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Environmental Pollution 113 (2001) 225–238

ENVIRONMENTAL POLLUTION

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Chlordane components and metabolites in seven species of Arctic seabirds from the Northwater Polynya: relationships with stable isotopes of nitrogen and enantiomeric fractions of chiral components

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Received 18 March 2000; accepted 5 June 2000

"Capsule": *The relative proportions of chlordane and its components in seabirds was related to phylogeny.*

Abstract

The Northwater Polynya (NOW) is a large area of year-round open water found in the high Arctic between Ellesmere Island and Greenland. NOW has high biological productivity compared with other arctic marine areas, and supports large populations of several seabird species. Seven species of seabirds, dovekie (*Alle alle*, DOVE), thick-billed murre (*Uria lomvia*, TBMU), black guillemot (*Cepphus grylle*, BLGU), black-legged kittiwake (*Rissa tridactyla*, BLKI), ivory gull (*Pagophila eburnea*, IVGU), glaucous gull (*Larus hyperboreus*, GLGU) and northern fulmar (*Fulmaris glacialis*, NOFU) were collected in May and June 1998 to determine chlordane concentrations in liver and fat and to examine species differences, relationships with stable isotopes of nitrogen, and enantiomeric fractions (EFs) of chiral components. ΣCHLOR concentrations varied over an order of magnitude among species, from a low of 176±19 ng/g (lipid corrected) in TMBU liver to a high of 3190±656 ng/g (lipid corrected) in NOFU liver. Lipid-corrected concentrations of chlordane did not vary between sex for any species or between fat and liver except for the DOVE, that had fat concentrations that were significantly greater than the liver. $\delta^{15}N$ values described a significant percentage of the variability of concentrations for most chlordane components, although less than what has been reported for whole food chains. Slopes of $\delta^{15}N$ versus concentration of chlordane components and ΣCHLOR were similar with the exception of those which were metabolized (*trans*-chlordane) or formed through biotransformation (oxychlordane). The relative proportions of chlordane components in seabirds were related to phylogeny; the procellariid (NOFU) had the greatest percentage of oxychlordane (>70%), followed by the larids (BLKI, IVGU and GLGU; 40–50%) and the alcids (DOVE and BLGU; 10–20%). The exception was TBMU, an alcid, where oxychlordane made up >40% of its chlordane. EFs of chiral components failed to predict concentration or trophic level, but did identify biotransformation differences between species and chlordane components. TBMU appeared to have a greater capacity to metabolize and eliminate chlordane, based on high proportions of oxychlordane, the highest EFs for oxychlordane and heptachlor epoxide, and a $\delta^{15}N$-ΣCHLOR value which was well below the relationships developed for all seabird species.

Keywords: Bioaccumulation; Biotransformation; Nitrogen-15; Chiral compounds

1. Introduction

Chlordane was one of the most heavily used pesticides until its restriction in the 1980s and subsequent world-wide ban in 1997 (Bidleman and Falconer, 1999). Various components of technical chlordane, including *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, and the metabolites heptachlor epoxide and oxychlordane, have been measured in Arctic abiotic and biotic environments (AMAP, 1998). Many of these components bioaccumulate and biomagnify in aquatic food chains (Muir et al., 1988), and relatively high concentrations have been measured in upper trophic-level organisms

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0269-7491/01/$ - see front matter
PII: S0269-7491(00)00153-6

such as polar bears (*Ursus maritimus*; Norstrom et al., 1988, 1998) and glaucous gulls (*Larus hyperboreus*; Braune, 1994a, b). Chlordanes are one of the most predominant persistent organic pollutants (POP) found in Arctic seabirds (Braune et al., 1999) and a number of components and metabolites are toxic and even carcinogenic (AMAP, 1998).

The ratio of the heavier to lighter stable isotopes of nitrogen ($^{15}N/^{14}N$), expressed as $\delta^{15}N$, generally increases with trophic position in aquatic food chains, providing a continuous variable with which to assess both trophic level (Michener and Schell, 1994; Hobson et al., 1995) and food chain transfer of POPs (Kidd, 1998). Biomagnification factors can be estimated from slopes of logarithmic concentration of contaminants versus $\delta^{15}N$ (e.g. Broman et al., 1992; Rolff et al., 1993; Jarman et al., 1996). More recently, enantiomeric ratios (ERs) of chiral pollutants have been used to assess bioaccumulation or food chain transfer (Wiberg et al., 1998, 2000). 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 (Buser and Müller, 1993). Therefore, differences in biotransformation rates between species can often be detected by changes in ERs or enantiomeric fractions (EFs) of chiral pollutants (Buser et al., 1992; Harner et al., 2000). Chiral components in technical chlordane are racemic (Buser et al., 1992; Buser and Müller, 1993), that is having equal amounts of the (+) and (−) enantiomers. The biotransformation ability in aquatic food chains generally increases from invertebrates to fish to birds and mammals (Norstrom et al., 1978; Boon et al., 1989), and therefore EFs have the potential to describe or quantify bioaccumulation and food-chain transfer (Wiberg et al., 2000). The assessment of bioaccumulation using $\delta^{15}N$ values and EFs of chiral POPs in foodweb components has proven to be effective for the analysis of whole food chains but assessment of their utility within single groups of animals (e.g. seabirds) has not been investigated previously.

Polynyas are areas of open water, often surrounded by sea ice, which persist throughout the winter in polar seas. They are one of the most important and least-understood phenomena in polar ecology (Stirling, 1980). The Northwater (NOW) in northern Baffin Bay is the largest and most productive polynya in the Canadian Arctic, supporting large populations of seabirds. Seabirds feed at several trophic levels, from small pelagic zooplankton and fish through seabird chicks and the carrion of polar bear kills, and thus can potentially influence energetics of arctic systems including polynyas.

Despite the importance of seabirds in Arctic marine ecosystems, there is limited data on the concentration of chlordane in their tissues and factors influencing these concentrations in adults. The majority of seabird contaminant data from the Canadian Arctic is derived from eggs. An extensive multi-disciplinary study on NOW afforded the opportunity to collect seven seabird species within the same area and at the same time to address this data and knowledge gap. These species cover a range of trophic levels and have a number of feeding and migration strategies and include: dovekie (*Alle alle*, DOVE), thick-billed murre (*Uria lomvia*, TBMU), black guillemot (*Cepphus grylle*, BLGU), black-legged kittiwake (*Rissa tridactyla*, BLKI), ivory gull (*Pagophila eburnea*, IVGU), glaucous gull (*Larus hyperboreus* GLGU) and northern fulmar (*Fulmaris glacialis*, NOFU). Concentrations and EFs of chiral chlordane components in liver and fat, and $\delta^{15}N$ values in muscle, were measured to assess differences among species and the utility of $\delta^{15}N$ trophic estimates and EFs of chiral chlordane components to predict concentrations of chlordane components in Arctic seabirds.

2. Methods and materials

2.1. Field collection, species and sample size

As part of a larger study, seabirds were collected from the NOW Polynya (Fig. 1) in May and June 1998 by shotgun. Seabirds were dissected shortly after death, and liver, subcutaneous or abdominal fat, and pectoral muscle samples were placed in Whirl Pak bags and frozen until analyzed for stable isotope analysis and/or chlordane components. Mass measurements were taken from all seabirds prior to dissection (Table 1). A total of 62 and 60 liver and fat samples were analyzed, respectively. Two birds did not have sufficient fat for collection and analysis. The number of samples analyzed per seabird species and the sex of the seabirds are presented in Tables 2 and 3.

2.2. Chemicals and standards

All solvents (pesticide grade) and sodium sulfate (Na_2SO_4) were obtained from BDH Inc. (City, SI, USA). Pesticide grade Florisil, 60–100 mesh was obtained from the Floridin Corp. (Berkeley Spring, WV, USA). Biobeads SX-3 used in the GPC column were purchased from Analytical Biochemistry Laboratories Ltd., (Columbia, MO, USA). Non-racemic standards of oxychlordane, heptachlor epoxide, and *cis*- and *trans*-chlordane were obtained from EQ Laboratories (Atlanta, GA, USA). Standards of MC5 and MC7 were donated by M. Oehme (University of Basel, Switzerland).

2.3. Stable isotope analysis

Stable-carbon and nitrogen isotope assays were performed on 1-mg subsamples of homogenized materials by loading into tin cups and combusting at 1800°C

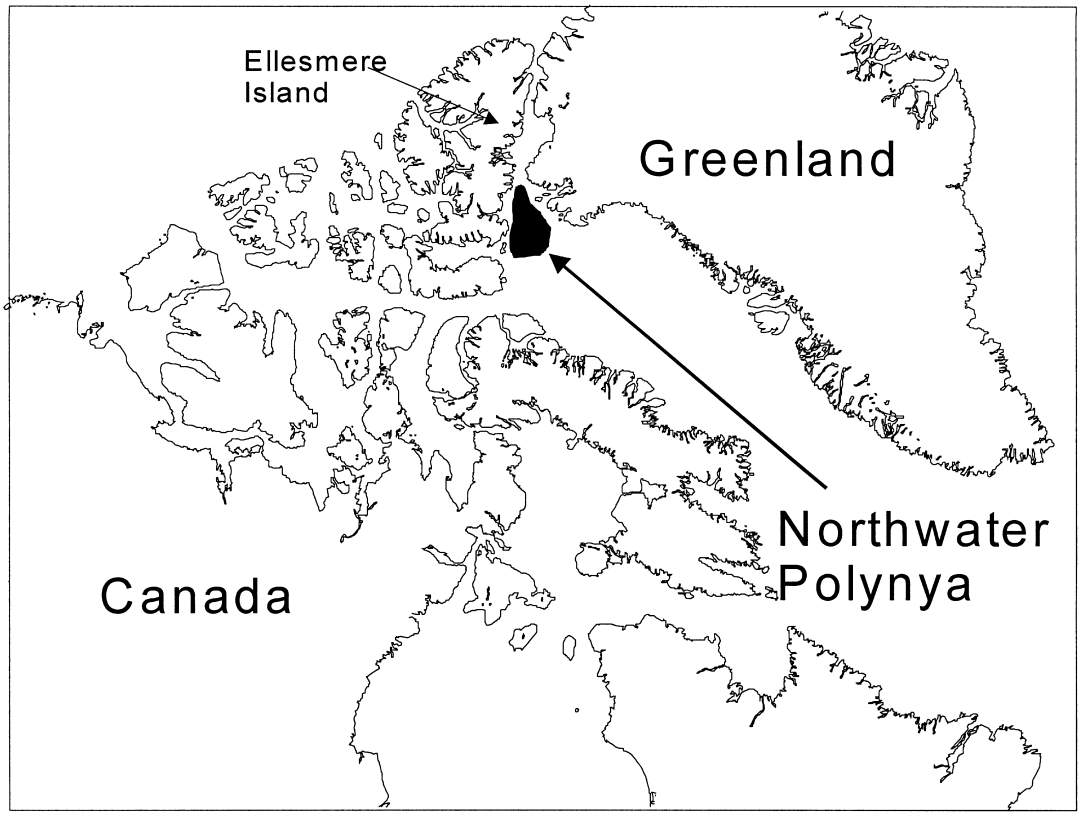

Fig. 1. Approximate location and size of the Northwater Polynya in May/June.

Table 1
Characteristics of seabird species collected in the Northwater[a]

Species[b]	*n*	Muscle $\delta^{15}N$ (‰)	Relative trophic level[c]	Weight (g)	Diet	Migrating species	Winter habitat	Ref.
DOVE	10	11.6±0.3 D	3	161±3.5	Zooplankton/fish	Yes	St. Lawrence estuary and eastern seaboard of NA	Gaston and Jones (1998)
TBMU	10	13.8±0.2 B	3.6	957±11.9	Zooplankton/fish	Yes	North Atlantic, coastal Newfoundland to southern Greenland	Gaston and Jones (1998)
BLGU	9	14.9±0.2 B	3.9	415±13.4	Zooplankton/fish	No	Arctic ice edges	Gaston and Jones (1998)
BLKI	10	13.3±0.2 C	3.5	387±9.8	Zooplankton/fish	Yes	Eastern seaboard of NA	Baird (1994)
IVGU	5	14.1±0.3 BCD	3.7	537±25.6	Zooplankton/fish/ carrion	No	Arctic ice edges	Maney and Macdonald (1998)
GLGU	11	16.4±0.3 A	5.3	1590±84.6	Zooplankton/fish/ carrion/seabird chicks	Yes	Eastern seaboard of NA and Great Lakes	Godfrey (1986)
NOFU	10	14.0±0.15 B	3.6	680±34.2	Zooplankton/fish/ carrion	Yes	North Atlantic east of Greenland	Hatch and Nettleship (1998)

[a] $\delta^{15}N$ values with the same letter are not significantly different ($P < 0.05$).

[b] Seabirds: dovekie (*Alle alle*, DOVE), thick-billed murre (*Uria lomvia*, TBMU), black guillemot (*Cepphus grylle*, BLGU), black-legged kittiwake (*Rissa tridactyla*, BLKI), ivory gull (*Pagophila eburnea*, IVGU), glaucous gull (*Larus hyperboreus*, GLGU) and northern fulmar (*Fulmaris glacialis*, NOFU).

[c] The relative trophic level was calculating assuming the DOVE occupied trophic level 3 and using the equation $TL = (\delta^{15}N - \delta^{15}N_{DOVE})/3.8 + 3$, modified from equations described in Hobson et al. (1995).

in a Robo-Prep elemental analyzer. Resultant CO_2 and N_2 gases were then analyzed using an interfaced Europa 20:20 continuous-flow isotope ratio mass spectrometer with every five unknowns separated by two laboratory standards. Stable isotope abundances were expressed in δ notation as the deviation from standards in parts per thousand (‰) according to the following equation:

$$\delta X = [(R_{sample}/R_{standard}) - 1] \times 1000, \quad (1)$$

Table 2
Chlordane component concentrations (mean±1 S.E. [in parentheses], ng/g wet weight) in liver of Northwater seabirds[a]

	DOVE		TBMU		BLGU		BLKI		IVGU		GLGU		NUFU	
	Female	Male	Female	Male	Female	Male	Female	Male	Female	Male	Female	Male	Female	Male
n	3	4	5	5	2	7	6	4	2	3	6	5	5	5
Lipid (%)	3.9 (0.33)	4.2 (0.11)	5.1 (0.82)	3.1 (0.46)	4.0 (0.78)	3.2 (0.38)	4.5 (0.85)	3.4 (0.44)	4.7 (0.12)	2.4 (0.42)	5.5 (0.36)	5.8 (0.68)	4.3 (0.99)	3.3 (0.52)
HE	2.5 (0.93)	2.4 (0.46)	1.6 (0.24)	1.0 (0.12)	2.5 (2.5)	4.9 (1.3)	6.5 (1.4)	7.2 (2.0)	33 (12)	9.8 (2.8)	24 (4.0)	27 (3.3)	16 (1.7)	11 (2.1)
Oxychlordane	1.6 (0.28)	2.0 (0.72)	3.7 (0.60)	2.5 (0.33)	5.8 (0.59)	4.4 (0.54)	11 (2.2)	11 (2.0)	70 (39)	23 (8.3)	57 (14)	63 (5.6)	69 (9.1)	79 (11)
t-chlordane	0.26 (0.12)	0.26 (0.094)	0.046 (0.046)	0.045 (0.045)	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
c-chlordane	0.40 (0.10)	0.44 (0.17)	0.42 (0.031)	0.29 (0.060)	0.92 (0.11)	0.99 (0.15)	0.44 (0.029)	0.56 (0.082)	1.9 (0.020)	1.3 (0.28)	1.3 (0.16)	2.2 (0.42)	0.35 (0.066)	0.37 (0.079)
t-nonachlor	2.8 (0.62)	3.4 (0.93)	0.77 (0.064)	0.47 (0.10)	5.5 (0.83)	6.0 (0.97)	2.7 (0.33)	3.6 (0.55)	15 (1.7)	22 (10)	15 (2.1)	29 (4.2)	3.4 (0.28)	3.9 (0.71)
c-nonachlor	1.3 (0.26)	1.5 (0.62)	1.2 (0.23)	1.1 (0.24)	8.3 (1.4)	6.3 (0.70)	1.9 (0.18)	2.4 (0.33)	3.4 (0.36)	6.2 (1.8)	5.7 (0.92)	9.3 (1.5)	0.32 (0.038)	0.22 (0.073)
MC5	0.64 (0.066)	0.69 (0.30)	0.084 (0.021)	0.053 (0.009)	2.9 (1.1)	2.0 (0.33)	1.9 (0.33)	2.7 (0.60)	5.1 (1.5)	2.5 (0.95)	4.7 (0.86)	4.9 (1.1)	2.8 (0.50)	4.7 (0.73)
MC7	nd	nd	0.053 (0.008)	0.028 (0.005)	nd	nd	0.039 (0.010)	0.039 (0.011)	nd	nd	0.17 (0.041)	0.13 (0.024)	0.069 (0.010)	0.085 (0.015)
Σchlordanes	9.5 (1.9)	11 (3.2)	7.8 (1.1)	5.5 (0.67)	26 (1.5)	25 (3.3)	25 (3.9)	28 (5.3)	130 (51)	64 (24)	110 (20)	140 (8.9)	92 (8.2)	99 (12)

[a] nd, not detected (detection limits ~0.02 ng/g).

Table 3
Chlordane component concentrations (mean±1 S.E. [in parentheses], ng/g wet weight) in fat of Northwater seabirds[a]

	DOVE		TBMU		BLGU		BLKI		IVGU		GLGU		NOFU	
	Female	Male	Female	Male	Female	Male	Female	Male	Female	Male	Female	Male	Female	Male
n	5	5	5	5	1	6	5	3	2	2	6	5	5	5
Lipid (%)	65 (1.0)	62 (4.3)	60 (3.1)	61 (2.6)	39	64 (5.7)	73 (5.6)	72 (6.2)	73 (5.2)	89 (2.9)	76 (2.4)	70 (9.3)	78 (1.7)	65 (7.0)
HE	94 (9.1)	96 (9.1)	35 (4.2)	35 (9.0)	110	100 (11)	71 (12)	63 (16)	380 (190)	210 (47)	410 (54)	390 (39)	140 (32)	81 (8.4)
Oxychlordane	63 (6.2)	77 (8.2)	61 (6.4)	72 (12)	140	140 (18)	150 (26)	160 (21)	860 (360)	540 (78)	850 (180)	940 (110)	740 (47)	950 (170)
t-chlordane	5.8 (0.78)	12 (3.9)	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
c-chlordane	15 (2.4)	21 (4.2)	6.5 (0.60)	8.9 (1.3)	19	23 (3.4)	11 (1.3)	12 (2.1)	38 (8.2)	52 (33)	49 (10)	39 (7.5)	11 (1.0)	11 (1.6)
t-nonachlor	140 (16)	150 (16)	13 (0.84)	17 (2.3)	120	140 (20)	51 (6.0)	74 (2.7)	236 (37)	620 (290)	390 (82)	420 (69)	130 (21)	120 (22)
c-nonachlor	70 (6.2)	73 (8.8)	22 (2.2)	28 (8.0)	210	190 (28)	32 (2.7)	37 (3.0)	38 (8.3)	150 (72)	140 (22)	130 (23)	5.0 (0.72)	4.2 (0.41)
MC5	43 (4.4)	54 (410)	9.1 (0.66)	9.0 (2.1)	100	87 (13)	54 (7.5)	59 (8.3)	65 (4.7)	55 (0.39)	110 (18)	100 (9.4)	79 (6.1)	120 (13)
MC7	0.59 (0.13)	0.96 (0.19)	0.34 (0.041)	0.46 (0.18)	1.1	0.81 (0.15)	0.67 (0.065)	0.63 (0.11)	0.81 (0.81)	0.91 (0.91)	1.5 (0.26)	1.5 (0.26)	0.60 (0.071)	0.74 (0.097)
Σchlordanes	430 (43)	484 (52)	150 (6.9)	170 (30)	700	680 (90)	370 (51)	400 (43)	1600 (510)	1600 (520)	2000 (330)	2000 (270)	1100 (84)	1300 (160)

[a] nd, not detected (detection limits ~ 0.02 ng/g).

where X is ^{15}N and R is the corresponding ratio $^{15}N/^{14}N$. The $R_{standard}$ values were based on atmospheric N_2 (AIR) for ^{15}N. Replicate measurements of internal laboratory standards (albumen) indicate measurement errors of ±0.3 ‰ for stable-nitrogen isotope measurements.

2.4. Extraction, cleanup and analysis of samples for chlordanes

Sample extraction and cleanup procedures have been published previously (Norstrom et al., 1988; Letcher et al., 1995). Briefly, a representative sample of tissue (approximately 2 and 5 g for fat and liver, respectively) was ground with anhydrous sodium sulfate, spiked with the internal standard (δ-hexachlorocyclohexane, 2,2′,3,4,4′,5,6,6′-octachlorobiphenyl (PCB 204), tris(4-chlorophenyl)methane and octachloronaphthalene) and extracted with 100 ml (1:1) methylene chloride/hexane. A fraction of the extract was used to determine lipids gravimetrically. Lipids were removed from the sample by automated gel permeation chromatography. The lipid-free eluate, containing the chlordane components, was evaporated to 1 ml and applied to a Florisil column (8 g, 1.2% deactivated). Chlordanes were recovered by consecutive elution with 35 ml hexane (Fraction 1 [F1]), 38 ml of 85% hexane: 15% DCM (F2), and 52 ml of 50% hexane: 50% DCM (F3). F1 contained 75% of *trans*-nonachlor and 100% of PCB 204. F2 contained 25% of *trans*-nonachlor and δHCH, 100% of oxychlordane, *trans*-chlordane, *cis*-chlordane, *cis*-nonachlor, MC5, MC7 and OCN. F3 contained 75% of δHCH, and 100% of heptachlor epoxide and TCPMe. All fractions were roto-evaporated, transferred to 2,2,4-trimethyl pentane and were evaporated to approximately 125 and 1000 μL for liver and fat samples, respectively. Aldrin was added as a volume corrector.

Samples were analyzed on a Hewlett Packard 5890 gas chromatograph (GC) equipped with a 60 m×0.25 mm DB-5 column (J & W Scientific, CA, USA) and a ^{63}Ni-electron capture detector (ECD). The carrier gas was H_2 and N_2 was used as the make-up gas for the ECD. External standards were run after every six samples. MC5 and MC7 were quantified based on the response of a *cis*-chlordane standard.

2.5. Chiral analysis

Two chiral columns were used for the analysis of chiral chlordane components. A 30-m fused silica column, 0.25 mm id., β DEX 120 (20% nonbonded permethylated β cyclodextrin), (Supelco Chromatography Products, ON, CA) was used for analysis of *cis*- and *trans*-chlordane, MC5 and MC7. A 30-m fused silica column 0.25 mm ID, 0.18-um film thickness, BGB-172 (BGB Analytic) was used for the analysis of oxychlordane and heptachlor epoxide. To reduce chiral column bleed from entering the mass spectrometer a 1-m section of a DB5 MS column was also joined to the exit end of the analytical column using Supelco Glasseal capillary column connector.

Chiral analyses were performed on a HP 5890 Series II GC coupled with a 5972 mass selective detector (MSD). The temperature program was as follows: injector temp 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. Splitless injection mode was used, with an injection volume of 3.0 μl. The MS was operated in the selected ion monitoring 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 chiral and racemic standards. Peak height was used for quantification of enantiomers. Six samples per tissue per species, only four and five for IVGU, were run for chiral analysis. Samples were combined to improve the detection limits and reduce the number of samples analyzed. Standards were run after every 7th sample. Ratio of enantiomers have been expressed as EFs, rather than more commonly used ER because EF is more easily compared and used in mathematical equations (Harner et al., 2000). The EF was calculated using the following equation:

$$EF = (+)/[(+) + (-)]$$

where (+) and (−) is the height of corresponding enantiomer. The designations of the MC5 enantiomers are unknown, so the EF = (1)/[(1) + (2)], where 1 and 2 correspond to the first and last eluting enantiomer, respectively.

Column resolution (R) for each enantiomeric pair was calculated using a racemic standard for all compounds except MC5. A Ringed Seal reference standard was used to calculate values for MC5. R values were calculated on a 'middle aged column' to provide a mean value since enantiomeric resolving ability of the columns tended to decrease with age. The column resolution was calculated by the following equation:

$$R = 2 \times (rt2 - rt1)/wb1 + wb2$$

where rt1 = retention time of the first eluting enantiomer, rt2 = retention time of the second eluting enantiomer, wb1 = baseline width of the first eluting enantiomer, wb2 = baseline width of the second eluting enantiomer.

2.6. Statistical analysis

Due to statistically significant differences in lipid content between tissues and species and non-normally distributed data, all concentrations were lipid normalized and log transformed prior to statistical analysis.

Differences in $\delta^{15}N$ and ΣCHLOR concentrations between species, tissues and sex were compared with an ANOVA by a Newman–Keuls aposteriori test. Due to insufficient sample numbers for the number of chlordane components, chlordane component concentrations were first analyzed using principal component analysis (PCA) to reduce the number of variables. Factor scores from the PCA were then used to assess differences among species, tissues and sex using analysis of variance (ANOVA). *Trans*-chlordane and MC7 were eliminated from the statistical analysis due to the large number of non-detects.

3. Results

3.1. Characteristics of seabirds analyzed

All seabirds used for chlordane analysis appeared in good nutritional condition. Mean $\delta^{15}N$ values in muscle and masses of seabird species are presented in Table 1. $\delta^{15}N$ values differed significantly among seabird species ($P<0.001$; ANOVA, F-value = 47.1, df = 6,44, $n=58$) but not between sexes ($P=0.25$; ANOVA, F-value = 1.36, df = 1,44, $n=58$) or species × sex ($P=0.73$; ANOVA, F-value = 0.59, df = 6,44, $n=58$). Based on $\delta^{15}N$ results, GLGU were feeding at the highest trophic level followed by BLGU, IVGU, NOFU, TBMU, BLKI and DOVE.

3.2. Concentrations and proportions of chlordane components

Chlordane component concentrations (wet weight) in liver and fat are presented in Tables 2 and 3. Most of the chlordane components were quantified in all samples, the exceptions were *trans*-chlordane and MC7 which fell below detection limits for many of the samples. Although concentrations of *trans*-chlordane were not quantified, EFs of *trans*-chlordane were determined by GC–MSD. The chiral analysis used pooled samples and reduced final volumes which resulted in greater concentrations and better detection. However, because the samples were pooled and brought to a low final volume, we could not confidently determine a concentration from the chiral analysis.

Lipid corrected concentrations of ΣCHLOR were not significantly different between sexes ($P=0.18$; ANOVA, F-value = 1.8, df = 1,100, $n=122$), within species ($P=0.78$; ANOVA, F-value = 0.54, df = 6,100, $n=122$), or within tissue ($P=0.92$; ANOVA, F-value = 0.01, df = 1,100, $n=122$). After removing sex as a variable, lipid-corrected concentrations of ΣCHLOR were significantly different among species ($P<0.001$; ANOVA, F-value = 78.39, df = 6,100, $n=122$) and tissues ($P=0.02$; ANOVA, F-value = 5.58, df = 1,100, $n=122$; Fig. 2). There was also significant interactions of species and tissues concentrations of ΣCHLOR ($P<0.001$; ANOVA, F-value = 6.0, df = 6,100, $n=122$). DOVE was the only seabird where lipid corrected concentrations of ΣCHLOR were significantly different between liver and fat. In general, ranked by ΣCHLOR concentrations GLGU, IVGU, NOFU > BLGU, BLKI, DOVE fat > TBMU, DOVE liver (Fig. 2).

Lipid-corrected concentrations of individual chlordane components varied between species and components (Fig. 3). Statistical analysis of each of the individual chlordane components was not performed because the number of samples was not sufficient. A PCA produced two significant factors (loadings > 0.7). Heptachlor epoxide, oxychlordane, *trans*-chlordane and MC5 loaded significantly on to Factor 1 (eigenvalue = 3.19, variance explained = 0.53), and *cis*-chlordane and *cis*-nonachlor loaded on to Factor 2 (eigenvalue = 2.12, variance explained = 0.35). However, further analysis on the factors did not provide any insight on species differences, although an ANOVA of factor loadings did show that sex was not a significant variable. Liver and fat concentrations of individual chlordane components were generally equal within species, although there were exceptions (Fig. 3). In general, IVGU, GLGU and NOFU had the highest concentrations, but differences were observed for *cis*-nonachlor, *trans*-nonachlor, MC5, MC7 and *trans*-chlordane.

Relative proportions of the chlordane components were similar between liver and fat for each species, but differed among species (Fig. 4). Oxychlordane was the predominant chlordane in TBMU, BLKI, IVGU, GLGU and NOFU, *trans*-nonachlor in BLGU and *cis*-nonachlor in DOVE.

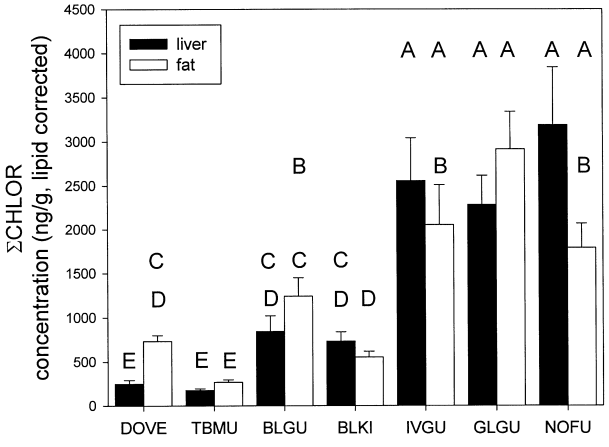

Fig. 2. Lipid corrected concentrations (mean±S.E., ng/g) of ΣCHLOR in liver and fat of Northwater seabirds. Male and females did not significantly differ ($P>0.05$) and were combined. Bars with the same letter do not differ significantly ($P>0.05$). DOVE, dovekie; TBMU, thick-billed murre; BLGU, black guillemot; BLKI, black-legged kittiwake; IVGU, ivory gull; GLGU, glaucous gull; NOFU, northern fulmar.

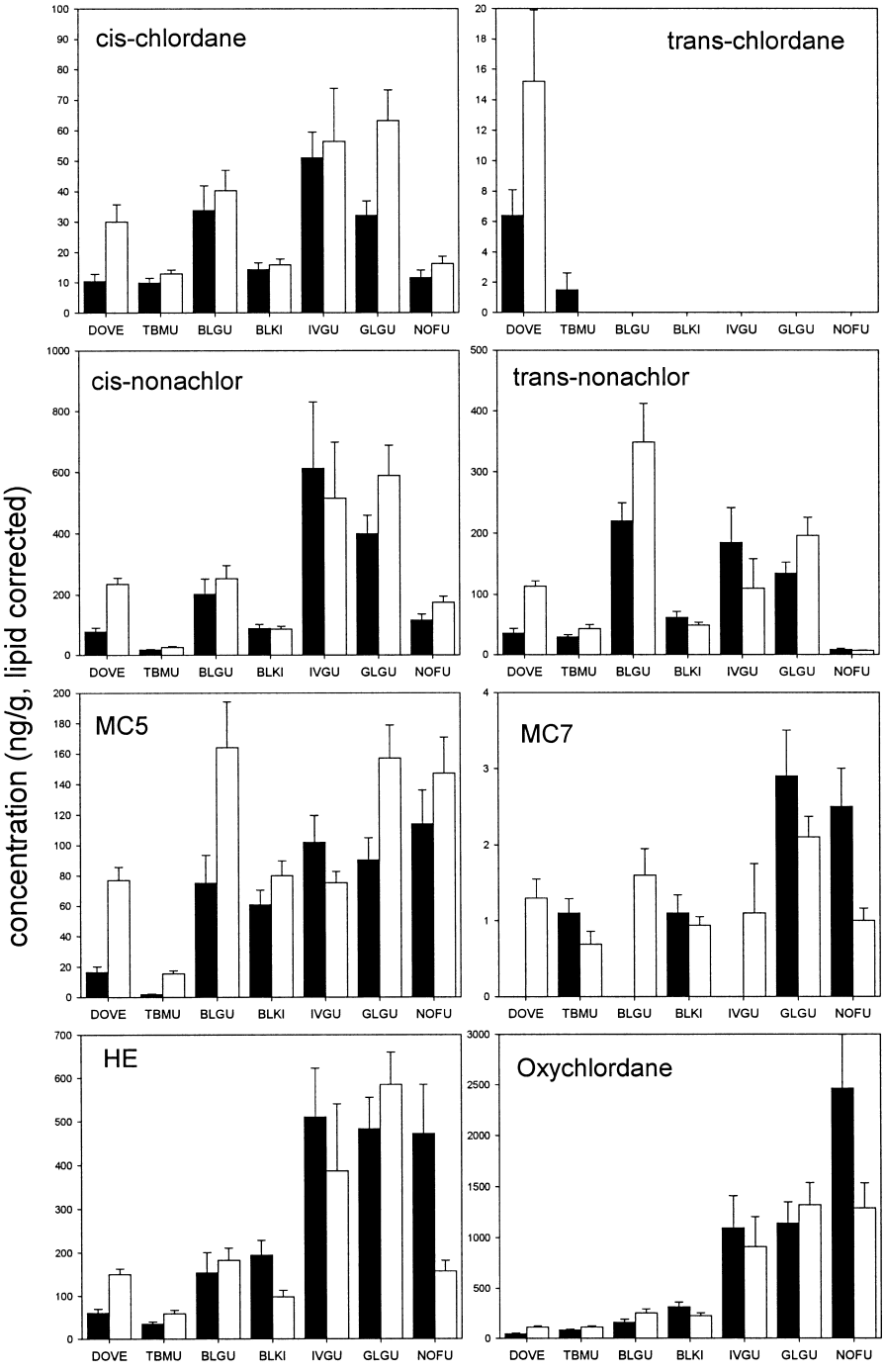

Fig. 3. Lipid-corrected concentrations (mean±S.E., ng/g) of chlordane components in liver (black bars) and fat (open bars) of Northwater seabirds. Male and females did not significantly differ ($P > 0.05$) and were combined. DOVE, dovekie; TBMU, thick-billed murre; BLGU, black guillemot; BLKI, black-legged kittiwake; IVGU, ivory gull; GLGU, glaucous gull; NOFU, northern fulmar.

3.3. Chlordane-$\delta^{15}N$ relationships

$\delta^{15}N$ values explained a significant proportion of the variability in ΣCHLOR and all chlordane component concentrations, except MC7, in seabirds (Fig. 5, Table 4). $\delta^{15}N$-chlordane component slopes were greater in liver than fat, and overall ranged from −1.4 to 0.46 (Table 4). Relationships for *trans*-chlordane were not determined because *trans*-chlordane was only quantified in DOVE and TBMU liver.

3.4. Enantiomer fractions

All of the chiral chlordane components were separated from each other and interference from other chlorinated compounds was not a problem. Good

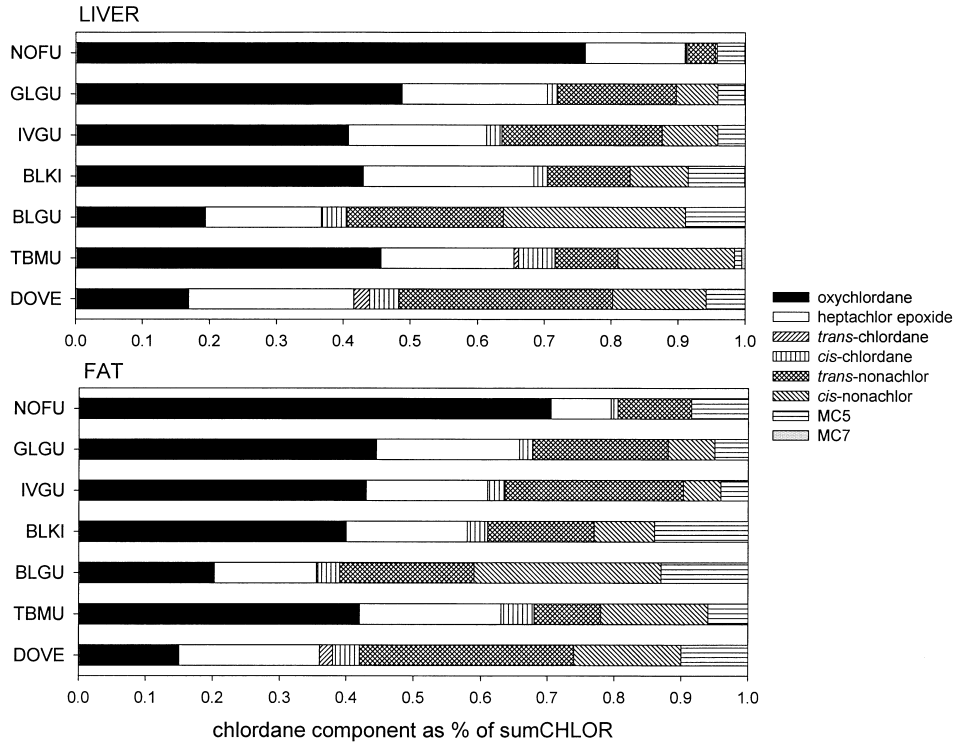

Fig. 4. Relative proportions of chlordane components in liver and fat of Northwater seabirds. DOVE, dovekie; TBMU, thick-billed murre; BLGU, black guillemot; BLKI, black-legged kittiwake; IVGU, ivory gull; GLGU, glaucous gull; NOFU, northern fulmar.

separation of enantiomers was achieved for all of the chiral chlordane components. *R* values were good for all chiral compounds: *cis*-chlordane = 0.62, *trans*-chlordane = 0.50, MC5 = 0.57, oxychlordane = 1.67 and heptachlor epoxide = 1.6.

Trans-chlordane levels were low in seabird livers and in most cases were not detected during chiral analysis. In a number of samples the (−)enantiomer of *trans*-chlordane appeared to be present but not at levels which were quantifiable. The EFs of the (+)enantiomers for the chiral chlordanes are summarized in Table 5. Non-racemic EFs were found for most chlordanes in most samples. There was a depletion of the (+)enantiomer of *cis*- and *trans*-chlordane and MC5 (EF < 0.5) and enrichment of the (+)enantiomer of oxychlordane and HE (EF > 0.5). These EFs were generally consistent between tissues and seabirds. There was an enrichment of (+)enantiomer of *cis*-chlordane in NOFU fat and liver and an enrichment of the (+)enantiomer of MC5 in DOVE and BLKI fat and liver which was not consistent with the other birds.

4. Discussion

This study represents one of the most comprehensive analyses of chlordane in Arctic seabirds in that seven species and two tissues were analyzed and stable isotopes of N and EF of chiral chlordanes were determined. Concentrations of chlordane were related to trophic level as described by $\delta^{15}N$ values, but the amount of variation explained was low and other factors may have influenced concentrations. The $\delta^{15}N$ values of seabird pectoral muscle tissue provided trophic-level estimates corresponding to about a month of dietary integration (Hobson, 1993). Thus, factors influencing chlordane concentrations outside the breeding season may have contributed to the variance in our trophic model. The relative proportions of chlordane components and EFs of the chiral components suggest that biotransformation is related to phylogeny, with the exception of the TBMU.

4.1. Chlordane-$\delta^{15}N$ relationships

The amount of variability in chlordane concentrations in seabirds described by $\delta^{15}N$ values was statistically significant for all but MC7, but less than what has been reported for complete food chains (Kidd et al., 1998a). The reduced ability of $\delta^{15}N$ values to describe differences in chlordane concentrations in seabirds may be due to the fact that $\delta^{15}N$ in these seabirds only gives a measure of short-term trophic position. Migration from areas of different relative contamination may confound these relationships in Arctic seabirds. Such factors are minimized in the lake food chain described by Kidd et al. (1998a). Other factors which could reduced the ability to $\delta^{15}N$ to describe chlordane concentrations are smaller

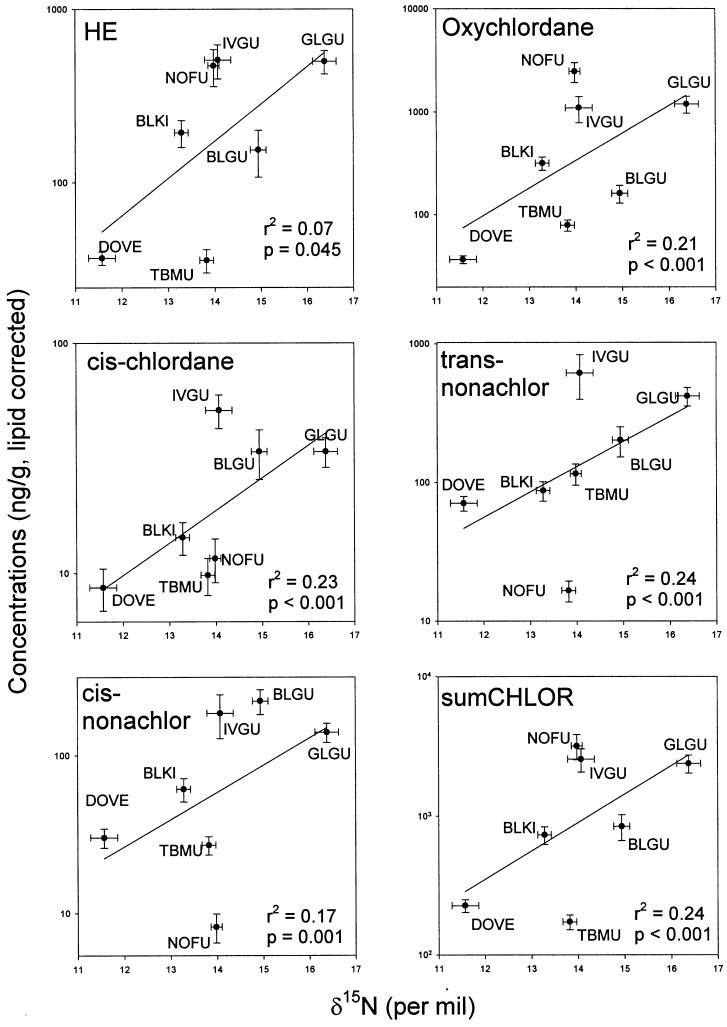

Fig. 5. δ^{15}N–chlordane component concentration relationships in Northwater seabirds. Chlordane concentrations were determined in liver and δ^{15}N in pectoral muscle. Each point is the mean±1 S.E. for both the concentration and δ^{15}N. Intercepts and slopes are provided in Table 4. DOVE, dovekie; TBMU, thick-billed murre; BLGU, black guillemot; BLKI, black-legged kittiwake; IVGU, ivory gull; GLGU, glaucous gull; NOFU, northern fulmar.

differences in trophic levels among seabird species and more variable biotransformation capabilities in the seabirds compared to those in complete food chains. δ^{15}N measurements are a poor predictor of PCB concentrations in Great Lakes herring gull eggs (*Larus argentatus*; Hebert, personal communication). Chlordane concentrations in a number of seabird species from this study were lower or higher than expected based on chlordane–δ^{15}N relationships. For example, ΣCHLOR concentrations in TBMU were well below what would be predicted from the ΣCHLOR–δ^{15}N relationships. This reveals a potential limitation in using δ^{15}N to predict concentrations in Arctic seabirds but it also provides insights on biotransformation ability of the seabirds and assists in interpretation of other results, such as EFs and relative proportions of chlordane components.

Slopes of the δ^{15}N-chlordane component relationships give a relative rate of bioaccumulation or biomagnification in aquatic food chains (Broman et al., 1992; Rolff et al., 1993). Comparisons of the δ^{15}N-chlordane slopes for the NOW seabirds to other food chains are limited because of the lack of published relationships for chlordane. Norstrom (1994) calculated a δ^{15}N–ΣCHLOR slope ($b = 0.49$) for the Resolute marine food chain (Hobson and Welch, 1992) which included zooplankton, fish, seabirds and marine mammals which was similar to the ΣCHLOR slope calculated in this study. A slope of 0.16±0.05 was reported for ΣCHLOR for a freshwater food web in a subarctic lake (Kidd et al., 1998b) and a slope of 0.17±0.02 for *trans*-nonachlor in an arctic freshwater food web (Kidd et al., 1998a), both of which included only phytoplankton, zooplankton and fish. Based on these other studies and the results of this work, birds bioaccumulate chlordane to a greater extent than fish or zooplankton.

Differences in bioaccumulation of organic contaminants in aquatic food chains are related to the hydrophobicity, or octanol–water partition coefficient

(K_{ow}) of the chemical (Kidd et al., 1998a) and the susceptibility of the contaminant to biotransformation. Birds, in general, have a greater ability to biotransform contaminants than fish and zooplankton (Norstrom et al., 1978), making biotransformation a more important variable than chemical hydrophobicity in the bioaccumulation of organic contaminants by seabirds (Braune and Norstrom, 1989). However, there is some evidence that hydrophobicity does influence the bioaccumulation rate of recalcitrant POPs in seabirds (Braune and Norstrom, 1989) and there is a need to do toxicokinetic studies of chlordanes and other POPs in birds. Slopes of the $\delta^{15}N$–ΣCHLOR relationships for most of the individual components were similar to ΣCHLOR. The exceptions were the components which are biotransformed (i.e. *cis*- and *trans*-chlordane [Buser et al. 1992]) and those which are formed, at least partially, through biotransformation (i.e. oxychlordane [Nomeir and Hajjar, 1987]). *Cis*- and *trans*-chlordane had the lowest slopes suggesting that the rate of metabolism increased and/or the rate of exposure decreased with trophic level. Oxychlordane had the highest slope, suggesting greater formation and/or exposure with trophic level. Slopes were lower for fat, which may be driven by the higher concentrations in DOVE fat as compared with DOVE liver, the seabird with the lowest $\delta^{15}N$ muscle value.

Table 4
Intercepts, slopes, coefficient of determination and *P*-values for $\delta^{15}N$-chlordane component relationships in Northwater seabird livers and fat

Chlordane component	Intercept	Slope	r^2	*P*-value
Liver				
HE	6.7	0.36	0.07	0.045
Oxychlordane	6.1	0.46	0.21	<0.001
cis-chlordane	5.8	0.28	0.23	<0.001
trans-nonachlor	5.6	0.42	0.24	<0.001
cis-nonachlor	5.5	0.37	0.17	0.001
MC5	5.8	0.33	0.10	0.02
MC7	−3.8	0.61	0.06	0.06
ΣCHLOR	8.1	0.40	0.24	<0.001
Fat				
HE	7.8	0.30	0.23	<0.001
Oxychlordane	6.4	0.45	0.34	<0.001
cis-chlordane	6.7	0.24	0.25	<0.001
trans-nonachlor	8.2	0.26	0.11	0.01
cis-nonachlor	7.7	0.23	0.08	0.04
MC5	8.2	0.21	0.08	0.03
MC7	3.8	0.21	0.05	0.10
ΣCHLOR	9.3	0.23	0.25	<0.001

4.2. EF

EFs of the chiral chlordane components were generally consistent among seabird species. Notable exceptions were the enrichment of the (+)enantiomer of *cis*-chlordane in NOFU and the first eluting (F1) enantiomer of MC5 in BLKI, the opposite of what was observed for the other seabird species. No differences in the ratio of the enantiomers were observed between tissues of any seabird species. Inverse ERs have been observed in the liver and blubber of individual seals (Wiberg et al., 1998). There was a depletion of the (+) or (F1) enantiomer of chiral components found in technical chlordane, namely *cis*- and *trans*-chlordane and MC5. Depletion of the (F1) enantiomer of MC5 has

Table 5
Enantiomeric fractions of the (+) enantiomer in NOW seabirds[a]

Species	Tissue	*n*	*cis*-chlordane	*trans*-chlordane	MC5[b]	Oxy chlordane	Hepatchlor epoxide
Standards							
Racemic	exp.		0.50	0.50	ns	0.50	0.50
	obs.	18	0.49±0.002	0.49±0.001	ns	0.50±0.001	0.50±0.001
Seabirds							
DOVE	liver	5	0.37±0.11	nq	0.52±0.01	0.54±0.01	0.63±0.03
	fat	6	0.19±0.01	0.35±0.05	0.52±0.01	0.55±0.02	0.56±0.01
TBMU	liver	6	0.25±0.02	nq	0.34±0.05	0.67±0.03	0.77±0.03
	fat	6	0.34±0.02	0.33±0.05	0.32±0.02	0.71±0.02	0.80±0.02
BLGU	liver	6	0.30	nq	0.35±0.03	0.56±0.02	0.54±0.01
	fat	5	0.25±0.01	0.23±0.05	0.34±0.01	0.56±0.01	0.61±0.05
BLKI	liver	6	0.16±0.01	nq	0.57±0.01	0.66±0.01	0.69±0.01
	fat	5	0.27±0.02	0.40±0.07	0.67±0.01	0.60±0.01	0.62±0.01
IVGU	liver	5	0.29±0.03	nq	0.37±0.02	0.60±0.01	0.60±0.01
	fat	4	0.22±0.04	0.22±0.02	0.39±0.01	0.64±0.05	0.61±0.08
GLGU	liver	6	0.30±0.04	nq	0.31±0.02	0.63±0.01	0.62±0.01
	fat	7	0.26±0.04	0.23±0.04	0.27±0.01	0.65±0.03	0.61±0.07
NOFU	liver	6	0.69±0.12	nq	0.35±0.01	0.61±0.01	0.57±0.02
	fat	6	0.53±0.01	0.22±0.05	0.33±0.01	0.61±0.05	0.61±0.05

[a] Compounds which were not quantified due to low concentrations or were not detected are indicted by nq.
[b] There was a limited amount of MC5 standard, so replicate samples could not be analyzed.

been reported in Antarctic penguins (*Pygoscelis adelis*) and Baltic Sea grey seals (*Halichoerus grypus*; Buser et al., 1992) and in ringed seals (*Phoca hispida*) and polar bears from the Canadian Arctic (Wiberg et al., 2000). Wiberg et al. (1998) reported a depletion of the (+)enantiomer of *cis*- and *trans*-chlordane in most of the grey, ringed and harp seals (*Pagophilus groenlandicus*) samples analyzed, although there were a number of exceptions.

There was an enrichment of the (+)enantiomer of oxychlordane, a metabolite of *cis*- and *trans*-chlordane, and heptachlor epoxide in all seabirds. This suggests that there is a direct conversion of the (+)enantiomer of *cis*- and *trans*-chlordane into the (+)enantiomer of oxychlordane in these seabirds. Enrichment of the (+)enantiomer of oxychlordane and heptachlor epoxide were reported in penguins and grey seals (Buser and Müller, 1993), and Arctic cod (*Boreogadus saida*) and polar bear (Wiberg et al., 2000). However, a depletion of (+)enantiomer of heptachlor epoxide has been observed in grey, ringed and harp seals (Wiberg et al., 1998, 2000). Some differences in the relative abundance of chlordane enantiomers suggest that the metabolic pathways for chlordane metabolism, e.g. CYP2B enzymes, may differ between seabirds and marine mammals, particularly seals.

The magnitude of the EFs of *cis*- and *trans*-chlordane, MC5 and heptachlor epoxide were consistent between seabird tissues, sex and species. Wiberg et al. (1998) reported that there were few differences in ERs of chiral chlordane between sex and species of seal. Oxychlordane EFs measured in the alcids (DOVE and BLGU) were lower than those observed in the larids (BLKI, IVGU and GLGU) and the procellariid (NOFU), suggesting lower biotransformation ability or activity. TBMU had the highest oxychlordane and heptachlor epoxide EFs of any seabird species, suggesting greater biotransformation ability or activity. Significant metabolism of heptachlor epoxide may be limited to the TBMU. EFs of heptachlor epoxide were consistent for the other 6 seabirds and in the range of those reported for Arctic seawater (Jantunen and Bidleman, 1998) and Arctic cod (Wiberg et al., 2000), suggesting that EFs in these seabirds may reflect what was bioaccumulated.

EFs were not related to the chlordane concentrations or to trophic level, and appear to have very little potential to predict these variables in seabirds. Previous work on EFs of chiral pollutants, in particular α-HCH, in various components of food chains have shown differences between groups of organisms (i.e. fish vs. seals; Wiberg et al., 1998, 2000). Wiberg et al. (2000) also concluded that there was no trend in chlordane EFs with trophic level in an Arctic marine food web that included Arctic cod, ringed seal and polar bear. The metabolic similarities of closely-related organisms, such as seabirds, probably precludes using EFs to predict their concentration or trophic level. However, the relative amounts of oxychlordane were somewhat related to the EFs and suggest that EFs provide information on relative biotransformation ability or activity.

4.3. Relative contribution to ΣCHLOR

The proportions of chlordane components in ΣCHLOR were similar between the liver and fat of the seabirds and, with the exception of TBMU, appeared to follow the phylogenetic relationship among the seabirds species. Alcids, BLGU and DOVE, had the smallest relative amount of oxychlordane, less than half of what was observe in the larids, BLKI, IVGU and GLGU. The procellariid, NOFU, had the greatest percentage of oxychlordane but also low levels of *cis*- and *trans*-nonachlor. A high percentage of oxychlordane, along with differences in EF of *cis*-chlordane, suggest that the NOFU is distinct with respect to chlordane metabolism from the other seabird species. However, the apparent difference in metabolic capability of the NOFU did not translate into reduced ΣCHLOR concentrations.

Variations in the relative proportions of chlordane components in seabird species in this study could be due to differences in species metabolic capability, enzyme induction and/or bioaccumulation, although the evidence suggests that it is mainly due to metabolism. Oxychlordane is a metabolite of *cis*- and *trans*-chlordane formed by mammals and birds, but evidence that invertebrates and fish form it are lacking. However, oxychlordane is found in Arctic fish and invertebrates but at much lower levels than *cis*- and *trans*-chlordane (Hargrave et al., 1992). BLGU and DOVE feed only on invertebrates and fish, therefore, oxychlordane in these species is likely predominantly formed in these birds. Since some of the larids and procellarids are know to scavenge dead marine mammals, oxychlordane in these birds could be due to metabolism and/or bioaccumulation. However, BLKI had chlordane concentrations that were much lower than the other larids suggesting minimal marine mammal scavenging. As well, NOFU concentrations or $\delta^{15}N$ values (see later) do not suggest that the NOFU fed at a higher trophic level (i.e. on marine mammals), which could account for such a large percentage of oxychlordane in this species. Due to the low levels of *cis*- and *trans*-nonachlor, NOFU may produce oxychlordane from chlordane components other than *cis*- and *trans*-chlordane. As well, the GLGU, which preys on the chicks of other seabirds during the late breeding season, and was at the highest trophic level based on $\delta^{15}N$ values, had only a slightly higher percentage of oxychlordane than the other larids.

The exception to the phylogeny-related proportions of chlordane components was the TBMU. The relative

amount of oxychlordane in the TBMU was greater than twice that observed in the other alcid seabirds, DOVE and BLGU. Since the alcids do not feed on carrion, the high proportion in TBMU is not due to bioaccumulation. TBMU were consistently below the chlordane–δ^{15}N relationships for many of the chlordane components and ΣCHLOR, but not for oxychlordane or *cis*-nonachlor. This suggests that TBMU was efficient at eliminating chlordane, or alternatively, fed on a diet relatively depleted in chlordane. A diet depleted in chlordane seems unlikely because fish found in the stomachs of the TBMU and the other seabirds were similar. It would appear that the TBMU is distinct from the other seabirds with respect to chlordane metabolism and elimination based on the highest EFs for oxychlordane and hepatchlor epoxide, the lowest chlordane concentrations, and different relative proportions of chlordane components. The relative amount of ΣCHLOR compared with other persistent organic pollutants in the common murre (*Uria aalge*) were found to be low compared to three other seabird species collected in the Gulf of the Farallones (Jarman et al., 1996). It would appear that the ability to more rapidly eliminate chlordane might be common to the Genus *Uria*.

4.4. Comparisons to other studies

Comparisons of the NOW data with other Canadian Arctic data are somewhat difficult in that most POP data are for seabird eggs and there are limited data for adult tissues and individual chlordane components. Lipid-corrected concentrations of PCB and DDT in NOFU, BLKI and TBMU eggs and livers collected from the arctic were not different (Nettleship and Peakall, 1987), and therefore comparisons of lipid-corrected liver and egg data are legitimate. The most recent chlordane data for Canadian Arctic seabirds were collected in 1993 for eggs. Concentrations (l.c.) of ΣCHLOR reported in eggs of BLGU, BLKI and TBMU collected in the Canadian high Arctic (Braune, 1994a, b) were similar to those reported for liver in this study. However, concentrations in GLGU and NOFU eggs were higher and lower, respectively (Braune, 1994a, b), than those reported for liver in this study. Braune (1994a, b) measured the same chlordane components except MC5, a minor component, as were measured in this study. Lipid-corrected concentrations of oxychlordane and heptachlor epoxide in the liver of TBMU, BLKI and NOFU and eggs of IVGU collected in 1975–1976 in the Canadian high Arctic were similar and lower, respectively, than those reported in this study (Noble and Elliott, 1986). ΣCHLOR concentrations (lipid corrected) reported for liver and fat tissues of BLKI and GLGU collected in the southern Barents Sea (Savinova et al., 1995) are also similar to those reported in this work. In general, chlordane concentrations do not appear to have declined, and for some components may have increased, in Arctic seabird species between 1975–1976 and 1998.

ΣCHLOR concentrations of seabirds were in the range of Arctic marine mammals. Weis and Muir (1997) reported ΣCHLOR concentrations (355–516 ng/g l.c.) in blubber of Resolute Bay ringed seals that were in the same range as the more obligate zooplankton and fish-eating birds (i.e. DOVE, TBMU, BLGU and BLKI). Concentrations of ΣCHLOR reported in polar bear blubber (670–5502 ng/g, l.c.) from the same region (Norstrom et al., 1998) were higher but similar to seabirds which are known to scavenge polar bear kills (i.e. IVGU, NOFU and GLGU).

Lipid-corrected concentrations of ΣCHLOR were not significantly different between sexes or between tissues for any seabird species except for tissues in DOVE. This suggests that despite migration and reproductive effort of many of these birds just prior to collection, ΣCHLOR is in relative steady state between tissues of the bird. The lower concentrations in liver of DOVE relative to fat suggest that this bird has recently moved from an area of high to low chlordane contamination and/or has changed to a less-contaminated diet. The liver is a more high-profused tissue than fat, and therefore would likely respond more quickly to changes in contaminant exposure and reflect more recent exposure to contaminants than fat (Ivie et al., 1974; Gobas et al., 1999). Chlordane concentrations in fat may be more reflective of long-term exposure which would include the winter diet of the DOVE. The DOVE migrates from the St. Lawrence estuary and eastern seaboard of Canada/US, areas of high contamination (Muir et al., 1990) consistent with this conclusion. However, many of the other species (TBMU, BLKI, NOFU and GLGU) also migrate from more contaminated temperate regions. Lower concentrations in the liver of DOVE may reflect post migration and pre-breeding diet of pelagic zooplankton (*Calanus hyperboreus* and *Calanus glacialis*) in May and June (Karnovsky, unpublished data). During the winter and for the remainder of their time in the NOW, July–September, DOVE fed predominately on pelagic fish (Karnovsky, unpublished data). Such an increase in feeding at this low trophic level would result in a pulse of less-contaminated lipid into the liver, pushing the concentration of chlordane in liver below those in fat. DOVEs are the only one of the migratory seabirds that fed predominantly on this zooplankton during this time (Karnovsky, unpublished data).

Acknowledgements

Funding for this project was provided in part by an Natural Science and Engineering Research Council

(NSERC) Network grant on the Northwater Polynya, Canadian Wildlife Service, Polar Continental Shelf Project and the National Science Foundation (NSF) to George Hunt. The authors would like to thank the many scientists of the NOW project, especially Ian Stirling, George Hunt, Dennis Andriashek and Nick Lunn, Michael Mulvihill for chemical analysis, and Patricia Healy for assistance with sample preparation. Stable isotope analysis were performed by Garth Parry, University of Saskatchewan. We thank the HTA of Grise Fjord for granting us permission to collect birds and other samples in their area. Birgit Braune and Craig Hebert provided insightful reviews of the manuscript. This project would not have been possible without the dedicated and skillful assistance of the *Pierre Radisson* Coast Guard captains and crew.

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