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Hexachlorocyclohexane (HCH) Isomers and Chiral Signatures of α-HCH in the Arctic Marine Food Web of the Northwater Polynya

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Concentrations of hexachlorocyclohexane (HCH) isomers (α, β, and γ) and enantiomer fractions (EFs) of α-HCH were determined in the Northwater Polynya Arctic marine food web. Relative food web structure was established using trophic level models based on organic δ^{15}N values. Concentrations of HCH in the samples collected, including water, sediment, benthic invertebrates (four species), pelagic zooplankton (six species), Arctic cod, seabirds (seven species), and ringed seal, were in the range previously reported for the Canadian Arctic. The relative proportion of the HCH isomers varied across the food web and appeared to be related to the biotransformation capacity of each species. For invertebrates and fish the biomagnification factors (BMFs) of the three isomers were >1 and the proportion of each isomer and the EFs of α-HCH were similar to water, suggesting minimal biotransformation. Seabirds appear to readily metabolize γ- and α-HCH based on low BMFs for these isomers, high proportions of β-HCH (62−96%), and high EFs (0.65−0.97) for α-HCH. The α- and β-HCH isomers appear to be recalcitrant in ringed seals based on BMFs >1 and near racemic EFs for α-HCH. The β isomer appears to be recalcitrant in all species examined and had an overall food web magnification factor of 3.9. EFs of α-HCH and the proportion of β-HCH in Σ-HCH in the food web were highly correlated ($r^2 = 0.92$) suggesting that EFs were a good indicator of a species capability to biotransform α-HCH.

Introduction

Hexachlorocyclohexane (HCH) has been used as a pesticide since 1942 when the insecticidal properties of the γ isomer were discovered. Technical HCH consists of an approximate composition of 60−70% α, 5−12% β, 10−15% γ, 6−10% δ, and 3−4% ε (*1*). The δ and ε isomers are not commonly observed in biological samples. Although γ is the isomer with the dominant insecticidal properties, the remaining isomers have toxic properties (*2*). The technical mixture, also known as benzene hexachloride (BHC) was banned in Canada and the United States in 1971 in 1978, respectively, but is still used in some developing countries. Currently lindane, consisting of 90−100% of the γ isomer, is still registered for use in many countries, including Canada and the United States.

Persistent organic pollutants (POPs), which include HCH isomers, are dispersed by long-range atmospheric transport to higher latitudes. At temperate and tropical temperatures, where the majority of POPs are used, they volatilize and then recondense to the surface at colder polar temperatures, a process termed "the cold-condensation effect" (*3*, *4*). HCH is a classic example of this process because water concentrations of HCH are highest in the Arctic despite the fact that it has never been used at these higher latitudes. Furthermore, HCH concentrations in abiotic and lower trophic level biotic samples collected in the Arctic are often greater than all other POPs combined (*5*). Historical changes in the HCH products used (technical verse pure γ-HCH) and slight differences in the physical chemical properties and susceptibility to environmental degradation of the HCH isomers have resulted in the use of HCH isomer ratios to trace environmental phenomena. For example, Rice and Shigaev (*6*) used ratios of the α- and γ-isomers to identify ocean currents.

Of the five isomers commonly found in technical HCH, only the α isomer is chiral. Chiral pollutants exist in two forms as optical isomers called enantiomers. Enantiomers have identical physical-chemical properties and abiotic degradation rates, but can have different rates of biotransformation (*7*). The chemical manufacturing process results in a mixture containing approximately 50% of each chiral compound, termed racemic. Selective biotransformation of one chiral component over another can occur and result in an enantiomeric enrichment (*7*). This selective enrichment originates from the inability of one of the enantiomers to meet the steric requirements of an enzyme or receptor site or in the differences of the resulting chemical properties caused by the coupling of enantiomers with active sites (*8*). This resulting selective accumulation of a single enantiomer can provide information on fate and dynamics of the chemical and may have significant toxicological ramifications (*7*). It has been proposed that comparison of enantiomeric ratios (ERs) may provide information on trophic transfer of contaminants in a food web (*9*). In some instances, this technique could have similar potential to stable isotope analysis (*10*).

The Northwater Polynya (NOW) in northern Baffin Bay, an area of year round open water (*11*), is the largest and most productive polynya in the Canadian Arctic and supports large populations of seabirds and marine mammals. An extensive multidisciplinary study on the NOW afforded the opportunity to collect a large number and range of biota (zooplankton, fish, seabirds, and a marine mammal) within the same area and time and to assemble a comprehensive food web to examine trophic transfer of HCHs. The Arctic provides an excellent opportunity to examine food-web relationships because the food webs are less complicated and there are very limited local point sources of pollution. This paper examines HCH isomer concentrations in the Arctic marine food web of the NOW and compares the food web transfer of individual isomers and the utility of using EFs of α-HCH to predict food web structure and metabolic differences between species.

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TABLE 1. Trophic Level (based on tissue $\delta^{15}N$ Values), Lipid Content, Concentrations of HCH Isomers, and Enantiomeric Fractions of α-HCH in Various Samples Collected from the NOW in 1998 (mean ± 1 SE)[a]

sample[b]	*n*	trophic level	lipid	α-HCH	β-HCH	γ-HCH	ΣHCH	*n* (EF)	EF (+α)
water	19	N/A	N/A	1.1 ± 0.07	0.07 ± 0.01	0.2 ± 0.01	1.4 ± 0.1	19	0.45 ± 0.01
sediment	14	N/A	N/A	0.10 ± 0.01	0.03 ± 0.004	0.04 ± 0.004	0.16 ± 0.004	3	0.39 ± 0.01
Benthic invertebrates									
A. nugax	5	2.6 ± 0.2	2.5 ± 0.3	315.9 ± 109.6	nd	31.7 ± 7.2	347.6 ± 115.4	0	N/A
basket star	3	2.5 ± 0.1	8.5 ± 1.2	33.5 ± 8.4	nd	7.1 ± 2.1	40.7 ± 10.4	1	0.41
starfish	2	2.6 ± 0.1	1.6 ± 0.2	1.0 ± 0.5	0.4 ± 0.4	0.3 ± 0.04	1.7 ± 1.0	1	nd
clam	2	1.3 ± 0.1	2.2 ± 0.0	0.7 ± 0.4	0.3 ± 0.3	0.1 ± 0.1	1.1 ± 0.7	1	nd
zooplankton									
M. occulata	7	2.0 ± 0.02	5.0 ± 0.5	7.4 ± 1.7	0.9 ± 0.4	1.9 ± 0.4	10.3 ± 2.4	1	0.34
Sagitta spp.	6	1.8 ± 0.1	2.1 ± 0.3	7.7 ± 2.1	0.9 ± 0.5	2.5 ± 0.6	11.1 ± 2.8	1	0.43
E. glacialis	3	2.3 ± 0.03	5.4 ± 0.3	20.4 ± 1.0	nd	4.2 ± 0.3	24.6 ± 1.2	0	N/A
C. hyperboreus	20	1.2 ± 0.1	6.3 ± 0.7	24.9 ± 6.7	0.5 ± 0.2[c]	5.5 ± 1.5	30.5 ± 7.7	3	0.42 ± 0.01
M. longa	3	1.7 ± 0.1	2.1 ± 0.1	18.6 ± 2.7	nd	17.7 ± 0.6	36.2 ± 3.3	1	nd
T. libellula	4	1.9 ± 0.04	2.2 ± 0.4	45.9 ± 7.5	nd	36.4 ± 12.0	82.3 ± 13.9	1	0.43
fish									
arctic cod	8	2.9 ± 0.04	1.2 ± 0.3	39.8 ± 7.8	27.1 ± 4.7	23.3 ± 4.6	90.2 ± 13.7	3	0.45 ± 0.01
seabirds									
dovekie	8	2.6 ± 0.05	66.0 ± 1.5	66.7 ± 13.6	138.5 ± 9.6	16.8 ± 2.9	222.0 ± 19.9	6	0.65 ± 0.05
thick-billed murre	9	3.2 ± 0.04	60.0 ± 2.2	20.1 ± 1.7	57.7 ± 8.9	6.7 ± 0.5	84.5 ± 9.6	6	0.85 ± 0.02
black guillemot	7	3.5 ± 0.1	60.0 ± 6.0	76.0 ± 10.0	199.0 ± 36.8	10.0 ± 1.8	285.0 ± 46.7	5	0.92 ± 0.01
black-legged kittiwake	8	3.1 ± 0.05	72.4 ± 3.9	6.9 ± 0.9	36.0 ± 5.0	4.4 ± 0.6	47.3 ± 6.3	5	0.86 ± 0.02
glaucous gulls	10	3.9 ± 0.07	71.6 ± 4.5	14.9 ± 4.0	424.2 ± 49.1	3.6 ± 0.5	442.7 ± 51.9	10	0.97 ± 0.01
ivory gull	4	3.3 ± 0.09	81.1 ± 5.0	11.4 ± 3.4	127.5 ± 31.3	4.1 ± 0.8	143.0 ± 32.7	4	0.93 ± 0.02
northern fulmar	10	3.3 ± 0.03	71.9 ± 4.0	19.8 ± 1.5	41.2 ± 4.5	4.1 ± 0.6	65.1 ± 5.8	6	0.83 ± 0.02
marine mammal									
ringed seal	57	3.8 ± 0.02	88.7 ± 1.5	99.5 ± 8.1	43.8 ± 4.9	7.2 ± 1.0	150.5 ± 13.1	57	0.51 ± 0.05

[a] Concentrations are nanograms per liter (ng/L) in water, nanograms per gram (ng/g) (dry weight) in sediment and nanograms per gram (ng/g) (lipid) in biota (nd = not detected; N/A = not applicable or analyzed). [b] Invertebrate, zooplankton and fish samples were whole body, seabird and seal samples were fat (or blubber). [c] Only four samples of *C. hyperboreus* had measurable amounts of β-HCH.

Methods and Materials

Sample Collection. Samples were collected during the April−July 1998 voyage of the CCGV Pierre Raddisson. Water samples were collected from 2 m below the surface, with a submersible pump, into 40 L stainless steel cans on the deck of the ship. These cans were solvent washed (acetone and hexane) prior to collection and sealed until extracted. Samples were collected 4−5 m off the bow of the ship, with the ship positioned into the prevailing water current to minimize contamination. Zooplankton samples were obtained from vertical net tows (bottom to surface) using large zooplankton nets (1 m^2, 520 μm mesh). Samples, consisting of numerous whole individuals, included the species *Calanus hyperboreus*, *Euchaeta glacialis*, *Metridia longa*, *Mysis occulata*, *Themisto libellula*, and *Sagitta spp*. Samples of the amphipod *Anonyx nugax*, a benthic invertebrate, were obtained using bait traps, containing squid or mackerel wrapped in *nilex* mesh to prevent feeding, on the ocean floor for 8−12 h. Sediment and other benthic invertebrates including basket stars, (*Gorgonocephalus arcticus*), starfish (*Ctenodiscus crispatus*), and clams (*Yoldia thraciaeformis*) were collected from the sediment surface of box cores taken in July. Arctic cod (*Boreogadus saida)* were opportunistically sampled when observed swimming near the surface in broken ice at one location in May. Seabirds were collected opportunistically by shotgun from a zodiac. Subcutaneous fat (for HCH analysis) and muscle (for stable isotope analysis) tissue was collected from seven seabird species, including dovekie (*Alle alle*, three male, five female), thick-billed murre (*Uria lomvia*, four male, five female), black guillemot (*Cepphus grylle*, seven male, one female), black-legged kittiwake (*Rissa tridactyla*, three male, five female), ivory gull (*Pagophila eburnea*, one male, three female), glaucous gull (*Larus hyperboreus*, four male, six female), and northern fulmar (*Fulmaris glacialis*, five male, five female). Blubber (HCH analysis) and muscle (stable isotope analysis) tissue from ringed seals (*Phoca hispida*, 28 male, 29 female) were obtained from Inuit hunters from Grise Fijord, Canada, and Qânâq, Greenland, during the spring of 1998. The number of samples analyzed is presented in Table 1.

Chemicals and Standards. All solvents (pesticide grade) and sodium sulfate (Na_2SO_4) were obtained from BDH (Toronto, ON, Canada). Pesticide grade Florisil, 60−100 mesh was obtained from the Floridin Corp. (Berkeley Spring, WV). Biobeads SX-3 used in the GPC column was purchased from Analytical Biochemistry Laboratories Ltd., (Columbia, MO). Individual enantiomeric standards of (+) and (−) α-HCH were obtained from EQ Laboratories (Atlanta, GA).

Extraction and Cleanup. Because of the variety of samples a number of different validated extraction techniques were employed. For all analyzes, blanks samples were analyzed after every 6−10 samples using identical extraction and clean up procedures.

HCHs were extracted from water samples on the ship. Each sample consisted of ~80 L (two 40 L stainless steel cans). Equal amounts of a recovery standard 2,4,6-trichlorobiphenyl (PCB 30) and δ-HCH were spiked into each stainless steel can and stirred with a stainless steel rod prior to extraction. Water was pumped through oven-baked filters and then extracted with XAD-2 resin. The XAD-2 resin was kept at 2 °C until analyzed back at the lab. HCHs were extracted from the resin using a Soxhlet extractor with methanol and dichloromethane (DCM). The methanol and DCM were exchanged with hexane, cleaned up and fractionated on silica gel. The percent recoveries of PCB 30 and δ-HCH in the water samples were 77 ± 2.7 (mean ± 1 SE) and 82 ± 2.3, respectively.

Samples of zooplankton and the benthic invertebrate, *A. nugax*, consisted of composite samples of numerous individuals. Clam samples consisted of the soft tissue from two individuals. Starfish and basket stars were 10 g subsamples of the soft tissue of one individual. Samples were freeze-dried, spiked with two internal standards (PCB 30 and OCN) and extracted with DCM/hexane (1:1) using a Dionex ASE

200 accelerated solvent extractor (Dionex Canada Ltd., Oakville, ON, Canada). Sulfur containing compounds were removed from the sediment samples by the addition of reduced copper. A fraction of the zooplankton and benthic invertebrate eluants were used to determine lipids gravimetrically, and lipids were subsequently removed from the samples by gel permeation chromatography (GPC). The percent recoveries of PCB 30 and OCN in sediment were 94 ± 5.8 and 86 ± 3.3, respectively, and in zooplankton and benthic invertebrates were 76 ± 3.2 and 77 ± 2.9, respectively.

Arctic cod, seabirds and ringed seal samples were extracted using the methods of Norstrom et al. (*12*), with slight modifications. Arctic cod samples were subsamples of the whole body, seabird, and ringed seal samples were fat or blubber. Samples were ground with anhydrous sodium sulfate and spiked with internal standards. The Arctic cod and seabird samples were spiked with three internal standards [2,2′,3,4,4′,5,6,6′-octachlorobiphenyl (PCB 204), OCN, and δ-HCH]. Ringed seal samples were spiked with a series of ^{13}C-labeled chlorobenzenes (tetra, penta, and hexachloro) and PCB congeners (PCBs 28, 52, 118, 153, 180, and 194). Samples were extracted with 100 mL (1:1) dichloromethane (DCM)/hexane and fraction of the eluant was used to determine lipids gravimetrically. Lipids were subsequently removed from the samples by gel permeation chromatography (GPC). The percent recoveries of PCB 204, OCN, and δ-HCH in Arctic cod were 90 ± 0.4, 90 ± 0.5, and 54 ± 3.2, respectively, and in seabirds were 89 ± 1.5, 100 ± 1.8, and 73 ± 2.2, respectively. The mean percent recovery of the ^{13}C internal standards in the ringed seals was 83 ± 1.6.

The lipid-free (biota) and sulfur-free (sediment) eluates were evaporated to 1 mL and applied to a Florisil column (8 g, 1.2% deactivated). For the sediment, zooplankton, benthic invertebrates, arctic cod and seabird samples, HCHs were recovered by consecutive elution with 35 mL of hexane [fraction 1 (F1)], 38 mL of 85% hexane: 15% DCM (F2), and 52 mL of 1:1 DCM: hexane (F3). F1 contained 5% of α-HCH and 100% of PCB 30 and 204, F2 contained 25% of δ-HCH, 95% of α-HCH, and 100% of β-HCH, γ-HCH, and OCN and F3 contained 75% of δ-HCH. For seals, HCHs were eluted by a single elution of 100 mL of 1:1 DCM:hexane. Only one fraction was collected for ringed seal samples because they were analyzed by mass selective detector (MSD) (see below). All fractions were rotoevaporated, transferred to 2,2′,4-trimethyl pentane and evaporated to approximately 125 (sediment, zooplankton, *A. nugax*, Arctic cod, and seabird livers) or 570 μL (*P. hispida*). Aldrin (zooplankton, *A. nugax*, Arctic cod, and seabird livers) or ^{13}C-labeled PCB 138 (ringed seal blubber) were added as a volume corrector or an instrument performance standard, respectively.

Quantitative Analysis. Water samples were analyzed on a dual column [30 m × 0.25 mm DB-1 and 30 m × 0.25 mm DB-5 column (Supleco)] Hewlett-Packard 5890 gas chromatograph (GC) equipped with a dual ^{63}Ni-electron capture detector (ECD). All sediment, zooplankton, benthic invertebrates, Arctic cod, and seabird samples were analyzed on a Hewlett-Packard 5890 GC equipped with a 60 m × 0.25 mm DB-5 column (J & W Scientific, CA) and an ECD. N_2 was used as the makeup gas for samples analyzed on ECD. Ringed seals samples were analyzed on a Hewlett-Packard 5890 gas chromatograph (GC) equipped with a 30 m × 0.25 mm DB-5 MS column (J & W Scientific) and an HP 5970 MSD. A 3.0 μL sample volume was injected by splitless mode and external standards were run after every six samples for all samples. Concentrations were not corrected for internal standard recoveries.

Chiral Analysis. A 30 m fused silica 0.25 mm i.d., β-DEX 120 (20% nonbonded permethylated β-cyclodextrin) (Supelco Chromatography Products, ON, Ca) was used for chiral analysis of α-HCH. When interference was present a second column, a 30 m fused silica 0.25 mm i.d., 0.18μm film BGB 172 (BGB Analytik AG, Switzerland), was used to verify results. To reduce column bleed from entering the mass spectrometer a 1 m section of a DB5 MS column was joined to the MS end of the analytical column using a Supelco Glasseal capillary column connector.

Water, sediment, zooplankton, benthic invertebrates, and Arctic cod were analyzed by high-resolution gas chromatography/mass spectrometry on a VG AutoSpec double focusing mass spectrometer. A 2.0 μL sample volume was injected by splitless mode. The temperature program used in the analysis was injector temperature 250 °C, initial temperature and hold time 90 °C for 1 min, first ramp at 15 °C /min to 130 °C, second ramp 2 °C /min to 250 °C and held for 7 min, total run time of 71 min. Seabird and ringed seal samples were analyzed on an HP 5890 Series II gas chromatograph (GC) coupled with a 5972 mass selective detector (MSD). Splitless injection mode was used, with an injection volume of 3.0 μL. The GC temperature program was identical to that used above. The MS was operated in the selected ion monitoring (SIM) mode, an interface temperature of 280 °C, and a source temperature of 250 °C. The compounds of interest with their respective retention times were determined from the fragmentation pattern of appropriate enriched and racemic standards. Peak height was used for quantification of enantiomers because of the presence of a closely eluting compound with a similar fragmentation, but different ion ratio pattern. In extreme instances, baseline resolution of the two peaks was not possible. Since the target and the interfering compound were not baseline resoled, quantitation by height eliminated this partial peak overlap problem. Results of chiral analysis in samples that were not compromised by interfering compounds showed that using peak height produced the same results as peak area. Samples were combined, with the exception of ringed seals, to improve the detection limits and reduce the number of samples analyzed. Standards were run after every seventh sample. The elution order of enantiomers was monitored with enantio-enriched standards (EQ laboratories). The enantiomeric fraction (EF) of racemic α-HCH standards was 0.50 ± 0.01 (mean ± 1 SE).

Stable Isotope Analysis and Trophic Level Calculations. Prior to stable isotope analyses, all tissue samples were washed in distilled water and then freeze-dried, powdered, and treated with a 2:1 chloroform:methanol solution to remove lipids. Samples were then dried under a fume hood. Zooplankton and starfish samples were soaked in 0.1 N HCl to remove carbonates and allowed to dry without rinsing. Details on stable isotope analysis can be found in Fisk et al. (*13*). Stable isotope abundances were expressed in δ notation as the deviation from standards in parts per thousand (‰) according to the following equation:

$$\delta^{15}N = [(^{15}N/^{14}N_{\text{sample}}/^{15}N/^{14}N_{\text{standard}}) - 1] \times 1000 \quad (1)$$

The $^{15}N/^{14}N_{\text{standard}}$ values were based on atmospheric N_2 (AIR). Replicate measurements of internal laboratory standards (albumen) indicate measurement errors of ±0.3 ‰ for stable-nitrogen isotope measurements.

Trophic levels were determined using equations modified slightly from those reported in Hobson et al. (*14*). Trophic level was determined relative to the copepod *Calanus hyperboreus*, which we assumed occupied trophic level 2 (i.e., primary herbivore). For zooplankton, fish and marine mammals the relationship used was

$$\text{TL} = 2 + (\delta^{15}N_{\text{consumer}} - \delta^{15}N_{C.\ hyperboreus})/3.8 \quad (2)$$

where TL is trophic level, $\delta^{15}N_{C.\ hyperboreus}$ is equal to 7.74 (mean $\delta^{15}N$ for *C. hyperboreus*) and 3.8 the isotopic enrichment factor (*15*). Captive-rearing studies on birds suggest that diet-tissue isotopic fractionation factor of +2.4‰ is appropriate

for these taxa (*16*), and therefore, we used this value and the relationships $TL_{birds} = TL_{prey} + 1$ and $\delta^{15}N_{C.hyperbopreus} = \delta^{15}N_{consumer} - 2.4$ in order to modify eq 2 to

$$TL_{bird} = 3 + (\delta^{15}N_{bird} - 9.2)/3.8 \quad (3)$$

Bioconcentration (BCF) and Biomagnification Factor (BMF) Calculations. BCFs were only calculated for the pelagic zooplankton *C. hyperboreus* because this species was abundant in samples and was at the lowest trophic level and, hence, was considered to experience minimal effects of trophic transfer. The following equation was used:

$$BCF = [C.\ hyperboreus]/[water] \quad (4)$$

where [*C. hyperboreus*] is the mean concentration in *C. hyperboreus* (ng/g, lipid basis) and [water] is the mean concentration in water (ng/mL).

BMFs were calculated for individual species using the equation:

$$BMF_{pred/prey} = [predator]/[prey] \quad (5)$$

where [predator] is the concentration (lipid basis) in the predator and [prey] is the concentration (lipid basis) in the prey. A BMF for the entire food web, termed a food web magnification factor (FWMF) was also determined from the relationship between trophic level (based on organic $\delta^{15}N$ values) and concentration using simple linear regression:

$$\ln \text{concentration} = a + (b \times \text{trophic level}) \quad (6)$$

All concentrations were calculated on a lipid basis due to a large range in lipid content among species. The slope of the relationship (b) was used to calculate FWMF using the equation:

$$FWMF = e^{b} \quad (7)$$

Results and Discussion

Concentrations. HCH concentrations are presented in Table 1. In most samples, three isomers were found and quantified. The exceptions were a number of zooplankton and benthic invertebrate samples in which β-HCH was not quantifiable. Arctic cod were all of similar length (18 ± 1.3 cm) and likely of the same year class and concentrations did not vary with sex. There is no method to age seabirds and HCH concentrations did not vary with sex, which is consistent with the results of chlordane concentrations in these same seabirds (*17*). HCH concentrations in the NOW ringed seals were not found to vary with sex or age. Although other POPs tend to vary with sex and age in ringed seals this is not observed with HCH (*18*). Therefore, all HCH concentration data were combined for each species with no separation for sex or age.

Water and sediment concentrations and relative proportions of α-HCH and γ-HCH in the NOW are consistent with the geographical trends recently observed for these compounds in the Arctic (*19*−*21*). HCH concentrations in Arctic zooplankton collected in 1986−87 were similar to those found in the NOW zooplankton (*22*, *23*), suggesting minimal temporal change in HCH concentrations in Arctic zooplankton. Σ-HCH concentrations of 670 and 45 ng/g (lipid basis) were reported in Arctic cod collected in the Barrow Strait area in 1984 [whole fish, Muir et al. (*24*)] and 1993 [muscle and skin only, Muir et al. (*25*)], respectively. Σ-HCH concentrations reported for Arctic cod in this study are approximately twice those reported in 1993. Higher concentrations in the 1984 samples and the present study may be due to the use of the whole fish, but these concentration differences require further study.

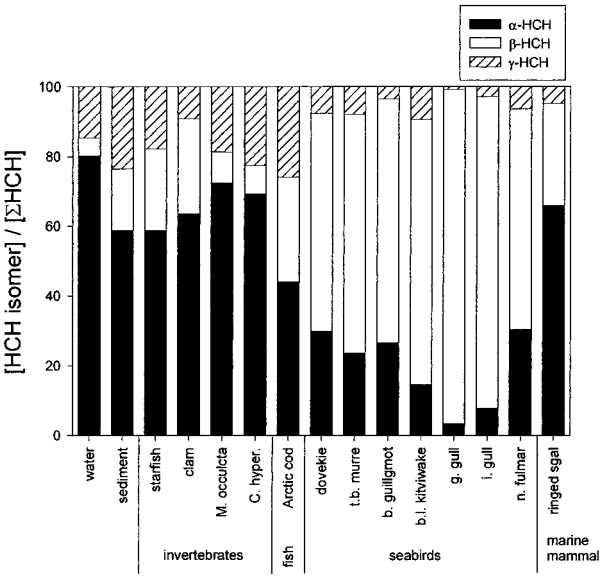

FIGURE 1. Relative proportions of α-, β-, and γ isomers of HCH in the NOW marine food chain. Only species and samples that had measurable amounts of each isomer were included.

Σ-HCH concentrations (lipid basis) in seabirds were similar to those reported previously in muscle of thick billed murre (*26*), black guillemot, and glaucous gull (*27*) from the Canadian Arctic. Concentrations of Σ-HCH in NOW glaucous gulls were approximately four times greater than those reported in fat of Barents Sea glaucous gulls (*28*), consistent with trends observed in other species, such as the ringed seal (*18*). Concentrations of Σ-HCH in ringed seals from the NOW are in good agreement with levels reported recently in blubber of ringed seals from similar locations in the Arctic (*18*, *21*).

Relative Proportions of HCH Isomers. The relative proportions of the HCH isomers varied dramatically across the species of NOW marine food web (Figure 1). The ratio of α/γ in the NOW water samples was 5.5, slightly less than the ratio reported for water collected in the Barrow Strait in 1993 [α/γ = 7 (*29*)] but slightly greater than those reported for Arctic Ocean water collected north of Greenland [α/γ = 3.4−4.2 (*19*)]. HCH concentrations have been declining in Arctic air, and volatilization from ocean water is now considered the predominant source of HCH to air (*19*). Therefore, it is likely that the source of HCHs in the NOW reflect the mixing of water from the Arctic Ocean above Greenland, the Canadian Archipelago and the north Atlantic. There was a greater proportion of β- and γ-HCH in the NOW sediment than in water, which likely reflects greater degradation of the α-HCH on particles in the water column (*20*) and by microorganisms in the sediment. It has been reported that under aerobic conditions in sediment/water experiments α-HCH can be iosmerized into β-HCH (*30*), which could contribute to differences between sediment and water.

The relative proportions of the HCH isomers in pelagic zooplankton are similar to water and those in benthic invertebrates are similar to sediment (Figure 1), suggesting minimal biotransformation of HCH in invertebrates. In general, invertebrates are considered to have limited ability to metabolize POPs (*31*), so changes in the HCH isomer patterns between source and invertebrate are unlikely. The proportions of the HCH isomers in Arctic zooplankton reported by Hargrave et al. (*22*) varied over time, but were in general agreement with those observed in this study. The relative percentage of α-HCH in Arctic cod was lower than those observed in zooplankton and water, suggesting greater bioaccumulation of β- and γ-HCH, greater metabolism of

TABLE 2. Apparent Bioconcentration (BCF), Biomagnification (BMF), and Food Web Magnification (FWMF) Factors for Various Pelagic Species of the NOW Marine Food Web[a]

predator	prey	α	(+) α	(−) α	*β*	*γ*	ΣHCH
log BCF							
C. hyperboreus	water	4.4	4.3	4.4	3.9	4.4	4.3
BMF							
T. libellula	*C. hyperboreus*	1.8	1.9	1.8		6.6	2.7
Arctic cod	*C. hyperboreus*	1.6	1.7	1.5	54	4.2	3.0
dovekie	*C. hyperboreus*	2.7	4.1	1.6	280	1.5	7.3
dovekie	Arctic cod	1.7	2.4	1.1	5.1	0.7	2.5
thick-billed murre	Arctic cod	0.5	1.0	0.1	2.1	0.3	0.9
black guillemot	Arctic cod	1.9	3.9	0.3	7.3	0.4	3.2
black legged kittiwake	Arctic cod	0.2	0.3	0.04	1.3	0.2	0.5
glaucous gull	Arctic cod	0.4	0.8	0.02	15.7	0.2	4.9
ivory gull	Arctic cod	0.3	0.6	0.04	4.7	0.2	1.6
northern fulmar	Arctic cod	0.5	0.9	0.2	1.5	0.2	0.7
ringed seal	Arctic cod	2.5	2.8	2.2	1.6	0.3	1.7
food web FWMF		1.8 ± 1.1			3.9 ± 1.2	1.1 ± 1.1	2.4 ± 1.1

[a] FWMF include ± 1 standard error.

α-HCH and/or different exposure. Due to similar log K_{ow} of the three congeners (*32*), it would seem unlikely that different bioaccumulation could account for these changes. These results suggest that Arctic cod have some ability to biotransform α-HCH.

The most marked change in HCH isomers was observed in the seabirds. *β*-HCH was the dominant HCH isomer in seabirds (Figure 1), and in some species accounted for >90% of the total HCH. High relative proportions of *β*-HCH, most >90%, have been noted previously in the eggs of a large number of Arctic seabird species (*33*). This suggests high biotransformation of α- and *γ*-HCH. There was variability among seabird species, with alcids (thick-billed murre, dovekie, and black guillemot) and fulmars (northern fulmar) having a greater percentage of α-HCH than "gulls" (black-legged kittiwake, glaucous gulls, and ivory gull) (Figure 1).

In ringed seals, the proportion of α-HCH was similar to zooplankton and water, but the percentage of *γ*-HCH was much less and *β*-HCH was greater. It would appear that ringed seals can biotransform *γ*-HCH, but are less efficient at metabolizing α- and *β*-HCH. These percentages are somewhat different than those reported for ringed seals collected in eastern Hudson Bay (*34*), which reported a high percentage of α-HCH (~60%) and similar percentages of *β*- and *γ*-HCH (~15%). Similar HCH proportions to the NOW ringed seals were recently reported for ringed seals collected in the White Sea, Russia (*35*).

Bioconcentration and Biomagnification Factors. Bioconcentration (BCF) and biomagnification factors (BMF) provide insight on the fate and dynamics of HCH isomers in the NOW marine food web. BCFs have been found to have a log-linear relationship with the octanol−water partition coefficient (K_{ow}) (*36*). This relationship can vary due to biomagnification (i.e., BCFs higher than expected based on K_{ow}) or biotransformation (i.e., BCFs lower than expected) (*37*). BMFs have also been found to be related to K_{ow} (*13, 38*), where greater than expected BMFs can be attributed to "*bioformation*", those contaminants formed from other contaminants due to biotransformation, and lower than expected BMFs can be attributed to biotransformation (*13*).

Log BCF values of HCH isomers observed for *C. hyperboreus* did not vary between isomers and were similar to their respective log K_{ow} values [log K_{ow} of α- and *β*-HCH is 3.9 and *γ*-HCH is 4.1 (*32*)]. This suggests that at the base of the food web there are no significant differences in the bioaccumulation of the HCH isomers and that there is minimal biotransformation.

BMFs of the HCH isomers varied dramatically among isomers and species of the NOW food web. Caution should be used when interpreting these BMFs, as the diet of many of these predators are varied and in many cases the predator is not a full trophic level above the assumed prey based on stable isotopes of nitrogen (Table 1). BMFs of all three isomers were greater than 1 in zooplankton and Arctic cod (Table 2), showing the potential for biomagnification of HCH isomers in aquatic food webs. BMFs of α-HCH were generally <1 in seabirds, the exception being dovekie and black guillemot. On the basis of the high BCF and BMFs of α-HCH in zooplankton and fish but low BMF values in seabirds, it would appear that seabirds can readily biotransform α-HCH preventing biomagnification of this congener. This is not the case with ringed seal, which had a BMF of 2.5 for α-HCH using seal blubber, suggesting limited biotransformation, but this BMF may have varied if another seal tissue was used. Wiberg et al. (*9*) and Muir et al. (*24*) reported similar BMFs for Arctic cod to ringed seal blubber. BMFs of *β*-HCH were >1 in all seabird species and ringed seal, suggesting limited biotransformation in these species. *γ*-HCH appears to be the most readily metabolized HCH isomer based on the lowest BMFs in ringed seal and seabirds.

It would appear that HCH isomers have the potential to biomagnify in aquatic food webs unless biotransformed by one or more species in the food web. The *γ*-isomer, which appears to be readily biotransformed by seabirds and ringed seals, did not have a significant relationship with trophic level based on organic $\delta^{15}N$ values (Figure 2), although *γ*-HCH did biomagnify at lower trophic levels where biotransformation was minimal. Whereas *β*-HCH, which appears to be recalcitrant in all species of the NOW food web, had a strong positive relationship with trophic level (Figure 2) and an FWMF of 3.5 (Table 2). The magnitude of biomagnification for HCH is low compared to other hydrophobic recalcitrant POPs, such as PCB 153 (*13*). This is likely due to the relatively high water solubility of HCHs compared to other POPs. Kidd et al. (*39*) found that α-HCH biomagnified in a freshwater food web that included only zooplankton and fish but to a lesser extent than more hydrophobic POPs, such as PCB 153 and DDE.

α-HCH Enantiomeric Trends. In a majority of samples, both enantiomers of α-HCH could be quantified and separated. For some samples one or both of α-HCH enantiomers were below detection limits on the MSD. This was likely due to the nature of the chiral analysis, which splits the α-HCH peak, resulting in a smaller area per enantiomer and an increase in the detection limit. In the rare instance when

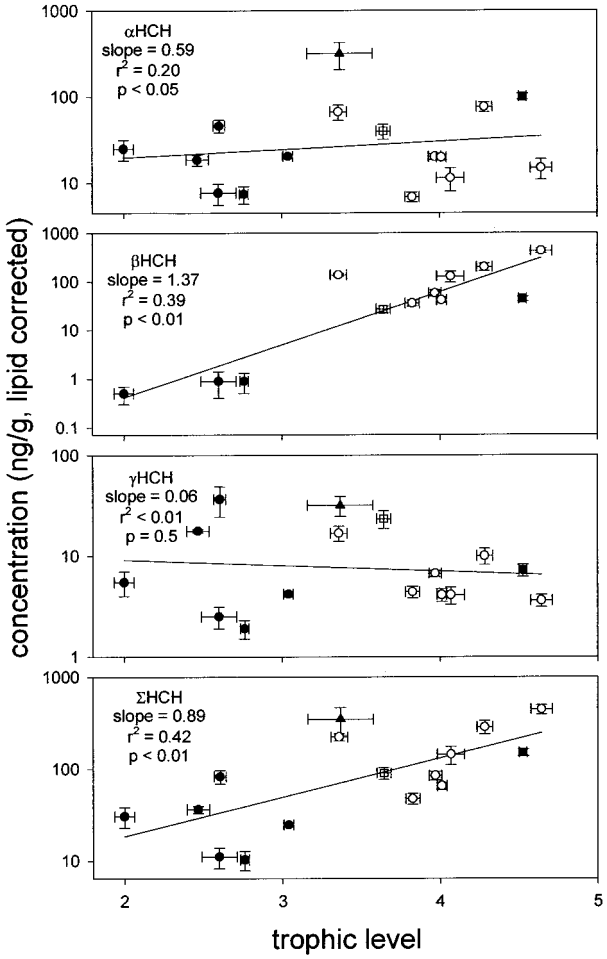

FIGURE 2. Relationship between HCH isomer ln concentrations (lipid basis) and trophic level in the NOW pelagic marine food web. Benthic organisms were not included in the regression analysis with the exception of *A. nugax*, a scavenging amphipod. Regression results are based on all data points and not the mean values. See text for details regarding sample types analyzed. (●) zooplankton; (▲) *A. nugax*; (□) arctic cod; (■) ringed seals; and (○) seabirds.

interference was present, enantiomers could be resolved by reanalyzing the sample on the second chiral column (BGB 172). This was verified and monitored by the use of pure external enantiomeric standards.

Enantiomeric composition of chiral chemicals can be expressed as fractions (EFs) or ratios (ERs). EFs are more easily compared and used in mathematical equations (*40*). EF was used in this paper and was calculated using the following equation:

$$EF = (+)/[(+) + (-)] \quad (8)$$

where (+) and (−) is the height of the corresponding enantiomer. For comparison with past work, where ERs are more commonly used, ERs were converted to EF with the equation:

$$EF = ER/(ER + 1) \quad (9)$$

Table 1 indicates that surface water samples displayed a slight depletion of the (+) α-HCH enantiomer (EFs < 0.5), consistent to values previously reported for the Arctic Ocean (*19*, *20*, *41*). There was a greater depletion of the (+) enantiomer of α-HCH in sediment (EF = 0.39) than observed in the water (Table 1). This is likely due to degradation of α-HCH as it decends the water column (*19*) or in the sediment.

The EFs of α-HCH in zooplankton and Arctic cod were similar to values obtained for water, with the exception of *M. occulata* (Table 1). It should be noted that only one sample of each species of zooplankton were analyzed. BMFs of the α-HCH enantiomers were also similar (Table 2). EFs of α-HCH reported for blue mussel (*Mytilus edulis*) and flounder (*Platychthys flesus*) were similar to water (*42*). Wiberg et al. (*9*) noted near racemic EFs for Arctic cod collected in the Canadian Arctic. Similar EFs of α-HCH in zooplankton and water, which was collected at the surface, are consistent with results of the relative proportions of HCH isomers, suggesting no, or limited, biotransformation. The relative proportions of HCH isomers in Arctic cod suggested some biotransformation of α-HCH, which was inconsistent with the EFs of α-HCH in this species. Biotransformation of α-HCH in Arctic cod could be nonenantiospecific, i.e., neither enantiomer is biotransformed at a great rate, which would result in a change in the relative proportions of HCH isomers compared with source but with no change in the chiral signature of α-HCH.

EFs of α-HCH in ringed seal blubber were near racemic, suggesting no enantiospecific biotransformation in these seals. Although seals could degrade both enantiomers of α-HCH, and thus have a racemic chiral signature, the α-HCH BMF in ringed seals was >1, suggesting that the NOW ringed seals were not efficiently biotransforming α-HCH. Similar, near racemic, EFs for ringed seal blubber have been reported (*9*, *43*). Similar EFs of α-HCH were noted in blubber, liver, and lung tissue of neonatal northern fur seals (*Callorhinus ursinus*) but with high EFs in the brain, which was attributed to selective crossing of the blood brain barrier (*44*).

The highest EFs of α-HCH were observed in the seabirds, and for some species were close to 1 (Table 1). It is clear that these seabirds preferentially metabolized the (−) enantiomer of α-HCH. EFs of α-HCH in double-crested cormorant (*Phalacrocorax auritus*) collected in the Great Lakes were near racemic (*45*), much lower than those found in this study. The proportion of α-HCH in these seabirds was also much higher than in the seabirds from this study. High EFs in the NOW seabirds were generally associated with α-HCH BMFs that were <1, the exceptions were the black guillemot and dovekie. This suggests that most seabirds can metabolize both enantiomers, although the (−) is metabolized at a much greater rate. It would appear that in the black guillemot, the (+) enantiomer is not biotransformed based on a BMF of 3.9. The high EF (0.92) of α-HCH measured in BLGU is consistent with this observation. The dovekie was the only seabird that had BMFs of >1 for both enantiomers, consistent with the least nonracemic EFs among seabirds. It would appear that BMFs of >1 for the (+) enantiomer is common for all of the alcids (thick-billed murre, black guillemot, and dovekie), and suggests a lack of significant metabolism of this enantiomer in this group of seabirds.

EFs of α-HCH appear to be a good indicator of a species ability to biotransform α-HCH. A strong relationship between the EF of (+) α-HCH and the proportion of β-HCH of the total HCH was found (Figure 3). The assumption being that biotransformation of α-HCH results in a greater percentage of the Σ-HCH being β-HCH. Since, as described above, β tends to be the most recalcitrant HCH isomer, higher ratios of β-HCH to Σ-HCH indicate an increased metabolic capability. Conversely, a low EF of (+) α-HCH should also be associated with a higher proportion of β-HCH because in this case the (+) α-HCH is metabolized. However, organisms with a low EF for (+) α-HCH had a lower percentage of β-HCH in the NOW food web. These organisms were at a lower trophic level and EFs were similar to their abiotic environment (water or sediment). Therefore biomagnification is also an important component of this relationship. Tanabe et al. (*46*) observed an opposite trend in small cetaceans, with a decrease in the proportion of β-HCH with increasing EFs of

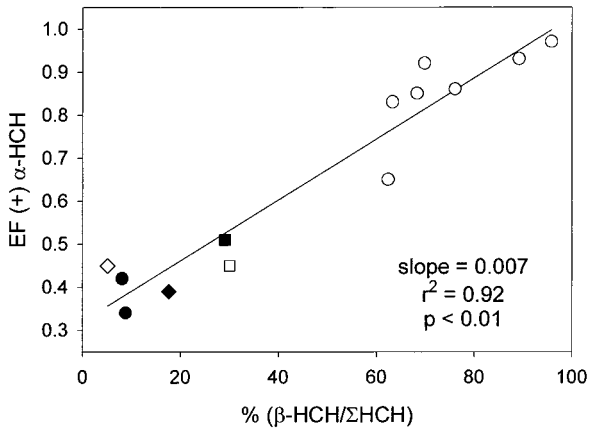

FIGURE 3. Relationship between EFs of (+)-α-HCH and proportion of *β*-HCH of the Σ-HCH in the NOW marine food web. (◇) water; (◆) sediment; (●) invertebrates (pelagic zooplankton and benthic); (□) arctic cod; (○) seabirds; and (■) ringed seal.

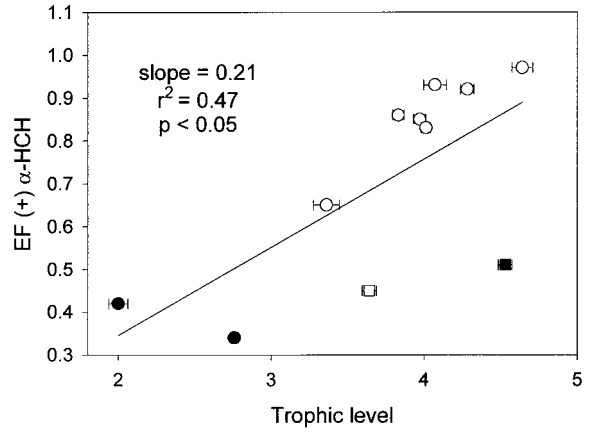

FIGURE 4. Relationship between EFs (+) α-HCH and trophic level (based on δ^{15}N) in the NOW marine food web. (●) pelagic zooplankton; (□) arctic cod; (○) seabirds; and (■) ringed seal.

α-HCH, although the elution order of the enantiomers was not verified with enatio-enriched standards. They also suggested that differences in EFs of α-HCH between cetaceans and seabirds could be interpreted as the efficiency to degrade α-HCH.

EFs of α-HCH were also positively related to trophic level (Figure 4). This suggests that the ability to degrade α-HCH increases with trophic level. In general, higher trophic levels are associated with greater biotransformation capacity (*31*). However, this relationship appears to be driven by the seabirds and nearly racemic values in the zooplankton, with Arctic cod and ringed seal falling well below the relationship. This relationship was very strong for the seabirds as a single group. Since the enzyme systems of birds are likely more similar than groups within the food web, these relationship may be driven by enzyme induction. Data from a larger number of species, particularly fish and mammals, might result in a nonsignificant relationship.

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