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- **Temporal trends of brominated flame retardants, cyclododeca-1,5,9-triene and mercury in eggs of four seabird species from Northern Norway and Svalbard**

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1. Preface

This project investigated the levels and temporal trends of brominated flame retardants (BFRs) and mercury in seabird eggs from Northern Norway and Svalbard. In this report, levels of polybrominated diphenyl ethers (PBDEs), hexabromocyclododecanes (HBCDDs), cyclododeca-1,5,9-triene (CDT) and mercury in eggs of herring gulls (*Larus argentatus*), Atlantic puffins (*Fratercula arctica*) and black-legged kittiwakes (*Rissa tridactyla*) from Røst and Hornøya (Northern Norway), in addition to glaucous gulls (*Larus hyperboreus*) from Bjørnøya (Svalbard) were compared to previously reported levels in eggs from seabirds. Furthermore, species-specific regional and temporal trends (1983-2003/1997-2002) of BFRs and mercury were examined.

The present study was a collaboration between the Norwegian Polar Institute, Tromsø University Museum, National Veterinary Institute, Norway and the Norwegian School of Veterinary Science. Funding was provided by the Norwegian Pollution Control Authority (SFT).

Norwegian Polar Institute, December 2005

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Front page photos

From left to right: herring gull (Rob Barrett, Tromsø University Museum), Atlantic puffin (Hallvard Strøm, Norwegian Polar Institute), black-legged kittiwake (Geir Wing Gabrielsen, Norwegian Polar Institute) and glaucous gull (Hallvard Strøm, Norwegian Polar Institute).

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3. Sammendrag

Formålet med denne studien var å kartlegge mulige forskjeller mellom arter og regioner, samt tidstrender av bromerte flammehemmere (BFR), cyclododeca-1,5,9-triene (CDT) og kvikksølv fra sjøfuglegg. Egg fra gråmåke (*Larus argentatus*), lundefugl (*Fratercula arctica*) og krykkje (*Rissa tridactyla*) ble samlet inn i 1983, 1993 og 2003 på Røst og Hornøya (Nord-Norge). Egg fra polarmåke (*Larus hyperboreus*) ble samlet inn i 1997 og 2002 på Bjørnøya (Svalbard). Nivåer og kongener/isomer-mønster ble sammenlignet med nivåer og mønster som er rapportert tidligere i fugl fra Europa og Nord-Amerika.

Polybromerte difenyletere

Med unntak av BDE-153, ble det ikke rapportert konsentrasjonsforskjeller av BDEer mellom regioner i denne studien. Nivåer av BDE-153 var signifikant høyere på Røst sammenlignet med Hornøya.

Konsentrasjoner av \sum BDE var høyere i gråmåkeegg og polarmåkeegg sammenlignet med lundefuglegg og krykkjeegg.

Gjennomsnittsverdier av BDEer i egg fra denne studien var lavere enn verdier rapportert i egg fra vandrefalk (*Falco peregrinus*) fra Sør-Grønland og Sverige, gråmåke fra Canada, og norske rovfugl.

BDE-47 utgjorde 34.6 til 77.4 % av \sum BDE. Konsentrasjonene av de ulike BDE kongenerne i dette studiet var avtagende i rekkefølgen BDE-47 > BDE-99 > BDE-100 > BDE-153 \approx BDE-154 \approx BDE-183 > BDE-28 > BDE-209.

Alle BDEer, med unntak av BDE-209, hadde økende konsentrasjoner fra 1983 til 1993. I tillegg var BDE-153, BDE-154 og BDE-183 også økende mellom 1983 til 2003. Det var ikke statistiske forskjeller i BDE-209 konsentrasjoner mellom år (miljøgifter tilstede versus ikke tilstede), selv om konsentrasjoner av BDE-209 generelt var høyere i 2003 enn i 1983 og 1993 både i egg fra gråmåke og krykkje. I lundefuglegg var konsentrasjoner av BDE-209 over deteksjonsgrensen i 1993, men verken funnet i 1983 eller 2003. Konsentrasjoner av BDEer fra 1997 og 2002 rapportert i polarmåkeegg fra Bjørnøya var tilnærmet like.

Hexabromocyclododecane (HBCDDs) og CDT

I denne studien var α -HBCDD isomeren over målbare grenseverdier i alle egg. β -HBCDD og γ -HBCDD isomeren derimot var under målbare grenseverdier i alle prøver.

Såvidt vi vet er dette studiet det første studiet med malinger på CDT i eggprøver. CDT nivåene var imidlertid under deteksjonsgrensen i alle prøver.

De høyeste nivåer av α -HBCDD ble målt i egg fra krykkje, nivåene avtok deretter i følgende rekkefølge gråmåke \approx polarmåke $>$ lundefugl. Som for BDEer, var det ingen geografiske trender av HBCDDer og CDT.

Total-HBCDD i denne studien var betydelig lavere enn tidligere rapporterte verdier i vandrefalkeegg fra Sverige og terneegg (*Sterna hirundo*) fra Belgia, men høyere enn konsentrasjoner i vandrefalkeegg fra Grønland og kirkeugleegg fra Belgia.

Nivåer av α -HBCDD var økende fra 1983 fram til 2003.

Kvikksølv

Som for BDEer og HBCDD, var det ingen geografiske trender av kvikksølv konsentrasjoner i datasettet. Polarmåkeegg hadde høyest konsentrasjoner av kvikksølv.

Videre var nivåer av kvikksølv i denne studien generelt lavere enn rapporterte nivåer i forskjellige fugleegg fra Nord-Amerika, som for eksempel, islom (*Gavia immer*), hvithodet havørn (*Haliaeetus leucocephalus*), prærieterne (*Sterna forsteri*), makrellterne (*Sterna hirundo*), skarv (*Phalacrocorax carbo*) og gråmåke. Nivåer av kvikksølv i denne studien var også lavere enn tidligere rapporterte nivåer i egg fra smålom (*Gavia stellata*) fra Sverige, men lik rapporterte nivåer i krykkjeegg fra Norge rapportert på 1970-tallet. Konsentrasjonene av kvikksølv i denne studien var lik rapporterte nivåer i egg fra skarv fra Lake Michigan, herodiashegre (*Ardea herodias*) og brudeand (*Aix sponsa*) fra nordøstlige deler av Nord-Amerika.

Ingen signifikante tidstrender av kvikksølv ble funnet i denne studien.

4. Summary

The aim of the present study was to investigate possible species and regional differences, in addition to temporal trends of brominated flame retardants (BFRs), cyclododeca-1,5,9-triene (CDT) and mercury in seabird eggs. Eggs were collected from herring gulls (*Larus argentatus*), Atlantic puffins (*Fratercula arctica*) and black-legged kittiwakes (*Rissa tridactyla*) in 1983, 1993 and 2003 at Røst and Hornøya (Northern Norway). Furthermore, eggs of glaucous gulls (*Larus hyperboreus*) were collected in 1997 and 2002 at Bjørnøya (Svalbard). Current levels and congener/isomer patterns were compared to previously reported levels in eggs of free-ranging seabirds.

Polybrominated diphenyl ethers

No spatial difference of BDEs was apparent in the dataset, with the exception of BDE-153. Levels of BDE-153 were significantly higher at Røst than those at Hornøya.

Σ BDE was higher in eggs of herring gulls and glaucous gulls than in eggs of Atlantic puffins and black-legged kittiwakes.

The summed mean concentrations of BDEs in the eggs from 2003 were lower compared to reported levels in eggs of peregrine falcons (*Falco peregrinus*) from South Greenland and Sweden, eggs of herring gulls from the Great Lakes and Norwegian predatory bird eggs.

For all seabird eggs, BDE-47 was the most abundant BDE congener, constituting from 34.6 to 77.4 % of Σ BDEs. Levels of the different BDE congeners in the present study decreased in the order BDE-47 > BDE-99 > BDE-100 > BDE-153 \approx BDE-154 \approx BDE-183 > BDE-28 > BDE-209.

For all BDEs, with the exception of BDE-183 and 209, concentrations increased from 1983 through 1993 in eggs of herring gulls, Atlantic puffins and black-legged kittiwakes. Between 1983 and 2003, only BDE-153, BDE-154 and BDE-183 levels increased. For BDE-209 no statistical significant difference (frequencies of detected versus non-detected) was found between sampling years, although concentrations of BDE-209 in eggs of herring gulls and black-legged kittiwakes tended to be higher in 2003 than in 1983 and 1993. In eggs of Atlantic puffins levels were above the detected limit only in 1993. Concentrations of BDEs in glaucous gull from Bjørnøya sampled in 1997 and 2002 were practically identical.

Hexabromocyclododecane (HBCDDs) and CDT

The isomer α -HBCDD was detected in all eggs of glaucous gulls, herring gulls, Atlantic puffins and black-legged kittiwakes. Levels of β -HBCDD and γ -HBCDD isomers were below the limits of detection in all samples.

The present study is, to our knowledge, the first survey of CDT in egg samples. CDT levels were, however, below the limits of detection in all samples.

Levels of α -HBCDD were highest in the eggs of black-legged kittiwakes and decreased in the order herring gulls \approx glaucous gulls $>$ Atlantic puffins. As for BDEs, no spatial trend of HBCDDs was apparent in the dataset.

Levels of total-HBCDD in the present study were lower than that in eggs of Swedish peregrine falcons and common terns (*Sterna hirundo*) from Belgium, but higher than mean concentrations reported in eggs of peregrine falcons from South Greenland and little owl (*Athene noctua*) eggs from Belgium.

Generally, levels of α -HBCDD increased from 1983 to 2003.

Mercury

Likewise to BDEs and HBCDD, there was no significant spatial variation in mercury concentrations. Mercury levels in eggs of glaucous gulls were higher than those in eggs of the other seabirds.

Levels of mercury determined in this study were generally lower than those reported in various species from North America, e.g., eggs of common loons (*Gavia immer*), bald eagles (*Haliaeetus leucocephalus*), Forster (*Sterna forsteri*) and common terns (*Sterna hirundo*), cormorants (*Phalacrocorax carbo*) and herring gulls. Mercury levels in the present study were also lower than those reported in eggs of red-throated divers (*Gavia stellata*) from Sweden, but similar to those previously reported in eggs of black-legged kittiwakes from Norway in the 1970s. Furthermore, mercury concentrations in the present study were similar to levels reported in eggs of cormorants from Lake Michigan, great blue herons (*Ardea herodias*) and wood ducks (*Aix sponsa*) from northeastern North America.

No significant temporal trend of mercury was found in the present study.

5. Background

Brominated flame retardants (BFRs) are a recent class of chemicals of global environmental concern. Current reports have demonstrated that BFRs are widespread in the environment and detected far from the locations where they are produced and used (de Wit et al. 2004; Verreault et al. 2005). Within the group of BFRs are the polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD) isomers (α -, β -, γ -HBCDD).

PBDEs comprise 209 potential congeners that are structurally comparable to PCBs and PCDD/Fs (Birnbaum and Staskal 2004). It has been shown that the lower-brominated PBDEs degrade more slowly and are more bioaccumulative and toxic than the higher-brominated PBDEs, e.g., deca-BDE. Nevertheless, PBDEs (including deca-BDE) are ubiquitous in the environment, and have been reported in a variety of animals, including eggs of aquatic and terrestrial birds (Lindberg et al. 2004; Sellström et al. 2003; Verreault et al. 2004; 2005).

HBCDDs are produced by bromination of cyclododecane, resulting in a technical product comprising several isomers, in which the most abundant enantiomers are α -HBCDD, γ -HBCDD and β -HBCDD (AMAP 2004; Birnbaum and Staskal 2004). Although γ -HBCDD is the most dominant enantiomer in technical mixtures (~ 80 %) and sediments (Alaee et al. 2003; Morris et al. 2004), α -HBCDD is the primary congener detected in biota samples (Morris et al. 2004; Zegers et al. 2005; Janak et al. 2005). HBCDDs have been detected in a number of environmental samples, and have shown potential for biomagnification in the aquatic food web as they are very hydrophobic and resistant to degradation by biological and abiotic processes (Birnbaum and Staskal 2004). Transformation of hexabromocyclododecane have been shown to occur via a step-wise reductive dehalogenation of HBCDD to cyclododecatriene in sludge, sediment and soil. There were no indications of further transformation of cyclododecatriene (EU 2005).

The total production of BFRs, including PBDEs and HBCDD, has increased in the United States and in Canada over the last decades. In Europe there has been an overall decline of the lower-brominated PBDEs as a result of a ban of penta- and octa-BDE mixtures within countries of the European Union (EU) (AMAP 2004; Birnbaum and Staskal 2004). There is currently no restriction in the production and use of deca-BDE and HBCDD, which together account for 36 % of the worldwide production of BFRs (BSEF 2004; SFT 2005). However, risk assessments and testing of deca-BDE and HBCDDs are being carried out within the EU.

Mercury is a highly toxic naturally-occurring element that is found in rocks, sediments and organic matter. Human activities, such as metal processing, incineration of coal and mining of gold, release mercury into the food web. Mercury exists in many different forms in the environment, e.g., gaseous elemental mercury which can be transported by air and the highly toxic methylmercury, and has no known biological function. However, it has been shown to bioaccumulate in the food web, resulting in high levels in top predators such as marine mammals and seabirds (AMAP 2005).

Emissions of mercury have been reduced in western European countries and North America over the last 25 years. Nevertheless, there is evidence that global emissions of mercury are escalating, especially from Asian countries (AMAP 2005).

The aim of the present study was to investigate possible species and regional differences and temporal trends of levels of BFRs, cyclododeca-1,5,9-triene (CDT) and mercury in seabird eggs. Eggs were collected from herring gulls (*Larus argentatus*), Atlantic puffins (*Fratercula arctica*) and black-legged kittiwakes (*Rissa tridactyla*) in 1983, 1993 and 2003 at Røst and Hornøya (Northern Norway). Furthermore, eggs of glaucous gulls (*Larus hyperboreus*) were collected in 1997 and 2002 at Bjørnøya (Svalbard). Current levels and congener/isomer patterns were compared to previously reported levels in eggs of free-ranging seabirds.

Little is known about levels, congener/isomer patterns and temporal trends of PBDEs, HBCDDs, CDT and mercury in seabirds. Eggs of seabirds have been recognized as useful monitoring tools to examine temporal trends of contaminants in the marine environment (Bignert et al. 1995; Braune et al. 2001; Braune et al. 2002; Hebert et al. 1997). This is mainly because lipid soluble halogenated organic contaminants (HOCs) are transferred along with lipids during egg formation (Bargar et al. 2001; Drouillard and Norstrom 2001). Accordingly, eggs can be utilized as non-invasive indicators of BFR and mercury exposure.

6. Materials and methods

6.1 Species studied and fieldwork

A total of 156 fresh eggs were collected from herring gulls, black-legged kittiwakes, Atlantic puffins and glaucous gulls in 1983, 1993/1997, and 2002/2003 (Table 1). The Atlantic puffins and black-legged kittiwakes feed mainly on fish such as herring and capelin, whereas the herring gulls and glaucous gulls have a more varied and opportunistic diet composed of fish, seabird chicks and eggs (Anker-Nilssen et al. 2000). Bjørnøya, Hornøya and Røst were selected as sampling areas to assess if there were a north-south (spatial) gradient in the dataset. Due to the lack of herring gull eggs from Røst in 1983, herring gull eggs from Hekkingen were included.

The eggs were wrapped in aluminium foil and stored frozen until laboratory analyses. For the brominated compounds all eggs were analyzed recently (2005), whereas mercury, were analyzed in 1983, 1993 and 2005 (Barrett et al. 1985; 1996).

Table 1. Number of eggs analyzed for a) brominated flame retardants (BFRs) and mercury in 2005, and b) mercury in 1983 and 1993 from herring gulls (H. gull), black-legged kittiwakes, Atlantic puffins and glaucous gulls collected from Røst, Hekkingen (in brackets), Hornøya and Bjørnøya in 1983, 1993/1997 and 2002/2003.

a) Number of eggs analyzed for BFRs and mercury (marked ^m) in 2005

Species	Røst/(Hekkingen)			Hornøya			Bjørnøya	
	1983	1993	2003	1983	1993	2003	1997	2002
H. gull	(5)	5	5 ^m	5	5	5 ^m	-	-
Puffin	5	4	5 ^m	5	5	5 ^m	-	-
Kittiwake	5	5	5 ^m	5	5	5 ^m	-	-
Glaucous gull	-	-	-	-	-	-	4 ^{m*}	3 ^m

* 201 pooled eggs.

b) Number of eggs previously analyzed for mercury in 1983 and 2003

Species	Røst		Hornøya	
	1983	1993	1983	1993
H. gull	5	5	5	5
Puffin	5	5	5	5
Kittiwake	5	5	5	5
Glaucous gull	-	-	-	-



Figure 1. Map showing the localities from which eggs were collected in 1983, 1993/1997 and 2002/2003.

6.2 Chemical analyses

6.2.1 Determination of BFRs and CDT

The chemical analyses of BDE-209 were performed at the Laboratory of Environmental Toxicology at the Norwegian School of Veterinary Science in Oslo. The content of the eggs were homogenized in a food blender. The egg homogenates (~3 g) were weighed in 80 ml centrifugation tubes and spiked with the internal standards BDE-77, BDE-119, and BDE-181 (for PBDEs and HBCDDs) and ^{13}C -BDE-209 (for BDE-209) (Cambridge Isotope Laboratories, Inc., Andover, MA, USA). The lipids were extracted twice using cyclohexane and acetone (3:2) and an ultrasonic homogenizer

(4710 Series, Cole- Palmer Instruments, Chicago). The supernatants of both extractions were merged and concentrated to about 1 ml using a Zymark® evaporation system (TurboWap II, Zymark Corporation, Hopkinton, MA, USA) at 40°C and by a gentle flow of nitrogen (pressure 0.6 bar). The concentrated lipid extracts were quantitatively transferred to volumetric flasks, and the final volume adjusted to 5 ml with cyclohexane. The lipid determination was done gravimetrically using 1 ml aliquot of the sample. For cleanup (i.e. removal of lipids) the rest of the lipid extracts were treated twice with ultra clean (purity 98.8%) concentrated H₂SO₄ (Scanpure, Chemsan AS, Elverum, Norway). The sample concentrates were transferred to GC vials and put in dark containers

Detection of BDE-28, -47, -100, -99, -154, -153, -183 and total HBCDD

Aliquots (1 µl) of the concentrates were automatically injected (Agilent Auto sampler, Agilent Technologies, Avondale, PA, USA) and the gas chromatographic separation of BFRs was performed by a Hewlett Packard 6890 Series, equipped with a pulsed split less injector (at 250°C) connected to a MS quadrupol detector (Agilent Technologies, Avondale, PA, USA). The PBDEs and HBCDDs were monitored using negative chemical ionization (NCI) in selected ion monitoring (SIM) at m/z ratios 79 and 81. The mean recoveries of the BDEs were 91-102 %, but 166 % for BDE-183. The coefficients of variation (CV) were 6-9 %, but 26 for BDE-153 and 69 for BDE-183. The detection limits ranged between 0.01 and 0.08 µg/kg wet weight (For BDE-183: 0.17 µg/kg wet weight in 1983 and 1993, 0.81 µg/kg wet weight in 2003). The mean recovery of total HBCDD was 102 % and the detection limits ranged between 0.15-0.83 µg/kg wet weight. In general method blank samples were analyzed to monitor contamination. In situations where an analyte indicated background contribution in blank samples (i.e., for BDE-153 and BDE-183) the detection limits were increased.

Detection of BDE-209

For detection of BDE-209 a programmable temperature vaporization (PTV) injector (Agilent Technologies) was used. 10 µl were injected for analysis on a GC and mass spectrometer (J&W Scientific, Agilent Technologies). The separation and identification of the compound was performed by a 10 m long column with a 0.1 µm film layer. BDE-209 was monitored using negative chemical ionization (NCI) in selected ion monitoring (SIM) at m/z 484.5 and 486.5 and ¹³C-BDE-209 at 494.5 and 498.5. The mean recovery of BDE-209 was 101 %, with a coefficient of variation (CV) of 7 %. The detection limits ranged between 0.03 and 0.125 µg/kg wet weight. Method blanks were used as described earlier.

Detection of Cyclododeca-1,5,9-triene (CDT)

For detection of CDT 1 µl was injected in a GC-MS (for details see detection of BFRs). The mass spectrometer was operated in the electron ionization (EI) mode and monitored at m/z ratios 133.134 and 162. The detection limit was 0.5 µg/kg wet weight. The validation of the analytical method for this compound was not finished at the time of reporting.

Detection of α -, β - and γ -HCBDD

For determination of α -, β - and γ -HCBDD, the extracts were analyzed using an API 3000 LC-MS-MS system (triple quadrupole) (Applied Biosystem, USA) connected to a C18 column (15 cm x 2.1 mm, 5 μ m) (Supelco). As mobile phases Ammoniumacetate in water (A) and Ammoniumacetate in 99% acetonitrile and 1% water (B) were used with a flow of 0.2 ml/min and gradients of 70 % B to 100 % B in 10 min, hold 5 min at 100 %B. The detection was performed by MRM (multiple reaction monitoring) and the mass transition ion-pair was selected as m/z 640.7-m/z 80.8. The mean recoveries of the α -, β - and γ -HCBDDs were 115, 111 and 90 %.

6.2.2 Quality Assurance BFRs

The laboratory is accredited for analysis of PBDEs in biological samples according to the requirements of the NS- EN ISO/IEC 17025 (TEST 051) and has successfully participated in several intercalibration tests, of which the most relevant and up to date were: QUASIMEME 2002 (Exercise 524, round 28 on BFRs), FIRE 2003 (EU-project internal exercise on BFRs in food and egg of common tern), QUASIMEME 2003 (Exercise 565, round 33 on BFRs) and NIST/NOAA/MMHSRP 2003 (Intercomparison Exercise Program of Organic Contaminants and Trace Elements in Marine Mammal Tissues on PCBs, pesticides, PBDEs, toxaphenes).

6.2.3 Determination of Mercury:

Mercury was analysed by the National Veterinary Institute, Oslo, Norway. The analytical method is accredited after NS-EN ISO/IEC 17025 by the Norwegian Accreditation.

In short, the samples were digested by a mixture of nitric acid and hydrogen peroxide in a closed system using a microwave oven (Milestone) and analysed by cold vapour atomic absorption spectrometry (CVAAS) using Stannous Chloride (SnCl_2) (Varian inc.). For a detailed description of the methods see Sturman 1985.

6.2.4 Quality Assurance mercury

Certified reference materials such as TORT-2 and LUTS-1 were analysed together with the samples for assurance of quality.

6.3 Data analyses

The statistical results of the present study have to be interpreted with caution due to the very low sample sizes.

Univariate statistics were conducted using R version 2.1.1 (<http://r-project.org>, Denmark, 2005). A generalized linear model with parametric bootstrapping ($N = 2000$) was conducted to examine whether there were any species, regional or temporal differences in the dataset. The bootstrapped, bias corrected and accelerated confidence intervals (BC_a) were used to check whether the coefficients of the different factors were different from zero (Davison and Hinkley 1997). For compounds included in the statistical analyses, values below the method detection limits were set to half the compound-specific

detection limit (i.e., for BDE-183). In the statistical interpretations of BDE-209 frequencies of detected versus non-detected were used instead of absolute concentrations of BDE-209. This was preferred because only 20 %, 42 % and 40 % of the samples from 1983, 1993 and 2003, respectively, were above the BDE-209 detection limit. Fisher's exact test was conducted to assess species-specific and spatial trends of BDE-209 in the dataset ($\alpha = 0.05$).

Multivariate statistics were conducted using CANOCO 4.5 for Windows (Microcomputer Power, Ithaca, USA, 2002). A direct (constrained) multivariate ordination analysis (redundancy analysis (RDA)) was performed to examine the species-specific, spatial and temporal trends of BFRs (log-transformed wet weight (ww) concentrations). The three seabird species (herring gull, Atlantic puffin and black-legged kittiwake), sampling area (Hornøya and Røst) and sampling year (1983, 1993 and 2003) were included in the model as nominal explanatory variables, whereas lipid percentage was included as a fixed covariate. The significance of the model was evaluated using Monte Carlo permutations (499) ($\alpha = 0.05$) (Ter Braak and Šmilauer 1998).

A principal component analysis (PCA) was used to investigate BFR profile characteristics in the egg samples. The samples' wet weight concentrations of PCB congeners were standardized by norm prior to the analysis (Ter Braak and Šmilauer, 1998). Lipid percentage, sampling area and sampling year were included as covariates.

Eggs of glaucous gulls were not included in any of the statistical analyses because of different sampling location (Bjørnøya) and sampling years (1997 and 2002) compared to the other species. Furthermore, eggs collected in 1997 were "pooled" without testing the homogeneity, and therefore not comparable to the individual eggs from other locations and sampling years.

7. Results and Discussion

7.1 Polybrominated diphenyl ethers

For details on the concentrations of the different congeners see Tables 2 and 3. BDE-28, 47, 100, 99, 154 and 153 were detected in all the herring gull, Atlantic puffin and black-legged kittiwake egg samples. BDE-183 and BDE-209 were detected in 60 % and 30 % of the eggs, respectively (Figure 2). All BDE congeners were detected in 100 % of the egg samples of glaucous gulls.

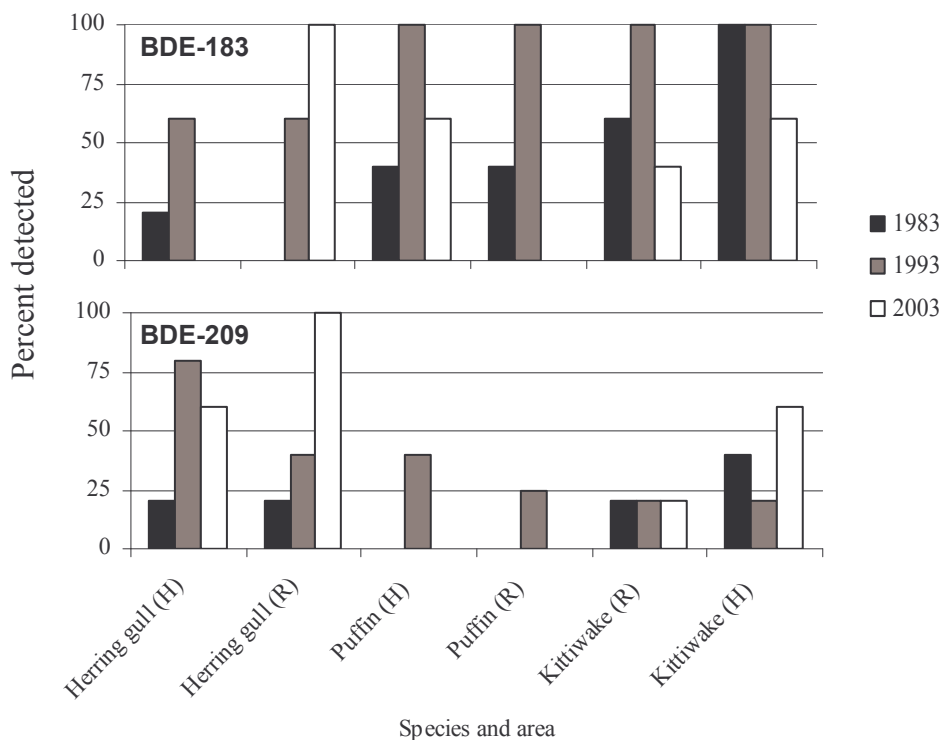


Figure 2. Percent BDE-183 and 209 detected in herring gull eggs, Atlantic puffin eggs and black-legged kittiwake eggs at Hornøya (H) and Røst (R) in 1983, 1993 and 2003.

7.1.1 Spatial trends of BDEs

No clear spatial difference of BDEs was apparent in the dataset, with the exception of BDE-153 of which levels were significantly higher at Røst than those at Hornøya (Appendix 1; Figure 4).

A lack of geographical difference in BDE levels in seabird eggs from Hornøya and Røst could be explained by:

a) similar feeding habits at these two colonies, b) similar local exposure and/or, c) similar migration patterns/exposure during winter and early spring in birds from different colonies.

7.1.2 Species-specific trends of BDEs

Concentrations of Σ BDE, BDE-47 and 100 were significantly higher in eggs of herring gulls than in eggs of Atlantic puffins and black-legged kittiwakes (Appendix 1; Figures 4 and 5).

BDE-209 was detected in 64 % of the herring gull eggs, 10 % of the Atlantic puffin eggs and 32 % of the black-legged kittiwake eggs. However, there was no significant species-specific trend ($p > 0.05$).

Differences between eggs of glaucous gulls and the other seabird eggs were not tested statistically (see Materials and methods). A qualitative assessment suggested that levels of BDEs in glaucous gulls were similar to those in eggs of herring gulls.

Higher concentrations of BDEs in eggs of herring gulls and glaucous gulls, than in eggs of Atlantic puffins and black-legged kittiwakes, may be the result of the predatory habit of the former, which occasionally eat seabird eggs and chicks. This was corroborated by findings of higher contaminant burdens in glaucous gulls feeding primarily on seabirds and eggs compared to those feeding mainly on fish (Bustnes et al. 2000).

7.1.3 Levels of BDEs

The BDE concentration in seabirds is determined mainly by their feeding habits and ability to biotransform BDE congeners. Seabirds, which feed exclusively on crustaceans and fish such as Atlantic puffins and black-legged kittiwakes, exhibit lower BDE concentrations compared to predatory terrestrial and marine birds, e.g., peregrine falcons, herring gulls and glaucous gulls (Gabrielsen 2005). For example, levels of BDEs in eggs of Atlantic puffins and black-legged kittiwakes were considerably lower than levels reported in eggs of predatory aquatic birds. Furthermore, concentrations of BDEs in herring gull and glaucous gull eggs were similar to levels reported in marine predatory birds, but higher than in common guillemot eggs from the Baltic Sea (Table 3).

Generally, seabirds in arctic regions are contaminated by the same chemicals as seabirds living further south, although the concentrations and proportions differ (Gabrielsen 2005). For instance, BDE levels in the present study were lower than in herring gull eggs from the Great Lakes (Table 3).

Levels of BDEs in eggs of herring gulls and glaucous gulls of the present study were lower than levels reported for predatory bird eggs from Norway, South Greenland and Sweden (Table 3). It is possible that the predatory birds in these referred studies feed at a higher trophic level than the seabirds of the present study. Alternatively, the high levels of BDEs in the predatory birds might be linked to their migratory habits, e.g., winter migration to more contaminated areas in central and southern Europe. Glaucous gulls and herring gulls generally migrate to areas in Northern Europe during the winter (Anker-Nilssen et al. 2000).

7.1.4 Congener patterns of BDEs

For all seabird eggs BDE-47 was the most abundant BDE congener, constituting from 34.6 to 77.4 % of Σ BDEs. Levels of the different BDE congeners in the present study decreased in the order BDE-47 > BDE-99 > BDE-100 > BDE-153 \approx BDE-154 \approx BDE-183 > BDE-28 > BDE-209 (Figure 3).

Higher proportions of BDE-47 compared to other BDE congeners were also found in eggs of common guillemots sampled from the Baltic Sea (Sellström et al. 2003), eggs of herring gulls from the Great lakes (Norstrom et al. 2002), eggs of black guillemots (*Cephus grylle*) from Greenland (Vorkamp et al. 2004), and eggs of goshawks, white-tailed eagles and ospreys from Norway (Herzke et al. 2005). A higher biomagnification factor for BDE-47, compared to the other BDE congeners, could explain the observed patterns as previously confirmed in common guillemots (*Uria aalge*) where biomagnification factors decreased in the order BDE-47 > BDE-99 > BDE-100 (de Wit 2002).

Congener patterns in eggs of other birds of prey, such as merlins and peregrine falcons (Linberg et al. 2004; Herzke et al. 2005; Vorkamp et al. 2005) were different by showing a predominance of BDE-153 proportions. Although the habitat and migratory behavior may explain differences in BDE patterns between birds, it has been suggested that birds feeding in the terrestrial environment, and on other birds, may be more highly exposed to higher brominated BDE congeners than marine species (Law et al. 2003). Other factors such as metabolism, bioavailability and uptake efficiencies also may influence the overall BDE profiles.

7.1.5 Temporal trends of BDEs

For all BDEs, with the exception of BDE-183 and 209, concentrations increased from 1983 through 1993 in eggs of herring gulls, Atlantic puffins and black-legged kittiwakes. Between 1983 and 2003, only BDE-153, 154 and 183 levels increased (Appendix 1). There is great uncertainty connected to the analytical methods of BDE-183. Consequently, results of this congener should be interpreted with particular caution.

For BDE-209 no statistical significant difference (frequencies of detected versus non-detected, bootstrap estimate 0.04 ± 0.04 , $p = 0.2$) was found between sampling years although concentrations of BDE-209 in eggs of herring gulls and black-legged kittiwakes tended to be higher in 2003 than in 1983 and 1993. In eggs of Atlantic puffins levels were above the detected limit only in 1993. Concentrations of BDEs in glaucous gull from Bjørnøya sampled in 1997 and 2002 were practically identical.

Reports on temporal trends of BDEs in seabird eggs are scarce. However, the results of the present study were corroborated with findings from a recent temporal trend study (1969 to 2001) of tetra- and penta-brominated diphenyl ethers in eggs of common guillemots from the Baltic Sea that has reported increasing trends of BDEs from the 1970s to the 1980s. BDE levels peaked around the mid-1980s to the late 1980s, followed by a rapid decrease in BDE concentrations during the rest of the study period (Sellström et al. 2003). A reduction in emission from the European production could be the

explanation for the decreasing concentrations of BDEs. Nevertheless, concentration changes of BDEs might be associated with local use and emissions.

In contrast to the reports above, studies in the Great Lakes and South Greenland revealed increasing concentrations of BDEs up until today. For example, a temporal trend study of BDEs (BDE-99, 100, 153 and 209) in peregrine falcon eggs from South Greenland reported a statistically significant 6-10 % increase in BDEs per year from 1986 to 2003 (Vorkamp et al. 2005). Furthermore, BDE concentrations (BDE-28, 99, 100, 153 and 154) in herring gull eggs from the Great Lakes increased 20-75-fold from 1981 to 2000 (Norstrom et al. 2002).

7.1.6 Risk assessment of BDEs

A wide range of effects associated with PBDE exposure has been reported. This includes reproductive and developmental effects, induction of cytochrome P450-dependent (CYP) monooxygenases and cancer (Viberg et al. 2003; de Wit 2002; de Wit et al. 2004).

Furthermore, debromination of BDE-209 to more bioavailable products such as BDE-47 and 99, has been shown to occur in the presence of UV light and sunlight (de Wit 2002). Moreover, several apparent debrominated products of BDE-209 were observed following a feeding experiment exposing juvenile carp (*Cyprinus carpio*) to BDE-209 (Stapleton et al. 2004). Generally, the lower brominated congeners are more bioaccumulative, persistent and toxic than BDE-209. Consequently, debromination of BDE-209 may pose a great threat to animals because it leads to potentially more toxic, stable and persistent degradation products (Birnbaum and Staskal 2004). Another group of very toxic compounds produced by the degradation of BDEs under combustion conditions is the polybromo-polychlor-dibenzo-*p*-dioxins and dibenzofurans (Söderström and Marklund 2002; 2004).

The toxicity thresholds of BDEs have not been extensively studied. There are, however, dietary studies reporting threshold levels for various biological effects in fish and laboratory rodents (e.g., Darnerud et al. 2001; Hardy 2002; Birnbaum and Staskal 2004). Because these studies do not report tissue thresholds (thresholds levels in eggs) it is not possible to directly compare threshold levels obtained in these studies with the present study. Furthermore, laboratory rodents and fish might be more, or less, sensitive to BDE exposure than free-ranging seabirds.

Table 2. Mean \pm standard error (se.) of the estimated mean, range (min-max) and median concentrations (ng/g wet weight) of brominated diphenylethers (BDEs) measured in eggs of herring gulls, Atlantic puffins and black-legged kittiwakes collected from Hornøya (a) and Røst (b) in 1983, 1993 and 2003. The number of samples above the detection limit is shown relative to the number of samples analyzed (N).**a) Hornøya**

Species	Year	Lipid %	BDE-28	BDE-47	BDE-100	BDE-99	BDE-154	BDE-153	BDE-183	BDE-209	Σ BDE	
Herring gull	1983	Mean \pm se	11.0 \pm 2.2	44.0 \pm 8.8	6.1 \pm 1.2	5.4 \pm 1.1	0.7 \pm 0.1	1.0 \pm 0.2	-	-	59.0 \pm 11.8	
		min-max	8.1 - 12.6	16.3 - 78.1	2.1 - 11.2	1.3 - 8.9	0.2 - 1.5	0.2 - 1.6	nd - 3.7	0.1 - 0.3	20.5 - 103	
		N	5/5	5/5	5/5	5/5	5/5	5/5	1/5	2/5	5/5	
		median	10.8	46.9	6.7	4.5	0.5	1.0	3.7	0.2	64.5	
	1993	Mean \pm se	10.2 \pm 2.0	28.3 \pm 5.7	4.5 \pm 0.9	5.9 \pm 1.2	0.8 \pm 0.2	0.9 \pm 0.2	1.2 \pm 0.2	0.2 \pm 0.0	0.2 \pm 0.0	42.3 \pm 8.5
		min-max	9.8 - 10.9	17.9 - 38.8	2 - 6.36	2 - 11.6	0.5 - 1.0	0.5 - 1.2	nd - 3.0	nd - 0.5	nd - 0.5	24.5 - 54.6
	N	5/5	5/5	5/5	5/5	5/5	5/5	3/5	4/5	5/5		
	median	9.9	32.3	5.7	5.6	0.9	1.0	0.4	0.1	0.1	49.6	
2003	Mean \pm se	9.7 \pm 1.9	0.7 \pm 0.1	38.8 \pm 7.8	7.4 \pm 1.5	4.4 \pm 0.9	1.6 \pm 0.3	1.3 \pm 0.3	-	4.1 \pm 0.8	56.6 \pm 11.3	
		min-max	8.5 - 11.1	0.5 - 0.9	27.3 - 72.3	6.4 - 10.1	2.8 - 9.4	1.1 - 2.8	0.6 - 3.0	-	nd - 11.2	39.3 - 90.9
		N	5/5	5/5	5/5	5/5	5/5	5/5	0/5	3/5	5/5	
		median	9.4	0.7	32.5	6.7	3.2	1.3	1	nd	0.7	46.1
	A. puffin	1983	Mean \pm se	12.6 \pm 2.5	8.3 \pm 1.7	2.0 \pm 0.4	4.4 \pm 0.9	0.6 \pm 0.1	0.5 \pm 0.1	-	-	16.7 \pm 3.3
			min-max	10.8 - 14.2	6.4 - 9.9	1.5 - 2.4	3.1 - 5.1	0.4 - 0.8	0.3 - 0.6	nd - 0.3	-	12.4 - 19.7
		N	5/5	5/5	5/5	5/5	5/5	5/5	2/5	0/5	5/5	
		median	12.4	9.1	2.2	4.8	0.6	0.5	0.2	-	-	18.7
1993		Mean \pm se	11.7 \pm 2.3	13.3 \pm 2.7	2.8 \pm 0.6	6.3 \pm 1.3	1.4 \pm 0.3	1.8 \pm 0.4	3.2 \pm 0.6	-	-	30.8 \pm 6.2
		min-max	10.8 - 12.5	8.5 - 16.5	2.1 - 3.9	5.0 - 8.8	1.1 - 1.8	0.9 - 4.4	0.2 - 13.7	nd - 0.7	nd - 0.7	19.4 - 43.6
	N	5/5	5/5	5/5	5/5	5/5	5/5	5/5	2/5	5/5		
	median	11.7	14.1	2.8	5.8	1.4	1.0	0.4	0.4	0.4	28.0	
2003	Mean \pm se	13.7 \pm 2.7	0.6 \pm 0.1	6.2 \pm 1.2	1.4 \pm 0.3	2.5 \pm 0.5	1.2 \pm 0.2	0.8 \pm 0.2	-	-	13.5 \pm 2.7	
		min-max	10.9 - 15.3	0.4 - 1.0	1.1 - 2.1	1.7 - 3.3	0.7 - 1.5	0.4 - 1.2	nd - 1.8	-	-	9.0 - 20.7
		N	5/5	5/5	5/5	5/5	5/5	5/5	3/5	0/5	5/5	
		median	14.5	0.5	1.3	2.2	1.2	0.7	1.7	-	-	13.0
	Kittiwake	1983	Mean \pm se	9.8 \pm 2.0	5.9 \pm 1.2	0.7 \pm 0.1	1.2 \pm 0.2	0.6 \pm 0.1	0.4 \pm 0.1	1.5 \pm 0.3	-	10.2 \pm 2.0
			min-max	9.2 - 10.5	4.9 - 7.1	0.6 - 0.8	1.1 - 1.6	0.1 - 2.1	0.2 - 1.0	nd - 3.7	nd - 0.2	7.8 - 14.5
		N	5/5	5/5	5/5	5/5	5/5	5/5	3/5	1/5	5/5	
		median	9.7	5.4	0.7	1.2	0.2	0.3	0.5	0.2	10.1	
1993		Mean \pm se	9.6 \pm 0.1	6.4 \pm 0.3	0.8 \pm 0.1	2 \pm 0.1	0.3 \pm 0	0.6 \pm 0.1	0.4 \pm 0.1	-	-	11.0 \pm 0.5
		min-max	9.2 - 10.1	5.3 - 9.1	0.7 - 1.3	1.6 - 2.8	0.2 - 0.6	0.4 - 1.3	0.03 - 1.7	nd - 0.1	nd - 0.1	8.8 - 15.7
	N	5/5	5/5	5/5	5/5	5/5	5/5	5/5	1/5	5/5		
	median	9.5	6.0	0.8	1.8	0.2	0.4	0.1	0.1	0.1	10.1	
2003	Mean \pm se	9.5 \pm 1.9	0.4 \pm 0.1	8.8 \pm 1.8	1.0 \pm 0.2	1.8 \pm 0.4	0.5 \pm 0.1	1.0 \pm 0.2	-	-	14.9 \pm 3.0	
		min-max	8.7 - 11.1	0.2 - 0.6	4.6 - 14.2	0.6 - 1.9	0.7 - 3.2	0.2 - 0.8	0.5 - 1.6	nd - 5.4	nd - 0.2	7.3 - 23.8
		N	5/5	5/5	5/5	5/5	5/5	5/5	2/5	1/5	5/5	
		median	8.9	0.5	7.6	1.8	0.5	0.8	3.5	0.2	0.2	15.8

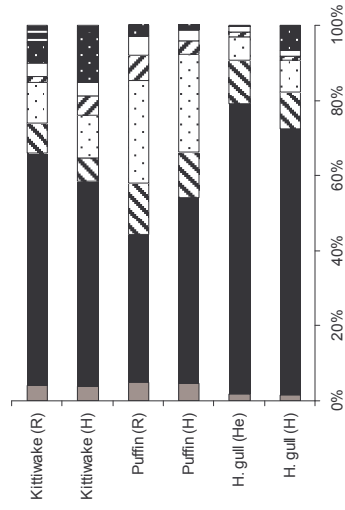
b) Røst/(Hekkingen)

Species	Year	Lipid %	BDE-28	BDE-47	BDE-100	BDE-99	BDE-154	BDE-153	BDE-183	BDE-209	ΣBDE
Herring gull (1983)	Mean ± se	9.5 ± 1.9	0.7 ± 0.1	29.3 ± 5.9	4.5 ± 0.9	2.3 ± 0.5	0.5 ± 0.1	0.6 ± 0.1	-	-	37.9 ± 7.6
	min-max	8.7 - 10.4	0.4 - 1.2	11.0 - 61.9	1.4 - 10.2	1.2 - 4.0	0.2 - 0.9	0.2 - 1.7	-	nd - 0.1	14.4 - 80.0
	N	5/5	5/5	5/5	5/5	5/5	5/5	5/5	0/5	1/5	5/5
1993	Mean ± se	8.3 ± 1.7	2.1 ± 0.4	61.5 ± 12.3	9.5 ± 1.9	19.0 ± 3.8	1.9 ± 0.4	2.4 ± 0.5	0.4 ± 0.1	-	96.8 ± 19.4
	min-max	6.9 - 10.1	1.7 - 2.7	30.7 - 106	4.8 - 17.6	2.9 - 47.9	1.1 - 3.2	0.8 - 4.6	nd - 0.8	nd - 0.2	44.5 - 183
	N	5/5	5/5	5/5	5/5	5/5	5/5	5/5	3/5	2/5	5/5
2003	Mean ± se	10.4 ± 2.1	0.7 ± 0.1	37.9 ± 7.6	6.0 ± 1.2	4.5 ± 0.9	1.5 ± 0.3	2.4 ± 0.5	4.0 ± 0.8	3.5 ± 0.7	60.5 ± 12.1
	min-max	6.3 - 15.0	0.2 - 1.1	10.2 - 51.1	2.4 - 8.4	1.4 - 10.8	0.9 - 1.8	1.0 - 4.5	1.7 - 8.3	0.1 - 14.4	45.3 - 82.6
	N	5/5	5/5	5/5	5/5	5/5	5/5	5/5	5/5	5/5	5/5
A. puffin	Mean ± se	12.2 ± 2.4	0.5 ± 0.1	3.7 ± 0.7	1.3 ± 0.3	2.6 ± 0.5	0.6 ± 0.1	0.5 ± 0.1	-	-	9.2 ± 1.8
	min-max	10.8 - 13.3	0.3 - 0.6	2.5 - 5.3	0.9 - 2.0	1.7 - 3.9	0.5 - 1.2	0.3 - 0.9	nd - 0.3	-	6.4 - 14.1
	N	5/5	5/5	5/5	5/5	5/5	5/5	5/5	2/5	0/5	5/5
1993	Mean ± se	12.2 ± 3.1	1.2 ± 0.3	8.8 ± 2.1	2.6 ± 0.6	7.6 ± 1.9	1.5 ± 0.4	1.9 ± 0.5	1.6 ± 0.4	-	25.1 ± 6.3
	min-max	10.8 - 13.5	0.8 - 1.6	6.8 - 11.8	2.1 - 3.5	6.6 - 9.1	1.4 - 1.8	1.5 - 2.6	0.3 - 5.5	nd - 0.1	19.5 - 30.2
	N	4/4	4/4	4/4	4/4	4/4	4/4	4/4	4/4	1/4	4/4
2003	Mean ± se	13.7 ± 2.7	0.4 ± 0.1	5.0 ± 1.0	1.5 ± 0.3	3.3 ± 0.7	1.3 ± 0.3	0.9 ± 0.2	-	-	12.5 ± 2.5
	min-max	11.2 - 17.1	0.3 - 0.8	2.8 - 9.7	0.8 - 3.1	1.7 - 6.5	0.2 - 3.0	0.5 - 1.6	-	-	7.0 - 24.7
	N	5/5	5/5	5/5	5/5	5/5	5/5	5/5	0/5	0/5	5/5
Kittiwake	Mean ± se	7.1 ± 1.4	0.6 ± 0.1	9.3 ± 1.9	1.2 ± 0.2	1.6 ± 0.3	0.2 ± 0.0	0.5 ± 0.1	0.9 ± 0.2	-	14.6 ± 2.9
	min-max	6.2 - 8.2	0.4 - 0.8	6.7 - 12.7	0.9 - 1.7	0.9 - 2.0	0.2 - 0.4	0.3 - 1.0	0.2 - 2.5	nd - 1.3	10.4 - 20.8
	N	5/5	5/5	5/5	5/5	5/5	5/5	5/5	5/5	2/5	5/5
1993	Mean ± se	9.4 ± 1.9	1.7 ± 0.3	28.3 ± 5.7	3.8 ± 0.8	9.6 ± 1.9	1.1 ± 0.2	3.3 ± 0.7	0.8 ± 0.2	-	48.7 ± 9.7
	min-max	8.3 - 10.7	1.1 - 3.3	14.8 - 58.5	1.9 - 8.5	3.6 - 26.2	0.5 - 1.9	0.9 - 10.7	0.2 - 2.7	nd - 0.2	23.1 - 112
	N	5/5	5/5	5/5	5/5	5/5	5/5	5/5	5/5	1/5	5/5
2003	Mean ± se	9.9 ± 2.0	1.0 ± 0.2	18.7 ± 3.7	2.0 ± 0.4	1.2 ± 0.2	0.8 ± 0.2	0.8 ± 0.3	2.5 ± 0.5	0.2 ± 0.0	26.1 ± 5.2
	min-max	7.3 - 11.0	0.6 - 1.4	10.8 - 24.8	1.1 - 2.7	0.6 - 1.5	0.5 - 1.1	0.5 - 1.1	nd - 3.4	nd - 0.2	18.3 - 32.0
	N	5/5	5/5	5/5	5/5	5/5	5/5	5/5	3/5	3/5	5/5
median	10.5	0.9	19.6	2.2	1.4	0.7	0.9	3.0	0.2	0.2	26.5

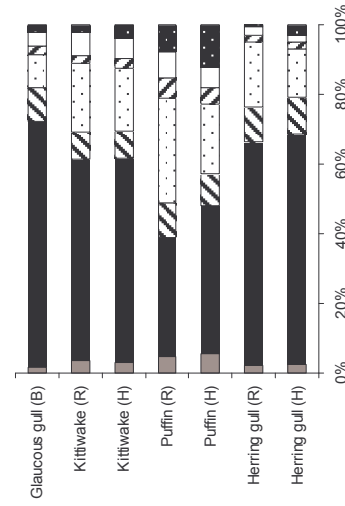
Table 3. Mean \pm standard error (se.), range (min-max) and median concentrations (ng/g wet weight) of brominated diphenylethers (BDEs) measured in eggs of glaucous gulls from Bjørnøya in 1997 (pooled eggs) and 2002. The number of samples above the detection limit is shown relative to the number of samples analyzed (N).

Species	Year	Lipid %	BDE-28	BDE-47	BDE-100	BDE-99	BDE-154	BDE-153	BDE-183	BDE-209	Σ BDE
Glaucous gull	2002	Mean \pm se	0.7 \pm 0.0	32.9 \pm 0.2	4.6 \pm 0.0	4.4 \pm 0.0	1.2 \pm 0.0	1.8 \pm 0.0	0.9 \pm 0.1	0.1 \pm 0.0	46.7 \pm 0.4
		min-max	0.7 - 0.8	31.7 - 33.5	4.5 - 4.8	4.3 - 4.5	1.1 - 1.3	1.6 - 1.9	0.1 - 1.4	0.1 - 0.2	44.1 - 48.0
		N	3/3	3/3	3/3	3/3	3/3	3/3	3/3	3/3	3/3
	median	9.0	0.7	33.3	4.6	4.4	1.2	1.9	1.1	0.1	47.4
1997	Mean \pm se	8.5 \pm 0.2	0.6 \pm 0.1	31.6 \pm 2.6	5.0 \pm 0.4	5.4 \pm 0.6	1.4 \pm 0.3	4.2 \pm 0.7	0.4 \pm 0.1	1.2 \pm 0.4	49.7 \pm 4.7
	min-max	8.0 - 9.2	0.4 - 0.9	24.4 - 39.9	4.1 - 6.5	3.6 - 7.3	0.9 - 2.4	1.7 - 5.6	0.1 - 0.8	0.1 - 2.6	40.6 - 66.1
	N	4/4	4/4	4/4	4/4	4/4	4/4	4/4	4/4	4/4	4/4
	median	8.2	0.5	30.5	4.4	5.2	1.0	5.2	0.3	0.8	42.5

a) 1983



b) 1993/1997



c) 2002/2003

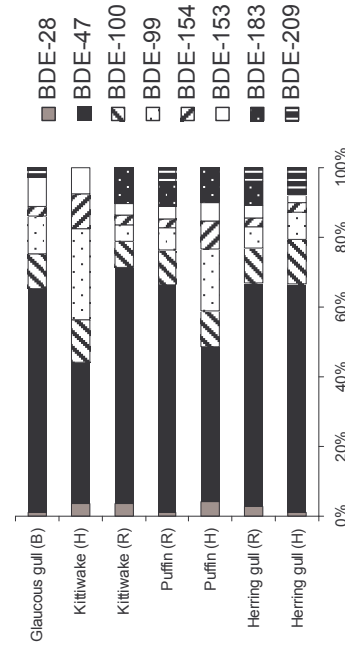


Figure 3. Percentage congener distribution to the sum of eight BDEs in eggs of herring gulls, Atlantic puffins and black-legged kittiwakes collected from Hornøya (H), Hekkingen (He) and Røst (R) in a) 1983, b) 1993 and c) 2003, in addition to eggs of glaucous gulls collected at Bjørnøya (B) in 1997 and 2002.

Table 4. BDE concentrations (ng/g lw) in bird eggs reported in the scientific literature. Values are ranges or mean concentrations.

Species	Scientific name	BDE-47	BDE-99	BDE-209	ΣBDE	Study area	Source
Peregrine falcon	<i>Falco peregrinus</i>	110	-	3.8-250	300-12 900	South Greenland	Vorkamp et al. 2005
		270	1100	130	3900	South Sweden	Lindberg et al. 2004
		360	860	110	4560	North Sweden	Lindberg et al. 2004
Herring gull	<i>Larus argentatus</i>	2270	1165	-	5460	Lake Ontario	Norstrom et al. 2002
		5900	3170	-	13 390	Lake Michigan	Norstrom et al. 2002
		1310	801	-	3175	Lake Huron	Norstrom et al. 2002
Glaucous gull	<i>Larus hyperboreus</i>	-	-	23.2-52.5	550	Bjørnøya	Verreault et al. 2004
				(4 out of 32 analysed eggs)			
Osprey	<i>Pandion haliaetus</i>	790	250	nd	1470	Norway	Herzke et al. 2005**
White-tailed sea eagle	<i>Haliaeetus albicilla</i>	2750	670	nd	4600	Norway	Herzke et al. 2005**
Golden eagle	<i>Aquila chrysaetos</i>	25	25	nd	150	Norway	Herzke et al. 2005**
Peregrine falcon	<i>Falco peregrinus</i>	140	500	nd	3100	Norway	Herzke et al. 2005**
Merlin	<i>Falco columbarius</i>	60	140	nd	415	Norway	Herzke et al. 2005**
Goshawk	<i>Accipiter gentilis</i>	515	180	nd	980	Norway	Herzke et al. 2005**
Great black-backed gull	<i>Larus marinus</i>	38.3*	2.8*	-	-	Northern Norway	Gabrielsen GW., unpublished results
Herring gull	<i>Larus argentatus</i>	51.6*	1.7*	-	-	Northern Norway	Gabrielsen GW., unpublished results
Common guillemot	<i>Uria aalge</i>	89	12	-	-	Baltic Sea	Sellström et al. 2003
Tawny owl	<i>Strix aluco</i>	-	-	nd-337	-	Norway	Bustnes JO., unpublished results
Little owl	<i>Athene noctua</i>	-	-	17	-	Belgium	Jaspers et al. 2004
				(1 out of 40 analyzed eggs)			

* lipid % not reported, ng/g ww

** Median concentrations

7.2 Hexabromocyclododecane (HBCDDs) and cyclododeca-1,5,9-triene (CDT)

For details on the concentrations of the different congeners see Tables 6 and 7. The isomer α -HBCDD was present in all eggs of glaucous gulls, herring gulls, Atlantic puffins and black-legged kittiwakes. Levels of β -HBCDD and γ -HBCDD isomers were below the limits of detection in all samples. This corroborated previous findings reporting strong prevalence of α -HBCDD in seabirds (Gabrielsen et al. 2005; Morris et al. 2004). One explanation for the high proportion of α -HBCDD in seabirds can be related to different biotransformation propensities of the enantiomers. For example, Wistar rats and harbor seals (*Phoca vitulina*) have been shown to metabolize and eliminate β -HBCDD and γ -HBCDD more efficiently than α -HBCDD (Zegers et al. 2005). Furthermore, it has been shown that α -HBCDD has a higher water solubility than β -HBCDD and γ -HBCDD, which may cause a preferential transfer of α -HBCDD from particles to the water phase and into organisms (Hunziker et al. 2004; Morris et al. 2004).

CDT levels were below the limits of detection in all samples in the present study. Since the analytical method for CDT was not validated at the reporting date, we recommend extended studies on this compound before final conclusions are made.

7.2.1 Spatial trends of HBCDDs

As for BDEs, no spatial variation of HBCDDs was apparent in the dataset (Appendix 1; Figure 3).

7.2.2 Species-specific trends of HBCDDs

Levels of α -HBCDD were highest in the eggs of black-legged kittiwakes and decreased in the order herring gulls \approx glaucous gulls $>$ Atlantic puffins (Appendix 1; Figures 3 and 4).

Higher levels of HBCDDs in eggs of black-legged kittiwakes compared to herring gull and glaucous gull eggs is unexpected because herring gulls and glaucous gulls generally feed at a higher trophic level. The higher levels of HBCDD in black-legged kittiwakes can be related to differences in biotransformation capacities of HBCDD. Even in seabirds with similar diets, the levels and patterns of HOCs have been shown to differ substantially due to differences in presence and activity of biotransformation enzymes (Walker 1990; Borgå et al. 2004). For example, high levels of HOCs in glaucous gulls have been linked to the low metabolic capacity in this species (Henriksen et al. 2000).

7.2.3 Levels of HBCDDs

No clear north-south trend or differences between predatory birds and piscivorous birds can be found when comparing present results to previous studies of HBCDD in seabird eggs (Table 5).

Levels in eggs of glaucous gulls and black-legged kittiwakes from 2002/2003 were similar or higher than those reported in common guillemots from the Baltic Sea and glaucous gulls eggs from Bjørnøya, peregrine falcon eggs from South Greenland and Belgium little owl eggs.

The levels of HBCDD were, however, somewhat lower than those reported in Swedish peregrine falcon eggs and in Belgium common tern eggs.

Table 5. HBCDD concentrations (ng/g lw) reported in bird eggs in the scientific literature. Values are ranges or mean concentrations.

Species	Scientific name	HBCDD	Study area	Source
Peregrine falcon	<i>Falco peregrinus</i>	17	South Greenland	Vorkamp et al. 2005
		79-2400	South Sweden	Lindberg et al. 2004
		34-590	North Sweden	Lindberg et al. 2004
Glaucous gull	<i>Larus hyperboreus</i>	142	Bjørnøya	Verreault et al. 2004
Little owl	<i>Athene noctua</i>	20 and 50 (2 out of 40 analyzed eggs)	Belgium	Jaspers et al. 2004
Common tern	<i>Sterna hirundo</i>	1501	Belgium	Morris et al. 2004
Common guillemot	<i>Uria algae</i>	140	Baltic Sea	Sellström et al. 2003

7.2.4 Temporal trends of HBCDDs

There was a statistically significant increase in concentrations of α -HBCDD from 1983 to 2003 (Appendix 1; Figure 3). This increase is probably caused by enhanced emission of this compound (BSEF 2004).

The results of the present study were in contradiction with findings in a study of total-HBCDD in common guillemot eggs from the Baltic Sea between 1969 and 2001 (Sellström et al. 2003). In eggs of common guillemots, peak concentrations were reported in the mid-1970s, followed by a gradual decrease from the mid-1970s to the 1980s, an increase during the latter part of the 1980s and finally no significant change over the last 10 years (Sellström et al. 2003).

Furthermore, a temporal trend study (1986-2003) of peregrine falcon eggs from South Greenland reported a tendency towards a decrease in total-HBCDD levels during the study period (Vorkamp et al. 2005).

7.2.5 Risk assessment of HBCDDs

Subtle biological effects, neurobehavioral alterations and disruption of the thyroid hormone system have been associated with HBCDD exposure (Eriksson et al. 2002; Birnbaum and Staskal 2004). Furthermore, HBCDD seems to have an inhibitory effect on CYP1A's activity (Ronisz et al. 2004), which may eventually enhance the toxicity of other compounds, as levels of coplanar PCBs might, for example, increase. Nevertheless, most studies conducted so far have been on laboratory animals using technical products

of HBCDD in which the γ -isomer dominates. Thus, little is known about the biological effects associated to HBCDD exposure in free-ranging animals, including seabirds.

As for BDEs, not only the “precursor” compounds of HBCDDs may give rise to detrimental effects following HBCDD exposure. In fact, HBCDD has been demonstrated to produce degradation products during waste combustion processes such as polybromopolychlor-dibenzo-*p*-dioxins and dibenzofurans (Söderström and Marklund 2002; 2004).

There are few studies of the threshold levels for biological effects subsequent to HBCDD exposure. The studies available have shown conflicting results. For example, in two different studies reporting lowest observed effect levels (LOEL) and no observed adverse effect levels (NOAEL) in rats the threshold levels were reported to be 13 mg/kg/day and 1000 mg/kg/day, respectively (e.g., review Birnbaum and Staskal 2004). Furthermore, significant decreases of thyroid hormone levels were reported at 100 mg/kg/day in rats, whereas reproductive/developmental studies (of rats) suggested no developmental effects after dietary exposure to HBCDD of > 500 mg/kg/day (Birnbaum and Staskal 2004). As for BDEs, it is difficult to assess the likelihood that the seabirds (eggs) in the present study may have been at risk for adverse biological effects based on the above presented effect studies.

Table 6. Mean \pm standard error (se.), range (min-max) and median concentrations of hexabromocyclododecane (HBCDDs) (ng/g wet weight (ww)), cyclododeca-1,5,9-triene (CDT) and mercury (mg/kg ww) measured in eggs of herring gulls, Atlantic puffins and black-legged kittiwakes collected from Hornøya (a) and Røst (b) in 1983, 1993 and 2003. The number of samples above the detection limit is shown relative to the number of samples analyzed (N).

a) Hornøya

Species	Year		Lipid %	α -HBCDD	Lipid %	Hg
Herring gull	1983	Mean \pm se	11.0 \pm 1.9	1.8 \pm 0.4	7.0 \pm 0.2	0.1 \pm 0.0
		min-max	1.9 - 12.6	0.4 - 3.1	5.4 - 7.9	0.1 - 0.1
		N	5/5	5/5	5/5	5/5
		median	10.8	2.0	7.3	0.1
	1993	Mean \pm se	10.2 \pm 0.46	1.9 \pm 0.4	8.8 \pm 0.2	0.1 \pm 0.0
		min-max	9.8 - 10.9	1.6 - 2.2	8 - 9.7	0.0 - 0.1
		N	5/5	5/5	5/5	5/5
		median	9.9	1.9	9.1	0.1
	2003	Mean \pm se	9.7 \pm 1.2	9.3 \pm 1.9	9.7 \pm 0.2	0.1 \pm 0.0
		min-max	8.5 - 11.1	5.7 - 17.8	8.5 - 11.1	0.1 - 0.2
		N	5/5	5/5	5/5	5/5
		median	9.4	6.7	9.4	0.1
A. puffin	1983	Mean \pm se	12.6 \pm 1.4	1.9 \pm 0.4	8.2 \pm 0.2	0.1 \pm 0.0
		min-max	10.8 - 14.2	1.2 - 2.3	7.5 - 9.4	0.1 - 0.2
		N	5/5	5/5	5/5	5/5
		median	12.4	2.0	8.0	0.1
	1993	Mean \pm se	11.7 \pm 0.8	4.9 \pm 1.0	8.9 \pm 0.9	0.2 \pm 0.0
		min-max	10.8 - 12.5	4.0 - 5.6	7.9 - 10.3	0.2 - 0.2
		N	5/5	4/4	4/4	4/4
		median	11.7	5.2	8.8	0.2
	2003	Mean \pm se	13.7 \pm 1.9	10.1 \pm 2.0	13.7 \pm 1.9	0.1 \pm 0.0
		min-max	10.9 - 15.3	6.3 - 13.1	10.9 - 15.3	0.1 - 0.1
		N	5/5	5/5	5/5	5/5
		median	14.5	10.5	14.5	0.1
Kittiwake	1983	Mean \pm se	9.8 \pm 0.5	1.7 \pm 0.3	11.3 \pm 0.2	0.1 \pm 0.0
		min-max	9.2 - 10.5	1.2 - 2.4	10.3 - 12.3	0.1 - 0.2
		N	5/5	5/5	5/5	5/5
		median	9.7	1.5	11.3	0.1
	1993	Mean \pm se	9.6 \pm 0.4	3.0 \pm 0.7	9.6 \pm 0.4	0.1 \pm 0.0
		min-max	9.2 - 10.1	2.4 - 3.7	9.2 - 10.1	0.1 - 0.1
		N	5/5	5/5	5/5	3/3
		median	9.5	2.7	9.5	0.1
	2003	Mean \pm se	9.5 \pm 1.0	10.8 \pm 2.2	9.5 \pm 0.2	0.1 \pm 0.0
		min-max	8.7 - 11.1	7.9 - 12.7	8.7 - 11.1	0.1 - 0.1
		N	5/5	5/5	5/5	5/5
		median	8.9	11.5	8.9	0.1

b) Røst/(Hekkingen)

Species	Year		Lipid %	α -HBCDD	Lipid %	Hg
Herring gull	1983	Mean \pm se	9.5 \pm 0.7	1.4 \pm 0.3	7.9 \pm 0.3	0.1 \pm 0.0
		min-max	8.7 - 10.4	0.6 - 2.7	6.3 - 10.5	0.1 - 0.2
		N	5/5	5/5	5/5	5/5
		median	9.5	0.8	7.8	0.1
	1993	Mean \pm se	8.3 \pm 1.3	3.7 \pm 0.7	8.1 \pm 0.2	0.1 \pm 0.0
		min-max	6.9 - 10.1	2.4 - 5.3	6.6 - 9.3	0.1 - 0.1
		N	5/5	5/5	5/5	5/5
		median	8.3	3.1	8.6	0.1
	2003	Mean \pm se	10.4 \pm 4.1	11.6 \pm 2.3	10.8 \pm 4.1	0.1 \pm 0.0
		min-max	6.3 - 15.0	6.5 - 16.5	6.3 - 15.0	0.1 - 0.2
		N	5/5	5/5	5/5	5/5
		median	8.0	11.3	9.8	0.1
A. puffin	1983	Mean \pm se	12.2 \pm 0.9	1.1 \pm 0.2	10.3 \pm 0.1	0.2 \pm 0.0
		min-max	10.8 - 13.3	0.8 - 1.7	9.6 - 10.8	0.1 - 0.5
		N	5/5	5/5	5/5	5/5
		median	12.2	1.0	10.4	0.2
	1993	Mean \pm se	12.2 \pm 1.2	2.2 \pm 0.5	11.9 \pm 0.3	0.1 \pm 0.0
		min-max	10.8 - 13.5	1.6 - 2.9	9.6 - 13.2	0.0 - 0.1
		N	4/4	4/4	5/5	5/5
		median	12.3	2.1	12.8	0.1
	2003	Mean \pm se	13.7 \pm 2.8	6.1 \pm 1.2	13.7 \pm 0.6	0.1 \pm 0.0
		min-max	11.2 - 17.1	3.5 - 11.2	11.2 - 17.1	0.1 - 0.1
		N	5/5	5/5	5/5	5/5
		median	12.0	5.0	12.0	0.1
Kittiwake	1983	Mean \pm se	7.1 \pm 0.9	2.9 \pm 0.6	6.3 \pm 0.1	0.1 \pm 0.0
		min-max	6.2 - 8.2	0.7 - 7.9	5.4 - 7.3	0.1 - 0.1
		N	5/5	5/5	5/5	5/5
		median	6.7	2.0	6.3	0.1
	1993	Mean \pm se	9.4 \pm 1.1	7.1 \pm 1.4	9.5 \pm 0.1	0.1 \pm 0.0
		min-max	8.3 - 10.7	4.6 - 10.5	8.6 - 10.2	0.1 - 0.2
		N	5/5	5/5	5/5	5/5
		median	9.5	5.7	9.4	0.1
	2003	Mean \pm se	9.9 \pm 1.5	17.3 \pm 3.5	9.9 \pm 0.3	0.1 \pm 0.0
		min-max	7.3 - 11.0	11.7 - 26.5	7.3 - 11.0	0.0 - 0.1
		N	5/5	5/5	5/5	5/5
		median	10.5	17.0	10.5	0.1

Table 7. Mean \pm standard error (se.), range (min-max) and median concentrations of hexabromocyclododecane (HBCDDs) (ng/g wet weight (ww)), cyclododeca-1,5,9-triene (CDT) and mercury (Hg) (mg/kg ww) measured in eggs of glaucous gulls from Bjørnøya in 1997 and 2002. The number of samples above the detection limit is shown relative to the number of samples analyzed (N).

Species	Year		Lipid %	α -HBCDD	Lipid %	Hg
Glaucous gull	1997	Mean \pm se	9.0 \pm 0.0	2.3 \pm 0.2	8.5 \pm 0.6	0.5 \pm 0.0
		min-max	8.9 - 9.1	1.7 - 2.9	8.0 - 9.2	0.5 - 0.5
		N	3/3	3/3	3/3	3/3
		median	9.0	2.3	8.2	0.5
	2002	Mean \pm se	8.5 \pm 0.2	12.0 \pm 3.3	9.0 \pm 0.1	0.3 \pm 0.0
		min-max	8.0 - 9.2	5.2 - 23.4	8.9 - 9.1	0.3 - 0.4
		N	4/4	4/4	4/4	4/4
		median	8.2	7.5	9.0	0.3

7.3 Multivariate presentation

7.3.1 Redundancy analysis

A RDA of the relationship between BFRs, the three seabird species (herring gull, Atlantic puffin and black-legged kittiwake), sampling area (Hornøya and Røst) and sampling year (1983, 1993 and 2003) was conducted (Figure 3). Response variables (BFRs) are given as arrows. Explanatory variables, seabird species, sampling area and sampling year are given as nominal centroids.

BFRs point in the direction of increasing concentrations. The approximated correlation between variables in a RDA is positive when the angle between their arrows is minimal and negative when the angle is larger than 90 degrees. Centroids close to the coordination origin are less different than centroids that are widely scattered (Legendre and Legendre 1998).

From the RDA it is possible to interpret the following:

Generally

- a) There appear to be no clear spatial difference of BFRs as Hornøya and Røst are gathered close to the coordination origin.

BDEs

- b) Herring gull eggs contain higher concentrations of most BDEs compared to Atlantic puffin and black-legged kittiwake eggs, as the herring gull centroid correlates positively to most BDE congeners.
- c) Highest BDE concentrations are reported in 1993. The 1993 centroid points in the direction of increasing values of most BDEs.

HBCDD

- d) Highest concentrations of HBCDD in 2003. The 2003 centroid is closely correlated to HBCDD.

The variance in BFRs was significantly related to the explanatory variables (Monte Carlo, $F = 15.8$, $p = 0.002$). 54 % of the variance in concentrations of BFRs could be explained by the selected explanatory variables.

7.3.2 Principal component analysis

A PCA of BFRs, with species given as centroids (mean of samples) was conducted. Arrows of BFRs point in the direction of increasing values.

- a) Herring gull eggs contain proportionally higher levels of BDE-47 (to \sum BFR) compared to the other two species.
- b) Black-legged kittiwake eggs contain proportionally higher levels of HBCDD compared to herring gull and Atlantic puffin eggs.
- c) Atlantic puffin eggs contain proportionally higher levels of other BFRs than BDE-47 and HBCDD.

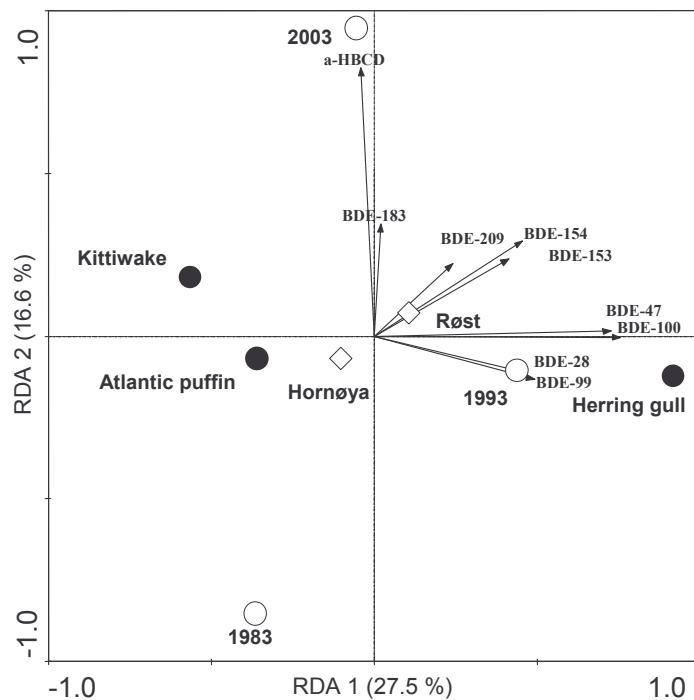


Figure 3. Redundancy analysis (RDA) of the relationship between brominated flame retardants (BFRs), the three seabird species (herring gull, Atlantic puffin and black-legged kittiwake), sampling area (Hornøya and Røst) and sampling year (1983, 1993 and 2003). Nominal explanatory variables are given as centroids (sampling area = diamond, species = filled circle, year = empty circle). The fraction of the unconstrained variance accounted for by each axis is given for the two ordination axes (brackets). $N = 89$.

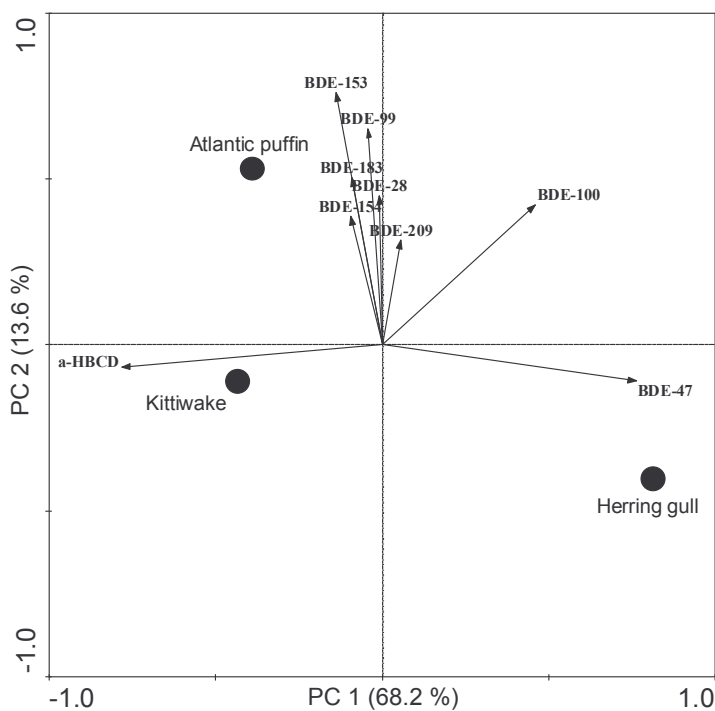


Figure 4. Principal component analysis (PCA) of brominated flame retardants (BFRs), with species as supplementary variables (given as centroids). The percentage variance in BFR concentrations explained by each axis is given in brackets. $N = 89$.

7.4 Mercury

For details of Hg concentrations see Tables 6 and 7.

7.4.1 Spatial trends of mercury

As for BDEs and HBCDD, there was no significant spatial variation of mercury concentrations in the present study.

7.4.2 Species-specific trends of mercury

Concentrations of mercury in eggs of Atlantic puffins were significantly higher than those in eggs of herring gulls (Appendix 1). However, lipid normalized levels of mercury in eggs of herring gulls sampled in 2003 were higher than in eggs of Atlantic puffins.

Higher levels of mercury in Atlantic puffin were unexpected because they generally feed at a lower trophic level than herring gulls. Mercury levels in eggs of glaucous gulls were approximately five times higher than those in eggs of the other seabirds.

7.4.3 Levels of mercury

Levels of mercury presented in the present study were generally similar to or lower than those reported in various species from North America, e.g., eggs of common loons, bald eagles, Forster and common terns, cormorants, great blue herons, wood ducks and herring gulls (Table 8).

Mercury levels in the present study were also lower than those reported in eggs of red-throated divers from Sweden, but similar to previously reported levels in eggs of black-legged kittiwakes from Norway in the 1970s (Table 8).

Table 8. Hg concentrations ($\mu\text{g/g ww}$) reported in bird eggs in the scientific literature. Values are ranges or mean concentrations.

Species	Scientific name	Hg	Study area	Source
Common loons	<i>Gavia immer</i>	0.2-0.9	North America	Evers et al. 2003
Bald eagles	<i>Haliaeetus leucocephalus</i>	0.5	North America	Evers et al. 2005
Herring gulls	<i>Larus argentatus</i>	0.6	North America	Evers et al. 2005
Wood ducks	<i>Aix sponsa</i>	0.1	North America	Evers et al. 2005
Great blue herons	<i>Ardea herodias</i>	0.1	North America	Evers et al. 2005
Forster tern	<i>Sterna forsteri</i>	0.2-0.9	North America	Scheuhammer et al. 2001
Common tern	<i>Sterna hirundo</i>	0.1-1.1	North America	Scheuhammer et al. 2001
Cormorants	<i>Phalacrocorax carbo</i>	0.3-0.8	Lake Huron	Scheuhammer et al. 2001
Cormorants	<i>Phalacrocorax carbo</i>	0.1-0.3	Lake Michigan	Scheuhammer et al. 2001
Red-throated divers	<i>Gavia stellata</i>	1.5-3.5	Sweden	Scheuhammer et al. 2001
Black-legged kittiwakes	<i>Rissa tridactyla</i>	<0.05-0.5	Norway	Fimreite et al. 1974

7.4.4 Temporal trends of mercury

No significant temporal trend in mercury levels was found in this study. This was similar to findings in a study by Appelquist et al. (1985) who reported stable levels of mercury in feathers of black-guillemots and Brünnich's guillemots (*Uria lomvia*) from Greenland over the past 100 years. In a study conducted on white-tailed sea eagles (feathers) (*Haliaeetus albicilla*) collected along the northern Norwegian coast from 1850 to 1995, large increases in mercury concentrations were seen in the 1950s and 1960s, after which concentrations decreased and then stabilized at about twice the background levels (AMAP 2005). Furthermore, no significant increase in mercury concentrations was found between the mid-1980s and the mid-1990s in liver samples of glaucous gulls from Greenland (Riget and Dietz 2000).

In contrast to the stable or declining concentrations of mercury reported in the European Arctic, studies from the Canadian Arctic have provided compelling evidence for increasing trends of mercury (Braune et al. 2005). For example, mercury concentrations nearly doubled between 1975 and 1998 in eggs of Brünnich's guillemots from the Canadian Arctic, whereas a 50 % increase was observed in eggs of northern fulmars (Braune et al. 2002).

7.4.5 Risk assessment of mercury

Laboratory studies have identified a number of effects of mercury exposure such as: effects of genetic and enzyme systems, decreased cardiovascular functions, altered blood parameters, inhibition of the immune response, reproductive failure, development damage, reduced growth, suppression of appetite, weight loss and decreased kidney functions (AMAP 2005). The toxic effects of mercury depend on the amount, its chemical form and the route of exposure. For example, organic mercury is more toxic than inorganic salts. Birds assimilate organic mercury (methylmercury, CH₃Hg) more readily than inorganic compounds. Egg Hg levels in seabirds are primarily in the methylmercury form (> 90 %) (Fimreite et al. 1974).

In the present study, mercury levels in one egg of the Atlantic puffin from Røst sampled in 1993, and in all the pooled egg samples of glaucous gulls sampled in 1997 at Bjørnøya (*N*=4), were nearly equal to the critical threshold level for biological effects (0.5 mg/kg ww) in eggs (Derome and Fairbrother 2005). Nevertheless, comparisons between species should be made cautiously as there are large differences in sensitivity to pollutant exposure between avian species. For example, seabirds are at least an order of magnitude less sensitive than freshwater birds (AMAP 2005). Furthermore, efficient detoxification strategies of mercury have been demonstrated in some birds. For example, Procellariiformes (e.g., Northern fulmars (*Fulmarus glacialis*)) demethylate organic mercury more efficiently than other seabirds, resulting in extremely high levels of inorganic mercury (Wolfe et al. 1998). Mercury is normally a problem only where the rate of the methylation of inorganic mercury is higher than the demethylation of organic mercury. Thus, high levels of total mercury do not necessarily result in adverse biological effects (Wolfe et al. 1998).

Finally, recent studies, including data of atmospheric mercury levels at the Zeppelin Station for Atmospheric Monitoring and Research (Ny-Ålesund, Svalbard), have revealed that the Arctic may become a global sink for atmospheric mercury during polar sunrise (spring/summer) (Berg and Aspmo 2003). Reactions involving sunlight and bromine remove elemental gaseous mercury from the atmosphere and transfer it to the ground as reactive mercury. Reactive mercury is efficiently converted to its bioavailable form and thus has the potential to accumulate to high levels in arctic animals, including breeding seabirds from the coast of Northern Norway, during the arctic summer. It is unknown if the mercury depletions in air are caused by a changing climate or levels of anthropogenic pollutants (AMAP 2005). If so, arctic animals may be at risk for adverse biological effects as mercury concentrations would follow these trends.

8. Conclusions

Spatial trends

- There were no spatial trends in the dataset for BFRs, CDT and mercury.

Species-specific differences

- Levels of BDEs were higher in eggs of herring gulls and glaucous gulls than in eggs of Atlantic puffins and black-legged kittiwakes.
- Levels of α -HBCDD were highest in the eggs of black-legged kittiwakes and decreased in the order herring gulls \approx glaucous gulls $>$ Atlantic puffins.
- Concentrations of mercury in eggs of glaucous gulls were higher than in eggs of Atlantic puffins, herring gulls and black-legged kittiwakes.

Levels

- BDEs, with the exception of BDE-183 and BDE-209, the α -HBCDD isomer and mercury were above the limits of detection in all samples.
- Levels of BDE-183 and BDE-209 were detected in 43-86 % and 20-42 % respectively, of the analyzed eggs.
- Levels of β -HBCDD, γ -HBCDD and CDT were below the limits of detection in all samples.

Temporal trends

- Levels of most BDEs increased from 1983 to 1993. Only BDE-153, BDE-154 and BDE-183 levels increased between 1983 and 2003. For BDE-209 no statistical difference was found between the sampling years, although concentrations of BDEs in eggs of herring gulls and black-legged kittiwakes tended to be higher in 2003 than in the other sampling years.
- Levels of α -HBCDD increased from 1983 to 2003.
- No significant temporal trend of mercury was found in this study.

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11. Appendix

Appendix 1. Generalized linear models with 2000 parametric bootstrap replicates ($\log(\text{BDE}/\text{HBCDD}/\text{mercury}) \sim \text{herring gull} + \text{Hornøya} + 1983 + (\text{lipid} \% \text{ covariate})$) were conducted on the dataset. Bootstrap estimates for species-, spatial- and temporal- trends of brominated diphenyl ethers, hexabromocyclododecane and mercury, in addition to bias corrected and accelerated confidence intervals (BCa) (used to check whether the coefficients of the different factors were different from zero) are given in the table. $N = 89$. Bold = significant. Italic = marginal significant.

	BDE-28	BDE-47	BDE-100	BDE-99	BDE-154	BDE-153	BDE-183	ΣBDE	HBCDD	mercury
(Intercept)	-0.4	3.0	1.0	0.2	-1.5	-1.4	-1.6	3.1	-0.4	-2.1
Kittiwake	-0.4	-1.2	-1.4	-0.7	-0.7	-0.3	0.3	-1.1	0.7	0.0
BCa	-0.6 - -0.1	-1.5 - -0.9	-1.7 - -1.1	-1.1 - -0.3	-1.0 - -0.4	-0.7 - -0.1	-0.4 - 0.9	-1.3 - 0.8	0.4 - 0.9	-0.1 - 0.5
Atl. puffin	-0.2	-1.7	-1.2	-0.4	-0.2	-0.4	0.2	-1.3	-0.1	0.2
BCa	-0.5 - 0.3	-2.1 - -1.2	-1.6 - -0.8	-0.7 - 0.1	-0.6 - 0.2	-0.7 - 0.1	-0.5 - 1.2	-1.6 - -1.0	-0.4 - 0.2	0.2 - 0.8
Røst	0.2	0.1	0.2	0.1	0.3	0.4	0.2	0.2	< 0	-2.1
BCa	-0.1 - 0.4	-0.1 - 0.4	-0.2 - 0.5	-0.1 - 0.4	> 0 - 0.5	0.2 - 0.6	-0.3 - 0.7	> 0 - 0.5	-0.3 - 0.1	-0.2 - 0.2
1993	0.7	0.5	0.5	0.9	0.9	1.1	0.6	0.6	1.0	-0.2
BCa	0.4 - 0.9	0.2 - 0.8	0.2 - 0.8	0.5 - 1.2	0.6 - 1.2	0.7 - 1.4	-0.1 - 1.2	0.3 - 0.9	0.7 - 1.3	-0.6 - 0.0
2003	< 0.1	0.2	0.2	-0.1	0.8	0.7	1.5	0.2	1.8	-0.2
BCa	-0.3 - 0.1	-0.1 - 0.5	-0.1 - 0.5	-0.3 - 0.2	0.4 - 1.1	0.4 - 1.0	0.9 - 2.1	< 0 - 0.5	1.6 - 2.1	-0.4 - 0.1

Appendix 2. Median and range (min-max) concentrations of BDEs (ng/g lipid weight (lw)) measured in eggs of herring gulls, Atlantic puffins and black-legged kittiwakes collected from Hornøya in 1983, 1993 and 2003, in addition to glaucous gulls from Bjørnøya in 1997 and 2002.

1983

	Herring gull			Atlantic puffin			Black-legged kittiwake					
	Hornøya			Hornøya			Hornøya			Røst		
	Median	Min	Max	Median	Min	Max	Median	Min	Max	Median	Min	Max
BDE-28	7.6	4.6	14.6	4.8	3.4	13.0	3.8	2.3	5.0	4.1	3.2	6.2
BDE-47	371	201	618	128	121	649	30.2	18.5	41.3	58.3	52.4	71.5
BDE-100	52.6	26.1	88.9	17.4	15.4	106	10.1	6.4	15.8	7.1	6.4	7.8
BDE-99	41.0	15.6	70.2	13.4	12.0	42.7	21.3	12.8	30.3	12.2	11.1	15.0
BDE-154	4.4	2.5	11.9	2.5	2.0	10.0	4.5	3.8	9.1	2.3	0.8	23.1
BDE-153	8.1	2.6	14.6	2.1	1.8	17.7	3.2	2.5	6.8	3.2	1.7	9.8
BDE-183	nd	nd	34.7	-	-	-	nd	2.0	2.6	5.7	2.0	37.9
BDE-209	nd	nd	2.2	nd	nd	1.4	-	-	-	nd	nd	1.6
ΣBDE	510	253	814	163	160	838	134	115	158	101	77.7	149
							73.0	48.4	111			
										208	134	267

1993/1997

	Herring gull			Atlantic puffin			Black-legged kittiwake			Glaucous gull											
	Hornøya			Røst			Hornøya			Bjørnøya											
	Median	Min	Max	Median	Min	Max	Median	Min	Max	Median	Min	Max									
BDE-28	12.9	6.3	14.2	27.0	19.6	34.0	15.0	12.1	18.3	10.2	6.2	11.7	3.6	2.66	4.6	13.9	13.6	34.3	8.1	7.7	9.0
BDE-47	325	173	375	578	446	1050	121	72.8	133	73.0	53.5	87.2	62.9	53.2	99.3	232	179	617	367	356	374
BDE-100	57.1	20.6	61.6	79.8	69.7	174	25.5	17.7	31.7	20.1	17.0	25.9	8.1	6.94	14.1	28.7	23.4	89.2	50.8	50.6	53.1
BDE-99	54.5	18.3	118.3	62.4	34.6	474.1	53.3	42.4	70.5	64.0	52.3	67.3	19.4	17.1	30.2	65.7	43.1	276	48.4	48.0	50.3
BDE-154	9.0	5.4	10.4	16.8	15.5	31.5	12.6	9.4	15.2	12.3	11.4	13.3	2.3	2.22	6.48	10.3	6.6	20.5	12.8	12.2	14.9
BDE-153	8.7	4.6	12.6	21.1	9.8	45.2	9.5	7.5	40.4	13.5	12.1	23.9	4.2	4.03	14.6	17.2	10.6	113	20.6	17.8	21.0
BDE-183	4.2	2.5	27.2	4.4	2.6	8.0	3.4	1.9	12.7	3.2	2.1	51.0	0.8	0.3	17.8	3.1	2.3	29.0	11.7	0.9	15.6
BDE-209	0.9	0.5	5.2	nd	nd	1.5	nd	nd	6.2	nd	nd	1.2	nd	nd	1.0	nd	nd	2.5	1.3	0.8	1.7
ΣBDE	499	248	558	782	646	1811	258	165	404	208	155	259	104	87.3	171	351	279	1182	523	494	535

2002/2003

	Herring gull			Atlantic puffin			Black-legged kittiwake			Glaucous gull											
	Hornøya			Røst			Hornøya			Bjørnøya											
	Median	Min	Max	Median	Min	Max	Median	Min	Max	Median	Min	Max									
BDE-28	7.9	5.7	8.5	8.4	1.1	9.9	3.7	3.4	6.7	3.2	2.1	4.4	4.1	2.6	6.7	12.0	5.1	12.9	5.2	4.6	11.2
BDE-47	322	302	654	512	67.9	640	38.9	32.1	77.0	30.7	20.7	56.6	84.8	53.6	159	195	98.4	235	331	297	497
BDE-100	75.1	62.0	91.2	71.5	16.1	105	9.7	8.3	14.8	8.6	7.1	18.1	8.8	6.5	20.9	22.3	10.4	25.9	50.4	47.3	80.5
BDE-99	33.9	27.7	107	28.7	14.0	76.4	17.1	14.7	22.8	17.5	13.3	37.7	17.0	8.0	36.2	13.6	5.7	20.7	63.9	38.7	90.9
BDE-154	13.4	10.5	25.4	19.7	5.8	22.8	8.3	6.1	10.1	8.3	1.3	17.7	4.7	2.6	9.4	7.2	4.6	10.7	11.0	10.7	29.9
BDE-153	8.6	7.5	33.9	18.8	10.9	55.7	5.1	3.6	7.7	6.4	4.5	9.4	7.7	5.1	17.7	8.4	4.8	14.9	63.2	18.4	70.1
BDE-183	-	-	-	21.6	18.5	103	11.4	6.8	11.7	-	-	-	nd	nd	55.0	31.0	9.7	41.2	3.5	1.5	10.4
BDE-209	7.1	4.8	128	2.7	0.9	95.7	nd	nd	0.0	-	-	-	nd	nd	2.2	1.5	1.5	1.9	8.9	0.8	32.2
ΣBDE	464	425	821	720	305	1035	84.5	74.2	143	75.4	56.0	144	143	84.3	267	297	166	308	494	462	822

Appendix 3. Median and range (min-max) concentrations of HBCDD (ng/g lipid weight (lw)) and Hg (mg/kg lw) measured in eggs of herring gulls, Atlantic puffins and black-legged kittiwakes collected from Hornøya in 1983, 1993 and 2003, in addition to glaucous gulls from Bjørnøya in 1997 and 2002.

Year	Species	Area	HBCDD		Hg	
			Median	Min - Max	Median	Min - Max
1983	Herring gull	Hornøya	15.5	4.7 - 28.1	1.0	0.8 - 1.9
	Herring gull	Lofoten	7.9	6.3 - 20.4	1.7	0.7 - 3.3
	Puffin	Hornøya	14.4	11.1 - 203	1.9	1.2 - 2.7
	Puffin	Lofoten	8.4	7.8 - 28.5	2.0	1.1 - 4.6
	Kittiwake	Hornøya	15.9	12.3 - 54.9	0.9	0.8 - 1.4
	Kittiwake	Lofoten	32.8	10.0 - 168	1.9	0.8 - 2.2
1993	Herring gull	Hornøya	18.0	16.1 - 18.2	0.6	0.5 - 1.0
	Herring gull	Lofoten	37.6	34.7 - 52.0	1.4	0.2 - 1.6
	Puffin	Hornøya	41.7	31.8 - 85.3	2.5	2.0 - 3.0
	Puffin	Lofoten	16.3	12.7 - 13.7	0.6	0.2 - 0.8
	Kittiwake	Hornøya	27.0	25.2 - 39.5	1.3	1.1 - 1.4
	Kittiwake	Lofoten	65.0	55.4 - 26.4	1.2	0.7 - 3.4
2003	Herring gull	Hornøya	67.2	62.0 - 25.5	0.7	0.7 - 1.5
	Herring gull	Lofoten	110	71.3 - 136	1.0	0.3 - 2.6
	Puffin	Hornøya	72.3	57.5 - 124	0.8	0.6 - 1.0
	Puffin	Lofoten	44.2	21.6 - 65.1	0.7	0.7 - 1.0
	Kittiwake	Hornøya	114.3	91.6 - 110	0.8	0.6 - 1.3
	Kittiwake	Lofoten	161.3	106 - 362	1.0	0.7 - 1.3
1997	Glaucous gull	Bjørnøya	25.3	19.1 - 32.2	5.2	5.2 - 5.3
2002	Glaucous gull	Bjørnøya	81.4	62.8 - 292	3.2	3.0 - 4.7



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Forfatter(e) Lisa Bjørnsdatter, Geir Wing Gabrielsen, Jonathan Verreault, Rob Barrett, Anuschka Polder, Janneche Utne Skåre, Elisabeth Lie
Tittel - norsk og engelsk Temporelle trender av bromerte flammehemmere, cyclododeca-1,5,9-triene og kvikksølv i egg av fire arter sjøfugl fra Nord-Norge og Svalbard Temporal trends of brominated flame retardants, cyclododeca-1,5,9-triene and mercury in eggs in four seabird species from Northern Norway and Svalbard
Sammendrag – summary The present study investigated possible species and regional differences, in addition to temporal trends of brominated flame retardants (BFRs), cyclododeca-1,5,9-triene and mercury in seabird eggs. Eggs were collected from herring gulls (<i>Larus argentatus</i>), Atlantic puffins (<i>Fratercula arctica</i>) and black-legged kittiwakes (<i>Rissa tridactyla</i>) in 1983, 1993 and 2003 at Røst and Hornøya (Northern Norway). Additionally, eggs of glaucous gulls (<i>Larus hyperboreus</i>) were collected in 1997 and 2002 at Bjørnøya (Svalbard). Current levels and congener/isomer patterns were compared to previously reported levels in eggs of free-ranging seabirds.

4 emneord Sjøfugl egg, BFR, Hg, tidstrender	4 subject words Seabird eggs, BFRs, Hg, temporal trends
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