li and Macdonald 2005).

$\Sigma$ HCH levels in 2015 averaged  $199.3 \pm 8.2$  ng/g lw which is in the same range as those reported in beluga whales from Cook Inlet (1992–2005, Hoguet et al. 2013), Svalbard (1995–1997, Andersen et al. 2001) but half the levels reported in individuals from Hudson Bay (Stern et al. 2005) and the St Lawrence Estuary (1987–1990, Muir et al. 1996).

#### Trends in other OC pesticides

There was no significant change in HCB concentrations in beluga whales between 1989 and 2015 (Fig. 2). HCB was used as a fungicide starting in the 1930s. While emissions peaked

in the late 1970s, early 1980s before it was regulated, HCB is still being released in the environment through its presence as an impurity in other pesticides, industrial combustion and by-production during manufacture of chlorinated compounds (Barber et al. 2005). Significant declining trends have been reported for the St Lawrence beluga whales (1986–2002, Lebeuf et al. 2007) and ringed seals from Ulukhaktok (1972–2010, Addison et al. 2014) and West Greenland (1994–2006, Vorkamp et al. 2011). Although the decrease observed here was slower (0.43% per year, Table 2) than what has been reported before in ringed seals from various locations across the Arctic (2.8% to 3.8% per year, AMAP 2004), it was similar to that observed in ringed seals from the Beaufort Sea (0.86% per year) potentially suggesting that the current emissions of HCB might be more significant in the Western Arctic.

There was no significant change in  $\Sigma$ chlordanes concentrations between 1989 and 2015 (Fig. 2). Chlordane was used for a variety of insecticides and pesticides until the 1980s. As opposed to what was found here, declining trends have been observed in ringed seals from various Arctic locations (Addison et al. 2014) with declining rates ranging from 1.2% to 7.4% per year as well as in ringed seals from west Greenland (Vorkamp et al. 2011). However, in ringed seals from the Beaufort Sea, a positive trend was reported with a rate of 0.3% per year, similar to what was found here (0.1% per year, Table 2).  $\Sigma$ chlordanes was not representative of the behavior of individual congeners (Table 2). *Trans*-nonachlor dominated the pattern with a contribution ranging from 49% to 63% depending on the year followed by oxychlordanes with 22% to 43% contribution. These two congeners drove the lack of significant trend in  $\Sigma$ chlordanes. Oxychlordanes is known to be the primary metabolite of chlordane so the ratio between oxychlordanes and  $\Sigma$ chlordanes is usually used to provide an indication of the age of chlordane in the environment. In the present study, the ratio increased, although not significantly ( $p = 0.065$ ), between 1989 and 2012 which is consistent with what has been reported in ringed seals and polar bears from East Greenland (AMAP 2004). This suggests no new input of chlordanes to the marine environment during this time period. However, this ratio decreased in recent years from 0.38% in 2012 to 0.27% in 2015. In addition, there were significant relationships between  $\Sigma$ chlordanes and  $\delta^{15}\text{N}$  ( $r = 0.32$ ,  $p = 0.002$ ) and  $\delta^{13}\text{C}$  ( $r = 0.24$ ,  $p = 0.017$ ). While adjusting the data for  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  (not shown) reduced inter-year variation, the overall trend remained not significant over the study period. Taken together these results suggest that this shift might be the result of a shift in diet and/or recent new input of chlordanes to the Arctic environment. In 2015,  $\Sigma$ chlordanes levels ( $1556.6 \pm 199.7$  ng/g lw) were in the same range as levels reported in beluga whales from Nunavut and Hudson Bay (1995–1998, Stern et al. 2005) but slightly lower than those reported in individuals from Cook Inlet and the eastern Chukchi Sea (1992–2005, Hogue et al. 2013).

There were no significant changes in dieldrin and mirex concentrations in beluga whales between 1989 and 2015 (Fig. 2). Less information is available for those two OC pesticides. Dieldrin and mirex were used as insecticide until the late 1970s when they were regulated. Dieldrin is structurally very similar to aldrin, also a legacy OC pesticide, which breaks down into dieldrin in abiotic and biotic environments (Bann et al. 1956). In accordance with our current results, stable or slowly declining levels of dieldrin have been reported in the Arctic reflecting equilibrium between residual historical accumulation of dieldrin and aldrin in the Arctic environment and declining input of dieldrin to the Arctic marine food web (AMAP 2004). Similarly, there was no significant change in mirex concentrations in the blubber of the St Lawrence beluga whales between 1986 and 2002 (Lebeuf et al. 2007). Dieldrin and mirex were significantly correlated with  $\delta^{15}\text{N}$  ( $r = 0.23$ ,  $p = 0.022$  and  $r = 0.28$ ,  $p = 0.006$ , respectively) and  $\delta^{13}\text{C}$  ( $r = 0.21$ ,  $p = 0.045$  and  $r = 0.25$ ,  $p = 0.013$ , respectively) indicating that diet might play a role in the observed trends.

## Conclusion

Overall, our results showed no changes or a very slow decline for most legacy POPs analyzed. It is likely that most of the decline happened before 1989, right after the regulations were put into place (late 1970s for most of the compounds). While regulations have had a positive impact on the marine environment by preventing concentrations from increasing, the lack of continuing decline further demonstrates the persistence of those compounds but also suggests continuing inputs via atmospheric and oceanic pathways. While diet only appeared to be a factor influencing concentrations for some of the POPs:  $\Sigma$ PCBs,  $\Sigma$ chlordanes, mirex, and dieldrin, studies suggest that dietary changes associated with climate change could significantly impact contaminant concentrations. For example, McKinney et al. (2010) showed that, in polar bears, a change in diet associated with climate change could result in increased POP concentrations. In addition, climate change will also have some direct and indirect implications for contaminant cycling, deposition, and processing with the potential for legacy contaminants to be re-introduced into Arctic aquatic food webs (Macdonald et al. 2000; Grannas et al. 2013). As shown for PCBs in this population of belugas (Desforges et al. 2013b; Noël et al. 2014), these environmental contaminants continue to present health risks to high trophic level predators with marine mammals being particularly vulnerable to potential contaminant-associated health risks due to their long life spans and limited metabolism and excretion capability as well as low reproductive rates (Boon et al. 1994). This highlights the need to keep monitoring “legacy” POPs in the more than ever changing Arctic environment.

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