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Marine Mammal Tissue Archival Project**

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ABSTRACT

Liver and blubber samples from 50 juvenile male northern fur seals (*Callorhinus ursinus*) were collected on St. Paul Island from four different seal rookeries between 1987 and 2007. Samples were analyzed for vitamins and for both legacy and current-use persistent organic pollutants (POPs). The resultant data set provides information on how concentrations of the two groups of POPs are changing with time in the northern fur seal population. Legacy POPs determined included polychlorinated biphenyl congeners (PCBs), dichlorodiphenyltrichloroethane (DDT) and metabolites, chlorobenzenes, toxaphene, chlordane-related compounds, and mirex. With the exception of mirex, all legacy compounds showed statistically significant declines with sampling year consistent with trends observed in marine mammals from other northern areas such as the Canadian Arctic. Overall, concentrations of legacy POPs are similar to levels seen in seal samples from other areas of the North Pacific Ocean and the Bering Sea and are considerably lower (factor of 10 or greater) than observed in marine mammals from temperate regions of the United States. The current-use brominated flame retardants, polybrominated diphenyl ether (PBDE) and hexabromocyclododecane (HBCD), showed exponential increases with sampling year although PBDE concentrations may be reaching a plateau. Likewise, perfluorinated alkyl acids (PFAAs) also show exponential increases with sampling year indicating continued release and incorporation into the North Pacific food web. Concentrations of retinol and α - and γ -tocopherol (vitamin E) in liver were not related to POP concentrations, year of sampling, or the rookery where the seal sample originated. POP concentrations do not appear to influence vitamin concentrations as observed in other wildlife studies. Further monitoring of northern fur seals is warranted because of increasing concentrations of current-use POPs and declining northern fur seal populations.

Introduction

The monitoring of marine mammals from northern regions for temporal and spatial changes in persistent organic pollutant (POP) concentrations has provided important information often driving regulatory action on chemical production and use. For instance, the detection of temporal increases in polybrominated diphenyl ethers (PBDEs) in ringed seals and the initial detection of perfluorooctane sulfonate (PFOS) in polar bears in Canada helped fuel the eventual phasing-out of these chemicals in North America (Ikonomou et al., 2002; Kannan et al., 2001). Recent monitoring of marine mammals from Arctic and sub-Arctic regions have identified new compounds, such as the flame retardant hexabromocyclododecane (HBCD), that are in current use and are being considered for phase-out (de Wit et al., 2010).

Detecting time trends of POPs in marine mammals requires samples collected at multiple time points, generally over several decades, and stored in proper conditions. To address this need, specimen banks were established in several countries including the US, Canada, Denmark, Scandinavia, and Japan to collect and archive marine mammal tissues in order to detect pollutant temporal trends (Becker et al., 2006). In the US, the National Institute of Standards and Technology (NIST) maintains the Marine Environmental Specimen Bank (Marine ESB) consisting of several different marine mammal tissue collections (Becker and Wise, 2006). One of these collections, the Alaska Marine Mammal Tissue Archival Project (AMMTAP), is ideal for assessing temporal changes of POPs in marine mammal populations. The AMMTAP collection spans from 1987 to the present day and contains liver, kidney and blubber samples obtained only from freshly dead animals using sampling protocols designed to reduce the likelihood of accidental sample contamination. Samples are processed in the field and then shipped to the NIST Marine ESB at liquid nitrogen vapor-phase temperatures (-150 °C) prior to permanent storage in liquid nitrogen vapor-phase freezers located in a clean room facility. The AMMTAP collection presently contains over 5,000 samples from 710 animals from 18 species of cetaceans, pinnipeds, and fissipeds collected from a variety of maritime regions in Alaska.

The objective of the present work was to use samples from the AMMTAP collection to assess temporal trends of POPs in northern fur seals (*Callorhinus ursinus*) occurring at summer rookeries in the Bering Sea (Table 1; Figure 1). Temporal trend data for both legacy persistent organic pollutants (legacy POPs), such as polychlorinated biphenyls (PCBs), and current or recently used POPs, such as the polybrominated diphenyl ethers (PBDEs), are needed from this region. Such data would provide a better understanding of POP concentrations trends in species from northern climates in response to climate change and source reductions mandated in the Stockholm Convention or other management activities (Riget et al., 2010). Fur seals are also consumed as food at the St. Paul Island collection site; therefore, understanding human exposure to current-use and legacy POPs was therefore also a motivation for this project. Vitamins were determined in samples as vitamin levels have been linked to POP concentrations in other work.

The Northern Fur Seal

The northern fur seal (NFS) is an otariid (externally eared) seal that has a wide geographic range in the North Pacific Ocean extending from Japan and Northern California northward to the Bering Sea (Gentry, 1997). The NFS tends to be smaller than most other species of otariid seals and weighs up to 250 kg. Adults are sexually dimorphic with mature males being much larger than females. The seals are polygynous and have established large breeding colonies (rookeries) at several locations in the Bering Sea, including at St. Paul in the Pribilof Islands (Fig.

1). Dominant males compete for harems of females while younger males gather in groups on or near their natal beaches. These younger immature males (2 to 3 years old) are harvested for food (and historically for pelts) at some locations, such as St. Paul Island. Breeding and pupping occur in the summer with peak pupping occurring typically during the first two weeks of July. Females raise a single pup and forage in local waters between feedings. When not on rookeries, NFSs are pelagic in the northern North Pacific Ocean. Animals using the St. Paul Island rookeries (Fig. 1), mainly subsist on pollock, sand lance, and squid (Antonelis et al., 1997). Pup production on St. Paul Island has declined nearly three-fold since the early 1970s, possibly as a result of declining forage fish availability due to climate change or from competition with commercial fisheries (Towell and Ream, 2006).

Persistent Organic Pollutants

Persistent organic pollutants can be categorized as either legacy or current-use. Legacy POPs are those that have been banned or phased-out from use in North America or were part of the original “dirty dozen” compounds specified in the Stockholm Convention (see www.pops.int). Legacy POPs include the PCBs, DDTs, chlordanes, toxaphene, hexachlorobenzene (HCB), aldrin, endrin, dieldrin, heptachlor, and mirex that were measured in this study in addition to polychlorinated dioxins and furans that were not determined in this work (Table 1). In 2009 and 2011, α -, β - and γ -hexachlorocyclohexane (HCH), pentachlorobenzene, perfluorooctane sulfonate (PFOS), endosulfan, and PBDEs, all measured in this study, were added to the Stockholm Convention POPs list. Chlorodecone (Kepone) and hexabromobiphenyl, which were not measured in the present work, are also on the Stockholm POPs list. Non-legacy POPs include those that still have some limited production or are still in wide-spread use, the perfluoro alkyl acids (PFAAs), PBDEs, and HBCDs fall into this category. In general, levels of these compounds are increasing in Arctic and nearby regions (de Wit et al., 2010; de Wit and Muir, 2010; Hoguet et al., 2013; Reiner et al., 2011).

There has been keen interest in determining both temporal and spatial trends of POPs in the Arctic and nearby environs such as the Bering Sea and North Pacific Ocean to understand concentration trajectories or possible toxic effects in marine food webs. Both of these ocean basins are believed to be major receptors of POP emissions and are proximate to Asia where legacy POPs are either still in use or where use has only recently been curtailed. Consequently, temporal trends of POPs in these regions are of interest. In addition, the accumulation of POPs in marine mammals such as polar bears, dolphins, and seals has been linked to toxic effects including suppressed immune function and endocrine disruption (Beckmen et al., 2003; Lahvis et al., 1995; Letcher et al., 2010). Determining POP concentrations in northern fur seals was thereby important to place their contaminant burdens in perspective to burdens in marine

mammals where levels are linked to toxic effects. From a human health perspective, Native Alaskans frequently rely on marine mammals for subsistence thus placing humans in a similar trophic position as marine mammals. Therefore, understanding human exposure to POPs was an additional motivation for this study.

There have been two prior investigations of POPs in northern fur seals. Beckman et al. (Beckmen et al., 2003) collected blood and milk samples from NFS mother/pup pairs on St. Paul Island and examined the relationship between blood POP concentration and immune response. Primiparous females had higher POP concentrations and mounted weaker immune response following vaccination than multiparous females. The results suggest an effect of higher POP levels in blood leading to decreased immune response in NFS (Beckmen et al., 2003; Beckmen et al., 1999). Temporal trends of POPs in NFS have been reported in blubber from mature female fur seals collected from 1971 through 2008 off of the Sanriku Coast in Japan (Kajiwara et al., 2004). Significant declines were observed for PCBs from 1975 onward. PBDEs accumulation mirrored PBDE usage in Japan during that time span with concentrations increasing until 1994 and then declining in more recent years. HCB did not show significant declines. Aside from this work, there have been no studies documenting temporal trends of POPs in NFSs highlighting the need for the present work.

Materials and Methods

Samples

Samples of liver and blubber were collected opportunistically from two to three year old male fur seals after they were taken during subsistence hunts on Saint Paul Island, Alaska (Figure 1 and Table 2). Samples were collected from 50 seals in 1987 (n=5), 1990 (n=10), 1997 (n=5), 2000 (n=10), 2006 (n=10), and 2007 (n=10). Data on seal weight, length and blubber thickness were only available for samples collected prior to 2000.

Blubber and liver samples were processed in the field using established protocols that were designed to minimize sample contamination (Becker and Wise, 2006). After processing, samples were placed in liquid nitrogen vapor-phase shippers and then sent to the NIST Marine Environmental Specimen Bank (Marine ESB) for storage and later homogenization following established protocols (Pugh et al., 2007). While in the NIST Marine ESB, samples were stored in liquid nitrogen vapor-phase freezers (-150 °C) in a clean room environment. Samples designated for analysis and not long term archival were cryogenically homogenized using the method described elsewhere (Zeisler et al., 1983).

Vitamins

Sample Preparation

QC97LH2, a beluga liver control material, and 49 NFS liver samples were provided by the NIST Marine ESB. Calibration solutions (n=5) were prepared of the fat-soluble vitamins A (retinol) and E (α -tocopherol and γ -tocopherol, respectively) (all concentrations determined spectrophotometrically). A solution of benzo[a]pyrene- d_{12} in methanol (approximately 7 $\mu\text{g/g}$) was prepared and used as the internal standard (IS).

NFS liver samples (approximately 1 g) and a liver quality control sample (QC97LH2) (approximately 1 g) were gravimetrically weighed into hot block digestion cups. Six milliliters of a 1:1 (volume fraction) tetrahydrofuran/methanol mixture with 200 mg/g pyrogallol was added to each sample. The samples were allowed to stir for 15 min. After stirring, 1.2 mL of 50 % (volume fraction) KOH in methanol was added to the sample. The samples were stirred at 40 °C for 1 h followed by the addition of 500 μL of acetic acid to neutralize the base and an additional 10 mL of water containing 10 % NaCl (mass fraction).

Liquid-liquid extraction was performed three times using 10 mL of a 1:1 (volume fraction) hexane:petroleum ether mixture. The combined extract was evaporated to approximately 20 mL and washed with water. The organic layer was removed, evaporated to dryness, and then reconstituted in 1 mL of 9:1 (volume fraction) ethanol:ethyl acetate. All of the above steps were performed in a room with subdued lighting, and when possible, the samples were fully shielded from the light.

Instrumental method

Analysis was performed on an Agilent 1100 liquid chromatograph (LC) coupled with a Dionex UltiMate 3000 LC/UV with a Jasco FP-1520 fluorescence detector. Samples (20 μL) were injected onto a Hypersil Green PAH column (250 mm x 4.6 mm i.d., 5 μm particles) held at 5 °C. An isocratic mobile phase consisting of 97 % acetonitrile with 3 % methanol by volume for 50 min was used for the separation. A response factor was calculated for each calibrant injection, and the average response factor was used for the calculation of retinol and tocopherol concentrations in the liver samples.

PCB Congeners, DDT compounds, Mirex, Chlorinated Cyclodienes, HCHs, PBDE Congeners, HCB, and HBCDs

Sample Preparation

Calibrants were prepared by gravimetrically combining the following solutions: Standard Reference Materials (SRMs) 2261, 2262, 2274, 2275, two additional PCB congener solutions, a solution of 26 PBDE congeners, a solution of PBDE 209, commercially purchased solutions of octachlorosytrene (Accustandard, Inc., New Haven, CT), three isomers of HBCDs, and toxaphene congeners Parlar 26, 32, 50 and 62 (Cambridge Isotope Laboratories, Andover, MA). Concentrations of components in the non-SRM solutions used to prepare the calibrants were not corrected for purity although the stated purities were all >98 %. The six-point calibration curve, processed alongside each sample set, contained from approximately 0.05 ng to 230 ng of each component of interest.

An internal standard (IS) solution in iso-octane contained the following compounds obtained from Wellington Laboratories (Guelph, Ontario, Canada), Cambridge Isotope Laboratories, Accustandard, and Chiron (Trondheim, Norway): ¹³C-labeled PCB congeners (28, 52, 77, 126, 169, 118, 153, 180, 194, and 206), 6-F-PBDE 47, PBDE 104, 4'-F-PBDE 160, 4'-F-PBDE 208, ¹³C-labeled PBDE 209, ¹³C-labeled pesticides (HCB, *trans*-chlordane, *trans*-nonachlor, oxychlordane, 4,4'-DDE, 4,4'-DDD, 4,4'-DDT), ¹³C-labeled methyl-triclosan and ¹³C-labeled α-, β- and γ-HBCDs. This solution contained approximately 300 ng/g (mass fraction) of each compound.

Blubber samples (~1 g) including subsamples of SRM 1945 Organics in Whale Blubber as a control sample were extracted using pressurized fluid extraction (PFE; ASE Dionex, Sunnyvale, CA). Three samples, FRSL-187, 808, and 828, were run in duplicate with one analysis in each of two batches. Cryohomogenized blubber was mixed with diatomaceous earth (Hydromatrix, Restek, Bellefonte, PA) or sodium sulfate and then packed into stainless steel extraction vessels. The IS solution was gravimetrically added to the top of each PFE vessel. The mixtures were extracted using dichloromethane. The cell temperature and pressure were set to 100 °C and 13.8 MPa, respectively, and there were three extraction cycles. Extracts were frozen, and the residual water was removed. Total extractable organic content (or lipid content) was determined gravimetrically by subsampling the extract. Solvent volume was reduced to 1 mL under nitrogen and lipid was removed by size exclusion chromatography column (PLGel 10 μm, 100 Å). The resultant fraction was evaporated to 0.5 mL and solvent exchanged to hexane. An additional step was performed using solid phase extraction cartridges packed with 30 % sulfuric acid/silica (mass fraction) in order to further clean up the samples and fractionate the more polar HBCD isomers from the other analytes as described previously (Hoguet et al., 2013). Fraction One (F1) extracts were solvent exchanged to *iso*-octane, evaporated, and transferred to amber auto sampler vials with a final volume of 0.5 mL. Fraction Two (F2) extracts

containing HBCD isomers were solvent exchanged to *iso*-octane then methanol, evaporated, and transferred to auto sampler vial (ASV) inserts and reduced to a final volume of approximately 200 μ L. F1 extracts were analyzed using gas chromatography with mass spectrometric detection (GC/MS) for the majority of the compounds using two ionization modes as described below, and F2 extracts were analyzed using liquid chromatography with a triple quadrupole MS (LC-MS/MS) for HBCDs.

Instrumental method

GC/MS-EI for PCB Congeners, DDT-related compounds, and Mirex

Samples were analyzed using two GC/MS injections, one for the pesticides and one for the PCB congeners. Both were on an Agilent 6890N GC coupled to an Agilent 5973 MS (Agilent Technologies, Santa Clara, CA) operated in the electron impact (EI) mode with selected ion monitoring (SIM). The column used for the first injection was a DB-XLB (0.25 mm x 60 m x 0.25 μ m; Agilent Technologies) coupled to a 5 m x 0.25 mm retention gap. Cool-on-column injections of 1 μ L were done for the pesticide run. The column temperature for the pesticide run was held for 1 min at 60 °C, ramped to 180 °C at a rate of 40 °C/min, held for 10 min at 180 °C, then ramped to 280 °C at a rate of 2 °C/min, and finally held at 280 °C for 5 min.

The second run, for PCB congeners, utilized the same column as the first injection. Cool-on-column injections of 1 μ L were done for the PCB run. The column temperature for the PCB run was held for 1 min at 60 °C, ramped to 180 °C at a rate of 40 °C/min, held for 10 min at 180 °C, then ramped to 280 °C at a rate of 2 °C/min, and finally held at 280 °C for 46 min.

GC/MS-NCI for PBDEs, Cyclodienes, HCHs and Chlorobenzenes

The PBDEs, cyclodienes, HCHs, and chlorobenzenes were analyzed using an Agilent 6890 GC and 5975B MS (Agilent Technologies) operated in the negative chemical ionization (NCI) mode with SIM. Methane was used as the reagent gas. The column used was a 30 m x 0.18 mm x 0.18 μ m DB-5ms (Agilent Technologies) equipped with a 5 m x 0.25 mm retention gap. Sample extracts (reduced to 0.5 mL) were injected (2 μ L) using cool on-column injection. The column temperature for the NCI run was held for 1 min at 60 °C, ramped to 170 °C at a rate of 25 °C/min, then ramped to 270 °C at a rate of 2 °C/min, next ramped to 325 °C at a rate of 25 °C/min, and finally held at 325 °C for 15 min.

LC-MS/MS for HBCDs

LC-MS/MS was used with the electrospray ionization (ESI) source. A 20 µL injection of F2 extracts was made, and HBCD isomers were separated on an Agilent Eclipse Plus C18 column (3.0 mm x 150 mm x 3.5 µm column) connected to an API 4000 (Applied Biosystems, Foster City, CA). The solvent program was as follows: solvent A was 2.5 mM ammonium acetate in 12.5 % water in methanol (volume fraction) and solvent B was acetonitrile. Flow was 0.3 mL/min and the following mobile phase gradient was used. Initial conditions began at 90 % A, ramped to 67 % A by 12 min, and held at 67 % A for 3 min. At 15 min, the solvent was ramped to 0 % A by 20 min, held for 3 min, and then ramped from 23 min to 28 min to 90 % A and held for 5 min.

Perfluorinated Alkyl Acids

Sample Preparation

One aliquot of NIST SRM 1946 Lake Superior Fish Tissue and one aliquot of SRM 1947 Lake Michigan Fish Tissue were run as control materials in this analysis and compared to concentrations determined previously (Reiner et al., 2012). A calibration series (n = 7) was prepared from three independent weighings of neat PFAAs in methanol (see Table 3 for list). A solution of $^{13}\text{C}_8$ -PFOA, $^{13}\text{C}_9$ -PFNA, $^{13}\text{C}_9$ -PFDA, $^{13}\text{C}_2$ -PFUnA, $^{13}\text{C}_2$ -PFDoA, $^{18}\text{O}_2$ -PFBS, $^{13}\text{C}_4$ -PFOS, and $^{18}\text{O}_2$ -PFOSA in the water was used as the IS solution for PFAAs. A methanolic solution of $^{13}\text{C}_2$ -PFOA was used as the recovery standard.

Samples and calibrants were extracted using the potassium hydroxide (KOH) in methanol method as described previously (Reiner et al., 2011). Briefly, approximately 0.5 g of each sample was transferred to a 50 mL polypropylene centrifuge tube using a stainless steel spatula. MilliQ water (0.5 mL) was added to each sample. The aqueous IS (0.2 mL) was then gravimetrically added to each sample. The centrifuge tubes were vortexed for 10 sec and allowed to settle for 20 min. Three milliliters of 0.01 mol/L of KOH in methanol was added to the samples and sonicated for 30 min. Samples were then centrifuged at 2500 rpm for 5 min, and the supernatant was transferred to a glass culture tube with a glass pipet. The original sample tube received an additional 3 mL of 0.01 mol/L of KOH in methanol, and the extraction procedure was repeated, placing the additional round of supernatant in the glass culture tube as well. Extracts were evaporated under nitrogen, at 35 °C, to approximately 3 mL and filtered using a Whatman UniPrep 0.2 µm filter (Stanford, ME). Samples were further evaporated to 1 mL (under nitrogen) and 10 mL of 50 % (volume fraction) formic acid (98 %, Fluka; Buchs, Switzerland) in water was added to each sample. Samples were loaded onto Waters Oasis WAX

SPE columns (3 cc, 60 mg, 30 μ m; Milford, MA) on a RapidTrace (Biotage, Uppsala, Sweden) workstation. The fraction of interest from the SPE column was concentrated under nitrogen to approximately 0.5 mL. They were spiked with 0.2 mL of the recovery standard, vortexed, and transferred to ASVs for instrumental analysis.

Instrumental method

The extracts were run using the LC-MS/MS (Agilent 1100 LC (Palo Alto, CA) interfaced to a negative electrospray ionization tandem mass spectrometer (API 4000, Applied Biosystems-MDS Sciex, Foster City, CA). Samples (5 μ L) were injected onto a Phenomenex Kinetex 2.6 μ PFP column (50 mm x 3.0 mm). The solvent gradient given in % by volume (flow rate 0.3 mL/min) started at 40 % methanol and 60 % 20 mmol/L ammonium acetate in water and then increased to 65 % methanol by 5 min, held for 5 min, and then increased to 95 % methanol by 12 min, and held for 3 min, before reverting back to original conditions at 15.5 min with a 14.5 min hold time. The two most abundant transitions for each PFAA were monitored.

Quantification of Analytes of Interest

Amounts of each compound of interest in the samples were calculated using the slope and y-intercept of at least a three point calibration curve that bracketed the peak area ratios observed in the samples. Concentrations were determined by dividing the calculated analyte mass by the extracted sample mass. The limits of detection (LOD) were determined as the maximum value of either the average ng measured in procedural blanks plus 3 times the standard deviation of the blanks or the lowest calibrant detectable, all divided by the sample mass. For the PFAAs, final concentrations are corrected for the salt content of the neat materials, and concentrations are totals including branched and linear isomers.

Statistical methods

Statistical comparisons were performed using JMP 10 (SAS Institute, Cary, NC). POP concentrations and vitamins were log transformed to provide a normal distribution. Lipid content did not require log transformation. Comparisons were made by performing multiple linear regression including the rookery and year as independent variables.

Results and Discussion

Vitamins

Vitamin levels in free-ranging marine mammals are reflective of many factors (physiological, biological, and chemical). While vitamins A and E are essential for reproduction, development, cellular and immune function, little is known about the “normal” levels in wildlife (Simms and Ross, 2000). This study provides much needed information about the mass fractions of vitamin A (retinol) and vitamin E (α - and γ -tocopherol) in one species of marine mammal. Mass fractions of total retinol (Σ retinol), α - and γ -tocopherol are shown for the control material in Table 4. The relative standard deviation for retinol and γ -tocopherol were less than 10 %, however the relative standard deviation for α -tocopherol was 30 % indicating higher variability in the measurement of this vitamin. Vitamin concentrations in the NFS liver samples are given in Table 5 with means by rookery shown in Table 6. Mass fractions of the vitamins in livers were highest for α -tocopherol with the concentration ranging from below the limit of detection to 23400 mg/kg in frsl-54 (median 10300 mg/kg). Log transformed vitamin mass fractions did not significantly differ among rookeries or year, nor was there a significant statistical interaction between rookery and year. In other words, vitamin concentrations did not differ among rookeries or among sampling years in the seals and mass fraction of vitamins exhibited considerable variability among individual seals. The relative standard deviation of the three vitamins were 134 %, 43 % and 42 % for Σ retinol, α - and γ - tocopherol, respectively. Since there is no baseline information on the concentrations of Σ retinol, α - and γ - tocopherols in NFS, we can assume that the differences seen in this study is due to feeding/post feeding intervals, seal condition and prey sources. Vitamin concentrations were also not significantly related to summed POP concentrations.

Legacy Organochlorine POPs

Since lipid content in tissues is often correlated to concentrations of lipophilic (fat-soluble) pollutants such as PCBs and DDTs, lipid content was determined in all NFS blubber samples. Lipid content ranged from 42.9 % (mass fraction) to 84.3 % with an average value of 68.5 % (SD = 9.5 %; Table 2). POPs were not correlated to lipid content therefore POP concentrations were not lipid normalized and are expressed on a wet mass basis.

The quality control of sample analysis for the organochlorine POPs included analyzing four aliquots of SRM 1945. Results from the analysis of SRM 1945 are shown in Tables 7 and 8. The mean concentration of the four aliquots was within the uncertainty or within 20 % of the certified values listed on the Certificate of Analysis except for *trans*-chlordane (Table 7). The

reason for the higher *trans*-chlordane value relative to the certified value for SRM 1945 (17.8 ng/g (1.7 ng/g SD) vs. 11.8 ng/g \pm 0.5 ng/g) may result from the coelution of *trans*-chlordane enantiomers in this study that separated during the certification measurements. Toxaphene congeners determined in SRM 1945 were between 88 % and 43 % higher than reference values derived from interlaboratory comparisons and prior analysis of toxaphene congeners by NIST. The discrepancy is likely due to non-agreement of stated concentrations of toxaphene congeners in an earlier lot compared to the lot used for this work. This earlier lot was used by NIST and was likely used by other laboratories that submitted data to NIST during the recertification of SRM 1945. Purity was assessed in the most recently used lots of toxaphene congeners; however concentration values stated by the vendor could not be verified. Regardless, the toxaphene values in this study are internally consistent and are useful to gauge the decline of toxaphene concentrations in NFS with time.

The average percent difference of values determined in duplicate measurement on the same homogenized seal blubber ranged from 14 % for pesticides determined by GC-MS/NCI to 2.5 % for PCBs determined by GC-MS/EI.

Concentration values for legacy POPs are given in Tables 9 and 10 and summarized in Tables 11 and 12. Median concentrations of legacy POPs were highest for Σ PCBs at 1540 ng/g (range 558 ng/g to 2210 ng/g) and lowest for the Σ CIBz at 2.93 ng/g (range 1.37 ng/g to 7.54 ng/g). Σ DDTs concentrations in NFS were highly variable ranging from 223 ng/g to 83200 ng/g (Tables 10 and 11). The variability in Σ DDT concentrations was partially driven by a significant ($p < 0.001$) decline with year of sampling (Figure 2), but among colony differences in Σ DDTs irrespective of year were not significantly different however colonies were not sampled all years (Table 2 and Figure 3). While concentrations generally declined with year of sampling, there was a significant ($P < 0.001$) and nearly 3-fold increase in Σ DDT concentration from 2006 until 2007 (from 464 ng /g (207 ng/g) to 1230 ng/g (590 ng/g) (mean (1SD))). This may be due to differences in forage location that appears to be colony dependent (Call et al., 2008) or possibly differences in dispersal during the non-breeding season. An annual difference of this magnitude in global sources of Σ DDT between these two years seems unlikely. Other POPs also showed significant declines with year (log concentration versus year) including Σ CIBzs ($P < 0.001$), Σ HCH ($P < 0.001$), Σ Chlor, and Σ Tox. Σ PCB and mirex concentrations did not significantly decline with year.

There are few other seal data from this region of the north Pacific to compare to POP concentrations seen in this study. The majority of seal data are from regions further north and east of the study site, primarily in the Canadian Arctic (Riget et al., 2010). The two most relevant studies to this work with regard to location and compounds measured were studies conducted on ringed seals from Alaska (Kucklick et al., 2006) and NFS from the northeastern

coast of Honshu, Japan (Kajiwara et al., 2004). Median concentrations of legacy POPs in male ringed seals collected in 1996 from near Nome, AK in comparison to legacy POPs in NFS from 1997 are shown in Figure 4. Median concentrations of Σ HCHs and Σ PCBs were similar between the two seal species. Σ DDT in NFSs was approximately twice the concentration observed in ringed seals while Σ Chlor was about half. Declining trends in chlordane concentration going from east to west have been observed for marine mammals in this region and the further west sampling location of NFS relative to the ringed seals would be consistent with this trend (Kucklick et al., 2006). The higher concentration of Σ DDTs in NFS relative to ringed seals suggests an Asian source of DDT to the NFS population.

Concentrations of legacy POPs from adult female NFS collected from the waters of northern Japan (Kajiwara et al., 2004) are also shown in Figure 4. The concentrations shown are the average concentrations from collections of seals made in 1994 and 1997 (n=5 female seals each year). With exception of Σ PCBs, POP concentrations for seals collected from the two locations (northeast Honshu coast and St. Paul Island) are very similar. While concentrations appear to be similar between the two locations, it should be noted that the samples from Japan were from reproductively mature female seals that have lower blubber concentrations than the juvenile male NFS seals examined in this study. Hickie et al. (Hickie et al., 2005) showed that adult female ringed seal PCB concentrations are reduced to about half of juvenile male concentrations. Extending this observation in ringed seals to the NFS, one would expect to see POPs in juvenile male NFS from coastal Japan at levels twice those observed at St. Paul Island.

Current-use POPs

Current-use POPs include the PBDE and HBCD flame retardants, as well as the PFAAs. Quality control of the measurement of PFAAs included the analysis of two aliquots of SRM 1946 Organics in Lake Superior Fish Tissue and two additional aliquots of SRM 1947 Organics in Lake Michigan Fish Tissue. With the exception of perfluorooctane sulfonate (PFOS), most PFAAs were below the limits of detection (Table 13). Mean PFOS values determined in the control samples were 2.07 ng/g (range 1.84 ng/g to 2.29 ng/g) and 6.15 ng/g (range 5.42 ng/g to 6.88 ng/g) for SRMs 1946 and 1947, respectively compared to certified values of 2.07 ng/g \pm 0.08 ng/g and 6.15 ng/g \pm 0.39 ng/g, respectively. PBDEs determined in SRM 1945 Organics in Whale Blubber were within <20% of certified values (Table 7). α -HBCD in SRM 1945 averaged 6.1 ng/g (0.60 ng/g SD).

Concentrations of the PFAAs in NFS livers are given in Table 3. Concentrations of individual PFAAs that were detected were generally in the single ng/g to tens of ng/g range with concentrations being highest in the odd-chain PFAAs. This is unlike PFAA compound profiles

seen in aquatic mammals from temperate or subtropical latitudes where profiles are dominated by PFOS (Houde et al., 2006). The pattern observed in the present work is typical for Arctic and subarctic marine mammals and has recently been reported in beluga whales from relatively nearby coastal Alaska and Cook Inlet (Reiner et al., 2011). However, in that study PFOSA dominated in male beluga whales along with PFOS and PFUnA. The magnitude of concentrations was fairly similar to this work with detectable values generally averaging in the ng/g to tens of ng/g range per compound. Total PFAAs increased significantly ($\log \Sigma\text{PFAA}$ vs. year; $r^2 = 0.53$) and nearly four-fold in seals from 1987 until 2007 which was similar in magnitude to that observed in beluga whales from Alaska (Reiner et al., 2011). There was no significant difference in ΣPFAA values among rookeries; however, rookery concentration was highly dependent on year of sampling.

Brominated flame retardant concentrations in NFS are given in Table 14 and summarized in Tables 11 and 12. PBDEs were detected in all samples and HBCDs were detected in 48 of 50 samples. Both ΣPBDEs and ΣHBCDs concentrations ranged nearly three orders of magnitude with the lowest concentrations generally observed in seals collected during the early years of sampling. As with the perfluorinated compounds, both classes of brominated flame retardants showed dramatic and significant ($r^2 = 0.52$ and 0.53 for ΣPBDEs and ΣHBCDs , respectively) increases with sampling year (Figure 2). Similar increases in PBDEs have been reported in ringed seals from the Canadian Arctic (Ikonomou et al., 2002) and more recently in beluga whales from Alaska (Hoguet et al., 2013). In the current study, there was however a decline in ΣPBDE concentrations between 2006 and 2007 suggesting that PBDE concentrations may be plateauing in NFS in response to phasing out the use of PBDE mixtures in the northern hemisphere. PBDE 47 dominated the PBDE profile in samples comprising on average 61 % (10 % SD) of total PBDEs. This fraction was consistent among all sampling events suggesting little change in the PBDE profile in the NFS forage fish during the sampling period.

The ΣHBCDs were dominated by $\alpha\text{-HBCD}$ as seen in other studies (Hoguet et al., 2013). $\gamma\text{-HBCD}$ was detected in eight samples although the concentrations were less than 1 ng/g wet mass. The exponential increase in ΣHBCD observed here is similar to what was recently reported for beluga whales from Cook Inlet, AK and from the Chukchi Sea, AK (Hoguet et al., 2013). HBCD is still used in North America although this mixture has been recognized by the Stockholm Convention as a candidate for removal from use. The inclusion of HBCD as a POP has sparked debate as the biomagnification potential for this compound is not as high as the legacy POPs. Despite its lower bioaccumulation potential, concentrations in NFS are increasing although the rate of increase with time is half that observed in Alaska beluga whales (slopes 0.12/year versus 0.26/year).

Conclusions

Results from this study demonstrate the utility of using archived samples as a source for understanding both geographic and temporal patterns of persistent organic pollutants. The concentrations of POPs in NFS are considerably lower than those observed in other marine mammals from the lower 48 United States; however, the concentrations may still be of concern to animal health as suggested in prior studies. Fortunately, with the exception of mirex, the concentrations of legacy POPs are declining with time in NFS highlighting the success of efforts aimed at curbing POP emissions in the Pacific Region. Current use POPs, however, are increasing in concentration in NFS and continued monitoring of NFS for contaminants is therefore warranted. Based on suggestions from this dataset and other work, PBDEs concentrations have likely peaked and are now declining. This is not true for PFAAs or HBCDs which continue to increase both in this study and the recent work on beluga whales. Archival of NFS tissues should be continued as the population of this animal in the Northern Pacific appears to be declining, and information on contaminants will help in ruling out or ruling in causes for declines. Continued sampling also extends the timeline for monitoring and samples taken now may be useful for identifying trends in the next generation of POPs. When the AMMTAP collection was begun in the late 1980s, legacy POPs were well-known; however, PBDEs, PFAAs, and HBCDs were unrecognized as POPs at that time.

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does it imply that the materials or equipment identified are necessarily the best available for the purpose.

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Table 1: Compounds determined in this work

Compound or class	Legacy POP?	Current Use POP?	Listed in Stockholm Convention?	Comments
Polychlorinated biphenyl (PCBs)	yes		yes	Total is the sum of 58 congeners or congener pairs (Σ PCB)
Chlordanes	yes		yes	Total is the sum of <i>cis</i> - and <i>trans</i> -chlordane; <i>cis</i> - and <i>trans</i> -nonachlor, heptachlor, oxychlordane (Σ Chlor)
Hexachlorocyclohexanes (HCHs)	Yes		Yes	Total is the sum of α -, β -, and γ -HCH (Σ HCH)
Mirex	Yes		Yes	
Toxaphene (sum of four congeners)	Yes		Yes	Four congeners were determined Parlar numbers 26, 32, 50, and 62 (Σ Tox)
Penta- and hexachlorobenzene (PeCB and HCB)	Yes		Yes	Sum is the sum of penta- and hexachlorobenzene (Σ CBz)
Dichlorodiphenyl ethane related compounds (DDTs)	Yes		Yes	Total DDT is the sum of 2,4'- and 4,4'- DDE, DDD and DDT (Σ DDT)
Perfluorinated alkyl acids (PFAAs)		Yes	Yes for PFOS	Total PFAAs is the sum of four to 14 carbon perfluorocarboxylic acids, perfluorobutane sulfonate, perfluorohexane sulfonate, perfluorooctane sulfonate, and perfluorooctanesulfonamide (Σ PFAA)
Polybrominated diphenyl ethers (PBDEs)		Yes	Yes	Sum of PBDE is congeners 47, 99, 100, 153, 154, and 155 (SPBDE)
Hexabromocyclododecanes (HBCD)		Yes	Yes--proposed	HBCDs is limited to α -HBCD in this work

Table 2: Sample Information.

^a 692-FRSL-	Blubber Storage#	Liver Storage#	Collection Date	Rookery	Seal Mass (kg)	Seal length (cm)	Blubber thickness (cm)	Blubber Lipid content (%)
1	MM1B004	MM1L001	7/28/1987	Polavina	56.3	114	1.5	51.2
2	MM1B008	MM1L005	7/28/1987	Polavina	77.6	117.3	2.5	73.0
3	MM1B012	MM1L009	7/28/1987	Polavina	84	124	3	70.8
4	MM1B016	MM1L013	7/29/1987	Northeast	64.7	111.4	2.5	69.6
5	MM1B020	MM1L017 ^a	7/29/1987	Northeast	58.7	110.4	3	80.7
6	MM4B097	MM4L095	7/23/1990	Zapadni	21.8	97.8	1.2	62.5
7	MM4B100	MM4L098	7/23/1990	Zapadni	23	103.5	0.9	69.5
8	MM4B103	MM4L101	7/23/1990	Zapadni	21.3	103.5	1.1	63.2
9	MM4B106	MM4L104	7/23/1990	Zapadni	na ^b	na	na	72.4
10	MM4B109	MM4L107	7/24/1990	Reef	15.8	94.6	0.8	50.6
11	MM4B112	MM4L110	7/24/1990	Reef	17.3	43.2	0.7	45.1
12	MM4B115	MM4L113	7/24/1990	Reef	20.2	99.1	1	57.8
13	MM4B118	MM4L116	7/27/1990	Reef	20.2	na	1	76.4
14	MM4B121	MM4L119	7/24/1990	Reef	15.9	95.9	0.9	66.2
15	MM4B124	MM4L122	7/24/1990	Reef	17.4	104.1	1.2	42.9
16	MM12B648	MM12L646	8/6/1997	Zapadni	na	99.1	too thin	67.0
17	MM12B652	MM12L650	8/6/1997	Zapadni	na	110.5	too thin	57.0
18	MM12B656	MM12L654	8/6/1997	Zapadni	na	111.8	too thin	65.5
19	MM12B660	MM12L658	8/6/1997	Zapadni	na	123.2	too thin	57.0
20	MM12B664	MM12L662	8/6/1997	Zapadni	na	111.8	too thin	65.0
22	MM16B169C	MM16L167C	7/10/2000	Polavina	na	na	na	62.5
23	MM16B172C	MM16L170C	7/10/2000	Polavina	na	na	na	78.6
24	MM16B175C	MM16L173C	7/10/2000	Polavina	na	na	na	71.0
25	MM16B178C	MM16L176C	7/10/2000	Polavina	na	na	na	60.4
26	MM16B181C	MM16L179C	7/10/2000	Polavina	na	na	na	79.9
27	MM16B184C	MM16L182C	7/10/2000	Polavina	na	na	na	68.5
28	MM16B187C	MM16L185C	7/10/2000	Polavina	na	na	na	76.8
29	MM16B190C	MM16L188C	7/10/2000	Polavina	na	na	na	69.7
30	MM16B193C	MM16L191C	7/10/2000	Polavina	na	na	na	68.8
31	MM16B196C	MM16L194C	7/10/2000	Polavina	na	na	na	74.8
38	MM22B819C	MM22L818C	7/22/2006	Reef	na	na	na	75.8
39	MM22B822C	MM22L820C	7/22/2006	Reef	na	na	na	75.4
40	MM22B825C	MM22L823C	7/22/2006	Reef	na	na	na	65.3
41	MM22B828C	MM22L826C	7/22/2006	Reef	na	na	na	81.9
42	MM22B831C	MM22L829C	7/22/2006	Reef	na	na	na	70.4
44	MM22B796C	MM22L794C	7/22/2006	Reef	na	na	na	82.2
45	MM22B799C	MM22L797C	7/22/2006	Reef	na	na	na	84.3
46	MM22B808C	MM22L806C	7/22/2006	Reef	na	na	na	70.7
47	MM22B802C	MM22L800C	7/22/2006	Reef	na	na	na	77.5
48	MM22B836C	MM22L834C	7/22/2006	Reef	na	na	na	83.6
52	MM22B899C	MM22L897C	7/8/2007	Polavina	na	na	na	74.2
53	MM22B902C	MM22L900C	7/8/2007	Polavina	na	na	na	77.9
54	MM22B905C	MM22L903C	7/8/2007	Polavina	na	na	na	63.7
55	MM22B908C	MM22L906C	7/8/2007	Polavina	na	na	na	56.1
56	MM22B911C	MM22L909C	7/8/2007	Polavina	na	na	na	68.1
57	MM22B914C	MM22L912C	7/8/2007	Polavina	na	na	na	76.5
58	MM22B917C	MM22L915C	7/8/2007	Polavina	na	na	na	68.3
59	MM22B920C	MM22L918C	7/8/2007	Polavina	na	na	na	64.1
60	MM22B923C	MM22L921C	7/8/2007	Polavina	na	na	na	70.3
61	MM22B926C	MM22L924C	7/8/2007	Polavina	na	na	na	65.4

^aMM1L017 sample was never located and excluded from the analysis. ^b“na” not available

Table 3: Perfluorinated alkyl acids (ng/g wet mass) in northern fur seal liver

FRSL-	Storage ID	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnA
1	MM1L001	<3.7	<1.1	<0.5	<0.4	<1.1	0.92	<0.9	1.5
2	MM1L005	<3.9	<1.1	<0.6	<0.5	<1.1	0.68	<0.9	3.3
3	MM1L009	<4.6	<1.3	<0.7	<0.5	<1.3	3.3	1.7	10.9
4	MM1L013	<1.1	<1.2	<0.2	<2.00	<0.5	2.4	<0.4	17.8
5	missing	--	--	--	--	--	--	--	--
6	MM4L095	<2.5	<0.7	<0.4	<0.3	<0.7	1.5	<0.6	1.68
7	MM4L098	<1.7	<2.0	<0.2	<3.2	<0.8	1.4	<0.6	7.8
8	MM4L101	<5.9	<1.7	<0.8	<0.7	<1.7	1.3	<1.4	2.7
9	MM4L104	<7.4	<2.0	<1.1	<0.9	<2.1	1.2	<1.8	5.2
10	MM4L107	<2.1	<2.4	<0.3	<3.9	<1.0	3.7	<0.8	13.0
11	MM4L110	<1.2	<1.4	<0.2	<2.3	<0.6	0.98	<0.5	34.6
12	MM4L113	<4.5	<1.2	<0.7	<0.5	<1.3	3.3	<1.1	4.3
13	MM4L116	<4.7	<1.3	<0.7	<0.5	<1.3	0.56	<1.1	0.55
14	MM4L119	<5.4	<1.5	<0.8	<0.6	<1.5	1.5	<1.3	4.2
15	MM4L122	<4.9	<1.4	<0.7	<0.6	<1.4	1.0	<1.2	3.4
16	MM12L646	<2.4	<0.7	<0.3	<0.3	<0.7	2.7	0.87	9.3
17	MM12L650	<5.6	<1.6	<0.8	<0.7	<1.6	1.9	<1.3	6.7
18	MM12L654	<4.0	<1.2	<0.6	<0.5	<1.1	4.1	1.3	6.4
19	MM12L658	<4.2	<1.2	<0.6	<0.5	<1.2	4.5	1.2	6.9
20	MM12L662	<3.9	<1.1	<0.6	<0.5	<1.1	8.5	1.4	12.2
22	MM16L167	<1.8	<2.0	<0.3	<3.3	<0.8	<1.35	<0.7	8.8
23	MM16L170	<2.2	<2.5	<0.3	<4.1	<1.1	5.0	<0.8	75.7
24	MM16L173	<1.5	<1.8	<0.2	<2.9	<0.7	6.0	<0.6	57.0
25	MM16L176	<1.4	<1.6	<0.2	<2.6	<0.7	1.9	<0.5	41.5
26	MM16L179	<2.1	<2.4	<0.3	<3.9	<1.0	18.8	<0.8	11.5
27	MM16L182	<1.3	<1.5	<0.2	<2.5	<0.6	3.3	<0.5	24.6
28	MM16L185	<1.0	<1.2	<0.2	<2.0	<0.5	2.5	<0.4	26.9
29	MM16L188	<3.0	<3.5	<0.4	<5.7	<1.4	6.0	<1.1	13.3
30	MM16L191	<1.6	<1.9	<0.2	<3.1	<0.8	2.3	<0.6	10.4
31	MM16L194	<1.5	<1.7	<0.2	<2.8	<0.7	4.3	<0.5	22.8
38	MM22L818	<1.0	<1.1	<0.1	<1.9	<0.5	3.3	<0.4	51.7
39	MM22L820	<1.4	<1.6	<0.2	<2.6	<0.7	11.8	<0.5	115
40	MM22L823	<1.3	<1.5	<0.2	<2.5	<0.6	7.6	<0.5	34.5
41	MM22L826	<1.5	<1.7	<0.2	<2.7	<0.7	13.1	<0.5	105
42	MM22L829	<1.2	<1.4	<0.2	<2.3	<0.6	10.1	<0.5	21.0
44	MM22L794	<1.4	<1.6	<0.2	<2.6	<0.7	3.7	<0.5	15.5
45	MM22L797	<1.2	<1.3	<0.2	<2.2	<0.6	9.7	<0.4	138
46	MM22L806	<1.4	<1.6	<0.2	<2.6	<0.7	4.1	<0.5	23.1
47	MM22L800	<1.8	<2.1	<0.3	<3.4	<0.9	7.0	<0.7	31.6
48	MM22L834	<1.5	<1.7	<0.2	<2.8	<0.7	5.8	<0.6	17.3
52	MM22L897	<1.2	<1.4	<0.2	<2.2	<0.6	6.0	<0.4	122
53	MM22L900	<1.3	<1.5	<0.2	<2.5	<0.6	7.9	<0.5	22.2
54	MM22L903	<2.5	<2.8	<0.4	<4.6	<1.2	3.0	<0.9	38.0
55	MM22L906	<2.6	<3.0	<0.4	<4.9	<1.3	7.9	<1.0	16.8
56	MM22L909	<1.4	<1.6	<0.2	<2.7	<0.7	4.0	<0.5	70.4
57	MM22L912	<1.2	<1.3	<0.2	<2.2	<0.6	19.8	<0.4	17.2
58	MM22L915	<1.4	<1.6	<0.2	<2.7	<0.7	6.0	<0.5	90.9
59	MM22L918	<1.3	<1.5	<0.2	<2.4	<0.6	8.2	<0.5	70.3
60	MM22L921	<2.2	<2.6	<0.3	<4.2	<1.1	5.5	<0.8	29.9
61	MM22L924	<2.0	<2.3	<0.3	<3.8	<1.0	15.5	<0.8	66.9

PFBA= perfluorobutanoic acid, PFPeA = perfluoropentanoic acid, PFHxA = perfluorohexanoic acid, PFOA = perfluorooctanoic acid, PFNA = perfluorononanoic acid, PFDA = perfluorodecanoic acid, PFUnA = perfluoroundecanoic acid

Table 3 (cont.): Perfluorinated alkyl acids (ng/g wet mass) in northern fur seal liver

FRSL-	Storage ID	PFD _o A	PFTriA	PFTA	PFBS	PFHxS	PFOS	PFOSA
1	MM1L001	<0.4	0.77	<0.3	<0.1	0.11	2.9	0.73
2	MM1L005	<0.4	0.76	<0.3	<0.2	0.12	1.7	1.6
3	MM1L009	2.2	1.8	0.97	<0.2	0.47	3.4	1.6
4	MM1L013	<0.2	1.8	<0.3	<0.1	0.14	4.9	<0.2
5	missing	--	--	--	--	--	--	--
6	MM4L095	<0.2	0.82	<0.2	<0.1	0.18	3.6	0.82
7	MM4L098	<0.3	2.5	<0.5	<0.2	0.26	7.7	<0.3
8	MM4L101	<0.6	1.4	<0.4	<0.2	0.19	3.1	0.57
9	MM4L104	<0.7	1.8	0.72	<0.3	0.26	5.2	1.5
10	MM4L107	<0.4	0.81	<0.7	<0.2	0.20	4.0	0.63
11	MM4L110	<0.2	9.1	<0.4	<0.1	<0.1	4.7	0.99
12	MM4L113	<0.4	2.2	<0.3	<0.2	0.40	8.0	1.4
13	MM4L116	<0.5	1.9	<0.3	0.62	0.08	1.1	0.08
14	MM4L119	<0.5	1.2	<0.4	<0.2	0.20	3.8	1.2
15	MM4L122	<0.5	1.8	<0.3	<0.2	0.12	3.0	0.70
16	MM12L646	2.8	3.0	1.3	<0.1	0.19	5.4	3.2
17	MM12L650	<0.6	2.4	0.79	<0.2	0.25	4.1	2.4
18	MM12L654	<0.4	1.7	<0.3	<0.2	0.24	8.1	1.2
19	MM12L658	<0.4	2.4	<0.3	<0.2	0.11	7.2	0.98
20	MM12L662	1.4	4.7	0.81	<0.1	0.36	13.6	2.8
22	MM16L167	<0.3	<0.3	<0.6	<0.2	<0.2	2.9	<0.3
23	MM16L170	<0.4	2.2	<0.7	<0.2	<0.2	3.1	<0.3
24	MM16L173	<0.3	2.7	<0.5	<0.2	0.67	15.3	<0.2
25	MM16L176	<0.3	4.7	<0.4	<0.2	0.30	3.1	3.4
26	MM16L179	<0.4	5.4	<0.7	<0.2	<0.2	2.2	1.0
27	MM16L182	<0.2	6.6	<0.4	<0.1	0.21	1.7	4.0
28	MM16L185	<0.2	1.2	<0.3	<0.1	<0.1	0.93	<0.2
29	MM16L188	<0.6	<0.5	<1.0	<0.3	0.31	4.6	1.1
30	MM16L191	<0.3	0.32	<0.5	<0.2	0.25	3.4	<0.3
31	MM16L194	<0.3	1.0	<0.5	<0.2	<0.2	2.5	0.86
38	MM22L818	<0.2	0.67	<0.3	<0.1	0.22	3.6	<0.2
39	MM22L820	<0.3	24.0	<0.4	<0.2	0.22	2.4	0.70
40	MM22L823	<0.2	2.62	<0.4	<0.1	0.20	2.7	0.23
41	MM22L826	<0.3	25.9	<0.5	<0.2	0.41	17.5	<0.2
42	MM22L829	<0.2	0.26	<0.4	<0.1	<0.1	5.6	<0.2
44	MM22L794	<0.2	<0.2	<0.4	<0.2	<0.1	1.3	<0.2
45	MM22L797	<0.2	11.4	<0.4	<0.1	<0.1	4.2	0.36
46	MM22L806	<0.3	5.4	<0.4	<0.2	0.46	8.8	<0.2
47	MM22L800	<0.3	5.4	<0.6	<0.2	<0.2	2.1	0.99
48	MM22L834	<0.3	4.2	<0.5	<0.2	<0.2	2.6	0.47
52	MM22L897	<0.2	1.6	<0.4	<0.1	0.24	7.7	<0.2
53	MM22L900	<0.3	9.1	<0.4	<0.1	0.36	3.9	1.47
54	MM22L903	<0.5	6.5	<0.8	<0.3	<0.3	2.7	0.48
55	MM22L906	<0.5	2.2	<0.8	<0.3	<0.3	10.0	0.78
56	MM22L909	<0.3	13.8	<0.4	<0.2	0.80	15.8	0.67
57	MM22L912	<0.2	1.6	<0.4	<0.1	0.33	3.3	0.42
58	MM22L915	<0.3	4.8	<0.5	<0.2	0.43	8.4	0.74
59	MM22L918	<0.2	4.7	<0.4	<0.1	<0.1	6.09	<0.2
60	MM22L921	<0.4	2.7	<0.7	<0.2	1.6	14.4	<0.3
61	MM22L924	<0.4	1.6	<0.6	<0.2	0.37	14.4	0.32

PFD_oA = perfluorododecanoic acid, PFTriA = perfluorotridecanoic acid, PFTA = perfluorotetradecanoic acid, PFHxS = perfluorohexansulfonate, PFOS = perfluorooctane sulfonate, PFOSA = perfluorooctanesulfonamide

Table 4: Mass fraction of vitamins (mg/kg wet mass) determined in the liver control material

Sample	Σ Retinol	α -Tocopherol	γ -Tocopherol
QC97LH02-1	151	12700	1600
QC97LH02-1	144	8280	1580
QC97LH02-1	123	7340	1370
Mean	139	9430	1520
1 SD	15	2850	131
RSD (%)	10	30	9

Table 5: Mass fraction of vitamins (mg/kg wet mass) determined in northern fur seal livers

FRSL-	Storage#	Σ Retinol	α -Tocopherol	γ -Tocopherol
1	MM1L001	300	14000	801
2	MM1L005	27.5	15900	735
3	MM1L009	157	7800	1310
4	MM1L013	52.4	4790	657
5	missing	--	--	--
6	MM4L095	43.9	9780	618
7	MM4L098	21.7	12800	nd
8	MM4L101	34.8	14700	nd
9	MM4L104	54.1	12600	1290
10	MM4L107	116	12700	902
11	MM4L110	12.3	10400	853
12	MM4L113	53.0	9370	2010
13	MM4L116	63.3	3000	685
14	MM4L119	61.6	10300	597
15	MM4L122	60.2	9140	1720
16	MM12L646	11.5	nd	nd
17	MM12L650	671	15300	<LOD
18	MM12L654	69.3	5680	571
19	MM12L658	59.9	13500	nd
20	MM12L662	39.6	6340	nd
22	MM16L167	409	11300	nd
23	MM16L170	98.3	11700	nd
24	MM16L173	12.1	5710	nd
25	MM16L176	165	14600	594
26	MM16L179	600	15400	nd
27	MM16L182	30.6	5260	nd
28	MM16L185	248	17700	nd
29	MM16L188	525	13600	nd
30	MM16L191	11.3	3650	736
31	MM16L194	251	11600	nd
38	MM22L818	21.9	6190	1600
39	MM22L820	21.8	2950	nd
40	MM22L823	26.4	10300	nd
41	MM22L826	11.1	4970	1280
42	MM22L829	12.9	nd	nd
44	MM22L794	14.4	11300	1200
45	MM22L797	98.2	16700	787
46	MM22L806	203	15200	578
47	MM22L800	38.8	5850	527
48	MM22L834	106	nd	nd
52	MM22L897	147	23400	729
53	MM22L900	20.2	nd	nd
54	MM22L903	13.0	9390	501
55	MM22L906	82.8	6740	741
56	MM22L909	44.9	10000	1220
57	MM22L912	4.80	nd	nd
58	MM22L915	75.6	7790	710
59	MM22L918	654	10900	1050
60	MM22L921	219	7820	996
61	MM22L924	134	nd	nd

nd: not detected

Table 6: Mass fraction (mg/kg-wet mass) of vitamins in northern fur seal livers by rookery

Rookery	n	Mean Σ Retinol	Mean α -Tocopherol	Mean γ -Tocopherol	Stdev Σ Retinol	Stdev α -Tocopherol	Stdev γ -Tocopherol
Northeast	1	52.4	4790	657			
Polavina	23	184	11200	843	195	4800	246
Reef	16	57.6	9160	1060	52.0	4180	497
Zapadni	9	112	11400	826	210	3700	402

Table 7: Mass fractions (ng/g wet mass) of compounds determined in aliquots of SRM 1945 used as a control material in this study. Italicized bold values are reference values. Reference values differ from certified values in that all sources of uncertainty have not been accounted for during certification.

Compound	SRM 1945 Aliquot						Certified or Reference Value
	1	2	3	4	mean	SD	
pentachlorobenzene	2.3	2.4	2.5	2.8	2.5	0.2	<i>2.09 ± 0.53</i>
α-HCH	20.5	19.3	20.5	20.2	20.1	0.6	16.9 ± 1.4
HCB	28.4	31.6	31.9	32.3	31.0	1.8	30.6 ± 1.5
γ-HCH	2.9	2.6	2.7	2.8	2.8	0.1	3.18 ± 0.01
oxychlordane	21.0	20.6	19.9	20.3	20.5	0.5	21.2 ± 1.1
trans-chlordane	20.3	16.8	16.9	17.2	17.8	1.7	11.8 ± 0.5
cis-chlordane	50.4	52.0	51.0	56.1	52.4	2.6	48.1 ± 1.6
trans-nonachlor	182	181	183	182	182	0.8	198 ± 16
cis-nonachlor	44.9	54.6	62.0	44.6	51.5	8.4	45.8 ± 3.3
Parlar 26	86.0	73.9	72.8	81.7	78.6	6.3	<i>41.8 ± 2.8</i>
Parlar 32	7.3	8.0	8.3	8.5	8.0	0.5	<i>no value</i>
Parlar 50	139	107	108	115	117	15	<i>69.7 ± 5.5</i>
Parlar 62	77.2	48.6	48.9	52.4	56.8	14	<i>37.2 ± 3.8</i>
PBDE 28	1.4	2.2	1.8	2.0	1.8	0.3	<i>2.66 ± 0.37</i>
PBDE 47	42.3	38.0	41.1	36.9	39.6	2.5	39.6 ± 0.2
PBDE 100	10.0	10.9	10.3	11.8	10.8	0.8	10.3 ± 1.1
PBDE 99	17.9	17.7	17.2	16.7	17.4	0.52	18.9 ± 2.3
PBDE 155	3.3	2.5	2.5	2.5	2.7	0.4	<i>4.45 ± 0.93</i>
PBDE 154	18.2	13.3	14.5	13.9	15.0	2.2	13.3 ± 1.7
PBDE 153	8.5	6.2	6.7	6.1	6.9	1.1	8.34 ± 0.55
α-HBCD	5.9	7.0	5.7	5.6	6.1	0.6	<i>no value</i>

Table 8: Concentrations (ng/g wet mass) of polychlorinated biphenyls congeners determined in aliquots of SRM 1945 used as a control material in this study. Italicized bold values are reference values. Reference values differ from certified values in that all sources of uncertainty have not been accounted for during certification.

Congener	SRM 1945 Aliquot						Certified or Reference Values
	1	2	3	4	Mean	SD	
PCB 18	4.3	4.4	4.3	4.7	4.4	0.2	4.35 ± 0.56
PCB 31/28	16.1	17.2	15.9	16.6	16.4	0.6	16.7
PCB 52	42.3	41.7	41.9	41.4	41.8	0.4	40.7 ± 1.3
PCB 49	17.8	17.0	18.8	18.4	18.0	0.8	18.3 ± 0.1
PCB 44	12.8	12.7	12.8	12.9	12.8	0.1	12.1 ± 0.5
PCB 95	31.9	32.8	31.9	34.1	32.7	1.0	33.9 ± 0.5
PCB 74	21.6	22.8	20.8	21.9	21.8	0.8	21.6 ± 7.8
PCB 66	23.1	22.4	21.7	22.0	22.3	0.6	22.4 ± 0.5
PCB 92	22.8	23.3	23.6	23.6	23.3	0.4	23.4 ± 3.9
PCB 56	7.8	7.3	7.9	8.0	7.8	0.3	7.5 ± 1.8
PCB 101	81.5	82.9	81.4	79.3	81.3	1.5	78 ± 12
PCB 99	60.0	62.9	56.9	60.1	60.0	2.5	58.5 ± 5.2
PCB 87	21.8	20.7	21.8	19.1	20.9	1.3	20.6 ± 2.6
PCB 110	34.3	34.7	34.2	34.7	34.5	0.3	33.8 ± 2.3
PCB 82	4.7	4.5	4.7	5.1	4.7	0.2	4.5 ± 1.1
PCB 151	29.4	28.6	28.1	29.1	28.8	0.6	28.6 ± 1.3
PCB 149	90.5	87.2	88.1	86.0	88.0	1.9	89.0 ± 6.9
PCB 118	77.1	73.8	76.5	77.4	76.2	1.6	76.5 ± 2.9
PCB 146	46.0	44.8	44.1	45.5	45.1	0.8	45.1 ± 4.0
PCB 153/132	257	258	258	239	253	9.3	249
PCB 105	28.4	28.8	28.2	28.0	28.4	0.3	28.6 ± 1.2
PCB 176	6.6	6.2	6.0	6.4	6.3	0.2	6.2 ± 1.0
PCB 137	7.76	7.55	7.18	7.7	7.55	0.3	7.7 ± 1.4
PCB 138/163	200	179	181	196	189	11	193
PCB 158	9.8	8.8	9.0	9.1	9.2	0.4	9.1 ± 1.0
PCB 178/175	28.7	24.2	23.4	25.4	25.4	2.3	25
PCB 187	115	124	128	125	123	5.6	121 ± 11
PCB 183	40.8	34.6	38.0	38.7	38.0	2.6	38.0 ± 1.8
PCB 128	23.5	21.6	23.4	23.9	23.1	1.0	23.0 ± 1.1
PCB 185	3.20	3.68	3.58	3.61	3.52	0.2	3.40 ± 0.32
PCB 174	25.8	25.8	24.8	25.4	25.5	0.5	25.2 ± 0.1
PCB 202	29.3	26.9	29.2	28.9	28.6	1.1	29.0 ± 7.0
PCB 177	29.1	27.3	29.1	28.9	28.6	0.9	28.6 ± 1.2
PCB 201	18.0	17.3	17.6	17.6	17.6	0.3	16.8 ± 1.3
PCB 156	11.9	11.3	11.3	11.7	11.6	0.3	11.4 ± 0.9
PCB 172	12.3	11.8	12.4	12.1	12.2	0.3	12.4 ± 3.4
PCB 157	3.4	3.4	3.0	3.4	3.3	0.2	3.27 ± 0.67
PCB 180/193	147	149	145	152	148	3.0	147
PCB 200	4.2	4.8	4.3	4.4	4.4	0.2	4.30 ± 0.32
PCB 191	1.8	1.6	1.6	1.4	1.6	0.2	1.57 ± 0.45
PCB 170	45.5	44.4	41.8	41.8	43.4	1.9	42.6 ± 2.2
PCB 199	85.4	87.9	80.9	85.9	85.0	3.0	84.2 ± 8.0
PCB 196	70.4	80.1	72.9	72.7	74.0	4.2	74.5 ± 8.4
PCB 208	22.1	21.9	23.6	24.6	23.1	1.3	23.9 ± 1.7
PCB 207	14.4	13.3	14.2	13.5	13.9	0.5	14.5 ± 2.4
PCB 195	13.9	13.5	14.5	14.8	14.2	0.6	14.3 ± 2.2
PCB 194	60.3	56.8	54.9	49.9	55.5	4.3	53.5 ± 5.2
PCB 206	43.9	43.4	46.2	41.1	43.7	2.1	44.9 ± 4.2
PCB 209	17.4	17.4	16.9	16.8	17.1	0.3	17.2 ± 1.9

Table 9: Mass fractions of polychlorinated biphenyl congeners (ng/g wet mass) in northern fur seal blubber

FRSL-	Storage ID	PCB 8	PCB 18	PCB 31/28	PCB 52	PCB 49	PCB 44	PCB 95	PCB 121	PCB 74	PCB 66	PCB 92	PCB 56
1	MM1B004	1.89	3.53	10.9	29.6	4.94	14.1	33.4	1.68	43.6	21.8	23.0	2.48
2	MM1B008	<1	2.75	7.73	17.3	3.95	10.8	17.1	<1.2	23.7	12.0	11.9	1.66
3	MM1B012	<1	3.20	11.1	21.1	4.68	12.8	14.6	<1.2	28.0	15.8	15.1	1.45
4	MM1B016	6.34	13.0	20.2	41.2	7.58	21.2	41.6	2.23	34.8	29.7	18.7	6.58
5	MM1B020	5.22	5.53	7.52	21.8	3.94	13.9	19.1	<0.26	22.9	20.6	10.4	3.64
6	MM4B097	1.39	2.89	7.21	21.0	4.22	12.0	20.7	<1.2	29.6	14.5	16.8	1.95
7	MM4B100	<1	2.82	8.17	20.7	3.98	12.1	19.8	<1.2	28.6	14.0	17.4	1.87
8	MM4B103	<1	4.18	11.0	29.3	4.11	13.5	36.3	<1.2	42.5	21.3	22.7	1.90
9	MM4B106	<1	2.59	8.00	24.7	3.39	11.1	25.9	<1.2	35.3	17.3	23.5	2.06
10	MM4B109	1.49	2.46	14.2	33.8	4.99	16.1	38.9	<1.2	58.8	28.4	28.0	2.05
11	MM4B112	<1	3.65	9.77	32.5	3.73	12.4	39.4	<1.2	48.7	25.2	26.5	1.98
12	MM4B115	<1	3.13	13.0	29.3	4.64	14.4	32.1	<1.2	50.1	26.2	25.2	1.78
13	MM4B118	<1	5.10	9.68	41.7	5.62	16.8	34.5	<1.2	35.3	17.9	27.2	2.13
14	MM4B121	<1	1.73	9.18	26.3	4.05	12.0	30.3	<1.2	39.7	19.9	28.3	2.22
15	MM4B124	1.47	2.76	10.5	21.5	3.91	11.9	23.3	<1.2	38.8	20.5	19.2	1.95
16	MM12B648	<1	2.19	8.74	19.5	2.99	10.6	31.9	<1.2	40.7	19.4	19.2	2.07
17	MM12B652	<1	1.79	5.24	17.0	3.40	8.37	17.8	1.35	25.2	12.5	16.2	1.99
18	MM12B656	<1	3.46	12.4	26.5	4.22	13.4	20.7	1.41	37.6	17.1	18.4	1.74
19	MM12B660	<1	3.17	8.98	20.8	3.89	10.8	21.2	<1.2	33.6	15.0	15.4	1.90
20	MM12B664	<1	2.49	5.96	17.3	3.29	9.85	16.9	1.41	28.9	13.5	13.0	1.89
22	MM16B169	0.55	3.76	3.27	34.1	7.41	7.94	38.1	<0.26	30.5	20.3	22.9	3.20
23	MM16B172	5.78	25.1	20.2	13.4	8.36	9.29	34.5	3.44	20.2	29.7	18.8	6.99
24	MM16B175	2.78	<0.58	7.63	21.6	2.69	11.1	23.4	<0.26	41.3	33.0	12.9	2.38
25	MM16B178	3.08	3.04	6.16	26.1	5.06	5.50	35.6	<0.26	37.0	22.5	19.4	<0.55
26	MM16B181	5.16	2.67	3.34	19.4	3.82	3.84	27.9	<0.26	45.9	30.0	15.8	<0.55
27	MM16B184	14.9	17.5	18.2	132	15.4	4.41	23.7	<0.26	28.4	15.2	12.3	<0.55
28	MM16B187	1.55	1.96	5.75	12.2	2.85	7.53	11.7	1.39	22.9	10.6	10.5	1.93
29	MM16B190	2.26	4.37	3.10	30.7	8.80	9.48	41.4	<0.26	34.4	39.6	26.3	6.42
30	MM16B193	2.84	2.57	3.49	11.2	7.27	16.9	16.2	<0.26	30.7	21.4	9.58	<0.55
31	MM16B196	11.4	12.9	14.0	18.6	6.25	7.15	31.6	<0.26	29.4	24.1	9.81	<0.55
38	MM22B819	<0.88	3.51	13.4	50.8	22.7	23.7	9.36	1.27	76.5	58.6	6.28	<0.55
39	MM22B822	2.85	4.54	9.95	15.6	7.74	17.6	14.5	<0.26	19.2	20.7	8.01	<0.55
40	MM22B825	4.44	8.28	8.78	17.2	6.17	16.0	18.5	<0.26	20.9	22.9	13.3	<0.55
41	MM22B828	<1	2.34	5.87	9.76	2.87	7.76	9.18	<1.2	18.5	9.17	8.05	1.99
42	MM22B831	7.75	12.3	15.3	32.6	7.49	18.9	9.24	<0.26	33.1	30.3	6.67	<0.55
44	MM22B796	17.7	10.4	7.20	7.5	5.43	6.34	11.5	2.41	17.9	28.7	13.7	7.25
45	MM22B799	12.8	11.7	13.4	21.3	5.83	11.8	28.6	<0.26	24.2	24.5	14.9	<0.55
46	MM22B808	<1	3.08	9.08	32.6	7.92	16.6	42.2	<1.2	27.0	20.2	22.3	3.20
47	MM22B802	2.26	2.39	4.08	17.6	4.98	7.00	17.7	<0.26	32.1	24.1	9.96	<0.55
48	MM22B836	1.84	8.22	4.58	56.2	10.6	30.9	13.5	<0.26	45.3	48.4	9.65	<0.55
52	MM22B899	2.70	3.45	8.23	19.9	2.58	12.1	24.5	<0.26	22.1	17.7	14.6	<0.55
53	MM22B902	1.57	2.76	9.65	25.7	6.69	6.78	31.7	<0.26	20.1	19.6	19.2	1.98
54	MM22B905	<1	2.21	7.93	14.4	3.48	9.42	15.2	<1.2	25.9	13.2	12.9	2.17
55	MM22B908	<1	1.92	14.5	14.6	2.62	7.77	14.6	1.49	39.4	18.4	17.7	1.42
56	MM22B911	<1	2.08	7.10	10.4	2.85	7.75	9.51	<1.2	20.9	9.64	8.67	1.99
57	MM22B914	<1	2.11	8.74	12.5	2.60	7.61	12.7	1.37	26.5	12.4	10.9	2.01
58	MM22B917	<1	2.15	7.94	11.4	3.01	7.71	10.7	<1.2	24.4	11.7	8.9	2.00
59	MM22B920	<1	2.48	12.3	23.2	4.01	11.1	17.1	<1.2	32.7	16.5	16.6	2.07
60	MM22B923	<1	2.23	16.9	27.5	3.80	12.3	23.9	1.73	51.1	27.2	24.5	2.52
61	MM22B926	<1	2.91	19.6	45.3	3.87	15.1	27.4	1.19	70.3	36.1	32.0	2.07

Table 9 (cont.): Mass fractions of polychlorinated biphenyl congeners (ng/g wet mass) in northern fur seal blubber

FRSL -	Storage ID	PCB 101	PCB 99	PCB 119/112	PCB 79	PCB 87	PCB 110	PCB 151	PCB 149	PCB 118	PCB 146	PCB 114	PCB 153/132
1	MM1B004	38.6	80.0	13.9	2.41	12.8	8.61	2.43	87.1	106	40.8	5.39	433
2	MM1B008	22.8	39.8	5.66	<2	9.09	6.05	5.90	43.9	52.6	25.7	4.12	179
3	MM1B012	32.8	42.1	6.31	<2	11.3	8.10	4.25	45.0	62.1	25.8	3.81	189
4	MM1B016	39.5	71.8	6.55	2.88	9.39	27.4	6.72	115.0	55.3	27.9	2.57	481
5	MM1B020	14.1	57.1	1.12	2.19	5.87	14.6	1.04	46.7	84.6	14.3	3.34	276
6	MM4B097	28.0	45.6	6.54	<2	10.1	7.04	7.79	60.9	67.0	30.8	4.61	190
7	MM4B100	29.9	46.2	6.28	<2	11.1	7.72	9.57	78.7	70.1	34.5	4.40	264
8	MM4B103	30.9	65.7	9.82	<2	10.9	7.58	12.9	90.0	95.7	31.9	4.78	317
9	MM4B106	25.5	83.8	13.6	<2	9.98	6.19	11.9	87.6	113	33.2	4.95	456
10	MM4B109	45.8	92.5	13.3	<2	17.7	10.6	15.1	112	137	38.9	5.81	460
11	MM4B112	28.3	79.6	12.2	<2	11.0	6.55	12.9	83.6	122	30.7	5.25	389
12	MM4B115	40.6	86.8	12.5	<2	15.5	9.85	12.8	92.8	132	36.8	5.74	439
13	MM4B118	41.2	61.3	8.72	<2	13.6	7.82	15.5	104	87.3	40.9	4.51	304
14	MM4B121	32.0	101	13.5	<2	10.8	7.16	17.5	83.2	127	44.4	5.99	535
15	MM4B124	29.8	84.2	11.6	<2	10.8	7.58	9.57	78.7	116	33.1	5.42	491
16	MM12B64	24.9	95.1	14.2	<2	7.60	6.04	9.13	59.3	128	27.5	5.56	445
17	MM12B65	22.5	46.7	7.19	<2	7.26	5.28	8.70	56.2	69.9	28.9	4.06	225
18	MM12B65	30.0	43.4	6.97	<2	9.05	7.87	9.31	69.5	67.5	32.5	4.55	216
19	MM12B66	22.7	56.8	8.81	<2	8.00	6.37	8.10	54.3	84.1	28.0	4.36	318
20	MM12B66	19.9	44.5	7.38	<2	6.36	5.30	6.89	45.9	70.0	27.1	4.27	235
22	MM16B16	48.8	76.5	3.91	<1.35	12.7	37.5	5.91	112	146	31.0	8.06	430
23	MM16B17	58.1	60.6	14.6	5.40	16.8	50.4	8.92	147	123	36.6	3.32	475
24	MM16B17	23.1	61.7	9.67	<1.35	7.83	12.7	0.81	66.7	106	19.6	3.06	334
25	MM16B17	39.2	100	5.41	<1.35	8.12	31.4	6.54	96.4	157	24.5	5.05	518
26	MM16B18	45.0	82.5	3.02	4.85	12.8	20.2	44.8	83.8	127	33.1	<1.53	407
27	MM16B18	31.5	73.5	5.47	<1.35	8.59	36.6	55.3	69.7	79.6	50.8	<1.53	389
28	MM16B18	13.4	27.6	4.13	<2	5.83	3.81	5.76	35.4	43.4	25.8	3.76	141
29	MM16B19	55.0	110	4.38	<1.35	16.8	46.3	6.53	104	178	28.2	4.90	514
30	MM16B19	19.3	45.1	1.94	<1.35	4.69	11.6	2.92	71.7	80.3	19.3	8.11	354
31	MM16B19	42.8	56.4	6.46	<1.35	4.84	9.95	3.02	88.1	78.9	22.9	<1.53	489
38	MM22B81	18.2	11.7	1.01	<1.35	3.94	9.01	8.71	140	44.1	43.0	3.06	406
39	MM22B82	26.2	34.8	3.38	<1.35	12.9	17.2	1.70	59.3	105	16.6	13.6	220
40	MM22B82	31.7	54.6	3.35	<1.35	19.4	56.3	4.00	96.3	99.4	32.7	17.1	406
41	MM22B82	13.8	21.9	3.59	<2	4.27	4.07	4.27	30.4	32.8	24.2	3.50	104
42	MM22B83	14.6	21.0	0.96	<1.35	3.38	11.2	8.14	137	39.2	33.8	1.91	496
44	MM22B79	35.2	36.4	8.11	<1.35	7.50	8.30	4.79	87.3	77.4	26.0	<1.53	319
45	MM22B79	58.7	53.8	2.72	<1.35	10.0	38.2	15.5	230	117	54.7	<1.53	71.1
46	MM22B80	46.9	85.3	3.47	2.93	12.8	33.7	4.14	96.7	132	25.7	2.57	446
47	MM22B80	21.3	53.7	1.48	<1.35	3.86	15.4	15.5	224	98.2	52.7	<1.53	83
48	MM22B83	19.9	54.2	0.90	<1.35	6.88	14.0	1.87	29.2	84.7	2.1	4.78	199
52	MM22B89	23.2	75.0	1.82	<1.35	3.90	13.9	2.68	69.4	117	17.5	5.40	377
53	MM22B90	40.0	83.2	4.08	<1.35	12.1	32.1	7.22	96.3	130	23.9	4.02	536
54	MM22B90	18.2	40.2	6.28	<2	6.06	4.70	7.20	47.0	64.2	27.1	4.22	258
55	MM22B90	22.9	80.3	12.3	<2	8.45	5.66	4.33	52.0	114	28.3	5.45	392
56	MM22B91	14.6	25.9	4.16	<2	5.30	4.41	4.76	33.3	40.6	25.0	4.00	149
57	MM22B91	13.7	33.6	5.38	<2	4.00	3.74	5.67	38.6	52.9	25.2	3.92	186
58	MM22B91	15.6	36.4	5.88	<2	5.02	4.15	4.97	31.2	50.8	24.0	4.01	217
59	MM22B92	31.8	41.6	6.67	<2	9.75	7.37	8.28	61.5	64.0	32.7	4.06	235
60	MM22B92	34.2	76.5	11.6	<2	10.3	8.68	12.2	83.9	117	39.3	5.52	409
61	MM22B92	37.2	88.1	12.8	<2	11.9	8.05	15.0	105	138	38.2	5.45	405

Table 9 (cont.): Mass fractions of polychlorinated biphenyl congeners (ng/g wet mass) in northern fur seal blubber

FRSL	Storage ID	PCB 105	PCB 176	PCB 127	PCB 137	PCB 138/163	PCB 158	PCB 178/175	PCB 197	PCB 187	PCB 183	PCB 159	PCB 128
1	MM1B004	43.5	4.49	2.88	14.6	215	15.4	8.54	5.57	61.9	38.4	3.27	31.5
2	MM1B008	21.9	2.78	<2	7.29	88.8	7.52	6.48	1.41	33.6	17.2	2.70	13.7
3	MM1B012	24.0	2.43	<2	7.15	93.8	7.76	6.04	1.90	39.8	17.1	2.47	13.9
4	MM1B016	20.6	5.39	<0.39	13.2	291	29.3	5.09	<0.51	44.3	23.6	<0.82	28.9
5	MM1B020	14.1	<0.23	<0.39	8.36	171	10.1	3.92	<0.51	79.3	62.4	<0.82	19.9
6	MM4B097	26.7	3.13	2.35	8.05	101	8.47	6.91	0.95	44.9	22.1	2.85	15.9
7	MM4B100	26.3	3.36	2.40	9.85	132	9.34	6.99	3.38	61.8	30.7	2.77	19.1
8	MM4B103	35.8	3.85	2.02	11.3	160	10.9	7.11	4.70	62.3	34.9	2.68	22.5
9	MM4B106	44.3	3.83	2.36	14.3	227	14.7	7.75	5.24	66.3	38.6	2.94	29.8
10	MM4B109	51.5	4.19	2.65	15.3	226	14.8	8.24	4.32	77.2	44.0	2.99	31.3
11	MM4B112	44.4	4.21	2.50	12.7	189	12.5	8.00	4.44	57.6	42.9	2.87	25.2
12	MM4B115	46.5	3.54	2.58	14.6	217	13.8	7.22	5.41	64.7	38.7	3.01	28.5
13	MM4B118	33.0	4.14	2.69	11.0	155	10.6	7.85	2.97	83.2	37.2	2.79	20.8
14	MM4B121	54.5	3.85	3.00	17.7	270	17.7	9.12	7.91	91.4	40.3	3.42	38.2
15	MM4B124	44.1	3.36	2.79	16.5	244	15.7	8.36	6.45	58.6	32.9	3.32	33.8
16	MM12B64	45.8	3.68	2.57	14.6	208	13.1	8.03	5.40	46.4	29.6	3.01	27.7
17	MM12B65	25.9	2.93	2.35	8.07	146	8.86	6.83	2.13	42.0	23.3	2.68	17.1
18	MM12B65	23.4	3.22	2.29	8.23	103	7.94	6.79	2.18	54.9	27.8	2.69	15.6
19	MM12B66	29.9	3.27	2.27	10.6	147	10.6	7.24	5.59	45.2	26.4	2.89	21.6
20	MM12B66	25.1	3.09	<2	8.92	113	8.83	6.72	1.19	37.8	20.8	2.64	18.0
22	MM16B16	36.1	10.0	<0.39	11.7	263	11.2	5.25	<0.51	88.5	50.4	<0.82	23.6
23	MM16B17	33.1	3.42	<0.39	7.87	271	27.7	9.66	<0.51	38.2	20.6	<0.82	17.7
24	MM16B17	25.6	<0.23	<0.39	7.00	194	12.4	9.04	<0.51	51.4	40.1	<0.82	18.7
25	MM16B17	9.01	5.05	<0.39	16.6	316	31.1	17.6	<0.51	44.8	33.5	<0.82	32.5
26	MM16B18	27.3	6.00	<0.39	1.40	261	1.48	1.68	<0.51	50.1	39.2	<0.82	25.7
27	MM16B18	25.7	<0.23	<0.39	22.1	400	13.1	11.5	<0.51	58.4	49.2	<0.82	21.5
28	MM16B18	16.8	2.95	<2	6.25	66.8	6.6	6.46	1.00	35.6	17.8	2.74	11.5
29	MM16B19	45.0	9.73	<0.39	38.6	312	22.1	27.6	<0.51	80.8	57.5	<0.82	28.7
30	MM16B19	22.3	3.25	<0.39	10.4	213	10.1	2.33	<0.51	32.7	19.9	<0.82	19.2
31	MM16B19	23.6	<0.23	<0.39	8.86	273	11.1	5.99	<0.51	35.3	22.1	<0.82	19.0
38	MM22B81	12.6	<0.23	<0.39	36.9	222	28.6	15.9	<0.51	53.5	24.7	<0.82	14.9
39	MM22B82	20.1	<0.23	<0.39	32.0	121	10.1	12.1	<0.51	51.1	43.2	<0.82	10.7
40	MM22B82	28.3	7.35	<0.39	11.8	236	23.2	17.2	<0.51	62.5	35.0	<0.82	25.4
41	MM22B82	13.2	2.80	<2	5.02	49.7	5.22	6.75	0.70	32.9	15.5	2.62	9.06
42	MM22B83	5.25	14.8	<0.39	61.8	296	18.5	25.8	<0.51	115	58.0	<0.82	21.0
44	MM22B79	19.0	4.56	<0.39	8.90	183	23.1	11.3	<0.51	51.9	26.3	<0.82	16.7
45	MM22B79	33.9	4.24	<0.39	9.08	386	36.4	88.2	<0.51	44.8	18.7	<0.82	18.7
46	MM22B80	35.3	15.4	<2	32.5	253	20.9	34.6	0.56	114	77.4	<2	22.5
47	MM22B80	27.8	8.19	<0.39	5.82	325	20.7	24.1	<0.51	77.6	51.2	<0.82	11.1
48	MM22B83	17.7	<0.23	<0.39	18.2	116	2.48	30.5	<0.51	169	99.7	<0.82	2.26
52	MM22B89	32.4	8.53	<0.39	28.7	216	11.1	16.0	<0.51	65.8	27.7	<0.82	20.7
53	MM22B90	32.6	0.61	<0.39	38.3	293	24.1	4.92	<0.51	8.08	6.42	<0.82	10.8
54	MM22B90	22.8	3.12	<2	8.12	106	8.58	7.33	3.29	41.2	25.3	2.84	16.8
55	MM22B90	38.2	2.80	2.53	12.2	178	11.5	7.29	4.36	50.7	20.1	2.92	23.6
56	MM22B91	15.5	3.14	<2	6.12	66.7	6.83	6.73	1.97	36.6	15.4	2.62	12.3
57	MM22B91	19.5	2.96	2.21	7.18	86.9	7.09	6.52	2.70	36.3	20.7	2.61	13.9
58	MM22B91	18.5	2.87	<2	7.69	96.2	7.57	7.05	1.78	36.6	16.4	2.76	14.7
59	MM22B92	21.4	3.43	2.19	8.16	104	7.81	6.36	2.31	58.8	23.4	2.58	15.3
60	MM22B92	39.8	3.80	2.70	12.7	185	12.9	8.70	5.07	72.5	36.7	3.03	24.5
61	MM22B92	48.9	3.62	<2	12.6	200	12.6	6.65	4.32	72.0	38.2	2.49	27.8

Table 9 (cont.): Mass fractions of polychlorinated biphenyl congeners (ng/g wet mass) in northern fur seal blubber

FRSL -	Storage ID	PCB 185	PCB 174	PCB 167	PCB 202	PCB 177	PCB 201	PCB 156	PCB 172	PCB 157	PCB 180/193	PCB 200	PCB 170
1	MM1B004	16.9	5.24	9.06	<2	6.15	2.30	11.83	5.20	5.66	219	3.28	44.1
2	MM1B008	13.1	2.71	3.98	<2	3.97	<1	4.73	3.10	3.32	71.6	<1.5	15.0
3	MM1B012	13.5	2.90	4.43	3.03	3.94	<1	4.89	3.30	3.30	81.1	<1.5	16.5
4	MM1B016	3.73	2.64	12.4	10.2	10.5	7.39	17.8	8.85	12.2	64.4	2.66	4.43
5	MM1B020	<0.45	2.32	5.62	<1.86	<0.28	<0.51	11.3	<0.30	<1.37	30.5	<1.75	7.79
6	MM4B097	12.8	4.18	5.54	3.03	5.33	<1	6.31	4.02	3.93	73.5	<1.5	16.7
7	MM4B100	20.3	5.48	5.61	3.66	6.35	1.40	6.07	4.35	4.10	131	<1.5	26.0
8	MM4B103	23.8	5.15	7.56	3.98	5.91	1.15	7.64	4.42	4.23	146	2.27	28.2
9	MM4B106	31.4	5.90	7.84	4.61	6.68	2.24	9.87	5.20	4.87	201	<1.5	38.6
10	MM4B109	30.8	6.31	10.1	4.32	7.07	1.56	8.94	5.05	5.26	189	<1.5	35.6
11	MM4B112	27.2	5.67	8.51	<2	6.45	<1	8.94	4.75	4.80	162	<1.5	32.8
12	MM4B115	29.9	5.12	9.19	4.54	6.01	2.32	9.00	4.58	4.96	182	<1.5	34.6
13	MM4B118	21.9	5.09	6.15	4.12	6.08	<1	7.09	5.44	4.39	128	<1.5	26.0
14	MM4B121	40.4	6.17	7.64	5.17	7.12	2.88	14.2	5.77	6.11	265	<1.5	51.5
15	MM4B124	33.4	5.04	7.80	3.99	6.04	<1	9.05	4.28	5.57	215	<1.5	44.7
16	MM12B64	30.4	4.22	8.41	4.00	5.23	3.05	8.72	4.15	4.99	183	<1.5	36.6
17	MM12B65	13.9	3.38	5.23	2.99	4.56	<1	5.49	3.55	3.84	88.4	<1.5	18.8
18	MM12B65	14.7	4.20	5.44	3.29	6.25	<1	4.62	4.16	3.58	80.3	<1.5	17.0
19	MM12B66	24.4	3.97	7.03	3.98	4.99	2.05	6.42	4.05	4.21	156	<1.5	29.7
20	MM12B66	15.2	3.79	5.37	<2	4.87	<1	5.93	3.38	3.93	80.7	<1.5	1.64
22	MM16B16	14.1	49.5	12.7	13.5	22.2	6.88	13.1	6.92	<1.37	135	7.37	30.3
23	MM16B17	4.51	22.1	10.7	17.1	31.0	6.98	14.3	2.28	6.93	48.6	<1.75	11.4
24	MM16B17	<0.45	3.87	3.65	<1.86	<0.28	<0.51	10.4	<0.30	<1.37	11.2	<1.75	4.32
25	MM16B17	<0.45	72.7	15.5	18.7	10.7	4.76	18.2	2.62	<1.37	88.1	33.6	26.7
26	MM16B18	10.47	32.0	8.93	15.2	<0.28	<0.51	13.1	<0.30	4.94	101	<1.75	25.6
27	MM16B18	<0.45	1.65	3.53	<1.86	<0.28	<0.51	9.43	<0.30	<1.37	82.4	<1.75	23.0
28	MM16B18	10.5	2.67	4.73	<2	3.96	<1	4.46	3.33	3.62	62.8	<1.5	13.6
29	MM16B19	<0.45	16.3	3.66	14.0	22.1	13.2	3.89	<0.30	6.57	151	3.71	38.2
30	MM16B19	<0.45	54.5	9.04	11.0	7.83	4.86	17.4	<0.30	<1.37	52.7	<1.75	14.8
31	MM16B19	<0.45	10.2	5.98	<1.86	<0.28	<0.51	13.3	<0.30	<1.37	41.1	<1.75	19.1
38	MM22B81	4.44	25.4	15.32	8.91	11.3	18.5	13.4	<0.30	<1.37	59.3	<1.75	14.9
39	MM22B82	17.1	3.36	3.12	<1.86	<0.28	<0.51	3.06	<0.30	<1.37	109	<1.75	38.9
40	MM22B82	<0.45	3.59	5.96	<1.86	<0.28	<0.51	5.84	<0.30	<1.37	148	<1.75	59.6
41	MM22B82	8.33	2.59	3.82	<2	3.90	<1	3.45	3.04	3.17	40.7	<1.5	10.5
42	MM22B83	<0.45	19.3	10.2	<1.86	37.2	18.8	14.1	<0.30	<1.37	153	13.8	35.4
44	MM22B79	<0.45	12.0	10.2	9.35	12.7	6.36	12.9	6.64	7.31	72.7	<1.75	41.7
45	MM22B79	<0.45	38.9	14.9	24.7	9.44	14.7	17.0	27.15	7.44	193	28.1	65.6
46	MM22B80	19.5	20.2	3.25	11.6	31.3	4.11	13.1	8.68	3.54	183	<1.5	43.6
47	MM22B80	<0.45	44.7	20.9	12.9	21.5	6.46	8.88	11.0	<1.37	130	<1.75	28.1
48	MM22B83	<0.45	10.1	1.88	<1.86	<0.28	<0.51	1.50	<0.30	<1.37	192	<1.75	48.4
52	MM22B89	<0.45	15.4	7.81	16.6	9.51	5.68	4.93	<0.30	<1.37	101	<1.75	62.6
53	MM22B90	4.83	4.69	3.60	14.0	1.90	10.0	8.41	<0.30	<1.37	32.8	<1.75	4.83
54	MM22B90	19.2	4.19	5.86	3.64	5.30	<1	5.94	4.61	4.17	125	<1.5	26.6
55	MM22B90	22.0	3.77	8.02	3.86	4.88	<1	7.61	4.00	4.64	142	<1.5	28.9
56	MM22B91	10.8	3.36	5.64	<2	4.53	<1	4.37	3.62	3.80	59.2	<1.5	15.1
57	MM22B91	13.4	3.29	5.55	<2	4.42	<1	5.01	3.28	3.77	78.0	<1.5	16.5
58	MM22B91	15.6	2.61	4.54	<2	3.94	<1	4.11	3.33	3.79	87.2	<1.5	18.6
59	MM22B92	15.8	3.99	6.05	<2	4.93	<1	4.75	3.63	3.79	87.3	<1.5	18.5
60	MM22B92	25.2	6.28	8.75	3.82	7.21	<1	6.86	5.06	4.65	140	<1.5	29.6
61	MM22B92	23.8	6.45	9.91	3.42	6.91	1.59	8.98	4.07	4.59	145	<1.5	31.2

Table 9 (cont.): Mass fractions of polychlorinated biphenyl congeners (ng/g wet mass) in northern fur seal blubber

FRSL-	Storage ID	PCB 191	PCB 199	PCB 196	PCB 208	PCB 207	PCB 195	PCB 194	PCB 205	PCB 206	PCB 209
1	MM1B004	1.92	16.0	19.1	4.32	2.19	4.73	13.5	<2.7	4.52	5.33
2	MM1B008	<1	5.34	8.86	<1	<1.2	<1.5	5.82	<2.7	<1	<3.4
3	MM1B012	1.27	6.51	9.82	<1	<1.2	<1.5	6.38	<2.7	<1	<3.4
4	MM1B016	<0.66	17.5	10.1	3.32	5.75	13.4	26.8	4.44	2.99	6.65
5	MM1B020	<0.66	13.5	5.42	<1.26	<0.59	<0.57	<0.14	<0.29	<0.48	<0.90
6	MM4B097	0.97	6.55	9.75	1.77	1.67	<1.5	5.50	<2.7	1.75	<3.4
7	MM4B100	1.74	10.6	12.6	2.49	1.80	2.76	8.29	<2.7	2.56	4.71
8	MM4B103	1.78	11.7	14.0	2.48	1.78	3.12	9.36	<2.7	3.79	<3.4
9	MM4B106	1.26	14.2	15.9	2.82	1.61	3.53	9.96	<2.7	3.00	<3.4
10	MM4B109	1.41	12.1	13.5	1.96	2.04	3.39	9.39	<2.7	2.71	<3.4
11	MM4B112	1.18	11.7	13.0	2.14	1.90	<1.5	8.98	<2.7	2.50	<3.4
12	MM4B115	1.68	12.7	16.2	3.53	2.62	1.93	9.00	<2.7	3.44	<3.4
13	MM4B118	1.52	8.82	12.3	2.61	2.16	<1.5	7.56	<2.7	2.39	<3.4
14	MM4B121	1.89	19.2	21.4	3.90	2.78	5.05	13.9	<2.7	4.97	<3.4
15	MM4B124	1.29	13.2	14.7	3.06	2.20	3.55	9.45	<2.7	3.07	<3.4
16	MM12B648	2.07	12.3	15.8	3.76	2.86	3.18	10.3	<2.7	4.05	5.94
17	MM12B652	1.25	7.24	10.5	<1	<1.2	<1.5	4.20	<2.7	<1	<3.4
18	MM12B656	<1	6.79	10.5	<1	<1.2	<1.5	6.35	<2.7	<1	<3.4
19	MM12B660	1.55	14.4	17.3	4.15	2.64	4.47	12.9	<2.7	3.60	6.34
20	MM12B664	<1	6.09	11.1	<1	<1.2	<1.5	5.83	<2.7	<1	<3.4
22	MM16B169	1.05	16.2	34.6	<1.26	<0.59	<0.57	10.1	<0.29	<0.48	<0.90
23	MM16B172	<0.66	33.2	34.6	<1.26	<0.59	6.10	15.3	<0.29	<0.48	<0.90
24	MM16B175	<0.66	7.85	5.06	<1.26	<0.59	<0.57	<0.14	<0.29	<0.48	<0.90
25	MM16B178	11.78	34.8	47.5	2.86	<0.59	11.0	14.5	<0.29	<0.48	<0.90
26	MM16B181	<0.66	16.8	18.4	<1.26	<0.59	<0.57	6.44	<0.29	<0.48	<0.90
27	MM16B184	<0.66	13.9	15.9	<1.26	<0.59	<0.57	<0.14	<0.29	<0.48	<0.90
28	MM16B187	<1	6.77	9.98	<1	<1.2	<1.5	7.74	<2.7	<1	<3.4
29	MM16B190	<0.66	18.3	22.0	<1.26	<0.59	<0.57	<0.14	<0.29	<0.48	<0.90
30	MM16B193	<0.66	16.5	20.1	<1.26	<0.59	9.45	12.0	<0.29	<0.48	<0.90
31	MM16B196	<0.66	21.6	22.9	<1.26	<0.59	<0.57	<0.14	<0.29	<0.48	<0.90
38	MM22B819	<0.66	19.5	18.6	<1.26	<0.59	10.8	11.4	<0.29	<0.48	<0.90
39	MM22B822	<0.66	14.1	13.4	<1.26	<0.59	<0.57	<0.14	<0.29	<0.48	<0.90
40	MM22B825	<0.66	46.5	40.0	<1.26	<0.59	<0.57	<0.14	<0.29	<0.48	<0.90
41	MM22B828	<1	6.29	5.53	<1	<1.2	<1.5	1.45	<2.7	<1	<3.4
42	MM22B831	<0.66	17.9	23.6	<1.26	<0.59	<0.57	23.4	<0.29	<0.48	<0.90
44	MM22B796	7.47	18.2	15.8	<1.26	<0.59	7.78	11.9	<0.29	<0.48	<0.90
45	MM22B799	11.17	53.7	28.2	<1.26	<0.59	11.4	36.5	<0.29	<0.48	<0.90
46	MM22B808	<1	20.6	21.8	1.20	<1.2	1.56	6.54	<2.7	<1	<3.4
47	MM22B802	<0.66	16.6	21.4	<1.26	<0.59	<0.57	<0.14	<0.29	<0.48	<0.90
48	MM22B836	<0.66	19.9	15.3	<1.26	<0.59	<0.57	<0.14	<0.29	<0.48	<0.90
52	MM22B899	<0.66	52.7	51.1	<1.26	<0.59	<0.57	<0.14	<0.29	<0.48	<0.90
53	MM22B902	<0.66	26.1	29.5	<1.26	<0.59	<0.57	11.90	<0.29	<0.48	<0.90
54	MM22B905	1.44	12.8	15.0	2.72	<1.2	4.15	8.63	<2.7	<1	<3.4
55	MM22B908	1.51	10.1	12.8	3.07	2.33	<1.5	8.14	<2.7	<1	4.51
56	MM22B911	1.81	7.22	9.23	<1	<1.2	<1.5	6.74	<2.7	<1	<3.4
57	MM22B914	0.94	9.05	11.8	<1	<1.2	<1.5	7.46	<2.7	<1	<3.4
58	MM22B917	1.48	7.05	10.3	<1	<1.2	<1.5	7.77	<2.7	<1	<3.4
59	MM22B920	1.95	6.88	11.9	<1	<1.2	<1.5	7.48	<2.7	<1	<3.4
60	MM22B923	1.12	10.0	13.7	2.20	2.19	<1.5	7.21	<2.7	3.21	<3.4
61	MM22B926	1.46	12.3	12.4	1.99	1.58	<1.5	7.31	<2.7	1.72	<3.4

Table 10: Mass fractions of chlorinated pesticides (ng/g wet mass) in northern fur seal blubber

FRS L-	Storage ID	Pe-CB	HCB	α -HCH	β -HCH	γ -HCH	Oxy-chlor	<i>trans</i> -chlordane	<i>cis</i> -chlor	<i>trans</i> -nonachlor	<i>cis</i> -nonachlor	Hepta-chlor	heptachlor epoxide
1	MM1B0	3.1	0.26	124	188	21.4	110	63.3	274	712	233	<1.4	<1.5
2	MM1B0	3.7	2.5	103	147	13.5	69.2	3.9	2.7	326	2.4	<1.4	<1.4
3	MM1B0	4.1	3.5	127	138	19.3	105	12.5	23.1	361	11.6	<1.4	<1.4
4	MM1B0	4.1	1.1	234	275	45.3	126	16.1	8.7	560	20.7	<3.1	<0.1
5	MM1B0	2.8	1.1	120	200	18.7	95.3	4.45	2.1	361	3.0	<3.7	6.70
6	MM4B0	3.9	1.7	56.3	148	7.2	72.3	3.02	1.9	323	4.9	<1.4	<1.5
7	MM4B1	1.9	0.50	71.2	126	10.5	67.6	4.65	1.9	375	6.2	<1.4	<1.4
8	MM4B1	3.8	0.66	100	245	23.7	113	23.3	220	667	90.8	<1.4	<1.4
9	MM4B1	2.9	2.3	97.3	241	14.1	117	4.9	3.2	702	3.9	<1.4	<1.4
10	MM4B1	2.2	0.24	84.0	194	9.7	168	6.7	2.9	834	21.2	<1.5	<1.5
11	MM4B1	2.8	0.36	82.5	233	16.4	178	9.0	4.3	736	11.4	<1.4	<1.4
12	MM4B1	3.8	0.24	83.5	203	11.0	123	6.2	2.4	703	4.6	<1.4	<1.4
13	MM4B1	2.4	0.53	184	262	30.6	134	8.4	7.5	559	7.95	<1.5	<1.5
14	MM4B1	5.7	1.7	46.7	151	4.6	156	5.3	1.8	786	11.8	<1.5	<1.5
15	MM4B1	2.0	0.32	53.9	178	6.9	87.2	3.0	0.93	568	2.6	<1.5	<1.5
16	MM12B	3.0	0.44	62.0	219	12.6	175	10.5	10.8	799	17.9	<1.4	<1.4
17	MM12B	1.7	0.30	39.7	145	6.7	83.6	5.5	2.7	373	6.1	<1.4	<1.4
18	MM12B	2.4	0.51	56.7	257	11.4	77.2	6.0	1.9	377	4.0	<1.4	<1.4
19	MM12B	3.7	0.39	61.7	218	12.0	90.1	5.3	2.4	479	4.4	<1.4	<1.4
20	MM12B	2.4	1.1	37.6	127	7.6	69.1	3.8	2.2	360	6.2	<1.4	<1.4
22	MM16B	1.4	0.16	48.3	248	11.7	84.7	3.9	1.4	357	4.3	9.78	<0.1
23	MM16B	1.5	0.68	42.3	141	9.3	51.9	4.8	2.0	243	3.9	<3.6	<0.1
24	MM16B	1.7	<0.1	50.9	222	10.9	72.5	6.2	1.1	328	4.1	<3.5	9.6
25	MM16B	1.8	<0.1	48.9	275	7.7	109	6.4	1.4	536	6.0	<3.7	<0.1
26	MM16B	2.2	0.67	41.8	195	7.1	48.8	2.8	1.1	292	2.3	15.8	23.3
27	MM16B	1.4	0.26	39.2	146	7.0	44.9	2.7	1.0	250	1.9	<3.3	1.4
28	MM16B	1.7	0.95	29.7	114	5.6	52.5	2.4	1.4	225	3.7	<1.5	<1.50
29	MM16B	1.5	<0.1	69.3	424	12.7	95.3	5.6	1.1	505	4.1	<3.3	28.4
30	MM16B	2.7	0.89	41.5	183	6.7	52.8	3.2	1.6	286	3.4	<3.7	<0.1
31	MM16B	1.6	0.50	49.4	178	9.3	55.1	5.2	2.2	307	4.4	<3.4	11.1
38	MM22B	2.3	<0.1	24.4	96.9	4.0	68.7	6.7	3.8	402	7.0	<3.7	<0.1
39	MM22B	1.4	<0.1	22.2	123	4.0	60.1	7.0	2.4	306	6.4	14.6	1.5
40	MM22B	2.9	0.28	22.1	128	5.0	39.2	3.1	0.81	216	2.9	<3.8	<0.1
41	MM22B	2.9	1.4	24.6	95.4	4.5	30.1	2.5	1.0	145	1.7	<1.5	<1.54
42	MM22B	3.2	0.16	33.7	166	5.3	43.9	2.1	0.45	212	1.2	<2.5	0.25
44	MM22B	2.2	0.59	24.3	87.5	3.5	41.0	2.8	1.2	216	2.1	<2.9	2.7
45	MM22B	1.9	0.83	24.0	119	4.5	42.4	3.9	1.1	216	2.6	<3.8	<0.1
46	MM22B	1.7	<0.1	33.9	186	11.8	84.2	11.1	116.5	456	98.8	<3.4	8.0
47	MM22B	3.2	0.22	24.0	131	6.1	61.6	4.8	1.5	383	4.8	<3.1	1.6
48	MM22B	1.7	0.61	25.5	72.8	5.1	31.9	3.7	3.0	148	4.0	<3.6	<0.1
52	MM22B	3.1	3.2	22.6	202	4.5	63.9	4.1	1.4	363	3.8	<3.7	<0.1
53	MM22B	2.4	0.77	24.1	224	5.3	78.4	3.8	1.5	502	4.3	<3.0	<0.1
54	MM22B	2.6	0.49	25.7	114	3.7	63.1	5.3	2.0	312	6.2	<1.5	<1.5
55	MM22B	5.0	0.38	22.1	231	4.0	137	9.2	1.9	578	6.5	<1.4	<1.4
56	MM22B	1.3	0.65	11.3	54.1	1.4	34.5	1.9	0.82	169	1.7	<1.5	<1.5
57	MM22B	2.0	0.89	20.4	84.6	3.2	48.1	2.2	1.0	259	3.4	<1.4	<1.4
58	MM22B	1.5	0.48	15.4	89.4	3.0	37.1	1.9	0.90	225	1.9	<1.5	<1.5
59	MM22B	1.4	0.59	15.3	101	4.0	63.5	5.8	3.1	329	8.0	<1.4	<1.4
60	MM22B	2.5	0.31	24.1	225	3.3	92.9	3.4	0.76	532	5.1	<1.4	<1.4
61	MM22B	3.3	0.30	40.8	205	8.1	163	9.5	2.9	654	24.3	<1.4	<1.4

Table 10 (cont.): Mass fractions of chlorinated pesticides (ng/g wet mass) in northern fur seal blubber

FRSL-	Storage ID	2,4'-DDE	4,4'-DDE	2,4'-DDD	2,4'-DDT	4,4'-DDD	4,4'-DDT	Parlar 26	Parlar 32	Parlar 50	Parlar 62	mirex
1	MM1B004	3.3	2460	18.65	102	184	160	575	13.2	375	152	17.5
2	MM1B008	1.0	844	6.83	29.6	96.7	71.6	21.2	1.65	11.5	8.59	7.90
3	MM1B012	0.17	1100	6.34	22.8	102	60.1	146	2.74	118	52.5	4.66
4	MM1B016	0.87	710	3.60	7.08	23.7	17.9	133	5.62	98.9	52.3	<0.1
5	MM1B020	<0.1	339	<0.1	<0.1	12.3	13.9	23.1	0.752	13.3	5.31	13.9
6	MM4B097	2.9	777	7.45	33.7	106	84.1	14.4	1.52	7.9	4.41	4.73
7	MM4B100	3.0	842	7.30	34.0	101	100	17.2	2.63	11.1	8.08	8.41
8	MM4B103	4.6	1660	21.8	101	195	129	351	4.15	282	78.9	10.5
9	MM4B106	2.9	3310	7.90	35.1	159	102	25.2	1.90	13.8	3.89	8.25
10	MM4B109	4.3	1550	12.1	62.6	145	164	52.0	2.38	27.4	11.2	13.4
11	MM4B112	3.4	1650	18.02	75.6	273	150	50.1	3.42	27.9	18.1	12.3
12	MM4B115	148	38000	11.9	53.8	150	138	27.1	1.54	13.9	7.02	10.6
13	MM4B118	5.1	1110	15.8	104	213	157	67.0	3.59	39.4	20.5	9.17
14	MM4B121	88.5	82700	4.88	39.9	105	189	38.3	2.01	20.9	12.3	12.9
15	MM4B124	142	46600	7.52	39.2	123	118	16.2	1.22	9.32	6.15	8.15
16	MM12B648	0.67	1740	9.27	38.4	135	71.1	91.9	4.07	54.7	21.8	15.4
17	MM12B652	0.49	1620	5.43	21.5	76.5	60.7	34.6	1.32	20.1	4.22	5.02
18	MM12B656	2.3	751	9.11	29.6	140	60.5	22.5	2.65	14.6	3.09	8.01
19	MM12B660	1.4	1740	8.90	29.5	140	61.6	27.9	2.24	15.4	4.72	11.5
20	MM12B664	1.3	959	8.12	31.0	113	52.5	19.1	2.46	12.7	8.23	5.19
22	MM16B169	1.7	491	1.58	0.281	20.0	34.5	25.4	1.21	19.3	6.76	<0.1
23	MM16B172	<0.1	285	<0.1	<0.1	9.17	6.53	22.3	<0.4	16.4	3.16	<0.1
24	MM16B175	<0.1	452	5.70	<0.1	15.3	6.75	22.8	1.53	17.4	6.40	11.8
25	MM16B178	<0.1	645	<0.1	<0.1	8.77	7.47	36.3	1.69	25.9	6.93	<0.1
26	MM16B181	<0.1	554	4.03	<0.1	14.5	1.45	14.2	<0.4	8.80	<2.2	<0.1
27	MM16B184	0.14	567	6.23	<0.1	9.22	<0.1	11.7	0.441	8.02	2.41	<0.1
28	MM16B187	0.69	796	4.46	13.3	52.3	32.7	15.8	1.19	10.4	<2.2	8.89
29	MM16B190	<0.1	275	<0.1	<0.1	10.3	7.16	21.9	1.67	15.9	7.93	<0.1
30	MM16B193	<0.1	430	<0.1	<0.1	8.85	4.05	16.0	0.971	11.4	3.94	<0.1
31	MM16B196	0.22	211	<0.1	<0.1	9.36	1.98	23.9	1.93	17.0	6.63	<0.1
38	MM22B819	0.51	782	11.1	<0.1	10.6	8.87	28.7	1.81	22.9	8.56	<0.1
39	MM22B822	0.11	453	<0.1	0.938	0.34	<0.1	33.3	2.04	25.5	7.38	9.76
40	MM22B825	0.47	303	<0.1	<0.1	7.42	3.48	14.9	1.02	10.4	3.29	<0.1
41	MM22B828	1.2	502	6.41	14.1	398	19.4	7.94	0.988	5.52	<2.2	5.69
42	MM22B831	0.12	415	<0.1	<0.1	13.9	5.12	9.76	0.57	6.58	<2.2	<0.1
44	MM22B796	0.44	228	<0.1	<0.1	5.68	2.19	10.8	<0.4	7.43	<2.2	15.7
45	MM22B799	<0.1	256	<0.1	<0.1	8.64	8.46	13.8	1.00	10.3	<2.2	<0.1
46	MM22B808	1.00	486	3.11	2.07	16.8	33.4	179	2.88	218	40.4	1.75
47	MM22B802	0.13	736	<0.1	<0.1	12.9	<0.1	21.2	1.61	17.2	9.27	<0.1
48	MM22B836	<0.1	230	<0.1	<0.1	5.09	5.32	21.4	1.46	16.9	6.06	<0.1
52	MM22B899	0.44	502	<0.1	<0.1	14.8	4.76	16.0	1.03	10.8	2.32	79.7
53	MM22B902	0.13	839	<0.1	<0.1	9.66	13.7	21.0	1.38	15.2	4.28	<0.1
54	MM22B905	0.89	1200	5.54	17.6	55.3	36.5	24.4	2.78	<2.2	9.07	6.99
55	MM22B908	<0.1	1310	4.50	13.7	108	56.0	50.3	1.38	33.1	13.1	14.7
56	MM22B911	<0.1	720	2.94	4.23	54.1	17.3	5.66	<0.4	3.38	<2.2	4.44
57	MM22B914	0.13	864	4.43	14.6	68.8	43.1	11.4	1.80	7.56	<2.2	6.38
58	MM22B917	<0.1	836	3.30	10.5	55.0	21.8	10.4	<0.4	5.24	2.24	5.58
59	MM22B920	0.78	927	7.15	24.3	100	49.1	33.2	1.19	17.4	4.74	7.71
60	MM22B923	1.4	1520	5.39	16.0	121	65.1	18.7	1.83	10.6	2.84	12.3
61	MM22B926	4.3	2100	16.5	77.2	225	153	91.4	3.64	47.3	16.4	10.2

Table 11: Sums of the mass fractions of organohalogen compounds (ng/g wet mass) by group in northern fur seals.

FRSL-	ΣPCB	ΣDDT	ΣChlor	ΣTox	ΣHCH	ΣCIBz	Mirex	ΣPBDE	α- HBCD	ΣPFAAs
1	1870	2920	1390	1120	333	3.3	17.5	7.3	2.6	6.9
2	848	1050	404	42.9	263	6.2	7.9	1.5	<0.10	8.1
3	935	1291	514	319	284	7.5	4.7	1.0	0.15	26.3
4	1800	763	732	290	554	5.2	<0.1	6.5	0.36	27.1
5	1100	366	472	42.4	339	3.9	13.9	2.7	<0.10	no value
6	996	1010	405	28.3	212	5.6	4.7	10.8	0.55	8.6
7	1260	1087	455	39.1	208	2.4	8.4	11.6	0.71	19.6
8	1510	2110	1110	716	368	4.5	10.5	13.9	3.6	9.3
9	1830	3620	830	44.8	353	5.3	8.3	25.1	0.21	15.9
10	2010	1940	1030	93.0	288	2.4	13.4	11.0	0.57	22.3
11	1690	2170	939	99.5	332	3.2	12.3	9.0	0.79	50.4
12	1870	38500	839	49.6	298	4.0	10.6	12.3	0.83	19.6
13	1510	1610	716	131	476	2.9	9.2	10.1	0.55	4.9
14	2190	83200	962	73.4	203	7.4	12.9	16.7	0.29	12.1
15	1890	47000	662	32.9	239	2.3	8.1	9.4	0.23	10.2
16	1740	2000	1010	173	293	3.4	15.4	14.4	4.7	28.6
17	1050	1790	471	60.3	191	2.0	5.0	25.7	2.2	18.6
18	1080	992	466	42.8	325	2.9	8.0	7.4	1.9	22.9
19	1360	1980	581	50.3	291	4.1	11.5	19.2	1.4	23.4
20	980	1160	441	42.5	172	3.5	5.2	10.3	3.3	45.9
22	1960	549	461	52.7	308	1.6	<0.1	39.6	2.0	11.7
23	1890	300	305	41.8	192	2.2	<0.1	16.8	1.5	86.1
24	1210	480	422	48.1	284	1.7	11.8	33.2	1.8	81.6
25	2080	661	659	70.9	331	1.8	<0.1	60.8	2.4	54.9
26	1690	574	386	23.0	243	2.9	<0.1	15.8	1.3	38.8
27	1830	583	302	22.6	192	1.7	<0.1	21.5	0.87	40.4
28	709	899	285	27.4	149	2.6	8.9	18.0	1.6	31.5
29	2210	293	639	47.4	506	1.5	<0.1	35.0	1.4	25.4
30	1310	443	347	32.3	231	3.6	<0.1	32.3	1.5	16.6
31	1500	223	385	49.5	237	2.1	<0.1	18.8	1.9	31.4
38	1600	813	488	62.0	115	2.3	<0.1	38.5	5.5	59.5
39	1140	454	398	68.2	148	1.4	9.8	57.4	4.3	154
40	1710	314	262	29.6	161	3.2	<0.1	49.2	1.7	47.9
41	558	583	181	14.4	231	4.3	5.7	10.0	1.6	162
42	1920	434	260	16.9	125	3.3	<0.1	39.8	1.2	37.0
44	1340	237	265	18.2	149	2.8	15.7	14.9	1.7	20.6
45	2040	273	266	25.1	155	2.8	<0.1	20.1	1.8	164
46	2080	543	775	440	125	1.7	1.8	93.2	22.4	41.9
47	1560	749	457	49.3	205	3.4	<0.1	55.7	3.7	47.2
48	1390	240	191	45.8	103	2.3	<0.1	28.9	2.2	30.3
52	1590	522	436	30.2	229	6.3	79.7	60.6	2.7	137
53	1690	863	590	41.8	254	3.1	<0.1	97.8	5.2	44.9
54	1090	1320	388	36.3	143	3.1	7.0	36.1	5.8	50.7
55	1490	1490	733	97.9	257	5.4	14.7	15.4	7.7	37.8
56	701	798	208	9.0	67	2.0	4.4	23.9	1.1	105
57	843	995	314	20.7	108	2.9	6.4	23.8	4.2	42.7
58	872	927	267	17.9	108	2.0	5.6	12.1	1.9	111
59	1070	1110	409	56.5	121	2.0	7.7	28.3	5.2	89.3
60	1690	1730	634	34.0	253	2.9	12.3	38.9	2.7	54.2
61	1840	2580	853	159	254	3.6	10.2	49.2	10.0	99.2

Table 12: Summary of organohalogen mass fractions (ng/g wet mass) determined in northern fur seals.

	Σ PCB	Σ DDT	Σ Chlor	Σ HCH	Σ Tox	Mirex	Σ CIBz	Σ PBDE	α -HBCD	Σ PFAAs
mean	1480	4370	540	240	103	11.4	3.29	26.2	2.78	47.7
geomean	1410	1060	481	219	54.1	9.09	3.00	18.7	1.71	33.7
median	1540	913	459	234	45.3	9.03	2.93	19.0	1.82	37.0
SD	437	14100	270	103	190	12.6	1.49	21.3	3.53	41.4
min	558	223	181	66.7	9.04	1.75	1.37	1.03	0.15	4.86
max	2210	83200	1390	554	1120	79.7	7.54	97.8	22.4	164
#>LOD	50/50	50/50	50/50	50/50	50/50	34/50	50/50	50/50	48/50	49/49

Table 13: Mass fractions (ng/g wet mass) of PFAAs determined in the fish reference materials

Compound	SRM 1946		mean	Certified Value	SRM 1947		mean	Certified Value
	Set 1	Set 2			Set 1	Set 2		
PFBA	<0.9	<6.3			<0.8	<8.2		
PFPeA	<1.1	<1.8			<1.0	<2.3		
PFHxA	<0.1	<0.9			<0.1	<1.2		
PFHpA	<1.8	<0.7			<1.5	<1.0		
PFOA	<0.5	<1.8			<0.4	<2.3		
PFNA	<0.7	<0.4			<0.6	<0.5		
PFDA	<0.4	<1.5			<0.3	<1.9		
PFUnA	<1.5	0.78			<1.3	<0.74		
PFDoA	<0.2	<0.6			<0.2	<0.8		
PFTriA	<0.2	0.37			<0.1	<0.3		
PFTA	<0.3	<0.4			<0.3	<0.6		
PFBS	<0.1	<0.2			<0.1	<0.3		
PFHxS	<0.1	0.12			<0.1	0.16		
PFOS	1.84	2.29	2.07	2.19 ± 0.08	6.88	5.42	6.15	5.9 ± 0.39
PFOSA	<0.1	0.11			<0.1	<0.1		

Table 14: Mass fractions of brominated flame retardants (ng/g wet mass) in northern fur seal blubber

FRSL-	Storage ID	BDE 47	BDE 100	BDE 99	BDE 155	BDE 154	BDE 153	α -HBCD
1	MM1B004	2.7	0.70	0.65	0.45	2.5	0.34	2.64
2	MM1B008	1.0	0.47	<0.100	<0.4	<0.1	<0.2	<0.1
3	MM1B012	1.0	<0.2	<0.100	<0.4	<0.1	<0.2	0.15
4	MM1B016	2.8	0.63	0.72	<0.4	1.8	0.52	0.36
5	MM1B020	1.5	0.25	0.26	<0.4	0.65	<0.2	<0.068
6	MM4B097	8.0	1.05	0.86	<0.4	0.60	0.29	0.55
7	MM4B100	7.3	1.1	0.92	0.60	0.90	0.83	0.71
8	MM4B103	6.2	2.7	1.1	0.48	2.6	0.85	3.5
9	MM4B106	12.3	3.6	4.2	0.53	2.8	1.6	0.21
10	MM4B109	7.4	0.92	0.99	0.47	0.87	0.33	0.57
11	MM4B112	4.9	1.6	1.0	<0.4	1.2	0.35	0.79
12	MM4B115	8.3	1.3	0.76	0.48	1.0	0.43	0.83
13	MM4B118	5.6	1.8	0.97	0.42	0.95	0.38	0.55
14	MM4B121	9.0	1.1	1.2	0.61	3.3	1.5	0.29
15	MM4B124	5.8	1.1	0.99	<0.4	0.78	0.77	0.23
16	MM12B648	7.2	2.3	1.6	1.09	1.7	0.65	4.7
17	MM12B652	16.7	3.1	3.5	0.43	1.4	0.48	2.0
18	MM12B656	5.4	1.5	<0.1	0.44	0.18	<0.2	1.9
19	MM12B660	9.8	3.4	2.2	0.63	2.0	1.2	1.4
20	MM12B664	6.1	1.6	1.4	0.43	0.51	0.21	3.3
22	MM16B169	24.3	6.7	4.7	0.59	1.8	1.4	2.0
23	MM16B172	10.8	2.4	2.4	<0.4	1.2	<0.2	1.5
24	MM16B175	21.5	3.8	5.2	0.45	1.3	0.85	1.8
25	MM16B178	40.2	3.3	8.7	1.01	3.3	4.2	2.4
26	MM16B181	7.6	2.4	2.0	0.48	1.8	1.5	1.3
27	MM16B184	11.9	2.6	3.6	0.57	1.5	1.3	0.87
28	MM16B187	12.1	3.0	1.9	<0.4	1.0	<0.2	1.6
29	MM16B190	24.0	3.8	4.5	0.45	1.5	0.86	1.4
30	MM16B193	19.3	4.3	5.0	0.57	1.9	1.3	1.5
31	MM16B196	10.8	2.3	3.5	<0.4	1.3	0.86	1.9
38	MM22B819	24.2	4.0	5.3	1.0	2.4	1.5	5.5
39	MM22B822	37.8	7.5	6.7	0.94	2.5	1.8	4.3
40	MM22B825	30.4	8.3	6.4	0.62	1.9	1.7	1.6
41	MM22B828	5.9	1.4	1.4	<0.4	0.71	0.71	1.6
42	MM22B831	25.8	5.0	6.0	<0.4	1.3	1.7	1.2
44	MM22B796	9.9	1.1	2.3	<0.4	0.81	0.77	1.7
45	MM22B799	11.8	4.3	2.0	0.50	0.83	0.70	1.71
46	MM22B808	57.8	18.3	10.2	1.2	3.9	1.7	21.7
47	MM22B802	31.4	8.0	9.4	1.0	2.9	2.9	3.6
48	MM22B836	18.7	4.8	2.7	0.51	1.4	0.75	2.1
52	MM22B899	38.2	9.0	7.7	0.94	2.5	2.1	2.7
53	MM22B902	60.3	19.1	8.7	1.6	3.9	4.2	5.1
54	MM22B905	27.5	3.1	2.6	0.82	1.3	0.76	5.8
55	MM22B908	9.1	2.4	1.2	0.91	1.2	0.54	7.7
56	MM22B911	13.8	5.3	2.8	0.52	0.87	0.62	1.1
57	MM22B914	14.6	3.1	2.8	0.84	1.4	0.98	4.2
58	MM22B917	5.6	2.7	2.1	<0.4	0.86	0.90	1.9
59	MM22B920	16.6	5.7	3.2	0.52	1.5	0.78	5.2
60	MM22B923	26.8	4.8	4.7	0.65	1.2	0.68	2.7
61	MM22B926	31.5	9.2	3.6	1.2	3.0	0.77	9.9

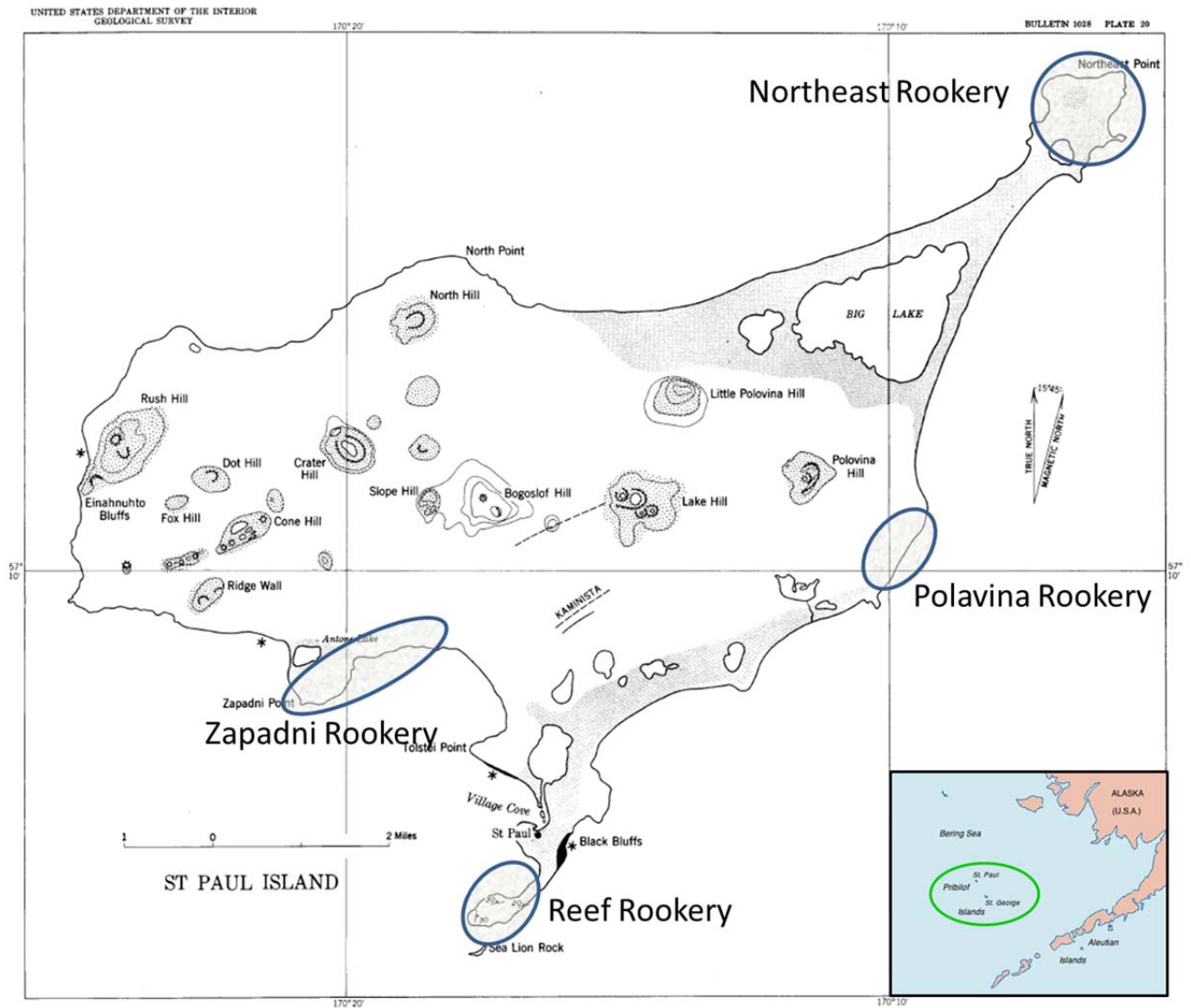


Figure 1: St. Paul Island and rookery locations

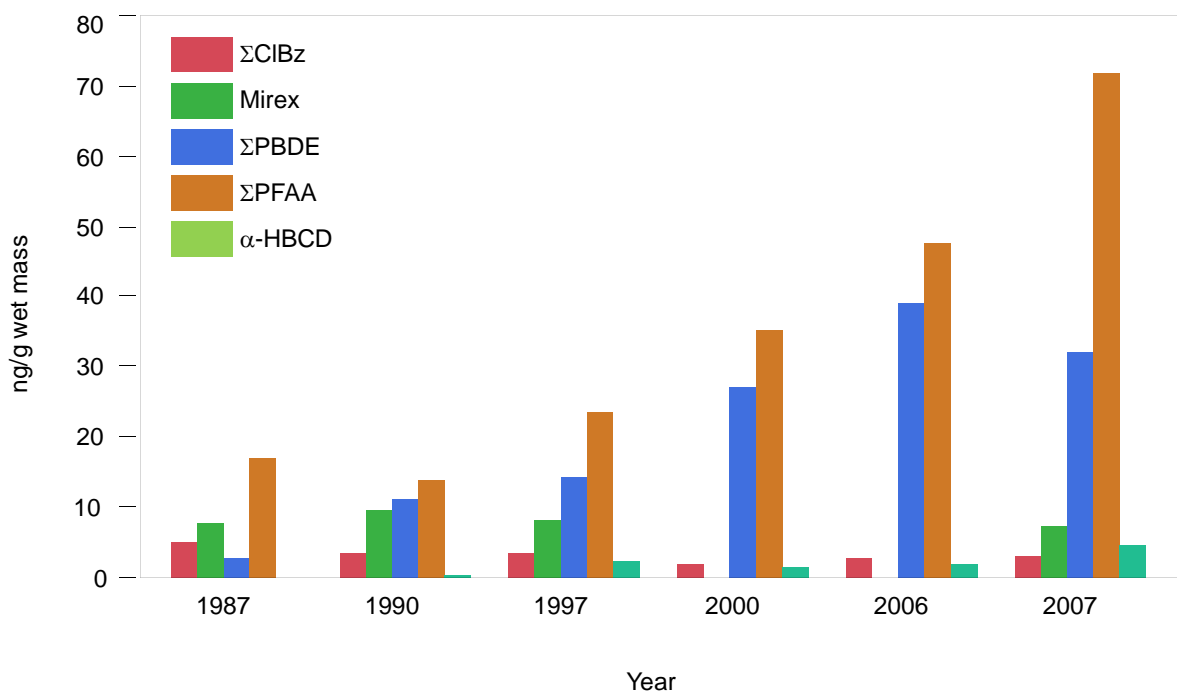
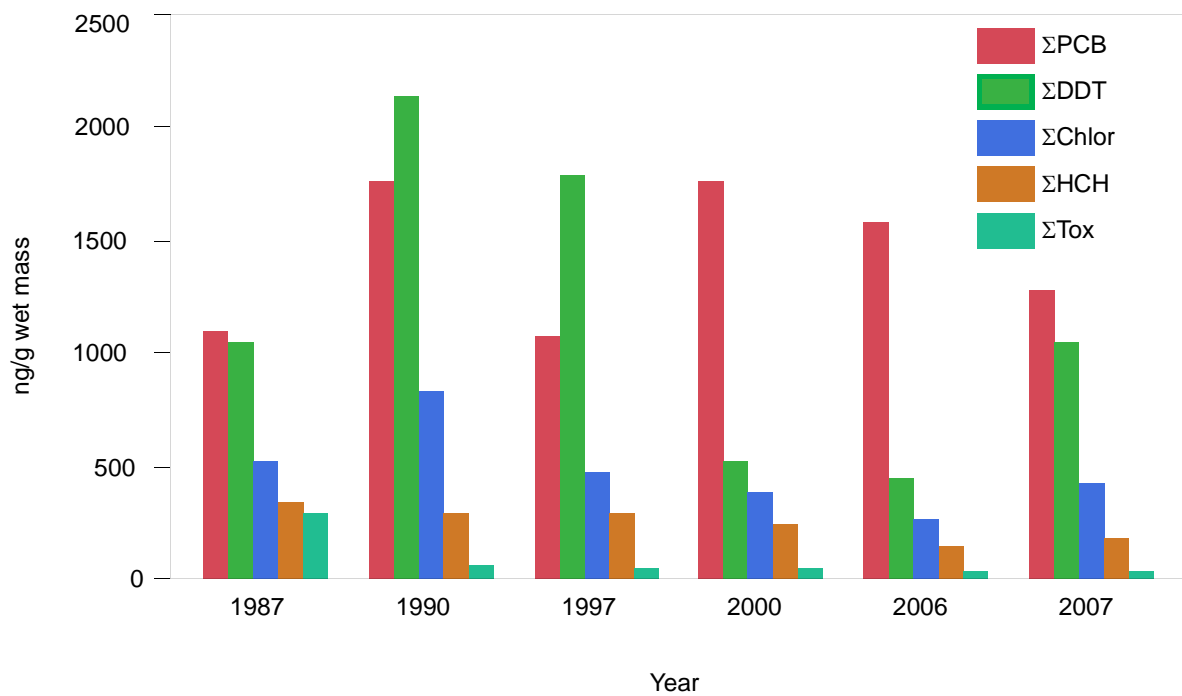


Figure 2: Median mass fractions of organohalogenes in fur seals by year

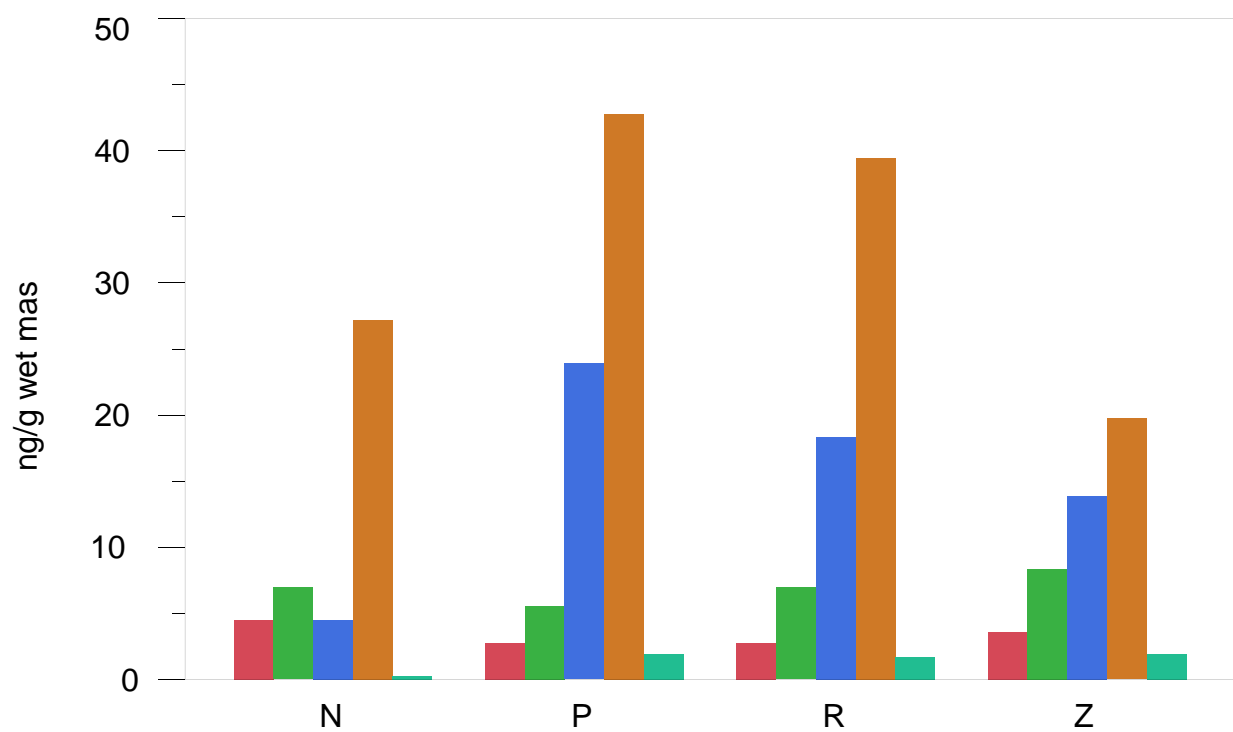
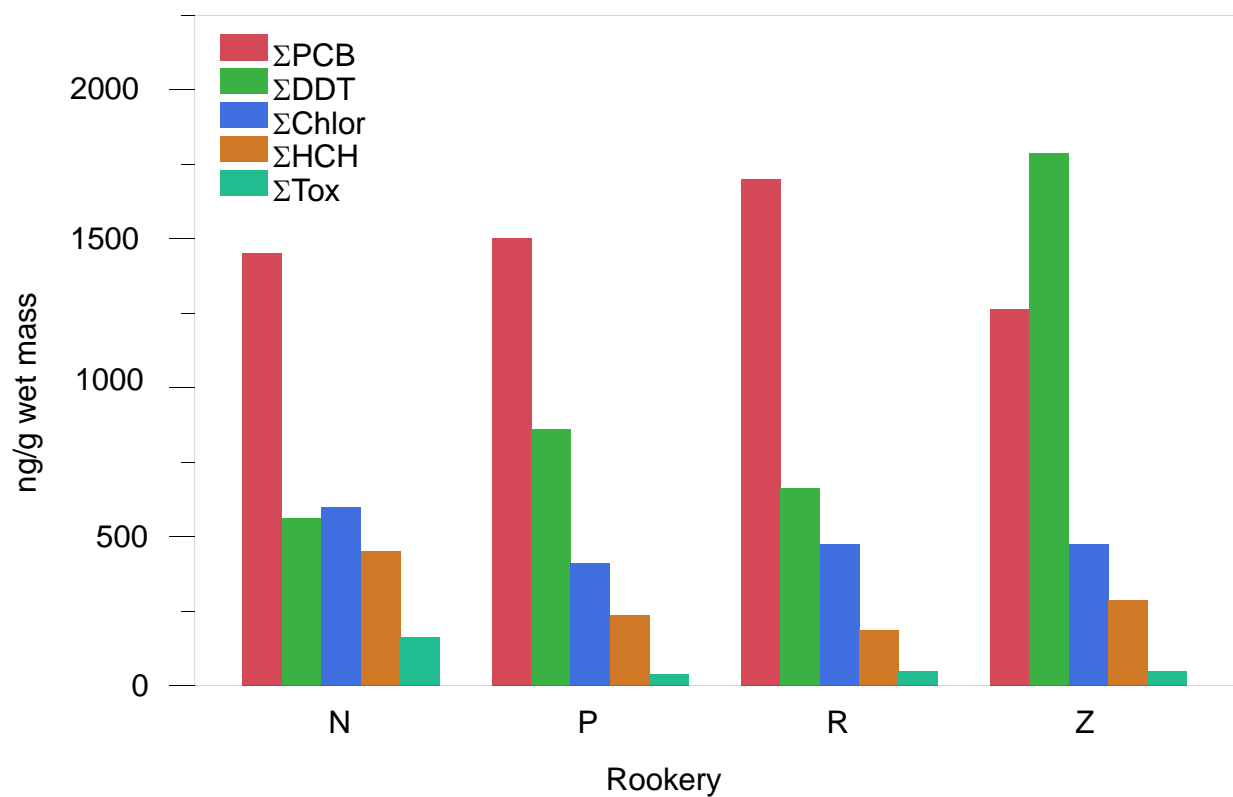


Figure 3: Median POP mass fractions by rookery (see Table 2 for number of seals per rookery). Rookery designations are Northeast (N), Polavina (P), Reef (R), and Zapadni (Z; See Fig. 1)

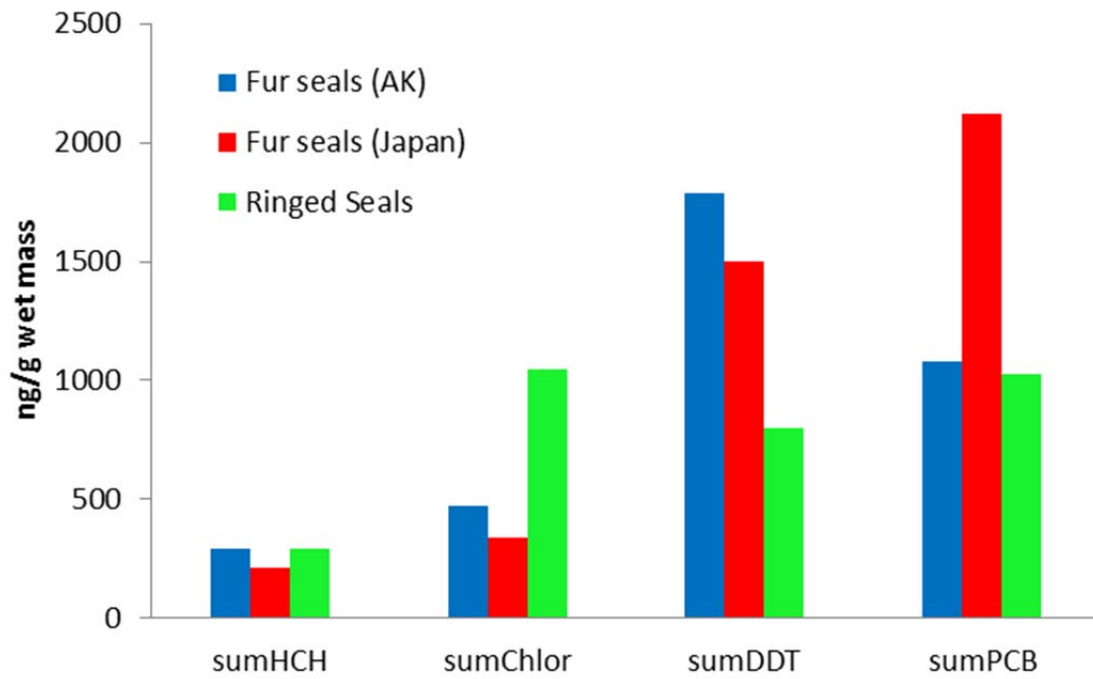


Figure 4: Mass fractions of legacy POPs in the current study (juvenile males) compared to NFS from the waters off of Japan and ringed seals from Norton Sound, AK.