



New and Established Organohalogen Contaminants and Their Metabolites in Plasma and Eggs of Glaucous Gulls From Bear Island

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Photo: Gunnar Sander



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1. Preface

The rationale of this project was to generate new knowledge on new organohalogen contaminants and metabolites of organohalogens in glaucous gulls (*Larus hyperboreus*) breeding at Bear Island (Bjørnøya). The congener patterns and the current levels in plasma and egg homogenate samples were examined, and compared to those reported in free-living mammals, seabirds, and eggs of seabirds. Future work ensuing this present investigation will aim at assessing the toxicological potential of these new environmental contaminants in glaucous gulls by ultimately investigating the correlative relationships between the current levels in plasma and eggs and biomarkers of biological effects.

The present study involved the collaboration between the Norwegian Polar Institute, and leading international analytical laboratories: the Great Lakes Institute for Environmental Research (University of Windsor, Windsor, Canada) and the National Water Research Institute (Environment Canada, Burlington, Canada). Funding was provided by the Norwegian Pollution Control Authority (SFT) and the Ecotoxicology Programme at the Norwegian Polar Institute. This project is part of a 3-year doctoral fellowship (Norwegian Research Council (NFR) project no. 160919/V10- Jonathan Verreault) assessing the effects of new and established organohalogen contaminants and their metabolites on the endocrine, enzymatic, and metabolic systems of glaucous gulls from Bear Island.

This screening study responds to the most recent recommendations formulated by PROFO (a programme initiated by the NFR), SFT, and Arctic Monitoring and Assessment Programme (AMAP).

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3. Summary

In the present report, we examined the congener patterns and the current levels of new and established organohalogen contaminants and their metabolites in plasma and egg homogenate samples of glaucous gulls breeding at Bear Island (Bjørnøya).

The chemical analyses of glaucous gull plasma and egg homogenate samples revealed the presence of several major classes of established and new organohalogen contaminants of concerns, and their metabolites. Besides the routinely-analyzed organochlorines (OCs) (i.e. polychlorinated biphenyls (PCBs) and a suite of pesticides and by-products), less studied and new organohalogen contaminants such as coplanar PCBs, toxaphenes, polychlorinated naphthalenes (PCNs), brominated flame retardants (BFRs), and perfluorinated compounds (PFCs) were detected in plasma and egg homogenate samples. Hexachlorobutadiene (HCB) was not detected at the established method detection limit in any of the plasma and egg homogenate samples of glaucous gulls. Levels of coplanar PCBs and PCNs quantified in plasma and egg homogenate samples in the present study were higher compared to those in tissue and egg samples of seabirds from Svalbard and the Canadian Arctic. Compounds of BFRs, i.e. hexabromocyclododecane (HBCDD), polybrominated biphenyls (PBBs), and polybrominated diphenylethers (PBDEs), were all detected in plasma and egg homogenate samples. PBDEs in glaucous gull samples were determined at levels generally lower than those of seabirds and eggs of seabirds of North America and central and southern Europe. Low concentrations of PBBs and HBCDD were quantified in plasma and egg homogenate samples suggesting that these chemicals are of low importance compared to PBDEs in glaucous gulls breeding at Bear Island. One congener of PBDEs, the highly brominated compound BDE-209 (decaBDE), was detected in 15% of the egg homogenate and 30% of the plasma samples. BDE-209 appears thus to have a bioaccumulative potential in the glaucous gull and possibly in the Barents Sea food web. Considering the male/female ratio studied and the fact that fewer egg homogenate samples had detectable levels of BDE-209, we suggest that the degree of bromination have impact on the transfer rate of PBDE congeners from mother to eggs. Perfluorooctane sulfonate (PFOS), a compound of PFCs, was quantified in plasma samples at levels higher than those of some legacy OCs, being comparable to e.g. levels of DDTs. Hence, PFOS appears to be a chemical of relatively high importance in glaucous gull plasma.

Furthermore, in plasma and egg homogenate of glaucous gulls was revealed the presence of metabolites of organohalogenes such as methylsulfonyl (MeSO₂) PCBs and *p,p'*-DDE, and hydroxylated (HO) PCBs and PBDEs. However, HO-PBDEs were not detected in any of the egg homogenate samples. Our study has demonstrated that PCB and PBDE metabolism is a source for generation of unique classes of persistent organohalogenes in glaucous gulls. However, our results suggested also a low metabolic ability of glaucous gulls towards congeners of PCBs and PBDEs relatively to e.g. marine mammals such as the polar bear. Metabolite formation may be a good indicator of the importance of organohalogen biotransformation in glaucous gulls vs other species.

4. Sammen drag

I denne rapporten ble sammensetning og nivåer av nye og etablerte organohalogene forbindelser, samt deres metabolitter, undersøkt i egg og plasma prøver hos hekkende polarmåker fra Bjørnøya.

De kjemiske analysene av polarmåke plasma og egg viste tilstedeværelsen av forbindelser som rutinemessig blir analysert, samt nye faretruende miljøgifter og deres metabolitter. Foruten rutinemessig analyserte klororganiske forbindelser (OCer) (som polyklorerte bifenyler (PCBer) og en rekke pestisider og biprodukter fra kjemisk industri), ble andre mindre studerte og nye miljøgifter som koplanare PCBer, toksafen, polyklorerte naftalener (PCNer), bromerte flammehemmere (BFRer) og perfluorerte forbindelser (PFCer) påvist i polarmåkene. Heksaklorbutadien (HCBDD) ble derimot ikke påvist over deteksjonsgrensen i noen av de analyserte plasma og egg prøvene. Nivåer av koplanare PCBer og PCNer var høyere sammenliknet med nivåer i andre sjøfuglearter fra Svalbard og Canada. BFR, dvs HBCDD, PBBer og PBDEer, ble påvist i alle plasma og egg prøver. PBDE nivåene i polarmåke var generelt lavere enn tidligere rapporterte nivåer i sjøfugl og sjøfugl egg fra Nord-Amerika og Sentral/Sør-Europa. Lave konsentrasjoner av PBBer og HBCDD er også rapportert i polarmåke plasma og egg. PBBer og HBCDD er foreslått kjemiske forbindelser av lav betydning i polarmåker på Bjørnøya sammenliknet med PBDEer. En høyt bromert PBDE, BDE-209 (dekaBDE), ble påvist i 15 % av eggene og 30 % av plasma prøvene. BDE-209 kan ha potensiale til å bioakkumulere i polarmåke, samt i marine næringskjede i Barentshavet. Etter en undersøkelse av forholdet mellom hann/hunn nivåer av BDE-209, samt det faktum at færre egg hadde påviste nivåer av BDE-209, ble det foreslått at antall brom atomer hadde en innvirkning på raten PBDE er overført fra mor til egg. Perfluoroktan sulfonat (PFOS) (inngår i gruppen PFCer) ble rapportert i høye konsentrasjoner i forhold til enkelte rutinemessig analyserte OCer. Nivåer av PFOS kunne sammenliknes med plasma nivåer av DDTer. Følgelig var PFOS en av de mest fremtredende kjemiske forbindelser i polarmåke plasma.

Både i polarmåke plasma og egg ble det påvist tilstedeværelsen av metabolitter av organohalogener, som metylsulfon (MeSO₂) PCBer og *p,p'*-DDE, og hydroksy (HO) PCBer og PBDEer. HO-PBDE derimot, ble ikke påvist i noen av polarmåke egg prøvene. I vårt studie har vi påvist at metabolisme av PCBer og PBDEer er kilden til utvikling av en rekke persistente organohalogener i polarmåke. Resultatene i denne rapporten indikerer også en lavere metabolsk aktivitet for PCBer og PBDEer i polarmåke sammenliknet med marine pattedyr som for eksempel isbjørn. Evnen til å danne metabolitter kan være en indikator på biotransformerings potensialet av organohalogene forbindelser i polarmåke kontra andre dyrearter.

5. Background

Recently, new concerns were raised as new chemical compounds of current widespread commercial use (e.g. some compounds of brominated flame retardants (BFRs) and perfluorinated compounds (PFCs)), were identified in Arctic biota. Several of these new chemicals are increasing temporally and have emerged as important classes of environmental contaminants as tissue residues in aquatic species worldwide, including the remote Arctic (AMAP, 2004). Recent research has focused particularly on BFRs and PFCs, and their potential effects on the long-term exposed organisms. Other new classes of environmental contaminants such as hexachlorobutadiene (HCB), toxaphenes, and polychlorinated naphthalenes (PCNs) have demonstrated toxicological properties in organisms. Although the commercial use of polybrominated biphenyls (PBBs) and certain polybrominated diphenylethers (PBDEs) has been banned in many countries or under increasing regulation, a global ban of these substances is inexistent. There is yet no ban or phase out contemplated for hexabromocyclododecane (HBCDD) (Alaee et al., 2003). For PFCs, chemicals related to perfluorooctane sulfonate (PFOS) have been withdrawn by the manufacturer (e.g. 3M) and were subsequently subjected to restrictions in the EU and USA, although no global ban has been proposed. Perfluorooctanoic acid (PFOA) and related perfluorocarboxylates continue to be used but are under increasing regulatory scrutiny. The ubiquitous presence of these substances in the environment is of major concern among environmental researchers. Hitherto, the general understanding of the dynamic and toxicology of these new contaminants in the environment and biota is limited. Further investigations are necessary to evaluate whether these compounds have a similar potential to alter biological functions of exposed animals compared to chemicals well established as environmental contaminants.

The biotransformation of organohalogenes ideally results in detoxification, and an increased likelihood for elimination from the organism via metabolite formation. However, metabolites of established and new organohalogen contaminants, i.e. methylsulfonyl (MeSO₂) PCBs and *p,p'*-DDE, and hydroxylated (HO) PCBs and PBDEs, are being detected as contaminant residues in tissues and blood of an increasing number of species (Letcher et al., 2000). In recent years, these classes of metabolites have emerged as important environmental contaminants in wildlife and humans. A number of studies have demonstrated the toxicological potential of MeSO₂-PCBs including tissue selective retention via non-covalent protein binding, induction of CYP enzymes, and endocrine-related effects (Letcher et al., 2000). More recently, HO-PCBs and other chlorinated phenolic compounds have gained greater scientific notoriety in environmental toxicology as a consequence of the capability of certain compounds to bind with the thyroid hormone transport proteins (e.g. transthyretin), and to interact with thyroid and estrogen hormone receptors (Sandau, 2000).

A wide range, and occasionally very high levels, of organochlorine (OC) contaminants have been reported in Svalbard/Bear Island glaucous gull tissue and blood samples (AMAP, 2004). High levels of polychlorinated biphenyls (PCBs) and OC pesticides and by-products in glaucous gulls have been associated to reproductive, behavioural, developmental, genotoxic, thyroid, and immunological effects (Sagerup et al., 2000; Bustnes et al., 2001, 2002, 2003; Verreault et al., 2004; Østby et al., in press). Large differences in OC levels reported between individuals of glaucous gulls breeding within the same colony were attributed to metabolic capacity (enzyme induction), tissue/blood lipid content, sex, nutritional status, and feeding ecology (Henriksen et al., 1998, 2000; Bustnes et al., 2000, 2003).

In this study we have determined a suite of new and established organohalogen contaminants and their metabolites in plasma and egg homogenate samples of glaucous gulls breeding at Bear Island.

6. Materials and Methods

A total of 109 adult glaucous gulls were captured at Bear Island during the incubation period in May-June 2002 and 2004 from two major breeding colonies (*Figure 1*). Blood samples were taken from all birds, and plasma was obtained by centrifugation of blood. Biological parameters, i.e. sex, size measurements, and body mass, were recorded (*Table 1*), and the birds were released in the colony subsequently to sampling. Additionally, 32 eggs of glaucous gulls were randomly collected early in the incubation period from the same colonies. All samples were stored at -20 or below prior to chemical analyses.

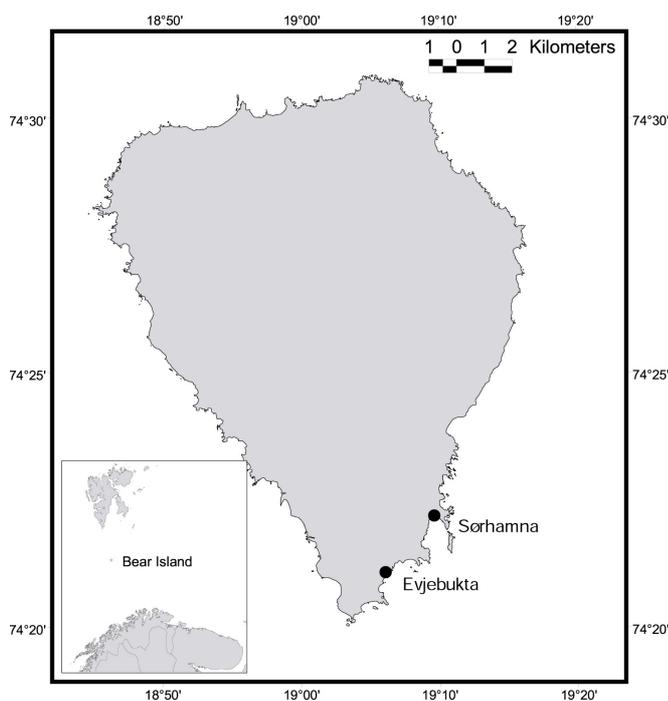


Figure 1. Map of the Barents Sea and Bjørnøya showing the two breeding colonies of glaucous gulls (*Evjebukta* and *Sørhamna*).

for linearity of signal response and quantitative precision and sensitivity. The minimum method detection limits (MDLs) for quantification of individual analytes using GC- μ ECD and GC-(ECNI)MS were determined as 10 times the noise level (S/N) at the baseline. For LC-MS-MS, the MDLs were determined as 3 times the standard deviation of the analyte concentrations in the blank samples (EPA, 2003). As a general rule for all analytes, the analytical reproducibility (inter-day comparison) was tested by repeated injections (duplicates) of standard compounds and glaucous gull samples at regular intervals of time. Method blank samples were run with each block of 5 samples to assess for background interferences. In cases where analyte concentrations in blanks indicated a background contribution above the minimum S/N ratio of 10, a blank-correction was performed on the samples using the mean analyte concentrations in blanks. The duplicate standard compounds and glaucous gull plasma and egg samples demonstrated 5% or less variation of analyte levels.

Chemical analyses were performed during fall 2003 at the Great Lakes Institute for Environmental Research (GLIER) (University of Windsor, Windsor, Canada), and were completed during summer 2004 at the National Water Research Institute (NWRI)

A suite of established and new organohalogen contaminants, and their metabolites was determined in plasma and egg homogenate samples of glaucous gulls based on published and widely cited analytical methods including the most recent modifications (Hansen et al., 2001; Helm et al., 2002; Covaci et al., 2003; Sandala et al., 2004). Briefly, the quantification of analytes was performed using a gas chromatograph equipped with a micro electron capture detector (GC- μ ECD), or a GC-mass spectrometer (GC-MS) in the negative chemical ionization (NCI) mode using a buffer gas in the electron impact source (EI) (GC-(ECNI)MS). The analysis of perfluorinated compounds was performed using a liquid chromatograph- tandem MS (LC-MS-MS).

Standard procedures were used to ensure quality assurance and control. The results for all analytes were within the laboratories accredited requirements

(Environment Canada, Burlington, Canada). These laboratories are certified as testing laboratories for PCBs and OC pesticides and by-products according to the requirements of the Canadian Environmental Analytical Laboratory (CAEAL) programme of the Canadian Standards Association, and are participants in the Northern Contaminant Program's (Indian and Northern Affairs of Canada, Ottawa, Canada) Quality Assurance Program.

Table 1. Means with standard errors (\pm SE) and ranges (Min–Max) of plasma fat content, head and bill length, and body mass for male and female glaucous gulls captured at Bear Island in 2002 and 2004.

| | Males (N = 57) | | Females (N = 52) | |
|-------------------------|------------------|---------------|------------------|---------------|
| | Mean \pm SE | Min – Max | Mean \pm SE | Min – Max |
| Plasma fat (%) | 1.43 \pm 0.03 | 1.11 – 1.97 | 1.38 \pm 0.03 | 1.04 – 1.85 |
| Head + bill length (mm) | 152 \pm 0.68 | 140 – 165 | 139 \pm 0.52 | 128 – 147 |
| Body mass (g) | 1,805 \pm 17.3 | 1,550 – 2,100 | 1,446 \pm 11.7 | 1,280 – 1,620 |

7. Results

The chemical analyses of glaucous gull plasma and egg homogenate samples revealed the presence of several major classes of established and new organic contaminants, and their metabolites. Concentrations of individual analytes and sums are summarized for plasma (*Table 2* (wet weight); *Appendix 1* (lipid weight)) and egg homogenate samples (*Table 3* (wet weight); *Appendix 2* (lipid weight)). Sexes are shown separately in *Table 2* and *Appendix 1* to stress the importance of sex-specific accumulation of compounds in plasma of glaucous gulls.

Table 2. Means ^a with standard errors (\pm SE) and ranges (Min–Max) of the sum (Σ) or individual concentrations (ng/g wet weight) of a suite of organohalogen contaminants in plasma samples of a nearly equal number of male and female glaucous gulls from Bear Island. The number of samples (N) with concentration above the method detection limit of the analyte is shown relatively to the number of samples analyzed. The number of congeners constituting the sums is specified in brackets. See appendix 3–10 for congener-specific concentrations (ng/g wet weight) in plasma of all birds, i.e. males and females combined.

| Plasma | | Males (N = 57) | | Females (N = 52) | |
|--|-----------|-----------------|-------------|------------------|-------------|
| | | Mean \pm SE | Min – Max | Mean \pm SE | Min – Max |
| Σ Polychlorinated biphenyl (Σ PCB) (41 congeners) ^b | N = 89/89 | 735 \pm 76.6 | 146 – 2,372 | 436 \pm 36.9 | 124 – 1,088 |
| Hexachlorobenzene (HCB) | N = 88/89 | 7.74 \pm 0.69 | 1.43 – 18.8 | 4.97 \pm 0.38 | 1.02 – 10.8 |
| Σ Hexachlorocyclohexane (Σ HCH) (α -, β -, γ -) | N = 89/89 | 1.58 \pm 0.16 | 0.30 – 6.63 | 1.01 \pm 0.10 | 0.35 – 2.60 |
| Octachlorostyrene (OCS) | N = 89/89 | 0.18 \pm 0.02 | 0.02 – 0.54 | 0.09 \pm 0.007 | 0.02 – 0.20 |
| Σ Chlordane (Σ CHL) (6 compounds) ^c | N = 89/89 | 39.4 \pm 3.56 | 9.38 – 121 | 28.2 \pm 2.18 | 8.17 – 64.8 |
| Σ Dichlorodiphenyldichloroethane (Σ DDT) (<i>p,p'</i> -DDT, -DDD, -DDE) | N = 89/89 | 218 \pm 14.4 | 67.4 – 486 | 141 \pm 8.98 | 45.5 – 386 |
| Σ mirex (Mirex, <i>photo</i> -Mirex) | N = 89/89 | 0.32 \pm 0.03 | 0.08 – 0.83 | 0.30 \pm 0.02 | 0.11 – 0.84 |
| Dieldrin | N = 89/89 | 2.99 \pm 0.44 | 0.34 – 14.6 | 2.43 \pm 0.25 | 0.31 – 7.67 |
| Hexachlorobutadiene (HCBd) | N = 0/20 | - | <0.001 | - | <0.001 |
| Hexabromocyclododecane (HBCDD) | N = 20/20 | 0.51 \pm 0.13 | 0.10 – 1.50 | 0.70 \pm 0.16 | 0.20 – 2.00 |
| Brominated biphenyl-101 (BB-101) | N = 31/89 | 0.22 \pm 0.04 | 0.09 – 0.76 | 0.19 \pm 0.03 | 0.09 – 0.56 |
| Σ Polybrominated diphenylether (Σ PBDE) (9 congeners incl. co-elution of BB-153 with BDE-154) | N = 89/89 | 22.4 \pm 2.34 | 7.35 – 75.7 | 15.8 \pm 1.71 | 2.70 – 52.6 |
| Σ Polychlorinated naphthalene (Σ PCN) (20 congeners) | N = 18/20 | 1.24 \pm 0.16 | 0.37 – 1.95 | 0.97 \pm 0.13 | 0.53 – 1.60 |
| Σ Coplanar PCB (CB-81, -77, -126, -169) | N = 19/20 | 1.38 \pm 0.17 | 0.79 – 2.25 | 1.84 \pm 0.32 | 0.65 – 4.21 |
| Σ Perfluorinated compound (Σ PFC) (12 compounds) | N = 20/20 | 222 \pm 36.5 | 101 – 427 | 198 \pm 29.2 | 97.2 – 411 |
| Σ Toxaphene (21 congeners) | N = 20/20 | 12.8 \pm 1.74 | 6.86 – 24.7 | 14.7 \pm 1.34 | 10.3 – 21.6 |
| Methylsulfone- <i>p,p'</i> -DDE (MeSO ₂ - <i>p,p'</i> -DDE) | N = 83/83 | 0.37 \pm 0.04 | 0.03 – 1.10 | 0.36 \pm 0.04 | 0.07 – 1.44 |
| Σ MeSO ₂ -PCB (17 congeners) | N = 83/83 | 5.09 \pm 0.53 | 1.22 – 14.5 | 5.40 \pm 0.66 | 1.18 – 16.3 |
| Pentachlorophenol (PCP) | N = 14/82 | 0.06 \pm 0.01 | 0.03 – 0.13 | 0.17 \pm 0.06 | 0.06 – 0.48 |
| 4-HO-Heptachlorostyrene (4-HO-HpCS) | N = 13/82 | 0.11 \pm 0.04 | 0.04 – 0.29 | 0.20 \pm 0.06 | 0.06 – 0.43 |
| Σ HO-PCB (12 congeners) | N = 82/82 | 7.07 \pm 0.77 | 1.44 – 22.4 | 4.74 \pm 0.45 | 0.68 – 12.0 |
| Σ HO-PBDE (14 congeners) | N = 27/27 | 0.70 \pm 0.16 | 0.21 – 2.12 | 0.95 \pm 0.27 | 0.29 – 3.76 |

^a Arithmetic means are reported for samples with analyte concentrations above the method detection limits only.

^b Σ PCB: sum of PCB congeners no. (IUPAC): 31, 28, 52, 49, 44, 42, 71/41/64, 74, 70/76/98, 66/95, 56/60, 101, 99, 97, 110, 151, 149, 118, 146, 153, 105, 141, 179, 163/138, 158, 129/178, 182/187, 183, 128, 174, 177, 171/202/156, 200, 172, 180, 170/190, 201, 203/196, 208/195, 194, and 206.

^c Σ CHL: sum of oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, *cis*-nonachlor, and heptachlor epoxide.

Table 3. Means ^a with standard errors (\pm SE) and ranges (Min–Max) of egg fat content and the sum (Σ) or individual concentrations (ng/g wet weight) of a suite of organohalogen contaminants in egg homogenate samples of glaucous gulls from Bear Island. The number of samples (N) with concentration above the method detection limit of the analyte is shown relatively to the number of samples analyzed. The number of congeners constituting the sums is specified in brackets.

| Egg homogenates | | Mean \pm SE | Min – Max |
|---|-----------|------------------|-------------|
| Egg homogenate fat (%) | N = 32 | 9.61 \pm 0.18 | 6.77 – 12.6 |
| Σ PCB (41 congeners) ^b | N = 32/32 | 1,151 \pm 72.5 | 515 – 2,162 |
| HCB | N = 32/32 | 20.1 \pm 1.12 | 10.6 – 36.2 |
| Σ HCH (α -, β -, γ -) | N = 32/32 | 4.69 \pm 0.58 | 1.16 – 20.1 |
| OCS | N = 32/32 | 0.42 \pm 0.05 | 0.13 – 1.33 |
| Σ CHL (6 compounds) ^c | N = 32/32 | 92.4 \pm 6.40 | 38.0 – 192 |
| Σ DDT (<i>p,p'</i> -DDT, -DDD, -DDE) | N = 32/32 | 343 \pm 15.7 | 180 – 586 |
| Σ Mirex (Mirex, <i>photo</i> -Mirex) | N = 32/32 | 2.03 \pm 0.13 | 0.68 – 4.18 |
| Dieldrin | N = 32/32 | 21.6 \pm 1.22 | 12.0 – 36.8 |
| HCBD | N = 0/10 | - | <0.001 |
| HBCDD | N = 10/10 | 13.3 \pm 6.42 | 2.00 – 70.0 |
| BB-101 | N = 20/32 | 0.26 \pm 0.03 | 0.06 – 0.54 |
| Σ PBDE (9 congeners incl. co-elution of BB-153 with BDE-154) | N = 32/32 | 52.9 \pm 3.84 | 23.7 – 104 |
| Σ PCN (20 congeners) | N = 10/10 | 1.58 \pm 0.20 | 0.23 – 2.54 |
| Σ Coplanar PCB (CB-81, -77, -126, -169) | N = 10/10 | 4.62 \pm 0.67 | 1.31 – 8.14 |
| Σ PFC (12 compounds) | - | Not analyzed | |
| Σ Toxaphene (21 congeners) | N = 10/10 | 92.6 \pm 13.8 | 16.0 – 159 |
| MeSO ₂ - <i>p,p'</i> -DDE | N = 32/32 | 2.84 \pm 0.37 | 0.57 – 9.05 |
| Σ MeSO ₂ -PCB (17 congeners) | N = 32/32 | 8.76 \pm 0.58 | 2.46 – 17.3 |
| PCP | N = 2/32 | 0.01 \pm 0.002 | 0.01 – 0.02 |
| 4-HO-HpCS | N = 3/32 | 0.04 \pm 0.003 | 0.04 – 0.05 |
| Σ HO-PCB (12 congeners) | N = 30/32 | 0.19 \pm 0.03 | 0.01 – 0.50 |
| Σ HO-PBDE (14 congeners) | N = 0/32 | - | <0.08 |

^a Arithmetic means are reported for samples with analyte concentrations above the method detection limits only.

^b Σ PCB: sum of PCB congeners no. (IUPAC): 31, 28, 52, 49, 44, 42, 71/41/64, 74, 70/76/98, 66/95, 56/60, 101, 99, 97, 110, 151, 149, 118, 146, 153, 105, 141, 179, 163/138, 158, 129/178, 182/187, 183, 128, 174, 177, 171/202/156, 200, 172, 180, 170/190, 201, 203/196, 208/195, 194, and 206.

^c Σ CHL: sum of oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, *cis*-nonachlor, and heptachlor epoxide.

8. Discussion

8.1. Organochlorine Compounds

8.1.1. Established Contaminants and HCBD

Plasma levels of established organochlorine (OC) contaminants, i.e. PCBs, HCB, HCHs (lindane), OCS, chlordane-related compounds (CHLs), DDTs, Mirex, and dieldrin, were comparable to levels previously reported for glaucous gull blood and egg homogenate samples (e.g. Bustnes et al., 2000, 2001, 2002; Verreault et al., 2004; Agudo, 2004). Studies of established OCs in blood samples of glaucous gulls from Bear Island have shown that levels in females were generally lower than those in males, which was consistent with the present study and several studies of e.g. Arctic seabirds (AMAP, 2004). This difference in OC concentrations between sexes was explained by the process where females excrete part of their contaminant load through egg formation.

Lipid weight-based concentrations of e.g. Σ PCB was approximately 3.5-fold higher in plasma samples relatively to egg homogenate samples (*Appendix 1* and *2*).

HCBD was not detected at the established method detection limit in any of the plasma and egg homogenate samples of glaucous gulls (*Table 2* and *3*). Measurements of HCBD in Arctic biota and in the North Atlantic Ocean were reported to be very low (AMAP, 2004). HCBD appears thus not to be a chemical of importance in glaucous gulls from Bear Island and in Arctic biota in general.

8.1.2. Coplanar PCBs

Plasma concentrations of Σ coplanar PCB (sum of the non-ortho Cl-substituted congeners CB-77, -81, -126, and -169) ranged between 0.65 to 4.21 ng/g ww (42.7 to 309 ng/g lw) (*Table 2* and *Appendix 1*). Σ coplanar PCB represented only 0.2–0.4% of Σ PCB in glaucous gull plasma samples. The contribution of individual coplanar PCB congeners to Σ coplanar PCB was approximately equal, although the proportion of CB-126 was slightly higher (*Appendix 3*). Reports of coplanar PCB levels for Arctic seabirds are scarce, and only few measurements have been undertaken in e.g. liver. Nonetheless, lipid-normalized concentrations in liver of lower-trophic level seabirds (kittiwake, fulmar, and guillemot) from the Canadian Arctic were considerably lower, ranging between 5.4 to 32.1 ng/g lw (Braune and Simon, 2003), relatively to those reported in the present study. However, results should be taken with caution considering the fact that different analytical matrices (liver vs plasma) were used. Similarly, lipid-normalized concentrations of coplanar PCB in liver samples of little auks, guillemots, and kittiwakes from Svalbard area (range 0.4 to 4.0 ng/g lw) (Borgå et al., in press), were also considerably lower. In general, similar trends in Arctic seabirds are observed for concentrations of other PCB congeners with respect to their geographic variations between locations in the Arctic and the species trophic position in the food web. Low trophic-level species (e.g. fish) tend to accumulate lower concentrations of lipophilic OC contaminants compared to species at higher trophic levels (e.g. glaucous gull) (Borgå et al., 2001).

Levels of Σ coplanar PCB in egg homogenate samples in the present study were higher compared to those of eggs of kittiwakes, fulmars, and guillemots from the Canadian Arctic (Braune and Simon, 2003). Levels of Σ coplanar PCB in eggs of glaucous gulls were consistent with levels of other PCB congeners that are generally higher in Svalbard/Bear Island compared to those of eggs from the Canadian Arctic (AMAP, 2004). Coplanar PCBs have been reported to have an significant contribution to total TCDD equivalents (TEQs) in Canadian Arctic seabirds (Braune and Simon, 2003).

8.1.3. PCNs

Only a few PCN measurements exist for Arctic biota, most of which have been determined for marine mammals (Helm et al., 2002; Muir et al., 2004), and more recently in seabirds and eggs of seabirds (Braune and Simon, 2004; Muir et al., 2004; Helm et al., in prep). The concentrations of Σ PCN (sum of 20 PCN congeners) in present glaucous gull plasma samples ranged between 0.37 to 1.95 ng/g wet weight (25.2 to 126 ng/g lw) (*Table 2* and *Appendix 1*), and the congener pattern of PCNs in plasma and eggs consisted uniquely of 5 detectable congeners including co-elutions (CN-52/60, -66/67, -71/72, -63, and -73) (plasma: *Appendix 4*). An assessment of new chemicals in liver and eggs of Canadian Arctic seabirds (kittiwake, fulmar, and two guillemot species) did not detect PCN congeners in any of the samples (Braune and Simon, 2004). Being cautious due to different analytical matrices used, lipid-normalized plasma levels of Σ PCN in glaucous gulls of the present study were significantly higher compared to those of liver of glaucous gulls (range 4.1–17.1 ng/g lw) from the Northwater Polynya in the Canadian Arctic (Helm et al., in prep). However, the present levels in glaucous gull plasma samples were generally lower compared to levels measured in liver of aquatic and predatory birds from the Baltic Sea (Falandysz, 1998). Although levels of PCNs in glaucous gull samples were relatively low comparatively to most OCs, they may exert high toxicological potential due to their dioxin-like toxicity (AhR mediated) (e.g. Villeneuve et al., 2000).

Lipid weight-based levels of Σ PCN in egg homogenate samples of glaucous gulls (mean 49.0 ng/g lw) (*Appendix 2*) of the present study were 35-fold compared to those reported in eggs of northern fulmars (mean 1.4 ng/g lw) from the Canadian Arctic (Muir et al., 2004).

8.1.4. Toxaphenes

Σ Toxaphene (sum of 21 toxaphene congeners) was fourth in importance among all OCs determined in glaucous gull plasma samples after Σ CHL (sum of 6 chlordane-related compounds) (*Table 2*). The dominating toxaphene congener in plasma samples among the 16 detected congeners was B8-789 (*Appendix 5*). Concentrations of 10 toxaphene congeners were previously determined in glaucous gull liver from Bear Island in 1999 (Herzke et al., 2003). In the Herzke et al. (2003) study, only 4 toxaphene congeners were detected in the liver samples. The sum of 3 of those congeners (lipid-normalized levels) in plasma samples of the present study were roughly 2.5 times lower than reported in the 1999 study of glaucous gull liver samples. However, results should be taken with caution considering the fact that different analytical matrices (liver vs plasma) were used. Concentrations of Σ toxaphene (sum of 3 congeners) in egg samples of great black-backed and herring gulls from colonies along the Norwegian coast (AMAP, 2004) were in average 2 times higher than those reported in the present study of glaucous gull egg homogenate samples from Bear Island. The present Σ toxaphene concentrations in egg homogenate samples were comparable to those found in eggs (sum of 8 congeners) of white-tailed sea eagle from Norway in 1991–1997 (AMAP, 2004). Even though toxaphene has never been used in Norway, terrestrial and aquatic birds of Norway and Svalbard have been exposed to relatively high concentrations.

8.2. Organobromine Compounds

The congener profile of PBDEs was dominated by BDE-47 that constituted on average 40% of Σ PBDE (sum of 9 congeners including co-elution of BB-153 with BDE-154) (*Appendix 6*). This observation was consistent with previous studies on liver and muscle samples of aquatic and terrestrial birds, as well as on liver samples of glaucous gulls from Bear Island (Herzke et al., 2003; Law et al., 2003). Levels of individual PBDE congeners in the present study were generally lower than those reported for bird populations from southern

regions of Europe and North America (reviewed by Law et al., 2003). In a study of glaucous gulls from Bear Island, BDE-47 and BDE-99 were detected in 15 liver samples, and the concentrations ranged between not-detected to 424 ng/g lipid weight (Herzke et al., 2003). In the present study, the range (14.7–2,037 ng/g lw) for lipid-normalized plasma concentrations of these two compounds was significantly higher.

Interestingly, BDE-209 was detectable in 30% of the present glaucous gull plasma samples. For samples where concentrations of BDE-209 were above the method detection limits, BDE-209 was the second PBDE congener in importance regarding to its contribution to Σ PBDE. However, results reported for BDE-209 in the present study and elsewhere should be interpreted with caution as this compound is shown to be unstable during instrument quantification, resulting therefore to higher method detection limits (2.73 ng/g ww in the present study (GC-(ECNI)MS)). The present lipid-normalized plasma concentrations of BDE-209 (range 202–1,055 ng/g lw) in glaucous gulls were generally comparable to those found in liver samples of terrestrial predatory birds of UK (UK Environment Agency, 2004). Similarly, plasma concentrations of BDE-209 (*Appendix 6*) in the present study were comparable to those reported previously for 20 liver samples of glaucous gulls collected in 2001 from Svalbard (range: not detected–10.0 ng/g ww; no lipid content available) (Savinova T; unpublished data). Again, results should be taken with caution considering the fact that different analytical matrices and methodology were used.

Lipid-normalized and wet weight-based concentrations of Σ PBDE in glaucous gull egg homogenate samples were significantly lower than those reported for herring gull eggs from the Great Lakes in Canada/USA (Norstrom et al., 2002). This suggests that North American- and local-sourced inputs are a major factor for PBDE exposure in herring gulls. For example, egg homogenates from a herring gull colony of this region, where the lowest mean Σ PBDE was reported, had 65% higher lipid-normalized mean Σ PBDE compared to glaucous gull egg homogenate samples of the present study. Likewise, Σ PBDE (lipid weight) reported in the present study was considerably lower than Σ PBDE measured for eggs of peregrine falcons from Sweden (Law et al., 2003). BDE-209 levels in the present study were below the method detection limit for 85% of the egg homogenate samples. Detectable levels were thus determined for 4 egg homogenate samples (range 23.2–52.5 ng/g lw). Given the difficulty of comparing levels between studies due to different methodologies (e.g. established method detection limit), the lipid-normalized concentrations of BDE-209 reported in the present study in eggs were nonetheless within the lowest range reported for peregrine falcon eggs from Sweden (28–430 ng/g lw) and UK (1.8–828 ng/g lw) (Law et al., 2003; UK Environment Agency, 2004). Considering the male/female ratio studied and the fact that fewer egg homogenate samples had detectable levels of BDE-209, we suggest that this compound is less mobilized relatively to lower-brominated PBDE congeners in the bird mother during egg formation. This observation is consistent with results for PCB congeners in eggs where the degree of chlorination has a strong effect on the transfer rate of congeners from mother to eggs.

Based on the results of the present study of plasma samples, we suggest that BDE-209 may have a bioaccumulation potential in glaucous gulls breeding at Bear Island and possibly in the Barents Sea food web. As the glaucous gulls breeding in the Barents Sea winter in the northern part of the Atlantic Ocean (Mehlum and Bakken, 2000; Gilchrist, 2001), the likelihood of PBDE/BDE-209 exposure of glaucous gulls via local-sourced inputs is unlikely. To present, few investigations dealing with the toxicokinetic and bioaccumulation of BDE-209 in biota have been carried out.

Two compounds of PBBs were determined in glaucous gull plasma and egg homogenate samples: BB-101 (pentabrominated) and BB-153 (hexabrominated) (co-eluting with BDE-154). Tetra- to hexabrominated biphenyl compounds in biota were previously

reported to be representative of the PBB mixtures used in e.g. USA (Law et al., 2003). Concentrations of BB-101 in the 35% of plasma samples where this compound was detected, were in the lower range of those of PBDE congeners. BB-101 was detected in 63% of egg homogenate samples. HBCDD was detected in all plasma and egg homogenate samples of this present study of glaucous gulls. Lipid-normalized concentrations of HBCDD in plasma (range: 6.13–122 ng/g lw) (*Appendix 1*) of glaucous gulls were 5–17 times lower than those of Swedish peregrine falcons (Linberg et al., 2004). In contrast to glaucous gulls from Bear Island, which migrate entirely across regions of the Arctic, the Swedish peregrine falcons overwinter along the coast and estuaries of central and southern Europe. It is suggested that exposure to HBCDD (and other BFRs) in predatory birds is closely linked to their migratory routes and overwintering sites (AMAP, 2004). However, the low concentrations of PBBs and HBCDD in plasma and egg homogenate samples show that these chemicals are of low importance compared to PBDEs in glaucous gulls from Bear Island.

8.3. Perfluorinated Compounds

A total of 8 PFCs were detected in glaucous gull plasma samples. The Σ PFC (sum of 12 PFC compounds), largely dominated by the compound PFOS (70% of Σ PFC), was generally comparable to levels of Σ DDT (sum of *p,p'*-DDT, -DDD, and -DDE) for the same plasma samples (*Table 2* and *Appendix 7*). PFOS was recently determined in 15 Svalbard polar bear plasma samples at a mean concentration of 97 ng/g wet weight (Gabrielsen et al., 2004), which was approximately 50% lower than those found in plasma samples of glaucous gulls of the present study. PFUnA, the second PFC in importance in glaucous gull plasma samples (24% of Σ PFC), was also among the top 3 compounds in polar bear liver samples from circumpolar locations (Smithwick et al., submitted). Levels of PFOS in plasma of glaucous gulls from the present study were comparable or lower than those reported for aquatic birds from the Great Lakes in Canada/USA (Geisy and Kannan, 2001). According to our findings, PFOS appears to be a chemical of relatively high importance in glaucous gull plasma.

8.4. Methylsulfonyl and Hydroxylated Metabolites

The methylsulfonyl congener profile of glaucous gull plasma samples was dominated consistently by the congeners 3'-MeSO₂-CB-132, 4-MeSO₂-CB-64, and an unknown MeSO₂-6Cl-PCB (*Appendix 8*). Σ MeSO₂-PCB (sum of 17 congeners) in glaucous gull plasma samples was approximately 134 and 81 times lower than Σ PCB for females and males, respectively. Furthermore, the mean ratio Σ MeSO₂-PCB to Σ PCB (0.01) was significantly lower in glaucous gulls relatively to e.g. polar bears (plasma levels), and other marine mammal species (Letcher et al., 2000; Sandala et al., 2004). The polar bear is a marine species known to have an enhanced metabolizing capability towards PCBs compared to other mammals (AMAP, 2004). The present ratio Σ MeSO₂-PCB/ Σ PCB for glaucous gulls was generally comparable to free-living aquatic and terrestrial bird species (reviewed by Letcher et al., 2000).

The major compounds making up Σ HO-PCB (sum of 14 congeners) in plasma were 4-HO-CB-187 (50% of Σ HO-PCB) and 4-HO-CB-146 (20% of Σ HO-PCB), the first being the most abundant organohalogen metabolite quantified in glaucous gull plasma samples (*Appendix 9*). The congener HO-CB-187, a confirmed metabolite of CB-187, was also reported to be the dominating HO-PCB congener in blood of polar bears, white-tailed sea eagles, and albatross (Klasson-Wehler et al., 1998; Letcher et al., 2000; Olsson et al., 2000; Sandala et al., 2004). A study of whole blood of white-tailed sea eagle from Sweden reported a mean concentration for 4-HO-CB-187 that was 13% lower than the present levels in glaucous gull plasma samples (Olsson et al., 2000). Low levels of PCP and 4-HO-HpCS were found in glaucous gull plasma samples (*Table 2*).

Only 6 of 14 HO-PBDE congeners analyzed were detected in plasma samples of glaucous gulls. The dominating congener 6-HO-BDE-47, which is a suggested metabolite of BDE-47, was the only congener detected in all plasma samples (*Appendix 10*). This observation was consistent with an investigation of HO-PBDEs in blood of Detroit River fish (Valters K; unpublished data). Among the 6 HO-PBDE congeners detected in plasma of glaucous gulls, 4 are suggested metabolites of 2 PBDE congeners: BDE-47 and BDE-49. HO-PBDEs were not detected in any of the egg homogenate samples of glaucous gulls (*Table 3*).

Most HO-PCB and MeSO₂-PCB/-*p,p'*-DDE congeners were found in egg homogenate samples of glaucous gulls, although few samples had concentrations above the established method detection limits (*Table 3*). A recent study has also demonstrated the occurrence of HO-PCBs and other phenolics in eggs of Norwegian predatory bird species (Berger et al., 2004). The presence and accumulation of these metabolites in eggs indicates the *in ovo* transfer of these compounds from mother to eggs during the process of egg formation. This may have a significant implication for the developing embryo as certain metabolites, e.g. HO-PCBs, have been shown to interfere with the endocrine systems of exposed organisms, and especially with the transport of hormones through plasma carrier proteins (i.e. transthyretin, albumin, TBG) (Sandau, 2000).

9. Toxicological Implications of New Contaminants and Metabolites

In the present study, novel classes of organohalogen contaminants of concerns have been detected in glaucous gull plasma and egg homogenates samples. A number of these compound groups, e.g., PCNs, BFRs, and PFCs, have already demonstrated toxicological potential in laboratory and free-living animals (AMAP, 2004). PCNs are planar molecules, likewise to coplanar PCBs, and exert their effects via the aryl hydrocarbon (Ah) receptor. Exposure to PCNs has led to effects similar to those seen for the highly toxic PCDDs/PCDFs and coplanar PCBs (e.g. Villeneuve et al., 2000). Exposure to PBDEs, including BDE-209, has been found to induce many biological effects similar to PCBs and OC pesticides (AMAP, 2004). A number of studies are ongoing to assess the toxicological effects of PFCs in biota. Yet, the few investigations performed on animals have raised concerns about the PFC congener PFOS potential developmental, reproductive, and systemic toxicity.

Our study has demonstrated that PCB and PBDE metabolism is a source for generation of unique classes of persistent organohalogens in glaucous gulls. However, the capacity for metabolism, and/or formation/retention of PCB and PBDE metabolites (methylsulfonyl and hydroxylated) in glaucous gulls is low relatively to, e.g., mammalian species such as the polar bear or some cetacean species. Nonetheless, a wide range of these metabolites are retained in plasma of glaucous gulls. Current levels of methylsulfonyl PCBs and hydroxylated PCBs/PBDEs in plasma of glaucous gulls may have a significant toxicological potential. Deleterious biological effects reported in several species have been associated to induction by e.g. HO-PCB, as for example through interference with the thyroid hormone T4 transport proteins (Sandau, 2000). We have demonstrated in a past study that levels of PCBs and OC pesticides were correlated negatively with circulating levels of thyroid hormones in male glaucous gulls (Verreault et al., 2004).

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11. Appendix 1–10

Appendix 1. Means ^a with standard errors (\pm SE) and ranges (Min–Max) of the sum (Σ) or individual concentrations (ng/g lipid weight) of a suite of organohalogen contaminants in plasma samples of a nearly equal number of male and female glaucous gulls from Bear Island. The number of samples (N) with concentration above the method detection limit of the analyte is shown relatively to the number of samples analyzed. The number of congeners constituting the sums is specified in brackets.

| Plasma | | Males (N = 57) | | Females (N = 52) | |
|---|-----------|---------------------------|------------------|--------------------|----------------|
| | | Mean \pm SE | Min – Max | Mean \pm SE | Min – Max |
| Σ PCB (41 congeners) ^b | N = 89/89 | 52,191 \pm 5,450 | 10,633 – 160,169 | 31,367 \pm 2,487 | 9,557 – 80,886 |
| HCB | N = 88/89 | 556 \pm 52.9 | 91.8 – 1,374 | 364 \pm 29.2 | 76.4 – 908 |
| Σ HCH (α -, β -, γ -) | N = 89/89 | 112 \pm 11.7 | 21.6 – 488 | 73.1 \pm 6.74 | 26.0 – 194 |
| OCS | N = 89/89 | 13.2 \pm 1.23 | 1.73 – 36.2 | 6.74 \pm 0.56 | 1.73 – 16.9 |
| Σ CHL (6 compounds) ^c | N = 89/89 | 2,809 \pm 265 | 657 – 8,918 | 2,068 \pm 166 | 628 – 5,618 |
| Σ DDT (<i>p,p'</i> -DDT, -DDD, -DDE) | N = 89/89 | 15,531 \pm 1,060 | 4,915 – 32,075 | 10,250 \pm 606 | 3,502 – 25,941 |
| Σ Mirex (Mirex, <i>photo</i> -Mirex) | N = 89/89 | 23.0 \pm 1.88 | 5.44 – 61.2 | 21.9 \pm 1.57 | 7.57 – 48.1 |
| Dieldrin | N = 89/89 | 208 \pm 29.6 | 26.2 – 1,024 | 178 \pm 17.9 | 18.5 – 549 |
| HCBD | N = 0/20 | - | <0.07 | - | <0.07 |
| HBCDD | N = 20/20 | 36.8 \pm 9.55 | 6.13 – 108 | 52.2 \pm 9.0 | 19.3 – 122 |
| BB-101 | N = 31/89 | 16.2 \pm 3.08 | 7.24 – 56.2 | 13.2 \pm 1.92 | 6.80 – 30.3 |
| Σ PBDE (9 congeners incl. co-elution of BB-153 with BDE-154) | N = 89/89 | 1,596 \pm 167 | 461 – 5,316 | 1,138 \pm 114 | 213 – 3,706 |
| Σ PCN (20 congeners) | N = 18/20 | 82.0 \pm 10.9 | 27.4 – 126 | 69.7 \pm 11.7 | 25.2 – 119 |
| Σ Coplanar PCB (CB-81, -77, -126, -169) | N = 19/20 | 92.4 \pm 12.2 | 51.8 – 154 | 121 \pm 22.7 | 42.7 – 309 |
| Σ PFC (12 compounds) | - | No report in lipid weight | | | |
| Σ Toxaphene (21 congeners) | N = 20/20 | 925 \pm 107 | 447 – 1,530 | 1,165 \pm 70.7 | 941 – 1,664 |
| MeSO ₂ - <i>p,p'</i> -DDE | N = 83/83 | 27.0 \pm 3.20 | 2.11 – 94.4 | 27.0 \pm 3.49 | 4.53 – 124 |
| Σ MeSO ₂ -PCB (17 congeners) | N = 83/83 | 367 \pm 40.6 | 67.6 – 1,112 | 396 \pm 48.2 | 66.4 – 1,192 |
| PCP | - | No report in lipid weight | | | |
| 4-HO-HpCS | - | No report in lipid weight | | | |
| Σ HO-PCB (12 congeners) | - | No report in lipid weight | | | |
| Σ HO-PBDE (14 congeners) | - | No report in lipid weight | | | |

^a Arithmetic means are reported for samples with analyte concentrations above the method detection limits only.

^b Σ PCB: sum of PCB congeners no. (IUPAC): 31, 28, 52, 49, 44, 42, 71/41/64, 74, 70/76/98, 66/95, 56/60, 101, 99, 97, 110, 151, 149, 118, 146, 153, 105, 141, 179, 163/138, 158, 129/178, 182/187, 183, 128, 174, 177, 171/202/156, 200, 172, 180, 170/190, 201, 203/196, 208/195, 194, and 206.

^c Σ CHL: sum of oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, *cis*-nonachlor, and heptachlor epoxide.

Appendix 2. Means^a with standard errors (\pm SE) and ranges (Min–Max) of egg fat content and the sum (Σ) or individual concentrations (ng/g lipid weight) of a suite of organohalogen contaminants in egg homogenate samples of glaucous gulls from Bear Island. The number of samples (N) with concentration above the method detection limit of the analyte is shown relatively to the number of samples analyzed. The number of congeners constituting the sums is specified in brackets.

| Egg homogenates | | Mean \pm SE | Min – Max |
|---|-----------|---------------------------|----------------|
| Egg homogenate fat (%) | N = 32 | 9.61 \pm 0.18 | 6.77 – 12.6 |
| Σ PCB (41 congeners) ^b | N = 32/32 | 12,051 \pm 777 | 5,383 – 25,743 |
| HCB | N = 32/32 | 211 \pm 12.6 | 103 – 430 |
| Σ HCH (α -, β -, γ -) | N = 32/32 | 49.1 \pm 6.0 | 11.2 – 204 |
| OCS | N = 32/32 | 4.36 \pm 0.54 | 1.27 – 14.1 |
| Σ CHL (6 compounds) ^c | N = 32/32 | 971 \pm 68.7 | 402 – 2,038 |
| Σ DDT (<i>p,p'</i> -DDT, -DDD, -DDE) | N = 32/32 | 3600 \pm 172 | 1,909 – 6,226 |
| Σ Mirex (Mirex, <i>photo</i> -Mirex) | N = 32/32 | 21.2 \pm 1.34 | 6.91 – 42.4 |
| Dieldrin | N = 32/32 | 227 \pm 13.8 | 122 – 402 |
| HCBD | N = 0/10 | - | <0.07 |
| HBCDD | N = 10/10 | 142 \pm 71.5 | 20.3 – 774 |
| BB-101 | N = 20/32 | 2.76 \pm 0.37 | 0.59 – 5.95 |
| Σ PBDE (9 congeners incl. co-elution of BB-153 with BDE-154) | N = 32/32 | 549 \pm 38.7 | 248 – 1,137 |
| Σ PCN (20 congeners) | N = 10/10 | 49.0 \pm 20.0 | 1.82 – 162 |
| Σ Coplanar PCB (CB-81, -77, -126, -169) | N = 10/10 | 149 \pm 66.2 | 10.6 – 573 |
| Σ PFC (12 compounds) | - | Not analyzed | |
| Σ Toxaphene (21 congeners) | N = 10/10 | 963 \pm 154 | 162 – 1,762 |
| MeSO ₂ - <i>p,p'</i> -DDE | N = 32/32 | 29.6 \pm 3.75 | 5.86 – 89.8 |
| Σ MeSO ₂ -PCB (17 congeners) | N = 32/32 | 91.0 \pm 5.50 | 30.7 – 155 |
| PCP | - | No report in lipid weight | |
| 4-HO-HpCS | - | No report in lipid weight | |
| Σ HO-PCB (12 congeners) | - | No report in lipid weight | |
| Σ HO-PBDE (14 congeners) | - | No report in lipid weight | |

^a Arithmetic means are reported for samples with analyte concentrations above the method detection limits only.

^b Σ PCB: sum of PCB congeners no. (IUPAC): 31, 28, 52, 49, 44, 42, 71/41/64, 74, 70/76/98, 66/95, 56/60, 101, 99, 97, 110, 151, 149, 118, 146, 153, 105, 141, 179, 163/138, 158, 129/178, 182/187, 183, 128, 174, 177, 171/202/156, 200, 172, 180, 170/190, 201, 203/196, 208/195, 194, and 206.

^c Σ CHL: sum of oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, *cis*-nonachlor, and heptachlor epoxide.

Appendix 3–10

The means (reported for samples with analyte concentrations above the method detection limits only) are reported with standard errors (\pm SE) and ranges (Min–Max) of the individual concentrations (ng/g wet weight) of a suite of organohalogen contaminants in all plasma samples (male and female glaucous gulls combined). The number of samples (N) with concentration above the method detection limit (MDL) of the analyte is shown relatively to the number of samples analyzed. MDLs (<MDL) are reported when the analyte was not detected in any of the plasma samples.

Appendix 3. Coplanar PCBs

| | | Plasma samples | |
|--------|-----------|-----------------|-------------|
| | | Mean \pm SE | Min – Max |
| CB-81 | N = 18/20 | 0.37 \pm 0.06 | 0.12 – 0.90 |
| CB-77 | N = 17/20 | 0.34 \pm 0.05 | 0.14 – 0.82 |
| CB-126 | N = 18/20 | 0.61 \pm 0.11 | 0.19 – 2.03 |
| CB-169 | N = 18/20 | 0.42 \pm 0.08 | 0.11 – 1.08 |

Appendix 4. Polychlorinated naphthalenes (PCNs)

| | | Plasma samples | |
|-------------|-----------|-----------------|-------------|
| | | Mean \pm SE | Min – Max |
| CN-33/34/37 | N = 0/20 | - | <0.03 |
| CN-47 | N = 0/20 | - | <0.02 |
| CN-28/43 | N = 0/20 | - | <0.03 |
| CN-32 | N = 0/20 | - | <0.01 |
| CN-35 | N = 0/20 | - | <0.02 |
| CN-52/60 | N = 12/20 | 0.54 \pm 0.08 | 0.21 – 1.10 |
| CN-58 | N = 0/20 | - | <0.02 |
| CN-61 | N = 0/20 | - | <0.02 |
| CN-57 | N = 0/20 | - | <0.01 |
| CN-62 | N = 0/20 | - | <0.02 |
| CN-53 | N = 0/20 | - | <0.02 |
| CN-59 | N = 0/20 | - | <0.02 |
| CN-66/67 | N = 18/20 | 0.42 \pm 0.08 | 0.14 – 1.25 |
| CN-64/68 | N = 0/20 | - | <0.02 |
| CN-69 | N = 0/20 | - | <0.02 |
| CN-71/72 | N = 10/20 | 0.26 \pm 0.04 | 0.16 – 0.58 |
| CN-63 | N = 10/20 | 0.31 \pm 0.05 | 0.15 – 0.68 |
| CN-65 | N = 0/20 | - | <0.02 |
| CN-73 | N = 1/20 | - | 0.20 |
| CN-74 | N = 0/20 | - | <0.03 |

Appendix 5. Toxaphenes

| | | Plasma samples | |
|-----------------|-----------|----------------|--------------|
| | | Mean ± SE | Min – Max |
| B6-923a | N = 0/20 | - | <0.001 |
| B7-1001 | N = 20/20 | 0.05 ± 0.007 | 0.02 – 0.13 |
| B8-1413 | N = 20/20 | 0.74 ± 0.09 | 0.12 – 1.82 |
| B8-1412 | N = 20/20 | 1.87 ± 0.13 | 0.95 – 2.92 |
| B7-1450 | N = 19/20 | 0.03 ± 0.006 | 0.003 – 0.09 |
| B7-515 | N = 0/20 | - | <0.001 |
| B7-1474/B7-1440 | N = 19/20 | 0.01 ± 0.002 | 0.002 – 0.03 |
| B8-789 | N = 20/20 | 4.99 ± 0.56 | 1.35 – 9.94 |
| B7-1059a | N = 1/20 | - | 0.008 |
| B8-531 | N = 17/20 | 0.02 ± 0.004 | 0.004 – 0.06 |
| B8-1414/B8-1945 | N = 20/20 | 0.42 ± 0.04 | 0.14 – 0.78 |
| B8-806 | N = 20/20 | 0.08 ± 0.009 | 0.03 – 0.15 |
| B8-2229 | N = 20/20 | 1.96 ± 0.18 | 0.93 – 4.33 |
| B8-810 | N = 20/20 | 0.37 ± 0.04 | 0.13 – 0.65 |
| B9-1679 | N = 20/20 | 1.80 ± 0.16 | 0.62 – 3.34 |
| B9-718 | N = 0/20 | - | <0.001 |
| B8-1471 | N = 20/20 | 0.93 ± 0.08 | 0.43 – 2.08 |
| B9-743/B9-2006 | N = 0/20 | - | <0.001 |
| B9-1046 | N = 0/20 | - | <0.001 |
| B9-715 | N = 20/20 | 0.13 ± 0.01 | 0.05 – 0.26 |
| B9-1025 | N = 20/20 | 0.36 ± 0.03 | 0.17 – 0.67 |

Appendix 6. Polybrominated diphenylethers (PBDEs)

| | | Plasma samples | |
|----------------|-----------|----------------|-------------|
| | | Mean ± SE | Min – Max |
| BDE-28 | N = 23/89 | 0.10 ± 0.01 | 0.01 – 0.23 |
| BDE-47 | N = 89/89 | 8.54 ± 0.57 | 1.87 – 28.0 |
| BDE-100 | N = 87/89 | 1.18 ± 0.08 | 0.38 – 3.43 |
| BDE-99 | N = 89/89 | 2.53 ± 0.24 | 0.19 – 10.9 |
| BDE-154/BB-153 | N = 89/89 | 1.69 ± 0.19 | 0.05 – 8.86 |
| BDE-153 | N = 89/89 | 3.19 ± 0.41 | 0.05 – 21.2 |
| BDE-138 | N = 33/89 | 0.15 ± 0.03 | 0.02 – 1.12 |
| BDE-183 | N = 58/89 | 0.32 ± 0.14 | 0.03 – 8.34 |
| BDE-209 | N = 28/89 | 5.71 ± 0.57 | 2.76 – 14.7 |

Appendix 7. Perfluorinated compounds (PFCs)

| | | Plasma samples | |
|--|-----------|----------------|-------------|
| | | Mean ± SE | Min – Max |
| Perfluorooctane sulfonate (PFOS) | N = 20/20 | 148 ± 17.1 | 57.9 – 328 |
| Perfluorononanoic acid (PFNA) | N = 20/20 | 3.27 ± 0.37 | 1.37 – 6.94 |
| Perfluorooctanoic acid (PFOA) | N = 17/20 | 0.60 ± 0.11 | 0.12 – 1.68 |
| Perfluoropentanoic acid (PFPA) | N = 4/20 | 0.38 ± 0.1 | 0.13 – 0.57 |
| Perfluorobutane sulfonic acid (PFBS) | N = 1/20 | - | 0.57 |
| Perfluorohexanoic acid (PFHxA) | N = 5/20 | 0.27 ± 0.06 | 0.15 – 0.43 |
| Perfluorohexane sulfonic acid (PFHxS) | N = 20/20 | 0.97 ± 0.12 | 0.19 – 2.49 |
| Perfluorodecanoic acid (PFDA) | N = 20/20 | 5.60 ± 0.70 | 3.08 – 14.0 |
| Perfluoroundecanoic acid (PFUnA) | N = 20/20 | 51.2 ± 6.23 | 24.5 – 147 |
| 1-hydroxyethane-2-perfluorooctanol (8:2 FTOH) | N = 0/20 | - | <0.26 |
| Perfluorooctane sulphonamide (PFOSA) | N = 0/20 | - | <0.30 |
| 1-hydroxyethane-2-perfluorodecanol (10:2 FTOH) | N = 0/20 | - | <0.26 |

Appendix 8. Methylsulfonyl PCBs (MeSO₂-PCBs)

| | | Plasma samples | |
|---|-----------|----------------|-------------|
| | | Mean ± SE | Min – Max |
| 3-MeSO ₂ -CB-52 | N = 3/83 | 0.26 ± 0.18 | 0.07 – 0.63 |
| 4-MeSO ₂ -CB-52 | N = 10/83 | 0.06 ± 0.006 | 0.04 – 0.09 |
| 3'-MeSO ₂ -CB-49 | N = 27/83 | 0.06 ± 0.006 | 0.03 – 0.17 |
| 4'-MeSO ₂ -CB-49 | N = 83/83 | 0.51 ± 0.11 | 0.03 – 7.26 |
| 4-MeSO ₂ -CB-64 | N = 6/83 | 2.53 ± 1.37 | 0.55 – 9.25 |
| 3-MeSO ₂ -CB-70 | N = 79/83 | 0.05 ± 0.006 | 0.01 – 0.33 |
| 4-MeSO ₂ -CB-70 | N = 32/83 | 0.10 ± 0.02 | 0.02 – 0.48 |
| 3'-MeSO ₂ -CB-101 | N = 33/83 | 0.16 ± 0.01 | 0.09 – 0.35 |
| 4'-MeSO ₂ -CB-101 | N = 80/83 | 0.19 ± 0.01 | 0.04 – 0.49 |
| 4-MeSO ₂ -CB-110/4'-MeSO ₂ -CB-87 | N = 82/83 | 0.16 ± 0.01 | 0.03 – 0.48 |
| 3-MeSO ₂ -CB-110 | N = 53/83 | 0.18 ± 0.02 | 0.04 – 0.96 |
| 3-MeSO ₂ -CB-149 | N = 83/83 | 0.27 ± 0.02 | 0.05 – 0.95 |
| 3'-MeSO ₂ -CB-132 | N = 74/83 | 1.45 ± 0.13 | 0.24 – 5.26 |
| 4'-MeSO ₂ -CB-132 | N = 76/83 | 0.25 ± 0.02 | 0.03 – 1.0 |
| 4-MeSO ₂ -CB-174 | N = 55/83 | 0.11 ± 0.01 | 0.03 – 0.56 |
| Unknown MeSO ₂ -6Cl-PCB | N = 83/83 | 2.04 ± 0.21 | 0.18 – 7.53 |

Appendix 9. Hydroxylated PCBs (HO-PCBs)

| | | Plasma samples | |
|--------------|-----------|----------------|-------------|
| | | Mean ± SE | Min – Max |
| 4'-HO-CB-104 | N = 19/82 | 0.24 ± 0.05 | 0.12 – 1.04 |
| 4'-HO-CB-120 | N = 82/82 | 0.57 ± 0.04 | 0.05 – 1.77 |
| 4-HO-CB-146 | N = 82/82 | 1.20 ± 0.09 | 0.16 – 4.47 |
| 4-HO-CB-107 | N = 55/82 | 0.10 ± 0.009 | 0.04 – 0.28 |
| 4-HO-CB-112 | N = 70/82 | 0.09 ± 0.008 | 0.02 – 0.39 |
| 4-HO-CB-165 | N = 40/82 | 0.96 ± 0.06 | 0.32 – 1.79 |
| 3'-HO-CB-85 | N = 49/82 | 0.12 ± 0.02 | 0.02 – 0.66 |
| 4-HO-CB-187 | N = 82/82 | 3.19 ± 0.37 | 0.29 – 17.5 |
| 4'-HO-CB-159 | N = 16/82 | 0.33 ± 0.11 | 0.04 – 1.70 |
| 3'-HO-CB-138 | N = 6/82 | 0.41 ± 0.09 | 0.18 – 0.71 |
| 4'-HO-CB-130 | N = 4/82 | 0.36 ± 0.29 | 0.06 – 1.23 |
| 3'-HO-CB-180 | N = 43/82 | 0.18 ± 0.02 | 0.05 – 0.73 |
| 4-HO-CB-193 | N = 20/82 | 0.11 ± 0.02 | 0.04 – 0.37 |

Appendix 10. Hydroxylated PBDEs (HO-PBDEs)

| | | Plasma samples | |
|--------------|-----------|----------------|-------------|
| | | Mean ± SE | Min – Max |
| 6'-HO-BDE-17 | N = 1/26 | - | 0.08 |
| 2'-HO-BDE-28 | N = 0/26 | - | <0.03 |
| 6'-HO-BDE-49 | N = 7/26 | 0.13 ± 0.03 | 0.06 – 0.27 |
| 2'-HO-BDE-68 | N = 0/26 | - | <0.05 |
| 6-HO-BDE-47 | N = 26/26 | 0.43 ± 0.07 | 0.09 – 1.59 |
| 3-HO-BDE-47 | N = 24/26 | 0.24 ± 0.05 | 0.09 – 1.29 |
| 5-HO-BDE-47 | N = 0/26 | - | <0.08 |
| 4'-HO-BDE-49 | N = 7/26 | 0.40 ± 0.18 | 0.06 – 1.22 |
| 4-HO-BDE-42 | N = 3/26 | 0.19 ± 0.05 | 0.14 – 0.29 |
| 6-HO-BDE-90 | N = 0/26 | - | <0.04 |
| 6-HO-BDE-99 | N = 2/26 | - | 0.09 – 0.62 |
| 2-HO-BDE-123 | N = 0/26 | - | <0.03 |
| 6-HO-BDE-85 | N = 0/26 | - | <0.05 |
| 6-HO-BDE-137 | N = 0/26 | - | <0.05 |



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| Forfattere Jonathan Verreault, Robert J. Letcher, Derek D.C. Muir, Shaogang Chu, Geir W. Gabrielsen |
| Tittel - norsk og engelsk Nye og etablerte organohalogene miljøgifter og deres metabolitter i plasma og egg prøver fra polarmåke på Bjørnøya New and established organohalogen contaminants and their metabolites in plasma and eggs of glaucous gulls from Bear Island |
| Summary The chemical analyses of glaucous gull plasma and egg homogenate samples revealed the presence of several major classes of established and new organohalogen contaminants of concerns, and their metabolites. Besides the routinely-analyzed organochlorines (OCs) (i.e. polychlorinated biphenyls (PCBs) and a suite of pesticides and by-products), less studied and new organohalogen contaminants such as coplanar PCBs, toxaphenes, polychlorinated naphthalenes (PCNs), brominated flame retardants (BFRs), and perfluorinated compounds (PFCs) were detected in plasma and egg homogenate samples. Hexachlorobutadiene (HCBd) was not detected at the established method detection limit in any of the plasma and egg homogenate samples of glaucous gulls. |

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|---|---|
| 4 emneord Nye miljøgifter, polarmåke, Bjørnøya | 4 subject words New contaminants, glaucous gull, Bear Island |
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