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ORGANOHALOGENS AND MERCURY IN IVORY GULL EGGS

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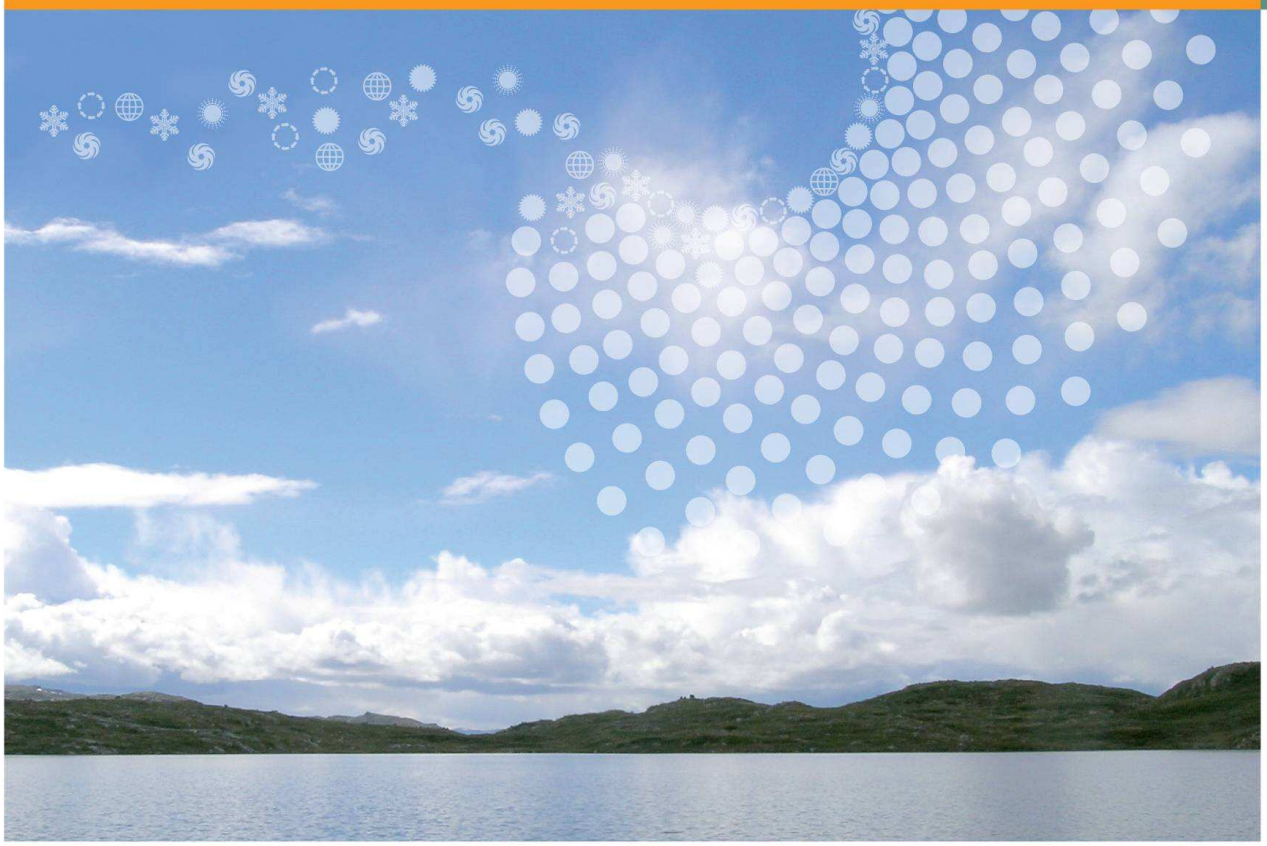

Arctic and Antarctic Research Institute

NTNU Norwegian University of Science and Technology

Norwegian School of Veterinary Science

Veterinærinstituttet
National Veterinary Institute

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: **Organohalogens and mercury in ivory gull eggs**

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Photo: Cecilie Miljeteig, Norwegian Polar Institute

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Preface

In the present study 35 ivory gull *Pagophila eburnea* eggs were sampled from four colonies in Svalbard (Svenskøya), Franz Josef Land (Nagurskoe and Klyuv Cape) and Severnaya Zemlya (Domashny). The eggs were analysed for polychlorinated biphenyls (PCBs), organochlorine pesticides, brominated flame retardants (BFRs), mercury (Hg), perfluorinated alkyl substances (PFAS), stable isotopes ($\delta^{15}N$ and $\delta^{13}C$) and vitamin A (retinol) and E (α-tocopherol). Furthermore, eggshell thickness was determined for all eggs.

This project was a collaboration between the Norwegian Polar Institute, Tromsø; Arctic and Antarctic Research Institute, St. Petersburg; Norwegian School of Veterinary Science, Oslo; Norwegian Veterinary Institute, Oslo; and Norwegian University of Science and Technology, Trondheim.

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1. Samandrag

Ismåka er ein sjeldan art i ein global samanheng, og er ein av dei minst kjende sjøfuglartane i verda. Kartleggjing av ismåkepopulasjonen i norsk og russisk Arktis vart starta etter at ein nedgang på 80 % i ismåkepopulasjonen i Canada vart dokumentert. Då ismåka er avhengig av sjøis og er på eit høgt trofisk nivå, er klimaendringar og miljøgifter to identifiserte miljøtruslar. Målet med denne studien var å kartleggje miljøgifter og kvikksølv i ismåkeegg frå Svalbard og russisk Arktis, og undersøkje moglege effektar av miljøgifter.

Totalt vart 35 egg samla inn frå ein koloni på Svalbard, to koloniar på Franz Josef Land og ein koloni på Severnaya Zemlya, Karahavet. Dei vart analysert for ei rekkje PCB-kongenerar, organoklorerte pestisider (OCP), bromerte flammehemmarar (BFR), perfluoroalkylstoffer (PFAS) og kvikksølv. I tillegg vart det analysert for tre responsvariablar i egga frå Russland (n=25); eggeskaltjukkleik og vitamina retinol og α-tocopherol.

Det vart funne høge nivå av miljøgifter i ismåkeegga samanlikna med nivå i egg frå ei rekkje sjøfuglartar (t.d. ismåke, polarmåke, krykkje) frå heile Arktis. Særskild nivåa av PCB-ar og OCP-ar, dominert av *p,p'*-DDE, var høge, medan nivåa av BFR-ar og PFAS-ar var vesentleg lågare. Skilnadar mellom koloniane i miljøgiftsnivå vart også funne. Generelt var nivåa av PCB-ar, OCP-ar og BFR-ar høgast i egg frå Nagurskoe-kolonien (Franz Josef Land), middels i egg frå Svenskøya-kolonien (Svalbard) og Klyuv Cape-kolonien (Franz Josef Land) og lågast i Domashny-kolonien (Severnaya Zemlya). Nivåa av kvikksølv og PFAS-ar var generelt sett like mellom koloniane.

Multivariat dataanalyse antyda assosiasjonar mellom dei tre responsvariablane og miljøgifter i egg frå dei tre koloniane i russisk arktis. Positive assosiasjonar vart funne mellom retinol (vitamin A) og nivå av miljøgifter. Indikasjonar på minkande konsentrasjonar av antioksidanten α-tocopherol med aukande miljøgiftskonsentrasjonar vart funne. Dette kan tyde på at ismåka er utsett for miljøgiftsindusert oksidativt stress. Det vart også funne

negative assosiasjonar mellom eggeskaltjukkleik og ei rekkje miljøgifter, noko som indikerer at eggeskaltjukkleiken er påverka av miljøgifter. Denne responsen er støtta av ei samanlikning av eggeskaltjukkleiken med ismåkeegg samla inn før DDT vart introdusert, då det vart funne ei 7 til 17% reduksjon i eggeskaltjukkleik i dei fire koloniane.

Dei høge nivåa av miljøgifter, særskild organokloriner, funne i ismåkeegg frå Svalbard og russisk Arktis er truleg så høge at dei påverkar ismåka. I tillegg til indikasjonane på effektar av miljøgifter på vitaminer og eggeskaltjukkleik funne i denne studien, er nivåa nær eller over ulike terskelverdiar for effektar. Ei samanlikning med egg frå før DDT-bruk indikerer at reduksjonen i eggeskaltjukkleik nærmar seg den kritiske grensa som er blitt assosiert med populasjonsnedgong i mange artar. Dette kan tyde på at nivåa er så høge at dei kan ha innverknad på populasjonsstatusen til ismåka. Dei høge nivåa av miljøgifter kan i tillegg verke som ein ytterlegare stressfaktor når ismåka møter utfordringane framtidige klimaendringar skapar.

2. Summary

The ivory gull is a rare species in a global context and is one of the most poorly known seabird species in the world. Following a documentation of an 80% population decline in the Canadian Arctic, mapping of the ivory gull population status in the Norwegian and Russian Arctic was initiated. Due to its dependence on sea ice and high trophic position, identified environmental threats are climate change and contaminants. The aim of the present study was to identify and quantify organic contaminants and mercury in ivory gull eggs from Svalbard and the Russian Arctic, and examine possible responses to contaminant exposure.

Thirty-five eggs were sampled from one colony in Svalbard, two colonies in Franz Josef Land and one colony in Severnaya Zemlya, Kara Sea. The eggs were analysed for a range of PCB-congeners, organochlorine pesticides (OCPs), brominated flame retardants (BFRs), perfluorinated alkyl substances (PFASs) and mercury. Additionally, three response variables were analysed in the eggs from the Russian Arctic (n=25); the vitamins retinol and α-tocopherol and eggshell thickness.

High contaminant levels were found in the ivory gull eggs when compared to levels in eggs from a range of seabird species (e.g. ivory gull, glaucous gull, black-legged kittiwake) throughout the Arctic. In particular the levels of PCBs and OCPs, dominated by *p,p*'-DDE, were high, whereas the levels of BFRs and PFASs were considerably lower. Differences between colonies in contaminant levels were also found. In general, PCBs, OCPs and BFRs were highest in eggs from the Nagurskoe colony (Franz Josef Land), intermediate levels in eggs from the Svenskøya (Svalbard) and Klyuv Cape (Franz Josef Land) colonies and the lowest concentrations found in eggs from the Domashny colony (Severnaya Zemlya). Levels of mercury and PFASs generally did not differ between colonies.

Multivariate data analysis indicated associations between the three response variables and contaminant variables in the eggs from the three colonies in the Russian Arctic. Positive associations between retinol (vitamin A) and contaminant levels were found. Indications of decreasing concentrations of the antioxidant α-tocopherol (vitamin E) with increasing concentrations of contaminants were found. This may indicate that the ivory gull is influenced by contaminant-induced oxidative stress. Negative associations were also found between eggshell thickness and a wide range of contaminants, indicating that the eggshell thickness is influenced by contaminants. The latter response is further supported by a comparison of eggshell thickness with ivory gull eggs collected before DDT was introduced, indicating a 7 to 17% decrease in eggshell thickness in the four colonies.

The high levels of contaminants, in particular organochlorines, found in ivory gull eggs from Svalbard and the Russian Arctic are likely to influence the ivory gull. In addition to the indications of effects on vitamin status and eggshell thickness found in the present study, the levels are approaching or higher than various threshold levels for effects. The degree of eggshell thinning relative to eggs from before DDT use is approaching the critical level associated with population declines in a range of species, indicating that eggshell thinning is of potential concern for the population status. Furthermore, the high levels of contaminants indicate that the ivory gull is under additional stress as it meets the challenges posed by future environmental change.

3. Introduction

Organohalogens of anthropogenic origin have been found in Arctic biota for decades, despite few local sources of contamination. Anthropogenic contaminants released in temperate regions in the northern hemisphere reach the Arctic by various routes, such as atmospheric and oceanic transport (Oehme 1991; Burkow and Kallenborn 2000). Recently, new classes of chemicals, such as brominated flame retardants (BFRs) and perfluorinated alkyl substances (PFASs), have become of global environmental concern in addition to the established organochlorines (OCs). Recent studies and reports have shown that both these compound groups are widespread in the environment and have been detected in the Arctic (de Wit et al. 2004; Smithwick et al. 2005; de Wit et al. 2006).

The ivory gull *Pagophila eburnea* is a characteristic high Arctic species, associated with sea ice throughout the year (Haney and MacDonald 1995). The ivory gull has an extremely northern distribution and has on average the northernmost breeding grounds of all birds (Blomqvist and Elander 1981). It has a patchy circumpolar breeding distribution, with scattered colonies in the Canadian Arctic, Greenland, Svalbard and Russia. The colonies in Russia are located in Franz Josef Land, on islands in the Kara Sea and in Severnaya Zemlya (Strøm 2006). The ivory gull is a colony breeder (Bakken and Tertitski 2000) and is versatile in its choice of nesting ground. It breeds on virtually inaccessible cliffs and nunataks in inland or coastal regions, as well as on flat ground on gravel-covered islands (Blomqvist and Elander 1981; Thomas and Macdonald 1987; Judin and Firsova 1990). The ivory gull lays 1-3 eggs, with a clutch size of two being most common, and egg laying has been reported from *medio* June to *medio* July (Blomqvist and Elander 1981; Gavrilo et al. in prep).

It is a rare species; the last population estimate suggests approximately 14,000 breeding pairs globally, with about 80% residing in the Russian Arctic (Bakken and Tertitski 2000). Gilchrist and Mallory (2005) demonstrated an 80% decline in the ivory gull breeding population in the Canadian Arctic during the last 20 years. Preliminary data from recent surveys in Russia and on Svalbard show some indications of population decline in Svalbard. Few of the known colonies were occupied in 2006, and those which were occupied consisted of fewer individuals than previously reported (Strøm and Gavrilo in prep).

The International Union for Conservation of Nature and Natural Resources (IUCN) classified the ivory gull as near threatened in 2006 on the IUCN red list of threatened species, with major threats identified as pollution and global warming (IUCN 2007). Gilchrist and Mallory (2005) suggested changing sea ice distribution and thickness, leading to altered wintering habitat, as a possible mechanism of the decline of ivory gulls in the Canadian Arctic. Furthermore, as a top predator, the ivory gull may be exposed to high levels of contaminants through biomagnification (Hobson et al. 2002; Borgå et al. 2004). Studies on contaminant levels in ivory gulls are scarce throughout the Arctic, with a few recent reportings from the Canadian Arctic, demonstrating high levels of organohalogen contaminants both in liver and fat (Fisk et al. 2001; Buckman et al. 2004), as well as in eggs (Braune et al. 2007) from ivory gulls. Additionally, some of the highest concentrations of Hg ever reported in seabird eggs from the Arctic have been found in ivory gull eggs from Canada (Braune et al. 2006).

Relationships between high levels of contaminants and various impacts on top predator birds in the Arctic have been demonstrated in a range of studies (recently reviewed in Gabrielsen 2007). Associations found include reduced reproductive performance (Helberg et al. 2005), alterations of the immune system (Bustnes et al. 2004), asymmetry in wing feathers (Bustnes et al. 2002), changes in circulating thyroid hormone levels (Verreault et al. 2004b) and altered

behaviour during nesting (Bustnes et al. 2001). Associations between contaminants and vitamin status have also been reported (e.g. Rolland 2000; Champoux et al. 2006; Murvoll et al. 2007), as well as genotoxic effects (Østby et al. 2005). Furthermore, contaminant-induced eggshell thinning is well documented (e.g. Cooke 1973; Lowe and Stendell 1991; Blus et al. 1997).

The main objective in the present study was to assess levels of mercury and a wide range of organohalogens, such as polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs), brominated flame retardants (BFRs) and perfluorinated alkyl substances (PFASs) in ivory gull eggs from four colonies in the Russian Arctic and Svalbard. Furthermore, eggshell thickness and levels of vitamin A (retinol) and E (α-tocopherol) was determined to evaluate associations with the contaminant variables.

4. Materials and Methods

4.1 Sampling procedures

A total of 35 eggs were sampled from individual nests within four colonies in Svalbard and north-western Russia; Nagurskoe and Klyuv Cape in Franz Josef Land and Domashny in Severnaya Zemlya (figure 1). Clutch size was noted for all sampled nests, and nests for sampling were chosen randomly. In Russia, only nests with two or three eggs were sampled. The eggs were weighed using a Pesola balance scaled at 50 g (Pesola AG, Baar, Switzerland) and width and length were determined to the nearest 0.1 mm with a still calliper in the field. The egg laying sequence was not determined to minimise disturbance. The eggs sampled in the Russian Arctic were marked, wrapped in aluminium foil and stored at -20 °C until further analyses. During transport the samples were kept frozen in thermos bottles. The eggs sampled in Svalbard were marked, wrapped in aluminium foil and stored on ice until the end of fieldwork (0-10 days). The samples were then stored at -20 °C until further analyses.

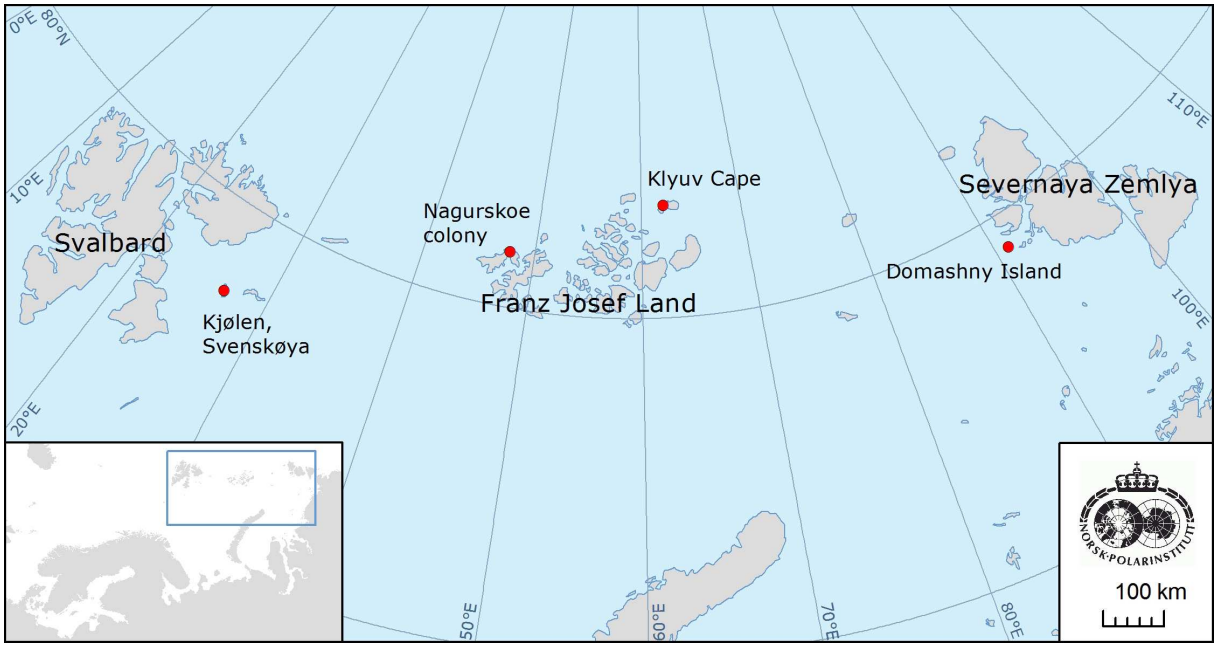

Figure 1 Map of Svalbard and the western Russian Arctic. Ivory gull eggs were sampled from Svenskøya (Svalbard), Nagurskoe and Klyuv Cape (both in Franz Josef Land) and Domashny Island (Severnaya Zemlya).

4.2 Analyses

All eggs were analysed for a suite of OCs, BFRs and PFASs. The organochlorine pesticides (OCPs) analysed and quantified were DDTs (*p,p'*-DDE, *p,p'*-DDT, *p,p'*-DDD, *o,p'*-DDT), chlordanes (oxychlordane, *trans*-nonachlor, *cis*-chlordane), HCHs (α-, β- and γ-), mirex, HCB, dieldrin, heptachlor, aldrin and toxaphenes (CHB-26, -40, -41, -44, -50 and -62). Other OCs analysed were PCBs (PCB-28, - 47, -52, -66, -74, -99, -101, -105, -114, -118, -128, -137, -138, -141, -149, -151, -153, -156, -157, -167, -170, -180, -183, -187, -189, -194, -196 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 and -209). The PFASs analysed and quantified were perfluorohexanoate (PFHxA), perfluoroheptanoate (PFHpA), perfluorooctanoate (PFOA), perfluorononanoate (PFNA), perfluorodecanoate (PFDcA), perfluoroundecanoate (PFUnA), perfluorododecanoate (PFDoA), perfluorotridecanoate (PFDTriA), perfluorotetradecanoate (PFTeA), perfluoropentadecanoate (PFPeDA), perfluorobutane

sulfonate (PFBS), perfluorohexane sulfonate (PFHxS), perfluorooctane sulfonate (PFOS), perfluorodecane sulfonate (PFDcS), perfluorooctane sulfonamide (PFOSA) and 6:2 fluorotelomer sulfonate (6:2 FTS). 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). Furthermore, total Hg, stable isotopes ($\delta^{15}N$ and $\delta^{13}C$) and vitamin A (retinol) and E (α-tocopherol) was analysed and eggshell thickness was determined.

4.2.1 Preparation of samples

The eggshell was thoroughly removed and the foetus or embryo (henceforth embryo) was removed from the thawed egg and weighed when visably present. Subsequently, the whole egg including the embryo was homogenised individually using a food blender (Melissa, Adexi group, Risskov, Denmark or Waring Commercial Laboratory Blender, Waring Laboratory, Torrington, CT, USA). Homogenate was separated into aliquots for different analyses and stored at -20 °C until analysed. Homogenate for vitamin analyses was kept in cryo tubes wrapped in aluminium foil to prevent light degradation of vitamins.

4.2.2 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) with mass spectrometer (MS) or electron capture detection (ECD). More details on the chromatographic separation and equipment is 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.

4.2.3 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 is given in Verreault et al. (2007).

For quality assurance were recovery rates of the stable isotope mass-labelled internal standards 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.

4.2.4 Analyses of Hg

The analyses of Hg were performed by the National Veterinary Institute (Oslo, Norway). Samples were decomposed with a mixture of nitric acid and hydrogen peroxide in a closed system using a microwave oven (Ethos Plus Microwave Labstation, Milestone Inc., Bergamo, Italy). The amount of Hg in the sample was determined using cold vapour atomic absorption spectrometry (CVAAS; Varian SpectrAA 600, Varian Inc., Palo Alto, CA, USA), using tin(II) chloride ($SnCl_2$) to reduce the Hg. The method is described in detail in Sturman (1985).

The laboratory is accredited by Norwegian Accreditation (Kjeller, Norway) according to NS-EN ISO/IEC 17025, and the laboratory's accredited analytical quality has been approved in several international intercalibration tests. Analyses of certified reference materials such as TORT-2, LUTS-1 and DORM-2 together with the samples gave acceptable results.

4.2.5 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 dichlormethane 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).

4.2.6 Analyses of vitamins

The vitamin analyses were carried out at the Department of Biology, Norwegian University of Science and Technology (Trondheim, Norway). The extraction of retinol and α-tocopherol was conducted in red light to prevent degradation of the vitamins. Samples were extracted three times with hexane using a high intensity ultrasonic processor (GEX400, Sonics and Materials, Inc., Newtown, CT, USA). The extract was evaporated to dryness and mobile phase (98:2% methanol:water) was added. The concentrations of retinol and α-tocopherol were determined by high-performance liquid chromatography (HPLC). More details on the extraction procedure and quantification is given in Murvoll et al. (2005).

All samples were extracted and analysed either in duplicate or triplicate and the coefficient of variation (CV%) was <20% for all samples. Control and blank samples were also analysed and acceptable results were achieved.

4.2.7 Eggshell thickness

The inner membrane of the eggshell was removed from the shell using running tap water and careful washing. Subsequently the egg was left to dry at room temperature for minimum two weeks. Eggshell thickness was then measured at or near the equator using a spring-loaded micrometer (0-25) with an accuracy of 0.01 mm. The mean of four measurements was recorded as the eggshell thickness.

4.3 Statistical analyses

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 analysed 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 with lipid content (%) included as a covariate.

Multivariate data analyses were performed using the multivariate program Unscrambler (version 9.2, Camo AS, Oslo, Norway). Principal component analyses (PCA) were conducted to consider differences and similarities between colonies and to evaluate intracorrelations. Multivariate regression; projection to latent structure (PLS), was carried out to evaluate associations between the response variables (vitamins and eggshell thickness) and the biological and contaminant variables for eggs from the Russian Arctic. More details on the statistical treatment of the data is given in Miljeteig (2007).

5. Results

The compounds α- and γ-HCH, PFHxA, PFHpA, PFBS, heptachlor and aldrin were below the detection limit in all samples analysed and are therefore not reported. *p,p'*-DDD, *o,p'*-DDT, *cis*-chlordane, BDE-209, PFOSA and 6:2 FTS were detected in less than 60% of the samples and were thus excluded from statistical analyses.

5.1 Levels of contaminants

The chemical analyses of ivory gull egg samples showed the presence of several major classes of contaminants. Concentrations of all quantified contaminants in the four colonies are summarised in table 1. OC contaminants were dominating the contaminant profile, with particularly PCBs (PCB-99, -118, -138, -153, -180, -183) and *p,p'*-DDE present in the highest concentrations. The two chlordane compounds, oxychlordane and *trans*-nonachlor, were also found in relatively high concentrations, as well as the toxaphenes CHB-26 and -50. Also PFASs and BFRs were present, however in considerably lower concentrations.

A PCA revealed a high degree of correlation between the majority of the OCs and BFRs, as indicated by the large extent of clustering in the loading plot (cluster 2 in figure 2). Cluster 1, with a high loading along PC2, contained the majority of the PFASs and showed a different distribution, and was thus not associated with the OCs and BFRs. A few compounds were separated from the main clusters, such as Hg, β-HCH, PFOA and PFNA. Hg showed a distribution similar to the PFAS-cluster, whereas β-HCH was negatively associated with Hg. PFNA was negatively associated with the OC/BFR-cluster and PFOA was not associated with any compound analysed. The score plot (figure 2) indicated colony differences in distribution of contaminants along PC1, with the highest levels of OCs and BFRs (the contaminants important along PC1) in the Nagurskoe colony, intermediate levels in the Svenskøya and Klyuv Cape colonies and the lowest levels in the Domashny colony (figure 3). Hg (figure 4) and most PFAS, however, did not differ between colonies (as seen along PC2 in figure 2).

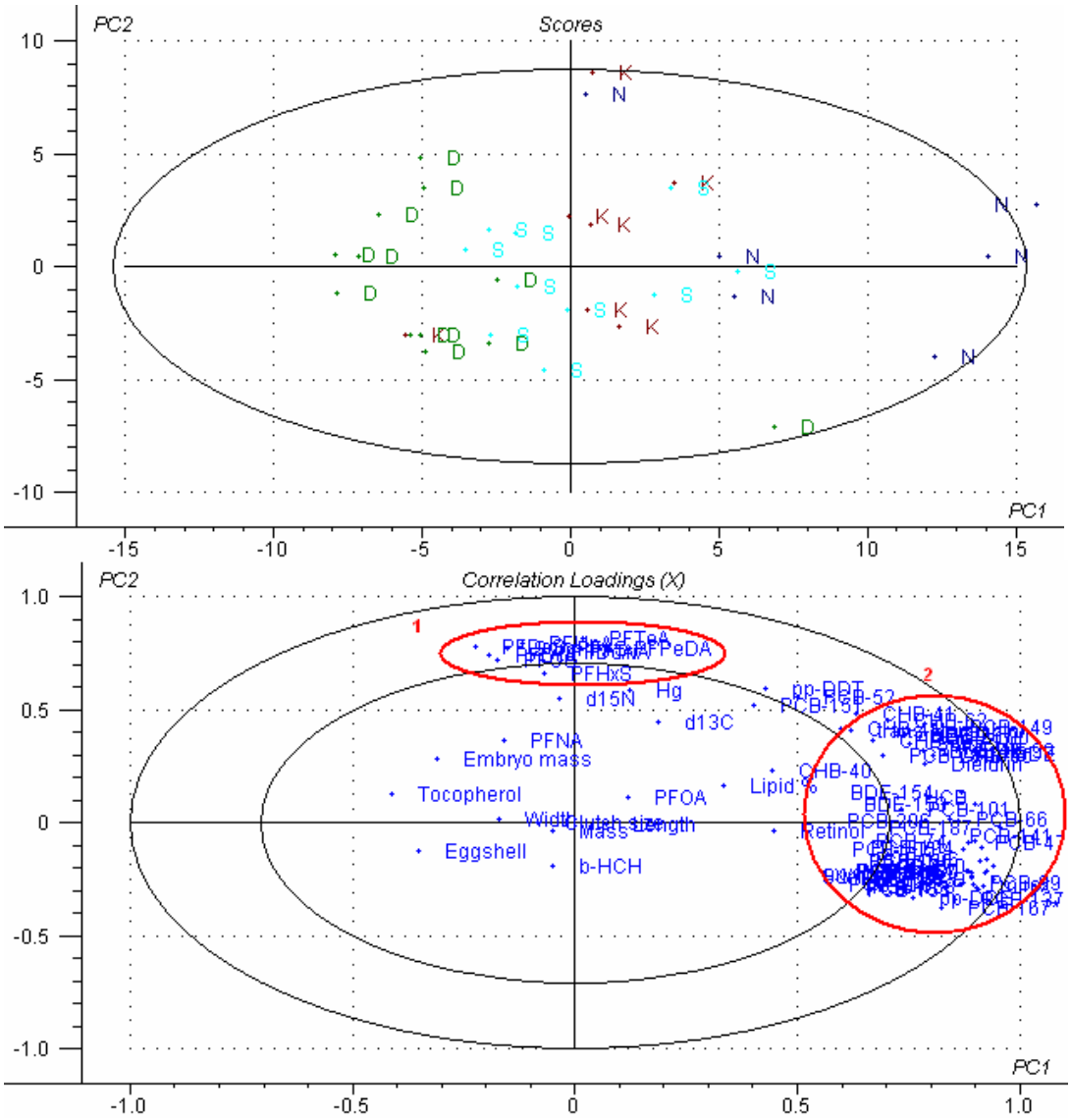

Figure 2 Score plot and loading plot from principal component analysis (PCA) of contaminants and biological variables measured in ivory gull *Pagophila eburnea* eggs from Svalbard and the Russian Arctic (n=35). PC1 explains 46% and PC2 explains 15% of the variance in the data set (validated: 50%). S, N, K and D in the score plot designate the Svenskøya, Nagurskoe, Klyuv Cape and Domashny colonies, respectively. Cluster 1 contains the majority of PFASs, cluster 2 contains the majority of OCs (PCBs, toxaphenes and other chlorinated pesticides) and BFRs. The analysis is based on wet weight values.

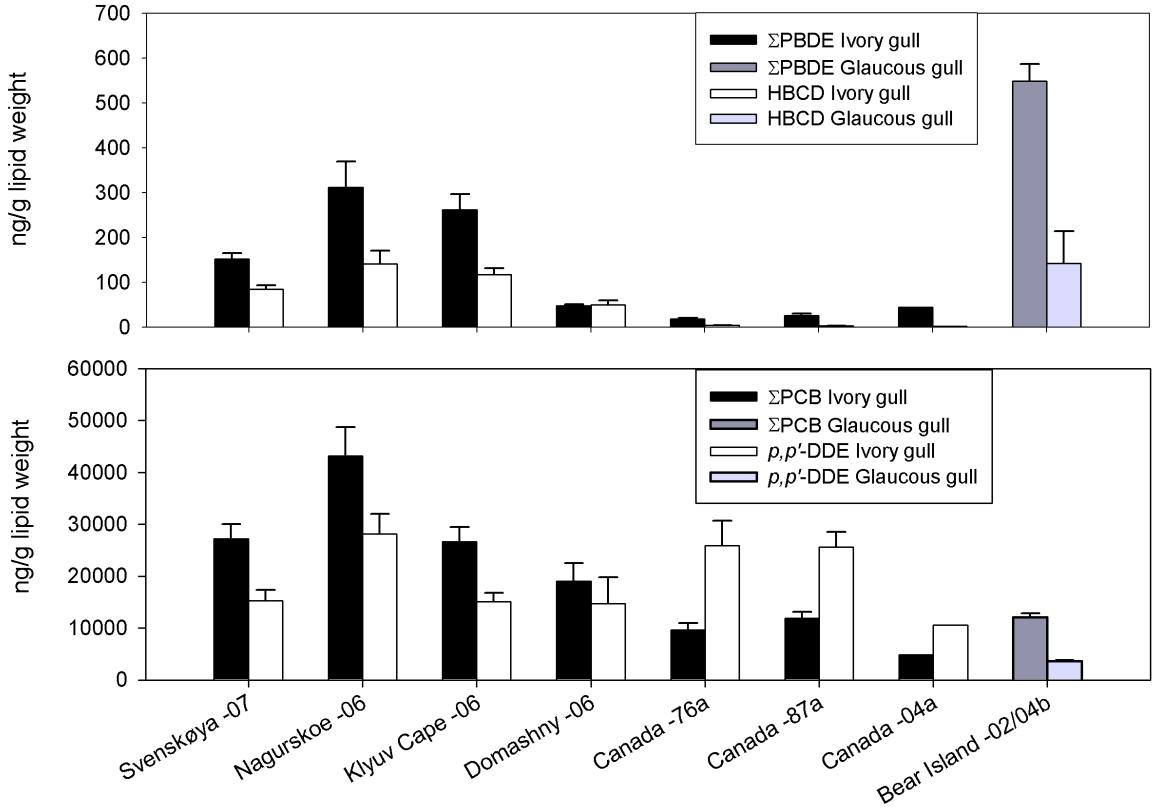

Figure 3 Comparison of mean concentrations with standard error of mean (±SEM) for ΣBDE, HBCD, ΣPCB and *p,p'*-DDE in ivory gull eggs from Svalbard, Russia and Canada and glaucous gull eggs from Bear Island. a denotes data from Braune et al. 2007, b denotes data from Verreault et al. 2004.

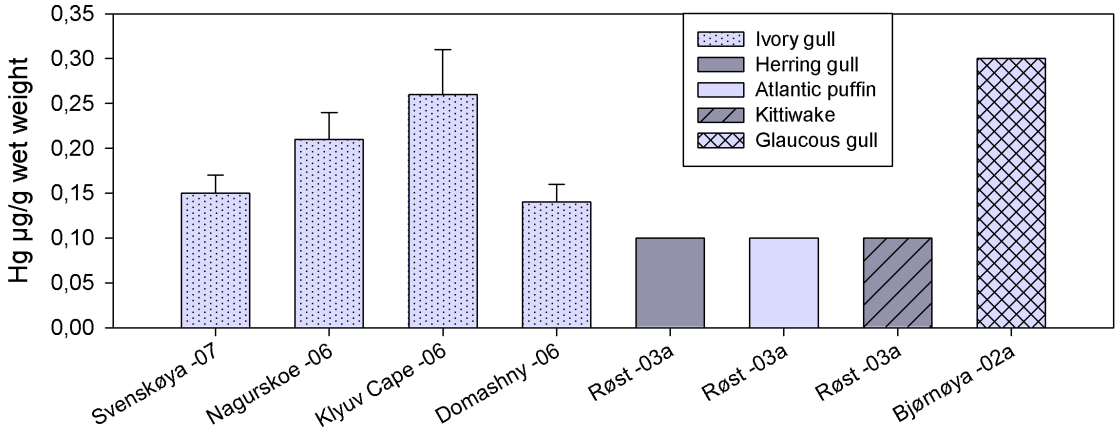

Figure 4 Comparison of mean concentrations with standard error of mean (±SEM) for mercury in ivory gull eggs from Svalbard and Russia, herring gull, Atlantic puffin and black-legged kittiwake eggs from Røst and glaucous gull eggs from Bear Island. a denotes data from Knudsen et al. 2005.

Table 1 Arithmetic mean with standard deviation (±SD) and ranges (min-max) for contaminant concentrations (ng/g ww) analysed in ivory gull egg homogenate samples from the Svenskøya colony on Svalbard and the Nagurskoe, Klyuv Cape and Domashny colonies in the Russian Arctic. Sample size is n=10, n=6, n=7 and n=12, respectively. Exceptions are BDE-209, PFOA, PFTeA, PFDcS and 6:2 FTS, where some individuals had levels below the detection limit, thus the sample size for BDE-209 were n=0, n=1, n=4 and n=2, for PFOA were n=0, n=5, n=6 and n=11, for PFTeA and PFDcS were n=9, n=6, n=7 and n=12 and for 6:2 FTS were n=0, n=4, n=3 and n=5 for the Svenskøya, Nagurskoe, Klyuv Cape and Domashny colonies, respectively. Due to small changes in the detection limit were PFOSA detected only in eggs from the Svenskøya colony (n=8). nd designates not detected. Dieldrin and PCB-167 was not analysed in eggs from the Svenskøya colony, whereas *cis*-chlordane, *p,p'*-DDD and *o,p'*-DDT was not analysed in eggs from the three colonies in the Russian Arctic, as designated by na (not analysed).

	Svenskøya			**Nagurskoe**			**Klyuv Cape**			**Domashny**		
	Mean ± SD	Median	Min - Max	Mean ± SD	Median	Min - Max	Mean ± SD	Median	Min - Max	Mean ± SD	Median	Min - Max
Lipid %	9.95 ± 1.18	9.88	8.63 - 12.3	10.5 ± 1.0	10.3	9.46 - 12.1	9.15 ± 0.99	9.1	8.11 - 10.4	9.57 ± 1.39	9.95	6.95 - 11.5
δ^{13}**C**	-20.2 ± 0.3	-20.2	-20.8 - -19.7	-20.1 ± 0.3	-20.3	-20.4 - -19.5	-20.0 ± 0.4	-20.1	-20.6 - -19.4	-20.6 ± 0.5	-20.6	-21.3 - -19.7
δ^{15}**N**	15.8 ± 0.3	15.8	15.3 - 16.2	16.2 ± 0.9	15.8	15.3 - 17.6	16.5 ± 0.6	16.4	15.9 - 17.7	16.2 ± 0.7	16.3	14.7 - 17.2
***p,p'*-DDT**	25.6 ± 12.5	22.7	10.1 - 47.0	40.1 ± 5.7	40.9	32.4 - 45.7	39.9 ± 19.1	32.5	20.9 - 69.5	23.7 ± 7.31	21.2	18.3 - 45.1
***p,p'*-DDE**	1,510 ± 700	1,330	933 - 3,240	2,910 ± 910	3,180	1,530 - 3,860	1,360 ± 340	1,370	813 - 1,920	1,460 ± 1,930	984	341 - 7,410
***p,p'*-DDD**	1.80 ± 2.48	0.49	nd - 6.89	na	na	na	na	na	na	na	na	na
***o,p'*-DDT**	3.32 ± 5.39	0.51	nd - 16.6	na	na	na	na	na	na	na	na	na
Oxychlordane	165 ± 69	156	95.3 - 319	287 ± 98	310	130 - 413	141 ± 34	142	98.9 - 188	134 ± 71	104	45.0 – 285
***trans*-nonachlor**	77.6 ± 37.7	67.1	39.7 - 170	177 ± 174	107	75.6 - 527	112 ± 52	124	26.5 - 175	36.0 ± 13.1	29.7	20.5 - 58.3
***cis*-chlordane**	23.1 ± 12.0	21.2	8.10 - 51.7	na	na	na	na	na	na	na	na	na
β-HCH	11.4 ± 3.5	10.6	6.47 - 17.9	18.1 ± 6.9	16.7	10.4 - 30.6	15.4 ± 8.7	11.6	8.88 - 31.5	28.4 ± 6.9	28.3	18.8 - 45.1
Mirex	31.2 ± 7.8	30.1	21.5 - 46.7	50.2 ± 13.6	49.6	28.3 - 67.7	30.6 ± 7.7	31	21.0 - 43.2	23.4 ± 14.1	19.5	13.2 - 65.4
Dieldrin	na	na	na	78.4 ± 46.3	61.9	51.7 - 172	43.4 ± 12.8	44.3	21.3 - 59.3	24.6 ± 9.1	23.1	13.7 - 44.2
HCB	62.1 ± 20.7	55.1	45.3 - 113	97.4 ± 11.2	97.4	79.2 - 112	59.1 ± 17.4	55.8	41.0 - 90.9	62.4 ± 15.9	59.8	42.4 - 93.5
CHB-26	38.6 ± 33.6	30.6	13.1 - 131	101 ± 105	63.9	40.2 - 314	47.5 ± 24.6	38.4	12.5 - 86.7	14.5 ± 5.83	13.2	7.01 - 24.9
CHB-40	7.26 ± 2.39	6.97	4.38 - 11.9	5.41 ± 1.03	5.41	4.27 - 6.65	3.98 ± 0.96	4.09	2.52 - 5.47	2.54 ± 0.80	2.42	1.56 - 3.94
CHB-41	3.53 ± 1.78	2.99	1.81 - 7.66	7.08 ± 4.98	5.12	3.18 - 16.7	4.87 ± 2.37	4.13	2.04 - 9.21	1.94 ± 0.64	1.83	1.30 - 3.69
CHB-44	6.32 ± 4.71	5.31	2.24 - 18.5	13.5 ± 15.1	7.14	5.33 - 44.2	8.09 ± 3.67	8.37	2.23 - 14.0	2.83 ± 0.87	2.77	2.02 - 5.16
CHB-50	75.3 ± 53.3	56.7	26.9 - 213	166 ± 94	144	82.2 - 349	72.0 ± 27.9	84.9	30.0 - 102	28.3 ± 9.78	30.7	13.7 - 43.4
CHB-62	9.70 ± 5.65	8.77	4.53 - 23.2	24.6 ± 8.0	22.8	15.3 - 39.3	17.1 ± 7.2	17.3	7.37 - 26.1	9.59 ± 4.10	8.42	3.35 - 16.9
$\sum_6$**CHB**	140 ± 100	111	54.1 - 405	318 ± 225	244	154 - 770	154 ± 60	155	57.2 - 221	59.7 ± 20.1	57.5	34.1 - 92.4
PCB-28	4.15 ± 1.50	3.83	1.95 - 7.21	5.86 ± 2.14	5.09	3.97 - 9.32	4.52 ± 1.48	4.11	2.31 - 6.97	4.10 ± 1.02	3.78	3.03 - 6.02
PCB-47	14.6 ± 7.0	11.7	9.82 - 27.9	33.5 ± 15.8	29.1	16.1 - 59.4	14.9 ± 3.3	16.5	8.12 - 17.6	14.4 ± 12.0	11.0	5.28 - 49.9
PCB-52	5.66 ± 2.57	6.41	2.33 - 9.51	14.3 ± 8.3	12.0	6.08 - 25.6	12.2 ± 13.5	4.52	2.46 - 36.2	3.28 ± 2.44	2.51	1.47 - 10.4
PCB-66	15.5 ± 5.7	13.5	10.4 - 26.4	31.3 ± 14.7	27.5	15.4 - 57.7	16.2 ± 2.8	17.1	10.6 - 18.8	15.1 ± 5.06	13.02	8.71 - 24.2

	Svenskøya			Nagurskoe			Klyuv Cape			Domashny		
	Mean ± SD	Median	Min - Max	Mean ± SD	Median	Min - Max	Mean ± SD	Median	Min - Max	Mean ± SD	Median	Min - Max
PCB-74	21.3 ± 7.72	18.5	13.5 - 35.4	33.3 ± 12.8	30.9	19.6 - 54.7	18.8 ± 4.1	19.7	10.7 - 23.8	16.9 ± 9.5	14.2	7.62 - 43.6
PCB-99	168 ± 71	151	99.3 - 346	362 ± 122	390	173 - 503	184 ± 44	191	120 - 260	175 ± 140	137	67.2 - 584
PCB-101	9.51 ± 3.68	9.40	4.53 - 18.0	18.3 ± 9.3	15.3	9.79 - 35.1	12.2 ± 9.2	8.67	5.18 - 31.1	6.15 ± 3.96	4.81	3.23 - 17.6
PCB-105	35.6 ± 11.8	31.6	21.9 - 57.4	69.0 ± 286	61.5	36.9 - 116	34.0 ± 6.6	35.7	20.5 - 40.4	32.0 ± 14.8	28.2	13.5 - 60.0
PCB-114	4.11 ± 0.82	3.85	3.16 - 5.21	5.51 ± 1.93	5.46	2.91 - 8.42	2.77 ± 0.56	2.81	1.73 - 3.52	2.71 ± 1.56	2.35	1.12 - 6.87
PCB-118	134 ± 40	125	89.1 - 188	244 ± 92	213	134 - 381	118 ± 24	125	74.7 - 145	119 ± 78.7	97.7	42.8 - 326
PCB-128	33.7 ± 21.5	25.7	13.2 - 79.9	94.8 ± 29.8	106	45.6 - 119	44.6 ± 15.3	44.9	21.3 - 70.6	36.2 ± 33.8	28.3	8.60 - 131
PCB-137	32.6 ± 11.2	31.3	18.9 - 57.3	66.6 ± 22.2	70.5	30.5 - 90.6	34.5 ± 7.0	36.4	24.6 - 45.3	32.3 ± 28.0	24.8	12.3 - 116
PCB-138	546 ± 213	522	312 - 1,030	903 ± 272	986	448 - 1,190	478 ± 117	491	302 - 662	382 ± 307	303	148 - 1,280
PCB-141	1.36 ± 0.37	1.32	0.81 - 2.05	3.07 ± 1.54	2.47	1.47 - 5.37	1.59 ± 0.54	1.61	0.78 - 2.41	0.87 ± 0.55	0.68	0.50 - 2.51
PCB-149	25.7 ± 7.5	25.4	15.5 - 41.1	44.3 ± 11.6	42.9	30.8 - 64.0	32.2 ± 15.1	28.1	12.9 - 61.3	17.7 ± 7.7	14.4	9.93 - 36.4
PCB-151	3.06 ± 1.84	3.04	0.74 - 6.54	7.02 ± 5.83	3.97	2.67 - 17.1	7.34 ± 9.79	2.51	0.87 - 27.8	1.10 ± 1.38	0.67	0.38 - 5.40
PCB-153	957 ± 346	892	566 - 1,770	1,410 ± 420	1,500	690 - 1,900	768 ± 172	794	516 - 1,020	610 ± 449	490	274 – 1,940
PCB-156	29.2 ± 8.5	26.8	19.4 - 43.7	42.9 ± 14.8	42.9	21.6 - 60.2	22.5 ± 5.3	22.1	14.3 - 30.7	16.4 ± 7.7	15.1	7.51 - 36.1
PCB-157	11.0 ± 3.6	10.4	6.88 - 18.9	16.7 ± 5.4	17.1	8.23 - 23.1	8.56 ± 2.11	8.53	5.33 - 12.0	7.09 ± 4.27	5.99	2.81 - 19.0
PCB-167	na	na	na	16.5 ± 5.6	14.8	9.74 - 23.4	7.60 ± 1.45	7.55	4.97 - 9.26	7.68 ± 6.69	6.40	1.96 - 26.6
PCB-170	105 ± 39	95.8	62.3 - 195	188 ± 64	185	94.5 - 281	102 ± 27	106	59.9 - 135	58.2 ± 41.2	48.4	25.1 - 181
PCB-180	341 ± 115	310	217 - 588	543 ± 178	536	267 - 800	298 ± 75	306	180 - 379	171 ± 127	139	78.2 - 557
PCB-183	61.3 ± 21.0	57.9	39.0 - 100	112 ± 34	112	61.8 - 162	62.2 ± 15.2	62.3	41.0 - 81.6	45.5 ± 38.7	35.4	19.6 - 163
PCB-187	44.5 ± 13.6	40.3	29.4 - 70.9	85.3 ± 21.8	79.8	64.4 - 121	49.7 ± 16.6	51.0	25.6 - 77.8	26.1 ± 26.0	19.4	10.4 - 107
PCB-189	3.51 ± 0.78	3.27	2.80 - 5.12	4.67 ± 1.63	4.55	2.24 - 7.05	2.34 ± 0.77	2.35	1.14 - 3.38	1.17 ± 1.01	0.93	0.38 - 4.15
PCB-194	38.7 ± 10.6	34.8	29.1 - 60.1	68.5 ± 24.3	65.0	32.6 - 101	37.3 ± 12.6	37.1	19.8 - 57.6	15.6 ± 9.2	13.9	8.68 - 43.5
PCB-196	24.6 ± 6.0	22.8	18.4 - 36.9	45.0 ± 16.3	42.7	23.5 - 67.9	24.1 ± 6.3	23.1	14.9 - 34.2	13.6 ± 8.9	11.6	6.88 - 40.9
PCB-206	7.28 ± 1.74	6.80	4.87 - 10.3	11.9 ± 3.5	10.8	7.34 - 16.3	6.54 ± 2.06	5.85	3.65 - 9.89	2.53 ± 0.76	2.36	1.66 - 4.62
$\sum_{10}$MO-PCB	218 ± 64	201	143 - 311	400 ± 147	358	216 - 613	196 ± 40	210	123 - 244	185 ± 114	157	70.3 - 478
$\sum_{28}$PCB	2,680 ± 930	2,520	1,660 - 4,800	4,480 ± 1,340	4,700	2,290 - 5,950	2,400 ± 550	2,460	1,500 - 3,130	1,830 ± 1,350	1,480	790 - 5,810
BDE-28	0.12 ± 0.06	0.10	0.05 - 0.22	0.29 ± 0.14	0.26	0.16 - 0.56	0.25 ± 0.11	0.25	0.06 - 0.38	0.05 ± 0.02	0.05	0.02 - 0.10
BDE-47	8.83 ± 3.19	7.98	5.66 - 13.9	21.6 ± 12.6	19.2	9.47 - 43.6	16.1 ± 6.75	17	4.72 - 24.9	2.72 ± 1.13	2.33	1.26 - 4.83
BDE-99	1.82 ± 0.56	1.72	1.09 - 3.15	3.45 ± 1.12	3.17	2.38 - 5.54	2.43 ± 0.8	2.76	0.92 - 3.39	0.60 ± 0.23	0.52	0.33 - 1.02
BDE-100	1.11 ± 0.32	1.14	0.60 - 1.53	1.87 ± 0.86	1.73	0.99 - 3.46	1.43 ± 0.63	1.37	0.31 - 2.37	0.29 ± 0.11	0.26	0.16 - 0.50
BDE-153	1.34 ± 0.35	1.31	0.77 - 2.01	1.99 ± 0.54	2.01	1.17 - 2.62	1.33 ± 0.42	1.28	0.56 - 1.84	0.43 ± 0.28	0.34	0.19 - 1.27
BDE-154	1.84 ± 0.54	1.62	1.13 - 2.87	3.65 ± 1.12	3.42	2.19 - 5.39	2.35 ± 0.85	2.4	0.78 - 3.42	0.49 ± 0.21	0.43	0.26 - 1.04

	Svenskøya			Nagurskoe			Klyuv Cape			Domashny		
	Mean ± SD	Median	Min - Max	Mean ± SD	Median	Min - Max	Mean ± SD	Median	Min - Max	Mean ± SD	Median	Min - Max
BDE-209	nd	nd	nd	0.031	0.031	0.031	0.051 ± 0.026	0.053	0.021 - 0.075	0.04 ± 0.01	0.04	0.03 - 0.04
$\sum_7$BDE	15.1 ± 4.4	13.9	9.35 - 23.5	32.9 ± 15.3	30.8	16.7 - 58.9	23.9 ± 8.93	26.2	7.35 - 35.9	4.59 ± 1.58	4.43	2.52 - 6.44
HBCD	8.41 ± 2.91	7.52	3.81 - 12.9	14.9 ± 8.0	14.1	7.19 - 28.4	10.6 ± 3.3	11.7	4.40 - 14.0	4.74 ± 3.20	3.85	1.42 - 11.9
PFOA	nd	nd	nd	0.32 ± 0.06	0.34	0.25 - 0.40	0.25 ± 0.06	0.24	0.17 - 0.31	0.24 ± 0.06	0.22	0.16 - 0.37
PFNA	1.25 ± 0.66	1.04	0.40 - 2.70	1.19 ± 0.31	1.34	0.77 - 1.48	0.94 ± 0.19	0.99	0.65 - 1.21	1.44 ± 0.39	1.49	0.83 - 2.15
PFDcA	2.48 ± 1.04	2.41	0.85 - 4.35	3.03 ± 1.50	3.11	1.42 - 5.63	2.86 ± 1.19	3.36	1.10 - 4.43	3.26 ± 1.36	3.50	1.21 - 5.61
PFUnA	12.1 ± 5.2	12.6	3.16 - 19.1	12.9 ± 7.0	12.9	5.82 - 24.7	10.8 ± 4.7	11.7	4.66 - 17.6	12.0 ± 5.2	10.7	4.84 - 20.8
PFDoA	2.99 ± 1.06	3.44	0.94 - 4.09	2.50 ± 1.71	2.29	1.04 - 5.71	2.08 ± 1.15	2.12	0.89 - 3.98	2.00 ± 0.96	1.51	0.87 - 3.65
PFTriA	10.7 ± 4.5	10.7	2.7 - 17.7	9.61 ± 5.59	8.21	4.80 - 19.8	7.89 ± 4.66	7.86	3.54 - 15.9	6.98 ± 3.16	5.67	3.43 - 13.2
PFTeA	0.90 ± 0.34	0.88	0.41 - 1.37	1.27 ± 0.63	1.07	0.80 - 2.46	1.08 ± 0.78	0.97	0.31 - 2.61	0.88 ± 0.40	0.77	0.43 - 1.72
PFPeDA	0.59 ± 0.27	0.58	0.14 - 1.0	1.52 ± 0.99	1.19	0.85 - 3.50	1.22 ± 0.88	0.91	0.53 - 3.07	0.92 ± 0.45	0.76	0.48 - 1.69
PFHxS	0.49 ± 0.39	0.37	0.19 - 1.46	0.79 ± 0.36	0.77	0.30 - 1.38	0.66 ± 0.4	0.69	0.24 - 1.31	0.83 ± 0.44	0.79	0.30 - 1.90
PFOS	72.6 ± 30.5	79.2	24.2 - 113	55.8 ± 23.6	59.1	25.2 - 89.9	56.2 ± 29.4	66.1	20.9 - 97.3	66.5 ± 32.3	57.7	17.7 - 117
PFDcS	0.45 ± 0.19	0.47	0.22 - 0.76	0.62 ± 0.37	0.62	0.25 - 1.27	0.75 ± 0.41	0.86	0.28 - 1.32	0.79 ± 0.51	0.68	0.21 - 1.80
PFOSA	0.06 ± 0.06	0.05	0.03 - 0.20	nd	nd	nd	nd	nd	nd	nd	nd	nd
6:2 FTS	na	na	na	0.26 ± 0.04	0.25	0.22 - 0.32	0.29 ± 0.07	0.28	0.23 - 0.37	0.37 ± 0.07	0.37	0.27 - 0.47
$\sum_{12}$PFAS	104 ± 42	116	32.7 - 157	89.6 ± 41.2	91.3	42.7 - 156	84.8 ± 43.1	96.8	34.8 - 149	96.0 ± 44.0	86.9	30.8 - 164
Hg	0.15 ± 0.05	0.14	0.08 - 0.24	0.21 ± 0.06	0.23	0.08 - 0.24	0.26 ± 0.12	0.20	0.16 - 0.48	0.14 ± 0.07	0.11	0.06 - 0.30

5.2 Pattern

Clearly, the concentration differences between colonies contributed largely to the colony differentiation in the PCA of the contaminants. To evaluate pattern differences in distribution of compounds the data set was factor-normalised to exclude differences in concentrations between individuals, and a new PCA was performed. The score plot indicated that it was not possible to discriminate between the Svenskøya, Nagurskoe and Klyuv Cape colonies with respect to contaminant pattern. There was, however, a clear distinction between Svalbard and Franz Josef Land (Svenskøya, Nagurskoe and Klyuv Cape) and Severnaya Zemlya (Domashny), explained by PC2. The eggs from Svalbard and Franz Josef Land contained a higher proportion of BDEs compared to the eggs from Severnaya Zemlya, in addition to HBCD, a few toxaphenes (CHB-26, CHB-50 and Σ_6CHB) and an assortment of higher-chlorinated PCBs, such as PCB-170, -180, -187, -189, -194, -196 and -206. The eggs from Severnaya Zemlya on the other hand appeared to contain a larger proportion of MO-PCBs, along with PCB-99 and -137, oxychlordane and β-HCH. A PCB-99 to PCB-180 ratio can be used as an indicator of high versus low chlorinated PCB distribution (Letcher et al. 1995; Andersen et al. 2001). The PCB-99:PCB-180 ratios were 0.49, 0.67, 0.62 and 1.0 in the Svenskøya, Nagurskoe, Klyuv Cape and Domashny colonies respectively, supporting the trend of a decreasing proportion of higher chlorinated PCBs from west to east seen from the PCA. PFASs did not differ between colonies in neither concentration nor pattern. The colony differences were however minor, in an overall view the relative contribution of compound classes to the total contaminant burden followed PCB (49-54%) > OCP (37-41%) > Hg (2.7-5.8%) > CHB (2.0-3.6%) > PFAS (1.2-3.5%) > BFR (0.3-0.7%) for all four colonies.

Within compound classes, the distribution appeared to be similar between colonies (figure 5). Σ_{28}PCB consisted mainly of the congeners PCB-153 (31-36%) > -138 (20%) > -180 (9.5-13%) ≥ -99 (6.2-9.3%) > -118 (4.9-6.5%), which together constituted 77-80% of the total PCB concentration. The relative contribution of the main perfluorinated compounds to Σ_{12}PFAS followed PFOS (63-69%) > PFUnA (11-14%) > PFTriA (7.5-11%) > PFDcA (2.4-3.5%), which together contribute to 91-94% of Σ_{12}PFAS. An exception to the general pattern was BFR, where the relative contribution in particular from HBCD and BDE-47 varied between colonies. The four major compounds and congeners constituting 89-91% of ΣBFR were BDE-47 (37%) > HBCD (35%) > BDE-154 (8.1%) > BDE-99 (7.9%) for the Svenskøya colony, BDE-47 (44%) > HBCD (31%) > BDE-154 (8.7%) > BDE-99 (7.8%) for the Nagurskoe colony, BDE-47 (45%) > HBCD (32%) > BDE-99 (7.2%) ≈ BDE-154 (7.0%) for the Klyuv Cape colony and HBCD (47%) > BDE-47 (31%) > BDE-99 (6.8%) > BDE-154 (5.9%) for the Domashny colony.

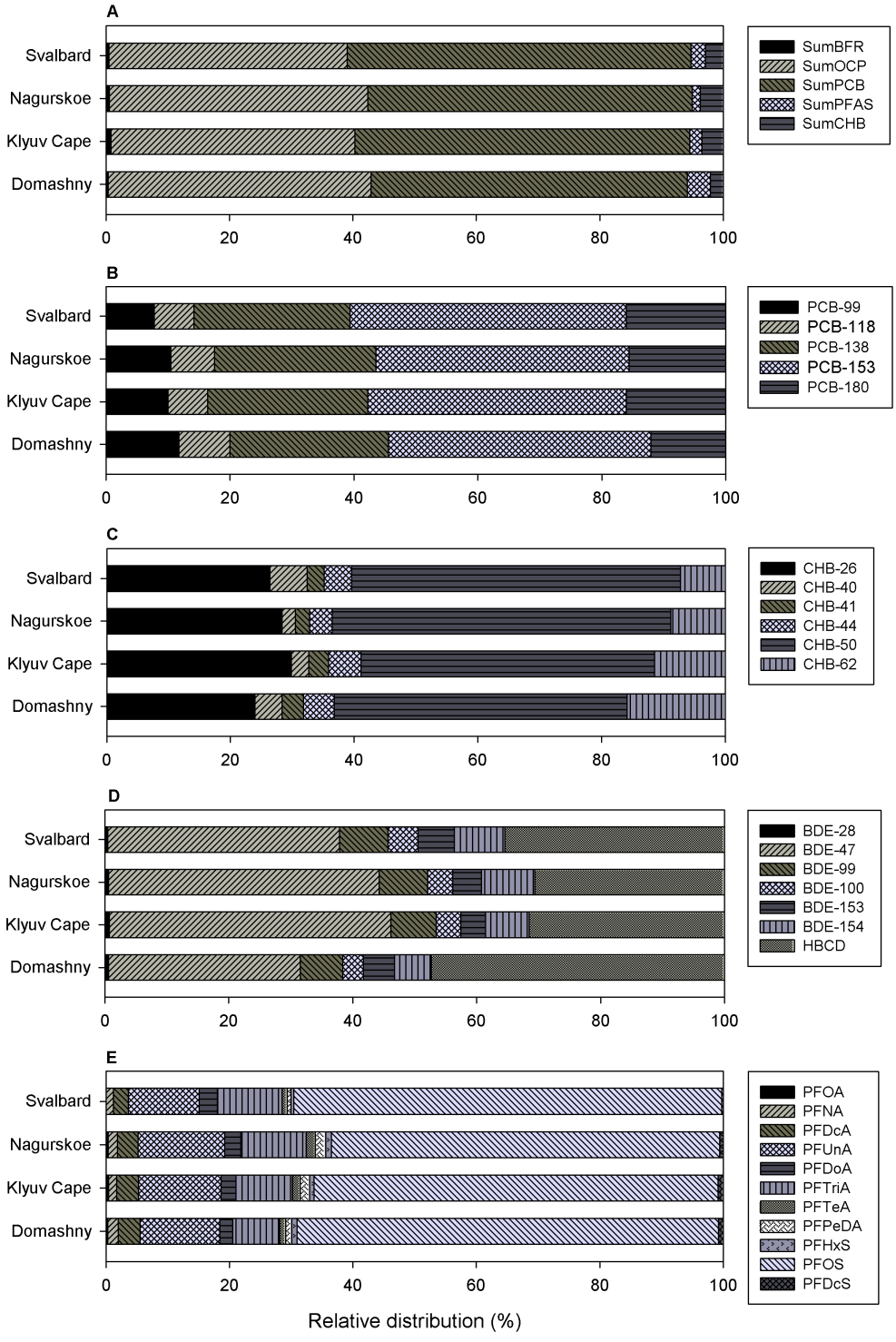

Figure 5 Relative distribution (%) of various classes or compounds in ivory gull *Pagophila eburnea* eggs from Svenskøya (n=10) colony on Svalbard and Nagurskoe (n=6), Klyuv Cape (n=7) and Domashny (n=12) colonies in the Russian Arctic. The graph is based on wet weight values. **A:** Relative distribution (%) of OCPs, PCBs, BFRs, PFASs and CHBs to total organohalogen. **B:** Relative distribution (%) of a selection of PCBs (main contributing congeners to Σ_{28}PCB (>5%)). **C:** Relative distribution (%) of CHBs to Σ_6CHB. **D:** Relative distribution (%) of BDEs and HBCD to ΣBFRs. **D:** Relative distribution (%) of PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFTriA, PFTeA, PFPeDA, PFHxS, PFOS and PFDcS to Σ_{12}PFASs.

5.3 Associations with response variables

A PLS analysis of retinol explained by all biological and contaminant variables, with a stepwise reduction of variables of low importance, resulted in a model explaining 65% (validated: 44%, q^2=0.42) of the variance in retinol with three PCs. A q^2-value of 0.42 indicate that this is an acceptable model for biological data (Lundstedt et al. 1998). Of the 26 x-variables remaining in the model were BDE-28, -47, -99, -100, Σ_7BDE, HBCD, PCB-28, -66, -128, -138, -153, -156, -170, -180, -189, -196, Σ_{28}PCB, mirex, oxychlordane and lipid content positively associated with retinol, whereas PFHxS, PFDcS, PFOS, Σ_{12}PFAS and embryo mass were negatively associated with retinol.

A PLS analysis of α-tocopherol explained by all biological and contaminant variables was performed, followed by a stepwise reduction of variables of low importance and a successive increase of q^2. This resulted in a model explaining 67% (validated: 51%, q^2=0.48) of the variance in α-tocopherol with two PCs. The q^2-value indicate that this was an acceptable model (Lundstedt et al. 1998). The ten x-variables remaining in the model were *p,p*'-DDE, *p,p*'-DDT, PCB-66, -101, -151, PFUnA, PFDoA, PFTriA and Hg. Of these were the OCs and Hg were negatively associated with α-tocopherol and the PFASs and lipid content were positively associated with α-tocopherol.

A PLS analysis with a successive improvement of the model resulted in a model explaining 56% (validated: 36%, q^2=0.33) of the variance in eggshell thickness with two PCs. A q^2-value of 0.33 indicated that this was a close to acceptable model based on biological data (Lundstedt et al. 1998). The 21 x-variables remaining in the model were clutch size, oxychlordane, *trans*-nonachlor, *p,p*'-DDE, PCB-28, -47, -99, -128, -137, -138, -170, -189, -194, -206, BDE-28, -47, HBCD, PFTeA and PFDcS, where all the variables were negatively associated with eggshell thickness.

6. Discussion

6.1 Levels of contaminants

Colony differences in contaminant levels were found for the majority of the analysed compounds, including most OCs and BFRs (figure 2, table 1). Concentrations of PFASs and Hg did generally not vary between colonies. Eggs from the Nagurskoe colony (Franz Josef Land) contained the highest levels of most OCs and BFRs, whereas the eggs from the Domashny colony (Severnaya Zemlya) displayed the lowest levels. In contrast, β-HCH showed an opposite distribution with approximately twice as high levels in the Domashny colony, when compared to levels in the two colonies in Franz Josef Land (Nagurskoe and Klyuv Cape) and nearly three times as high levels as in the Svenskøya colony on Svalbard. These distinct differences in contaminant levels within the remote Arctic may indicate local sources of contamination in the proximity of the Franz Josef Land colonies. Alternatively, the differences may be due to differential long range transport of contaminants, leading to exposure to different levels of contaminants on the breeding grounds or in wintering areas, or both. The ivory gull moves over large areas during the year (Haney and MacDonald 1995), thus the colony differences may be a result of regional differences in food web composition or contaminant levels. The ivory gull is an opportunistic feeder, foraging primarily on polar cod *Boreogadus saida* and crustaceans, as well as carrion of seals killed by polar bears *Ursus maritimus* and human waste (Haney and MacDonald 1995). Local variation in availability of the various food items may lead to regional differences in contaminant levels, as these food items contain different levels of biomagnifying contaminants.

The levels of contaminants measured in the ivory gull eggs from Svalbard and Russia were generally high in comparison with other studies on seabird eggs throughout the Arctic. This is in keeping with several studies of OC levels in polar bears (Andersen et al. 2001; Lie et al. 2003) and ringed seals *Phoca hispida* (Nakata et al. 1998; Muir et al. 2000), indicating that the western Russian Arctic is the most polluted region within the Arctic. This may be explained by the stronger association with multi-year sea ice in this region (Andersen et al. 2001) or be related to the continued use of PCBs and several pesticides in the former Soviet Union after ban in many other countries (Fedorov 1999) and the high extent of riverine input to the Kara Sea (de March et al. 1998).

A selection of organochlorine pesticides, including six toxaphene congeners, were analysed and found in relatively large quantities, with *p,p*'-DDE being the prevailing compound (table 1). The *p,p*'-DDE mean concentrations were higher in all four colonies than concentrations measured in ivory gull eggs from the Canadian Arctic sampled in 2004 (Braune et al. 2007, figure 3), and higher than or comparable to levels measured in eggs from a variety of seabird species sampled in the Barents Sea region over three decades with ten-year intervals (Barrett et al. 1996). Furthermore, the levels were four to eight times higher in the present study than reported in glaucous gull *Larus hyperboreus* eggs from Bear Island in 2004 (Verreault et al. 2004a). Similar indications of high levels of organochlorine pesticides were found when comparing the other organochlorine pesticides with other studies. Levels of chlordanes, mirex, β-HCH and HCB were higher than reported in glaucous gull eggs from Bear Island (Verreault et al. 2004a) and in eggs from a range of seabird species in the Barents Sea region (Barrett et al. 1996). The levels of these contaminants were also higher than or comparable to the levels measured in ivory gull eggs from the Canadian Arctic (Braune et al. 2007). Exceptions were the levels of chlordanes (oxychlordane and *trans*-nonachlor), which were lower in the Svenskøya and Domashny colonies compared to the ivory gull eggs from

Canada, and the levels of β-HCH, which were lower in the Svenskøya and Klyuv Cape colonies compared to the ivory gull eggs from Canada (Braune et al. 2007). Toxaphenes have not previously been quantified in ivory gull eggs. The level of Σ_6CHB in the Nagurskoe colony was however three times as high as the level of Σ_{21}CHB reported in glaucous gull eggs from Bear Island, whereas the level in the Domashny colony was approximately half of the level found in glaucous gull eggs (Verreault et al. 2004a).

The concentrations of Σ_{28}PCB in all four colonies were markedly higher than Σ_{85}PCB in ivory gull eggs from the Canadian Arctic sampled in 2004, 1987 and 1976; the concentrations were approximately four to eight times higher than the concentrations in the eggs from 2004 (Braune et al. 2007, figure 3). Furthermore, Σ_{28}PCB in all four colonies were clearly higher than Σ_{41}PCB reported in glaucous gull eggs from Bear Island sampled in 2004 (Verreault et al. 2004a) and higher than (Nagurskoe) or comparable to or slightly lower than (Klyuv Cape, Svenskøya and Domashny) Σ_{21}PCB reported in herring gull *Larus argentatus* and razorbill *Alca torda* eggs from northern Norway and glaucous gull eggs from Svalbard sampled in 1993 (Barrett et al. 1996).

The concentrations of individual BDE congeners in the two Franz Josef Land colonies (Svenskøya, Nagurskoe and Klyuv Cape) were markedly higher than individual BDE concentrations reported in ivory gull eggs from the Canadian Arctic sampled in 2004, whereas the Domashny colony generally displayed concentrations similar to those in Canada (Braune et al. 2007, figure 3). The Σ_7BDE levels in all four colonies were however lower than Σ_8BDE measured in herring gull eggs from northern Norway and glaucous gull eggs from Bear Island (Knudsen et al. 2005). The levels found in the Svenskøya, Nagurskoe and Klyuv Cape colonies were similar to levels in eggs from Atlantic puffin *Fratercula arctica* and black-legged kittiwake *Rissa tridactyla* in northern Norway (Knudsen et al. 2005), while they were clearly higher than reported in black guillemot *Cepphus grylle* eggs from east Greenland (Vorkamp et al. 2004). The distribution of HBCD was somewhat different, with less pronounced differences between colonies. The levels of HBCD were 20 to 70 times higher in ivory gull eggs from the Russian Arctic than in ivory gull eggs from Canada sampled in 2004 (Braune et al. 2007). The differences were less distinct when compared to seabird eggs from the European Arctic. Nagurskoe and Klyuv Cape displayed values similar to those reported in glaucous gull eggs from Bear Island, whereas the levels in the Svenskøya and Domashny colonies were lower (Verreault et al. 2004a). The levels in the Nagurskoe and Klyuv Cape colonies were higher than or similar to levels in eggs from herring gulls and black-legged kittiwakes from northern Norway, whereas the levels in the Svenskøya and Domashny colony were lower than in herring gulls and black-legged kittiwakes from northern Norway (Knudsen et al. 2005).

The presence of perfluorinated compounds has not previously been assessed in ivory gulls. The levels were similar in eggs from all four colonies and markedly higher than the levels of BFRs, with PFOS as the clearly dominating compound (table 1). Nevertheless, the levels of PFOS were about one order of magnitude lower than reported in common guillemot *Uria aalge* eggs from the Baltic Sea (Holmstrom et al. 2005), and less than half the concentration measured in glaucous gull eggs from Bear Island (Verreault et al. 2005). The concentrations of PFOS were however higher than reported in liver tissue from black-legged kittiwakes and glaucous gulls from eastern Canada (Tomy et al. 2004). Levels in eggs and liver have been shown to be fairly comparable in glaucous gulls (Verreault et al. 2005). Few studies report on perfluorinated compounds other than PFOS above the detection or quantification limit. These are now possible to quantify due to improvements in methods of extraction and quantification.

Generally, the levels of perfluorocarboxylic acids were found to be similar to or lower than levels reported in glaucous gull eggs from Bear Island (Verreault et al. 2005).

Hg was the only inorganic contaminant measured in the present study. The Hg concentrations did not differ between colonies and were roughly five to ten times lower than in reported in ivory gull eggs from the Canadian Arctic (Braune et al. 2006). The latter were however among the highest concentrations ever reported for seabird eggs from the Arctic. The concentrations were slightly lower than in northern fulmar *Fulmarus glacialis* eggs and similar to concentrations found in Brünnich's guillemot *Uria lomvia* and black-legged kittiwake eggs from Canada (Braune and Simon 2004). The concentrations were comparable to levels reported in glaucous gull eggs from Bear Island, herring gull, Atlantic puffin and black-legged kittiwake eggs from northern Norway (Knudsen et al. 2005, figure 4) and eggs from a variety of seabird species sampled from the Barents Sea region (Barrett et al. 1996).

6.2 Contaminant pattern

Not only were differences in concentration levels found between colonies, but also differences in contaminant pattern. Although the order of dominating compound classes did not differ regionally, the proportion of each group varied slightly between colonies. The colony on Svalbard and the two colonies on Franz Josef Land contained a higher proportion of BFRs, whereas PFASs contributed more to the total contaminant burden in eggs from the Domashny colony. The latter can be explained by the similar levels of PFASs in all four colonies, whereas the concentrations of other compounds varied largely between colonies. This further suggests that the levels of PFASs are a result of long range transport to the Arctic, and that the PFASs are evenly distributed in this part of the Arctic. The higher proportion of BFRs in the colonies on Svalbard and Franz Josef Land indicates a proportionally higher influx of BFRs to these colonies compared to the Domashny colony on Severnaya Zemlya, with Europe as a likely source region (Vinogradova 2000; de Wit et al. 2004). Nevertheless, PCBs and OCPs were undoubtedly the dominating compound groups in ivory gull eggs from all four colonies.

6.3 Associations with response variables

Elucidating effects from pollutants in wildlife is challenging. In nature, the biota is exposed to complex mixtures of anthropogenic contaminants (de Wit et al. 2004), the composition of which are qualitatively and quantitatively not fully known (Groten et al. 2001). Only a selection of compound classes and only a selection of compounds within these classes have been analysed and quantified. Impact of non-analysed compounds on the organism and possible interactions are not possible to account for. Furthermore, many of the congeners and compounds analysed were found to be highly correlated. This intercorrelation between variables complicates the elucidation of response from certain compounds (Esbensen 2001). Multivariate data analyses are generally a good approach for handling the vast amount of information associated with complex mixtures (Groten et al. 2001). A drawback is however the lack of certainty that the remaining variables are cause of the response or merely covary with a compound leading to the response.

Retinol, together with the other forms of vitamin A, is important for growth and development, reproduction, vision, epithelial maintenance and immune function (Zile 1998; Simms and Ross 2000). Due to the complex storage and transport mechanisms and the wide range of functions, vitamin A may be influenced by contaminants at several steps in the pathway from uptake to function, such as reduced dietary uptake, decreased liver stores or disruption of circulatory transport to tissues (Simms and Ross 2000). Thus, a range of contaminant-related

mechanisms may influence the retinol levels in many ways, with a possibility of both increase and decrease of retinol. The present study found positive associations between retinol and contaminant levels in eggs. This may indicate an increased mobilisation of retinol from the vitamin A stores. However, retinyl palmitate, the main storage form of vitamin A, was not possible to determine in the ivory gull eggs, thus the vitamin status was not possible to assess.

α-Tocopherol is the main chain-breaking membrane antioxidant and plays a major role in the cellular antioxidant defence system (Packer and Kagan 1993). Contaminants may enhance production of reactive oxygen species in several ways, i.e. through cytochrome P450 enzymes or disruption of the normal electron flow in the mitochondrial membrane (Boelsterli 2003), thus leading to a depletion of α-tocopherol (Di Mascio et al. 1991). This supports the findings in the present study with decreasing concentrations of α-tocopherol with increasing levels of contamination and may indicate that the ivory gull is influenced by contaminant-induced oxidative stress.

Contaminant-induced eggshell thinning was largely found to explain the population declines in birds of prey after 1945 in Europe and North America, and one of the main reasons leading to a ban of DDT in many developed countries in the early 1970s (Fiedler 2000; Walker et al. 2001). *p,p'*-DDE is a well-known eggshell thinner (Cooke 1973), however, other compounds, such as PCBs, methyl-Hg and dieldrin, have also shown eggshell modifying properties (e.g. Cooke 1973; Lowe and Stendell 1991; Lundholm 1995). The present study found associations between eggshell thickness and a wide range of contaminants, including PCBs, Hg, BFRs and DDTs, indicating that the eggshell thickness in ivory gull eggs from the Russian Arctic are influenced by contaminants.

6.4 Toxicological evaluation

Most studies regarding threshold levels for biological effects have focused on old and well-known contaminants such as PCBs and organochlorine pesticides. These are also the dominating compounds in the ivory gull eggs in the current study. Very little is known about the sensitivity of ivory gulls and to date no studies of potential effects have been performed on the species. Although threshold data have limitations and extrapolating across species should be done with care, these levels can still be used to judge the risk posed to the ivory gull from environmental contaminants.

Generally, the levels of contaminants in the ivory gull eggs were relatively high. In particular the high $\sum_{28}$PCB and *p,p'*-DDE concentrations were at levels that are likely to elicit effects in avian species. The mean $\sum_{28}$PCB in Nagurskoe, the colony with the highest levels, was above the no-effect level for hatching success in Forster's terns *Sterna forsteri* and above the low-effect level for egg mortality in double-crested cormorants *Phalacrocrax auritus* and bald eagles (de Wit et al. 2004). The mean level, however, was below the low-effect levels for egg mortality and deformities in herring gulls, for deformities in double-crested cormorants and for reproduction in common terns *Sterna hirundo* and night herons *Nycticorax nycticorax* (de Wit et al. 2004). The three remaining colonies had considerably lower levels of PCBs. The mean $\sum_{28}$PCB concentrations in eggs from the Svenskøya and Klyuv Cape colonies exceeded the low-effect level for hatching success in Forster's terns, whereas the mean $\sum_{28}$PCB concentrations in the eggs from the Domashny colony were below all effect thresholds listed in de Wit et al. (2004).

All levels of *p,p'*-DDE were well below levels associated with reproductive failure in peregrine falcon *Falco peregrinus* eggs and most eggs had levels below the threshold

associated with marked productivity decline in bald eagle *Haliaeetus leucocephalus* eggs (de Wit et al. 2004). Although the concentrations of *p,p*'-DDE were below critical threshold levels in bald eagles and peregrine falcons, none of the eggs displayed low levels of *p,p*'-DDE. Contaminant-induced eggshell thinning was largely found to explain the population decline in birds of prey after 1945 in Europe and North America. Although there is a wide inter-species variation in sensitivity, it is possible to define a critical degree of eggshell thinning, where thinning above 16 to 18% is associated with population declines (Walker et al. 2001). The mean values of eggshell thickness for each colony were compared to the mean value of eleven ivory gull eggs in the collections of the Western Foundation of Vertebrate Zoology (WFZV) sampled between 1885 and 1930 (R. Corado, pers. comm.). A mean thinning of 7 to 17% were found in the four colonies, indicating that the ivory gull may be influenced by contaminants at levels affecting the population status.

7. Conclusions

High levels of organohalogen contaminants were found in ivory gull eggs from Svalbard and the western Russian Arctic compared to levels in eggs from seabird species throughout the Arctic. In particular the levels of PCBs and *p,p'*-DDE were high, whereas the levels of BFRs and PFASs were considerably lower. Large differences between colonies were also found, in particular for PCBs, organochlorine pesticides and BFRs, being considerably higher in eggs from the Naguskoe colony, intermediate levels in eggs from the Svenskøya and Klyuv Cape colonies and the lowest concentrations found in eggs from the Domashny colony. Levels of Hg and PFASs generally did not differ between colonies. These findings indicate that there may be a possible local source of contamination, in particular of PCBs and organochlorine pesticides on Franz Josef Land, or large regional differences in long-range transported contaminants to the breeding grounds or wintering areas.

These high levels are likely to influence the ivory gull. The levels are approaching or higher than various threshold levels for effects (de Wit et al. 2004; Gabrielsen 2007) and the present studies found indications of effects on vitamin status and eggshell thickness. The degree of eggshell thinning relative to eggs from before DDT use is approaching the critical level associated with population declines in a range of species, indicating that eggshell thinning is of potential concern for the population status. Contaminants inflict stress on organisms, often in combination with natural stress factors, and the effects of the contaminants may become more severe when the organism is under additional environmental stress (Bustnes 2006). The future of the ivory gull is likely to involve changes in habitat and food availability due to changes in ice conditions. The high levels of contaminants indicate that the ivory gull is under additional stress as it meets the challenges posed by future environmental change.

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Tittel - norsk og engelsk
Organohalogener og kvikksølv i ismåkeegg

Organohalogens and mercury in ivory gull eggs

Samandrag – summary
In the present study 35 ivory gull *Pagophila eburnea* eggs from four colonies in Svalbard, Franz Josef Land and Severnaya Zemlya were analysed for halogenated organic contaminants and mercury. Very high levels of PCBs and organochlorine pesticides were found.

4 emneord	4 subject words
Ismåke, miljøgifter, Russland, Svalbard	Ivory gull, pollutants, Russia, Svalbard

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Statlig program for forurensningsovervåking omfatter overvåking av forurensningsforholdene i luft og nedbør, skog, vassdrag, fjorder og havområder. Overvåkingsprogrammet dekker langsiktige undersøkelser av:

- overgjødsling
- forsuring (sur nedbør)
- ozon (ved bakken og i stratosfæren)
- klimagasser
- miljøgifter

Overvåkingsprogrammmet skal gi informasjon om tilstanden og utviklingen av forurensningssituasjonen, og påvise eventuell uheldig utvikling på et tidlig tidspunkt. Programmet skal dekke myndighetenes informasjonsbehov om forurensningsforholdene, registrere virkningen av iverksatte tiltak for å redusere forurensningen, og danne grunnlag for vurdering av nye tiltak. SFT er ansvarlig for gjennomføringen av overvåkningsprogrammet.

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