Temporal trend (1983 – 2024) of polychlorinated biphenyls (PCBs) in male Eastern Beaufort Sea beluga whales (*Delphinapterus leucas*)

Marie Noël, Gary Stern, Magali Houde, Dana Neumann, Ashley Elliott, Shannon MacPhee, Derek Muir, Gerald Inglangasuk, Tuktoyaktuk Hunters and Trappers Committee, Lisa L. Loseto

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ABSTRACT

Noël, M., Stern, G., Houde, M., Neumann, D., Elliot, A., MacPhee, S., Muir, D., Inglangasuk, G., Tuktoyaktuk Hunters and Trappers Committee, Loseto, L.L. 2026. Temporal trend (1983 – 2024) of polychlorinated biphenyls (PCBs) in male Eastern Beaufort Sea beluga whales (*Delphinapterus leucas*). Can. Tech. Rep. Fish. Aquat. Sci. 3819: vii + 23 p. https://doi.org/10.60825/64pa-sw56

The Eastern Beaufort Sea (EBS) beluga (*Delphinapterus leucas*) population, one of the largest beluga populations in Canada, is under increasing anthropogenic pressures including climate-related ecosystem changes and exposure to contaminants. Despite their regulations in the 1970s, legacy polychlorinated biphenyls (PCBs) remain a contaminant group of concern in many marine mammal species. Here, we aimed at updating the temporal trends of PCB levels in the blubber of EBS beluga whales from 1983 to 2024. Concentrations of PCBs were quantified in 250 blubber samples collected during traditional harvests in the Mackenzie Estuary area, Northwest Territories. Total PCB concentrations (ΣPCBs) declined linearly between 1983 and 2024 ($R^2 = 0.15$; $p < 0.05$) with a decrease of 1.7% per year. This represented an overall decline of 51% over the 41-year period (1983 mean: 5664 ng/g lw; 2024: 2780 ng/g lw). The dominant congeners showed different temporal trends so that their respective contributions to total PCBs varied across the time series. For example, the percent contributions of the top two dominant congeners, CB-153 and -138, to the ΣPCB were significantly different ($p < 0.05$) between 1983 (8.1 ± 0.24 and 7.6 ± 0.26%, respectively) and 2024 (15.3 ± 0.55 and 12.1 ± 0.51%, respectively) suggesting a change in PCB congener patterns over time. While the main reason for these changes remain unclear, climate-associated changes in some sources (secondary emissions), transport and fate of PCBs to the remote Arctic and changes in the EBS beluga food web and diet could all play a role. Assessing PCB-associated health risks using a beluga-specific health threshold of 1.6 mg/kg lw, we found that 100% of the samples exceeded this threshold from 1983 to the mid-2000s followed by more inter-annual variations (40 – 100%) and an average of 84% of samples exceeding the threshold since 2017, suggesting a decrease, albeit slight, in health risks associated with PCBs over the 41-year period. Given these shifts and important changes occurring in the Arctic, continued monitoring of these legacy compounds in EBS belugas is key along with additional information on diet and contamination of beluga prey to better understand PCB congener pattern changes over time and associated health risks.

RÉSUMÉ

Noël, M., Stern, G., Houde, M., Neumann, D., Elliot, A., MacPhee, S., Muir, D., Inglangasuk, G., Tuktoyaktuk Hunters and Trappers Committee, Loseto, L.L. 2026. Temporal trend (1983 – 2024) of polychlorinated biphenyls (PCBs) in male Eastern Beaufort Sea beluga whales (*Delphinapterus leucas*). Can. Tech. Rep. Fish. Aquat. Sci. 3819: vii + 23 p.
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La population de bélugas de l'est de la mer de Beaufort (EBS) (*Delphinapterus leucas*), l'une des plus grandes populations de bélugas au Canada, subit des pressions anthropiques croissantes, y compris des changements écosystémiques liés au climat et une exposition aux contaminants. Malgré leur réglementation dans les années 1970, les biphényles polychlorés (BPC) restent un groupe de contaminants préoccupant pour de nombreuses espèces de mammifères marins. Ici, notre objectif était de mettre à jour les tendances temporelles des niveaux de BPC dans le tissu adipeux des bélugas de l'EBS de 1983 à 2024. Les concentrations de BPC ont été quantifiées dans 250 échantillons de tissu adipeux collectés lors de chasses traditionnelles dans l'estuaire du Mackenzie, dans les Territoires du Nord-Ouest. Les concentrations totales de BPC (ΣBPC) ont diminué de manière linéaire entre 1983 et 2024 ($R2 = 0,15$; $p < 0,05$) avec une diminution de 1,7 % par an. Cela représentait une baisse globale de 51 % sur la période de 41 ans (1983 : 5664 ng/g lw ; 2024 : 2780 ng/g lw). Les congénères dominants ont montré des tendances temporelles différentes, de sorte que leurs contributions respectives aux BPC totaux variaient au cours de la série temporelle. Par exemple, les contributions des deux congénères dominants, CB-153 et -138, à ΣBPC étaient significativement différentes ($p < 0,05$) entre 1983 (8,1 ± 0,24 et 7,6 ± 0,26 %, respectivement) et 2024 (15,3 ± 0,55 et 12,1 ± 0,51 %, respectivement), suggérant un changement dans le profil des BPC au fil du temps. Bien que la raison principale de ces changements reste inconnue, des changements de provenance (émissions secondaires) des BPCs et transport vers l'Arctique liés au climat ainsi que des modifications dans le réseau trophique et l'alimentation des bélugas de l'EBS pourraient tous jouer un rôle. En évaluant les risques sanitaires associés aux BPC en utilisant un seuil de santé spécifique au béluga de 1,6 mg/kg lw, nous avons constaté que 100 % des échantillons dépassaient ce seuil de 1983 jusqu'au milieu des années 2000, suivis par des variations interannuelles plus importantes (40 à 100 %) et une moyenne de 84 % des échantillons dépassant le seuil depuis 2017, suggérant une diminution, quoique légère, des risques pour la santé associés aux BPC sur la période de 41 ans. Compte tenu de ces changements et des modifications importantes qui se produisent dans l'Arctique, le suivi continu de ces composés chez les bélugas de l'EBS est essentiel, ainsi que l'obtention d'informations supplémentaires sur l'alimentation et la contamination des proies des bélugas afin de mieux comprendre les changements dans les profils de congénères de BPC au fil du temps et les risques pour la santé qui y sont associés.

PREFACE

It is important to recognize the data presented in this study resulted from the generosity of hunters who kindly shared their food to be part of this research. Inuvialuit lead subsistence lifestyles as their ancestors did, and beluga, qilalugak, remain the most revered socio-cultural species (FJMC, 2024). The well-being and health of beluga whales are connected to the health and well-being of Inuvialuit, thus Inuvialuit concerns of contaminants in belugas and supporting ecosystem drives research and contaminants monitoring in the Inuvialuit Settlement Region (ISR).

Our study presents findings of polychlorinated biphenyls (PCBs) measured in beluga blubber for the purpose of tracking temporal trends and examining PCB concentrations in relation to thresholds of physiological impacts to belugas. The temporal trends inform management and policy on the fate and transport of PCBs to Arctic belugas. Currently, there are no consumption advisories in place with regards to consuming beluga whales, and the most recent human health assessment (2007-2008) concluded 'the benefits of eating country foods outweigh the risks from contaminant exposure'. This current report is not intended to supplement or provide guidance for food safety consumption, however these data have been made available for an updated evaluation of risks and benefits of consuming country food.

INTRODUCTION

The Eastern Beaufort Sea (EBS) beluga whale (*Delphinapterus leucas*) population is recognized as one of the largest in Canada, the last survey in 2019 estimated the population to approximately 32,500 individuals (DFO, 2023). With one of the largest known home range of the species, EBS belugas migrate in the spring from the Bering Sea to the Beaufort Sea (Fraker 1977; Citta et al., 2017; Storrie et al., 2022). They spend their summer with high site fidelity to the Mackenzie Estuary where aggregations of more than 7,000 surfaced belugas have been observed during early to mid-July (Norton and Harwood, 1985). In these waters, Inuvialuit have harvested beluga for subsistence for hundreds of years (Alunik et al., 2003; McGhee 1988) and beluga continue to be part of a traditional way of life and provide food and cultural well-being (Day, 2002; Usher 2002; Waugh et al., 2018).

While wildlife species of the remote Arctic are generally exposed to moderate levels of contaminants compared to their southern counterparts, there are an increasing number of studies reporting contaminant-associated health effects in Arctic marine mammals including beluga whales (Letcher et al. 2010; Desforges et al. 2013; Brown et al. 2014; Noël et al. 2014; Dietz et al., 2019). Levels of polychlorinated biphenyls (PCBs) in the EBS beluga population have been reported to be seven-fold lower than those reported in the highly contaminated St. Lawrence beluga whales (Noël et al., 2014). However, PCB-related transcriptomic and endocrine disruption have been reported for the EBS beluga whales (Desforges et al., 2013; Noël et al. 2014).

PCBs were first commercially produced in 1929 after which they were heavily used in electrical and hydraulic equipment, as well as in paint additives, sealing and caulking compounds and inks. Given the significant threat these pose to the environment, PCBs were banned by numerous countries in the 1970s. PCBs were among the 12 initial persistent organic pollutants (POPs), referred to as the “dirty dozen,” under the Stockholm Convention, and they are listed under Annex A (elimination) and Annex C (unintentional production). The Stockholm Convention came into force in 2004, and with it, came obligations from the countries that signed the treaty to phase out and ultimately ban PCBs from all production and use (Othman et al., 2022). In Canada, the manufacture, sale, and import of PCBs were banned in 1977 and their use and disposal are strictly regulated under the Canadian Environmental Protection Act (CEPA, 1999). In addition, the PCB regulations (SOR/2008-273) came into force in September 2005 with an amendment in 2015 to protect the health of Canadians and the environment by preventing the release of PCBs into the environment and accelerating their phasing out (Environment and Climate Change, 2018). As a result, the amount of PCBs used in Canada in equipment subject to PCB Regulations went from 1279.9 tons in 2008 to 0.0036 tons in 2022 (Government of Canada, 2023).

However, despite their regulation, there are still known sources of PCBs in cities of Western Europe and North America such as in electrical transformers (Diamond et al., 2010; Gasic et al., 2010). In addition, due to their persistence and bioaccumulative properties, PCBs remain important contaminants of concern detected at high concentrations in various marine mammal species (Mos et al., 2010; Desforges et al., 2018). In efforts to describe and understand the fate and transport of PCBs to the Arctic, research in the 1980’s began measuring PCBs in beluga whales (and other marine mammals) across the north (e.g. Muir et al., 1988, 1990). In 1991, the Northern Contaminants Program (NCP) began work throughout the Canadian Arctic to fund and coordinate research and monitoring of contaminants in the environment and in key species important to traditional diets. The NCP’s mandate is to reduce, and if possible, eliminate contaminants in traditionally harvested foods. But importantly, the NCP also provides valuable information on contaminant levels and trends to northern communities so that they can make

informed decisions on their dietary choices. The NCP also supports a wide range of international initiatives, including assessments of circumarctic contaminant patterns and trends in collaboration with the Arctic Monitoring and Assessment Programme in support of international and national treaties and regulations (Government of Canada, 2018). These early and ongoing efforts followed by ongoing and sustained support for long term contaminant monitoring have resulted in over 40 years of PCB data for EBS belugas and a number of other species.

Here, we present updated temporal trends of PCB levels in the blubber of EBS beluga whales from 1983 to 2024.

MATERIALS AND METHODS

SAMPLE COLLECTION

In collaboration with the annual traditional subsistence beluga harvest by Inuvialuit hunters from Mackenzie Delta, the Canada/Inuvialuit Fisheries Management Committee (FJMC), together with local Hunters and Trappers Committees and Fisheries and Oceans Canada have been collecting tissue samples since the 1980s to monitor beluga contaminant levels (Muir et al., 1990; Lockhart et al., 2005; Stern et al., 2005; Noël et al., 2018; Loseto et al., 2015). Hendrickson Island is a primary site where harvesters from the community of Tuktoyaktuk land and flense their whales to bring home meat and muktuk for food preparation. It is also the location with the largest consistent harvest of belugas in the ISR (Harwood et al., 2020; FJMC unpublished data) and one of the NCP core long term monitoring location for annual collection and analyses of tissues. In addition, this area is also part of the Tarium Niryutait Marine Protected Area (TN MPA), designated in 2010 with the objective to “conserve and protect beluga whales and other marine species, their habitats and their supporting ecosystem” (Gazette, 2010).

Blubber samples were collected from belugas landed at Hendrickson Island by community-based monitors trained in scientific sample collection methods. Samples were stored in a portable -20°C freezer before shipment to DFO in Winnipeg, Manitoba, where they were archived frozen at -20°C until analysis. Morphometrics including length, girth and sex are recorded on site for each individual (Harwood et al., 2002; 2020). From the start of the study until 2015, the ages of whales were determined using teeth (typically one tooth per individual, pulled from the right lower mandible, either from position 2 or 5 or the tooth that is least eroded or miss-shaped) and counting individual growth layer groups in the dentine (Stewart et al., 2006); from 2016 onwards, aspartic acid racemization measured in the nucleus of the eye balls were used for aging following methods from Pleskach et al. (2016).

In addition to samples collected at Hendrickson Island, we are including 12 samples collected from beluga whales entrapped in Husky Lakes in 1989 (described in Harwood et al., 2020). When possible, basic biological information (total length, sex, age structures) was taken along with blubber samples. While these entrapped whales were stressed at the time of sampling (Harwood and Smith 2002), the % lipid in blubber was within the range of other samples and the total PCB concentrations for that year were fitting the overall trend. It was therefore decided that it was appropriate to use data from these whales as part of the temporal trend analyses.

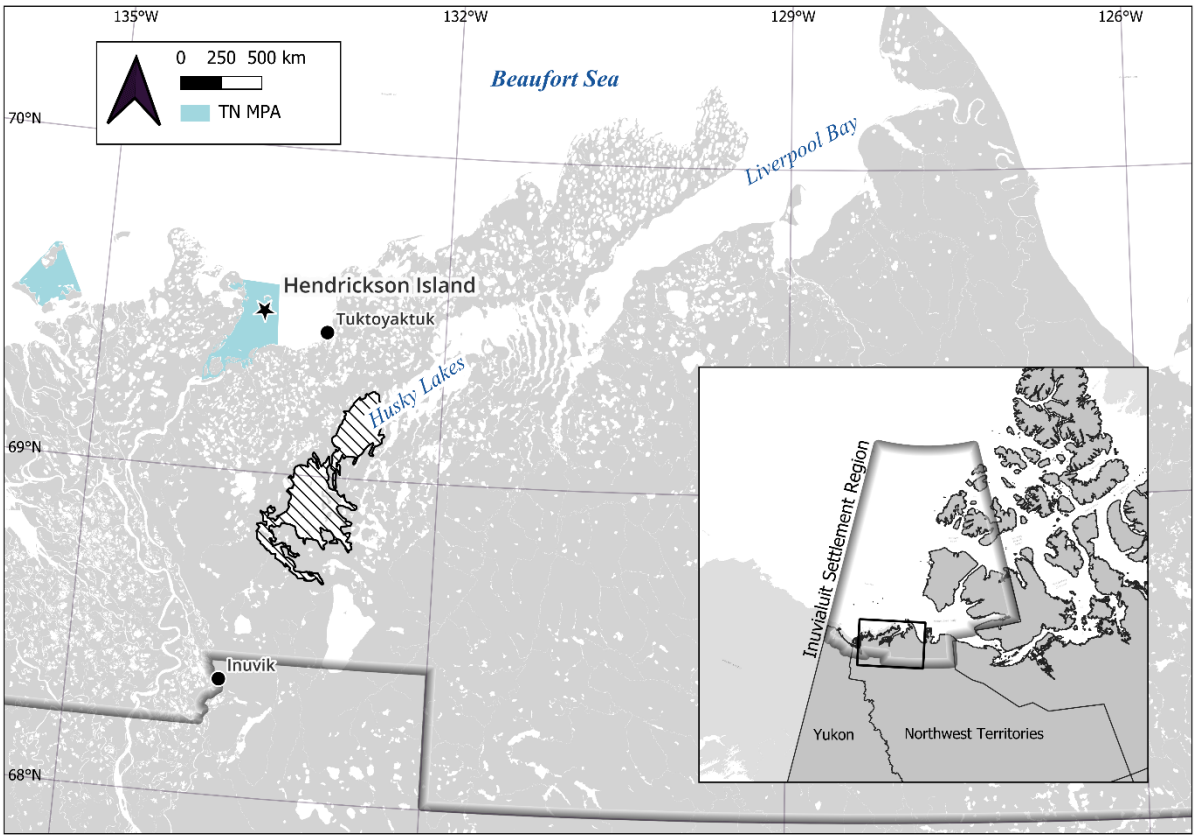


Figure 1. Beluga samples were collected during the annual beluga subsistence harvest between 1983 and 2021 on Hendrickson Island, Northwest Territories, Canada. Additional samples were collected in 1989 from an ice entrapment event at Husky Lakes (hatched area).

CONTAMINANT ANALYSIS

Tissue extraction and PCB quantification of full depth blubber samples were conducted at the DFO Freshwater Institute in Winnipeg, Manitoba, for samples collected between 1983- 2007 and 2011. Samples collected between 2008 and 2010 were analyzed at the DFO Laboratory of Expertise in Aquatic Chemical Analyses (LEACA), in Sidney, British Columbia (BC). Samples collected in 2012–2018, 2020 and 2024 were analyzed at SGS AXYS, Sidney, BC. Finally, samples collected in 2019, 2021 and 2023 were analyzed at the Environment Canada and Climate Change Laboratory (QLET: Québec Laboratory for Environmental Testing) in Montréal, Québec (QC) (Table 1).

All laboratories followed comparable, well-established, high-quality protocols for PCB extraction and quantification, although the specific methods varied among facilities. Extractions started with a subsample of blubber of approximately 2g.

Blubber samples analyzed at the DFO Freshwater Institute followed procedures described elsewhere (Muir et al., 1988; 1990; Stern et al., 2005). In brief, partially thawed blubber were combined with anhydrous Na_2SO_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 with the

fraction eluted with hexane used for PCB quantification. Each fraction was analyzed by high-resolution gas chromatography (GC) with ^{63}Ni electron capture detection, H_2 as the carrier gas and N_2 as the make-up gas. Details on quality assurance/quality control (QA/QC) can be found elsewhere (Stern et al. 2005).

Similarly, samples analyzed at the DFO-LEACA lab were ground with anhydrous sodium sulfate, packed onto a glass extraction column and extracted with hexane:dichloromethane (1:1, v/v). Following sample clean-up and separation on Florisil, contaminants were quantified by high-resolution gas chromatography–high resolution mass spectrometry (HRGC/HRMS). Details on the analytical conditions, criteria for identification and quantitation, and QA/QC are described in Ikonomou et al. (2001).

SGS AXYS Analytical Ltd. used the USEPA method 1668C EPA (EPA, 2010). Briefly, samples were extracted by a Soxhlet extraction using dichloromethane. Extracts were cleaned by gel permeation chromatography (GPC) and alumina column chromatography. Procedural blanks and extracts were spiked with ^{13}C-labeled surrogate standards (n = 29) and analyzed with isotope-dilution HRGC/HRMS.

Finally, samples analyzed at QLET were extracted using Soxhlet extraction with acetone:hexane ((1:1) v/v) and then run on GPC. After lipid removal, PCB extracts were passed on a two-stage cleanup: a multilayer column cleanup consisting of layers of $AgNO_3$-silica-NaOH-silica-H_2SO_4-silica followed by an alumina column. PCBs were then analyzed for each sample using a HRGC/HRMS.

DATA ANALYSIS

PCB levels in marine mammals are known to accumulate with age in males. In contrast, in females, PCB concentrations tend to decline once they reach sexual maturity and start offloading their contaminants to their offspring during pregnancy and lactation (Borrell et al., 1996; Ross et al., 2000). Therefore, sex and age are important factors to take into account when investigation PCB levels in beluga whales from a temporal trend aspect. Few females were collected as hunters generally select medium to larger sized males. For these reasons, and to avoid additional confounding factors that come with looking at PCB concentrations in females, we focused the current data set on male beluga whales only, with a length over 380 cm (n = 255; Table 1). Total PCB concentration outliers were determined for the whole dataset using the interquartile range method and removed (n = 4) from the data set.

Table 1: 255 adult male beluga whales sampled between 1983 and 2024 on Hendrickson Island (and Husky Lakes for 1989) were analyzed for PCB concentrations at four different laboratories over the 41 year period (DFO-FWI: Fisheries and Oceans Canada - Freshwater Institute in Winnipeg, MB; DFO-LEACA: Fisheries and Oceans – Laboratory of Expertise in Aquatic Chemical Analyses in Sidney, BC; SGS AXYS in Sidney, BC; QLET: Quebec Laboratory of Environmental Testing in Montréal, QC)

Year	N	Age (Years) Mean ± standard error (se)	Length (cm) Mean ± se	Lipid (%) Mean ± se	Lab
1983	6	23.0 ± 3.0	na	69.7 ± 3.5	DFO-FWI (ortho congeners only)
1987	3	na	na	84.9 ± 2.4	DFO-FWI (ortho congeners only)
1989	12	29.0 ± 4.3	na	90.5 ± 1.1	DFO-FWI
1993	5	28.8 ± 2.4	429.8 ± 11.5	86.0 ± 0.8	DFO-FWI
1994	8	31.3 ± 3.9	424.8 ± 8.1	94.4 ± 0.9	DFO-FWI
1995	13	32.7 ± 3.2	430.6 ± 5.8	81.7 ± 0.9	DFO-FWI

Year	N	Age (Years) Mean ± standard error (se)	Length (cm) Mean ± se	Lipid (%) Mean ± se	Lab
1996	8	27.0 ± 2.7	415.9 ± 8.4	92.5 ± 1.8	DFO-FWI
2001	14	32.8 ± 3.0	412.1 ± 5.5	82.5 ± 0.8	DFO-FWI
2002	8	31.3 ± 2.9	423.2 ± 5.6	91.0 ± 0.8	DFO-FWI
2003	8	28.8 ± 3.1	418.9 ± 7.9	90.9 ± 1.4	DFO-FWI
2004	9	25.6 ± 2.1	423.9 ± 6.3	84.9 ± 2.4	DFO-FWI
2005	10	26.8 ± 3.0	415.8 ± 8.3	92.4 ± 1.1	DFO-FWI
2006	11	24.8 ± 2.2	411.5 ± 5.5	89.5 ± 1.9	DFO-FWI
2007	9	25.3 ± 2.3	429.3 ± 8.9	86.1 ± 2.4	DFO-FWI
2008	17	32.1 ± 2.2	427.0 ± 5.7	88.8 ± 1.8	DFO-LEACA
2009	12	29.3 ± 2.4	415.8 ± 5.3	89.0 ± 0.7	DFO-LEACA
2010	8	24.8 ± 1.5	419.4 ± 14.1	89.4 ± 1.4	DFO-LEACA
2011	9	26.4 ± 2.0	412.2 ± 5.8	92.4 ± 1.3	DFO-FWI
2012	8	27.3 ± 1.6	416.6 ± 5.8	66.1 ± 4.1	SGS AXYS
2013	5	34.0 ± 2.4	423.6 ± 6.0	77.8 ± 8.2	SGS AXYS
2014	6	28.3 ± 2.6	406.0 ± 6.0	94.1 ± 2.1	SGS AXYS
2015	5	23.4 ± 1.9	397.4 ± 5.5	91.6 ± 2.1	SGS AXYS
2017	9	28.9 ± 6.7	421.9 ± 4.7	97.6 ± 1.2	SGS AXYS
2018	9	38.4 ± 4.1	435.7 ± 4.5	90.5 ± 3.6	SGS AXYS
2019	10	37.0 ± 4.3	428.6 ± 4.4	89.3 ± 1.0	QLET
2020	11	27.8 ± 3.5	415.7 ± 7.0	93.0 ± 1.0	SGS AXYS
2021	7	30.1 ± 3.9	422.7 ± 8.3	na	QLET
2023	5	29.8 ± 7.1	413.0 ± 5.1	85.4 ± 8.4	QLET
2024	7	34.3 ± 11.3	419.4 ± 8.0	86.4 ± 6.4	SGS AXYS

Despite the differences in sampling and analytical procedures among the participating laboratories, all laboratories follow well-documented QA/QC protocols to ensure that the results are of high quality and comparable. In addition, the beluga project participates in the NCP/AMAP inter-laboratory study (ILS) program, which assesses the quality of results produced for a wide range of organic and metallic contaminants in a number of matrices, including blubber. The ILS was established in 2005 to support quality assurance and quality control within the NCP and was subsequently extended to laboratories participating in AMAP activities. The ILS is administered by the Information and Quality Management Group (IQM) of Environment and Climate Change Canada (ECCC). The NCP/AMAP ILS program received accreditation in 2026.

A different number of congeners was quantified by each lab (Appendix 1). In order to combine datasets, we determined the dominant congener in each co-eluting combination based on Frame et al., 1996 and aligned the datasets (Appendix 1).

All contaminant data were blank-corrected and levels were reported on a lipid weight (lw) basis. Since not all congeners/compounds were detectable in all samples, substitutions had to be applied. For the calculations of totals, individual congener/compound concentrations below the detection limit were treated as zero (Vorkamp et al., 2011). Total PCB concentrations were calculated based on the 79 congeners (104 including co-eluting congeners) quantified by the

DFO-FWI laboratories (Appendix 1). Since the QLET lab quantified 41 congeners (analyses in 2019, 2021 and 2023), we evaluated the percent contribution of these 41 congeners to ΣPCBs in the years adjacent to the years where analyses were conducted in that lab and adjusted the total PCB concentrations accordingly (+8.9%).

All statistical analyses were performed in R (R Core Team, 2020). Normality and homogeneity of variances were tested for biological variables (age, total length) and total PCB data 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. Analysis of variance were used to evaluate differences amongst years. Total PCB results were presented as mean ± standard error.

Temporal trends in total PCB concentrations in blubber were assessed using a method used by AMAP (AMAP, 2020) and recently by Morris et al. (2022). Briefly, temporal trends of total PCBs were analyzed based on log-transformed data using individual concentration data rather than annual medians. Linear and non-linear models were explored as described in Morris et al. (2022), and best-fit models were selected using the Akaike Information Criterion corrected for small sample size (AIC_c) (Burnham and Anderson, 2002). If the difference in AIC between any candidate model and the top-ranked model was less than 2, the models were deemed statistically indistinguishable. In such scenarios, we applied the principle of parsimony and selected the simpler linear model for subsequent analysis.

Annual percent (%) change in concentration over the entire study period (1989–2024) was calculated. For significant linear trends, the slope of the log-linear trend was used to calculate the annual change as follows, based on Morris et al. (2022):

$$\text{Equation 1 : Trend (\%)} = 100\,(\exp(\beta) - 1)$$

where β was slope of the log-linear trend. The exponentiation back-transforms the annual absolute change in log concentration, measured by β, to the annual percentage change in concentration.

For significant non-linear trends, the change in total PCB concentrations between 1983 and 2024 was converted to the equivalent annual percentage change had the trend been linear as follows, based on Morris et al. (2022):

$$\text{Equation 2: Trend (\%)} = 100\,(\exp\,(s(2024) - s(1983) / 2024 - 1983) - 1)$$

Where s(2024) and s(1983) were the fitted log concentrations in 2024 and 1983, respectively.

RESULTS AND DISCUSSION

A total of 250 blubber samples from adult male beluga whales collected between 1983 and 2024 were analyzed for PCBs. In order to limit confounding factors in the temporal trend analysis, we focused on adult males with a length greater than 380 cm.

TOTAL PCB TEMPORAL TREND

For total PCB concentrations (log transformed), both linear and non-linear models were significant with similar AICs (Table 2). It was decided to select the linear model ($R^2 = 0.15$; $p < 0.05$) which showed an annual decrease of 1.7% per year. This represents a total decline of 51% over the 41-year period with total PCB levels averaging 5664 ng/g lw in 1983 and 2780 ng/g lw in 2024. This is different from a previous trend study conducted in this population between 1989 and 2015 that reported no trend (Noël et al., 2018). However, the Noël et al., (2018) study only used the sum of ten congeners as opposed to the 79 congeners used in the

present study and started in 1989 as opposed to 1983. The annual decrease reported here was in the same range of what has been reported for ringed seals (*Pusa hispida*) from the Western Arctic: -1.2% in Sachs Harbour for 1988-2016; -4.1% in Ulukhaktok for 1981-2015 (Houde et al., 2019) and 2.2% in Ulukhaktok for 1972-2010 (Addison et al., 2014).

Overall, across the Arctic, declines in PCBs have been observed in accordance with the regulations established in the 1970's. In their review on temporal trends of POPs in the Arctic environment, Riget et al. (2019) evaluated a total of 347 long-term time series in various biota (blue mussels, freshwater fish, marine fish, seabirds and marine mammals) across the Arctic for the sum of 10 PCB congeners. They reported that 38% of biota showed a significant log-linear decreasing trend and 13% showed a significant decreasing trend together with a non-linear trend component with an overall mean annual decrease across all significant trends of 3.7%.

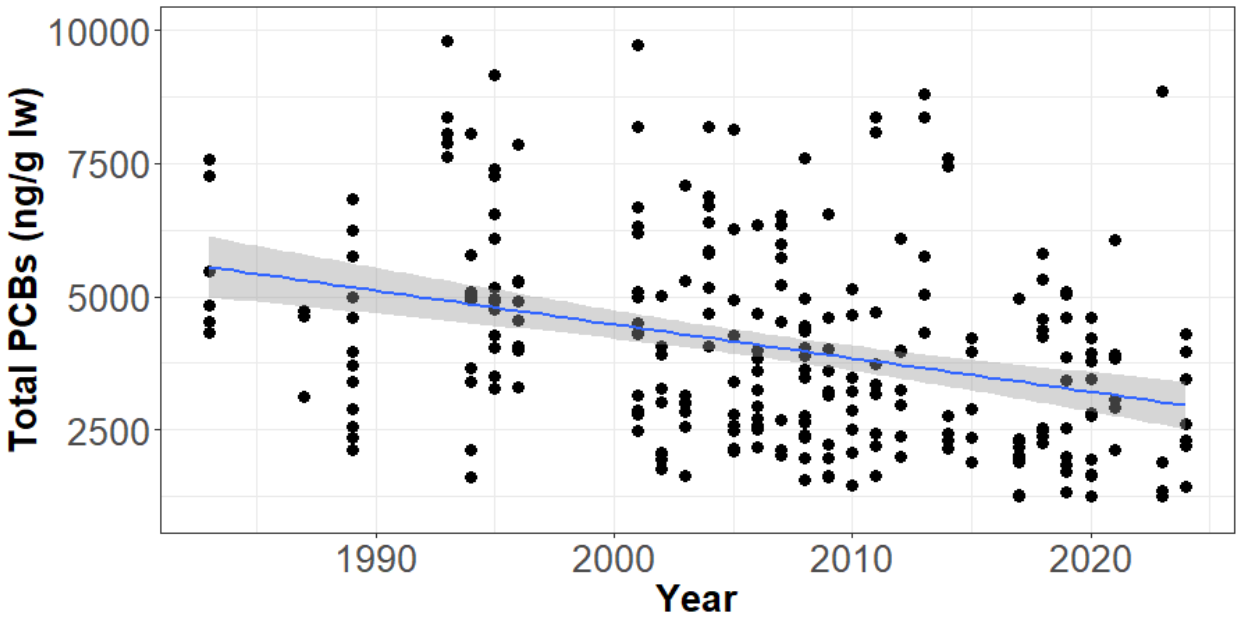


Figure 2: Linear decrease (R^2 = 0.15; $p < 0.05$) in total PCB concentrations (ng/g lipid weight) in 250 adult male beluga whales sampled in the eastern Beaufort Sea between 1983 and 2024. (Presented as non-log-transformed for visualization of actual concentrations).

TEMPORAL TRENDS OF DOMINANT PCB CONGENERS

CBs-153 and -138 were consistently in the top 2 congeners each year accounting for 13.2 ± 0.60 and 11.5 ± 0.47% of ΣPCBs on average over the entire study period, respectively (Appendix 2). These two congeners are resistant to metabolism in cetacean species and typically dominate PCB patterns (Ross et al., 2000; Desforges et al., 2013). Both of these congeners had significant non-linear trends (Table 2) with an annual decrease of 0.22% and 0.61%, respectively.

In their study, Houde et al. (2019) reported a non-significant increase of 0.7% for CB-153 in ringed seals harvested from Sachs Harbour, NT, (1988-2016) and a significant decrease of -2% in ringed seals harvested from Ulukhaktok, (1981-2015) both located in the Beaufort Sea. A study across the Arctic for CB-153 in various biota (blue mussels, freshwater fish, marine fish, seabirds and marine mammals that 34% of biota showed a significant log-linear decreasing

trend for long-term time series starting before the year 2000. In addition, another 15% of the trends in that study showed a significant decreasing trend together with a non-linear trend component. The overall mean annual decrease across all significant trends was 3.8% which is more than the decline reported here (Riget et al. 2019). In addition, in that review, the % annual decrease for ΣPCBs and CB-153 was similar which was different for the observations made here where the annual decrease of CB-153 was an order of magnitude lower than that of ΣPCBs suggesting a change in PCB pattern over time (see below).

The other congeners consistently present in the top 6 dominant congeners were CBs-101, -99, -52, -149, -118 accounting for 6.9 ± 0.3, 6.3 ± 0.2, 5.9 ± 0.3, 5.6 ± 0.2, 5.3 ± 0.2 % of ΣPCBs on average over the entire study period, respectively (Appendix 2). These congeners had non-linear trends (except CB-101) between 1983 and 2024 with percent annual change ranging from -2.3% for CB-99 to +0.3% for CB-149 (Table 2). The range in percent annual changes for these dominant congeners further suggest changes in PCB patterns over time (Table 2 and Figure 3). The percent annual changes reported here were in the lower range of what was observed in ringed seals from Ulukhaktok where all individual congeners showed a decline ranging from <1.5% to 4% per year between 1972 and 2010 (Addison et al., 2014). More recently, Houde et al. (2019) reported an annual decline for CB-99 of 0.4% and 1.7% for ringed seals from Sachs Harbour (1988-2016) and Ulukhaktok (1981-2015), respectively.

Table 2: Results from linear and non-linear models for total PCB concentrations and the concentrations of the dominant congeners in blubber of beluga. Models were selected based on the lowest AIC. When AICs were similar between the two models, the linear model was selected. Percent annual changes were calculated based on Equation 1[a] and Equation 2[b].

Compound	Model	p-value	R^2	AIC	% annual change
Total PCBs	**Linear**	**<0.05**	**0.15**	**308.1**	**-1.7%[a]**
	Non linear	<0.05	0.14	308.0	
CB-153	Linear	<0.05	0.1	385.4	
	Non linear	**<0.05**	**0.1**	**383.2**	**-0.22%[b]**
CB-138	Linear	<0.05	0.1	394.3	
	Non linear	**<0.05**	**0.1**	**388.6**	**-0.61% [b]**
CB-101	**Linear**	**<0.05**	**0.1**	**415.4**	**-1% [a]**
	Non linear	<0.05	0.1	415.3	
CB-99	Linear	0.559		391	
	Non linear	**<0.05**	**0.1**	**369.6**	**-2.3% [b]**
CB-52	Linear	<0.05	0.12	310.8	
	Non linear	**<0.05**	**0.18**	**299**	**-2.0% [b]**
CB-149	Linear	<0.05	0.1	408.6	
	Non linear	**<0.05**	**0.22**	**368.5**	**0.3% [b]**
CB-118	Linear	<0.05	0.1	330.8	
	Non linear	**<0.05**	**0.1**	**328.1**	**-1.0% [b]**

Evaluating the contribution of those dominant congeners to ΣPCBs is another way to visualize PCB patterns over time and further demonstrate the pattern change over the study period (1983–2024; Figure 3). The contribution CBs -153, -138, -101, -99 and -118 to ΣPCBs had significant linear increase while CB-52 did not exhibit a significant trend and CB-149 showed a significant non-linear trend over time (Figure 3). The contributions of CBs-153 and -138 to

ΣPCBs were indeed significantly different ($p < 0.05$) between 1984 (8.1 ± 0.24 and 7.6 ± 0.26%, respectively) and 2024 (15.3 ± 0.55 and 12.1 ± 0.51, respectively; Appendix 2). An earlier study on PCB temporal trends in EBS beluga whales spanning 1989-2015 also showed temporal changes in patterns and suggested a change in diet may have contributed to this change in PCB profile over time (Noël et al., 2018). Moreover, given the impacts of climate change on the transport and fate of contaminants in the Arctic environment (Vorkamp et al., 2022, Houde et al., 2019), altered PCB transport and uptake in the EBS beluga food web could also impact the shift in PCB patterns over time.

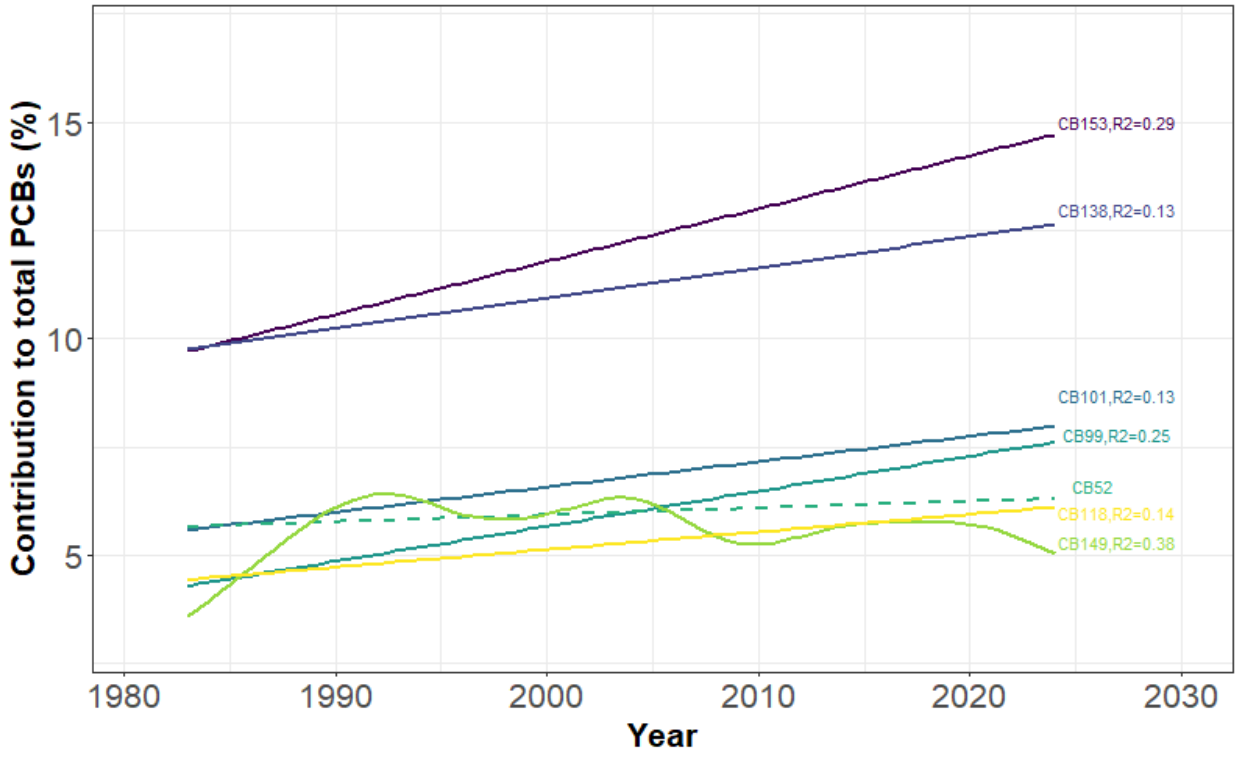


Figure 3: Temporal trends of the contribution of the dominant congeners to ΣPCBs. CBs -153, -138, -101, -99 and -118 had significant linear increase while CB-52 did not exhibit a significant trend and CB-149 showed a significant non-linear trend over time.

HEALTH RISK TO BELUGAS

In marine mammals, adverse health effects in relation to POP exposure have been reported at all levels of biological organization; from the molecular level to the population level with epizootic outbreaks of morbillivirus killing thousands of immuno-compromised European harbor and grey seals (*Halichoerus grypus*) (Dietz et al., 1989) and high rate of cancer among the St. Lawrence beluga population (DeGuise et al., 1995; Martineau et al., 1994). The immune, reproductive, and endocrine systems are all targets of various POPs (Lie et al., 2004; Mos et al., 2007, Tabuchi et al., 2006, Buckman et al., 2011, DeSwart et al., 1996). While marine mammals are exposed to a complex cocktail of contaminants, the weight of evidence from field, semi-field, and *in vitro* studies has shown that PCBs remain the number one contaminant of concern (Mos et al., 2010). Mos et al. (2010) conducted a risk-based evaluation of 13 POPs in harbor seals from the Salish Sea, British Columbia, and concluded that PCBs remain the number one threat. Similarly, Desforges et al. (2018) suggested that, despite their ban, PCBs still threaten the long-term viability of more than half of the populations of killer whales in the world with those living close to industrialized region being at high risk of population collapse.

For marine mammals, three PCB toxicity reference values (TRVs) have been derived and are widely used as benchmarks for conservation and/or risk management (De Swart et al., 1996; Ross et al., 1996; Hall et al., 2006; Mos et al., 2010). The least conservative TRV is 17 mg/kw lw derived from a study using captive harbor seal fed contaminated herring from the Baltic Sea which resulted in immunotoxicity including reduced natural killer cell function, T-cell function and *in vivo* responses to immunization (DeSwart et al., 1996; Ross et al., 1996). In their 2006 study, Hall et al. (2006) used an individual-based model framework to quantify the effects of PCBs on bottlenose dolphin population growth rate and predicted calf mortality to be 50% at a median effective PCB concentration (EC_{50}) of 10 mg/kg lw. Finally, the most conservative TRV of 1.3 mg/kg lw represents the 5% tissue residue concentration in harbor seals which exhibited dose-related alterations in endocrine and immune functions as well as gene expression (Mos et al., 2010).

For beluga specifically, a toxicity reference value of 1.6 mg/kg lw was proposed and associated with disruption of vitamin A and E in the liver and blubber (Desforges et al., 2013). More recently, Simond et al., (2025) identified a lowest EC_5 of 1.5 mg/kg at which adverse molecular effects may be expected. This value was similar to the most conservative TRV derived by Mos et al. (2010).

In this report, we evaluated total PCB levels measured in EBS beluga whales against the two beluga-specific thresholds (1.6 and 1.5 mg/kg). We looked at the percentage of samples, each year, that exceeded these thresholds. However, as they yielded the same results, we only present results using the 1.6 mg/kg threshold (Figure 4).

All whales sampled between 1983 and 2007 had total PCB levels over the 1.6 mg/kg health effect threshold for PCBs in beluga whales (Figure 4). From 2008 onward, there were more interannual variations with percentage of samples exceeding the threshold ranging from 40% to 100%. Since 2017, the majority of years had less than 100% of samples that surpassed the thresholds and the average across those years was 84% (Figure 4). While this only provides a snapshot given the limited number of samples analyzed each year, it provides high level information on the health of EBS beluga whales as it relates to PCB exposure.

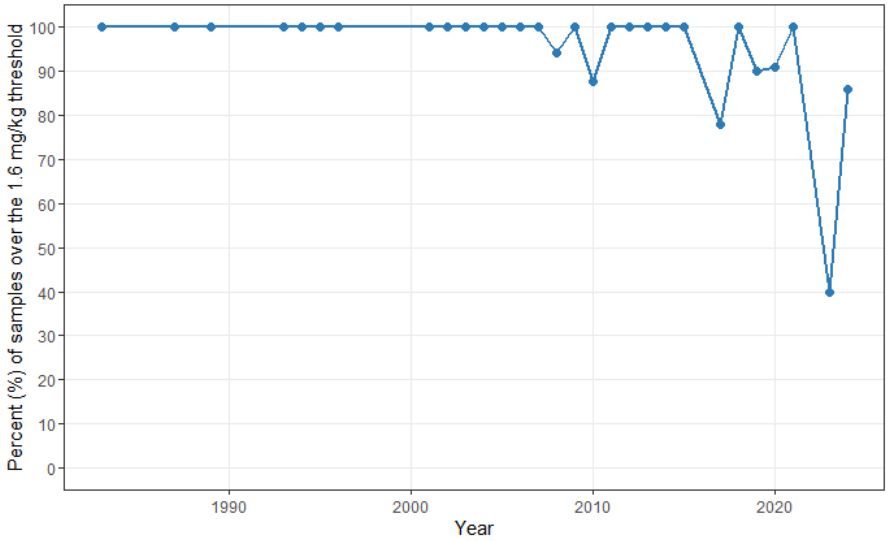


Figure 4: Percentage of samples over beluga-specific threshold for PCB-related health effects (1.6 mg/kg (Desforges et al., 2013)).

CONCLUSION

PCB concentrations in blubber of adult male beluga whales from the EBS population sampled between 1983 and 2024 declined by 51%, reflecting the impact of regulations put into place in the 1970s. Evaluation of the dominant congeners revealed different temporal trends and annual percent declines suggesting PCB patterns changing over time.

While regulations, and therefore changes in primary emissions, have driven PCB trends in Arctic biota in the past, recent studies have shown that climate change is also in play, affecting contaminant transport, fate and uptake into the Arctic food web (Vorkamp et al., 2022). For example, Houde et al. (2019) reported that atmospheric patterns and sea-ice coverage are influencing POP accumulation in ringed seals from the Beaufort Sea.

Previous studies have shown that climate-induced decreases in Arctic sea ice cover, changes in ocean temperature profiles, and food web productivity have influenced beluga distribution (range expansion) and feeding ecology, which would consequently change the accumulation of contaminants in belugas (Loseto et al., 2015; Desforges et al., 2021). In addition, climate-related changes may affect the condition and health of beluga whales by reducing the abundance of their preferred prey, Arctic cod (*Boreogadus saida*) (Loseto et al., 2018), impacting nutrition, and/or increasing energetic expenditures to search for prey potentially leading to additional stress and decrease ability to cope with contaminant-associated health effects.

In an ever-changing Arctic environment, continued monitoring of legacy compounds, including PCBs, is needed to evaluate the health of marine mammals such as beluga whales given that they remain prominent contaminants in arctic wildlife. In addition, complementary information on long-term trends of dietary biomarkers and/or trends in PCB levels in EBS beluga prey are needed in order to accurately interpret and understand the drivers of emergent temporal trends of PCBs measured in beluga presented here.

CREDIT AUTHORSHIP CONTRIBUTION STATEMENT

Marie Noel: Writing – original draft, Formal analysis, Visualization; Gary Stern: Conceptualization, Methodology, Writing – review & editing; Magali Houde: Methodology, Writing – review & editing; Dana Neumann: Methodology, Writing – review & editing; Ashley Elliot: Methodology, Writing – review & editing; Shannon MacPhee: Methodology, Writing – review & editing; Derek Muir: Conceptualization, Methodology, Writing – review & editing; Gerald Inglangasuk: Editing; Tuktoyaktuk Hunters and Trappers Committee: Editing; Lisa Loseto: Conceptualization, Funding acquisition, Visualization, Writing – review & editing.

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Appendix 1. Congeners quantified by each laboratory. The dominant congener from analytical co-elutions was determined from Frame et al., 1996 and used to align the PCB datasets from the different laboratories.

Congeners	Dominant co-eluting congener*	QLET	DFO-FWI	DFO-FWI (only ortho-congeners)	SGS AXYS/DFO-LEACA
PCB_1			Y		Y
PCB_2					Y
PCB_3			Y		Y
PCB_4	4		Y		Y
PCB_5_8			Y		Y
PCB_6			Y		Y
PCB_7_9	9		Y		Y
PCB_10					Y
PCB_11					Y
PCB_12_13	13				Y
PCB_14					Y
PCB_15					Y
PCB_16_32	16		Y	Y	Y
PCB_17		Y	Y		Y
PCB_18_30	18	Y	Y	Y	Y
PCB_19			Y		Y
PCB_20_28	28	Y	Y	Y	Y
PCB_21_33	33	Y	Y		Y
PCB_22			Y		Y
PCB_23_34	34				Y
PCB_24					Y
PCB_25			Y		Y
PCB_26_29	26		Y		Y
PCB_27	27		Y		Y
PCB_31		Y	Y		Y
PCB_35					Y
PCB_36					Y
PCB_37					Y
PCB_38					Y
PCB_39					Y
PCB_40				Y	
PCB_40_41_64_71	64		Y		Y
PCB_42_68	42		Y	Y	Y
PCB_43_49_69	49	Y	Y	Y	Y
PCB_44_47_48_65	44	Y	Y	Y	Y
PCB_45_51	45		Y		Y

Congeners	Dominant co-eluting congener*	QLET	DFO-FWI	DFO-FWI (only ortho-congeners)	SGS AXYS/DFO-LEACA
PCB_46			Y		Y
PCB_50_53	53				Y
PCB_52		Y	Y	Y	Y
PCB_54					Y
PCB_55					Y
PCB_56	56		Y		Y
PCB_57					Y
PCB_58					Y
PCB_59_62_75	59				Y
PCB_60				Y	Y
PCB_61_70_74_76	70	Y	Y	Y	Y
PCB_63					Y
PCB_66			Y	Y	Y
PCB_67					Y
PCB_71				Y	
PCB_72					Y
PCB_73					Y
PCB_77					Y
PCB_78					Y
PCB_79					Y
PCB_80					Y
PCB_81					Y
PCB_82		Y	Y		Y
PCB_83_99	99	Y	Y	Y	Y
PCB_84	84		Y		Y
PCB_85_111_116_117	85		Y	Y	Y
PCB_86_87_97_109_119_125	87	Y	Y	Y	Y
PCB_88_91	91		Y	Y	Y
PCB_89					Y
PCB_90_101_113	101	Y	Y	Y	Y
PCB_92					Y
PCB_93_95_98_100_102	95	Y	Y		Y
PCB_94					Y
PCB_96					Y
PCB_103					Y
PCB_104					Y
PCB_105		Y	Y	Y	Y
PCB_106					Y

Congeners	Dominant co-eluting congener*	QLET	DFO-FWI	DFO-FWI (only ortho-congeners)	SGS AXYS/DFO-LEACA
PCB_107_108_124	124			Y	Y
PCB_110_115	110	Y	Y	Y	Y
PCB_112					Y
PCB_114			Y	Y	Y
PCB_118		Y	Y	Y	Y
PCB_120			Y		Y
PCB_121					Y
PCB_122					Y
PCB_123			Y		Y
PCB_126			Y		Y
PCB_127					Y
PCB_128_166	128	Y	Y	Y	Y
PCB_129_138_160_163	138	Y	Y	Y	Y
PCB_130					Y
PCB_131			Y		Y
PCB_132		Y	Y		Y
PCB_133					Y
PCB_134_143	134		Y		Y
PCB_135_151_154	151	Y		Y	Y
PCB_136				Y	Y
PCB_137			Y	Y	Y
PCB_139_140	139				Y
PCB_141			Y	Y	Y
PCB_142					Y
PCB_144				Y	Y
PCB_145					Y
PCB_146			Y	Y	Y
PCB_147_149	149	Y	Y	Y	Y
PCB_148					Y
PCB_150					Y
PCB_152					Y
PCB_153_168	153	Y	Y	Y	Y
PCB_155					Y
PCB_156_157	156	Y	Y	Y	Y
PCB_158		Y	Y	Y	Y
PCB_159					Y
PCB_161					Y
PCB_162					Y

Congeners	Dominant co-eluting congener*	QLET	DFO-FWI	DFO-FWI (only ortho-congeners)	SGS AXYS/DFO-LEACA
PCB_164					Y
PCB_165	176		Y		Y
PCB_167			Y	Y	Y
PCB_169		Y			Y
PCB_170_190	170	Y	Y	Y	Y
PCB_171_173	171	Y	Y	Y	Y
PCB_172	172		Y	Y	Y
PCB_174			Y	Y	Y
PCB_175			Y		Y
PCB_176					Y
PCB_177		Y	Y	Y	Y
PCB_178	178		Y	Y	Y
PCB_179					Y
PCB_180_193	180	Y	Y	Y	Y
PCB_181					Y
PCB_182					Y
PCB_183_185	183	Y	Y	Y	Y
PCB_184					Y
PCB_186					Y
PCB_187		Y	Y	Y	Y
PCB_188					Y
PCB_189			Y		Y
PCB_191		Y	Y	Y	Y
PCB_192					Y
PCB_194		Y	Y	Y	Y
PCB_195		Y	Y	Y	Y
PCB_196				Y	Y
PCB_197_200	200		Y	Y	Y
PCB_198_199	199	Y	Y	Y	Y
PCB_201	201		Y	Y	Y
PCB_202					Y
PCB_203			Y		Y
PCB_204					Y
PCB_205		Y	Y		Y
PCB_206		Y	Y	Y	Y
PCB_207			Y		Y
PCB_208		Y	Y		Y
PCB_209		Y	Y	Y	Y

*Fame et al., 1996

Appendix 2. Top 6 PCB congeners (average ± standard error) for each year. Italics show their contribution (%) to ΣPCBs.

1983	CB-153	453.4 ± 40.4 8.1 ± 0.24 %	CB-138	427.0 ± 43.9 *7.55 ± 0.26 %*	CB-99	412.7 ± 43.5 *7.27 ± 0.15 %*	CB-101	353.2 ± 37.5 *6.22 ± 0.17 %*	CB-52	318.4 ± 33.5 *5.62 ± 0.15 %*	CB-66	247.8 ± 29.7 *3.89 ± 0.16 %*
1987	CB-153	408.4 ± 68.4 9.2 ± 0.57 %	CB-138	318.8 ± 46.7 *7.63 ± 0.47 %*	CB-99	311.5 ± 36.5 *7.50 ± 0.32 %*	CB-101	282.2 ± 34.4 *6.78 ± 0.22 %*	CB-52	226.7 ± 28.3 *5.45 ± 0.24 %*	CB-118	163.6 ± 21.0 *3.94 ± 0.25 %*
1989	CB-153	447.2 ± 48.4 10.3 ± 0.08 %	CB-138	403.7 ± 48.4 *9.77 ± 0.18 %*	CB-149	245.2 ± 25.1 *6.01 ± 0.06 %*	CB-101	225.8 ± 22.0 *5.58 ± 0.10 %*	CB-187	214.9 ± 22.1 *5.13 ± 0.43 %*	CB-52	207.4 ± 28.1 *4.93 ± 0.15 %*
1993	CB-153	674.1 ± 60.1 *8.4 ± 0.34 %*	CB-138	592.9 ± 54.3 *7.05 ± 0.34 %*	CB-149	567.6 ± 39.1 *6.77 ± 0.16 %*	CB-101	486.2 ± 17.9 *5.88 ± 0.38 %*	CB-99	450.7 ± 77.2 *5.57 ± 1.02 %*	CB-52	429.3 ± 20.0 *5.15 ± 0.19 %*
1994	CB-153	726.2 ± 156.2 *15.5 ± 1.21 %*	CB-138	652.2 ± 124.0 *14.2 ± 0.71 %*	CB-149	264.9 ± 39.9 *5.82 ± 0.11 %*	CB-101	255.4 ± 27.7 *6.11 ± 0.35 %*	CB-52	251.0 ± 25.7 *6.06 ± 0.44 %*	CB-95	220.9 ± 11.2 *5.43 ± 0.43 %*
1995	CB-153	618.3 ± 66.1 *11.6 ± 0.27 %*	CB-138	542.5 ± 59.3 *9.65 ± 0.28 %*	CB-149	337.8 ± 36.2 *6.22 ± 0.46 %*	CB-101	328.8 ± 32.9 *5.98 ± 0.33 %*	CB-99	303.8 ± 31.2 *5.48 ± 0.19 %*	CB-52	297.6 ± 22.3 *5.49 ± 0.12 %*
1996	CB-153	740.2 ± 99.1 *15.7 ± 0.57 %*	CB-138	731.3 ± 88.1 *15.1 ± 0.35 %*	CB-118	296.0 ± 29.0 *6.31 ± 0.14 %*	CB-149	279.4 ± 25.0 *5.96 ± 0.09 %*	CB-52	258.6 ± 15.8 *5.67 ± 0.20 %*	CB-101	258.5 ± 21.8 *5.52 ± 0.15 %*
2001	CB-153	567.7 ± 80.1 *10.8 ± 0.38 %*	CB-138	526.3 ± 79.2 *9.77 ± 0.39 %*	CB-101	301.3 ± 39.4 *5.78 ± 0.27 %*	CB-149	293.2 ± 29.8 *5.91 ± 0.40 %*	CB-52	282.5 ± 29.3 *5.66 ± 0.23 %*	CB-99	270.6 ± 29.4 *5.30 ± 0.13 %*
2002	CB-138	448.5 ± 63.5 *14.8 ± 0.57 %*	CB-153	444.8 ± 61.8 *14.9 ± 0.36 %*	CB-52	195.4 ± 18.1 *6.68 ± 0.34 %*	CB-118	181.4 ± 30.3 *6.93 ± 0.24 %*	CB-95	180.6 ± 16.1 *6.15 ± 0.29 %*	CB-149	178.9 ± 19.3 *6.07 ± 0.17 %*
2003	CB-153	549.8 ± 116.9 *14.10 ± 0.52 %*	CB-138	529.5 ± 124 *14.1 ± 0.69 %*	CB-52	226.9 ± 22.2 *6.87 ± 0.50 %*	CB-101	211.5 ± 29.3 *6.09 ± 0.27 %*	CB-149	211.0 ± 30.2 *6.08 ± 0.15 %*	CB-95	194.1 ± 19.2 *5.43 ± 0.43 %*
2004	CB-153	685.2 ± 50.6 *11.11 ± 0.09 %*	CB-138	642.1 ± 55.3 *10.7 ± 0.29 %*	CB-101	401.8 ± 30.1 *6.69 ± 0.14 %*	CB-149	375.9 ± 41.4 *6.31 ± 0.50 %*	CB-99	366.0 ± 22.6 *6.15 ± 0.08 %*	CB-52	337.8 ± 21.6 *5.67 ± 0.09 %*
2005	CB-138	517.3 ± 109.8 *12.4 ± 0.55 %*	CB-153	511.8 ± 100.0 *12.12 ± 0.39 %*	CB-149	305.9 ± 49.0 *7.88 ± 0.13 %*	CB-101	266.2 ± 50.6 *7.31 ± 0.13 %*	CB-118	255.9 ± 39.4 *7.02 ± 0.73 %*	CB-52	253.3 ± 28.9 *6.86 ± 0.29 %*
2006	CB-153	386.3 ± 42.3 *10.13 ± 0.30 %*	CB-138	365.4 ± 44.6 *10.2 ± 0.31 %*	CB-101	248.6 ± 25.6 *7.10 ± 0.28 %*	CB-52	232.3 ± 23.2 *6.70 ± 0.32 %*	CB-95	223.7 ± 19.2 *6.51 ± 0.29 %*	CB-110	218.6 ± 23.2 *6.42 ± 0.39 %*
2007	CB-138	563.0 ± 86.2 *11.9 ± 0.71 %*	CB-153	545.1 ± 80.8 *11.14 ± 0.56 %*	CB-101	289.0 ± 40.7 *6.34 ± 0.23 %*	CB-95	260.1 ± 18.2 *5.93 ± 0.24 %*	CB-149	233.3 ± 30.8 *5.10 ± 0.08 %*	CB-118	227.8 ± 32.7 *4.89 ± 0.18 %*

2008	CB-153	429.3 ± 56.6 *11.15 ± 0.36 %*	CB-138	424.4 ± 53.6 *11.7 ± 0.27 %*	CB-101	309.9 ± 26.5 *8.97 ± 0.24 %*	CB-99	255.0 ± 26.4 *7.21 ± 0.09 %*	CB-52	222.5 ± 19.9 *6.40 ± 0.16 %*	CB-118	208.1 ± 20.9 *5.97 ± 0.14 %*
2009	CB-153	453.7 ± 71.0 *13.16 ± 0.41 %*	CB-138	348.1 ± 55.9 *10.0 ± 0.37 %*	CB-101	277.5 ± 26.2 *8.66 ± 0.26 %*	CB-99	237.8 ± 30.4 *7.16 ± 0.09 %*	CB-118	215.0 ± 28.9 *6.42 ± 0.12 %*	CB-52	208.0 ± 20.4 *6.53 ± 0.27 %*
2010	CB-153	489.2 ± 87.1 *14.17 ± 0.72 %*	CB-138	333.9 ± 60.3 *10.0 ± 0.49 %*	CB-101	247.1 ± 26.9 *8.05 ± 0.35 %*	CB-52	217.8 ± 25.8 *7.05 ± 0.27 %*	CB-99	204.2 ± 30.7 *6.37 ± 0.13 %*	CB-118	185.3 ± 27.3 *5.79 ± 0.22 %*
2011	CB-153	524.8 ± 114.5 *12.18 ± 0.39 %*	CB-138	477.6 ± 107.3 *10.8 ± 0.44 %*	CB-99	345.9 ± 64.8 *8.34 ± 0.16 %*	CB-101	288.6 ± 50.1 *7.16 ± 0.25 %*	CB-52	243.0 ± 36.5 *6.22 ± 0.32 %*	CB-118	233.4 ± 45.6 *5.50 ± 0.17 %*
2012	CB-153	437.6 ± 81.5 *12.19 ± 0.47 %*	CB-138	372.2 ± 68.0 *10.9 ± 0.39 %*	CB-101	266.6 ±34.4 *8.10 ± 0.34 %*	CB-99	247.1 ± 36.7 *7.40 ± 0.21 %*	CB-149	193.1 ± 28.3 *5.81 ± 0.18 %*	CB-52	192.0 ± 24.0 *5.88 ± 0.27 %*
2013	CB-153	1162.1 ± 301.9 *17.20 ± 2.44 %*	CB-138	831.4 ± 118.0 *12.8 ± 0.41 %*	CB-99	502.1 ± 82.3 *7.70 ± 0.36 %*	CB-101	407.7 ± 36.8 *6.50 ± 0.40 %*	CB-149	392.0 ± 59.3 *6.06 ± 0.30 %*	CB-118	358.6 ± 62.3 *5.55 ± 0.45 %*
2014	CB-153	753.6 ± 289.8 *15.21 ± 2.11 %*	CB-138	666.3 ± 249.5 *14.0 ± 1.71 %*	CB-99	315.5 ± 88.9 *7.48 ± 0.17 %*	CB-101	311.6 ± 75.0 *7.78 ± 0.32 %*	CB-52	234.1 ± 54.6 *5.91 ± 0.30 %*	CB-149	195.1 ± 39.4 *5.10 ± 0.28 %*
2015	CB-153	504.1 ± 89.1 *16.22 ± 1.04 %*	CB-138	441.1 ± 78.3 *14.0 ± 0.85 %*	CB-99	258.2 ± 48.3 *8.23 ± 0.37 %*	CB-101	215.6 ± 33.3 *7.05 ± 0.39 %*	CB-52	188.3 ± 35.6 *6.04 ± 0.38 %*	CB-149	175.9 ± 25.3 *5.79 ± 0.30 %*
2017	CB-153	312.9 ± 60.1 *13.23 ± 0.33 %*	CB-138	273.4 ± 48.0 *12.0 ± 0.27 %*	CB-99	173.8 ± 31.6 *7.66 ± 0.12 %*	CB-101	161.7 ± 26.0 *7.27 ± 0.19 %*	CB-149	136.4 ± 20.4 *6.17 ± 0.14 %*	CB-52	134.5 ± 23.5 *5.97 ± 0.21 %*
2018	CB-153	557.4 ± 72.1 *14.24 ± 0.85 %*	CB-138	498.3 ± 66.2 *13.1 ± 0.43 %*	CB-99	280.9 ± 36.6 *7.37 ± 0.08 %*	CB-101	261.4 ± 34.6 *6.85 ± 0.26 %*	CB-118	236.3 ± 29.4 *6.27 ± 0.14 %*	CB-149	191.0 ± 25.5 *5.02 ± 0.28 %*
2019	CB-153	460.8 ± 70.2 *14.25 ± 0.32 %*	CB-138	397.5 ± 62.5 *12.4 ± 0.25 %*	CB-101	235.6 ± 39.0 *7.36 ± 0.20 %*	CB-99	215.0 ± 29.5 *6.94 ± 0.12 %*	CB-149	192.0 ± 28.3 *6.10 ± 0.11 %*	CB-52	186.2 ± 28.0 *5.91 ± 0.13 %*
2020	CB-153	426.0 ± 61.2 *14.26 ± 0.49 %*	CB-138	345.4 ± 46.5 *11.5 ± 0.28 %*	CB-99	217.4 ± 27.9 *7.37 ± 0.09 %*	CB-101	216.6 ± 23.9 *7.59 ± 0.23 %*	CB-118	176.9 ± 23.0 *6.00 ± 0.16 %*	CB-52	167.6 ± 18.9 *5.92 ± 0.26 %*
2021	CB-153	669.5 ± 104.2 *16.27 ± 0.51 %*	CB-138	531.6 ± 82.9 *13.1 ± 0.37 %*	CB-99	269.6 ± 37.9 *6.79 ± 0.21 %*	CB-118	250.9 ± 39.1 *6.22 ± 0.22 %*	CB-101	248.9 ± 31.6 *6.46 ± 0.60 %*	CB-52	232.5 ± 29.4 *6.03 ± 0.45 %*
2023	CB-153	183.9 ± 44.1 *13.28 ± 0.46 %*	CB-138	138.9 ± 32.1 *10.0 ± 0.30 %*	CB-101	135.3 ± 18.6 *10.2 ± 0.87 %*	CB-52	96.2 ± 17.5 *7.12 ± 0.21 %*	CB-99	93.6 ± 15.5 *6.98 ± 0.33 %*	CB-149	72.9 ± 16.1 *5.31 ± 0.09 %*
2024	CB-153	464.2 ± 76.0 *15.29 ± 0.55 %*	CB-138	362.4 ± 62.0 *12.1 ± 0.51 %*	CB-101	194.4 ± 20.6 *6.95 ± 0.37 %*	CB-99	186.2 ± 25.4 *6.44 ± 0.11 %*	CB-52	161.0 ± 17.6 *5.76 ± 0.32 %*	CB-118	160.6 ± 25.2 *5.44 ± 0.15 %*