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Abstract

Eggs from Brünnich's guillemots *Uria lomvia* collected in Kongsfjorden in 1993 (reanalysed from Barrett et al. 1996), 2002 and 2003 and in Bjørnøya in 2003 and 2007 were analysed in order to investigate the level of contaminant concentrations in the two areas and the temporal trends of the different contaminants in the time range from 1993 to 2007. All eggs were analysed for a range of contaminants, including organochlorines (OCs), brominated flame retardants (BFRs) and perfluorinated alkyl substances (PFASs). The eggs from Kongsfjorden in 2002 and 2007 and from Bjørnøya in 2003 and 2007 were also analysed for a suite of PAHs, organotin, trace elements and stable isotopes. The contaminant concentrations were generally within the range seen in eggs from other seabird species. The temporal trends of decreasing concentrations of OCs and BDEs were also confirmed. The trends of PFASs were less clear, and varied between compounds. The present study indicates that the concentrations of PFOS are decreasing, whereas other PFASs, such as PFUnA and PFTriA are increasing.

Sammendrag – Summary in Norwegian

Polarlomvi *Uria lomvia* er en alkefugl som er noe større enn lomvi *Uria aalge*. Det foregår ikke systematisk overvåking av miljøgifter i arten, men det ble tatt prøver (egg) i 1993 og igjen i 2002/2003 og 2007. Prøvene ble tatt på Bjørnøya og Kongsfjorden på Svalbard og analysert for organokloriner (OC), bromerte flammehemmere (BFR), perfluorerte alkylstoffer (PFAS), organiske tinnforbindelser, PAH-er og et utvalg av metaller.

Prøvene viser en reduksjon i konsentrasjonen av de fleste organohalogener fra 1993 til 2002/2003 med en ytterligere reduksjon til 2007. For organoklorinene og de bromerte flammehemmerene ser det generelt ut som at nivåene i Kongsfjorden 2002 og Bjørnøya 2003 og i Kongsfjorden 2007 og Bjørnøya 2007 er relativt like. Reduksjonen fra 2002/2003 til 2007 er omtrent lik i begge områder. Flertallet av organoklorinene (pesticider, toksafener, PCB-er), med noen få unntak, var signifikant lavere fra 1993 til 2002/2003 og fra 2002/2003 til 2007 (figur 3A). Alle organoklorinene med unntak av HCB og β-HCH var signifikant lavere i 2007 sammenlignet med 1993. Av de bromerte flammehemmerene var de polybromerte difenyl-eterene (PBDE) også signifikant lavere fra 1993 til 2007 (figur 3B). Forskjellene mellom 1993 og 2002/2003 var signifikant for noen av PBDE-ene, mens ingen var signifikant forskjellige mellom 2002/2003 og 2007. Dette indikerer at nivåene av bromerte flamme- hemmere begynner å stabilisere seg. HBCD-konsentrasjonene var på et stabilt nivå gjennom hele tidsperioden (figur 3B).

For de fleste perfluorerte alkylstoffene ble det funnet signifikante forskjeller mellom Kongs-fjorden og Bjørnøya. Disse ble derfor behandlet hver for seg statistisk. PFAS-nivåene var generelt høyere på Bjørnøya enn i Kongsfjorden ved sammenligning innenfor de to tids-periodene (2002/2003 og 2007). De perfluorerte alkylstoffene var den gruppen av forbindelser med størst variasjon fra år til år og den eneste av organohalogenene hvor det ble funnet en økning i konsentrasjoner for noen stoffer i tidsperioden. For Kongsfjorden var konsentra-sjonene av de perfluorerte karboksylsyrene (PFNA, PFDcA, PFUnA, PFDoA, PFTriA, PFTeA og PFPeDA) generelt sett høyest i 2002 og lavest i 1993. Konsentrasjonene av de perfluorerte sulfonatene (PFOSA, PFHxS og PFOS) var generelt sett høyest i 1993 og lavest i 2007. Et unntak blant sulfonatene var PFDcS som var høyest i 2007 og 2002 og lavest i 1993. Forskjellene var signifikante for de fleste av de perfluorerte alkylstoffene mellom 1993 og 2002, men bare for et fåtall mellom 2002 og 2007. For Bjørnøya var konsentrasjonene av perfluorerte karboksylsyrer generelt sett høyere i 2007 enn i 2003 med signifikant forskjellige verdier for PFDcA, PFUnA, PFDoA, PFTriA, PFTeA og PFPeDA. Konsentrasjonene av sulfonater var høyere i 2003 sammenlignet med 2007, med signifikante forskjeller for PFOSA og PFOS.

Økningen i konsentrasjoner av perfluorerte karboksylsyrer, kombinert med reduksjonen av PFOS-konsentrasjoner i samme tidsperiode, leder til at PFOS ikke var det dominerende perfluorerte alkylstoffet i polarlomviegg fra Bjørnøya i 2007 (figur 4). Både PFUnA og PFTriA var til stede i

omtrent dobbelt så høye konsentrasjoner som PFOS og disse to for-bindelsene utgjør omtrent 2/3 av ΣPFAS. For 5-10 år siden var PFOS den dominerende perfluorerte forbindelsen i biotiske prøver (Houde et al. 2006). PFOS ble faset ut av hoved-produsenten (3M, USA) i 2000-2002, og nedgangen i PFOS-konsentrasjoner i polarlomviegg kan være et resultat av reduksjonen i produksjon.

Egg fra Kongsfjorden i 1993 ble ikke analysert for PAH, organiske tinnforbindelser, metaller og stabile isotoper. Ingen av de analyserte organiske tinnforbindelser ble detektert over deteksjonsgrensen i noen prøver. PAH-er ble kun detektert over deteksjonsgrensen i et fåtall prøver. PAH-er ble detektert i flere prøver fra Bjørnøya i 2003 og fra Kongsfjorden i 2002 enn i de andre. ΣPAH-konsentrasjonene var signifikant høyere i 2002/2003, sammenlignet med i 2007.

Metallanalyse viste ikke noen klare generelle trender. Cadmium-nivåene var signifikant lavere i 2007 enn i 2002/2003 og nivåene var omtrent like i de to områdene. For kvikksølv var konsentrasjonene signifikant høyere i Kongsfjorden enn på Bjørnøya. Kvikksølvkonsentra-sjonene ble redusert mellom 2002/2003 og 2007 i begge områder (figur 6), men nedgangen var bare signifikant for Kongsfjorden (hvor ble konsentrasjonen nesten ble halvert). Ingen signifikante forskjeller ble funnet for bly-konsentrasjonene, verken mellom områdene eller over tid.

Konsentrasjonene av alle analyserte stoffer var generelt sett sammenlignbare med tidligere rapporterte konsentrasjoner i sjøfuglegg i Arktis og Barentshavet, og ofte i det lavere sjikt sammenlignet med disse (AMAP 2004, Helgason et al. 2008). Miljøgiftnivåene som er funnet i egg fra polarlomvi ligger under grenseverdier for effekter for alle stoffer som er vurdert.

Det er verdt å merke seg at målinger av nitrogenisotoper, et mål på trofisk nivå, viser signifikant høyere nivåer i Kongsfjorden enn på Bjørnøya og signifikant høyere nivåer i 2002/2003 sammenlignet med 2007 (figur 7). Dette kan virke forstyrrende på tolkningen av tidstrendene i denne studien, da nivåer av miljøgifter henger tett sammen med trofisk nivå og de høyere konsentrasjonene vi ser i 2002/2003 kan være, helt eller delvis, et resultat av at disse individene hadde spist mat som var på et høyere trofisk nivå sammenlignet med de fra 2007.

Contents

Introduction

Eggs of seabirds have been used to monitor contamination in the Arctic marine environment for decades (Barrett et al., 1996; Braune et al., 2001). The contaminants are being transferred to the egg during egg formation, and thus represent the maternal levels of contamination at the time of laying (Verreault et al., 2006).

There are few local sources of contamination within the Arctic. Thus, anthropogenic contaminants originating in temperate regions in the northern hemisphere reach the Arctic through various routes, such as atmospheric and oceanic transport (Oehme, 1991; Burkow and Kallenborn, 2000). Production and use of DDTs and PCBs were among the first pesticides and industrial chemicals to be restricted and banned after it was discovered that they represented a serious threat to human health and the environment in the 1970s, and they were followed by others (Fiedler, 2000). Decreasing organochlorine (OC) concentrations were observed during the 1980s and 1990s, however, the declines in organochlorine concentrations in Arctic biota are slow because of global cycling and long half-lives (AMAP, 2004).

As the OC concentrations started levelling off and decreasing, new emerging compound classes were detected in increasing concentrations, such as the brominated flame retardants (BFRs). AMAP (2004) estimated that if the same rate of increase of polybrominated diphenyl ethers (BDEs) and declines in PCBs were to continue, BDEs would reach parity with PCBs in Canadian Arctic ringed seals *Phoca hispida* sometime between 2015 and 2025. However, penta- and octa-BDE mixtures are now formally banned in the European Union (EU) (BSEF, 2009). Following the voluntary withdrawal and later formal ban, some temporal trend studies are reporting that the BDE concentrations are stabilising or decreasing (Sellström et al., 2003; Knudsen et al., 2005), whereas others document still increasing concentrations in the Arctic (Law et al., 2003; de Wit et al., 2006; Braune et al., 2007).

A second emerging compound class is the perfluorinated alkyl substances (PFASs), for which reliable analytical methods for quantification in environmental media have only recently been developed, despite being widespread in use for about 50 years (Kannan et al., 2001; Prevedouros et al., 2006). PFASs are a group of chemicals characterised by carbon chains of variable lengths where all hydrogen atoms have been replaced by fluorine. The many strong carbon-fluorine bonds result in virtually indestructible compounds, which are expected to redefine the term persistence as used about organic contaminants (AMAP, 2004). PFASs have been found widespread in biota, including in the Arctic (Giesy and Kannan, 2001; Tomy et al., 2004; Houde et al., 2006). Perfluorooctane sulfonate (PFOS), the dominating PFAS in biota (Martin et al., 2004; Houde et al., 2006), has been voluntarily phased out by one of the main manufacturers (AMAP, 2004) and a risk assessment is in process in the European Union (Miljøstatus, 2009). Temporal trend studies show increasing concentrations of PFASs, however a few indicate that the concentrations of PFOS and PFOA are starting to level off (Bossi et al., 2005; Holmström et al., 2005; Verreault et al., 2007).

On the contrary to the above-mentioned anthropogenic contaminants, trace elements including heavy metals are naturally occurring substances. However, anthropogenic activities such as mining and metal processing may increase environmental levels of trace elements (Walker et al., 2001). Thus, trace element contamination of the environment reflects both natural sources and anthropogenic activities. Some trace elements, such as cobalt (Co), copper (Cu), manganese (Mn), molybdenium (Mo), selenium (Se) and sink (Zn), are essential, thus deficiency produces functional or structural abnormalities. However, in higher concentrations they have potential for toxicity (Goyer and Clarkson, 2001).

Brünnich's guillemot *Uria lomvia* is one of the species in the Barents Sea which is monitored for environmental contaminants. The aim of this study was to provide data on levels of contaminants in two different areas (Kongsfjorden and Bjørnøya) and to look into temporal trends of the different contaminants with samples from 1993, 2002/2003 and 2007.

Materials and methods

Sampling procedures

A total of 25 Brünnich's guillemot eggs were sampled from Kongsfjorden and Bjørnøya in different years; 1993, 2002 and 2007 for Kongsfjorden and 2003 and 2007 for Bjørnøya (figure 1). Five eggs were sampled for each year and location, giving a total of 5 groups with 5 replicate eggs which were analysed individually. After sampling, the eggs were kept frozen until analyses. The eggs were weighed, and width and length were measured with a still calliper. The eggshell was thoroughly removed and whole eggs were homogenised with an Ultra-turrax T25 (Janke & Kunkel IKA-Labortechnik, Staufen, Germany). Homogenate was separated into aliquots for different analyses and stored in glass vials or cryo tubes at -20 °C until analysed.

The eggs from Kongsfjorden in 1993 have previously been assessed for contaminant levels (OCs and Hg) (Barrett et al., 1996) and the stored samples were reanalysed in this study to enable comparisons with eggs from Kongsfjorden and Bjørnøya in 2002/2003 and 2007 and to broaden the range of compounds analysed.

Figure 1 Map of Svalbard. Brünnich's guillemot *Uria lomvia* eggs were sampled from Kongsfjorden and Bjørnøya.

Analyses

All eggs were analysed for a suite of OCs, BFRs and PFASs. Eggs from Kongsfjorden in 2002 and 2007 and from Bjørnøya in 2003 and 2007 were also analysed for a suite of PAHs, organotin and trace elements. The organochlorine pesticides (OCPs) analysed and quantified were DDTs (*p,p'*-DDE, *p,p'*-DDT), chlordanes (oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, *cis*-nonachlor), HCHs (α-, β- and γ-), mirex, HCB and toxaphenes (CHB-26, -40, -41, -44, -50 and -62). Other OCs analysed were PCBs (PCB-28, - 47, -52, -66, -74, -99, -105, -114, -118, -123, -128, -137, -138, -141, -151, -153, -156, -157, -170, -180, -183, -187, -189, -194 and -206). The BFRs analysed and quantified were HBCD (sum of α-, β- and γ-HBCD) and BDEs (BDE-28, -47, -99, -100, -153, -154, 183, -206, -207, -208 and -209). The PCB and BDE congeners follow the numbering given in Ballschmiter and Zell (1980), later adapted by the International Union of Pure and Applied Chemistry (IUPAC).

The PFASs analysed and quantified were perfluorohexanoate (PFHxA), perfluoroheptanoate (PFHpA), perfluorooctanoate (PFOA), perfluorononanoate (PFNA), perfluorodecanoate (PFDcA), perfluoroundecanoate (PFUnA), perfluorododecanoate (PFDoA), perfluorotride-canoate (PFTriA), perfluorotetradecanoate (PFTeA), perfluoropentadecanoate (PFPeDA), perfluorooctane sulfonamide (PFOSA), perfluorobutane sulfonate (PFBS), perfluorohexane sulfonate (PFHxS), perfluorooctane sulfonate (PFOS) and perfluorodecane sulfonate (PFDcS). The PAHs analysed and quantified were naphtalene, phenanthrene, anthracene, acenaphtylene, acenaphtene, fluorene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, benzo[ghi]perylene and dibenzo[a,h]anthracene.

The organotin compounds analysed were monobutyltin (MBT), dibutyltin (DBT), tributyltin (TBT), monophenyltin (MPhT), diphenyl-tin (DPhT) and triphenyltin (TPhT). The following trace elements were analysed and reported; selenium (Se), cadmium (Cd), molybdenum (Mo), tin (Sn), mercury (Hg), thallium (Tl), lead (Pb), lithium (Li), vanadium (V), chromium (Cr), manganese (Mn), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn), antimony (Sb) and arsenic (As). Furthermore, stable isotope ratios ($\delta^{15}N$ and $\delta^{13}C$) were determined.

Analyses of OCs and BFRs

Analyses of OCs and BFRs were carried out at the Laboratory of Environmental Toxicology at the Norwegian School of Veterinary Science (Oslo, Norway). Lipids were extracted twice from egg homogenate by acetone/cyclohexane extraction. Lipid content was determined gravimetrically. Extracts were treated twice with sulphuric acid for sample clean up. An aliquot for toxaphene analyses required further separation on silica columns. Finally, contaminants were separated and quantified using high resolution gas chromatographs (GC) coupled to mass spectrometer (MS) or electron capture detection (ECD). More details on the chromatographic separation and equipment are given in Murvoll et al. (2006) for OCs, in Andersen et al. (2006) for toxaphenes and in Sørmo et al. (2006) for BFRs.

The laboratory is accredited by Norwegian Accreditation (Kjeller, Norway) according to NS-EN ISO/IEC 17025, test 137, and the analytical quality of the laboratory has been approved in several intercalibration tests. As standard procedure, recoveries of spiked samples, blanks and reference samples were analysed in each series and acceptable results were achieved.

Analyses of PFASs

PFAS analyses were conducted by the Analytical Environmental Chemistry Unit at the Stockholm University (Sweden). Samples were extracted twice from egg homogenates with acetonitrile in an ultrasonic bath. Concentrated extracts went through clean-up on graphitised carbon and acetic acid. Clean extract was added to ammonium acetate and precipitation followed. High performance liquid chromatography coupled to high resolution mass spectrometry (HPLC-HRMS, for sulfonates) or tandem mass spectrometry (MS-MS, for carboxylates) was applied. More details on the extraction procedure and quantification are given in Verreault et al. (2007).

For quality assurance, recovery rates of the stable isotope mass-labelled internal standards were determined, one sample was analysed twice and a fish tissue sample used in an interlaboratory comparison study analysed with the samples. Acceptable results were achieved.

Analyses of PAHs

PAH analyses were conducted by Unilab Analyse (Tromsø, Norway). Homogenised material was added methanol (100 ml) and potassium hydroxide (3 g) together with a solution containing 0.5 µg/ml of the following internal standards; naphthalene-d_8, biphenyl-d_{10}, anthracene-d_{10}, fenanthrene-d_{10}, pyrene-d_{10}, chrysene-d_{12} and perylene-d_{12}. The solution was left to reflux for 1.5 h, before extraction with pentane. The pentane extract was concentrated using a rotavapor.

Clean up of the extracts were performed on extraction columns with 500 mg silica. The PAH compounds were eluted with pentane, followed by dichloromethane. The extract was evaporated almost to dryness under nitrogen before clean up on gel permeation chromatography (GPC). Next, a solvent change was performed before instrument analysis. The separation and quantification of aromatic and bicyclic, aliphatic hydrocarbons was performed using GC-MS (Hewlett-Packard).

Analyses of organotin

Organotin analyses were conducted by the Norwegian Institute for Water Research (NIVA) (Oslo, Norway). Homogenate went through saponification followed by an adjustment of pH to 4-5. The components were ethylated and extracted into hexane. The extract was cleaned on basic alumina before analyses by GC/MS in SIM mode. The GC was equipped with a 30 m column with a stationary phase of 5% phenyl polysiloxane (0.25 mm i.d. and 0.25 µm film thickness), and the injection was pulsed splitless. The initial column temperature was 50 °C, which was raised to 290 °C stepwise during a 20 min period. Quantification of individual components was performed by using an internal standard.

Certified reference material was analysed together with the samples for quality control and was within ± 30% of certified value. The concentrations of the different compounds in the standards ranged from 1.5 - 4,500 ng/ml. No organotin compounds were detected in any of the samples. One of the samples was spiked with 10 ng of all components. All components were detected and were within ± 30% of the added amount. Thus, the eggs contained no detectable amount of organotin compounds.

Analyses of trace elements

Freeze-dried homogenate samples were added 50% HNO_3, giving a final volume of 50 ml. The biological material was digested using an UltraClave (Milestone Inc., Bergamo, Italy) using a pre-set temperature profile. The trace elements were quantified using high resolution inductive coupled plasma mass spectrometry (HR-ICP-MS; Element 2, Thermo Electronics). The measurements were verified against certified reference material.

Analyses of stable isotopes

Freeze-dried homogenate samples were analysed for stable isotope ratios ($\delta^{15}N$ and $\delta^{13}C$) at the Institute for Energy Technology (Kjeller, Norway). Lipids were removed by Soxhlet extraction with dichloromethane added 7% methanolin. The sample was then dried at 80 °C before rinsing with 2 N HCl to remove traces of carbonates. Next, the sample was rinsed with distilled water and dried at 80 °C, before combustion with O_2 and Cr_2O_3 in a Carlo Erba NCS Elemental Analyser.

Finally, the combustion products were separated on a Poraplot Q column and $\delta^{15}N$ and $\delta^{13}C$ were determined on a Micromass Optima mass spectrometer. International standards, Pee Dee Belemnite (PDB: USGS 24) for $\delta^{13}C$ and atmospheric air (IAEA-N-1 and 2) for $\delta^{15}N$, were generally run for each 10 samples. A detailed description of the method is given in Søreide et al. (2006).

Statistical analyses

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

For calculations of mean ± standard deviation (SD) concentrations only values above the respective detection limits were included, denoted by a lower sample size (n). Similarly, only values above the detection limit were included when calculating sums (Σ) of compound classes or congeners. Contaminants detected in less than 60% of the samples were excluded from further statistical analyses. Concentrations below the detection limit for contaminants detected in more than 60% of the samples were given values of half the corresponding detection limit for statistical analyses to avoid missing values in the data set. Statistical analyses were performed with contaminant concentrations given as wet weight values.

Multivariate data analyses were performed using the multivariate program Unscrambler (version 9.2, Camo AS, Oslo, Norway). Principal component analysis (PCA) was carried out to consider differences and similarities between groups and to evaluate intracorrelations. Next, univariate analyses were conducted using R 2.7.1 for Windows (R Development Core Team). Due to the small sample size, non-parametric tests were used. A Kruskal-Wallis test with a pairwise Mann-Whitney U *post hoc* test was used to test for differences between the three groups from Kongsfjorden (Dytham, 2003), whereas for the remaining a pairwise Mann-Whitney U test was used. Statistical significance level was set to $\alpha = 0.05$.

Results

The compounds *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, PCB-52, -141 and -151, BDE-183, -206, -207 and -208, PFHxA, PFHpA, PFBS, dibenzo[a,h]anthracene and all analysed organotin compounds (MBT, DBT, TBT, MPhT, DPhT and TPhT) were below the detection limit in all samples analysed. γ-HCH, PCB-128, BDE-28 and -209, PFOA, phenanthrene, anthracene, acenaphtylene, fluorene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[a]fluoroanthene, benzo[k]fluoroanthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene and benzo[ghi]perylene were detected in less than 60% of the samples and were thus excluded from statistical analyses.

Organic contaminants

Concentrations

Concentrations of all quantified compounds are summarised in table 1-4. The organochlorines were generally found in the highest concentrations, in particular *p,p*'-DDE and PCB-153. PFASs were detected in intermediate concentrations, whereas the lowest concentrations were generally found for BFRs and PAHs. No organotin compounds were detected in any of the samples.

A PCA revealed two groups of contaminant variables, with high degree of correlation between the contaminants in each of the two groups (figure 2). The smaller group consisted of the majority of PFASs (PFNA, PFTeA, PFDcS, PFPeDA, PFDcA, PFUnA, PFTriA, PFDoA and ΣPFAS), whereas the other cluster consisted of the remaining compounds included in the statistical analyses (organochlorine pesticides including toxaphenes, PCBs, BFRs, naphthalene, ΣPAH, PFOS, PFHxS and PFOSA). The objects were distributed temporally in the score plot, with the eggs from 1993 to the right and the eggs from 2007 to the left (figure 2), thus indicating that the eggs from 1993 contained the highest concentrations of the majority of the analysed organic contaminants, whereas the eggs from 2007 contained the lowest concentrations. Furthermore, the eggs from 2007 appeared to have higher concentrations of a selection of PFASs (the smaller cluster, figure 2). The score plot also indicates that eggs from Bjørnøya sampled in 2007 have the highest levels of these PFASs.

Differences between Kongsfjorden and Bjørnøya were analysed pairwise for the different years with univariate analyses. Overall, few significant differences were found and more differences were found between the two areas in 2007 than in 2002/2003 (appendix 2). Only a selection of PFASs (PFNA, PFPeDA, PFOSA, PFOS, PFDcS and ΣPFAS) were significantly or near significantly different in 2002/2003, of which all except for PFPeDA were present at the highest concentrations in Bjørnøya. In 2007 also HCB, PCB-28 and HBCD were significantly different between the two areas, as well as a variety of PFASs (PFNA, PFDcA, PFUnA, PFDoA, PFTriA, PFTeA, PFPeDA, PFOSA, PFDcS and ΣPFAS). All of these were present at higher concentrations in Bjørnøya than in Kongsfjorden. Thus, in the further analyses the PFAS were treated separately with respect to region, whereas for the OCs and BFRs the data for the two regions were combined for temporal analyses.

Univariate analyses confirm the temporal trends seen in the PCA. There were significant differences in contaminant concentration in Brünnich's guillemot eggs from Kongsfjorden and Bjørnøya between 1993 and 2007 for most OCPs and PCBs, with a few exceptions being HCB and β-HCH (Appendix 1, figure 3). The differences were also significant for the majority of the OCPs and PCBs between 1993 and 2002/2003 and between 2002/2003 and 2007. The concentrations were highest in 1993 and lowest in 2007 for all OCs in the samples. A similar trend was seen for the BDEs (figure 3); all the BDEs were significantly lower in 2007 than in 1993. However, fewer of the BDEs were significantly different between 1993 and 2002/2003, and none of the analysed BDEs were significantly different between 2002/2003 and 2007. On the contrary, HBCD was only significantly lower between 2002/2003 and 2007, and differed from the other BFRs by being found at the highest concentrations in 2002 and not in 1993.

The PFASs were the compound group with the largest variations in trends. In general for Kongsfjorden, concentrations of the carboxylic acids were highest in 2002 and lowest in 1993, whereas concentrations of the sulfonates were highest in 1993 and lowest in 2007 (figure 4). An exception was PFDcS, which was highest in 2007 and 2002, and lowest in 1993. In general for Bjørnøya, the concentrations of the carboxylic acids were higher in 2007 than in 2003, whereas the concentrations of the sulfonates were higher in 2003 than in 2007. An exception was PFDcS, which was higher in 2007 than 2003. Furthermore, significant differences were also found between 2002 and 2007 in Kongsfjorden and between 2003 and 2007 in Bjørnøya for naphthalene and ΣPAH.

Pattern

Chlorinated compounds clearly dominated the contaminant mixture, with PCBs and organochlorine pesticides including toxaphenes constituting 82-96% of the organic contaminants. There was, however, some differences in composition between the groups, both overall and within compound classes (figure 5). The largest variations were seen for PFAS. The samples from Bjørnøya (2003 and 2007) and from Kongsfjorden (2002 and 2007) followed the same overall contaminant pattern with the relative contribution of compound classes to the total contaminant burden in the following order; OCP (47-52%) > PCB (34-41%) > PFAS (6.2-17%) > BFR (1.1-1.4%). The samples from Kongsfjorden in 1993 differed from this and followed PCB (51%) > OCP (45%) > PFAS (2.6%) > BFR (1.5%).

The OCPs followed the same order of relative contribution to the total OCP concentrations in the five sample groups; *p,p'*-DDE (55-60%) > HCB (14-26%) > ΣCHB (6.3-13%) > oxychlordane (4.9-6.1%) > *p,p'*-DDT (1.8-4.3%) > *cis*-nonachlor (1.0-3.1%) ≈ β-HCH (1.3-1.8%) > mirex (0.7-1.2%) > α-HCH (0.1-0.4%). The dominating PCBs were PCB-153 (25-27%) > PCB-138 (16-18%) > PCB-118 (12-13%) > PCB-99 (6.9-7.8%) ≈ PCB-180 (5.9-7.8%) > PCB-187 (4.3-5.0%) ≈ PCB-66 (4.1-4.9%) ≈ PCB-105 (3.9-4.8%), which together constituted 82-84% of the total PCB concentrations.

The BFRs and PFASs however differed more in the relative contribution between groups. The five major compounds constituting 95-98% of ΣBFR were BDE-47 (48%) > HBCD (18%) > BDE-99 (16%) > BDE-100 (8.4%) > BDE-154 (5.4%) for Kongsfjorden in 1993, BDE-47 (38%) ≈ HBCD (38%) > BDE-100 (7.0%) ≈ BDE-99 (7.0%) > BDE-154 (6.2%) for Kongsfjorden in 2002 and HBCD (40%) > BDE-47 (35%) > BDE-100 (8.6%) > BDE-154 (6.3%) > BDE-99 (5.5%) for Kongsfjorden in 2007. For Bjørnøya the five major BFR compounds were HBCD (45%) > BDE-47 (38%) > BDE-100 (7.1%) > BDE-99 (4.5%) > BDE-154 (3.7%) for 2003 and HBCD (50%) > BDE-47 (29%) > BDE-100 (7.3%) > BDE-154 (5.6%) > BDE-99 (4.3%) for 2007.

Four compounds dominated the PFAS compounds and constituted 88-95% of ΣPFAS. Although the order was the same for all samples from Kongsfjorden in all three years, the relative contribution varied markedly between location and year. The relative contribution of the four major compounds contributing to ΣPFAS were PFOS (76%) > PFUnA (8.2%) ≈ PFTriA (8.1%) > PFDoA (2.5%) for Kongsfjorden in 1993, PFOS (37%) > PFUnA (21%) ≈ PFTriA (21%) > PFDoA (6.7%) for Kongsfjorden in 2002, PFOS (39%) > PFUnA (25%) > PFTriA (21%) > PFDoA (6.9%) for Kongsfjorden in 2007, PFOS (57%) > PFTriA (16%) > PFUnA (12%) > PFDoA (4.0%) for Bjørnøya in 2003 and PFTriA (32%) > PFUnA (31%) > PFOS (17%) > PFDoA (8.9%) for Bjørnøya in 2007.

Trace elements

Concentrations of trace elements are summarised in table 5. The samples from Kongsfjorden in 1993 were only analysed for organic contaminants, thus the temporal aspect for trace elements is minor. An exception is Hg, as Hg concentrations were reported for the 1993 Kongsfjorden eggs (0.20 ± 0.06 μg/g ww) in Barrett et al. (1996).

It should, however, be noted that in Barrett et al. (1996) the Hg concentrations were determined by hydride generator atomic absorption spectroscopy, whereas in the present study ICP-MS was utilised. The Hg concentrations in eggs from Kongsfjorden in 1993 (Barrett et al., 1996) and 2002 were similar. Significant differences in Hg concentrations were seen in Kongsfjorden between 2002 and 2007, with the highest concentrations in 2002. Furthermore, Hg was significantly higher in Kongsfjorden in 2002 than in Bjørnøya in 2003.

Table 1 Arithmetic mean concentrations (ng/g ww) with standard deviation (±SD) and ranges (min-max) for organochlorines (OCs) analysed in Brünnich's guillemot *Uria lomvia* egg homogenate samples from Kongsfjorden (1993, 2002 and 2007) and Bjørnøya (2003 and 2007). Sample size for each group is n=5, and number of samples above the respective detection limit is given for each compound. na denotes not analysed, nd denotes not above the detection limit.

	Kongsfjorden 1993			Kongsfjorden 2002			Kongsfjorden 2007			Bjørnøay 2003			Bjørnøya 2007		
	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max
Lipid%	5/5	12.7 ± 0.3	12.4 – 13.3	5/5	11.0 ± 2.1	7.64 – 13.6	5/5	13.3 ± 0.5	12.6 – 13.8	5/5	13.1 ± 1.5	11.8 – 15.6	5/5	13.0 ± 1.03	11.3 – 13.9
$\delta^{15}N$		na	na	5/5	12.6 ± 0.1	12.5 – 12.8	5/5	12.0 ± 0.3	11.6 – 12.5	5/5	12.1 ± 0.3	11.6 – 12.3	5/5	11.6 ± 0.2	11.3 – 11.7
$\delta^{13}C$		na	na	5/5	-19.9 ± 0.2	-20.3 – -19.7	5/5	-19.9 ± 0.2	-20.1 – -19.5	5/5	-19.8 ± 0.3	-20.1 – -19.5	5/5	-19.9 ± 0.2	-20.1 – -19.5
HCB	5/5	54.1 ± 20.4	22.9 – 73.6	5/5	43.9 ± 6.3	39.2 – 54.6	5/5	42.0 ± 4.0	38.7 – 48.5	5/5	51.3 ± 15.4	29.5 – 66.3	5/5	49.0 ± 2.01	47.1 – 52.1
α-HCH	5/5	1.46 ± 0.36	0.95 – 1.93	5/5	0.36 ± 0.12	0.23 – 0.54	5/5	0.24 ± 0.03	0.20 – 0.27	5/5	0.51 ± 0.20	0.26 – 0.81	5/5	0.31 ± 0.07	0.24 – 0.43
β-HCH	5/5	5.06 ± 1.78	2.19 – 6.89	5/5	4.09 ± 1.17	2.66 – 5.73	5/5	2.70 ± 0.61	2.09 – 3.71	5/5	2.99 ± 1.37	1.89 – 5.12	5/5	2.72 ± 0.65	1.75 – 3.47
γ-HCH	5/5	0.37 ± 0.10	0.22 – 0.47	1/5	0.21	nd – 0.21	0/5	nd	nd	1/5	0.27	nd – 0.27	0/5	nd	nd
ΣHCH	5/5	6.88 ± 2.19	3.35 – 8.91	5/5	4.49 ± 1.26	2.96 – 6.13	5/5	2.94 ± 0.61	2.36 – 3.97	5/5	3.55 ± 1.63	2.28 – 6.19	5/5	3.02 ± 0.69	1.99 – 3.76
Oxychlordane	5/5	23.4 ± 8.7	14.1 – 37.7	5/5	14.3 ± 3.6	11.0 – 20.3	5/5	9.38 ± 2.20	6.94 – 12.8	5/5	14.3 ± 6.7	8.25 – 24.9	5/5	9.26 ± 1.19	7.60 – 10.6
***trans*-Chlordane**	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd
***cis*-Chlordane**	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd
***trans*-Nonachlor**	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd
***cis*-Nonachlor**	5/5	12.4 ± 7.3	2.19 – 19.6	5/5	5.27 ± 3.70	1.86 – 10.4	5/5	1.85 ± 0.92	0.8 – 2.85	5/5	3.44 ± 3.52	1.49 – 9.67	5/5	2.23 ± 0.80	1.35 – 3.41
Mirex	5/5	4.64 ± 2.90	2.11 – 9.5	5/5	3.00 ± 1.21	1.78 – 4.95	5/5	1.61 ± 0.62	0.95 – 2.51	5/5	2.21 ± 0.83	1.46 – 3.57	5/5	1.31 ± 0.10	1.14 – 1.41
***p,p'*-DDE**	5/5	227 ± 112	158 – 424	5/5	129 ± 22	104 – 161	5/5	111 ± 18	92.9 – 132	5/5	131 ± 27	102 – 173	5/5	103 ± 8.50	97.4 – 118
***p,p'*-DDT**	5/5	17.1 ± 9.2	3.20 – 27.1	5/5	8.60 ± 3.70	3.66 – 13.5	5/5	3.30 ± 1.32	1.72 – 4.95	5/5	6.46 ± 3.83	3.63 – 13.1	5/5	4.70 ± 1.12	3.29 – 5.98
CHB-26	5/5	14.6 ± 6.1	6.79 – 22.6	5/5	6.30 ± 1.61	4.36 – 8.57	5/5	4.35 ± 1.77	2.65 – 6.94	5/5	7.26 ± 4.31	3.61 – 14.1	5/5	4.93 ± 1.59	2.67 – 6.77
CHB-40	5/5	10.5 ± 5.6	2.06 – 17.1	5/5	5.03 ± 2.13	2.15 – 7.41	5/5	1.84 ± 0.73	0.93 – 2.71	5/5	3.91 ± 2.21	2.32 – 7.71	5/5	2.87 ± 0.64	1.99 – 3.52
CHB-41	5/5	0.87 ± 0.63	0.11 – 1.52	5/5	0.49 ± 0.23	0.22 – 0.77	5/5	0.14 ± 0.05	0.09 – 0.22	5/5	0.38 ± 0.21	0.22 – 0.74	5/5	0.24 ± 0.07	0.17 – 0.32
CHB-44	5/5	8.62 ± 4.49	3.46 – 15.8	5/5	4.49 ± 1.74	2.33 – 6.97	5/5	2.29 ± 0.61	1.5 – 2.98	5/5	3.92 ± 1.75	2.45 – 6.76	5/5	2.63 ± 0.46	1.87 – 3.03
CHB-50	5/5	11.4 ± 9.1	1.62 – 24.0	5/5	5.58 ± 3.10	2.07 – 9.73	5/5	1.86 ± 0.79	0.95 – 2.79	5/5	4.64 ± 3.04	2.22 – 9.76	5/5	2.66 ± 0.65	1.81 – 3.40
CHB-62	5/5	7.36 ± 5.24	1.34 – 13.1	5/5	3.45 ± 1.67	1.27 – 5.68	5/5	1.16 ± 0.49	0.6 – 1.82	5/5	2.49 ± 1.50	1.42 – 5.10	5/5	1.55 ± 0.42	1.09 – 2.00
ΣCHB	5/5	53.3 ± 28.9	15.4 – 87.8	5/5	25.3 ± 9.8	12.4 – 36.4	5/5	11.6 ± 3.7	7.04 – 17.0	5/5	22.6 ± 12.9	12.6 – 44.2	5/5	14.9 ± 3.63	9.61 – 19.0
PCB-28	5/5	7.04 ± 1.52	5.46 – 9.20	5/5	4.01 ± 0.89	3.26 – 5.52	5/5	3.11 ± 0.21	2.9 – 3.41	5/5	3.79 ± 1.38	2.46 – 5.39	5/5	3.99 ± 0.22	3.70 – 4.24
PCB-47	5/5	4.60 ± 1.48	3.55 – 7.17	5/5	2.86 ± 0.66	2.32 – 3.94	5/5	1.99 ± 0.26	1.68 – 2.29	5/5	2.59 ± 1.47	0.72 – 4.65	5/5	2.00 ± 0.09	1.89 – 2.13
PCB-52	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd
PCB-66	5/5	18.9 ± 4.7	14.7 – 26.8	5/5	9.18 ± 1.97	7.6 – 12.5	5/5	6.36 ± 0.65	5.42 – 7.09	5/5	8.17 ± 3.46	4.37 – 13.2	5/5	6.46 ± 0.23	6.13 – 6.76
PCB-74	5/5	11.7 ± 3.0	9.29 – 16.9	5/5	5.90 ± 1.26	4.86 – 8.01	5/5	4.08 ± 0.45	3.59 – 4.58	5/5	5.32 ± 2.16	3.51 – 8.64	5/5	4.11 ± 0.26	3.65 – 4.29
PCB-99	5/5	30.6 ± 11.4	21.3 – 50.4	5/5	16.1 ± 4.0	13.0 – 22.8	5/5	10.7± 1.9	8.20 – 12.9	5/5	14.7 ± 10.5	3.75 – 31.8	5/5	9.72 ± 0.46	9.01 – 10.3
PCB-105	5/5	17.5 ± 4.9	12.7 – 25.2	5/5	8.68 ± 0.97	7.22 – 9.95	5/5	6.04 ± 0.77	4.99 – 6.79	5/5	7.78 ± 4.24	1.31 – 12.3	5/5	6.33 ± 0.37	5.74 – 6.68

PCB-114	5/5	1.84 ± 0.51	1.25 – 2.52	5/5	0.72 ± 0.11	0.56 – 0.88	5/5	0.49 ± 0.04	0.42 – 0.53	5/5	0.77 ± 0.24	0.58 – 1.14	5/5	0.54 ± 0.05	0.48 – 0.59
PCB-118	5/5	53.2 ± 15.6	42.0 – 77.9	5/5	28.2 ± 6.5	20.6 – 37.3	5/5	17.7 ± 2.86	13.7 – 21.1	5/5	24.1 ± 12.7	8.78 – 43.0	5/5	17.1 ± 0.81	15.8 – 17.7
PCB-123	5/5	1.98 ± 0.79	1.40 – 3.23	5/5	0.91 ± 0.24	0.71 – 1.32	5/5	0.54 ± 0.09	0.41 – 0.64	5/5	0.90 ± 0.39	0.57 – 1.43	5/5	0.56 ± 0.08	0.48 – 0.69
PCB-128	5/5	2.58 ± 2.23	0.66 – 6.32	3/5	1.02 ± 0.72	nd – 1.54	0/5	nd	nd	4/5	0.92 ± 0.92	nd – 2.28	1/5	0.48	nd – 0.48
PCB-137	5/5	2.99 ± 1.90	1.43 – 6.29	5/5	1.42 ± 0.69	0.93 – 2.56	5/5	0.75 ± 0.28	0.49 – 1.19	5/5	1.43 ± 1.20	0.69 – 3.22	5/5	0.61 ± 0.06	0.54 – 0.68
PCB-138	5/5	82.2 ± 45.8	46.2 – 161	5/5	40.1 ± 12.1	30.5 – 60.4	5/5	24.8 ± 6.3	17.4 – 32.9	5/5	34.3 ± 24.4	10.0 – 74.8	5/5	21.6 ± 1.57	20.0 – 24.1
PCB-141	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd
PCB-151	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd
PCB-153	5/5	111 ± 63	63.5 – 221	5/5	57.9 ± 17.6	44.4 – 87.3	5/5	39.0 ± 9.0	27.9 – 50.6	5/5	49.4 ± 32.8	13.2 – 102	5/5	33.6 ± 2.01	30.9 – 36.5
PCB-156	5/5	11.0 ± 3.2	7.60 – 15.5	5/5	4.31 ± 0.90	3.05 – 5.57	5/5	2.86 ± 0.37	2.32 – 3.28	5/5	4.14 ± 1.44	2.61 – 6.36	5/5	2.97 ± 0.23	2.72 – 3.27
PCB-157	5/5	3.8 ± 1.4	2.40 – 5.98	5/5	1.33 ± 0.32	0.94 – 1.77	5/5	0.91 ± 0.17	0.62 – 1.08	5/5	1.25 ± 0.54	0.68 – 2.09	5/5	0.92 ± 0.04	0.88 – 0.96
PCB-170	5/5	13.2 ± 8.6	7.55 – 28.0	5/5	5.60 ± 1.59	4.25 – 8.18	5/5	3.73 ± 0.84	2.59 – 4.95	5/5	4.19 ± 2.70	0.65 – 8.2	5/5	3.08 ± 0.05	3.01 – 3.14
PCB-180	5/5	35.1 ± 26.3	15.5 – 80.8	5/5	15.4 ± 6.0	11.1 – 25.2	5/5	9.77 ± 3.01	6.17 – 14.3	5/5	11.0 ± 8.00	1.82 – 23.7	5/5	7.73 ± 0.53	7.18 – 8.38
PCB-183	5/5	8.25 ± 4.20	5.45 – 15.6	5/5	3.74 ± 0.96	2.9 – 5.38	5/5	2.47 ± 0.44	1.81 – 3.04	5/5	2.94 ± 1.30	1.90 – 5.11	5/5	2.11 ± 0.15	1.99 – 2.37
PCB-187	5/5	20.6 ± 12.1	9.94 – 40.3	5/5	11.0 ± 3.1	7.82 – 16.0	5/5	6.89 ± 1.76	4.62 – 9.35	5/5	8.41 ± 3.27	5.65 – 14.0	5/5	5.63 ± 0.23	5.37 – 5.92
PCB-189	5/5	0.80 ± 0.34	0.50 – 1.33	5/5	0.30 ± 0.06	0.22 – 0.38	5/5	0.23 ± 0.02	0.21 – 0.25	5/5	0.30 ± 0.07	0.25 – 0.41	5/5	0.24 ± 0.02	0.22 – 0.26
PCB-194	5/5	6.02 ± 3.44	3.58 – 12.0	5/5	2.44 ± 0.58	1.73 – 3.33	5/5	1.85 ± 0.31	1.39 – 2.21	5/5	2.22 ± 0.61	1.73 – 3.2	5/5	1.76 ± 0.20	1.57 – 2.09
PCB-206	5/5	1.96 ± 0.88	1.28 – 3.45	5/5	1.04 ± 0.24	0.73 – 1.31	5/5	1.08 ± 0.08	1.00 – 1.17	5/5	1.18 ± 0.28	0.85 – 1.57	5/5	0.99 ± 0.17	0.82 – 1.26
ΣMOPCB	5/5	128 ± 35	97.6 – 184	5/5	63.6 ± 12.8	49.5 – 83.3	5/5	42.2 ± 5.4	34.5 – 48.3	5/5	56.3 ± 26.2	24.9 – 94.0	5/5	43.2 ± 2.0	39.8 – 44.8
ΣPCB	5/5	447 ± 214	297 – 817	5/5	222 ± 60	170 – 321	5/5	145 ± 29	108 – 183	5/5	189 ± 112	66.6 – 369	5/5	132 ± 6	123 – 139

Table 2 Arithmetic mean concentrations (ng/g ww) with standard deviation (±SD) and ranges (min-max) for brominated flame retardants (BFRs) analysed in Brünnich's guillemot *Uria lomvia* egg homogenate samples from Kongsfjorden (1993, 2002 and 2007) and Bjørnøya (2003 and 2007). Sample size for each group is n=5, and number of samples above the respective detection limit is given for each compound. nd denotes not above the detection limit.

	Kongsfjorden 1993			Kongsfjorden 2002			Kongsfjorden 2007			Bjørnøya 2003			Bjørnøya 2007		
	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max
BDE-28	5/5	0.20 ± 0.11	0.05 – 0.36	4/5	0.12 ± 0.06	nd – 0.2	1/5	0.06	nd – 0.06	2/5	0.12 ± 0.06	nd – 0.16	2/5	0.07 ± 0.03	nd – 0.09
BDE-47	5/5	6.39 ± 3.42	3.54 – 12.1	5/5	3.25 ± 2.23	0.87 – 6.15	5/5	1.32 ± 0.38	0.8 – 1.71	5/5	2.54 ± 2.23	0.29 – 6.09	5/5	1.34 ± 0.35	0.95 – 1.81
BDE-99	5/5	2.09 ± 1.47	0.69 – 4.55	5/5	0.59 ± 0.36	0.26 – 1.06	5/5	0.21 ± 0.07	0.12 – 0.29	5/5	0.30 ± 0.22	0.1 – 0.65	5/5	0.20 ± 0.06	0.13 – 0.28
BDE-100	5/5	1.13 ± 0.88	0.37 – 2.63	5/5	0.59 ± 0.52	0.06 – 1.17	5/5	0.32 ± 0.10	0.2 – 0.46	5/5	0.48 ± 0.34	0.15 – 1.04	5/5	0.33 ± 0.07	0.26 – 0.42
BDE-153	5/5	0.34 ± 0.27	0.14 – 0.81	5/5	0.18 ± 0.11	0.08 – 0.32	5/5	0.09 ± 0.02	0.07 – 0.12	5/5	0.09 ± 0.02	0.07 – 0.11	5/5	0.09 ± 0.02	0.06 – 0.11
BDE-154	5/5	0.72 ± 0.52	0.34 – 1.64	5/5	0.52 ± 0.34	0.15 – 0.96	5/5	0.24 ± 0.08	0.13 – 0.34	5/5	0.25 ± 0.16	0.07 – 0.5	5/5	0.26 ± 0.07	0.18 – 0.38
BDE-183	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd
BDE-206	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd
BDE-207	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd
BDE-208	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd
BDE-209	2/5	0.25 ± 0.28	nd – 0.44	0/5	nd	nd	1/5	0.27	nd – 0.27	0/5	nd	nd	1/5	0.07	nd – 0.07
ΣBDE	5/5	11.0 ± 6.70	5.42 – 22.5	5/5	5.23 ± 3.62	1.48 – 9.86	5/5	2.21 ± 0.72	1.25 – 2.97	5/5	3.69 ± 3.00	0.68 – 8.49	5/5	2.25 ± 0.61	1.57 – 2.94
HBCD	5/5	2.45 ± 1.14	1.09 – 3.94	5/5	3.23 ± 1.16	1.98 – 4.67	5/5	1.50 ± 0.26	1.17 – 1.84	5/5	3.04 ± 1.64	1.83 – 5.83	5/5	2.29 ± 0.60	1.44 – 3.07

Table 3 Arithmetic mean concentrations (ng/g ww) with standard deviation (±SD) and ranges (min-max) for perfluorinated alkyl substances (PFASs) analysed in Brünnich's guillemot *Uria lomvia* egg homogenate samples from Kongsfjorden (1993, 2002 and 2007) and Bjørnøya (2003 and 2007). Sample size for each group is n=5, and number of samples above the respective detection limit is given for each compound. nd denotes not above the detection limit.

	Kongsfjorden 1993			Kongsfjorden 2002			Kongsfjorden 2007			Bjørnøya 2003			Bjørnøya 2007		
	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max
PFHxA	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd
PFHpA	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd
PFOA	0/5	nd	nd	5/5	0.90 ± 0.26	0.54 – 1.26	0/5	nd	nd	5/5	1.69 ± 0.53	1.09 – 2.15	0/5	nd	nd
PFNA	4/5	0.48 ± 0.12	nd – 0.56	5/5	0.73 ± 0.10	0.58 – 0.86	3/5	0.58 ± 0.25	nd – 0.87	5/5	1.29 ± 0.40	0.88 – 1.95	5/5	1.17 ± 0.31	0.84 – 1.65
PFDcA	5/5	0.54 ± 0.06	0.46 – 0.63	5/5	1.15 ± 0.32	0.83 – 1.66	5/5	1.01 ± 0.39	0.60 – 1.55	5/5	1.02 ± 0.33	0.78 – 1.56	5/5	2.61 ± 0.92	1.37 – 3.51
PFUnA	5/5	1.91 ± 0.35	1.47 – 2.36	5/5	6.30 ± 1.35	4.71 – 7.91	5/5	5.47 ± 1.58	4.32 – 8.17	5/5	5.43 ± 1.85	3.97 – 8.48	5/5	20.2 ± 6.4	11.3 – 28.3
PFDoA	5/5	0.58 ± 0.13	0.39 – 0.70	5/5	1.97 ± 0.46	1.48 – 2.56	5/5	1.51 ± 0.41	1.24 – 2.22	5/5	1.81 ± 0.49	1.41 – 2.58	5/5	5.76 ± 1.45	3.69 – 7.41
PFTriA	5/5	1.88 ± 0.76	0.91 – 2.71	5/5	6.15 ± 1.35	4.86 – 7.90	5/5	4.56 ± 1.14	3.41 – 6.12	5/5	7.17 ± 1.34	5.89 – 9.21	5/5	20.6 ± 5.51	11.5 – 25.6
PFTeA	0/5	nd	nd	5/5	0.52 ± 0.15	0.36 – 0.72	1/5	0.39	nd – 0.39	5/5	0.43 ± 0.14	0.28 – 0.65	5/5	1.38 ± 0.37	0.82 – 1.71
PFPeDA	0/5	nd	nd	5/5	0.69 ± 0.17	0.46 – 0.83	3/5	0.47 ± 0.10	nd – 0.58	5/5	0.45 ± 0.09	0.36 – 0.56	5/5	1.99 ± 0.51	1.29 – 2.55
PFOSA	5/5	0.18 ± 0.06	0.09 – 0.25	5/5	0.04 ± 0.03	0.02 – 0.09	2/5	0.03 ± 0.00	nd – 0.03	5/5	0.12 ± 0.02	0.09 – 0.15	5/5	0.04 ± 0.01	0.03 – 0.05
PFBS	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd
PFHxS	5/5	0.14 ± 0.07	0.07 – 0.25	5/5	0.10 ± 0.03	0.07 – 0.13	4/5	0.07 ± 0.06	nd – 0.12	5/5	0.14 ± 0.10	0.04 – 0.28	5/5	0.04 ± 0.02	0.02 – 0.07
PFOS	5/5	17.5 ± 2.4	14.9 – 20.8	5/5	10.9 ± 1.7	9.00 – 12.7	5/5	8.48 ± 2.23	7.02 – 12.4	5/5	26.0 ± 11.2	16.0 – 43.0	5/5	10.8 ± 2.7	6.57 – 14.0
PFDcS	3/5	0.05 ± 0.01	0.04 – 0.05	5/5	0.14 ± 0.02	0.12 – 0.17	4/5	0.15 ± 0.02	0.13 – 0.17	5/5	0.27 ± 0.10	0.18 – 0.44	5/5	0.36 ± 0.11	0.20 – 0.46
ΣPFAS	5/5	23.2 ± 3.7	18.7 – 27.5	5/5	29.6 ± 5.2	24.1 – 35.0	5/5	21.9 ± 5.9	17.1 – 32.1	5/5	45.8 ± 15.3	33.8 – 70.7	5/5	64.8 ± 17.3	37.6 – 83.0

Table 4 Arithmetic mean concentrations (ng/g ww) with standard deviation (±SD) and ranges (min-max) for polyaromatic hydrocarbons (PAHs) analysed in Brünnich's guillemot *Uria lomvia* egg homogenate samples from Kongsfjorden (2002 and 2007) and Bjørnøya (2003 and 2007). Sample size for each group is n=5, and number of samples above the respective detection limit is given for each compound. nd denotes not above the detection limit.

	Kongsfjorden 2002			Kongsfjorden 2007			Bjørnøya 2003			Bjørnøya 2007		
	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max
Naphtalene	5/5	2.8 ± 0.7	1.6 – 3.4	1/5	0.2	nd – 0.2	5/5	4.5 ± 1.9	2.8 – 7.7	1/5	0.2	nd – 0.2
Phenanthrene	0/5	nd	nd	0/5	nd	nd	5/5	0.5 ± 0.3	0.2 – 0.9	0/5	nd	nd
Anthracene	1/5	1.2	nd – 1.2	0/5	nd	nd	2/5	0.1 ± 0.0	nd – 0.1	0/5	nd	nd
Acenaphtylene	0/5	nd	nd	0/5	nd	nd	1/5	0.1	nd – 0.1	0/5	nd	nd
Acenaphtene	1/5	0.4	nd – 0.4	0/5	nd	nd	1/5	0.8	nd – 0.8	1/5	0.3	nd – 0.3
Fluorene	1/5	0.2	nd – 0.2	0/5	nd	nd	0/5	nd	nd	1/5	0.1	nd – 0.1
Fluoranthene	1/5	0.3	nd – 0.3	1/5	0.2	nd – 0.2	3/5	0.7 ± 0.4	nd – 0.9	1/5	0.3	nd – 0.3
Pyrene	0/5	nd	nd	1/5	0.4	nd – 0.4	2/5	0.4 ± 0.1	nd – 0.4	0/5	nd	nd
Benzo[a]anthracene	1/5	1.5	nd – 1.5	0/5	nd	nd	3/5	0.5 ± 0.3	nd – 0.8	0/5	nd	nd
Chrysene	1/5	1.6	nd – 1.6	0/5	nd	nd	3/5	0.6 ± 0.2	nd – 0.7	0/5	nd	nd
Benzo[b]fluoranthene	1/5	0.4	nd – 0.4	0/5	nd	nd	3/5	0.9 ± 0.3	nd – 1.2	0/5	nd	nd
Benzo[k]fluoranthene	1/5	0.7	nd – 0.7	0/5	nd	nd	3/5	0.6 ± 0.1	nd – 0.8	0/5	nd	nd
Benzo[a]pyrene	0/5	nd	nd	0/5	nd	nd	1/5	0.3	nd – 0.3	0/5	nd	nd
Indeno[1,2,3-cd]perylene	0/5	nd	nd	0/5	nd	nd	1/5	0.1	nd – 0.1	0/5	nd	nd
Benzo[ghi]perylene	0/5	nd	nd	0/5	nd	nd	2/5	0.3 ± 0.2	nd – 0.4	0/5	nd	nd
Dibenzo[a,h]anthracene	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd	0/5	nd	nd
ΣPAH	5/5	4.1 ± 2.1	1.6 – 7.1	2/5	0.4 ± 0.3	nd – 0.6	5/5	7.5 ± 3.6	3.7 – 13.0	3/5	0.3 ± 0.1	nd – 0.5

Table 5 Arithmetic mean concentrations (µg/g dw) with standard deviation (±SD) and ranges (min-max) for a selection of trace elements analysed in Brünnich's guillemot *Uria lomvia* egg homogenate samples from Kongsfjorden (2002 and 2007) and Bjørnøya (2003 and 2007). Sample size for each group is n=5, and number of samples above the respective detection limit is given for each compound. nd denotes not above the respective detection limit.

	Konsfjorden 2002			Kongsfjorden 2007			Bjørnøya 2003			Bjørnøya 2007		
	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max	n	Mean ± SD	Min – Max
Drymass %	5/5	29.6 ± 1.0	28.2 – 30.5	5/5	30.0 ± 0.6	29.2 – 30.9	5/5	31.2 ± 2.9	29.1 – 35.9	5/5	30.0 ± 1.6	27.6 – 31.9
Li	5/5	0.0405 ± 0.0174	0.0264 – 0.0688	5/5	0.0530 ± 0.0562	0.0196 – 0.152	5/5	0.0379 ± 0.0039	0.0344 – 0.0436	5/5	0.0220 ± 0.0051	0.0140 – 0.0269
V	5/5	0.0043 ± 0.0033	0.0031 – 0.0092	5/5	0.0043 ± 0.0019	0.0027 – 0.0075	5/5	0.0073 ± 0.0032	0.0036 – 0.0103	5/5	0.0021 ± 0.0018	0.0014 – 0.0048
Cr	5/5	0.416 ± 0.527	0.019 – 1.286	5/5	0.041 ± 0.044	0.013 – 0.119	5/5	0.176 ± 0.325	0.020 – 0.757	5/5	0.041 ± 0.027	0.016 – 0.087
Mn	5/5	1.18 ± 0.14	1.01 – 1.34	5/5	0.907 ± 0.154	0.736 – 1.16	5/5	1.25 ± 0.61	0.750 – 2.01	5/5	1.07 ± 0.25	0.786 – 1.47
Co	5/5	0.0132 ± 0.0062	0.0065 – 0.0222	5/5	0.0135 ± 0.0046	0.0083 – 0.0183	5/5	0.0258 ± 0.0091	0.0159 – 0.0395	5/5	0.0132 ± 0.0018	0.0113 – 0.0154
Ni	5/5	0.219 ± 0.289	0.0097 – 0.706	5/5	0.0467 ± 0.0502	0.0086 – 0.128	5/5	0.138 ± 0.207	0.0209 – 0.507	5/5	0.0235 ± 0.0106	0.0163 – 0.0423
Cu	5/5	3.90 ± 0.40	3.35 – 4.35	5/5	3.83 ± 0.43	3.38 – 4.53	5/5	3.95 ± 0.42	3.33 – 4.31	5/5	3.87 ± 0.45	3.06 – 4.1
Zn	5/5	57.4 ± 3.1	53.6 – 60.7	5/5	52.7 ± 4.5	48.5 – 59.3	5/5	58.0 ± 6.1	51.6 – 65.4	5/5	57.1 ± 2.5	53.8 – 60.42
As	5/5	0.52 ± 0.18	0.39 – 0.83	5/5	0.50 ± 0.06	0.41 – 0.55	5/5	0.79 ± 0.24	0.58 – 1.18	5/5	0.42 ± 0.038	0.39 – 0.48
Se	5/5	2.33 ± 0.20	2.05 – 2.59	5/5	2.24 ± 0.12	2.12 – 2.42	5/5	2.56 ± 0.24	2.31 – 2.85	5/5	2.58 ± 0.36	2.05 – 2.99
Mo	5/5	0.152 ± 0.066	0.117 – 0.270	5/5	0.149 ± 0.015	0.124 – 0.164	5/5	0.185 ± 0.062	0.140 – 0.292	5/5	0.150 ± 0.029	0.110 – 0.185
Cd	5/5	0.0029 ± 0.0020	0.0016 – 0.0063	5/5	0.0021 ± 0.0006	0.0011 – 0.0028	5/5	0.0033 ± 0.0007	0.0022 – 0.0041	5/5	0.0014 ± 0.0008	0.0008 – 0.0028
Sn	5/5	0.0089 ± 0.0064	0.0043 – 0.0200	5/5	0.0050 ± 0.0028	0.0025 – 0.0094	5/5	0.0059 ± 0.0031	0.0037 – 0.0114	5/5	0.0063 ± 0.0007	0.0052 – 0.0069
Sb	4/5	0.0007 ± 0.0002	nd – 0.0010	1/5	0.0005	nd – 0.0005	3/5	0.0006 ± 0.0001	nd – 0.0008	4/5	0.0006 ± 0.0001	nd – 0.0008
Hg	5/5	0.684 ± 0.194	0.496 – 0.931	5/5	0.360 ± 0.154	0.251 – 0.614	5/5	0.164 ± 0.065	0.103 – 0.258	5/5	0.126 ± 0.099	0.065 – 0.298
Tl	5/5	0.0041 ± 0.0010	0.0028 – 0.0052	5/5	0.0063 ± 0.0009	0.0051 – 0.0075	5/5	0.0314 ± 0.0052	0.0249 – 0.0364	5/5	0.0056 ± 0.0019	0.0028 – 0.0082
Pb	5/5	0.0044 ± 0.0019	0.0024 – 0.0071	5/5	0.0020 ± 0.0013	0.0011 – 0.0044	5/5	0.0061 ± 0.0015	0.00458 – 0.008	5/5	0.0078 ± 0.0080	0.0013 – 0.0193

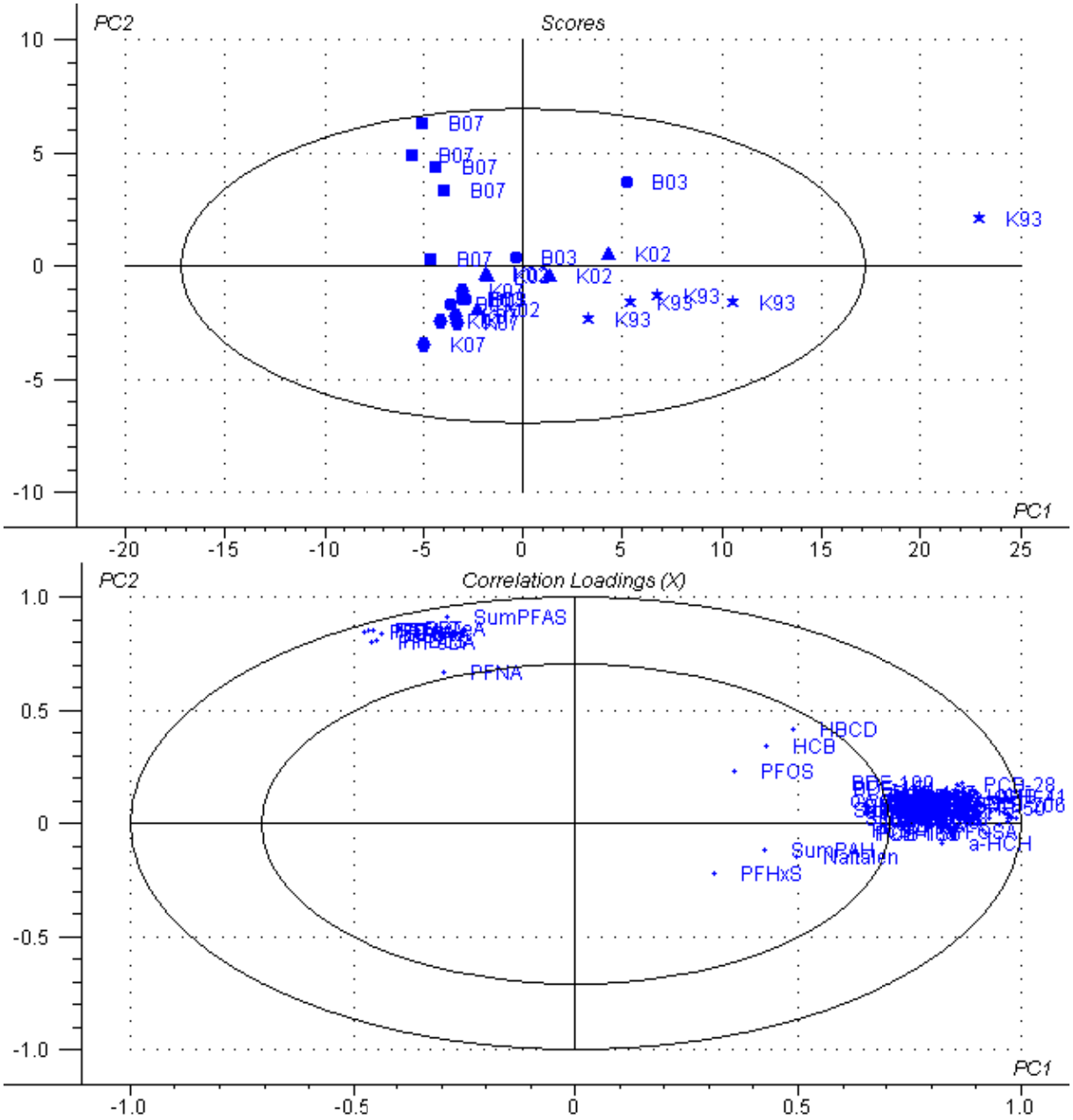

Figure 2 Score plot and loading plot from principal component analysis (PCA) of organic contaminant variables quantified in Brünnich's guillemot *Uria lomvia* eggs (n=25) from Kongsfjorden (1993, 2002 and 2007) and Bjørnøya (2003 and 2007). PC1 explains 69% and PC2 explains 11% of the variance in the data set (validated: 72%). K and B in the score plot designate samples from Kongsfjorden and Bjørnøya, respectively, followed by a two-digit number designating the year of sampling. The smallest cluster contained the majority of the PFASs (PFNA, PFTeA, PFDcS, PFPeDA, PFUnA, PFTriA, PFDoA, ΣPFAS), whereas the larger cluster (right) contained the OCs (PCBs, toxaphenes and other chlorinated pesticides), BFRs (BDEs and HBCD), naphthalene, ΣPAHs, PFOS, PFHxS and PFOSA. The analysis is based on wet weight values.

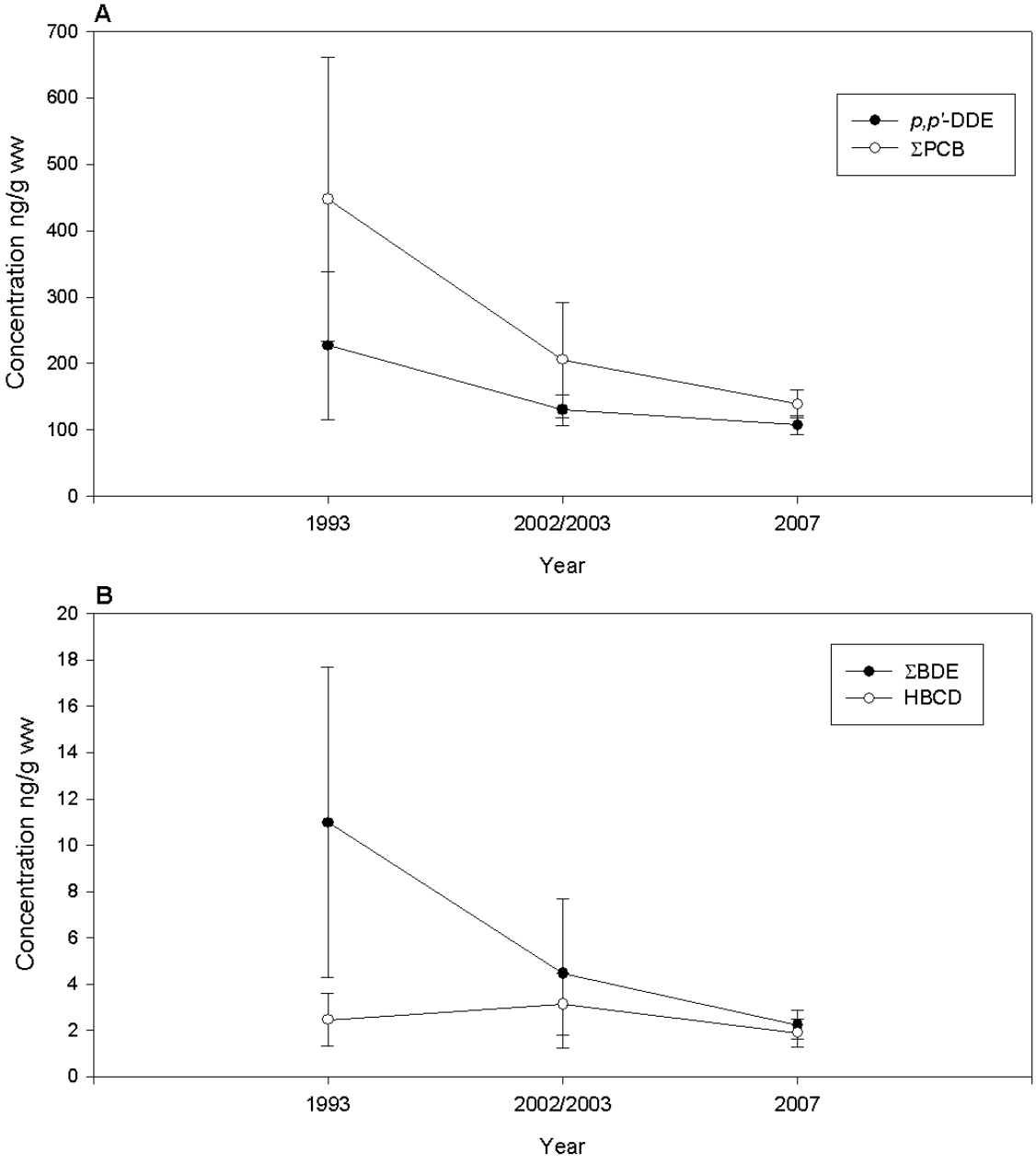

Figure 4 Arithmetic mean concentrations (ng/g wet weight) with standard deviation (±SD) for A) *p,p'*-DDE and ΣPCB and B) ΣBDE and HBCD in Brünnich's guillemot eggs from Kongsfjorden and Bjørnøya in 1993, 2002/2003 and 2007. Sample size for each group is n=5 for 1993 (only Kongsfjorden) and n=10 for 2002/2003 and 2007.

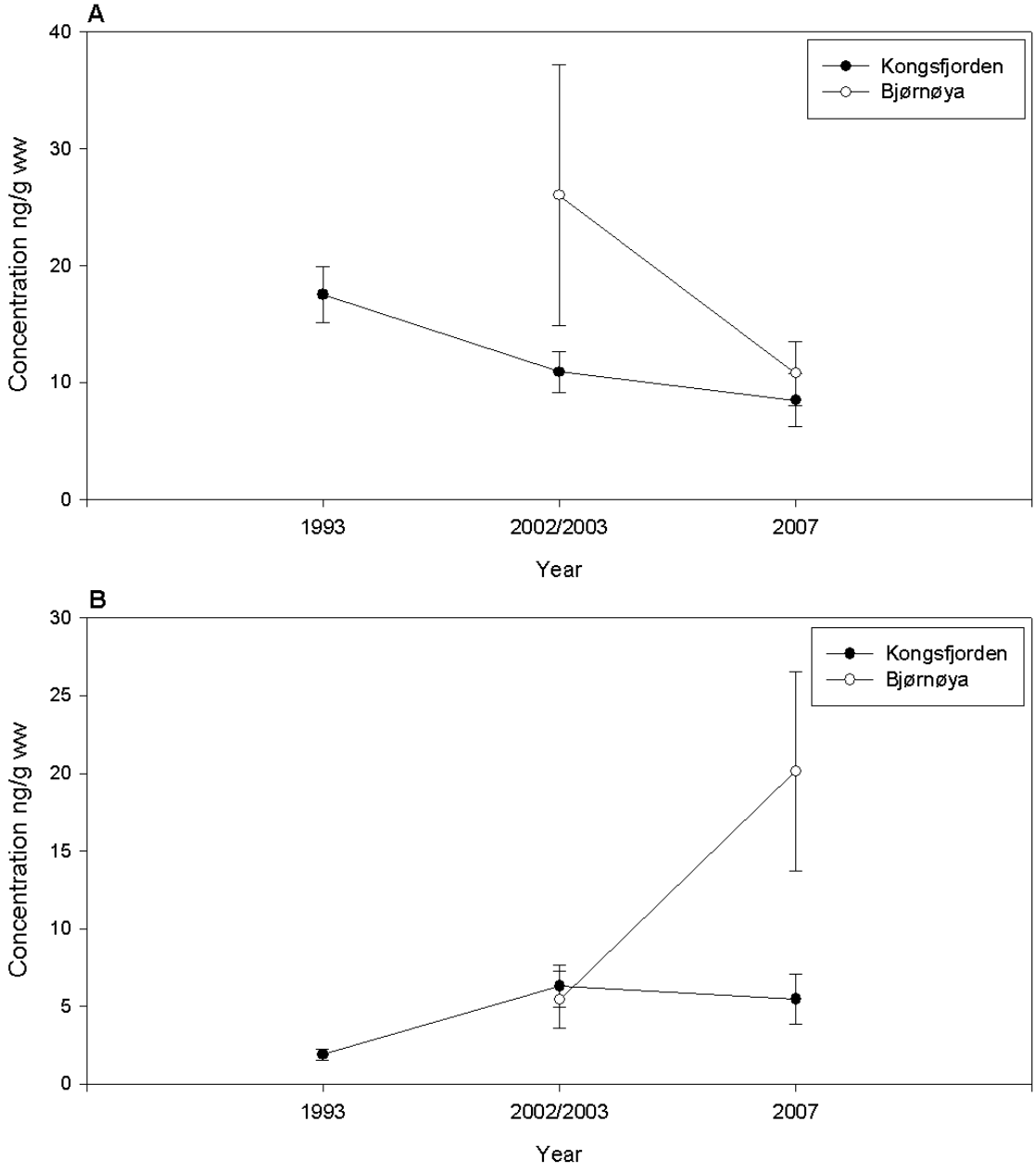

Figure 5 Arithmetic mean concentrations (ng/g wet weight) with standard deviation (±SD) for A) PFOS and B) PFUnA in Brünnich's guillemot eggs from Kongsfjorden and Bjørnøya in 1993, 2002/2003 and 2007. Sample size for each group is n=5.

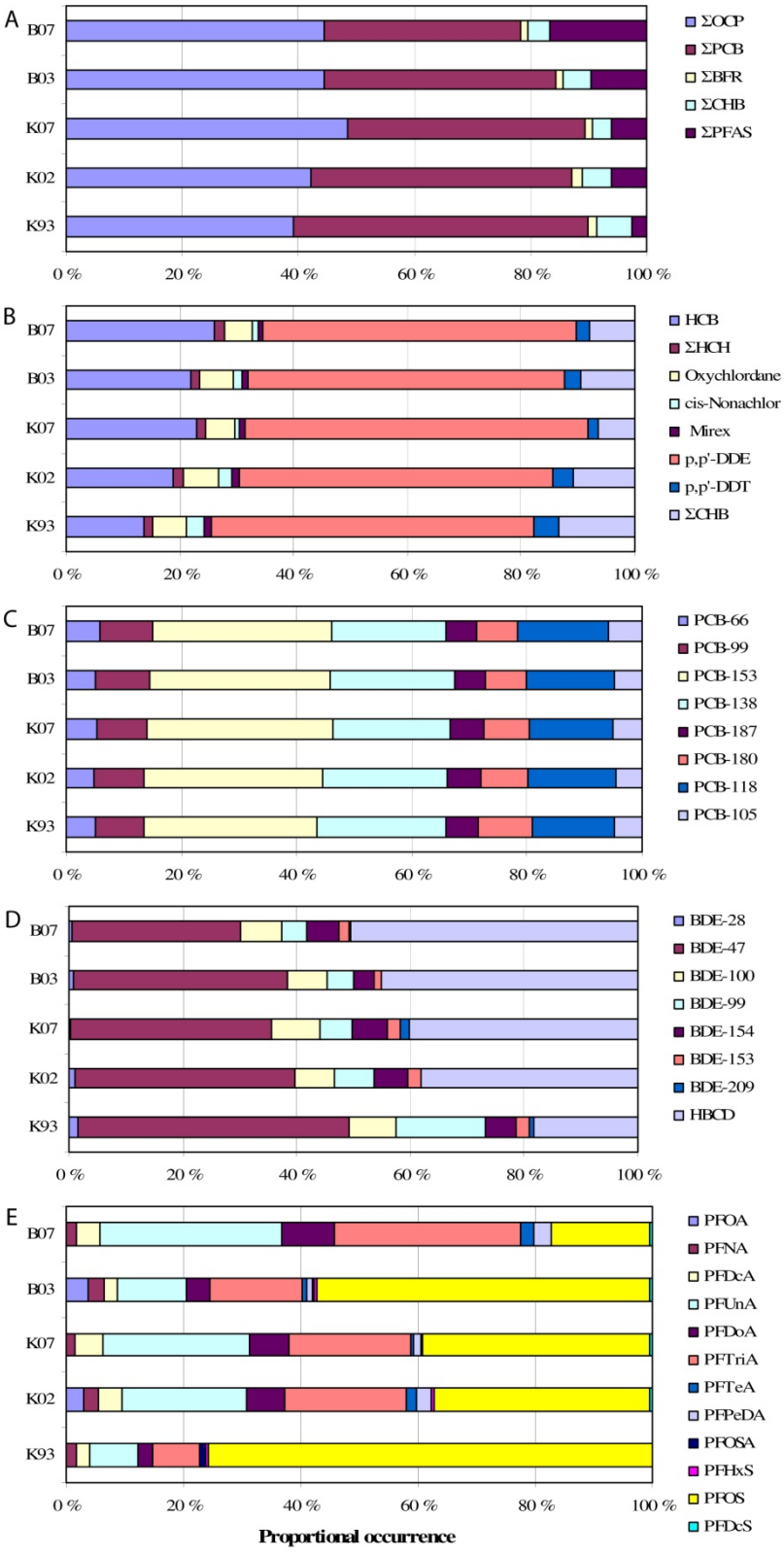

Figure 5 Relative distribution of various classes or compounds in Brünnich's guillemot *Uria lomvia* eggs from Kongsfjorden (1993, 2002 and 2007) and Bjørnøya (2003 and 2007). Sample size for each group is n=5. The graph is based on wet weight values. Plot A: Relative distribution (%) of OCPs, PCBs, BFRs, CHBs and PFASs to total organohalogen. Plot B: Relative distribution (%) of analysed organochlorine pesticides. Plot C: Relative distribution (%) of a selection of PCBs (main contributing congeners to ΣPCB (>4%)). Plot D: Relative distribution (%) of BDEs and HBCD to ΣBFR. Plot E: Relative distribution (%) of PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFTriA, PFTeA, PFPeDA, PFOSA, PFHxS, PFOS and PFDcS to ΣPFAS.

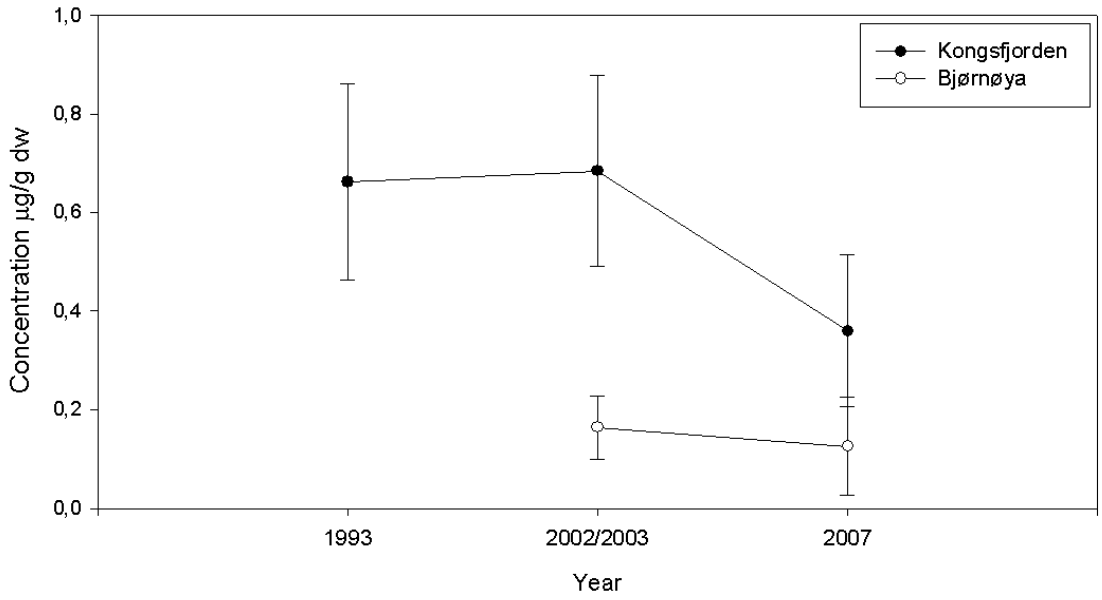

Figure 6 Arithmetic mean Hg concentrations (µg/g wet weight) with standard deviation (±SD) in Brünnich's guillemot eggs from Kongsfjorden and Bjørnøya in 1993, 2002/2003 and 2007. Sample size for each group is n=5. Data for Kongsfjorden 1993 are from Barrett et al. (2009) (wet weight values) and were estimated to dry weight values based on drymass % in the present study (n=25).

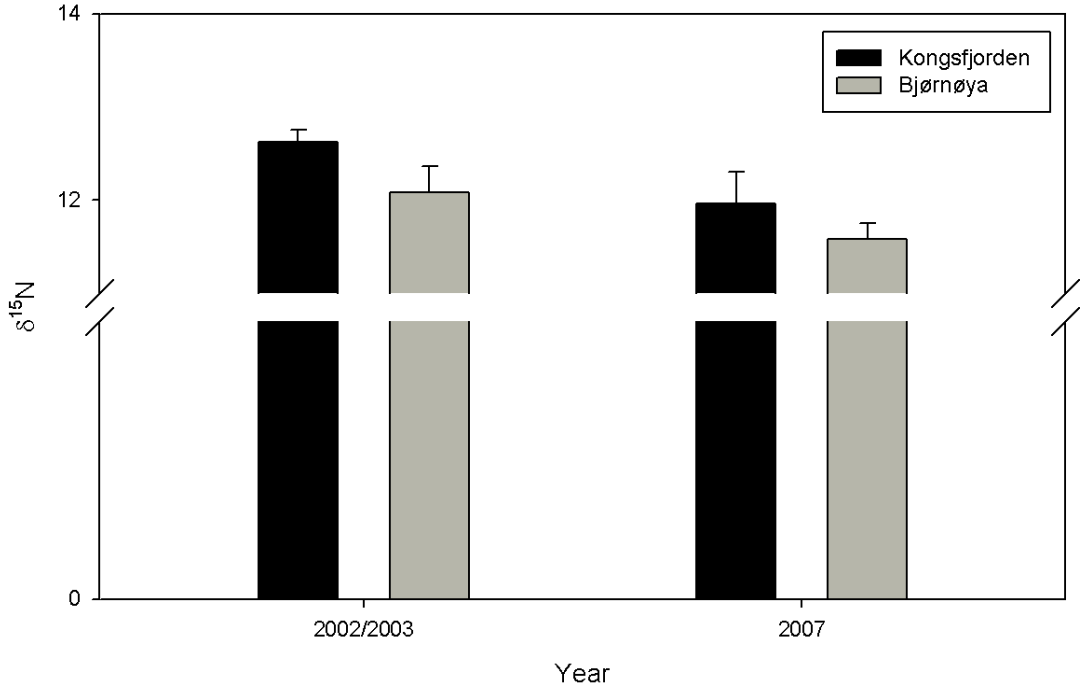

Figure 7 Arithmetic mean $\delta^{15}N$ ratios with standard deviation (±SD) in Brünnich's guillemot eggs from Kongsfjorden and Bjørnøya in 2002/2003 and 2007. Sample size for each group is n=5.

Discussion

Organic contaminants

Concentrations

The concentrations of HCB in the present study were roughly twice the concentrations found in glaucous gull *Larus hyperboreus* eggs from Bjørnøya (Verreault et al., 2005b), but lower than in European shag *Phalacrocorax aristotelis* yolk sac from central Norway (Murvoll et al., 2006), ivory gull *Pagophila eburnea* eggs from Svalbard and Russia (Miljeteig et al., 2009) and herring gull *Larus argentatus*, black-legged kittiwake *Rissa tridactyla*, common guillemot *Uria aalge* and Atlantic puffin *Fratercula arctica* eggs from northern Norway (Helgason et al., 2008). The concentrations were similar to that in Brünnich's guillemot and black guillemot *Cepphus grylle* eggs from Bjørnøya (AMAP, 2004).

The concentrations of HCHs were similar to that found in herring gull, black-legged kittiwake and Atlantic puffin eggs from northern Norway (Helgason et al., 2008) and glaucous gull eggs from Bjørnøya (Verreault et al., 2005b). The concentrations were however lower than in Brünnich's guillemot eggs from Canada (Braune et al., 2001). Thus, this is in line with the general findings of higher concentrations of HCHs in Canadian seabirds due to the proximity to Asia, where HCH have recently been used (AMAP, 2004).

The chlordane concentrations were lower than that found in northern fulmar *Fulmarus glacialis*, Brünnich's guillemot and black-legged kittiwake eggs from Canada (Braune et al., 2001) and glaucous gull eggs from Bjørnøya (Verreault et al., 2005b). The concentrations of oxychlordane were generally similar to that in herring gull, black-legged kittiwake and Atlantic puffin eggs from northern Norway; an exception was Brünnich's guillemot eggs from Kongsfjorden in 1993 which were higher (Helgason et al., 2008). The *cis*-nonachlor concentrations in the present study were lower than in herring gull, black-legged kittiwake and Atlantic puffin eggs from northern Norway (Helgason et al., 2008). In the present study oxychlordane is dominating and *cis*-nonachlor is the only other chlordane detected, whereas *trans*- and *cis*-nonachlor were the chlordanes present in highest concentrations in herring gull and Atlantic puffin eggs (Helgason et al., 2008).

The mirex concentrations were lower than in black-legged kittiwake and northern fulmar eggs from Canada and similar to Brünnich's guillemot eggs from Canada and glaucous gull eggs from Bjørnøya (Braune et al., 2001; Verreault et al., 2005b). The DDT concentrations in the present study were higher than in kittiwake eggs and similar to concentrations in common guillemot and Atlantic puffin eggs from northern Norway (Helgason et al., 2008). Compared to Canada, the DDT concentrations were similar to that in Brünnich's guillemot eggs and higher than in black-legged kittiwake eggs (Braune et al., 2001).

Toxaphenes have not been routinely analysed in seabird eggs, thus the reports on toxaphene concentrations are few. However, the toxaphene concentrations in the present study were generally lower than in ivory gull eggs from Svalbard and Russia (Miljeteig et al., 2009), glaucous gull eggs from Bjørnøya (Verreault et al., 2005b) and black-legged kittiwake and northern fulmar from Canada (Braune and Simon, 2004).

The PCB concentrations in the present study were similar to that found in eggs of black-legged kittiwakes, herring gulls, common guillemots and Atlantic puffins from northern Norway (Helgason et al., 2008). The PCB concentrations were also in the same range as in black-legged kittiwake, northern fulmar and Brünnich's guillemots from the Canadian Arctic (Braune et al., 2001). The concentrations were however considerably lower than in ivory gull and glaucous gull eggs from Svalbard and Bjørnøya (Verreault et al., 2005b; Miljeteig et al., 2009).

The BDE concentrations in the present study were generally lower than that reported in black-legged kittiwake, Atlantic puffin and herring gull eggs from northern Norway (Knudsen et al., 2005). The concentrations were also lower than in glaucous gull eggs from Bjørnøya (Verreault et al., 2004), ivory gull eggs from Svalbard and Russia (Miljeteig et al., 2009) and common guillemot eggs from the

Baltic Sea (Sellström et al., 2003). Concentrations reported in black guillemot eggs from East Greenland were similar to the highest concentrations in the present study (Kongsfjorden 1993) (Vorkamp et al., 2004). Nevertheless, the concentrations were higher than in northern fulmar eggs from the Faroe Islands (Fängström et al., 2005; Karlsson et al., 2006).

The HBCD concentrations were similar to that reported in common guillemot eggs from the Baltic Sea (Sellström et al., 2003). However, the concentrations were somewhat lower than that reported in glaucous gull eggs from Bjørnøya (Verreault et al., 2004), ivory gull eggs from Svalbard and Russia (Miljeteig et al., 2009) and black-legged kittiwake, herring gull and Atlantic puffin eggs from northern Norway (Knudsen et al., 2005).

The concentrations of PFOS, the most prevailing PFAS, were generally lower than concentrations reported in liver of seabirds from more polluted and industrialised regions, such as the USA, Italy and Korea (Giesy and Kannan, 2001; Kannan et al., 2001). The concentrations of the different PFAS, including PFOS, were generally lower than in glaucous gull eggs from Bjørnøya (Verreault et al., 2005a), herring gull eggs from northern Norway (Verreault et al., 2007), ivory gull eggs from Svalbard and Russia (Miljeteig et al., 2009) and common guillemot eggs from Iceland, Faroe Islands, Norway and Sweden (Löfstrand et al., 2008). The concentrations were however higher in the present study than in northern fulmar and black guillemot liver from the Canadian Arctic (Martin et al., 2004).

Very few studies report PAH concentrations in seabirds. The concentrations in the present study were lower than in a range of fish and invertebrates in Arctic marine ecosystems (Muir et al., 1992) and lower than in eggs from various species of birds in the Bajkal region (Lebedev et al., 1998). Furthermore, the concentrations were lower than that reported in livers from oiled common guillemot stranded on the east coast of England (Troisi et al., 2006). The highest concentrations in the present study (Kongsfjorden 2002 and Bjørnøya 2003), however, were similar to that reported in herring gull, cormorant *Phalacrocrax carbo* and chough *Pyrrhocorax pyrrhocorax* eggs from the British coast (Shore et al., 1999).

Temporal trends

The trend of decreasing OC concentrations with time is in line with other studies on contaminants in seabirds (e.g. Braune et al., 2001; Helgason et al., 2008). Barrett et al. (1996) reported decreasing concentrations of OCs from 1983 to 1993 in seabird eggs from the Barents Sea, a trend that has continued to 2003 (Helgason et al., 2008). The general trend in the Arctic is also decreasing concentrations of OCs, with regard to both the abiotic environment and biota (AMAP, 2004). Most OC pesticides were restricted or banned from the 1970s and although use of some pesticides is still continued in some developing countries, the global release into the environment has markedly decreased (Aguilar et al., 2002).

The decreasing BDE concentrations from 1993 to 2007 are in agreement with other studies on temporal BDE trends. Knudsen et al. (2005) reported an increase in BDEs in seabird eggs from Bjørnøya and northern Norway from 1983 to 1993 followed by a decrease from 1993 to 2003. Sellström et al. (2003) found a significant decrease in BDEs in common guillemot eggs from the Baltic Sea after the late 1980s. On the contrary, in peregrine falcon *Falco peregrinus* eggs from Greenland the BDE concentrations were increasing over the period 1986-2003 (Vorkamp et al., 2005). HBCD concentrations showed a less clear trend than the BDEs in the present study, but appear to peak in 2002/2003 before decreasing to 2007. Knudsen et al. (2005) reported an increase in HBCD concentrations from 1983 to 2003 in seabird eggs from Bjørnøya and northern Norway, whereas in common guillemot eggs from the Baltic Sea the HBCD concentrations were increasing up to recent periods and levelled out from the mid 1990s (Sellström et al., 2003). Vorkamp et al. (2005), however, reported a tendency towards a decrease in HBCD concentrations (not significant) in peregrine falcon eggs from Greenland over the time period studied (1986-2003).

In general, the present study found decreasing concentrations of sulfonates (PFOS, PFOSA) and increasing concentrations of carboxylates (PFNA, PFDcA, PFUnA, PFDoA, PFTriA, PFTeA, PFPeDA) from 1993 to 2002/2003 that either levelled off or increased to 2007. Similarly, Verreault et al. (2007) studied PFASs concentrations in herring gull eggs from northern Norway, and found an

increase in PFOS concentrations from 1983 to 1993, followed by a levelling off from 1993 to 2003. The concentrations of other PFASs also increased from 1983 to 1993, followed by a non-significant increase (PFOA, PFNA, PFDcA, PFUnA) or levelling off to 2003 (PFDoA, PFTriA) (Verreault et al., 2007).

A study investigating the time trend of PFOS and PFOA in common guillemot eggs from the Baltic Sea from 1968-2003 found a significant increase from 1968 to a peak in 1997-2000, followed by a decrease (Holmström et al., 2005). It was, however, suggested that the decrease appeared too early to be a response to the out-phasing of PFOS starting in 2000, but was more likely a result of a shift in diet (Holmström et al., 2005). The production of PFOS was out-phased by the main producer (3 M, USA) in 2000-2002. The observed decrease in PFOS in Brünnich's guillemot eggs may therefore be a result of the reduction of production of PFOS. A shift in diet may also describe the differences found in the present study, as the stable nitrogen isotope ratios ($\delta^{15}N$) were significantly higher in Kongsfjorden than in Bjørnøya in both 2002/2003 and 2007, and significantly higher in 2002/2003 than in 2007 (appendices 1 and 2).

Pattern

There were some differences in contaminant pattern between the different regions with time. The proportions of OCs were generally stable between years, whereas changes were seen for BFRs and PFASs. The OCs have been under regulation for decades, whereas the BFRs and PFASs have only been regulated for a few years (SFT, 2009). There was an increase in the proportion of HBCD relative to ΣBFR. HBCD is not yet regulated, although an EU risk assessment has been conducted and HBCD have been assigned as having persistent, bioaccumulative and toxic properties (BSEF, 2009). This may explain the proportional increase as HBCD is still in use, whereas pentaBDE and octaBDE were banned in all applications for the EU market in 2004 (BSEF, 2009).

There were large variations in the proportional occurrence of PFAS relative to ΣPFAS. Generally, there is a proportional decrease in PFOS with time and a proportional increase in perfluorinated carboxylates, dominated by PFUnA and PFTriA. Phase-out of PFOS started in 2000 (AMAP, 2004) and the decreasing proportions and concentrations of PFOS in the present study may indicate that the regulations are starting to be evident in biota. Other PFAS, such as PFUnA and PFTriA are still unregulated (SFT, 2008), and are thus still increasing. PFOS was clearly the predominant PFAS in biotic samples in previous studies (e.g. Houde et al., 2006), however, in the present study other PFAS are present in higher concentrations than PFOS in eggs from Bjørnøya in 2007. This is also reported in another recent study (Löfstrand et al., 2008).

Trace elements

Concentrations

The Hg concentrations in Brünnich's guillemot eggs in the present study were lower than in eggs from northern fulmars and Brünnich's guillemots from Canada (Braune et al., 2001; Braune and Simon, 2004). Furthermore, the Hg concentrations in eggs from Bjørnøya in 2003 and 2007 and Kongsfjorden in 2007 were lower than in black-legged kittiwake eggs from Canada, Brünnich's guillemot from Svalbard and common eider *Somateria mollissima* from Alaska (Barrett et al., 1996; Braune et al., 2001; Braune and Simon, 2004; Burger et al., 2008). The Hg concentrations in eggs from Kongsfjorden in 2002 were, however, higher than in eggs from black-legged kittiwakes from Canada and common eider from Alaska (Braune et al., 2001; Braune and Simon, 2004; Burger et al., 2008), but similar to concentrations in Brünnich's guillemot eggs from Svalbard (Barrett et al., 1996). The Hg concentrations in the present study were lower than in eggs from herring gull, black-legged kittiwake and atlantic puffin from northern Norway (Helgason et al., 2008).

Concentrations of other trace elements are not routinely analysed in seabird eggs. The Se concentrations in Brünnich's guillemot eggs in the present study were lower than in eggs from northern fulmars from Canada and similar to eggs from black-legged kittiwakes from Canada (Braune et al., 2001; Braune and Simon, 2004). The Se concentrations were, however, lower than the

concentrations reported in eggs from Brünnich's guillemots from Canada and common eider from Alaska (Braune et al., 2001; Braune and Simon, 2004; Burger et al., 2008). The Cu concentrations in the present study were lower than in eggs from northern fulmars, similar to Brünnich's guillemots and higher than black-legged kittiwakes from Canada (Braune and Simon, 2004). The Mn concentrations in the present study were higher than in common eider eggs from Alaska (Burger et al., 2008).

The Zn concentrations in the present study were higher than in eggs from black-legged kittiwakes and Brünnich's guillemots from Canada and similar to concentrations in northern fulmar eggs from Canada (Braune and Simon, 2004). The As concentrations were lower than (Bjørnøya in 2007 and Kongsfjorden in 2002 and 2007) or similar to (Bjørnøya 2003) the concentrations reported in common eider eggs in Alaska (Burger et al., 2008). The Cd concentrations were similar to (Bjørnøya 2007) or higher than the concentrations reported in common eider eggs from Alaska (Burger et al., 2008). The Cr concentrations were similar to (Kongsfjorden 2002) or lower than the Cr concentrations reported in common eider eggs from Alaska (Burger et al., 2008). The Pb concentrations were considerably lower in the present study than in eggs from common eider in Alaska (Burger et al., 2008).

Differences in Hg

The Hg concentrations in Brünnich's guillemot eggs from Kongsfjorden did not change from 1993 to 2003, however, in 2007 the concentration were approximately half of the concentrations in 2002. Barrett et al. (1996) and Helgason et al. (2008) found very little change in mercury levels when investigating seabird eggs from northern Norway and Svalbard from 1983 to 2003. The Hg concentrations in Kongsfjorden were clearly higher than the Hg concentrations in Bjørnøya in the present study. Savinov et al. (2003) also found considerably higher levels of Hg in Kongsfjorden than in Bjørnøya when investigating liver and muscle from several seabirds.

Conclusions

Contaminant concentrations in Brünnich's guillemot eggs were generally in the lower range of concentrations reported for other seabirds' eggs. The present study also confirms the temporal trend of decreasing concentrations of OCs and BDEs. The trends of PFASs were less clear and compound-dependent. The study indicates that the concentrations of PFOS are starting to decline, whereas the concentrations of the unregulated PFASs, such as PFUnA and PFTriA, are increasing. It should, however, be noted that the sample size for each year and site is low (n=5) and only a short time span is represented (1993-2007). The results are further confounded by significant differences in stable nitrogen isotope ratios ($\delta^{15}N$) between the two areas and between years (figure 7).

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Appendices

Appendix 1 U- and p-values for significant (in italics) and near significant (p<0.08) differences in contaminant concentrations between Kongsfjorden and in Bjørnøya (2002/2003 and 2007) tested pairwise with Mann-Whitney U test. The statistical test is performed using ww-values.

	Kongsfjorden vs Bjørnøya 2002/2003		**Kongsfjorden vs Bjørnøya 2007**	
	U	p	U	p
HCB			22	0.056
PCB-28			*25*	*0.008*
CHB-40			22	0.056
HBCD			*23*	*0.032*
PFNA	*25*	*0.008*	*24*	*0.021*
PFDcA			*24*	*0.016*
PFUnA			*25*	*0.008*
PFDoA			*25*	*0.008*
PFTriA			*25*	*0.008*
PFTeA			*25*	*0.007*
PFPeDA	3	0.057	*25*	*0.012*
PFOSA	*24.5*	*0.015*	*24*	*0.017*
PFOS	*25*	*0.008*		
PFDcS	*25*	*0.008*	*25*	*0.012*
ΣPFAS	*23*	*0.032*	*25*	*0.008*
Hg	*0*	*0.008*	3	0.059
Tl	*25*	*0.008*		
Co	*23*	*0.032*		
As			*2*	*0.036*
$\delta^{15}N$	*0*	*0.012*	3	0.056

Appendix 2 U- and p-values for significant (p<0.05, in italics) and near significant (p<0.08) differences in contaminant concentrations between the different sampling years in Kongsfjorden and Bjørnøya tested pairwise with Mann-Whitney U test. The samples were first tested for all three years with a Kruskal-Wallis test, where HCB, β-HCH and BDE-100 showed no significant differences and were thus excluded from the pairwise Mann-Whitney U test. The statistical test is performed using ww-values. na designates not analysed.

	1993 vs 2002/2003		**1993 vs 2007**		**2002/2003 vs 2007**	
	U	p	U	p	U	p
α-HCH	*50*	*0.003*	*50*	*0.003*	*82*	*0.017*
ΣHCH	*44*	*0.019*	*47*	*0.008*		
Oxychlordane	*43*	*0.028*	*50*	*0.001*	*85*	*0.007*
***cis*-nonachlor**	*44*	*0.019*	*46*	*0.008*		
Mirex			*49*	*0.001*	*90*	*0.002*
***p,p'*-DDE**	*47*	*0.005*	*50*	*0.001*	*81*	*0.019*
***p,p'*-DDT**	40	0.075	*42*	*0.040*	*82*	*0.015*

PCB-28	*49*	*0.001*	*50*	*0.001*		
PCB-47	*43*	*0.028*	*50*	*0.001*	*81*	*0.019*
PCB-74	*50*	*0.001*	*50*	*0.001*	*78*	*0.035*
PCB-66	*50*	*0.001*	*50*	*0.001*	*84*	*0.009*
PCB-99	*45*	*0.013*	*50*	*0.001*	*86*	*0.005*
PCB-153	*44*	*0.019*	*50*	*0.001*	*81*	*0.019*
PCB-137	*42*	*0.040*	*50*	*0.003*	*82*	*0.017*
PCB-138	*45*	*0.013*	*50*	*0.001*	*83*	*0.012*
PCB-187	*44*	*0.019*	*50*	*0.001*	*87*	*0.004*
PCB-183	*50*	*0.001*	*50*	*0.001*	*82*	*0.015*
PCB-180	*43*	*0.028*	*50*	*0.001*	*79*	*0.029*
PCB-170	*48*	*0.003*	*50*	*0.001*	*83*	*0.012*
PCB-194	*50*	*0.003*	*50*	*0.001*	*82*	*0.017*
PCB-206	*47*	*0.005*	*50*	*0.003*		
PCB-123	*49*	*0.001*	*50*	*0.001*	*84.5*	*0.010*
PCB-118	*48*	*0.003*	*50*	*0.001*	*87*	*0.004*
PCB-114	*50*	*0.003*	*50*	*0.003*	*97*	*0.000*
PCB-105	*50*	*0.001*	*50*	*0.001*	*90*	*0.002*
PCB-156	*50*	*0.001*	*50*	*0.001*	*87*	*0.004*
PCB-157	*50*	*0.001*	*50*	*0.003*	*85*	*0.009*
PCB-189	*50*	*0.003*	*50*	*0.003*	*88.5*	*0.004*
ΣMOPCB	*50*	*0.001*	*50*	*0.001*	*88*	*0.003*
ΣPCB	*46*	*0.008*	*50*	*0.001*	*84*	*0.009*
CHB-26	*45*	*0.013*	*49*	*0.001*		
CHB-40	40	0.075	*44*	*0.019*		
CHB-41			*42*	*0.043*	*91.5*	*0.002*
CHB-44	*43*	*0.028*	*50*	*0.001*	*80*	*0.023*
CHB-50			*42*	*0.040*	*85*	*0.007*
CHB-62			*45*	*0.013*	*86*	*0.005*
ΣCHB	*42*	*0.040*	*46*	*0.008*	*84*	*0.009*
BDE-47	*42*	*0.040*	*50*	*0.001*		
BDE-100			*46.5*	*0.010*		
BDE-99	*48*	*0.003*	*50*	*0.003*	76	0.053
BDE-154	40	0.075	*48.5*	*0.005*		
BDE-153	*42*	*0.043*	*50*	*0.003*		
ΣBDE	*43*	*0.028*	*50*	*0.001*		
HBCD					*83*	*0.012*
Naphtalene	na	na	na	na	*100*	*0.000*
ΣPAH	na	na	na	na	*100*	*0.000*
Cd	na	na	na	na	*82*	*0.017*
Pb	na	na	na	na	76	0.055
Li	na	na	na	na	*81*	*0.019*
V	na	na	na	na	76	0.054
Ni	na	na	na	na	*79*	*0.029*
As	na	na	na	na	*77*	*0.045*
$\delta^{15}N$	na	na	na	na	*87.5*	*0.005*

Appendix 3 U- and p-values for significant ($p<0.05$, in italics) and near significant ($p<0.08$) differences in contaminant concentrations between the different sampling years in Kongsfjorden and in Bjørnøya tested pairwise with Mann-Whitney U test. The two regions are tested separately for PFASs due to significant differences between the two regions (appendix 1). The samples from Kongsfjorden were first tested for all three years with a Kruskal-Wallis test, where PFHxS and ΣPFAS showed no significant differences and were thus excluded from the pairwise Mann-Whitney U test. The statistical test is performed using ww-values. na designates not analysed.

	Kongsfjorden 1993 vs 2002		**Kongsfjorden 1993 vs 2007**		**Kongsfjorden 2002 vs 2007**		**Bjørnøya 2003 vs 2007**	
	U	p	U	p	U	p	U	p
PFNA	*25*	*0.012*						
PFDcA	*25*	*0.008*	*24*	*0.016*			*1*	*0.021*
PFUnA	*25*	*0.008*	*25*	*0.008*			*0*	*0.008*
PFDoA	*25*	*0.008*	*25*	*0.008*	22	0.056	*0*	*0.008*
PFTriA	*25*	*0.008*	*25*	*0.008*			*0*	*0.008*
PFTeA	*25*	*0.007*			*25*	*0.007*	*0*	*0.008*
PFPeDA	*25*	*0.007*	20	0.072	*23*	*0.036*	*0*	*0.012*
PFOSA	*0.5*	*0.016*	*0*	*0.011*			*25*	*0.011*
PFOS	*0*	*0.008*	*0*	*0.008*	22	0.056	*25*	*0.008*
PFDcS	*25*	*0.012*						
Hg	na	na	na	na	*23*	*0.032*		