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Polychlorinated biphenyls and chlorinated pesticides in harbor seals (*Phoca vitulina concolor*) from the northwestern Atlantic coast

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Abstract

Concentrations of polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane (DDT) and its metabolites, chlordane-related compounds (CHLs), mirex, hexachlorocyclohexane isomers (HCHs), dieldrin, and hexachlorobenzene (HCB) were determined in blubber of harbor seals (*Phoca vitulina concolor*) from the northwestern Atlantic coast. PCBs, DDTs, and CHLs were the major persistent organochlorines in harbor seal blubber, while mirex, HCHs, dieldrin, and HCB were minor contaminants. Highest concentrations were found in the adult males, followed by the pups, yearlings, adult females, and fetuses. DDT and PCB concentrations have declined from the high levels reported in the early 1970s, but no declines were observed in our samples over the ten-year period 1991–2001. DDT/PCB ratios were indicative of a more rapid decline of DDTs than PCBs, while ratios of p,p′-DDE/DDT were indicative of a metabolic "weathering" of DDT. The population appears to be susceptible to disease outbreaks, as evidenced by a recurrence of viral epizootics since the late 1970s. The PCB burdens in these seals are similar to levels reported in seals from polluted regions of Europe and Asia, and exceed the estimated threshold levels for adverse reproductive and immune system effects in the species.

Keywords: Harbor Seals; Northwestern Atlantic; Epizootics; Temporal trends; PCBs; Organochlorine pesticides

1. Introduction

Harbor seals (*Phoca vitulina*) are widely distributed in the temperate near-shore waters of the northwestern Atlantic and are useful indicators of ecosystem contamination because they occupy a high trophic level and accumulate high concentrations of persistent organic pollutants (POPs) over a long lifetime. Concern has focused on the polyhalogenated aromatic hydrocarbons (PHAHs) including the polychlorinated biphenyls (PCBs), dioxins and furans (PCDD/Fs), and chlorinated pesticides (DDT, chlordane) because of their lipophilicity, ubiquity and stability in the marine environment, and their immune- and endocrine-disrupting potential in seals and other marine wildlife (De Guise et al., 2001; Shaw, 2001; Colborn et al., 1993). A large body of data suggests that PHAHs, particularly PCBs and DDT, have adversely affected the health of seals inhabiting industrialized regions, and may be a contributing factor in the recurring epizootics reported among harbor seals since the late 1970s (Geraci et al., 1982; Dietz et al., 1989; Duignan et al., 1993; Van Loveren et al., 2000;

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Harding et al., 2002). The sensitivity of harbor seals to the effects of PHAH exposure was previously demonstrated by captive feeding studies in which harbor seals fed PHAH-contaminated fish exhibited reproductive impairment (Reijnders, 1986), reduced plasma thyroid hormone and retinol levels (Brouwer et al., 1989) and suppression of immune functions (De Swart et al., 1994; Ross et al., 1995).

At present, there are an estimated 99,340 harbor seals (*Phoca vitulina concolor*) inhabiting the northwestern Atlantic region extending from the Gulf of Maine southward to the coast of New Jersey (Gilbert et al., 2001). Considered relatively nonmigratory, harbor seals feed in coastal and estuarine environments and are exposed to contaminated habitats and prey across their range. Isolated from the deeper waters of the Atlantic Ocean by Georges and Brown Banks and Nantucket Shoals, the large northern Gulf portion of their range is a semi-enclosed sea with a variable, principally estuarine circulation pattern receiving significant riverine, urban, agricultural, and industrial pollutant discharges from industrialized areas in the Northeast as well as inputs via long-range atmospheric transport (Hameedi et al., 2002). Because the removal rates of persistent chemicals are slow, biota in such semi-enclosed seas are at elevated risk (Loganathan and Kannan, 1994). In the southern areas, coastal urban development has produced some of the densest concentrations of human populations in North America, and organochlorine (OC) contamination has been a concern since at least the 1950s. In the 1980s, Massachusetts Bay and Long Island Sound were ranked as among the most polluted regions, having more "high organic contamination" sites, than any other coastal state or region in the United States (O'Connor, 1990). Throughout the region, OC levels remain sufficiently high to warrant human fish consumption advisories on marine fishes (Maine Bureau of Health, 2001; New Hampshire Department of Health and Human Services, 2001).

PHAHs, particularly PCBs and DDTs, have been frequently detected in marine wildlife inhabiting the northwestern Atlantic, including right whales (*Eubalaena glacialis*) (Weisbrod et al., 2000a), pilot whales (*Globicephala melaena*) (Weisbrod et al., 2000b), harbor porpoises (*Phocoena phocoena*) (Gaskin et al., 1983), harbor seals (*Phoca vitulina concolor*) (Gaskin et al., 1973), bald eagles (*Haliaeetus leucocephalus*) (Matz, 1998), and migratory waterfowl (Barclay, 2000). PCB-related adverse effects have been reported in marine species including reproductive impairment in Maine coastal bald eagles (Matz, 1998), liver neoplasms in bottom fish from Boston Harbor (Moore and Stegeman, 1994; Moore et al., 1996), and population declines of ducks on Long Island Sound (Barclay et al., 1995). PCBs were strongly implicated as contributing factors in the mass mortality of bottlenose dolphins (Phocena phocena) along the mid-Atlantic coast in 1987–1988 (Kuehl et al., 1991; Lahvis et al., 1995).

Over the past two decades, the northwest Atlantic harbor seal population has been affected by a series of mass mortalities associated with viral epizootics. In 1979–1980, a type A influenza virus infection spread northward from Cape Cod into the Gulf of Maine and ultimately resulted in the deaths of more than 500 seals (Geraci et al., 1982). In 1991–1992, a morbillivirus epizootic (phocine distemper virus, or PDV) of unknown magnitude was reported among harbor seals found stranded from southern Maine to New York (Duignan et al., 1993; Duignan et al., 1995). In late summer of 2004, an unusual mortality event was reported among harbor seals in southern Maine where approximately 300 animals, primarily pups, were found dead on beaches within a few weeks. The possible role of environmental chemicals (e.g., PCBs) in these outbreaks has not been investigated, although data from the 1970s (Gaskin et al., 1973) indicated that PCB and DDT body burdens in these seals exceeded or were approaching the 100 ppm concentration range (lipid basis), similar to the levels associated with reproductive failure and population declines among European harbor, grey (*Halichoerus grypus*), and ringed seals (*Phoca hispida*) (Reijnders, 1980; Helle et al., 1976a,b) and premature pupping in California sea lions (*Zalophus californianus*) (De Long et al., 1973).

The goal of the present study was to determine current burdens of persistent OCs in harbor seals along the northwestern Atlantic coast as part of an ongoing assessment of the effects of environmental contaminants on population health. Concentrations of PCBs and chlorinated pesticides were measured in blubber samples of stranded harbor seals collected in 2001 and 2002, and in archived samples of a subset of yearling seals collected in 1991. We compared OC levels and congener and isomer compositions in the 1991 and 2001–2002 samples, as well as with available region-specific data to examine temporal variability. Concentrations found in this study were also compared with those reported in stranded pinnipeds from different marine regions and with estimated threshold levels of adverse effects to evaluate the toxicological implications of current body burdens.

2. Methods and materials

2.1. Sample collection

Tissues were collected from stranded harbor seals during the spring and summer of 2001–2002 under a permit issued by the National Oceanographic and Atmospheric Administration (NOAA)/National Marine Fisheries Service (NMFS) and the Northeast Regional Stranding Network. The study area extended from

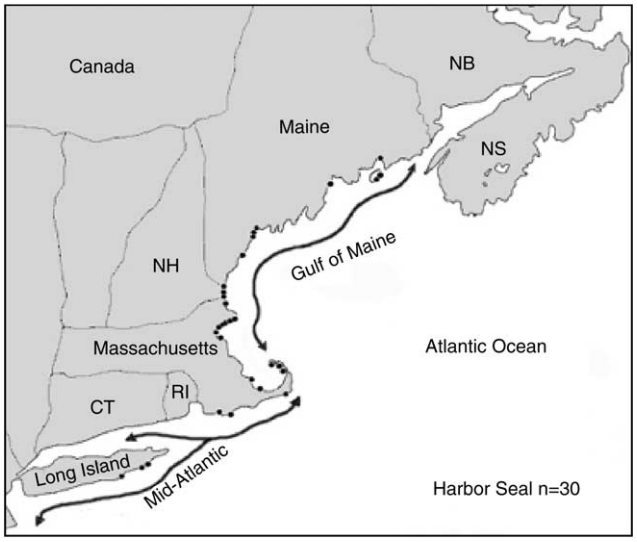

Fig. 1. Stranding locations of harbor seals along the northwestern Atlantic coast. Lines and arrows represent hypothesized seasonal movements of the population.

Mount Desert Island, Maine southward to the eastern shore of Long Island, New York (Fig. 1). Blubber samples were collected from 30 harbor seals (6 adults, 17 yearlings, 4 pups, and 3 fetuses) found at locations in the Gulf of Maine ($n = 24$; 12 males, 11 females, 1 of unknown gender) and along the mid-Atlantic coast ($n = 6$, 3 males, 3 females) (Table 1). In addition, blubber samples collected during 1991 from three harbor seal yearlings from Long Island shore were used to examine temporal trends in OC concentrations. Seal blubber samples were stored at −40 °C until analysis. Seals were weighed, and standard length and axillary girth were measured. Age was estimated based on body size and pelage. A condition index (CI) was estimated for each individual by dividing the axillary girth with the standard body length × 100 (McLaren, 1958; Hobbs et al., 2002) (Table 2).

2.2. Chemical analysis

Identification and quantification of PCBs and OCs was performed by NOAA Method ORCA 130 (NOAA, 1998). Briefly, blubber samples (≈1 g) were homogenized with hydromatrix (in proportion: 4:1 w/w) and extracted in an accelerated solvent extraction (ASE) cell (Dioinex, Inc. Salt Lake City, UT) with methylene chloride under 1500 psi of pressure at 80 °C for ≈11 min per sample. Surrogate, internal standard (1-bromo-2-nitrobenzene)

Table 1
Concentrations of PCBs and OC pesticides (ng/g lw) in blubber of northwestern Atlantic harbor seals

Sample ID	Location	Sex	Age class	Lipid (%)	∑PCB	∑DDT	∑CHL	∑HCB	Mirex
MERI 5	Manchester, MA	M	Adult	82	95 700	16 600	10 900	106	564
MERI 36	MA	F	Adult	85	13 700	6400	825	22	96
MERI 33	Blue Hill, ME	U*	Adult	55	5670	1390	317	49	49
MERI 23	Mt. Desert Isle, ME	M	Adult	80	14 200	4690	1120	73	136
MERI 15	Fairhaven, MA	M	Adult	57	54 900	15 800	3880	122	276
MERI 20	Marblehead, MA	F	Adult	93	8470	2790	724	43	3.3
MERI 40	Long Island E., NY	F	Yearling	77	11 000	4920	1240	51	42
MERI 44	Long Island E., NY	M	Yearling	46	48 600	19 000	3480	113	141
MERI 42	Long Island E., NY	M	Yearling	88	11 800	6430	1620	70	32
MERI 8	N. Truro, MA	M	Yearling	56	7530	2050	535	32	32
MERI 9	Newcastle Beach, MA	F	Yearling	73	15 100	7260	2040	104	44
MERI 10	Gloucester, MA	F	Yearling	18	151 000	57 500	17 600	368	605
MERI 11	Provincetown, MA	M	Yearling	84	6390	2480	571	25	34
MERI 2	Hampton, NH	F	Yearling	91	5870	2630	788	103	14
MERI 12	Hampton, NH	F	Yearling	84	14 500	6750	1970	151	4.3
MERI 13	East Sandwich, MA	F	Yearling	62	21 000	10 300	2410	104	74
MERI 3	Manchester, MA	M	Yearling	95	18 400	7490	1770	53	40
MERI 4	Truro, MA	F	Yearling	77	10 300	4530	1030	58	23
MERI 24	Orleans, MA	F	Yearling	62	22 700	9820	1960	68	40
MERI 6	Falmouth, MA	F	Yearling	25	65 700	25 900	6380	233	174
MERI 31	Cape Elizabeth, ME	F	Yearling	56	34 700	13 500	3050	134	5.0
MERI 30	Cape Elizabeth, ME	M	Yearling	37	110 000	34 200	7640	173	218
MERI 65	Saco, ME	M	Yearling	48	11 100	6050	871	59	7.7
MERI 18	Singing Beach, MA	F	Pup	88	45 200	13 500	3060	86	68
MERI 35	Lincolnville, ME	M	Pup	16	75 100	50 500	14 300	425	460
MERI 38	Mt. Desert Isle, ME	M	Pup	76	23 600	8200	2550	185	38
MERI 29	Cape Elizabeth, ME	M	Pup	62	28 100	12 200	4440	185	97
MERI 19	Newburyport, MA	M	Fetus	34	6950	2430	666	42	3.4
MERI 21	Marblehead, MA	F	Fetus	49	6180	2680	539	53	7.6
MERI 14	Manomet, MA	F	Fetus	45	3860	1500	798	62	22

* U = unknown.

Table 2
Biometry of northwestern Atlantic harbor seals

Age/sex	Total *n*	Weight (kg)/(*n*)	Standard length (cm)	Axillary girth (cm)	Condition index[a]
Adult					
Male	3	57.2 – (1)	146 ± 2.1 144–147 (2)	95 – (1)	66 – (1)
Female	2	68.5 ± 9.6 61.7–75.3 (2)	145 ± 3.5 142–147 (2)	99.5 ± 5 96–103 (2)	68.8 ± 1.7 67–70 (2)
Yearling					
Male/female	17	17.1 ± 3.9 11.3–25.3 (12)	91.8 ± 8.1 79–110 (17)	61.3 ± 7.1 53–69 (4)	65.9 ± 4.9 61.6–72.6 (4)
Pup					
Male/female	4	12.6 ± 0.9 11.8–13.5 (3)	84.8 ± 2.9 81–87 (4)	48.3 ± 6.4 41–53 (3)	57 ± 8.6 47–63 (3)
Fetus					
Male/female	3	5.4 ± 1.5 3.6–6.4 (3)	66 ± 8.2 59–75 (3)	37.7 ± 6.8 30–43 (3)	56.9 ± 5.8 50.9–62.5 (3)

[a] Condition indices calculated as axillary girth/standard length × 100 (McLaren, 1958).

and matrix spiking solutions were added to the cell prior to extraction. Lipid contents were determined gravimetrically from the aliquots of K–D (Kuderna–Danish) concentrated extracts. The remaining extracts were cleaned up using silica gel columns (column-Phenomenex 00W-3035-PO, 350 × 21.20 mm; guide column-Phenomenex 03R-3035-PO Envirosep ABC 60 × 21.20 mm) and gel permeation chromatography (OI, Inc, Austin, TX) with methylene chloride as the mobile phase to remove the lipid portion. The extract was concentrated and then solvent exchanged with hexane and concentrated to a volume of 1 ml. A final cleanup process was accomplished by passing the extract across an activated Florasil packed glass cartridge (Varian part #12102109) for fractionation. Samples were concentrated to a final volume of 0.5 ml using high purity nitrogen (Turbovap II, Zymark, Inc., Hopkington, MA).

Extracts were injected into a gas chromatograph with dual microelectron capture detectors (GC/uECD) and a gas chromatograph with a mass selective detector (GC-MSD) for quantification. OCs were quantified by GC/uECD using HRGC (Agilent model Hewlett Packard 6890 series) equipped with an auto injection system. Target analytes included 20 PCB congeners and 25 OC pesticides. Gas chromatographic separation was carried out by two 60-m long × 0.25-mm ID fused silica capillary columns, DB-XLB and DB-5 ms (J&W Scientific Folsom, CA) using helium as a carrier gas. The initial column oven temperature was programmed for 90 °C for 1.0 min and then programmed at 90–210 °C at 50 °C/min, held 5 min, then 210–235 °C at 1 °C/min, 235–245 °C at 2 °C/min, 245–320 °C at 4 °C/min and then held for 2 min. Injector and detector temperature were held at 250 and 320 °C, respectively. Helium and nitrogen were used as carrier and make-up gases, respectively. OC concentrations were quantified from the peak area of the sample relative to the corresponding internal standards. PCBs were quantified by individual congener using calibration mixtures (Accustandard five PCB congener mix #1–5 which contains 39, 36, 27, 22 and 20 components respectively with total of 144 PCB congeners) which includes PCB congeners contained in Aroclors. The standard, including 24 PCB congeners and 17 pesticides, was used as spiking and performance standards. The individual congener data were processed by Hewlett Packard ChemStation software. Concentrations of individual PCB congeners were summed to obtain total PCB concentrations. Individual PCB congeners are referred to by their IUPAC numbers. Most pesticides coeluted with PCBs on the GC column; therefore, pesticides were analyzed using a HP GC 6890 equipped with HP 5972 MSD and a DB-XLB column (60 m × 25 mm, 0.25 μm film thickness). Selected ion monitoring (SIM) was used for quantitation. The individual pesticide data were processed by HP ChemStation (for MSD) software.

Recovery of surrogates in each sample was reported with each sample analysis. All data were corrected to surrogate recovery and lipid normalized. OC concentrations were expressed as ng/g (ppb) on a lipid weight basis. The method detection limits (MDLs) for PCBs and OC pesticides were determined using the ultra standard mix at a low-level concentration in a procedural spike. A mean value and standard deviation for each

congener in the standards mix (Ultra Scientific) were established across 7 spikes. The MDLs are equal to 3 times the standard deviation.

For quality assurance/ quality control (QA/QC), a method blank, a lab control spike sample, and a MS/MSD pair was processed with each sample preparation batch, or every 20 samples. All sample QA/QC was within acceptable limits established by the USEPA National Coastal Assessment (USEPA, 2001). Specifically, the acceptable limit for percent recovery of a certified reference material was ±30%, the percent recovery for matrix spike and matrix spike duplicate samples were 50–120% with a relative percent difference of ±50%. The acceptance limits for surrogate recovery were 30–150%. Our analytical laboratory is certified by the National Environmental Laboratory Accreditation Conference (NELAC) and has successfully participated over the past five years in the Intercomparison Exercise for Persistent Organic Contaminants in Marine Mammal Blubber organized by the NOAA/National Institute of Standards and Technology (Gaithersburg, MD).

2.3. Statistical analysis

Statistical analyses were conducted using the SPSS 11.5 statistical package (SPSS, Inc., Chicago, IL, USA). The level of statistical significance was defined at $p \leqslant 0.05$. Data were examined for independence and normalcy, and where appropriate, were log normalized. Pearson's bivariate correlations were applied to identify relationships among contaminants. Means were compared using a Student's t-test, or using the nonparametric Mann–Whitney test. One-way analysis of variance (ANOVA) with a Bonferroni post-hoc test or regression analysis were used to determine the effects of possible confounders.

3. Results and discussion

3.1. PCBs and organochlorine pesticide concentrations

PCBs, DDT, and chlordane-related compounds (CHL) were the major persistent OCs in harbor seals relative to other contaminant groups (Fig. 2). Mirex, HCHs, and dieldrin were minor contaminants in seals relative to other groups (Table 3). HCB was detected at trace levels in these samples. Aldrin, endrin, endrin aldehyde, endrin ketone, and methoxychlor concentrations were lower than detection limits in all samples. The sum of PCB_{soc20} and DDT concentrations contributed up to 95% of the total POPs measured. PCBs were the predominant contaminant in seal blubber, with concentrations ranging from 5.7 to 151 μg/g on a lipid weight basis, followed by DDTs (1.4–57.5 μg/g), CHLs (0.3–17.6 μg/g), mirex (3.2–605 ng/g), HCHs (22–425 ng/g), and dieldrin (3–1060 ng/g) (Fig. 3). This distribution pattern (PCBs > DDTs > CHLs > mirex) is consistent with that found in our harbor seal yearling samples from Long Island, New York in 1991. The pattern also agrees with that reported in blubber of stranded harbor seals sampled in 1980 along the Massachusetts coast and in 1991–1992 from Long Island (Lake et al., 1995), as well as with that found in older male harbor seals from the St. Lawrence estuary (Hobbs et al., 2002). With the exception of dieldrin, OCs were highly correlated ($p < 0.001$) in our samples, reflecting parallel accumulation of these contaminants in the food chain. The different trend for dieldrin may reflect different sources of the compound and/or different transport mechanisms.

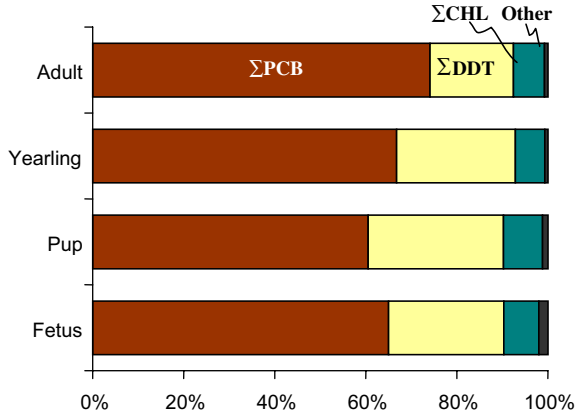

Fig. 2. Contribution of persistent organochlorines to the total OC burden in harbor seals by age.

Ten PCB congeners (52, 101, 105, 118, 128, 138, 153, 170, 180, and 187) accounted for the majority of PCB_{soc20} in blubber (Fig. 4). The $\sum$10PCB/$\sum$PCB ratio (mean ± SD) ranged from 0.98 ± 0.005 in fetuses to 0.96 ± 0.03 in adults. Ratios of the major DDT residue, *p*,*p*′-DDE to $\sum$DDT (mean ± SD) ranged from 0.8 ± 0.1 in fetuses to 0.86 ± 0.03 in adults. Pups had the highest *p*,*p*′-DDE/$\sum$DDT ratios, averaging 0.91 ± 0.03. *Trans*-nonachlor was the predominant chlordane-related compound in seal blubber samples, followed by oxychlordane, accounting for 62% and 32% of $\sum$CHL, respectively. HCH isomers were dominated by α-HCH, contributing 71% of $\sum$HCH in seal blubber, followed by β-HCH, γ-HCH (lindane), and δ-HCH.

3.2. Variations with gender, age, and condition

The accumulation of PHAHs in blubber of phocid seals is strongly influenced by age, sex, reproductive status, and condition of the animal, as well as internal physiological processes such as lipid metabolism and biotransformation (reviewed in Boon et al., 1992). For seals in areas with medium to low contamination, OC levels in blubber generally increase with age in mature

Table 3
Comparison of organochlorine residues[a,b] (ng/g lipid, wt) in blubber of northwestern Atlantic harbor seals by age

Age	Adult male	Adult female	Yearling	Pup	Fetus
n	3 Mean ± SD Range	2 Mean ± SD Range	17 Mean ± SD Range	4 Mean ± SD Range	3 Mean ± SD Range
Lipid (%)	73 ± 14 57–82	88.9 ± 5.3 85–93	63.5 ± 23.1 18–95	60.5 ± 31.6 16–88	42.7 ± 7.5 34.4–48.8
∑10 PCB	53 500 ± 39 500 (13 900–92 800)	10 700 ± 3860 (8000–13 500)	32 700 ± 39 800 (5740–149 000)	42 300 ± 22 700 (23 300–73 400)	5530 ± 1600 (3740– 6810)
∑PCB	**55 000 ± 40 800** (14 200–95 700)	**11 100 ± 3680** (8470–13 700)	**33 300 ± 40 500** (5870–151 000)	**43 000 ± 23 300** (23 600–75 100)	**5660 ± 1610** (3860–6950)
o,p′-DDD	138 ± 86 (45–213)	67 ± 6.9 (63–72)	253 ± 332 (22–1420)	230 ± 201 (130–576)	88 ± 82 (82–97)
o,p′-DDE	4.1 ± 0.8 (3.5–5)	3.3 ± 0.02 (3.25–3.28)	6.6 ± 5.3 (2.9–24)	10 ± 11 (3–27)	3.9 ± 1.9 (2.3–6)
o,p′-DDT	4.1 ± 0.8 (3.5–5)	3.3 ± 0.02 (3.25–3.28)	6.6 ± 5.3 (2.9–24)	10 ± 11 (3–27)	3.9 ± 1.9 (2.3–6)
p,p′-DDD	96 ± 48 (47–143)	78 ± 5.6 (75–82)	127 ± 212 (28.5–922)	204 ± 127 (119–391)	84 ± 56 (49–148)
p,p′-DDE	10 600 ± 5570 (4180–14 200)	3930 ± 2370 (2250–5600)	11 500 ± 12 700 (1820–50 400)	19 200 ± 18 500 (7030–46 900)	1810 ± 659 (1050–2200)
p,p′-DDT	1520 ± 963 (407–2110)	521 ± 194 (384–658)	1110 ± 1200 (151–4760)	1340 ± 830 (844–2580)	220 ± 188 (3.4–338)
∑DDT	**12 400 ± 6650** (4690–16 600)	**4600 ± 2560** (2790–6410)	**13 000 ± 14 400** (2050–57 500)	**21 100 ± 19 700** (8200–50 500)	**2210 ± 620** (1500–2680)
α-Chlordane	24 ± 35 (3.5–64.3)	12.1 ± 12.5 (3.3–20.9)	6.6 ± 5.3 (2.9–24)	10 ± 11 (3–27)	18 ± 24 (3.4–46)
γ-Chlordane	4.1 ± 0.8 (3.5–5)	3.3 ± 0.02 (3.25–3.28)	6.6 ± 5.3 (2.9–24)	10 ± 11 (3–27)	8.2 ± 6.3 (3.4–15)
Oxychlordane	1720 ± 1110 (563–2760)	228 ± 36 (202–253)	1150 ± 1420 (7.7–5720)	2110 ± 1940 (933–5000)	192 ± 164 (6–317)
Heptachlor	4.1 ± 0.8 (3.5–5)	3.3 ± 0.02 (3.25–3.28)	6.6 ± 5.3 (2.9–24)	10 ± 11 (3–27)	3.9 ± 1.9 (2.3–6)
Heptachlor Epoxide	175 ± 89 (73–227)	28.6 ± 12.6 (19.7–37.5)	190 ± 192 (16–815)	314 ± 196 (72–504)	61 ± 43 (34–110)
cis-nonachlor	48.5 ± 76.7 (3.5–137)	8.5 ± 7.4 (3.3–13.4)	93 ± 207 (3.7–886)	248 ± 210 (83–528)	25 ± 35 (3.4–66)
trans-nonachlor	3290 ± 3790 (469–7600)	482 ± 88.3 (420–545)	1780 ± 2410 (231–10 100)	3390 ± 3230 (1270–8200)	350 ± 130 (212–471)
∑CHL	**5290 ± 5020** (1120–10 900)	**775 ± 71.8** (724–825)	**3230 ± 4180** (535–17 600)	**6090 ± 5540** (2550–14 300)	**668 ± 130** (539–798)
Mirex	**325 ± 218** (136–564)	**50 ± 65** (3.3–96)	**90 ± 146** (4.3–605)	**165 ± 197** (38–460)	11 ± 10 (3.4–23)
α-HCH	74 ± 22 (58–99)	23 ± 15 (13–34)	80 ± 54 (8–197)	182 ± 116 (70–344)	34 ± 2 (32–35)
β-HCH	16 ± 10 (8–28)	3.3 ± 0.02 (3.25–3.28)	18 ± 29 (3.7–126)	17 ± 10 (6.6–27)	11 ± 10 (3.4–22)
γ -HCH	6.3 ± 3.7 (3.5–10.5)	3.3 ± 0.02 (3.25–3.28)	7.2 ± 5.2 (3–24)	11 ± 11 (3–27)	3.9 ± 1.9 (2.3–6)
δ-HCH	4.1 ± 0.7 (3.5–5)	3.3 ± 0.02 (3.25–3.28)	6.6 ± 5.3 (2.9–24)	10 ± 11 (3–27)	3.9 ± 1.9 (2.3–6)
∑HCH	**100 ± 25** (73–122)	**33 ± 15** (22.4–43.3)	**112 ± 85** (25–368)	**220 ± 144** (87–425)	**53 ± 10** (42–62)
Dieldrin	**52 ± 44** (5–93)	**11.3 ± 11.4** (3.3–19.4)	**53 ± 52.7** (3–202)	**364 ± 482** (27–1060)	80.7 ± 112 (3.4–209)
HCB	**8.2 ± 4.6** (3.5–12.8)	**6.6 ± 4.8** (3.3–10)	**7 ± 5.4** (2.9–24)	**12 ± 11** (3–27)	6 ± 2.6 (3.4–8.5)

[a] Values given are arithmetic mean ± standard deviation and range.
[b] ∑10 PCB calculated as sum of 52, 101, 105, 118, 128, 138, 153, 170, 180 and 187; ∑PCB calculated as sum of 20 individual congeners; ∑DDT calculated as sum of *o,p′*-DDD, *o,p′*-DDE, *o,p′*-DDT, *p,p′*-DDD, *p,p′*-DDE and *p,p′*-DDT; ∑CHL calculated as sum of α- and γ-chlordane, *cis*-nonachlor, *trans*-nonachlor, oxychlordane, heptachlor, and heptachlor epoxide; ∑HCH calculated as sum of α-, β-, δ-, and γ-HCH.

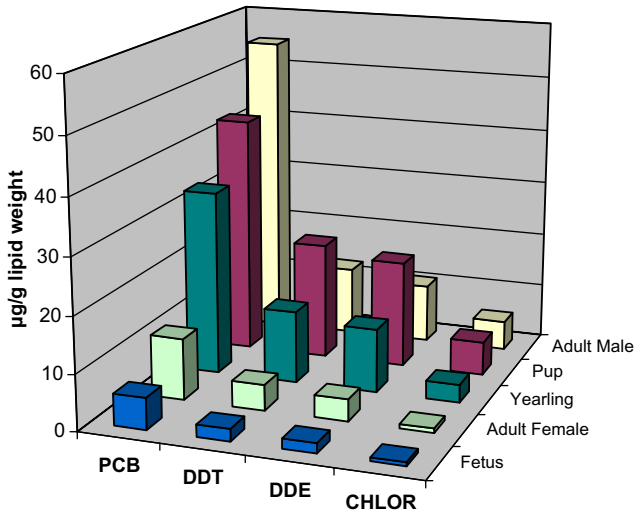

Fig. 3. Organochlorine concentrations (µg/g, lw) in northwestern Atlantic harbor seals by age.

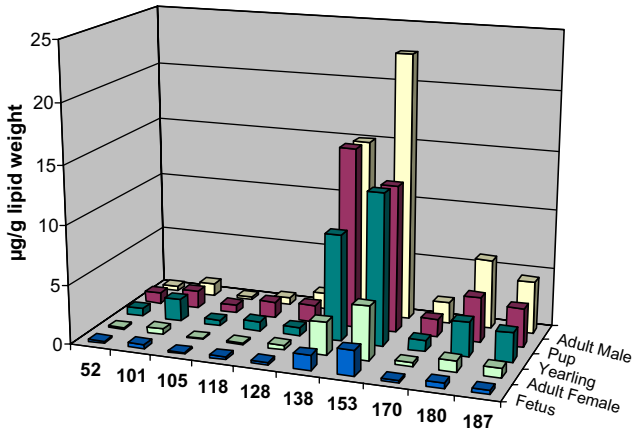

Fig. 4. Predominant PCB congener levels (µg/g, lw) in northwestern Atlantic harbor seals by age.

males but remain constant or decrease slightly in females, reflecting placental and lactational transfer of a proportion of the pollutant load to offspring (Addison and Brodie, 1977). In the present study, gender-related differences in residue levels were not observed among the younger seals but were apparent among the adults, with adult males having five-fold higher burdens of PCBs than females, three-fold higher concentrations of DDTs, and almost seven-fold higher levels of CHLs (Table 3, Fig. 3). These differences between the males and females are indicative of a contaminant exposure where females are able to keep their POP burden low by transferring an appreciable amount to their pup. This is in contrast to seals inhabiting highly contaminated environments such as the Baltic where lifetime residue burdens develop equally in both sexes because exposure levels exceed the capacity for excretion (Nyman et al., 2002).

The harbor seal pups had relatively high PCB and DDT concentrations (mean 43 and 21.1 µg/g lw, respectively), reflecting their exposure to maternal lipid stores during gestation and lactation (Addison and Brodie, 1977; Reijnders, 1980). The pups had significantly higher PCB burdens than the fetuses ($p = 0.002$), reflecting the greater importance of lactational transfer as an exposure route. In harbor seals, the average lactation period is 4-6 weeks, during which time, high levels of lipid-soluble OCs accumulate in nonpolar milk lipids, resulting in high transfer rates to nursing pups. The net effect of lactational transfer is that the lipid stores of PCBs and DDTs are often higher in pups at weaning than in their mothers (Addison and Brodie, 1977). Although we did not examine mother–pup pairs, PCB and DDT levels were four-fold higher in the pups than in the adult females; HCH and CHL levels were nearly seven- and eight-fold higher, respectively, in the pups (Table 3, Fig. 3). Lactational transfer may also pose an increased toxic risk to pups compared with that of adult exposure through feeding. As the lactating seal does not feed, the bulk of her circulatory lipids are derived from the blubber layer, rather than from chylomicra arising from lipid sources in her diet (Addison and Brodie, 1987). Thus, during the fasting period, as the mother loses weight, the nursing pup may be exposed to the more toxic PCB residues mobilized from the mother's depot lipids, as compared with lower chlorinated residues which, if the mother were feeding, would be obtained from fish.

The condition of a marine mammal is considered a measure of its energy reserves and is linked to the probability of its survival (Read, 1990). Condition indices (CIs) calculated for stranded harbor seals in this study were positively correlated with age ($R = 0.668$, $p = 0.013$), with mean CIs ranging from 57 in pups to 66 in adult males (Table 2). CIs in these seals were lower than those reported in live-captured adult harbor seals (Hobbs et al., 2002; Shaw et al., 2003), likely reflecting differences in nutritive state. Seals in this study were sampled in late spring and summer during the pupping and moulting season when animals are fasting and blubber stores are being depleted. During this period, the decrease in the proportion of blubber to the total body weight results in a release of contaminants to other body organs, creating equilibrium with increased contaminant levels in the body.

The lipid content (%) of the harbor seal blubber samples was positively correlated with condition indices ($p = 0.002$, $n = 30$) and with the exception of dieldrin, was negatively correlated with OC levels including PCBs ($p = .004$), DDTs ($p < .001$), CHLs ($p = .002$), HCHs

($p < .001$), mirex ($p = 018$), and HCB ($p < .001$). Lipid content was variable, ranging from 16% to 95%, indicating that some of the animals were in poor nutritive condition. Lipid content in samples from one pup and two yearlings was extremely low (16%, 18%, and 25%, respectively), and PCB levels in these seals were high (mean 97.1 μg/g, lw) compared with levels in the remaining samples. However, when these animals were removed from the sample, PCB levels (mean 32.3 and 23.3 μg/g, lw, in pups and yearlings, respectively) still exceeded the estimated threshold level of 17 μg PCB/g, lw in blubber for adverse effects (Kannan et al., 2000). The pups' congener profiles also varied, with lower concentrations of the hexa-congener 153 than PCB 138 in the pup with low lipid content (Fig. 4). This pup may have stranded early during lactation, and the variable PCB congener pattern could reflect the age-dependent accumulation of PCB 153 (Ruus et al., 2002) and the pup's metabolic capacity at the time of sampling (Addison et al., 1986). Given the past susceptibility of this population to viral epizootics, the elevated PCB concentrations found in these seals are of concern, particularly for the pups, taking into consideration that young seals seem to be less capable of metabolizing these compounds than adults (Addison et al., 1986; Boon et al., 1992), and are likely to be more vulnerable to their toxic effects (Shaw et al., 1999).

3.3. Temporal variation in organochlorine levels

In the northwestern Atlantic, PCB levels in harbor seals are reported to be consistently higher than DDT levels, which has been explained by a more rapid decline of DDT in the environment since these compounds were banned in the 1970s (Kennish, 1992), while PCBs are still being released from stockpiled residues (Marquenie and Reijnders, 1989). This observation is supported by the decreasing trend of the DDT/PCB ratio from 0.71 to 0.38 in harbor seal tissues between 1971 and 2001 (Table 4), which is consistent with temporal trends in many temperate areas of the northern hemisphere. In industrialized areas of Europe, PCB levels have remained constant or declined only slightly since the 1980s, reflecting an equilibrium in environmental cycling (Blomkvist et al., 1992; Nyman et al., 2002). Tanabe (1988) calculated that in the mid 1980s, only 30% of all the PCBs produced had so far dispersed into the environment. It was estimated by the late 1980s that only about 1% of all PCBs had reached the oceans while about 30% had accumulated in dumpsites and sediments of rivers, coastal zones, and estuaries (Marquenie and Reijnders, 1989). Based on their likely future dispersal into the oceans, Tanabe (1988) concluded that global PCB levels in marine biota are unlikely to decline in the near future, and certainly not before 2010–2030.

Comparison of the limited region-specific data in northwestern Atlantic harbor seals (Table 4) indicates

Table 4
Comparison of PCB and DDT residues (μg/g lipid weight) in harbor seals from the northwestern Atlantic 1971–2002

Sampling year/location	n	∑PCB	∑DDT	∑DDT/∑PCB	p,p'-DDE	p,p'-DDE/DDT	p,p'-DDT	p,p'-DDE/p,p'-DDT	Reference
1971	6	101	71.1	0.71	36.8	0.52	26.3	1.4	Gaskin et al. (1973)[a]
Boothbay Harbor Maine		(30–254)	(28–147)		(16.5–61.3)		(10–67.7)		
1980	6	17.1	–	–	15.6	–	–	–	Lake et al. (1995)[b]
Massachusetts coast		(10.4–34.7)			(9.3–31.3)				
1990–1992	9	9.5	–	–	5.9	–	–	–	Lake et al. (1995)[b]
Long Island, NY		(3.7–16.1)			(2.6–11.2)				
2001–2002	27	34.4	13.1	0.38	11.6	0.88	1.1	10.5	This study[c]
Mid-coast Maine to Long Island, NY		(5.7–151)	(1.4–57.5)		(1.2–50.4)		(0.07–4.8)		

[a] Males and females, dead, unspecified age and condition; ∑PCB calculated as Aroclor 1254 and 1260; wet weight data adapted using % lipid given by author.
[b] Males and females; unspecified age; stranded, dead; ∑PCB calculated as sum of 18 peaks; wet weight data adapted using an estimated lipid content of 70%.
[c] Males and females; varied ages (pups, yearlings, adults); stranded, dead; ∑PCB calculated as sum of 20 congeners.

that DDT and, to a lesser extent, PCB burdens have declined from the high levels reported in the 1970s (mean DDT 71.1 and PCB 101 μg/g, lw) (Gaskin et al., 1973). While comparisons with previous studies must be viewed with caution due to differences in analytical methods and sample variability (age and reproductive and health status of the animals), a significant decrease in DDT levels of ~82% ($p < 0.001$) along with a smaller decrease in PCB levels of ~66% ($p = 0.002$) can be observed over a thirty year period in harbor seal blubber, which is consistent with trends in many areas of the temperate east and west North Atlantic where these compounds were extensively used. Addison and Stobo (2001) reported an 85–90% decline in DDT residues in grey seals (*Halichoerus grypus*) from Sable Island, Nova Scotia, Canada, from the mid 1970s to 1998, while the decline in PCB concentrations was apparent only after the mid-1980s. In ringed (*Phoca hispida*) and grey seals from the highly polluted Baltic Sea, DDT levels have decreased by 72–85% since the 1970s (Nyman et al., 2002) while PCB levels showed only a minor decrease of 25 % in females and no decrease in males. A similar time trend of more rapidly declining DDT was observed in most Arctic marine mammal populations (AMAP, 2000).

The *p*,*p*′-DDE/DDT ratio is widely used to identify distribution patterns and time trends of the DDT group, wherein a ratio>0.6 is indicative of a stable system with a lack of new DDT inputs (Aguilar, 1984; Tanabe et al., 1997). In this region, the *p*,*p*′-DDE/DDT ratio in harbor seals increased from 0.52 in 1971 to 0.88 in 2001–2002 (Table 4), while concentrations of the technical residue, *p*,*p*′-DDT, decreased by 96% ($p < 0.001$) over the thirty year period. The higher *p*,*p*′-DDT levels and lower *p*,*p*′-DDE/DDT ratio in the 1971 samples are indicative of commercial DDT releases to marine and estuarine environments before uses were banned in North America in 1972. This is similar to the low *p*,*p*′-DDE/DDT ratio (0.56) reported in blubber of seals from Lake Baikal, Siberia, in 2002 (Tsydenova et al., 2004), where DDT contamination has been relatively high on a global scale (Tanabe et al., 2003). As *p*,*p*′-DDE is the "dead-end" metabolite of *p*,*p*′-DDT in seals (Addison and Stobo, 2001), the trend of increasing *p*,*p*′-DDE/DDT and declining *p*,*p*′-DDT levels is even more apparent if the ratio of *p*,*p*′-DDE/ *p p*′-DDT is considered. In northwestern Atlantic harbor seals, the ratio increased from 1.4 to 10.5 between 1971 and 2002, reflecting the aging of DDT in this ecosystem.

Lake et al. (1995) reported a decreasing trend in PCB and *p*,*p*′-DDE residues in stranded harbor seals from the northwestern Atlantic between 1980 and 1990–1992 (Table 4). While the present study found a similar decreasing trend between 1971 and 2001, PCB and *p*,*p*′-DDE levels in our 2001–2002 samples were 3.6 and 2 times higher, respectively, than those reported in the 1990–1992 samples by Lake and co-workers. This apparent discrepancy in concentrations could be due to differences in analytic methods; however, the lack of information provided on potentially confounding variables (sex, age, lipid content, and time of year samples were collected) precluded meaningful comparisons between the studies.

To estimate temporal variability over the past decade, we compared OC levels and congener and isomer patterns in blubber samples collected at the same time of year (spring and summer) in 1991 and 2001–2002, and analyzed by the same laboratory using the same analytic methods (Table 5, Fig. 5). All seals were yearlings in similar condition, although stranding locations of the animals varied. The 2001–2002 samples were collected along the coast from Maine to Long Island, while the 1991 samples were collected only in Long Island (Fig. 1). Harbor seals make seasonal migrations southward along this coast during fall and winter (Whitman and Payne, 1990), thus, despite their fidelity to pupping and breeding areas in spring and summer months, it is reasonable to assume that these seals came from similar populations. Due to the small sample size ($n = 3$) of the 1991 samples, no firm conclusions can be drawn, but the data suggest that absolute concentrations of the major contaminant groups, PCBs, DDTs, and CHLs, remained relatively stable in seal blubber over the ten-year period. Slight decreases were observed in concentrations of HCHs, mirex and dieldrin, and almost a two-fold decrease was observed in concentrations of HCB in seal blubber from 1991 to 2001–2002, which is consistent with a general decline of these compounds in the environment.

Table 5
Comparison of organochlorine residues (ng/g, lipid weight) in yearling harbor seals from the northwestern Atlantic 1991 and 2001–2002

Sample date	*n*	∑PCB	∑DDT	*p*,*p*′-DDE/ *p*,*p*′-DDT	*p*,*p*′-DDE/ ∑DDT	∑CHL	Mirex	HCH	Dieldrin	HCB
1991	3	28 300 ± 17 600 14 200–48 100	11 400 ± 7610 4700–19 700	11.3 ± 3 9–14.7	0.89 ± 0.02 0.89–0.92	2730 ± 2100 1120–5110	110 ± 61 40–155	118 ± 82 68–212	64 ± 24 44–91	7 ± 5.4 3–24
2001–2002	17	33 300 ± 40 500 5870–151 000	13 000 ± 14 400 2100–57 500	10.2 ± 2.16 7.32–15	0.88 ± 0.02 0.85–0.92	3230 ± 4180 535–17 600	90 ± 146 4.3– 605	112 ± 85 25–368	53 ± 53 3–202	4.6 ± 1.4 3.5–6.3

Values given are arithmetic mean ± standard deviation and range.

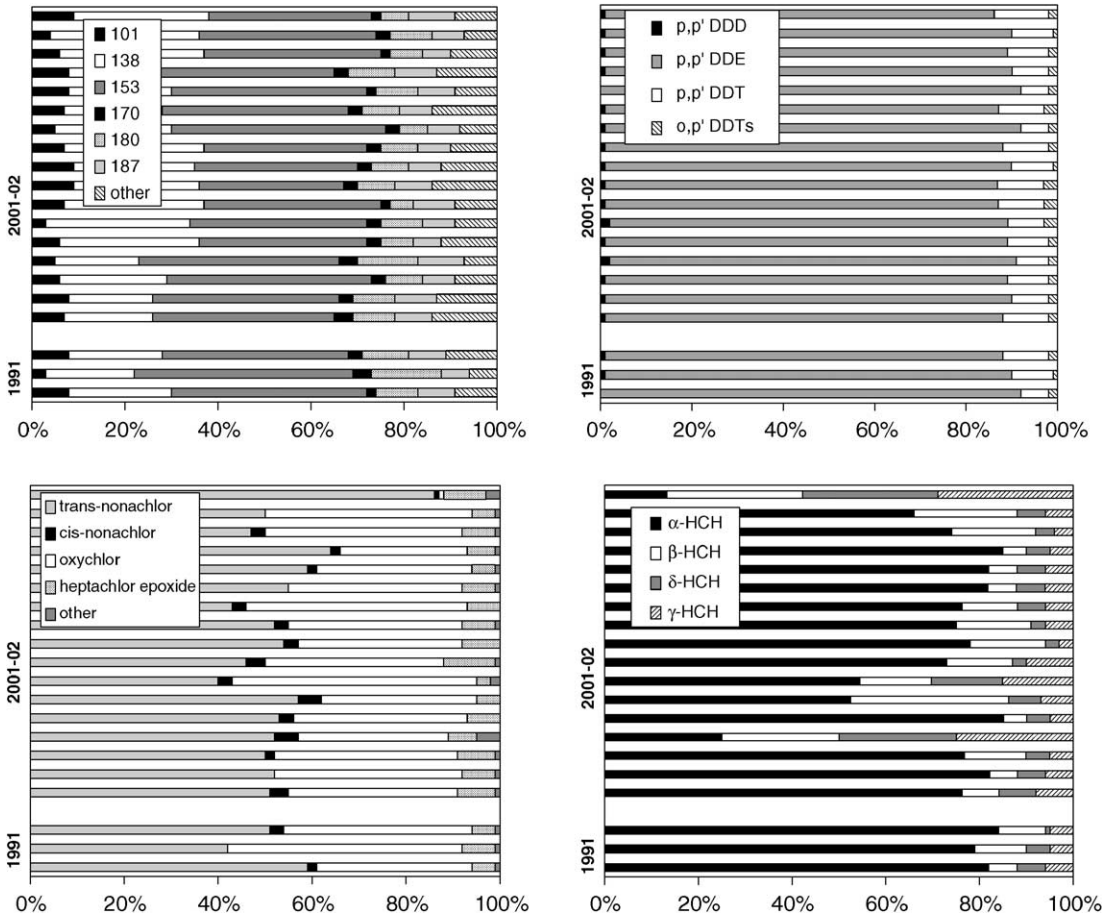

Fig. 5. Comparison of isomer and congener composition of PCBs, DDTs, CHLs and HCHs in harbor seal yearlings samples in 1991 and 2001–2002.

Congener compositions appeared to remain constant for PCBs, DDTs, and CHLs between 1991 and 2001–2002, while differences were observed in the HCH isomer profiles (Fig. 5). PCB congeners 101, 138, 153, 180, 170, and 187 were the predominant congeners in all samples and no differences were found in the proportions of tri-, tetra-, penta-, hexa-, and heptachlorobiphenyls between 1991 and 2001–2002. ∑DDT compositions also remained constant in these samples over the ten-year period. The extremely persistent DDT metabolite, *p*,*p*′-DDE, was the predominant residue in all samples, contributing 87–92% and 85–92% of the ∑DDT concentration in 1991 and 2001–2002, respectively. The stable *p*,*p*′-DDE/DDT ratios in these samples (0.89 and 0.88 in 1991 and 2001–2002, respectively) indicate that the northwestern Atlantic is a "weathered" system, similar to the St. Lawrence Estuary where *p*,*p*′-DDE/DDT ratios in harbor seals ranged from 0.84 to 0.81 in male and female seals, respectively (Bernt et al., 1999). The *p*,*p*′-DDE/*p*,*p*′-DDT ratio also remained stable in our samples between 1991 and 2001–2002, indicating the lack of recent inputs to the ecosystem. The stable ratios of *p*,*p*′-DDE/∑DDT and *p*,*p*′-DDE/*p*,*p*′-DDT over the period suggest that the decline of DDT residues in marine biota which began in the 1970s may have slowed or reached equilibrium in the northwestern Atlantic during the 1990s.

The composition of CHL-related compounds also remained similar in harbor seal blubber samples between 1991 and 2001–2002, reflecting the widespread historical application of this extremely persistent pesticide on agricultural and urban soils for ~40 years, including lawns and golf courses on the coast of New England (Phillips and Birchard, 1991). It is estimated that 25–50% of CHL produced in the US since the 1940s still remains unaltered in the environment. CHL readily biomagnifies in aquatic food chains (Norstrom et al., 1988), although the compositional pattern of CHL compounds is known to differ among different trophic levels and species (Kawano et al., 1988). For example, at the highest trophic level, polar bears retain only the metabolite oxychlordane (Hargrave et al., 1992). In harbor seal blubber samples, the ratio of the most abundant CHLs, trans-nonachlor (contributing ~40–60%), and oxychlordane (~30–50%) to total CHL did not change from 1991 to 2001–2002, and the contribution of heptachlor epoxide to ∑CHL (~10%) also remained the same, with one exception. In an animal sampled in 2001–2002, oxy-

chlordane was present only at trace levels, while trans-nonachlor contributed ~90% of the ∑CHL, followed by heptachlor epoxide. *Cis*-nonachlor, *cis*-chlordane (α), trans-chlordane (γ), and heptachlor were present at trace levels in all seal samples.

Some differences were observed in the composition of HCH isomers in seal blubber between 1991 and 2001–2002. Because of their stability, relatively higher volatility than other POPs, and global use for many years, persistent HCH isomers, particularly α-HCH and β HCH, appear to be universal environmental contaminants, the levels of which are only slowly decreasing in the environment (ATSDR, 2003). α-HCH dominated all the 1991 samples, comprising ~80% of ∑HCH, followed by β-HCH, γ-HCH (lindane), and δ-HCH. Technical-grade HCH, which contained a high proportion (60–70%) of the α-HCH isomer, was banned for production and use in the US in 1976; however, lindane (99.8% γ-HCH) is still imported by the US and formulated as a pesticide on seed crops and as a scabicide in humans (US EPA, 2002). Whereas the γ-HCH isomer is less persistent and can be rapidly metabolized in animals, β-HCH is considered to be the most persistent and bioaccumulative isomer of the HCHs, and thus a composition dominated by α-HCH suggests that these seals may be exposed to an HCH source such as a hazardous waste site at which technical-grade HCH was disposed. A similar composition dominated by α-HCH found in polar bears (Norstrom et al., 1988) and ringed seals (Muir and Norstrom, 2000) from the Canadian Arctic was attributed to sources from China and Southeast Asia (Li et al., 1996). A higher proportion of β-HCH/∑HCH was observed in 53% of our 2001–2002 harbor seal samples compared with that of the 1991 samples; the ratio of γ-HCH/ ∑HCH was also greater in 23% of the 2001–2002 samples. The increasing ratio of β-HCH/ ∑HCH along with the decreasing concentrations of HCHs in seal blubber over the ten-year period is consistent with a gradual decline of HCH in the ecosystem. The increasing ratio of γ-HCH/ ∑HCH in these samples likely reflects the ongoing importation and use of lindane in the region.

3.4. Comparison of organochlorine residues in stranded pinnipeds from various regions

OC residues detected in harbor seals from the northwestern Atlantic were compared with levels reported in stranded seals from various regions, including carcasses of seals that died during epizootics (Table 6). Geographic comparisons are necessarily limited because of the numerous sources of variability across the studies, particularly methodological differences. Other factors that strongly influence contaminant distributions (age, sex, reproductive status, condition) are often omitted, making interpretation of the data difficult. Therefore, only a few specific comparisons are discussed here together with the general trends.

On a global scale, the highest PCB and DDT loads are found in seals inhabiting the heavily industrialized mid-latitude areas of Europe, North America, and Asia, with the lowest levels occurring in the polar regions of both hemispheres (Iwata et al., 1994; Aguilar et al., 2002; Tanabe et al., 2003). Luckas et al. (1990) observed a decreasing gradient in OC levels from the northern hemisphere to the southern hemisphere, with the highest ∑PCB levels (~85 μg/g, lw) occurring in harbor seals from the Baltic and the Dutch Wadden Sea. High PCB levels (mean 72.6 μg/g, lw) were also observed in Caspian seals (*Phoca caspica*) that died during a canine distemper virus (CDV) epidemic (Kajiwara et al., 2002). Harbor seals from the northwestern Atlantic are in the middle of the global contamination spectrum, with mean ∑PCB concentrations of 55 μg/g, lw, in the adult males. These levels are lower than those in seals from the Baltic, the Wadden Sea, and the Caspian Sea, but exceeded the levels reported in harbor seals from the coasts of Denmark, eastern England, northern Ireland, and the southern coast of Norway during the 1988 morbillivirus epizootic. Levels in the adult male harbor seals were much higher than those reported in Mediterranean monk seals (*Monachus monachus*) from the relatively unpolluted Sahara coast (Atlantic Ocean), North Africa, but were approaching the concentrations (mean PCB 64.3 μg/g, lw) found in monk seals from the northern coast of Africa (western Mediterranean), an area considered to be one of the most highly polluted in the world (Borrell et al., 1997).

DDT residues (mean 12.3 μg/g, lw) were lower in adult male harbor seals from the northwestern Atlantic than those reported in harbor seals from the polluted Baltic Sea, western Mediterranean monk seals, and northern elephant seal yearlings from central California, but were two- to three-fold higher than levels in harbor seals from the coasts of Denmark, Germany, eastern England, and northern Ireland and four-fold higher than those in monk seals from the Sahara coast. While DDT levels have declined in many parts of the developed world, DDT residues (143.4 μg/g, lw) in the Caspian seals were ten times higher than those in northwestern Atlantic harbor seals, likely reflecting recent illegal uses of this pesticide in the former USSR (Federov, 1999).

Pups and yearling harbor seals in this study had much higher PCB concentrations (mean 34.3 μg/g, lw) than those reported in young seals from the US Pacific coast. Levels were two-fold higher than those reported in stranded harbor seal pups from southern Puget Sound, Washington, an area considered relatively polluted (Shaw, 1998; Hong et al., 1996), and an order of magnitude higher than levels reported in stranded, rehabilitated harbor seal pups from the central California

Table 6
Comparison of organochlorine residues (ng/g lipid weight) in pinnipeds from various regions

Species	Location	Year sampled	*n*	Lipid (%)	Age/sex	PCBs	DDTs	DDT/PCB	CHLs	HCHs	Reference
Harbor Seal	NW Atlantic	2001–2002	3	73	Adult >4 years	55000	12400	0.22	5290	100	This study[a]
(*Phoca vitulina*)				57–82	M	14200–95700	4690–16600		1120–10900	73–122	
Harbor Seal	NW Atlantic	2001–2002	21	63	Pup/yearling	34300	14200	0.41	3710	134	This study[b]
(*Phoca vitulina*)				16–95	M/F	5870–151000	2050–57500		534–17600	25–425	
Harbor Seal	S. Puget Sound	1990–1991	4	78	Pup < 1 week	16900	3750	0.22	NR	NR	Shaw (1998)[c]
(*Phoca vitulina*)	Washington			69–92	M/F	11800–20600	2040–5160				
Harbor Seal	Central coast	1990–1991	11	79	Pup ~4 weeks	3340	3970	1.19	NR	NR	Shaw (1998)[d]
(*Phoca vitulina*)	California				M/F	1400–5290	715–13500				
Harbor Seal	Baltic Sea	1990	9	NR	NR	84700	22500	0.27	NR	90	Luckas
(*Phoca vitulina*)	Sweden										et al. (1990) [e]
Harbor Seal	Wadden Sea	1990	32	NR	NR	85300	3900	0.05	NR	39	Luckas
(*Phoca vitulina*)	W. Germany										et al. (1990) [f]
Harbor Seal	Wadden Sea	1988	3	NR	Adult 5 years	41500	3010	0.07	NR	NR	Storr-Hansen
(*Phoca vitulina*)	Denmark				M						and Spliid (1993)[f]
Harbor Seal	E. England	1988	9	72	<1–12 years	33800	6510	0.19	NR	32	Law et al. (1989) [g]
(*Phoca vitulina*)				51–89	M/F	16700–57900	3280–13900			17–84	
Harbor Seal	N. Ireland	1988	44	57.3	Pup to >2 years	49000	4800	0.1	383	66	Mitchell and
(*Phoca vitulina*)				22.6–92	M/F						Kennedy (1992) [h]
Harbor Seal	South Coast	1988	10	73.1	<1-Adult	15000	NR	NR	NR	NR	Bernhoft and
(*Phoca vitulina*)	Norway			59.2–87	M	3400–29000					Skaare (1994)[i]
Elephant Seal	Central coast	1991–1992	4	74	Yearling	19200	35600	1.85	2940	380	Kajiwara
(*Mirounga augustirostris*)	California			19–93	M/F	5000–58000	830–110000		830–8600		et al. (2001)[j]
Mediterranean Monk Seal	Sahara Coast	1992–1994	31	18.7	<1-Adult	7090	2910	0.4	NR	NR	Borrell
	(Atlantic Ocean)										et al. (1997)[k]
(*Monachus monachus*)	No. Africa			0.2–56	M/F	260–23100	60–10700				
Mediterranean Monk Seal	No. Coast Africa	1991	2	6.1	Adult 3–10 years	64300	19000	0.3	NR	NR	Borrell
(*Monachus monachus*)	(Western			2–10	M/F	21300–107000	11300–26700				et al. (1997)[l]
	Mediterranean)										
Caspian Seal	Caspian Sea	2000	6	73	<1–27 years	72600	143000	2.0	3720	6890	Kajiwara
(*Phoca caspica*)	Azerbaijan,			37–84	M	3500–320000	9500–470000		210–14000	890– 9900	et al. (2002)[m]
	Kazakhstan,										
	Turkmenistan										

Caspian Seal (*Phoca caspica*)	Caspian Sea Azerbaijan, Kazakhstan, Turkmenistan	2000	7	79 25–96	<1–23 years F	24 200 2400–120 000	47 300 6300–230 000	2.0	1010 210–4800	1670 690–5300	Kajiwara et al. (2002) [n]

Values given are arithmetic means, with ranges, unless otherwise noted.
M: male, F: female.
NR: not reported.
[a] Stranded, dead; ∑PCB calculated as sum of 20 congeners.
[b] Stranded, dead; ∑PCB calculated as sum of 20 congeners.
[c] Stranded, dead; ∑PCB calculated as sum of 73 congeners; ∑DDT given as *p,p′*-DDE.
[d] Stranded, sampled alive during rehabilitation (biopsy) or at necropsy; ∑PCB calculated as sum of 73 congeners; ∑DDT given as *p,p′*-DDE.
[e] Unspecified sex, age, and condition, dead; ∑PCB calculated from PCB 138 and 153 as Clophen 60; ∑HCH calculated as sum α- and γ-HCH; DDT calculated as sum of *p,p′*-DDD, *p,p′*-DDE and *p,p′*-DDT.
[f] Diseased, died during morbilli virus epidemic; ∑PCB calculated as sum of 31 congeners; ∑DDT given as *p,p′*-DDE; wet weight data adapted using % lipid given by author.
[g] Diseased, died during morbilli virus epidemic; ∑PCB calculated as Aroclor 1254; DDT calculated as sum of *p,p′*-DDE and *p,p′*-DDT; ∑HCH given as α-HCH; wet weight data adapted using % lipid given by author.
[h] Diseased, died during morbilli virus epidemic; ∑PCB calculated as Aroclor 1254; ∑HCH given as α-HCH.
[i] Diseased, died during morbilli virus epidemic; ∑PCB calculated as sum of 21 congeners, data given as median and ranges.
[j] Stranded, diseased, died during rehabilitation, sampled at necropsy.
[k] Stranded, dead; ∑PCB calculated as sum of 18 individual peaks.
[l] Stranded, dead; ∑PCB calculated as sum of 18 individual peaks.
[m] Stranded, died during CDV epizootic.
[n] Stranded, died during CDV epizootic.

coast (Shaw, 1998). While DDT levels (mean 14.2 μg/g, lw) in the younger seals in this study were higher than levels reported in both the Puget Sound and California harbor seal pups, the higher DDT/PCB ratio in the California pups (1.2) compared with ratios in the northwestern Atlantic pups (0.4) and Puget Sound pups (0.2), reflects the higher prevalence of DDT contamination in central California associated with massive agricultural releases in the 1950s and 1960s (Kennish, 1992). PCB levels in the young harbor seals in this study were nearly two-fold higher than those reported in yearling northern elephant seals (*Mirounga augustirostiris*) from central California with advanced skin disease (Kajiwara et al., 2001), while DDT levels in the elephant seals were more than two-fold higher. The higher DDT/PCB ratios observed in the northern elephant seals and Caspian seals (1.85 and 2.0, respectively) reflect the extent of historical and recent DDT contamination in these areas.

Compared with DDTs, PCBs, and CHLs, which tend to remain concentrated near their sources, more volatile compounds such as HCHs are subject to long-range transport and high concentrations of these compounds are found in polar regions (Iwata et al., 1994). On a global scale, the levels of HCHs were an order of magnitude higher in Caspian seals (mean 6.8 and 1.7 μg/g, lw in males and females, respectively) than those in seals from all other areas, reflecting the continued use of technical HCH in the former USSR until it was banned in the early 1990s (Li et al., 1996). Adult male harbor seals from the northwestern Atlantic had the highest levels of CHLs (mean 5.3 g/g, lw), although concentrations reported in Caspian seals and northern elephant seal yearlings from California were in a similar concentration range, reflecting the ubiquity and extreme persistence of these compounds in the global environment.

3.5. Toxicological implications

The levels of persistent OCs found in northwestern Atlantic coast harbor seals are at the middle of the contamination spectrum on a global scale. PCB levels detected in the adult males exceed or are in the lower range of concentrations associated with tumors and uterine stenosis in northern European harbor, ringed, and grey seals (Reijnders, 1980; Helle et al., 1976a,b; Baker, 1989). PCB burdens found in our adult male, pup, and yearling harbor seals exceed the estimated threshold level of 17μg PCB/g lw in blubber for adverse effects on immune function (Kannan et al., 2000), and fall within the estimated threshold level of 25–77 μg PCB/g lw for reproductive effects in marine mammals (AMAP, 2000). Moreover, PCB burdens in the pups are an order of magnitude higher than the concentrations associated with significantly reduced lymphocyte proliferative responses and plasma retinol and thyroid

hormone levels in stranded harbor seal pups from the central California coast (Shaw et al., 1999). These observations, together with reports of at least two large-scale viral epizootics among these seals since the late 1970s, suggest that the population is currently at risk for adverse health effects. The harbor seal population in this region has increased since the 1970s when bounty hunting was discontinued, but population growth rates (pup production) have declined in some areas for reasons that are poorly understood (Gilbert and Guldager, 1998). Although the present study was limited by a small sample size distributed over a large geographic area, the toxicological impacts of the current OC levels in these seals would be expected to be considerable, particularly among the pups, leading to developmental deficits and compromised immune resilience, which in turn, may increase susceptibility to infections.

4. Conclusions

To our knowledge, these are the first extensive data reported on OC residues in harbor seals from the northwestern Atlantic coast in 25 years. Given their relatively high tissue burdens of PCBs and chlorinated pesticides, harbor seals inhabiting this region accumulate levels that place them at risk for adverse health effects including effects on reproduction and immune and endocrine function. Levels in the pups resulting from perinatal exposure are of special concern given that young seals appear to be vulnerable to immune- and endocrine-disrupting effects of PCBs and DDT when levels are an order of magnitude lower. In view of the past vulnerability of these seals to viral epizootics, there is a clear need for further research to ascertain the average levels of a broad range of OCs and other halogenated compounds in the population in conjunction with an assessment of immune status, reproductive rates, and overall health.

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