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Persistent organic Pollutants (POPs) in the Greenland environment – Long-term temporal changes and effects on eggs of a bird of prey

NERI Technical Report No. 509

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Persistent organic Pollutants (POPs) in the Greenland environment – Long-term temporal changes and effects on eggs of a bird of prey

NERI Technical Report No. 509
2004

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Data sheet

Title:	Persistent organic Pollutants (POPs) in the Greenland environment - Long-term temporal changes and effects on eggs of a bird of prey
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Serial title and no.:	NERI Technical Report No. 509
Publisher:	National Environmental Research Institute © Ministry of the Environment
URL:	http://www.dmu.dk
Date of publication:	November 2004
Editing complete:	October 2004
Referee:	Cynthia de Wit, Institute of Applied Environmental Research (ITM), Stockholm University.
Financial support:	Danish Cooperation for Environment in the Arctic (DANCEA), Ministry of the Environment.
Please cite as:	Sørensen, P.B., Vorkamp, K., Thomsen, M., Falk, K. & Møller, S. 2004. Persistent organic pollutants (POPs) in the Greenland environment - Long-term temporal changes and effects on eggs of a bird of prey. National Environmental Research Institute, Denmark - 126 pp. NERI Technical Report No 509. http:\\technical-reports.dmu.dk
Abstract:	The project studied the long-term time trend of brominated flame retardants, polychlorinated biphenyls (PCBs) and organochlorine pesticides in peregrine falcon *(Falco peregrinus)* eggs. Furthermore, possible effects of the contamination on the eggshell thickness were investigated using multivariate statistical methods. The contamination profile of the eggs was dominated by PCBs and organochlorine pesticides, but the polybrominated diphenyl ethers (PBDEs), including the fully brominated congener BDE-209, were also found in all eggs analysed. All compound groups were found at very high concentrations, reaching median summed concentrations of 55 µg/g lw for PCBs. Indications of an increase in PBDE concentrations during the last 17 years were found, while concentrations of organochlorine compounds seemed to decrease or remain constant. The correlation coefficient between the concentration and the eggshell thickness was negative, indicating a negative influence of the contaminants on the eggshell thickness. Thus, it has not been possible to identify remarkable improvement in the ecotoxicological pressure on the peregrine falcons during the period of investigation.
Keywords:	Arctic, bird eggs, brominated flame retardants, eggshell thickness, hexabromocyclododecane, organochlorine pesticides, peregrine falcon, persistent organic pollutants, polybrominated diphenyl ethers, polychlorinated biphenyls, time trend, tetrabromobisphenol A.
Layout:	Ann-Katrine Holme Christoffersen
ISBN:	87-7772-832-7
ISSN (electronic):	1600-0048
Number of pages:	126
Internet version:	The report is available only in electronic format from NERI's homepage http://www2.dmu.dk/1_viden/2_Publikationer/3_fagrapporter/rapporter/FR509
For sale at:	Ministry of the Environment Frontlinien Rentemestervej 8 DK-2400 København NV Tel.: +45 70 12 02 11 frontlinien@frontlinien.dk

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Preface

This report presents results of from the project "Persistent organic contaminants in the Greenland environment: Long-term temporal changes and effects on eggs of a bird of prey". The work is performed based on co-operation between the Roskilde University Library, the Danish EPA and the National Environment Research Institute in Denmark. The consulting company PFA Consult and the analytical laboratory of RIVO in the Netherlands have been sub contractors in the project. The academic staff behind the report can be seen from the author list. The laboratory technician Birgit Groth at NERI is gratefully acknowledged for her high quality work.

Sammenfatning

Problem

Vandrefalken *(Falco peregrinus*) er top predator fugl og har derfor et stort potentiale for kontaminering med persistente organiske stoffer ved opkoncentrering gennem fødekæden. Vandrefalkens færd leder den gennem relativt kontaminerede områder i både det nordlige og centrale Amerika. Det er velkendt at nogle organochlorerede forbindelser kan indicere indirekte eller direkte toksikologiske effekter, som f.eks. en reduktion i æggeskalstykkelsen. Flere studier over de sidste årtier har påpeget effekten af chlorinerede forbindelser som DDT (og dets nedbrydningsprodukter) og PCBer, mens der kun findes få langtidsundersøgelser på kontaminering af biota i Arktis. Nyligt er der fundet uventet høje kontamineringsniveauer af bromerede flammehæmmere i æg fra svenske vandrefalke.

Formål

Formålet med dette projekt er at belyse eventuelle sammenhænge mellem kontamineringsniveauer og mønstre med en ændring i æggeskalstykkelsen på vandrefalkeæg fra Sydgrønland. Undersøgelsen dækker de klassiske chlorinerede forbindelser som PCBer, DDT og dets nedbrydningsprodukter, organopesticider og de nyere identificerede bromerede flammehæmmere. Tidstrends og mulige korrelationsprofiler undersøges.

Rammerne for undersøgelsen

Falkeæg samlet i Sydgrønland i årene fra 1981 til 2003 er analyseret for kontaminering med gamle og nyere persistente organiske stoffer. Æggeprøverne er taget fra 28 forskellige reder og inkluderer 41 æg. Koncentrationsniveauerne i æggene er identificeret ved kemisk analyse af 55 kemiske stoffer bestående af PCBer, DDT og nedbrydningsprodukter, HCH, HCB, toxaphen congenerer, chlordaner og de bromerede flammehæmmere PBDEer, HBCD og TBBPA. Æggeskalstykkelsen er målt for samtlige analyserede æg, samt for æggeskalsfraktioner opsamlet i 47 andre reder i løbet af undersøgelsesperioden fra 1981 til 2003.

Resultater

Kontamineringsniveauer

Spredningen i koncentrationen mellem forskellige kemiske stoffer spænder over 5 størrelsesordener. Variationen i koncentrationsniveauer for det samme stof, men mellem æg, er meget mindre end variationen mellem kemiske stoffer i samme æg.

En sammenligning af den gennemsnitlige kontaminering af æg viser at summen af PCBer og DDT er de dominerende stofgrupper, idet de tilsammen dækker 95 % af den totale kontaminering i æggene. Summen af mediane koncentrationer er 55 µg/g lipid for PCBerne og 40 µg/g lipid for DDT og dets nedbrydningsprodukter. Det højeste koncentrationsniveau for en enkelt komponent, dvs. et enkelt kemisk stof, er målt for p,p'-DDE. Koncentrationen i enkelte æg er 0.7-9.1 µg/g vådvægt henholdsvis 9-170 µg/g lipid. Den høje variabilitet i kontamineringsniveauer samt den usikkerhed som er forbundet med analysen taget i betragtning, er resultaterne for p,p'-DDE stadigt tæt på det rapporterede effektniveau på 20 µg/g vådvægt som medfører fald i populationen. Dertil kommer at 42% af de analyserede æg overstiger NOAEL (No Observable Adverse Effect Level) grænseværdien på 3 µg/g vådvægt for p,p'-DDT. Dette indikerer tilstedeværelsen af en stærkt bekymrende og problematisk kontaminering af de Sydgrønlandske falkeæg.

De bromerede flammehæmmere (PBDEerne) blev fundet i alle æg, i en middelsummeret koncentration på 1.9 µg/g vådvægt. Dette fund er blandt de højeste målte PBDE koncentrationsniveauer som hidtil er fundet i biologiske matricer i naturen. BDE-209, som er den tungeste af de bromerede flammehæmmere har man hidtil troet ikke var i stand til at ophobe sig gennem fødekæden, hvilket denne undersøgelse kan afkræfte idet stoffet er fundet om end i relativt lave koncentrationer. HBCD er ligeledes fundet i æggene i lave koncentrationer, mens kun det methylerede nedbrydningsprodukt af TBBPA, dimethyl-TBBPA, blev detekteret. PCB-koncentrationerne i nærværende undersøgelse ligger lavere end i tilsvarende undersøgelser af norske falkeæg, men højere end i prøver fra Alaska. Målte DDT koncentrationer i nærværende undersøgelse af falkeæg fra Grønland ligger på samme niveau som i Norge og Alaska. Summen af de bromerede flammehæmmere i denne undersøgelse er tilsvarende PBDE-koncentrationer i falkeæg fra Sverige.

Korrelationen mellem stofferne er generelt positiv og stærkest mellem stoffer indenfor en stofklasse. En undtagelse er dog de bromerede flammehæmmere som generelt er negativt korreleret til de resterende stofklasser.

Korrelationen mellem kontamineringsniveauer og æggeskalstykkelse

Det generelle billede er at et højere kontamineringsniveau inducerer en tyndere æggeskalstykkelse. Mere specifikt for de enkelte kemiske stoffer ses en sammenhæng mellem molekylets fleksibilitet og æggeskalstykkelse. Således at en øget molekyle fleksibilitet giver en øget effekt på æggeskalstykkelsen. Dette indikerer en tydelig reduktion af æggeskalstykkelsen forårsaget af en øko-toksikologisk effekt. Den høje grad af korrelation mellem kemiske stoffer udelukker identificeringen af enkelte stoffer som havende stærkest effekt på æggeskalstykkelse.

Tidsmæssig udvikling i kontamineringsniveau og æggeskalstykkelse

Koncentrationsniveauet for flere af PCBerne viser en faldende tendens, mens flertallet af PBDEerne synes at have et stigende kontamineringsniveau gennem undersøgelsesperioden fra 1981 til 2003. Dette er sammenfaldende med at PCBerne er udfaset mens anvendelsen af PBDEerne har været stigende i undersøgelsesperioden. Resultaterne på falkeæg i nærværende undersøgelse er dog modsat hvad man har fundet for langnæbbet lomvie æg fra Østersøen, men i overensstemmelse med studier fra Nordamerika. Forskellen i resultaterne fra studier af Europæiske og Nordamerikanske æg kan skyldes forskellen i de regulatoriske indgreb, idet der kun i Europa er blevet gjort en indsats for at begrænse anvendelsen af de bromerede flammehæmmere. Kontamineringsniveauet af DDT og dets nedbrydningsprodukter ligger mere eller mindre konstant i perioden. Ligeledes ligger tidstrenden for summen af alle analyserede kontaminanter på et konstant niveau gennem undersøgelsesperioden. p,p'-DDE som har højest koncentrationsniveau, er en dominerende årsag til den ikke tilstedeværende tidslige udvikling det generelle kontamineringsniveau. I multivariate dataanalysemetoder er det muligt at differentiere mellem forskelligt rettede systematiske variationer i enkelte stoffers koncentration og mønstre i æggeskalstykkelser over år. PLS-regressionerne viser at en negativt korreleret sammenhæng mellem de enkelte PBDE koncentrationer og æggeskalstykkelsen som er i samtidig overensstemmelse med en negativ tidstrend i æggeskalstykkelsen. Både æggeskalsmålingerne og den multivariate dataanalyse viser en positiv tidstrend for æggeskalstykkelsen. Tidstrenden i æggeskalstykkelsen er dog svag og ikke statistisk signifikant.

Generelle konklusioner

Vandrefalkens æg er kontamineret med xenobioter i en relativt høj grad. Den tidslige udvikling i kontamineringsniveauet synes at være status quo. Æggeskallerne synes at være influeret af kontamineringen i en grad der gør at det ikke er muligt at se en tydelig forbedring i det øko-toksikologiske tryk på vandrefalken i perioden 1981 til 2003.

Summary

The problem and purpose

Peregrine falcons *(Falco peregrinus*) are top predators and thus subject to biomagnification leading to accumulation of Persistent Organic Pollutants (POPs). Furthermore, the route of migration leads the birds through relatively contaminated areas in both the north and central parts of the continental America. It is well known that some organochlorine chemicals can induce indirect or direct toxicological effects, including a thinning of eggshells. Several studies over the past decades have addressed the effects of chlorinated compounds such as DDT (and breakdown products) and PCBs, although long term trends in the occurrence in the arctic have seldom been conducted. Recently, unexpectedly high contamination levels of brominated flame retardants were observed in Swedish peregrine falcon eggs. The contamination by some xenobiotic chemicals may decrease temporally due to regulation in usage, while the contamination level may increase for others. This induces a profile of contamination level that may change with time and influence the eggshell thickness.

The purpose of this project is to study the contamination by xenobiotics and shell thickness of peregrine falcon eggs from Southern Greenland. The xenobiotics include both the classic chlorinated compounds, some pesticides and the more newly identified brominated flame retardants. Time trends and possible correlation profiles are investigated.

Frame of investigation

The contamination by xenobiotics is measured for Peregrine falcon *(Falco peregrinus*) eggs collected in South Greenland between 1981 and 2003. Egg samples are taken from 28 different clutches and includes 41 single eggs. The egg tissue concentration level is identified for a broad suite of 55 single chemicals including Polychlorinated biphenyls (PCBs), DDT (including DDD, DDE), HCH, HCB, toxaphene congeners and chlordane-related compounds and the newly identified contaminants brominated flame-retardants PBDES, HBCD and TBBPA. The eggshell thickness is measured for the same eggs as used in the chemical analysis and also for shell fractions collected in other 47 clutches during the period of investigation.

Results

Contamination level

The concentration range between the different substances is large, covering 5 orders of magnitude. So, the variability in concentration level for the same substance but between eggs is much smaller than the variability between compounds in the same egg.

On average, the sum of PCBs and DDT dominates the contamination by accounting for 95 % of the total contaminant load analysed in the eggs. Median summed concentrations were 55 µg/g lipid weight for PCBs and 40 µg/g on lipid weight basis for DDT (and its degradation products). The highest concentration level for a single component was measured for p,p'-DDE in the range for single eggs of respectively 0.7-9.1 µg/g wet weight and 9-170 µg/g lipid weight. Taking into account the high variability in contamination levels and the actual uncertainty inherent in the effect assessment, this range seems close to the reported limit of 20 µg/g wet weight above which population declines are reported to occur. Furthermore, 42 % of the eggs analysed exceeded the NOAEL level of 3 µg/g wet weight for p,p'-DDE. This seriously indicates a problematic contamination of the eggs.

PBDEs were detected in all eggs, with a medium summed concentration of 1.9 µg/g lipid weight. This is among the highest PBDE concentration ever detected in wildlife. The main congener was BDE-153. Relatively low but measurable concentration levels were detected for BDE-209, which indicates some degree of bioavailability and accumulation potential in biota. HBCD was likewise detected in the eggs, however, the concentrations were low, while TBBPA was only detected in terms of the degradation product dimethyl-TBBPA. PCB concentrations were lower than in Norwegian peregrine falcon eggs, but higher than in samples from Alaska. DDT concentrations were similar in Greenland, Norway and Alaska. Summed PBDE concentrations were similar to the results for wild peregrine falcons in Sweden.

The correlation between substances is positive, showing the strongest correlation within the same class of substances.

Correlation between contamination level and eggshell thickness

A higher contaminant level is seen to induce a thinner eggshell. Also the molecular flexibility is shown to have negative influence on the eggshell thickness as suggested by several investigators. This indicates a clear reduction in eggshell thickness due to an ecotoxicological effect. The observed intercorrelation between the single substances does not allow the identification of particular chemicals with the strongest effect on eggshell thickness. However, both the PLS-regression and single chemical correlation analysis with the eggshells, indicate that the PBDEs seem to have a negative influence on eggshell thickness.

Temporal development in contamination level and eggshell thickness

The concentration level for several PCBs shows a decreasing tendency while the majority of the PBDEs was seen to increase in contamination level during the period of investigation. This coincides with the fact that the PCBs are not in usage any more, while the PBDEs are used in increasing quantities during the period of investigation. This is in contrast to results for guillemot eggs from the Baltic Sea, but in agreement with studies on biota from North America. The

discrepancy between the European and North American study areas might be caused by regulatory measures only taken in Europe to regulate the PBDEs. DDT and the degradation products remain constant in time. The time trend for the sum of all measured contaminants shows a constant level during the period of investigation. As p,p'-DDE dominates the level of contamination the constant temporal level of this single substance was the single factor most responsible for the constant contamination level in general. Both eggshell measurements and multivariate statistics show a positive time trend for the eggshell thickness, which, however, is only relatively weak and not statistically significant.

General conclusion

The Peregrine falcon eggs are contaminated to a relatively high degree with xenobiotics and the temporal development of the contamination seems to be status quo. The eggshells seem influenced by this contamination and it has not been possible to identify remarkable improvement in the ecotoxicological pressure on the Peregrine falcons during the period of investigation.

1 Introduction

Top predators often accumulate high levels of Persistent Organic Pollutants (POPs). Peregrine falcons *(Falco peregrinus)* feed almost exclusively on other birds. As their prey species often contain contaminants from overwintering areas, the potential for biomagnification is great, leading to high concentrations in the peregrine falcons. Some organochlorine chemicals have shown indirect or direct toxic effects, including the thinning of eggshells in birds of prey which caused widespread declines in the wild populations. Several studies over the past decades have addressed the effects of chlorinated compounds such as DDT (and its breakdown products) and PCBs, however, long-term trends in the occurrence of POPs in peregrine falcons of the Artic have not been conducted.

Brominated flame retardants (BFR) have been identified as ubiquitous contaminants and potential POPs. Recently, unexpectedly high contamination levels of brominated flame retardants were observed in Swedish peregrine falcon eggs, including the fully brominated congener BDE-209, which had previously not been considered bioavailable (Sellström et al. 2001, Lindberg et al. 2004).

The purpose of this project was to study the temporal trend in the contamination of peregrine falcon eggs from Southern Greenland. Furthermore, possible relationships between the eggshell thickness and the contamination of a broad range of xenobiotics were analysed, with emphasis on the long-term trend of BFR and possible effects on peregrine falcon eggs.

The xenobiotic contamination and the eggshell thickness were measured for eggs collected in South Greenland between 1981 and 2003. This includes a broad suite of 55 single chemicals including Polychlorinated biphenyls (PCBs), DDT (including DDD, DDE), HCH, HCB, toxaphene and chlordane-related compounds and has focussed on the newly identified contaminants brominated flame-retardants (BFRs).

2 Data and Methods

2.1 Sampling area and period

The chemical analyses included 37 addled eggs collected between 1986 and 2003 (no samples are available from 1993, 1996 and 1997). The eggs represent 28 different clutches – in 8 cases more than one egg derives from the same nest and year. Eggshell thickness was measured for the same eggs, but also for some whole eggs not included in the chemical analyses (total n=41 whole eggs). In addition, small eggshell fragments were collected between 1981 and 2003 after the birds had hatched, increasing the sample coverage to include nests where all eggs were hatched (i.e. no addled eggs were collected). In total 75 clutches provided an adequate amount of shell fragments (>20, see below) to include in the long-term analyses of changes in eggshell thickness.

The sample areas for the eggs are shown in Figure 1.

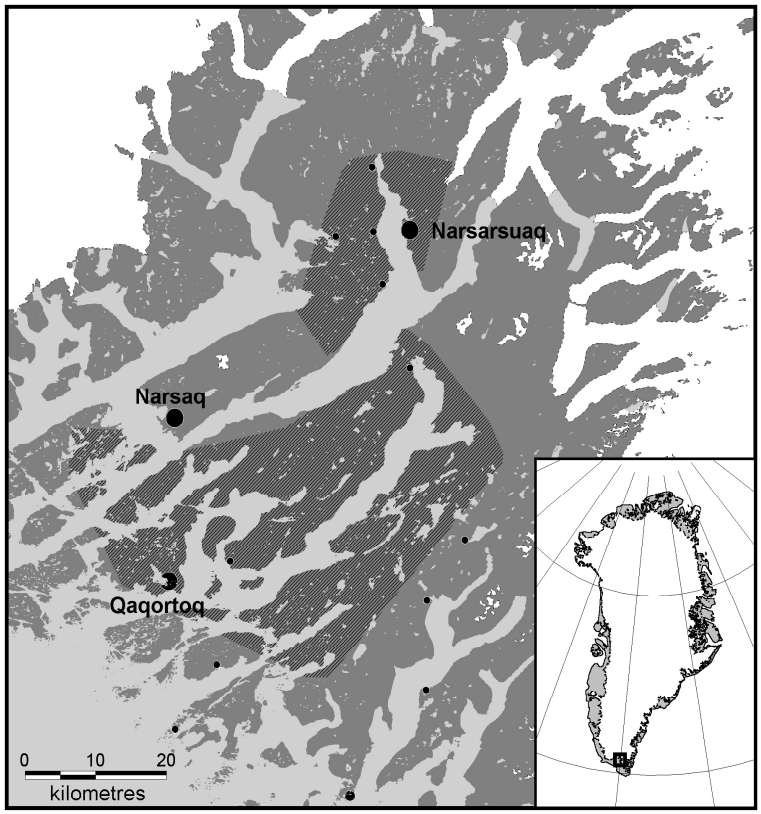

Figure 1. Map of sampling area in South Greenland spanning from outer coast to inland areas; all nest sites sampled in this study (n=13 different locations) are located inside the hatched area. White areas with dashed line edge are ice/glaciers.

2.2 Chemicals selected for analysis

The compounds analysed in this project include various chlorinated compounds, such as polychlorinated biphenyls (PCBs) and organochlorine pesticides, as well as BFRs (Table 1). With the exception of HCH, all organochlorine compounds are listed in the Stockholm Convention of persistent organic pollutants (POPs), which restricts or prohibits the production, trade and use of these compounds because of their persistence, bioaccumulation, long-range transport and adverse health effects. Table 1 gives an overview of compounds included in this study. A description of the use and properties of the individual compounds is given in Appendix 1.

Table 1. Chemicals included in this project

Compound group	Acronym	Congeners and analytes	Legal status in Denmark (de March et al., 1998)
Polychlorinated biphenyls	PCB	CBs 28, 31, 44, 49, 52, 99, 101, 105, 110, 118, 128, 138, 149, 151, 153, 156, 170, 180, 187, 188, 194, 209	Prohibited
DDT and degradation products	DDT	p,p'-DDT, p,p'-DDE, p,p'-DDD, o,p'-DDT, o,p'-DDE	Prohibited for plant protection use
Toxaphene	CHB	CHBs 26, 40, 41, 44, 50, 62	Banned since 1987
Chlordane-related compounds		oxychlordane, cis-chlordane, trans-chlordane, cis-nonachlor, trans-nonachlor	Prohibited for plant protection use
Hexachlorocyclohexanes	HCH	α-HCH, β-HCH, γ-HCH	Mixed isomers prohibited for plant protection use
Hexachlorobenzene	HCB	HCB	Banned
Polybrominated diphenyl ethers	PBDE	BDEs 17, 28, 47, 49, 66, 99, 100, 153, 154, 183, 209	Restriction of penta- and octa-BDE, future ban
Hexabromocyclododecane	HBCD	HBCD	no restrictions known
Tetrabromobisphenol A	TBBPA	TBBPA	no restrictions known

Most of these compounds were analysed at the National Environmental Research Institute (NERI) except for TBBPA and HBCD, which were analysed at the Netherlands Institute for Fisheries Research (RIVO). The analytical methods are described in Appendix 2.

2.3 Reported data

Concentration measurements - Appendix 3

Reported results on the measurement of chemical substances are provided in Appendix 3. Each egg sample has a ring number, a registration number, a batch number referring to the trial of chemical analysis, and a year and place of sampling. The sample weight, lipid content and dry matter are given for each egg sample. The concentrations of the chemical substances are given in ng/g wet weight in the upper half of the tables, as well as in ng/g lipid weight in the lower half of the tables in Appendix 3. Results are divided into chemical classes, i.e. the brominated flame retardants, the PCBs, and finally the chlorinated pesticides, e.g. DTT and degradation products, toxaphene congeners, and chlordanes.

Eggshell measurements – Appendix 4
Measurements of the eggshell thickness over years are provided in Appendix 4. A more detailed description of these measurements is given in Falk and Møller, 2004, in prep (cf. Appendix 5).

2.4 Data Analysis

The purpose of this project is to gain information on the contamination of peregrine falcon eggs over a longer period of time. Furthermore, possible relationships between the eggshell thickness, time and the contamination patterns of xenobiotics have been studied.

To fulfil the purpose of the study several types of data analysis have been performed. The data analysis is briefly described below with reference to specific Appendices, where a detailed presentation of the results and individual methods of data analysis are presented.

Pearson Correlation Matrix - Appendix 6
A Pearson correlation matrix showing the correlation between all possible pairs of variables is provided in Appendix 6. Variables are chemicals and eggshell thickness, respectively. A chemical variable is made up by the concentration of contamination in each egg sample as given in Appendix 3. The corresponding eggshell thickness, identified by the egg sample registration number, is given in Appendix 4. As such, the correlation coefficient between two variables expresses the degree of covariation in the contamination pattern between eggs. The correlation coefficients yield no information about the covariation of compounds over years. The correlation matrix is given in Appendix 6.

Multivariate Data Analysis – Appendix 7
Principal Component Analysis (PCA) has been performed on the data from Appendix 2. The variables are the individual chemicals and egg samples are objects. The concentration of each chemical in egg samples was mean centred, i.e. the mean between samples was subtracted from each measurement. Furthermore data was standardized, i.e. each measurement was divided by the standard deviation between measurements to obtain unit variance for each chemical. As such the PCA analysis shows relative patterns in concentration profiles between egg samples. Furthermore, the egg samples are assigned by an id expressing the year of sampling, the batch number of the chemical analysis and the ring number of the mother bird. The data analysis, in Appendix 5, includes an inspection of patterns in egg samples identified by a time, batch and mother bird of the samples, explained by so-called latent variables, which are vectors comprised of the original chemical variables. A more in depth presentation of the multivariate data analysis including Partial Least Square Regression is presented in Appendix 7, while the overall results and conclusions partially based on the results in Appendix 7 are given in the following Chapters 3 and 4, respectively.

Partial order rank correlations – Appendix 8
The Rank correlation method is a non-parametric method. Appendix 6 includes an analysis of the existence of a possible relation between eggshell thickness and the ortho-substitution patterns in two-ringed chemical structures. This case study is based on a hypothesis of a possible existence of a structure-activity relationship analogous to the known influence on toxicity of the planarity of the PCBs, which is related to the degree of co-planarity between the two phenyl rings (Thomsen and Carlsen, 2002).

Time trend of the single chemicals – Appendix 9
This Appendix includes time trend analyses for each chemical. The time trend analysis is based on ln-transformed concentrations in the individual egg samples. For each year on the x-axis, there are as many concentration measurements, y-axis, as egg samples. For each regression line, showing the concentration in egg samples as a function of sampling years, the slope is given. Furthermore, a hypothesis of the possibility of an inverse slope was tested by Monte Carlo calculations on random concentration and year combinations. The probability of an inverse slope is 1 minus the significance value given below the value of the slope in each graph.

3 Results

3.1 Concentrations

The concentration of the compounds analysed is related to the lipid content of the samples. The ln-transformed concentration values show general normality as seen from Figure 2 for pp′-DDE.

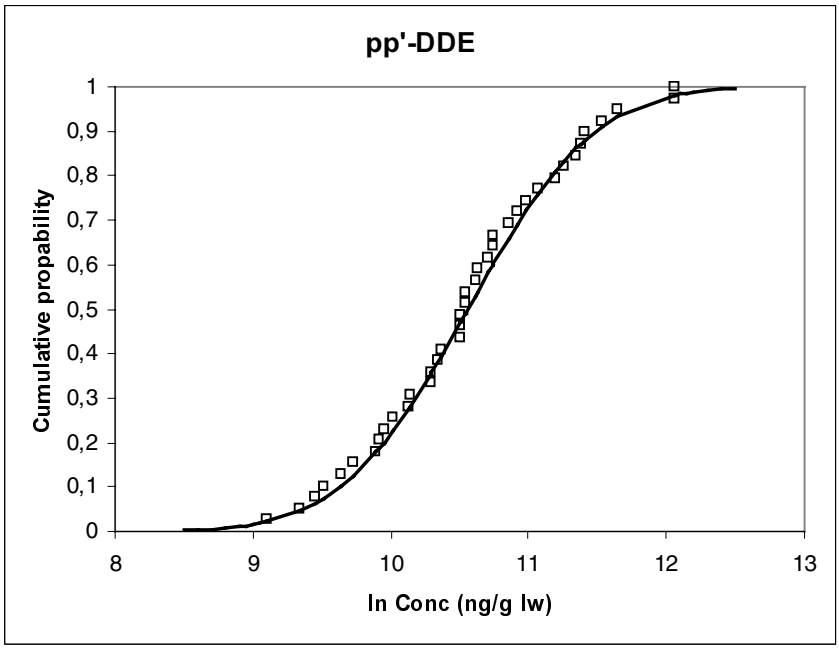

Figure 2. The ln-transformed concentration values referring to the lipid concentration fitted to a normal distribution (based on the data included in this study).

The variability in concentration level for specific compounds is rather constant for the ln-transformed values with a standard deviation around 0.9. This means that the relative variability is constant for the chemicals and Figure 3 shows the general variability relative to the average concentration.

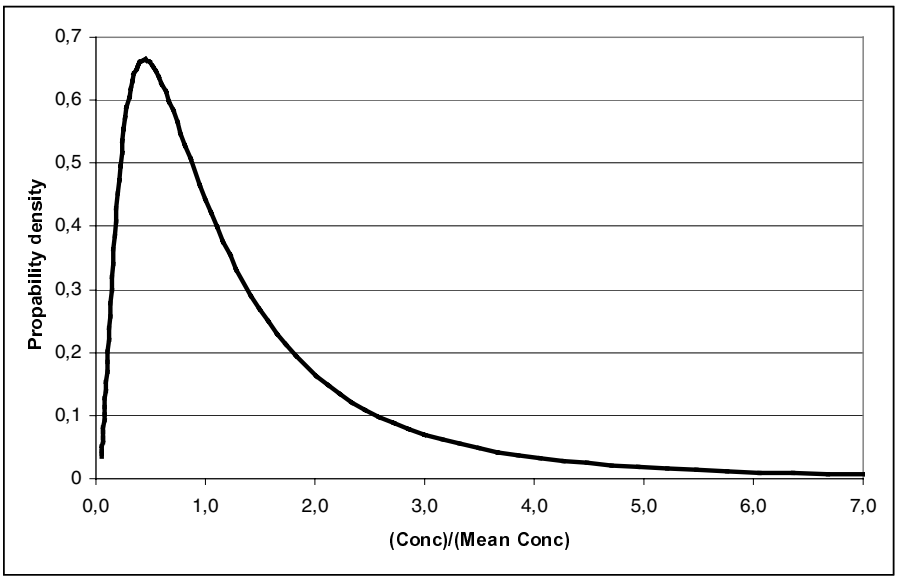

Figure 3. The variability of the concentration measurements relative to the mean value for a single chemical using a standard deviation of 0.9 All substances are close to this distribution except: CB-151, p,p´-DDD, CHB-26, BDE-17, BDE-28, HCBD, Me-TPBBP-A, where the variability is higher.

The x-axis in Figure 3, represents centred ln-transformed concentrations values ($lnC - lnC_{mean}$), backward transformed into real concentration units (C/C_{mean}). The probability distribution shows that a specific measurement very seldom will exceed 6 times the average value and often will be far below. However, a few substances show a standard deviation general higher than 0.9: CB-151, p,p´-DDD, CHB-26, BDE-17, BDE-28, HCBD, Me-TPBBP-A.

Figure 4 shows the mean values for every compound estimated based on the ln-transformed lipid concentration. The chemicals are further divided into categories, which clearly illustrates that the PCB congeners are the dominating contaminants. The variability in contamination level is considerable and covers 5 orders of magnitude. Low concentrations were found for HBCD, and TBBP-A was not detected in the eggs, but the metabolite (Me-TBBP-A) was found, which is more hydrophobic (lipophilic) than the parent compound and thus can be expected to bioaccumulate more effectively (Leslie et al., 2003).

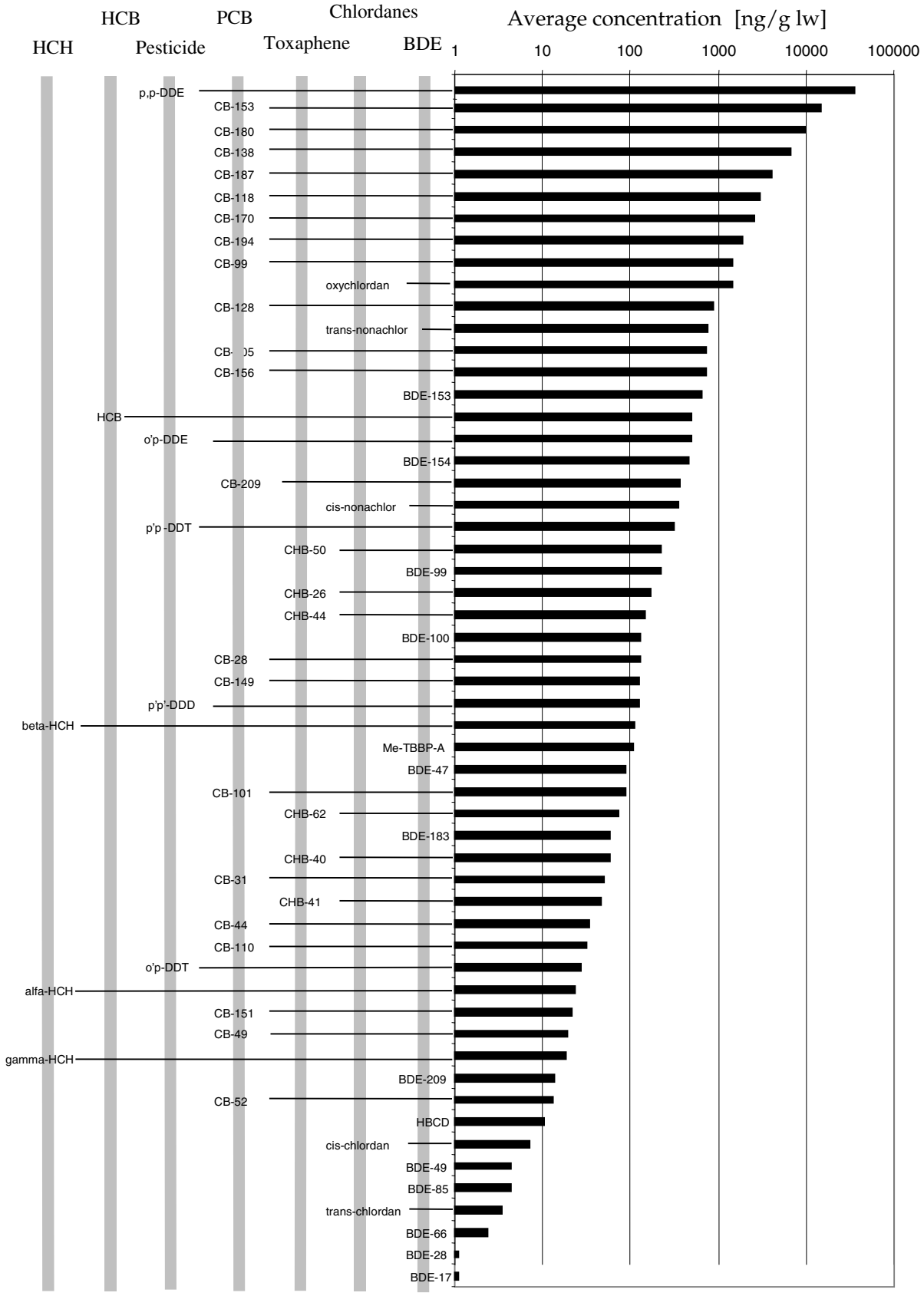

Figure 4. Mean concentration in ng/g lipid weight, for all substances ranked according to the contamination level in decreasing order. The chemicals are grouped into chemical classes to give an impression of the contamination levels between chemical classes.

As seen from Figure 4, p,p´-DDE has the highest contamination level of all chemicals. Besides p,p´-DDE, top concentrations are dominated by the PCBs. However, some of the brominated flame retardants and

single chlordanes and toxaphene congeners are also represented in the upper part of the figure. Accumulated values for each chemical class are shown in Figure 5. Median and mean values of all eggs are presented for each compound group.

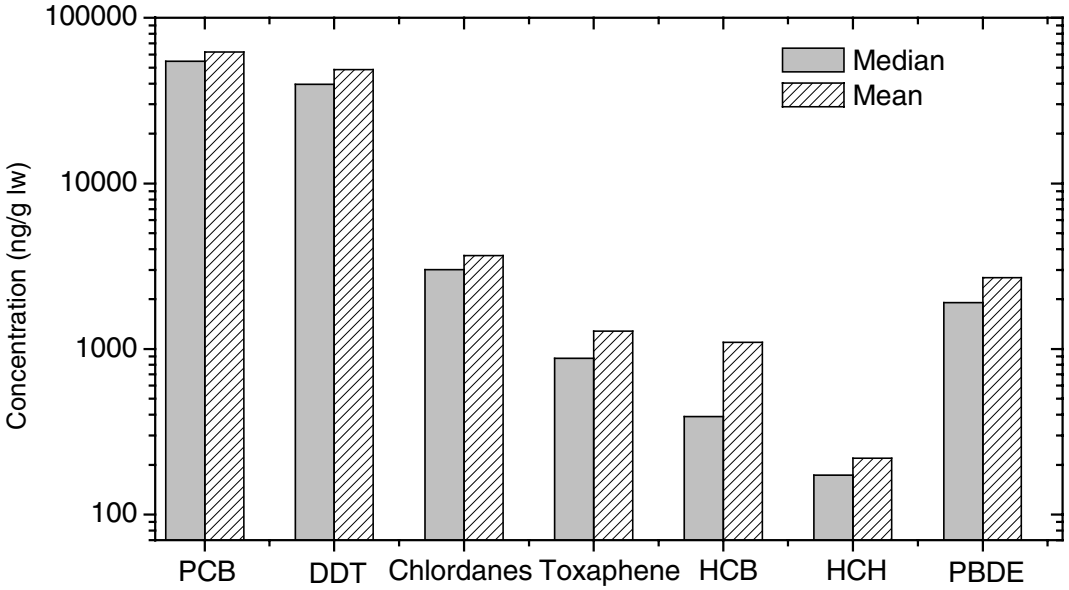

Figure 5. Median and mean values for the different compound classes analysed.

DDT and its degradation products and PCBs (21 compounds) together dominate the egg contamination as seen in Figure 5. The accumulated median concentration for the PBDEs (12 compounds) is 1900 ng/g lipid weight, which is in the same range as reported for Canadian biota (marine whale fat) (Hale et al., 2003).

The summed concentration of the 12 BDE congeners analysed ranges from 400-15130 ng/g lipid weight, with mean and median concentrations of 2700 and 1900 ng/g lipid weight, respectively. Data for comparison are available from Lindberg et al. (2004) who analysed eggs of three populations of peregrine falcons in Sweden: Wild populations from North and South Sweden and a captive population feeding on chicken only. They found significantly higher concentrations in eggs of the wild populations, which they attributed to the differences in diet.

In both the Swedish and the present studies, the concentrations cover a wide range and reach extremely high concentrations, which are among the highest concentrations seen in wildlife so far (Lindberg et al., 2004). Summing the concentrations of the dominating congeners, BDE-47, -99, -100, -153 and –154, results in values as high as 39000 ng/g lipid weight in the Swedish samples, and 15000 ng/g lipid weight in the samples from Greenland.

In spite of the large range, it can be noted that the Greenland samples are similar to the samples of the wild population from South Sweden, with regard to summed concentrations. For the sum of the 5 congeners mentioned above, the mean concentration is 2370 ng/g lipid weight in the samples from South Sweden and 2600 ng/g lipid

weight in the Greenland samples. The respective values for the population from North Sweden and for captive population are 4070 ng/g lipid weight and 47 ng/g lipid weight, respectively. The studies from Greenland and Sweden also agree in that respect that the total concentration of brominated compounds makes up approximately 2% of the sum of PCBs and DDT (incl. DDE and DDD).

Apparently, peregrine falcon eggs contain relatively more of the highly brominated congeners, compared with other biota samples. The main congener in the Swedish and the Greenland samples was BDE-153, while other biota samples, including bird eggs, usually are dominated by BDE-47 (Law et al., 2003). Interestingly, on average BDE-153 accounted for 34% of the summed concentration in both the Swedish and the Greenland sample provided the sum includes the same congeners. However, large differences can be seen for BDE-154 which accounts for 32% in the Greenland samples, but only 6.3% in the samples from South Sweden. For all these calculations, it has to be noted that mean values do not correctly represent the data which are affected by temporal trends, but have been chosen for the sake of comparison with the literature data.

As pointed out by Hale et al., (2003) BDE-209 (deca-BDE) is highly used in the USA (72 % of total demand), but the concentration level of BDE-209 is relatively low compared to other BDEs. This can be due to a larger molecule and extreme hydrophobicity inducing low bio-availability. Also the fact that the lower brominated congeners can be formed by de-bromination of BDE-209 can help to explain the relatively low concentrations of BDE-209 compared to the other congeners. However, even though the concentration level of BDE-209 is not high, it still indicates some degree of release and bioaccumulation as shown also by Sellström et al. (2001) and Lindberg et al. (2004).

Sellström et al. (2001) and Lindberg et al. (2004) found BDE-209 in 18 out of 21 eggs of peregrine falcon from Sweden, at concentrations ranging from <20-430 ng/g lipid weight in eggs of the wild falcons and <7-9 ng/g lipid weight in the captive falcons. All 36 eggs analysed of the South Greenland peregrine falcon population contained detectable amounts of BDE-209, which ranged from 3.8-250 ng/g lipid weight. However, the high concentrations were only detected in two eggs from 1995 and 2002. The median concentration of all eggs is 11 ng/g lipid weight, which is closer to the levels found in eggs of captive peregrine falcons.

The Swedish study showed for the first time that BDE-209 was bioavailable and could be accumulated in living organisms (Lindberg et al., 2004). The results from Greenland confirm this conclusion. The eggs of wild peregrine falcons had significantly higher levels of BDE-209 than eggs of captive falcons feeding on chicken. This indicates that BDE-209 is present in the environment and is taken up by falcons (Lindberg et al., 2004).

Total HBCD was detected in over half of the egg samples analysed, whereas the individual HBCD-isomers could not be detected due to higher limits of detection. The HBCD concentrations range from <0.1 ng/g lipid weight to 230 ng/g lipid weight in a sample from 1990.

The median concentration is 9.5 ng/g lipid weight, and the mean concentration is 28 ng/g lipid weight. The range of HBCD in this study is somewhat lower than that in the Swedish peregrine falcon eggs, which had HBCD concentrations of <4-2400 ng/g lipid weight (Lindberg et al., 2004).

The concentrations of the organochlorine compounds also cover a wide range. The ΣPCB concentration ranges from 12 µg/g lw to 162 µg/g lw. The lowest concentration occurs in an egg sample from 1989, which also has the lowest concentrations of HCB (0.17 µg/g lw) and ΣChlordane (0.72 g/g lw). The highest summed concentration of PCBs is found in a sample from 1987. This bird also has the highest concentrations of ΣToxaphene (5.3 µg/g lw) and ΣChlordane (12.1 µg/g lw). This indicates that some compound groups co-occur, while others follow a different pattern.

The comparison with Norwegian data indicates slightly lower PCB concentrations in the Greenland peregrine falcon eggs. 5 peregrine falcon eggs from Norway collected between 1991 and 1997 had maximum PCB concentrations of 25 µg/g wet weight and an average PCB concentration of 9.1 µg/g ww (Herzke et al., 2002). The lipid content of the eggs was not given. Our data were recalculated with the same congeners and converted to the wet weight basis. The same sample period was considered as in the Norwegian study. The recalculation indicated lower PCB concentrations in the Greenland samples, with a maximum value of 7.74 µg/g ww and an average concentration of 3.15 µg/g ww.

Even though the PCB concentrations were lower in Greenland than in Norwegian eggs, the pesticide concentrations were similar. The organochlorine pesticides analysed by Herzke et al. (2002) included p,p'-DDE, the three HCH isomers, dieldrin and the same chlordane-related compounds as the present study, including heptachloroepoxide. Furthermore, HCB was added to the sum of pesticides. As our data do not include dieldrin and heptachloroepoxide, the recalculation may underestimate the actual sum of organochlorine pesticides. The average concentration in the Greenland and the Norwegian samples were 3220 and 3118 ng/g lipid weight, respectively. However, the Greenland samples had a clearly larger concentration range.

The PCB concentrations are higher in the eggs from Greenland compared to concentrations found in peregrine falcon eggs from Alaska. Organochlorines in peregrine falcon eggs have been monitored in Alaska since 1979, including the two subspecies American peregrine falcon *(Falco peregrinus anatum)* and Arctic peregrine falcon *(F.p. tundrius)* (Ambrose et al., 2000). In the sampling period 1988-1995, PCB concentrations for *F.p. anatum* ranged from 0.4 to 15.0 µg/g ww, while PCB concentrations in eggs of *F.p. tundrius* ranged from 0.6 to 14.8 µg/g ww. Regarding the same period, the Greenland data range from 1.1 to 27.5 µg/g ww.

Besides higher PCB concentrations, the Greenland peregrine falcon eggs also had higher concentrations of oxychlordane compared to the two populations from Alaska. The other compounds analysed in both

studies (p,p'-DDE, p,p'-DDT, p,p'-DDD, β-HCH, HCB) had very similar concentrations with regard to the geometric mean. The Alaskan eggs, however, had a larger range and higher maximum concentrations of these chemicals. *F.p. tundrius* overwinters in areas from the Great Lakes in Canada through the USA to Texas, while *F.p. anatum* breeds in forested areas fromt the treeline south to Californa and Mexico (de March et al., 1998).

Peakall et al. (1975) concluded that populations of peregrine falcons would decline if DDE levels exceeded 20 ppm. Herzke et al. (2002) stated a *no observed adverse effect levels* (NOAEL) for peregrine falcons of 3 µg/g wet weight for p,p'-DDE. All the Greenland eggs analysed had p,p'-DDE concentrations below 20 ppm, however, 42% of the eggs exceeded the NOAEL, including eggs from the whole time period analysed. An embryonic LD50 for herring gull eggs was reported at 4.3 ppm, which also is clearly above the concentrations measured in the peregrine falcon eggs (Jarman et al., 1993).

3.2 Correlation analysis between single substances

A complete correlation matrix is given in Appendix 6 using the linear correlation coefficient (Pearson's r). A correlation coefficient close to 1 indicates a strong positive linear correlation and a value close to -1 indicates a strong negative correlation. A value close to 0 indicates independence. Cells having numeric values in the interval 0.80-0.89 have a weak grey spotted pattern and cell values between 0.90-1.00 have a denser grey spotted pattern. The significance of correlation has not been tested so the correlation matrix only indicates the internal differences in correlation within the data set.

The most obvious clustering, i.e. intercorrelated chemical substances regarding the contamination pattern between egg samples from all years, is summarised in Table 2. Typically, the correlation between substances is positive, i.e. if an egg is highly contaminated by one chemical then that egg also tends to be highly contaminated by other chemicals. However, the correlation between the level of concentration and the eggshell thickness is negative for nearly all chemicals. This indicates a general tendency for the contamination to have a negative influence on the eggshell thickness. At this stage it is not possible to judge the single chemicals according to an actual effect on eggshell thickness. It is not possible to discriminate between chemicals having a negative effect on the eggshell and chemicals, which have no effect, but instead covariating exposure patterns. This aspect is further described in section 3.3.

Table 2. Clustering of substances based on the most obvious correlation in the correlation matrix given in Appendix 6.

CB-44, CB-49, CB-52
CB-99, CB-105, CB-110, CB-118, CB-128, CB-138, CB-153, CB-156, CB-170, CB-180, CB-187, CB-194
Cis-nonachlor, Trans-nonachlor (and a tendency for positive correlation with a series of PCBs)
CHB-26, CHB-41, CHB-44, CHB-50
BDE-47, BDE-99, BDE-100

The general tendency for the correlation between chemicals to be positive in the correlation matrix in Appendix 6 is supported in the PCA (Appendix 7, Figure 4), and in Figure 6.

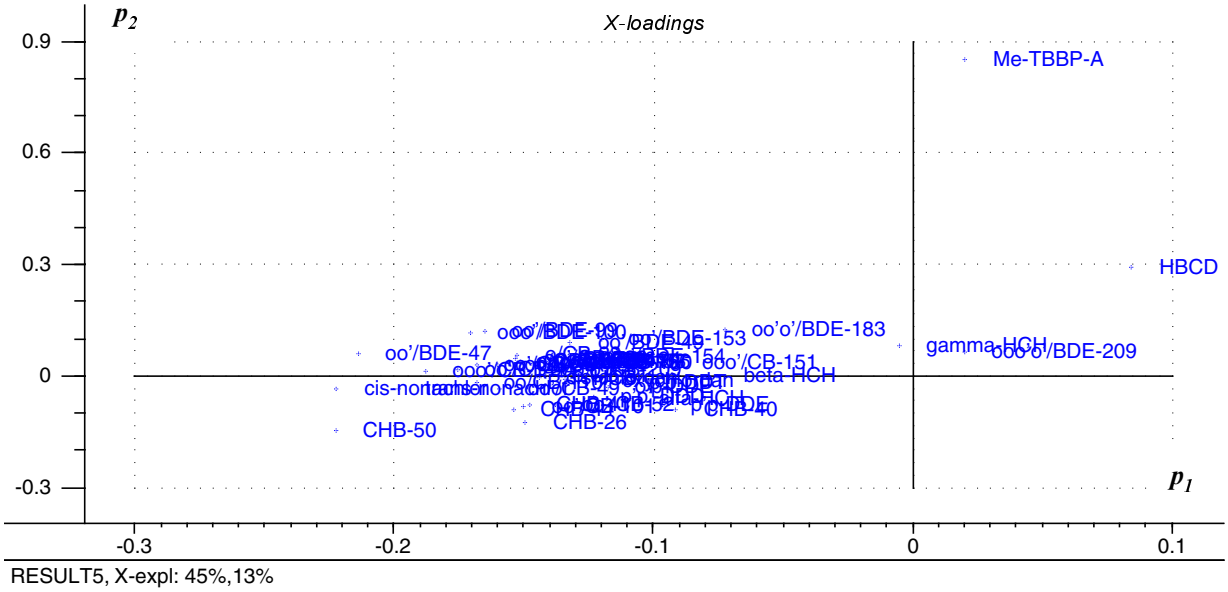

Figure 6. Loading plot of the two latent variables, p_2 versus p_1, showing visible groupings or clusters of intercorrelated variables. The first principal component, p_1, explains 45 % of the variance, whereas p_2 explains 13 % of the variance in the X-space.

The loading plot shows a high degree of intercorrelation between chemical variables, with negative loading in p_1 γ-HCH and BDE-209 have small loading values in both p_2 and p_1, which means that these variables have low explanatory capacity regarding the patterns in the score plot in Figure 7 compared to the remaining chemical variables. There is a tendency for Me-TBBP-A and HBCD to be inversely correlated to the remaining compound variables, which is in agreement with the results given in Appendix 6. However, the PCA shown in Figure 6 and 7 is based on mean values and the loading of Me-TBBP-A and HBCD seems to be due to very high concentration in few of the samples (e.g. cf. Figure 7, egg sample 2003d13). It should be mentioned that BDE-17, -28, 85, o,p´-DDT, trans-chlordane and CBH-62 were eliminated due to high frequency of missing data.

Figures 6 and 7 differ from the presentation of exactly the same data in Appendix 7 (Figure 3 and 4) by being autoscalled, i.e. mean centred and standardised to variance 1 prior to analysis. The overall patterns are the same, but the effect of difference in concentration levels has been eliminated from the PCA analysis in Figure 6 and 7. Still the PBDEs have highest positive loading in p_2, the majority of the PCBs have positive loadings as well except for CB-49, -52, -101 and –110. All of the chlordanes, the organochlorinated pesticides, HCB and toxaphene congeners - most dominating, have negative loading values in p_2. The corresponding patterns in egg samples are shown in Figure 7.

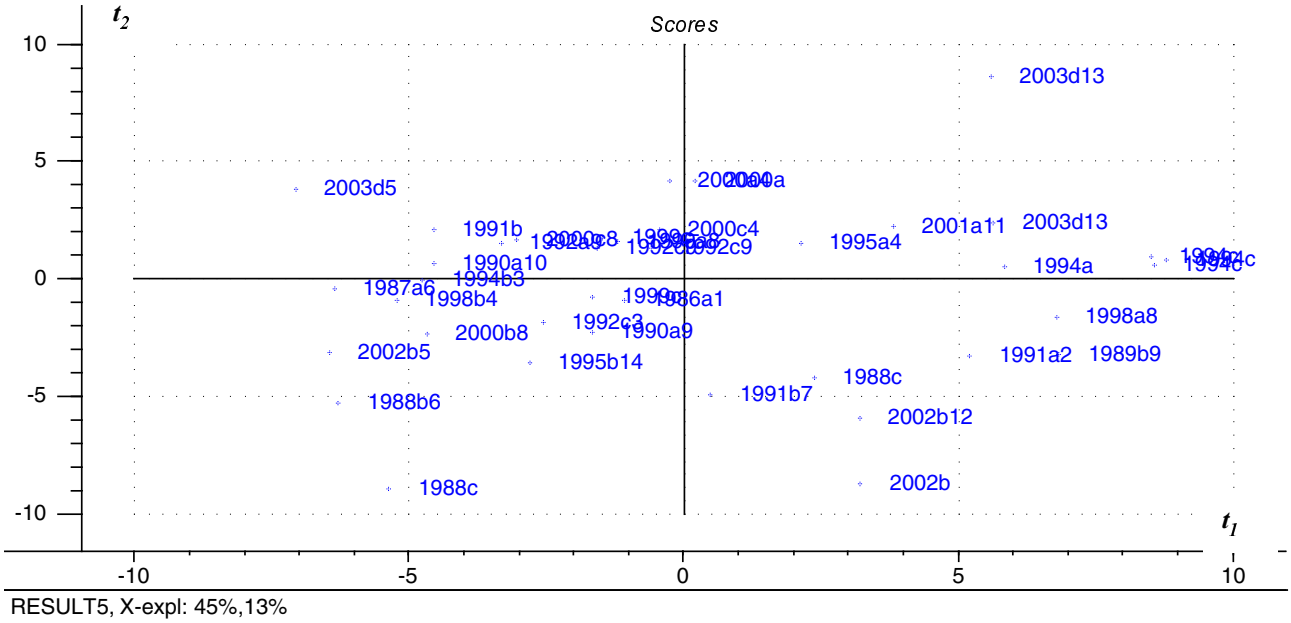

Figure 7. Score plot of t_2 versus t_1, showing the scores, or the position, of the objects in the hyperplane spanned by p_2 and p_1,. By looking at the score plot it seems difficult to identify any patterns in the egg sample scores indicating the presence of a time trend. There may be a tendency for lowest years to have negative score values in t_2 and of newest egg samples, i.e. years, to have positive score values in t_2.

The majority of chemicals within a chemical class are clustered together in p_1. Egg samples with high negative score values in t_1 have highest concentrations of the majority of chemicals with high negative loadings in p_1. On the other hand, egg samples with high positive score values in t_1 seem to differ from the egg samples with high negative score values in t_1 being better explained by the few chemical compound variables Me-TBBP-A, HBCD and to a lesser degree BDE-209.

The patterns in the score plot do not indicate the presence of any significant time trend, not even by removing the variables with positive loading values from the PCA. There is a small tendency for lowest years to have negative score values in t_2 and of more recent egg samples to have positive score values in t_2. This is supported by the BDEs having high loadings positive loading values in t_2 (cf. Appendix 5, Figure 4, section 3.4 describing time trends and the conclusions in chapter 4).

The egg samples with positive score values in t_2 are best described by the PBDEs having highest positive loadings in p_2. The egg samples with high negative score values in t_2 are best described by the toxaphene congeners, the organochlorine pesticides and the chlordanes having negative loading values in p_2. The same information can be obtained from Figure 3 and 4 in Appendix 5, where the concentration levels are included giving higher weight to chemical compound variables with a high concentration level.

3.3 Correlation between eggshell thickness and contamination

The correlation between eggshell thickness and mean concentration levels is shown for every chemical in Figure 8 (expressed as mean concentrations as shown in Figure 3 above). There is a clear tendency for the correlation coefficient to be negative, indicating a negative influence of contamination on eggshell thickness. The few chemicals for which the correlation coefficient is positive are all in the lower concentration range.

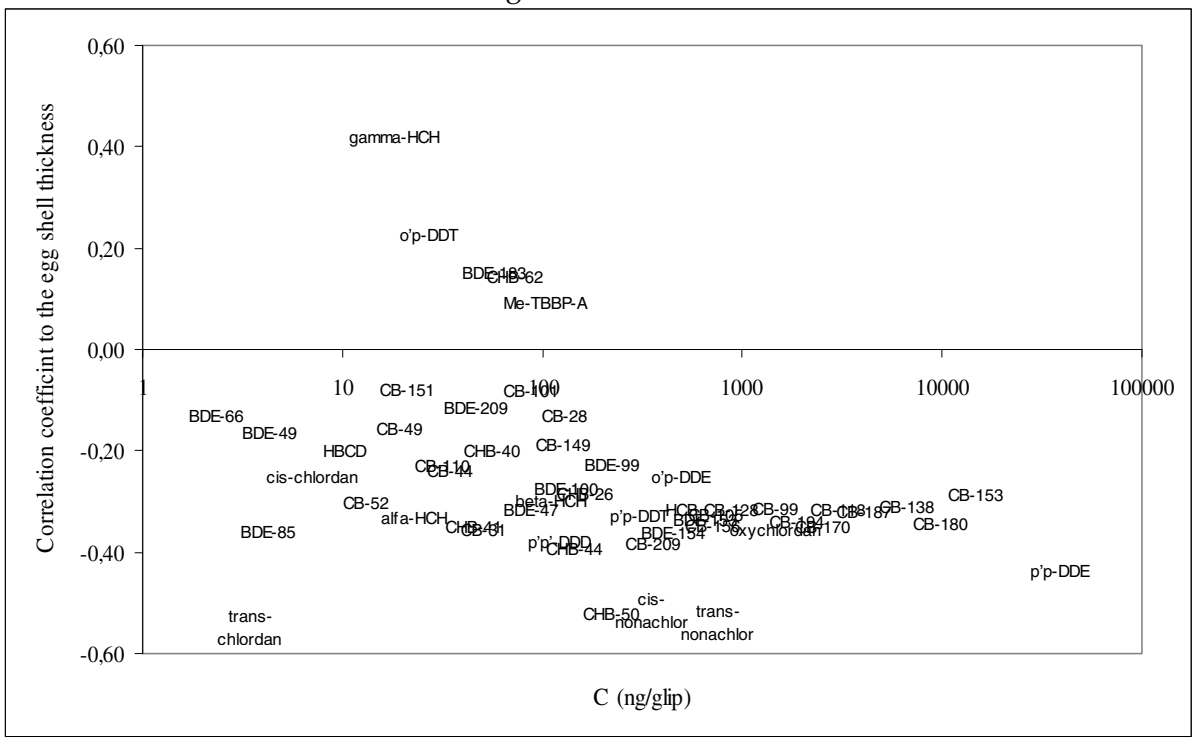

Figure 8. Correlation coefficients as a function of the concentration level for the specific substances. The substances BDE-17 and BDE-28 are removed due to few data points and thus higher statistical uncertainty. o,p'-DDT was not eliminated from this calculation, in spite of several values below the detection limit.

3.3.1 Influence of angle between two-ringed molecules in addition to concentration level on the eggshell thickness

The correlation between eggshell thickness and concentration level and flexibility of the molecules, respectively, has been investigated for chemical structures having a two-ringed structure analogous to the PCBs. The flexibility of the molecule is expressed by the number of ortho-substituents on the two-ring structures, which ranges from 1 to 4 as there are two ortho-positions on each of the connected ring-structures, i.e. the o, o, o´, o´ positions. This case study, presented in Appendix 6, shows a highly significant correlation between the egg-shell thickness and the levels of contamination as well as the flexibility of the molecules. For these structures, i.e. the PBDEs, the PCBs and the organochlorine pesticides, a combination of high contamination level and high molecular flexibility is suggested to induce thinning of eggshells. Top candidates are p,p'-DDE, p,p'-DDT while the o,p-configurations have lower rank in both rankings, i.e. based on correlation coefficients in set 1 and concentration and substitution parameters in set 2. Analogously, the PCBs with lowest degree of

ortho-substitutions have some tendency for higher rank in both rankings, with the exception of e.g. CB-153 which has lower rank in set 1 compared to set 2. There are visible tendencies for the existence of this structure-activity type relationship, which is well known for the PCBs. There are several investigations showing the influence of coplanarity, e.g. low or no ortho-substituents, determining the exposure-effect relationships in biological matrices as well as physical-chemical properties (Thomsen & Carlsen, 2002). For the PCBs it has been shown that the non-ortho-substituted PCBs, i.e. higher flexibility due to lower angle strain, increases the probability for a molecule to fit into a receptor and thus having lower effect concentration compared to ortho-substituted PCBs. However, the effect concentration level has to be exceeded for this relationship to be significant, as observable effects are a function of concentration no matter what the mechanism of action. Herzke et al. (2002) give a NOAEL of 3 µg/g ww for p,p'-DDE, while Peakall et al. (1975) state a threshold value of 20 ppm (p,p'-DDE) for declines in population. In spite of the negative correlation observed, the concentrations may be below the threshold levels.

The inverse correlation patterns between chemicals and eggshell thickness was further analysed by Partial Least Square (PLS) regression. The relative importance of chemicals in explaining the variation in the eggshell thickness is shown in Figure 9.

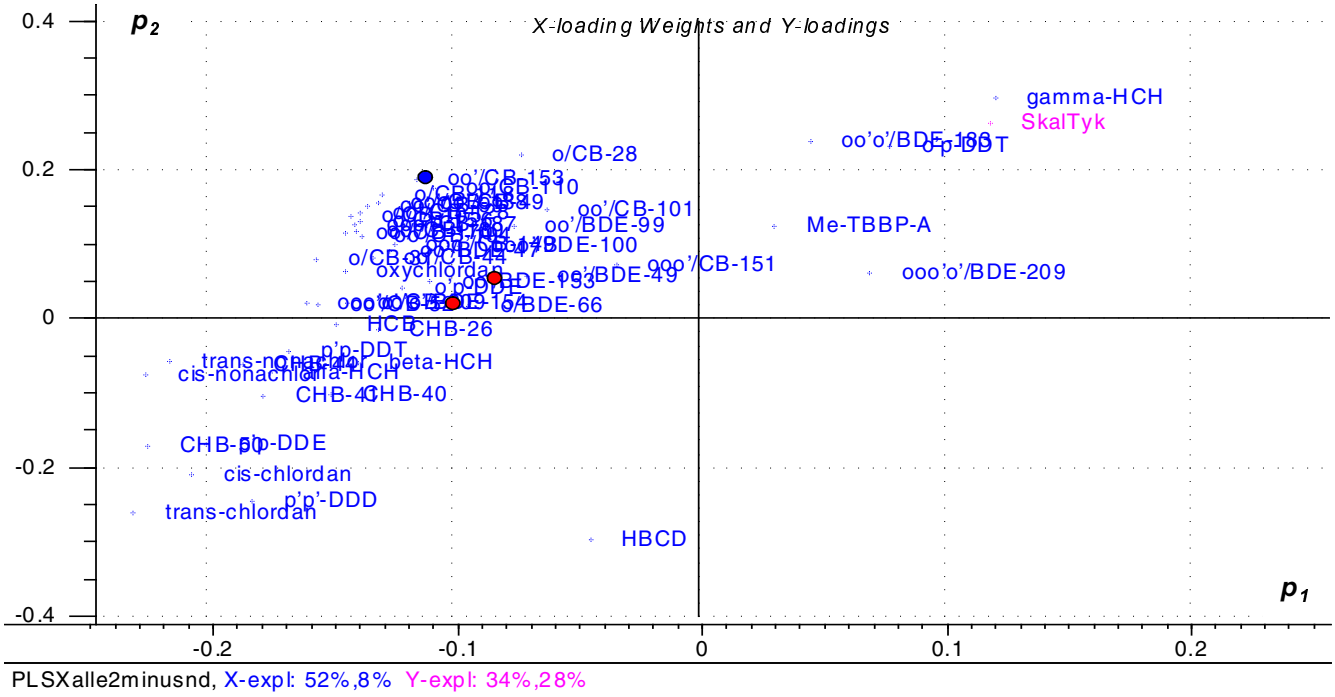

Figure 9. X-loading weights and Y-loading. The loading weights show the relative importance of chemical variables for explaining the variation in Y (SkalTyk_). 52% of the X- variance is used for explaining 34 % Y-variance in p_1, whereas 8 % X-variance is used for explaining 28% of the Y-variance in p_2. The red circles highlight the loading weights of BDE-153 and -154, whereas the blue circle represents CB-153. BDE-153 and CB-153 represent approximately one third of the total amount of PBDEs and PCBs, respectively.

The PLS-regression shows that the chemical compounds with high negative loading weights in both p_1 and p_2 are the chemicals with

highest importance regarding eggshell thickness, i.e. they show high importance for the explanatory capacity of both p_1 and p_2. These chemicals are positioned in the third quadrant in Figure 9, and include p,p´-DDT and degradation products as well as HBCD, trans- and cis-chlordane, CHB-26, -44, -41, -40 and -50, alfa- and beta-HCH and HCB. It should be mentioned that trans-chlordane has many missing data, but still the chlordanes and toxaphene congeners seem to dominate with high importance in both p_1 and p_2, which are a group of chemicals with increased effect on the eggshell thickness.

The BDEs do not have negative loading weights in p_2, where the chemicals with highest explanatory capacity, i.e. in both p_1 and p_2 are located. In the direction of p_2, the dominating PCB congener, CB-153, marked by a blue circle is not inversely related to the eggshell thickness. The most dominating BDEs, i.e. BDE-153 and –154, marked by red circles have low explanatory capacity in p_2. According to Figure 9, there are several potential chemicals with low effect concentration among the toxaphene congeners as well as the chlordanes. These chemicals are positioned in the third quadrant together with p,p´-DDT and its degradation products. This may indicate that the effects of these candidates are not zero even though the time trends are decreasing.

The high model performance of the PLS-regression based on BDEs alone suggests that the BDEs may already have some negative effect on the eggshell thickness at the present contamination level. The possible negative effects of the BDEs may not be visible yet due to the decreasing effects of the majority of the POPs showing a decrease in exposure level towards the peregrine falcons (cf. section 3.4 and Appendix 7).

In the PLS analysis (Appendix 5, Table 1) the goodness of fit of the regression to eggshell thickness, is shown for every group of chemicals and for all chemicals together. A relatively good correlation is seen for most of the chemical groups. The 12 BDEs have a better goodness of fit compared to the 31 PCBs and pesticides (cf. Appendix 7, Table 1). This may seem surprising because the highest concentration levels belong to the PCBs and organochlorine pesticides (cf. Figure 4) and this suggests a lower effect concentration of the BDEs with regard to eggshell thickness.

3.4 Time trend analysis

3.4.1 Chemicals

The temporal development for the total contamination is shown in Figure 10, in terms of the total concentration sum (ng/g lipid weight) of all compounds analysed. Apparently, there is no time trend in the overall contamination of the eggs, i.e. while the concentration of the "old" POPs in general decreases, the concentration of "new" compounds increases, causing a constant pollutant load. The concentrations of the "old" POPs are, however, still much higher than the increasing concentrations of the brominated flame retardants.

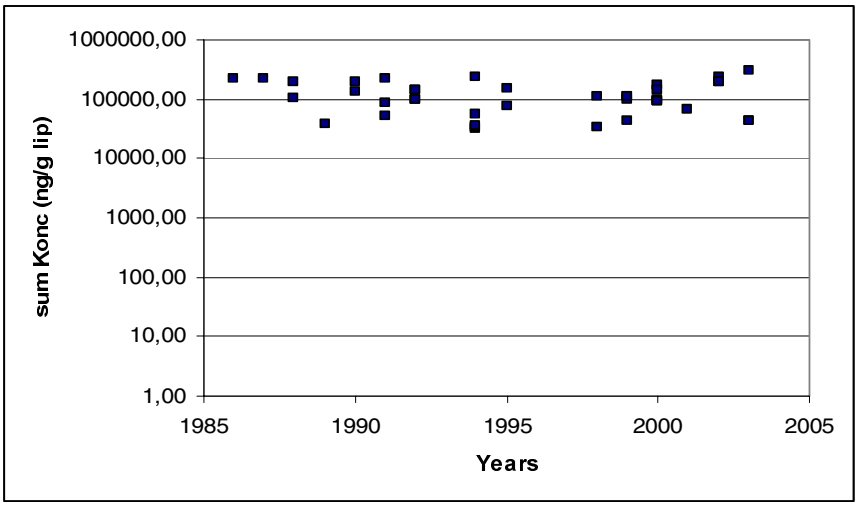

Figure 10. The temporal development in the concentration sum (i.e. all analysed compounds).

The time trend of the lipid normalised concentration is shown for all individual chemicals in Appendix 9 as ln-transformed values. There are rather clear trends both upwards and downwards for some of the chemicals. In general the downward trends belong to chemicals, which are no longer in use such as the PCBs. A representative example of such a downward trend is shown in Figure 11 for CB-110. The significance is defined as the probability for the "true" slope to have the same direction (upwards or downwards) as the fitted line slope.

Studies from the Baltic Sea have shown decreasing concentrations of PCBs and DDT since the 1970s, for instance in baltic guillemot *(Uria aalge)* eggs collected between 1969 and 1995 (Bignert et al., 1995; Bignert et al., 1998). The same temporal trend was found for freshwater fish from the Arctic regions in Sweden (Bignert et al., 1998). DDT in guillemot eggs started to decrease in the beginning of the 1970s, immediately after the international ban of DDT, while PCB did not decrease before 1975-1977. In the middle of the 1990s, DDT had decreased to concentrations less than 4% of that in the late 1960s. The decrease in PCB concentration occurred at a lower rate than that of DDT, indicating that there is ongoing PCB pollution (Bignert et al., 1998).

For both compound groups, the decrease seems to level out in the end of the 1980s and the beginning of the 1990s. While the concentrations have been almost unchanged for DDT since the mid-1980s, PCBs still decrease at a very low rate. These temporal trends observed for Baltic guillemot may be similar to the development observed in the peregrine falcon eggs. The time series studied in this project starts in 1986 and continues until 2003. Possibly, the main changes in the concentrations of PCBs and DDT occurred prior to the study period. Since 1986, an ongoing, but less pronounced drop in PCB concentration has occurred in the peregrine falcon eggs. The DDT, DDE and DDD chemicals tend to decrease in concentration but the tendency is not strong. The trend of p,p'-DDT and p,p´-DDE, which has the highest mean concentration, is constant in time,

whereas the decrease is a bit more significant for the o´,p-DDT and DDE (cf. Appendix 9). The constant concentrations of p,p'-DDE also agree with the findings by Bignert et al. (1998).

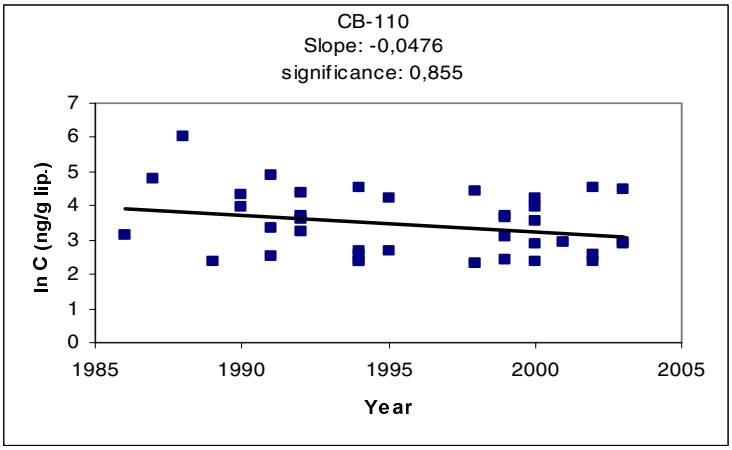

Figure 11. The time trend for CB-110. The significance is defined as the probability for the "true" slope to have the same direction (upwards or downwards) as the fitted line slope.

The chlordanes and the HCHs have the most significant decrease. The toxaphene congeners have a weak trend downward, similarly to the PCBs and the DDTs. As the only group of chemicals, the BDEs show an increasing trend as, e.g., shown in Figure 12 for BDE-99.

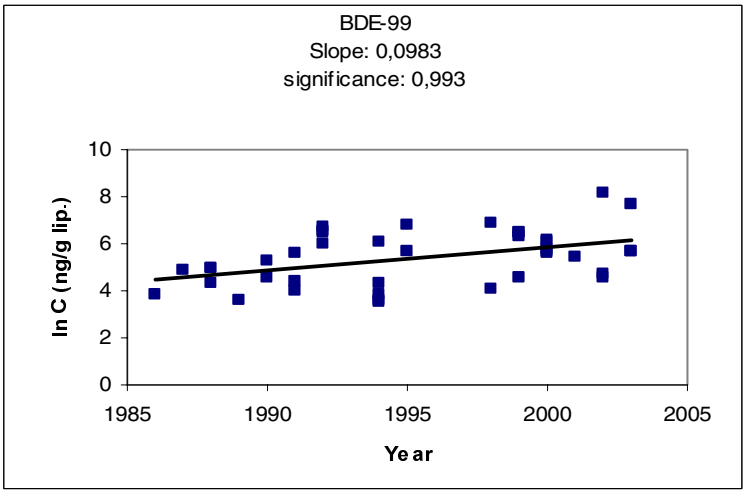

Figure 12. The time trend for BDE-99. The significance is defined as the probability for the "true" slope to have the same direction (upwards or downwards) as the fitted line slope.

The slope value in Figure 12 is around 0.1 and represents one of the steepest upward slope values for the brominated flame retardants. This corresponds approximately to a 10 % increase in concentration per year, or about a 3-fold increase in concentration over a 10-year period. In the USA, the production volume of the brominated flame retardants was 60000 tons in 1992, 95000 tons in 1995 and 155000 tons in 2004 (BKH, 2000) yielding an increase of about 8 % per year. The steepest linear slope for the brominated flame retardants in Appendix

7 has approximately the same value as the increase in production volume in the USA.

Ikonomou et al., (2002) shows a 10-fold increase in BDE contamination in ringed seals during approximately 20 years from year 1982 to year 2000 and thus an increase not much different from the increase observed in this investigation. However, other investigations have shown a more rapid increase in BDE contamination: up to a 300-fold increase during 20 years for Lake Ontario lake trout (Luross et al., 2000; Hale et al., 2003).

Brominated flame retardants in guillemot eggs from the Baltic Sea were analysed by Sellström et al. (2003) including eggs sampled between 1969 and 2001. The concentrations of respectively BDE-47 and BDE-99 show a peak value in the middle of the 1980s followed by a rapid decrease during the 1990s. Such a concentration peak is not seen in Figure 12, which shows a steady increase during the whole period. This difference might be related to geographical differences in the production and use of PBDEs, as well as the regulatory measures taken in Europe.

The time trend has also been investigated using multivariate statistical methods (cf. Appendix 7). From the PLS-regression the scoring of the two first principle components are related to the year in a regression analysis (Appendix 7, Table 3). Only a weak tendency is observed in years of egg samples and the scorings.

3.4.2 Egg shells

Measured eggshell thickness, listed in Appendix 3, includes more eggs than used for chemical analysis. The measurements are mainly based on large numbers of small fragments collected in all nests including those where all eggs hatched and, therefore, no whole eggs were available. This part of the investigation is described and discussed in detail in Appendix 8. Samples from a total of 93 clutches were measured and provided from 3 to 91 membrane-free measurements. However, since eggshell thickness varies within the egg there is a risk that too few samples may bias results. Hence, we chose to include only 75 clutches that provided 20 or more measurable fragments. The same threshold was selected by Odsjö (1982), in a study of Swedish Ospreys, and it is assumed that they represented the thickness of the entire clutch.

During the period 1981-2003 there was a weak but significant increase in the average thickness of eggshells (P=0.0253, N=79). The slope of the linear regression shows an average increase of 0.21% per year. This would correspond to a change in eggshell thinning from 12.8% in 1981 to 8.2% thinning in 2003 when compared to pre-DDT eggs collected in Greenland (0.336 mm, 48 eggs from 16 clutches, Falk & Møller 1990). If we assume the trend to be linear, the regression line can be extended backwards for a rough assessment of when/if the thinning exceeded the critical empirical "threshold" of about 17% (Peakall & Kiff 1988). The shell thinning might have been near the critical limit around 1950 – probably too short after DDT became widespread (introduced 1947) to have had a marked effect on the

Greenlandic Peregrine population as supported by evidence of a strong population since the 1970s (Burnham & Mattox 1984). This is despite the fact that the Arctic subspecies in Greenland migrates through and/or to areas (Latin America) where phasing out of the pesticides has been slower than in North America, or where a renewed use has been deemed necessary to fight Malaria.

To our knowledge, this is the first time a long-term increase in eggshell thickness has been detected in a Peregrine Falcon population. Nygård (1999) observed a slight increase in shell thickness of eggs from another bird of prey, Norwegian Merlins (*Falco columbarius*), when comparing eggs from the 1990s (8-11% reduction) to eggs from the 1960s and 1970s (15% reduction).

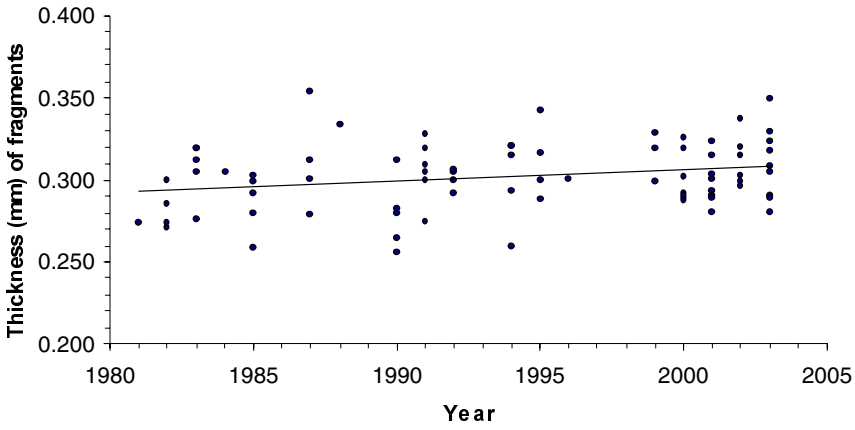

Figure 13. The eggshell fragment thickness data including.

4 Conclusion

Levels of contamination

- The concentration range between the different chemicals is large, covering 5 orders of magnitude. The variability between eggs is still considerable, but much smaller than between compounds.
- PCBs and persistent organochlorine pesticides dominate the contamination profile. On average, the sum of PCBs and DDT account for 95 % of the total contaminant load analysed in the eggs.
- Median summed concentrations were 55 µg/g lw for PCBs and 40 µg/g lw for DDT (and its degradation products).
- The concentration of p,p'-DDE did not exceed the threshold of 20 ppm (wet weight) beyond which population declines occur. However, 42 % of the eggs analysed exceeded the NOAEL level of 3 µg/g wet weight for p,p'-DDE.
- PBDEs were detected in all eggs, with a medium summed concentration of 1.9 µg/g lw. This is among the highest PBDE concentration ever detected in wildlife. The main congener was BDE-153.
- BDE-209 was detected in all eggs analysed, which proves that BDE-209 is bioavailable and accumulates in biota.
- HBCD was likewise detected in the eggs, however, the concentrations were low. TBBPA was only detected in terms of the degradation product dimethyl-TBBPA.
- The large concentration ranges make comparison with the literature data difficult. PCB concentrations were lower than in Norwegian peregrine falcon eggs, but higher than in samples from Alaska. DDT concentrations were similar in Greenland, Norway and Alaska. Summed PBDE concentrations were similar to the results for wild peregrine falcons in Sweden.

Correlations between chemicals

- The correlation between chemicals is positive, and a strong correlation exists within the same class of chemicals.

Correlation between contamination and eggshell thickness

- The correlation coefficient between the concentration and the eggshell thickness is negative. This indicates a negative influence of the contaminants on the eggshell thickness.
- The intercorrelation between the contaminants analysed does not allow the identification of particular chemicals with the strongest effect on eggshell thickness.
- Molecular flexibility is negatively correlated with eggshell thickness.
- Both the PLS-regression and single chemical- eggshell thickness correlation matrix, suggests that the PBDEs have a negative influence on eggshell thickness.
- Overall the eggshell thickness is showing a small increasing tendency which could indicate that the concentration levels of the BDEs are below the critical effect concentration.

Time trend in chemical contaminations

- The total concentration of all chemicals analysed is constant over the time period studied.
- Chemicals such as PCBs, which are no longer in use, have a downward time trend. DDT remains constant. The main decrease in PCB and DDT concentration might have occurred prior to the time period studied in this project.
- As the only group of chemicals, the majority of the PBDEs show an increasing trend. This is in contrast to results for guillemot eggs from the Baltic Sea, but in agreement with studies from North America. This difference might be caused by regulatory measures taken in Europe.
- Ongoing increases in PBDE concentrations might affect eggshell stability in a similar manner to organochlorine compounds.

Time trend in eggshell thickness

- Both eggshell measurements and multivariate statistics show a positive time trend for the eggshell thickness, which, however, is not statistically significant.

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Appendix 1

Compounds selected for analysis

Introduction

The compounds analysed in this project included various chlorinated compounds, such as polychlorinated biphenyls (PCBs) and organochlorine pesticides, as well as brominated flame retardants (BFR) (Table 1). With the exception of HCH, all organochlorine compounds are listed in the Stockholm Convention of persistent organic pollutants (POPs), which restricts or prohibits the production, trade and use of these compounds because of their persistence, bioaccumulation, long-range transport and adverse health effects. The following characterisation is mainly based on information published in relation to monitoring of POPs in the Arctic (de March et al., 1998; AMAP, 2004).

Table 1 Compounds selected for analysis

Compound group	Acronym	Congeners and analytes	Legal status in Denmark (de March et al., 1998)
Polychlorinated biphenyls	PCB	CBs 28, 31, 44, 49, 52, 99, 101, 105, 110, 118, 128, 138, 149, 151, 153, 156, 170, 180, 187, 188, 194, 209	Prohibited
DDT and degradation products	DDT	p,p'-DDT, p,p'-DDE, p,p'-DDD, o,p'-DDT, o,p'-DDE	Prohibited for plant protection use
Toxaphene	CHB	CHBs 26, 40, 41, 44, 50, 62	Banned since 1987
Chlordane-related compounds		oxychlordane, cis-chlordane, trans-chlordane, cis-nonachlor, trans-nonachlor	Prohibited for plant protection use
Hexachlorocyclohexanes	HCH	α-HCH, β-HCH, γ-HCH	Mixed isomers prohibited for plant protection use
Hexachlorobenzene	HCB	HCB	Banned
Polybrominated diphenyl ethers	PBDE	BDEs 17, 28, 47, 49, 66, 99, 100, 153, 154, 183, 209	Restriction of penta- and octa-BDE, future ban
Hexabromocyclododecane	HBCD	HBCD	no restrictions known
Tetrabromobisphenol A	TBBPA	TBBPA	no restrictions known

Persistent organochlorine pesticides

The technical product **dichlorodiphenyltrichloroethane (DDT,** Figure 1) contains the p,p'- and o,p'-DDT isomers as well as p,p'-DDD and o,p'-DDD. DDE is the more persistent and more toxic metabolite of DDT. DDT was introduced as an insecticide in 1945 and has found broad application all over the world. Being restricted or prohibited in the USA, Canada and Western Europe, it is likely that its use continues in other parts of the world. The regulatory measures have led to decreasing levels in the environment, however, high levels of DDT and DDE in top predators of the Arctic are still a great concern.

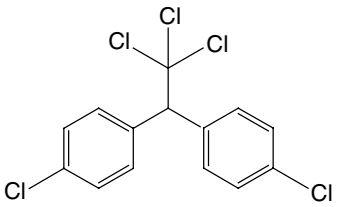

Figure 1: Chemical structure of DDT

The organochlorine pesticide **toxaphene** (Figure 2) is a complex mixture of polychlorobornanes and camphenes with six to ten chlorine atoms and was widely used in the USA on cotton crops in the 1970s. Having been banned in the USA since the 1980s, it may still be used in other countries for pest control on cotton. With half-lives in soil of up to 14 years and a relatively high vapour pressure (1.9×10^{-3} Pa at 25°C) it has been dispersed ubiquitously. Monitoring results from the Arctic indicates that it follows the fate of other persistent organochlorine pesticides and PCBs.

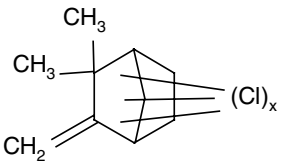

Figure 2: Chemical structure of toxaphene

Cis- and trans-**chlordane** as well as cis- and trans-nonachlor are among the main components of technical chlordane mixtures, which contain at least 120 different compounds. Oxychlordane is a metabolite of chlordane with high acute toxicity. The mixture was mainly used as an insecticide in the 1970s and early 1980s and was further used in termite control in the USA up to 1988. Especially cis- and trans-chlordane have high Henry's Law coefficients of 85 and 132 $Pa \cdot m^3/mol$, respectively, which make them more volatile than other pesticides. Combined with their persistence in the environment, they are likely to undergo long-range transport and have been detected extensively in the Arctic. The wide occurrence of the toxic metabolite oxychlordane has been of particular concern.

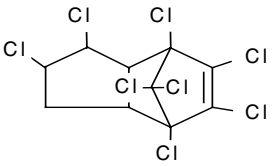

Figure 3: Chemical structure of chlordane

The technical **hexachlorocyclohexane (HCH**, Figure 4) mixture is comprised of 55-80 % α-HCH, 5-14 % β-HCH, 8-15 % γ-HCH and 5-21 % δ- and ε-HCH (Li et al., 1996). γ-HCH (lindane) is the isomer with highest insecticidal effect and may still be applied in pest control, whereas the use of the other isomers or the technical mixtures has been prohibited in most countries since the late 1970s. α-HCH is the most volatile isomer, but all isomers are capable of long-range transport. The Henry's Law coefficient of β-HCH is about 200 times lower

than that of α-HCH, which leads to differences in the spatial distribution of the isomers (Li et al., 2002). HCH does not bioaccumulate to the same extent as other organic compounds. It has the lowest K_{OW} of the compounds analysed in this study, which is consistent with the lowest biomagnification.

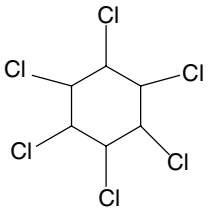

Figure 4: Chemical structure of HCH

Chlorinated industrial products

The **PCB** group consists of 209 congeners with different degrees of chlorination (Figure 5). PCBs were widely used as transformer and capacitor oils, and hydraulic and heat exchange fluids until the 1970s and are still in use in some closed systems today. They have proven very stable in the environment, in particular the congeners lacking adjacent unsubstituted positions on the biphenyl rings, e.g. 2,4,5-, 2,3,5, or 2,3,6-substitution. The half-lives of PCBs in air have been estimated to range from three weeks to two years, thus enabling long-range transport to remote regions. PCBs have been detected in a number of species in Greenland and other Arctic regions, with a tendency of bioaccumulation and biomagnification in the marine food web. The highly active co-planar PCBs were not included in this study.

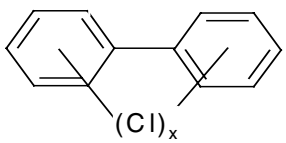

Figure 5: Chemical structure of PCBs

Hexachlorobenzene (HCB, Figure 6) was once used as a fungicide on grains, however, its major source is now thought to be as a production by-product of a large number of chlorinated compounds, particularly lower chlorinated benzenes, and several pesticides (Bailey, 2001). It is mainly emitted to the atmosphere in flue gases generated by waste incineration and industry and persists in the environment with a half-life of several years. With a $\log K_{OW}$=5.5, HCB is lipophilic and bioaccumulates in lipid-rich tissues. It has been detected in numerous species in the Arctic.

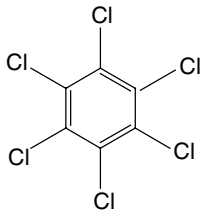

Figure 6: Chemical structure of HCB

Brominated flame retardants

Polybrominated diphenyl ethers (PBDEs) are aromatic compounds structurally similar to PCBs (Figure 7). In their function as flame retardants in electric and electronic equipment, textiles and paint, they are added to polymers without forming covalent bonds and can therefore leach into the environment during production, use and disposal of the products (Sjödin et al., 2001). Since the early 1980s, PBDEs have been detected in all compartments of the environment word-wide, making them global and ubiquitous contaminants. So far, their occurrence and fate in the Arctic have not been studied as extensively as for PCBs, however, the knowledge available regarding PBDEs is increasing. Tetra- and penta-BDEs have $\log K_{OW}$-values of 5.9-7.0 and have been shown to bioaccumulate similarly to PCBs. For octa- through deca-BDE, $\log K_{OW}$-values of 8.4-10 have been estimated, but little is known about the bioaccumulation potential. A Swedish study detected PBDEs in peregrine falcon eggs and observed a relatively high amount of higher brominated congeners, such as BDE-99 and BDE-153. In contrast to the general assumption that deca-BDE was not bioavailable, it was also detected in peregrine falcon eggs (Sellström et al., 2001, Lindberg et al., 2004).

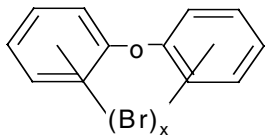

Figure 7: Chemical structure of PBDEs

Tetrabromobisphenol A (TBBPA, Figure 8) is the most abundant brominated flame retardant currently in use (Hakk, 2001). Between 1992 and 1998, the global demand for TBBPA had increased from 50 000 to 145 000 tons per year and is expected to keep increasing. In Europe, the annual demand for TBBPA is estimated to be 40 000 tons (RIKZ, 2000). About 90 % of TBBPA use is for the production of resins used in printed circuit boards (Hakk, 2001). TBBPA has a very low vapour pressure (< 133×10^{-6} Pa at 20°C). The water solubility is reported as 720 µg/l at 20°C, but increasing to 4.2 mg/l at 25°C. The log K_{OW} has been determined as 4.5-5.3 (RIKZ, 2000). Information about TBBPA in the environment is still rare. In water and air, TBBPA was only found in traces, even near production sites, probably as a consequence of sorption to sediment and soil. A Swedish study showed increasing TBBPA levels downstream a plastic-producing plant (Sellström and Jansson, 1995). In soils, TBBPA has half-lives of 50-100 days and is degraded both aerobically and anaerobically. Even

though its hydrophobicity (log K_{OW}= 4.5) indicates bioaccumulation, both TBBPA is rapidly excreted from biological species (WHO, 1995).

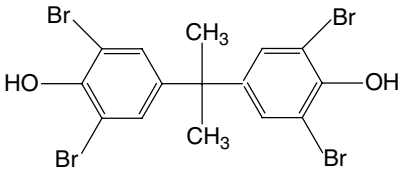

Figure 8: Chemical structure of tetrabomobisphenol A (TBBPA)

Hexabromocyclododecane (HBCD) is a non-aromatic brominated flame retardant mainly used in polystyrene resins and textiles (Lund et al., 2001). Its demand in the European Union is estimated to be about 10 000 tons per year. HBCD has a vapour pressure of $62.7x10^{-6}$ Pa at 20°C and low water solubility (3.4-8 µg/l). The log K_{OW} is 5.8-7.0 (RIKZ, 2000). Similarly to TBBPA, data on HBCD in the environment are still rare. However, HBCD was included in a Swedish study on peregrine falcon eggs, yielding concentrations in a range of 34-2400 ng/g lw (Sellström et al., 2001, Lindberg et al., 2004).

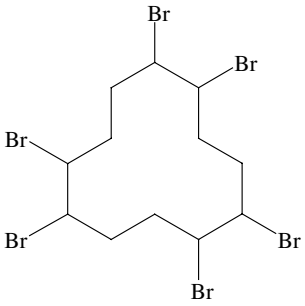

Figure 9: Chemical structure of hexabromocyclododecane (HBCD)

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Appendix 2

Analytical Methods

Introduction

The compounds analysed in this project are listed in Appendix 1 and include organochlorine pesticides, polychlorinated biphenyl ethers (PCBs) and various brominated flame retardants. As most of these contaminants have similar physical-chemical characteristics, it was possible to combine the analyses of several compound groups. Wherever possible, the sample extraction, clean-up and preconcentration procedures as well as the instrumental analyses included more than one compound group. Therefore, in order to avoid repetition, the analytical methods will not be described for each compound group separately.

The main steps are illustrated in Figure 1. A detailed description of the analysis of PCBs, HCB, the HCH isomers and DDT and its degradation products is given by Cleemann et al. (1999). This analytical method is based on gas chromatography – electron capture detection (GC-ECD). The analysis of PBDEs is further described by Christensen et al. (2002) and Vorkamp et al. (2004). PBDEs, chlordane-related pesticides and toxaphene are analysed by GC- mass spectrometry (GC-MS) in the negative chemical ionisation mode (NCI).

The brominated flame retardants HBCD and TBBPA as well as the metabolite dimethyl-TBBPA were analysed by the Netherlands Institute for Fisheries Research (RIVO). The analytical method is based on the procedures described by de Boer et al. (2001) and summarised in the laboratory report C077/03 prepared by Leslie et al. (2003).

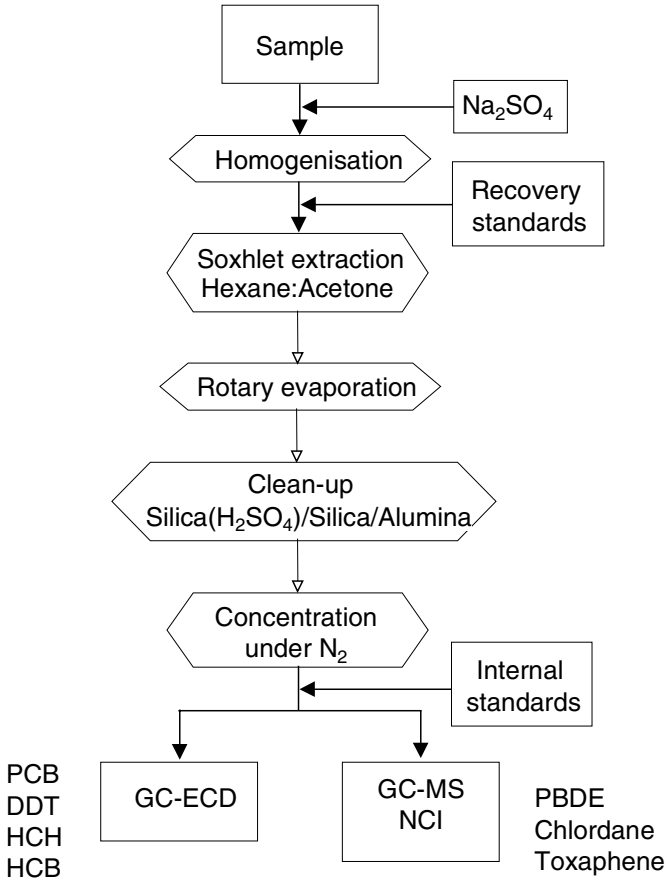

Figure 1: Analytical methods

Extraction and purification

After homogenisation, about 3.5 g of each egg sample were dried with anhydrous Na_2SO_4, and spiked with the recovery standards. In the first sample batch, approximately 6 g of sample were taken. As the POP concentrations in the egg samples generally were high, the amount of sample was reduced in the following analyses. Thus, matrix effects were reduced and more of the valuable material could be kept for other purposes. Special care was taken to avoid UV-influence, as BDE-209 is likely to be degraded by UV-radiation.

The samples were Soxhlet extracted using 350 ml of a mixture of *n*-hexane and acetone (4:1, v/v) and concentrated by rotary evaporation to a volume of 1 ml. The extracts were cleaned on a multilayered glass column packed with 5 g deactivated aluminium oxide containing 10 % water, 1 g activated silica (24 h at 160°C), 5 g activated silica impregnated with concentrated sulphuric acid and 1 cm anhydrous Na_2SO_4, and eluted with 250 ml *n*-hexane. The cleaned extracts were concentrated to about 1 ml by rotary evaporation with iso-octane as keeper and under nitrogen. After defined amounts of the internal standards were added, the samples were adjusted to a precise volume of 1 ml.

For analysis of TBBPA and HBCD, the egg samples were Soxhlet extracted using a mixture of *n*-hexane and acetone (3:1, v/v). The extracts were purified by gel permeation chromatography and on a silica column. The recovery and internal standards added to the sam-

ples are summarised in Table 1. Details on purchase sources, chemical purity etc. are given in the references listed in the introduction.

Table 1: Standards and instruments used in the analyses of peregrine falcon eggs

Compound group	Recovery standards	Internal standards	Analytical instrument
PCB	CB-3	CB-53	GC-ECD
DDT	CB-40	CB-155	
HCH	CB-198		
HCB			
Toxaphene	CB-198	^{13}C-Mirex [a)]	GC-MS
Chlordane-related compounds	CB-198	^{13}C-Transchlordane	GC-MS
PBDE	BDE-77	BDE-71	GC-MS
BDE-209	^{13}C-BDE-209	^{13}C-BDE-209	GC-MS
HBCD	^{13}C-HBCD	[b)]	LC-MS and GC-MS
TBBPA	^{13}C-TBBPA		

[a)] In these analyses, quantification of toxaphene was also based on ^{13}C-transchlordane since the matrix was found to interfere with ^{13}C-mirex. [b)] Quantification was based on external standards.

Instrumental analysis

PCBs, DDT, HCH and HCB were analysed by GC-ECD (Table 1). The technical details regarding capillary columns, temperature programming and calibration are described by Cleemann et al. (1999) and Vorkamp et al. (2004). The analysis by GC-ECD includes two chromatographic columns of different polarity (DB-5 and DB-1701, each 60 m long, 0.25 mm internal diameter, 0.25 µm film thickness). Thus, each analysis produces two results, which ideally should be identical. For four compounds (CB-28, CB-31, CB-99, o,p'-DDE) only one result is available due to co-elution on the other column. The selection of one signal becomes necessary in case of chromatographic or calibration problems on one of the columns. In the usual procedure, an average of the two results is calculated. In case of >10% difference, the lower value is taken, as higher values might reflect interference. Quantification is based on two internal standards and a duplicate 7-point-calibration.

The methods for chlordane-related compounds, toxaphene and PBDEs are based on GC-MS with negative chemical ionisation (NCI). Methane is used as the ionisation gas. Chlordanes and toxaphene are analysed in one analytical run, while PBDEs are analysed separately. Details on m/z-values, temperature programmes and calibration are given by Christensen et al. (2002) and Vorkamp et al. (2004). The same column (DB-5) was chosen for the three compound groups. Quantification is based on duplicate 8-point-calibrations and the internal standards given in Table **1**.

The deca-brominated congener BDE-209 was analysed on a DB-1 capillary column with a length of 15 m (0.25 mm internal diameter, 0.25 µm film thickness). The shorter column is necessary to minimise thermal degradation of BDE-209, which is favoured by long exposure to elevated temperatures in the GC oven (de Boer et al., 2003). Other technical specifics were identical to the method for the lower brominated PBDEs.

Data on TBBPA and HBCD were obtained by liquid chromatography-mass spectrometry (LC-MS). This analytical method allowed the

separation of the three stereomers α-HBCD, β-HBCD and γ-HBCD. Besides, the TBBPA-metabolite dimethyl-TBBPA was determined. Quantification was based on external standards. Since the LC-MS method consistently yielded concentrations below the detection limit for HBCD, the samples were re-analysed by GC-MS. Thus, concentrations of total HBCD could be determined for most of the samples.

Quality assurance and quality control

Each batch consisted of 12 samples, one of which was analysed in duplicate and three blanks, two of which contained ^{13}C-BDE-209. Furthermore, four samples of internal reference material (sand launch oil) were analysed per batch in order to assess the precision of the analysis. Since the reference material does not contain detectable amounts of BDE-209, two samples of reference material were spiked with BDE-209. Precision was monitored by plotting the results of the internal reference material in control charts with warning and action limits (2 and 3 times the standard deviation of the target value, respectively).

The overall quality of the analyses was monitored by regular participation in QUASIMEME intercalibration exercises on PCBs and toxaphene in biota as well as the BSEF/QUASIMEME development exercises on brominated flame retardants (de Boer et al., 2002). Results of the time period 1999-2002 are summarised by Asmund et al. (in press).

The RIVO method for TBBPA and HBCD basically used the same means of quality assurance, including eel and sediment as internal reference materials. The method was tested by participation in the BSEF/QUASIMEME interlaboratory study on brominated flame retardants and was confirmed with good results (de Boer et al., 2002).

References

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Appendix 3

Concentration levels for brominated flame retardants

Ring No.:		3050122	3050142		3050526	3050541
Reg. No.:	01-1597	01-1601	01-1607	01-1609	01-1616	01-1618
Batch No.:	#03-16	#03-16	#03-16	#03-16	#03-16	#03-16
Year of sampling:	1988	1992	2000	1988	2000	1990
Place of sampling:	Morten	Eqaluit	Sdr. Igaliko	Igaliko	Upernaviarsuk	Skyggesø
Sample weight (g):	3.04	3.34	3.56	3.52	3.05	3.54
Fat content (%):	20.31	9.31	3.58	22.42	4.63	n.a.
Dry matter (% weight):	38.73	18.25	13.34	45.16	17.54	21.09
Substance	Concentration in ng/g wet weight					
BDE-17	<0.10	<0.09	<0.09	0.26	<0.10	<0.09
BDE-28	<0.10	<0.09	<0.09	0.22	<0.10	<0.09
BDE-49	0.27	0.36	0.22	1.28	0.46	0.21
BDE-47	5.93	12.46	2.86	41.12	6.54	9.03
BDE-66	0.07	0.21	<0.04	0.88	0.13	0.11
BDE-100	6.86	21.33	6.96	18.59	15.70	3.91
BDE-99	15.57	36.63	10.01	32.46	20.29	6.10
BDE-85	<0.10	0.11	<0.09	0.26	0.12	<0.09
BDE-154	66.98	31.95	19.54	175.44	44.15	18.69
BDE-153	33.12	62.71	55.83	82.48	74.53	38.19
BDE-183	8.13	17.85	5.37	7.24	2.67	6.40
BDE-209	2.98	0.89	0.36	1.58	0.51	0.48
	Concentration in ng/g fat weight					
BDE-17	< 0.5	< 1.0	< 2.5	1.1	< 2.2	
BDE-28	< 0.5	< 1.0	< 2.5	1.0	< 2.2	
BDE-49	1.3	3.8	6.3	5.7	9.8	
BDE-47	29.2	133.8	80.0	183.4	141.3	
BDE-66	0.4	2.3	< 1.0	3.9	2.7	
BDE-100	33.8	229.1	194.3	82.9	339.1	
BDE-99	76.7	393.5	279.7	144.8	438.3	
BDE-85	< 0.5	1.2	< 2.4	1.2	2.7	
BDE-154	329.8	343.2	545.7	782.5	953.6	
BDE-153	163.1	673.6	1559.6	367.9	1609.6	
BDE-183	40.0	191.7	150.0	32.3	57.7	
BDE-209	14.7	9.5	10.0	7.1	11.0	
HBCD		1.20	14.00		< 0.10	230.00
Me-TBBP-A		< 0.10	700.00		900.00	400.00

Ring No.:	3050541	3050541				
Reg. No.:	01-1620	01-1622	01-1630	01-1631	01-1633	02-1789-1
Batch No.:	#03-16	#03-16	#03-16	#03-16	#03-16	#03-16
Year of sampling:	1992	1992	1994	1994	1994	1999
Place of sampling:	Skyggesø	Skyggesø	Havnen	Havnen	Havnen	Upernaviarsuk
Sample weight (g):	3.62	3.45	3.61	3.67	3.57	3.71
Fat content (%):	6.84	5.63	6.74	7.92	5.73	3.98
Dry matter (% weight):	17.74	15.58	17.87	20.85	15.54	16.04
Substance	Concentration in ng/g wet weight					
BDE-17	< 0.09	< 0.09	< 0.09	< 0.09	< 0.09	< 0.08
BDE-28	< 0.09	< 0.09	< 0.09	< 0.09	< 0.09	< 0.08
BDE-49	0.15	0.13	< 0.09	< 0.09	< 0.09	0.59
BDE-47	14.85	11.02	0.84	0.73	0.60	7.34
BDE-66	0.07	0.05	< 0.03	< 0.04	< 0.04	0.06
BDE-100	18.69	13.60	1.18	1.64	1.44	11.45
BDE-99	48.36	36.01	2.32	2.90	2.66	22.67
BDE-85	0.18	0.12	< 0.09	< 0.08	< 0.09	0.60
BDE-154	28.50	20.93	11.39	14.51	12.57	28.96
BDE-153	52.47	36.45	10.59	13.94	11.14	43.92
BDE-183	3.79	1.51	1.19	1.30	1.17	1.19
BDE-209	1.09	0.59	0.50	0.41	0.35	0.48
	Concentration in ng/g fat weight					
BDE-17	< 1.3	< 1.6	< 1.3	< 1.1	< 1.5	< 2.1
BDE-28	< 1.3	< 1.6	< 1.3	< 1.1	< 1.5	< 2.1
BDE-49	2.2	2.3	1.3	1.1	1.5	14.9
BDE-47	217.1	195.8	12.4	9.2	10.5	184.5
BDE-66	1.1	1.0	< 0.5	< 0.4	< 0.6	1.6
BDE-100	273.2	241.6	17.5	20.6	25.1	287.7
BDE-99	707.0	639.6	34.4	36.6	46.5	569.5
BDE-85	2.6	2.2	< 1.3	< 1.1	< 1.5	15.1
BDE-154	416.6	371.7	169.0	183.3	219.4	727.8
BDE-153	767.0	647.4	157.2	175.9	194.4	1103.5
BDE-183	55.4	26.8	17.7	16.4	20.4	30.0
BDE-209	16.0	10.6	7.4	5.2	6.1	12.0
HBCD	26.00	32.00	77.00	67.00	< 0.10	< 0.10
Me-TBBP-A	430.00	480.00	230.00	240.00	270.00	905.00

Ring No.:		3050501	3050541	3050502		3050122
Reg. No.:	02-1789-2	01-1610	01-1617	01-1611	01-1629	01-1602
Batch No.:	#03-16	#03-14	#03-14	#03-14	#03-14	#03-14
Year of sampling:	1999	1988	1989	1991	1991	1994
Place of sampling:	Upernaviarsuk	Igaliko	Skyggesø	Bagerfalken	Hosp.dal	Eqaluit
Sample weight (g):	3.38	3.40	3.41	3.52	3.30	3.36
Fat content (%):	3.93	18.93	10.69	6.84	5.15	7.48
Dry matter (% weight):	16.27	35.29	22.42	18.97	17.45	22.36
Substance	Concentration in ng/g wet weight					
BDE-17	< 0.09	0.17	< 0.09	< 0.09	< 0.09	< 0.09
BDE-28	< 0.09	0.18	< 0.09	< 0.09	< 0.10	< 0.09
BDE-49	0.57	0.82	< 0.09	0.11	0.13	0.43
BDE-47	8.25	29.26	1.31	2.81	8.38	11.00
BDE-66	0.07	0.71	< 0.09	< 0.09	0.28	0.29
BDE-100	13.21	14.92	2.63	3.56	12.67	30.37
BDE-99	25.50	27.13	3.92	5.56	13.58	33.61
BDE-85	0.55	0.13	< 0.09	< 0.09	< 0.09	0.32
BDE-154	32.57	138.02	17.43	18.80	46.05	50.45
BDE-153	47.11	72.08	37.86	14.50	50.52	101.41
BDE-183	1.10	7.02	10.76	3.43	2.40	9.74
BDE-209	0.39	1.31	1.01	0.43	0.35	1.10
	Concentration in ng/g fat weight					
BDE-17	< 2.4	0.9	< 0.8	< 1.3	< 1.7	< 1.2
BDE-28	< 2.4	0.9	< 0.8	< 1.3	< 1.9	< 1.2
BDE-49	14.5	4.3	< 0.8	1.6	2.5	5.8
BDE-47	210.0	154.6	12.2	41.1	162.8	147.1
BDE-66	1.9	3.7	< 0.8	< 1.3	5.5	3.8
BDE-100	336.2	78.8	24.6	52.1	246.0	406.0
BDE-99	648.8	143.3	36.7	81.3	263.7	449.3
BDE-85	13.9	0.7	< 0.8	< 1.3	< 1.7	4.3
BDE-154	828.6	729.1	163.0	274.9	894.2	674.4
BDE-153	1198.7	380.8	354.1	212.0	981.1	1355.8
BDE-183	28.0	37.1	100.7	50.1	46.7	130.2
BDE-209	10.0	6.9	9.4	6.3	6.8	14.7
HBCD			7.80	10.00	22.00	2.40
Me-TBBP-A			< 0.10	< 0.10	940.00	< 0.10

Ring No.:	98764543	3050142		3050526	3050491	3050563
Reg. No.:	02-1785	01-1604	01-1626	01-1615	02-1788	02-1787-1
Batch No.:	#03-14	#03-14	#03-14	#03-14	#03-14	#03-14
Year of sampling:	1995	1998	1999	2000	2002	2002
Place of sampling:	Lejren	Sdr.Igaliko	Lejren	Upernaviarsuk	Enoraq	Qanisartut
Sample weight (g):	3.52	3.45	3.38	3.42	3.34	3.47
Fat content (%):	6.30	5.80	6.02	3.94	6.87	5.27
Dry matter (% weight):	18.96	34.20	17.26	13.65	20.05	15.91
Substance	Concentration in ng/g wet weight					
BDE-17	< 0.09	< 0.09	< 0.09	< 0.09	< 0.09	< 0.09
BDE-28	< 0.09	< 0.09	< 0.09	< 0.09	< 0.09	< 0.09
BDE-49	0.34	1.64	0.09	0.35	0.40	0.09
BDE-47	28.53	31.89	2.85	4.76	82.66	1.84
BDE-66	0.22	1.34	< 0.09	0.11	0.37	< 0.09
BDE-100	23.60	31.57	4.06	13.89	87.70	3.51
BDE-99	54.47	56.65	5.68	19.41	234.50	4.89
BDE-85	0.40	0.72	< 0.09	0.11	3.55	< 0.09
BDE-154	51.35	35.73	12.77	42.74	98.18	5.60
BDE-153	43.75	75.08	19.14	69.10	382.61	27.29
BDE-183	2.49	13.76	0.93	3.31	37.77	0.94
BDE-209	0.27	0.94	0.37	0.75	16.97	0.38
	Concentration in ng/g fat weight					
BDE-17	< 1.4	< 1.6	< 1.5	< 2.3	< 1.3	< 1.7
BDE-28	< 1.4	< 1.6	< 1.5	< 2.3	< 1.3	< 1.7
BDE-49	5.4	28.3	1.5	8.8	5.8	1.6
BDE-47	452.9	549.9	47.4	120.7	1203.1	34.8
BDE-66	3.5	23.1	< 1.5	2.9	5.3	< 1.7
BDE-100	374.6	544.2	67.5	352.6	1276.6	66.6
BDE-99	864.6	976.8	94.4	492.7	3413.4	92.7
BDE-85	6.3	12.4	< 1.5	2.8	51.6	< 1.7
BDE-154	815.0	616.0	212.2	1084.7	1429.1	106.2
BDE-153	694.5	1294.4	318.0	1753.8	5569.2	517.9
BDE-183	39.5	237.2	15.4	84.1	549.8	17.9
BDE-209	4.3	16.2	6.1	19.1	247.0	7.2
HBCD	3.20	2.60	< 1.20	2.70	1.60	< 1.00
Me-TBBP-A	2.00	24.00	0.40	0.10	0.90	1.70

Ring No.:	3050563				362867	3050501
Reg. No.:	02-1787-2	03-0542-1	03-0542-2	03-0543	01-1593	01-1608
Batch No.:	#03-14	#03-19	#03-19	#03-19	#02-17	#02-17
Year of sampling:	2002	2003	2003	2003	1986	1987
Place of sampling:	Qanisartut	Skyggesø	Skyggesø	Enoraq	Igaliko	Igaliko
Sample weight (g):	2.99	3.58	3.72	3.57	6.6544	5.9662
Fat content (%):	5.27	6.04	5.89	4.02	6.956	9.4145
Dry matter (% weight):	15.88	18.18	18.16	20.38	19.7007	28.9606
Substance	Concentration in ng/g wet weight					
BDE-17	< 0.09	< 0.35	< 0.34	< 0.35	< 0.05	0.14
BDE-28	< 0.09	< 0.35	< 0.34	< 0.35	0.05	0.12
BDE-49	0.09	< 0.35	< 0.34	0.71	0.20	0.55
BDE-47	1.88	5.37	5.40	23.50	1.96	14.81
BDE-66	< 0.09	< 0.35	< 0.34	< 0.35	0.10	0.59
BDE-100	3.28	6.75	7.12	54.79	2.93	7.10
BDE-99	5.70	18.38	17.82	89.66	3.17	12.47
BDE-85	< 0.10	0.31	0.32	1.07	< 0.05	nd
BDE-154	6.19	11.23	11.43	71.47	907.67	78.78
BDE-153	24.42	39.87	39.45	239.86	131.49	33.77
BDE-183	0.93	48.14	43.08	15.04	2.76	3.76
BDE-209	0.42	1.13	1.40	4.71	1.99	0.64
	Concentration in ng/g fat weight					
BDE-17	< 1.7	< 5.8	< 5.7	< 8.7	< 0.7	1.4
BDE-28	< 1.7	< 5.8	< 5.7	< 8.7	0.7	1.3
BDE-49	1.7	< 5.8	< 5.7	17.8	2.8	5.9
BDE-47	35.7	88.8	91.6	585.0	28.2	157.3
BDE-66	< 1.7	< 5.8	< 5.7	< 8.7	1.5	6.3
BDE-100	62.2	111.6	120.8	1364.2	42.1	75.4
BDE-99	108.1	304.1	302.4	2232.2	45.6	132.4
BDE-85	< 1.8	5.1	5.4	26.6	< 0.7	nd
BDE-154	117.4	185.8	194.0	1779.5	13048.7	836.8
BDE-153	463.4	659.7	669.3	5971.7	1890.3	358.7
BDE-183	17.7	796.4	730.9	374.4	39.7	39.9
BDE-209	8.0	18.6	23.7	117.3	28.5	6.8
HBCD		< 0.10		27.00	< 0.8	34.0
Me-TBBP-A		290.00		760.00	49.0	120.0

Ring No.:	3050543	3050541	3050107	3050541		3050142
Reg. No.:	01-1623	01-1619	01-1598	01-1621	01-1632	01-1603
Batch No.:	#02-17	#02-17	#02-17	#02-17	#02-17	#02-17
Year of sampling:	1990	1990	1991	1992	1994	1995
Place of sampling:	Igaliko	Skyggesø	Kirkeruin	Skyggesø	Havnen	Igaliko
Sample weight (g):	7.1024	6.7776	6.7367	7.0974	6.8119	6.9215
Fat content (%):	6.3047	6.3009	7.5179	4.8213	3.5919	6.5446
Dry matter (% weight):	22.8597	25.9584	27.5136	23.2656	21.0737	45.4613
Substance	Concentration in ng/g wet weight					
BDE-17	0.08	0.05	< 0.05	< 0.04	< 0.05	< 0.05
BDE-28	0.12	0.12	0.05	0.05	< 0.05	< 0.05
BDE-49	0.66	0.20	0.07	0.15	0.06	0.19
BDE-47	11.14	8.74	1.73	16.12	0.75	4.46
BDE-66	0.28	0.17	0.05	0.09	0.05	0.09
BDE-100	9.31	3.56	2.41	17.10	1.27	14.54
BDE-99	12.30	5.88	4.25	41.42	2.66	19.87
BDE-85	0.10	< 0.05	< 0.05	< 0.04	< 0.05	< 0.05
BDE-154	42.26	16.34	8.81	28.92	11.78	19.41
BDE-153	36.54	33.14	10.80	42.30	9.96	31.59
BDE-183	6.37	4.28	1.77	2.88	1.29	1.78
BDE-209	0.93	0.55	0.28	1.23	0.59	15.35
	Concentration in ng/g wet weight					
BDE-17	1.3	0.8	< 0.7	< 0.8	< 1.4	< 0.8
BDE-28	1.8	1.9	0.6	1.1	< 1.4	< 0.8
BDE-49	10.4	3.1	0.9	3.1	1.5	2.9
BDE-47	176.7	138.7	23.1	334.2	21.0	68.2
BDE-66	4.5	2.7	0.7	1.8	< 1.4	1.4
BDE-100	147.7	56.5	32.1	354.7	35.3	222.1
BDE-99	195.1	93.4	56.5	859.2	74.1	303.5
BDE-85	1.5	< 0.8	< 0.7	< 0.8	1.4	< 0.8
BDE-154	670.2	259.3	117.2	599.8	328.0	296.6
BDE-153	579.5	526.0	143.6	877.4	277.4	482.6
BDE-183	101.1	67.9	23.5	59.8	35.8	27.2
BDE-209	14.8	8.8	3.8	25.4	16.4	234.6
HBCD	9.0	4.1	< 1.1	< 1.1	2.1	< 8.0
Me-TBBP-A	160.0	12.0	15.0	750.0	270.0	440.0

Ring No.:	3050526	3050526	3050142	3050142	3050556	
Reg. No.:	01-1613	01-1614	01-1606-1	01-1606-2	01-1645	
Batch No.:	#02-17	#02-17	#02-17	#02-17	#02-17	
Year of sampling:	1998	1999	2000	2000	2001	
Place of sampling:	Upernaviarsuk	Upernaviarsuk	Igaliko	Igaliko	Egaluit	
Sample weight (g):	6.76	7.4561	7.5774	7.4622	7.022	
Fat content (%):	7.6171	3.8854	2.7293	2.8214	6.4711	
Dry matter (% weight):	33.1614	21.707	28.907	23.4434	24.5998	
Substance	Concentration in ng/g wet weight					
BDE-17	< 0.05	< 0.04	< 0.04	< 0.04	< 0.04	
BDE-28	< 0.05	< 0.04	< 0.04	< 0.04	< 0.04	
BDE-49	0.12	0.60	0.19	0.21	0.18	
BDE-47	1.93	8.60	2.11	2.16	2.73	
BDE-66	0.06	0.09	< 0.04	< 0.04	0.07	
BDE-100	3.77	13.95	6.37	5.88	10.89	
BDE-99	4.49	26.02	8.71	8.24	15.24	
BDE-85	< 0.05	0.63	< 0.04	0.12	0.07	
BDE-154	19.56	33.80	17.57	15.93	14.28	
BDE-153	27.90	38.73	44.09	43.18	33.20	
BDE-183	1.05	1.18	3.97	3.79	1.33	
BDE-209	0.46	0.43	0.88	1.11	0.74	
	Concentration in ng/g wet weight					
BDE-17	< 0.7	< 1.0	< 1.5	< 1.4	< 0.6	
BDE-28	< 0.7	< 1.0	< 1.5	< 1.4	< 0.6	
BDE-49	1.6	15.5	6.8	7.6	2.8	
BDE-47	25.3	221.4	77.2	76.5	42.1	
BDE-66	0.8	2.4	< 1.5	< 1.4	1.1	
BDE-100	49.5	359.0	233.3	208.4	168.4	
BDE-99	59.0	669.7	319.1	292.1	235.5	
BDE-85	< 0.7	16.1	< 1.5	4.3	1.1	
BDE-154	256.8	869.9	643.7	564.6	220.6	
BDE-153	366.3	996.7	1615.6	1530.6	513.0	
BDE-183	13.7	30.4	145.4	134.4	20.6	
BDE-209	6.0	11.0	32.4	39.2	11.4	
HBCD	< 8.0	< 9.0			< 0.8	
Me-TBBP-A	28.0	520.0			360.0	

Concentration levels for PCBs

Ring No.:		3050122	3050142		3050526	3050541
Reg. No.:	01-1597	01-1601	01-1607	01-1609	01-1616	01-1618
Batch No.:	#03-16	#03-16	#03-16	#03-16	#03-16	#03-16
Year of sampling:	1988	1992	2000	1988	2000	1990
Place of sampling:	Morten	Eqaluit	Sdr. Igaliko	Igaliko	Upernaviarsuk	Skyggesø
Sample weight (g):	3.04	3.34	3.56	3.52	3.05	3.54
Fat content (%):	20.31	9.31	3.58	22.42	4.63	n.a.
Dry matter (% weight):	38.73	18.25	13.34	45.16	17.54	21.09
Substance	Concentration in ng/g wet weight					
CB-28	11.92	21.88	4.66		5.11	11.35
CB-31	< 4.24	19.04	2.65		3.71	5.67
CB-44	< 3.71	3.67	2.65		1.06	2.28
CB-49	< 4.16	2.02	0.73		0.80	1.53
CB-52	< 4.10	1.20	0.36		0.49	1.95
CB-99	206.90	245.35	51.11		90.94	142.18
CB-101	8.96	14.73	5.52		6.92	14.51
CB-105	120.23	114.10	33.17		48.21	71.79
CB-110	< 3.16	7.53	1.25		2.42	4.75
CB-118	426.28	450.43	108.60		219.20	258.16
CB-128	171.47	133.06	36.19		60.70	78.23
CB-138	1038.38	801.40	227.15		487.59	658.61
CB-149	15.00	22.73	15.32		5.82	26.98
CB-151	< 3.43	1.61	0.67		< 0.44	1.43
CB-153	2650.19	1529.77	490.22		1240.08	1180.59
CB-156	186.84	75.49	28.07		67.47	56.99
CB-170	891.89	221.82	84.42		230.72	221.30
CB-180	3291.01	716.65	337.90		965.62	672.04
CB-187	773.75	721.59	128.31		258.25	421.23
CB-194	819.16	153.78	62.84		215.94	119.36
CB-209	68.50	87.60	14.47		16.59	59.73
	Concentration in ng/g fat weight					
CB-28	58.7	235.0	130.2		110.4	
CB-31	< 20.9	204.5	74.0		80.1	
CB-44	< 18.3	39.4	74.0		22.9	
CB-49	< 20.5	21.7	20.4		17.3	
CB-52	< 20.2	12.9	10.1		10.6	
CB-99	1018.7	2635.3	1427.7		1964.1	
CB-101	44.1	158.2	154.2		149.5	
CB-105	592.0	1225.6	926.5		1041.3	
CB-110	< 15.6	80.9	34.9		52.3	
CB-118	2098.9	4838.1	3033.5		4734.3	
CB-128	844.3	1429.2	1010.9		1311.0	
CB-138	5112.7	8607.9	6345.0		10531.1	
CB-149	73.9	244.1	427.9		125.7	
CB-151	< 16.9	17.3	18.7		< 9.5	
CB-153	13048.7	16431.4	13693.3		26783.6	
CB-156	919.9	810.9	783.9		1457.2	
CB-170	4391.4	2382.6	2358.2		4983.1	
CB-180	16203.9	7697.7	9438.5		20855.8	
CB-187	3809.7	7750.7	3584.1		5577.8	
CB-194	4033.3	1651.8	1755.2		4664.0	
CB-209	337.3	940.9	404.3		358.4	

Ring No.:	3050541	3050541				
Reg. No.:	01-1620	01-1622	01-1630	01-1631	01-1633	02-1789-1
Batch No.:	#03-16	#03-16	#03-16	#03-16	#03-16	#03-16
Year of sampling:	1992	1992	1994	1994	1994	1999
Place of sampling:	Skyggesø	Skyggesø	Havnen	Havnen	Havnen	Upernaviarsuk
Sample weight (g):	3.62	3.45	3.61	3.67	3.57	3.71
Fat content (%):	6.84	5.63	6.74	7.92	5.73	3.98
Dry matter (% weight):	17.74	15.58	17.87	20.85	15.54	16.04
Substance	Concentration in ng/g wet weight					
CB-28	22.07	17.47	1.94	2.39	1.81	2.78
CB-31	6.19	4.16	1.25	1.59	1.10	3.32
CB-44	6.09	1.17	0.66	0.69	0.53	0.72
CB-49	< 0.15	< 0.15	0.46	0.48	0.45	0.58
CB-52	0.38	0.32	0.28	0.30	0.30	0.41
CB-99	130.64	99.70	28.59	35.66	27.05	52.75
CB-101	2.43	1.43	4.78	5.06	4.57	6.36
CB-105	58.14	49.14	15.68	19.45	14.60	28.86
CB-110	2.51	2.30	0.71	0.83	0.82	1.57
CB-118	223.76	167.31	71.64	90.05	67.13	133.21
CB-128	73.31	58.94	19.33	24.55	18.13	39.19
CB-138	583.39	461.77	178.79	223.05	172.09	319.29
CB-149	5.26	3.33	2.39	2.59	2.33	3.97
CB-151	< 0.44	< 0.44	< 0.44	0.44	0.52	< 0.44
CB-153	1034.71	803.45	383.59	485.52	363.52	732.37
CB-156	57.10	42.42	21.52	27.30	20.60	48.06
CB-170	204.98	155.17	57.20	72.98	57.13	170.75
CB-180	721.51	583.60	239.94	296.93	236.55	618.76
CB-187	371.86	285.48	90.84	113.39	88.56	170.37
CB-194	149.59	104.86	43.44	52.50	42.76	164.24
CB-209	45.57	18.79	5.82	7.13	5.64	12.68
	Concentration in ng/g fat weight					
CB-28	322.7	310.3	28.8	30.2	31.6	69.8
CB-31	90.5	73.9	18.5	20.1	19.2	83.4
CB-44	89.0	20.8	9.8	8.7	9.2	18.1
CB-49	< 2.2	2.7	6.8	6.1	7.9	14.6
CB-52	5.6	5.7	4.2	3.8	5.2	10.3
CB-99	1909.9	1770.9	424.2	450.3	472.1	1325.4
CB-101	35.5	25.4	70.9	63.9	79.8	159.8
CB-105	850.0	872.8	232.6	245.6	254.8	725.1
CB-110	36.7	40.9	10.5	10.5	14.3	39.4
CB-118	3271.3	2971.8	1062.9	1137.0	1171.6	3347.0
CB-128	1071.8	1046.9	286.8	310.0	316.4	984.7
CB-138	8529.1	8202.0	2652.7	2816.3	3003.3	8022.4
CB-149	76.9	59.1	35.5	32.7	40.7	99.7
CB-151	< 6.4	< 7.8	< 6.5	5.6	9.1	< 11.1
CB-153	15127.3	14270.8	5691.3	6130.3	6344.2	18401.1
CB-156	834.8	753.4	319.3	344.7	359.6	1207.6
CB-170	2996.8	2756.0	848.7	921.5	997.1	4290.1
CB-180	10548.4	10366.0	3559.9	3749.2	4128.3	15546.7
CB-187	5436.6	5070.6	1347.8	1431.6	1545.6	4280.7
CB-194	2186.9	1862.5	644.5	662.8	746.2	4126.6
CB-209	666.2	333.7	86.3	90.0	98.5	318.6

Ring No.:		3050501	3050541	3050502		3050122
Reg. No.:	02-1789-2	01-1610	01-1617	01-1611	01-1629	01-1602
Batch No.:	#03-16	#03-14	#03-14	#03-14	#03-14	#03-14
Year of sampling:	1999	1988	1989	1991	1991	1994
Place of sampling:	Upernaviarsuk	Igaliko	Skyggesø	Bagerfalken	Hosp.dal	Eqaluit
Sample weight (g):	3.38	3.40	3.41	3.52	3.30	3.36
Fat content (%):	3.93	18.93	10.69	6.84	5.15	7.48
Dry matter (% weight):	16.27	35.29	22.42	18.97	17.45	22.36
Substance	Concentration in ng/g wet weight					
CB-28	2.66	112.44	7.92	5.29	11.18	23.93
CB-31	3.36	22.51	2.17	3.26	2.25	9.99
CB-44	0.63	23.61	1.84	2.31	6.09	5.15
CB-49	0.59	13.03	1.13	1.70	3.00	3.97
CB-52	0.40	8.64	0.98	1.36	2.55	2.50
CB-99	51.51	1247.54	39.28	59.46	215.40	311.27
CB-101	6.76	83.23	7.97	11.52	29.23	29.92
CB-105	27.53	437.24	22.86	30.71	101.68	152.48
CB-110	1.64	76.80	1.13	1.98	6.97	7.06
CB-118	126.30	1862.31	101.63	118.48	398.33	670.26
CB-128	39.02	379.10	21.73	36.86	146.91	190.12
CB-138	320.15	4671.33	144.97	276.18	1011.88	1280.32
CB-149	4.20	53.15	4.34	11.11	31.95	40.80
CB-151	0.54	7.26	0.47	1.50	3.88	4.33
CB-153	896.09	8906.17	415.03	600.34	1824.81	2323.24
CB-156	45.79	401.05	18.92	30.76	99.43	95.29
CB-170	164.47	1271.42	62.22	115.68	389.63	320.27
CB-180	734.79	5213.47	297.45	486.34	1390.69	1046.88
CB-187	161.85	1860.71	96.03	209.55	690.36	1122.27
CB-194	155.18	741.46	59.65	106.45	308.89	254.86
CB-209	12.31	60.31	11.99	20.57	41.85	122.01
	Concentration in ng/g fat weight					
CB-28	67.7	594.0	74.1	77.3	217.1	319.9
CB-31	85.5	118.9	20.3	47.7	43.7	133.6
CB-44	16.0	124.7	17.2	33.8	118.3	68.9
CB-49	15.0	68.8	10.6	24.9	58.3	53.1
CB-52	10.2	45.6	9.2	19.9	49.5	33.4
CB-99	1310.7	6590.3	367.4	869.3	4182.5	4161.4
CB-101	172.0	439.7	74.6	168.4	567.6	400.0
CB-105	700.5	2309.8	213.8	449.0	1974.4	2038.5
CB-110	41.7	405.7	10.6	28.9	135.3	94.4
CB-118	3213.7	9837.9	950.7	1732.2	7734.6	8960.7
CB-128	992.9	2002.6	203.3	538.9	2852.6	2541.7
CB-138	8146.3	24676.9	1356.1	4037.7	19648.2	17116.6
CB-149	106.9	280.8	40.6	162.4	620.4	545.5
CB-151	13.7	38.4	4.4	21.9	75.3	57.9
CB-153	22801.2	47047.9	3882.4	8776.9	35433.1	31059.4
CB-156	1165.0	2118.6	177.0	449.7	1930.8	1273.9
CB-170	4184.9	6716.4	582.0	1691.3	7565.6	4281.6
CB-180	18696.9	27540.8	2782.5	7110.3	27003.7	13995.7
CB-187	4118.4	9829.4	898.3	3063.5	13405.1	15003.7
CB-194	3948.6	3916.9	558.0	1556.2	5997.8	3407.3
CB-209	313.2	318.6	112.2	300.7	812.7	1631.1

Ring No.:	98764543	3050142		3050526	3050491	3050563
Reg. No.:	02-1785	01-1604	01-1626	01-1615	02-1788	02-1787-1
Batch No.:	#03-14	#03-14	#03-14	#03-14	#03-14	#03-14
Year of sampling:	1995	1998	1999	2000	2002	2002
Place of sampling:	Lejren	Sdr.Igaliko	Lejren	Upernaviarsuk	Enoraq	Qanisartut
Sample weight (g):	3.52	3.45	3.38	3.42	3.34	3.47
Fat content (%):	6.30	5.80	6.02	3.94	6.87	5.27
Dry matter (% weight):	18.96	34.20	17.26	13.65	20.05	15.91
Substance	Concentration in ng/g wet weight					
CB-28	13.57	17.29	4.14	5.74	15.80	3.66
CB-31	3.43	4.32	1.12	2.52	6.54	1.37
CB-44	4.45	12.20	1.39	2.85	6.12	0.91
CB-49	3.25	4.46	1.03	2.14	4.06	0.63
CB-52	2.36	2.91	1.02	1.58	2.67	0.64
CB-99	148.09	153.76	25.84	96.68	322.05	27.05
CB-101	20.90	20.42	7.78	20.85	39.23	4.46
CB-105	74.41	98.07	10.19	50.99	119.26	15.40
CB-110	4.20	4.73	0.67	2.67	6.32	0.56
CB-118	311.99	275.18	59.13	226.24	596.18	59.79
CB-128	72.27	77.40	14.92	59.25	188.32	15.74
CB-138	668.39	466.05	139.72	469.72	1596.11	107.34
CB-149	13.45	28.18	2.96	12.99	39.76	5.33
CB-151	1.58	3.02	0.43	1.83	5.39	0.48
CB-153	1370.01	842.24	437.72	1320.37	3217.90	244.13
CB-156	55.29	41.81	15.35	53.20	119.23	12.75
CB-170	177.41	154.96	63.35	223.69	512.60	43.67
CB-180	699.64	594.09	281.16	999.82	1920.62	144.53
CB-187	366.45	275.72	64.27	259.45	1462.55	75.74
CB-194	96.49	117.96	46.60	218.78	433.40	27.02
CB-209	11.18	50.11	6.28	19.26	163.96	16.85
	Concentration in ng/g fat weight					
CB-28	215.4	298.1	68.8	145.7	230.0	69.4
CB-31	54.4	74.5	18.6	64.0	95.2	26.0
CB-44	70.6	210.3	23.1	72.3	89.1	17.3
CB-49	51.6	76.9	17.1	54.3	59.1	12.0
CB-52	37.5	50.2	16.9	40.1	38.9	12.1
CB-99	2350.6	2651.0	429.2	2453.8	4687.8	513.3
CB-101	331.7	352.1	129.2	529.2	571.0	84.6
CB-105	1181.1	1690.9	169.3	1294.2	1736.0	292.2
CB-110	66.7	81.6	11.1	67.8	92.0	10.6
CB-118	4952.2	4744.5	982.2	5742.1	8678.0	1134.5
CB-128	1147.1	1334.5	247.8	1503.8	2741.2	298.7
CB-138	10609.4	8035.3	2320.9	11921.8	23233.0	2036.8
CB-149	213.5	485.9	49.2	329.7	578.7	101.1
CB-151	25.1	52.1	7.1	46.4	78.5	9.1
CB-153	21746.2	14521.5	7271.1	33511.8	46839.8	4632.5
CB-156	877.7	720.8	255.0	1350.2	1735.5	241.9
CB-170	2816.0	2671.6	1052.3	5677.4	7461.4	828.6
CB-180	11105.4	10242.9	4670.4	25376.2	27956.6	2742.5
CB-187	5816.6	4753.7	1067.6	6584.9	21289.0	1437.2
CB-194	1531.6	2033.8	774.2	5552.7	6308.5	512.8
CB-209	177.5	864.0	104.4	488.9	2386.6	319.7

Ring No.:	3050563				362867	3050501
Reg. No.:	02-1787-2	03-0542-1	03-0542-2	03-0543	01-1593	01-1608
Batch No.:	#03-14	#03-19	#03-19	#03-19	#02-17	#02-17
Year of sampling:	2002	2003	2003	2003	1986	1987
Place of sampling:	Qanisartut	Skyggesø	Skyggesø	Enoraq	Igaliko	Igaliko
Sample weight (g):	2.99	3.58	3.72	3.57	6.6544	5.9662
Fat content (%):	5.27	6.04	5.89	4.02	6.956	9.4145
Dry matter (% weight):	15.88	18.18	18.16	20.38	19.7007	28.9606
Substance	Concentration in ng/g wet weight					
CB-28	3.61	10.06	9.43	20.11	8.10	65.31
CB-31	1.33	0.87	0.76	7.65	7.44	22.23
CB-44	0.94	0.31	0.30	3.47	3.19	12.70
CB-49	0.57	0.36	0.33	1.26	0.83	6.66
CB-52	0.46	0.21	0.20	0.57	1.47	3.74
CB-99	26.32	35.19	33.66	241.73	161.92	714.85
CB-101	4.72	2.38	2.18	10.41	3.38	12.25
CB-105	17.43	20.90	19.80	118.63	80.96	247.21
CB-110	0.70	1.12	1.07	3.45	1.63	11.38
CB-118	61.60	78.39	74.66	487.25	349.39	1055.75
CB-128	16.58	23.82	22.09	157.90	102.66	223.13
CB-138	107.26	186.84	179.04	1012.17	825.34	2541.65
CB-149	5.94	2.76	2.59	27.90	6.80	15.28
CB-151	1.00	< 0.38	< 0.38	1.41	0.21	1.21
CB-153	243.43	480.45	466.61	2261.29	1565.26	4741.97
CB-156	13.74	19.27	18.27	99.01	86.08	235.78
CB-170	41.54	78.58	73.23	406.55	331.51	743.17
CB-180	144.62	294.91	281.33	1251.86	1246.50	3050.56
CB-187	74.10	94.50	89.11	971.03	493.37	1097.38
CB-194	25.42	50.07	46.14	321.40	233.15	428.72
CB-209	15.56	8.90	8.31	112.86	41.89	28.39
	Concentration in ng/g fat weight					
CB-28	68.5	166.4	160.0	500.7	116.4	693.7
CB-31	25.2	14.4	12.9	190.5	107.0	236.1
CB-44	17.8	5.1	< 5.1	86.4	45.9	134.9
CB-49	10.8	6.0	5.6	31.4	11.9	70.7
CB-52	8.7	3.5	3.4	14.2	21.1	39.7
CB-99	499.4	582.2	571.1	6018.3	2327.8	7593.1
CB-101	89.6	39.4	37.0	259.2	48.6	130.1
CB-105	330.7	345.8	335.9	2953.5	1163.9	2625.8
CB-110	13.3	18.5	18.2	85.9	23.4	120.9
CB-118	1168.9	1296.9	1266.7	12130.9	5022.9	11214.1
CB-128	314.6	394.1	374.8	3931.2	1475.8	2370.1
CB-138	2035.3	3091.2	3037.6	25199.7	11865.2	26997.2
CB-149	112.7	45.7	43.9	694.6	97.7	162.3
CB-151	19.0	< 6.3	< 6.4	35.1	3.0	12.9
CB-153	4619.1	7948.9	7916.5	56298.7	22502.3	50368.8
CB-156	260.8	318.9	310.0	2465.1	1237.5	2504.4
CB-170	788.2	1300.1	1242.5	10121.8	4765.9	7893.8
CB-180	2744.1	4879.2	4773.1	31167.2	17919.7	32402.8
CB-187	1406.2	1563.4	1511.8	24175.3	7092.8	11656.3
CB-194	482.3	828.3	782.9	8001.9	3351.7	4553.8
CB-209	295.3	147.3	140.9	2809.9	602.2	301.6

Ring No.:	3050543	3050541	3050107	3050541		3050142
Reg. No.:	01-1623	01-1619	01-1598	01-1621	01-1632	01-1603
Batch No.:	#02-17	#02-17	#02-17	#02-17	#02-17	#02-17
Year of sampling:	1990	1990	1991	1992	1994	1995
Place of sampling:	Igaliko	Skyggesø	Kirkeruin	Skyggesø	Havnen	Igaliko
Sample weight (g):	7.1024	6.7776	6.7367	7.0974	6.8119	6.9215
Fat content (%):	6.3047	6.3009	7.5179	4.8213	3.5919	6.5446
Dry matter (% weight):	22.8597	25.9584	27.5136	23.2656	21.0737	45.4613
Substance	Concentration in ng/g wet weight					
CB-28	33.61	9.51	2.35	22.61	2.34	4.00
CB-31	6.14	2.77	2.70	3.13	0.98	1.54
CB-44	9.63	1.50	0.93	5.32	0.57	1.05
CB-49	4.40	0.95	0.42	< 0.08	0.08	0.44
CB-52	2.23	1.90	0.66	0.78	0.46	0.42
CB-99	250.77	129.74	47.29	141.82	26.00	94.36
CB-101	13.01	5.22	1.76	1.07	0.72	1.30
CB-105	127.89	62.40	16.55	61.28	13.79	48.16
CB-110	4.69	3.40	0.93	1.26	0.46	0.95
CB-118	380.79	207.87	89.19	209.80	68.89	201.91
CB-128	142.59	68.33	22.48	76.31	17.14	61.53
CB-138	901.37	643.08	210.75	530.54	143.19	387.77
CB-149	85.22	20.07	2.04	4.67	0.83	3.75
CB-151	0.99	26.35	0.13	0.26	1.67	4.86
CB-153	1585.64	1218.06	432.62	1042.52	360.34	882.91
CB-156	72.50	45.89	22.42	49.57	17.22	45.10
CB-170	292.01	190.63	79.58	189.72	59.08	144.13
CB-180	951.67	769.57	307.62	769.71	232.75	506.73
CB-187	610.96	449.46	145.78	373.13	91.77	241.78
CB-194	148.67	116.01	51.05	154.62	45.79	84.57
CB-209	41.08	59.38	9.51	48.15	6.08	18.51
	Concentration in ng/g fat weight					
CB-28	533.1	150.9	31.3	469.0	65.1	61.1
CB-31	97.4	44.0	35.9	64.9	27.3	23.5
CB-44	152.7	23.8	12.4	110.3	15.9	16.0
CB-49	69.8	15.1	5.6	1.7	< 2.2	6.7
CB-52	35.4	30.2	8.8	16.2	12.8	6.4
CB-99	3977.5	2059.1	629.0	2941.5	723.9	1441.8
CB-101	206.4	82.8	23.4	22.2	20.0	19.9
CB-105	2028.5	990.3	220.1	1271.0	383.9	735.9
CB-110	74.4	54.0	12.4	26.1	12.8	14.5
CB-118	6039.8	3299.1	1186.4	4351.5	1917.9	3085.1
CB-128	2261.6	1084.4	299.0	1582.8	477.2	940.2
CB-138	14296.8	10206.2	2803.3	11004.1	3986.5	5925.0
CB-149	1351.7	318.5	27.1	96.9	23.1	57.3
CB-151	15.7	418.2	1.8	5.4	46.5	74.2
CB-153	25150.1	19331.5	5754.5	21623.2	10032.0	13490.6
CB-156	1150.0	728.3	298.2	1028.1	479.5	689.1
CB-170	4631.5	3025.5	1058.5	3935.0	1644.8	2202.3
CB-180	15094.6	12213.7	4091.9	15964.8	6479.9	7742.7
CB-187	9690.6	7133.3	1939.0	7739.3	2554.8	3694.3
CB-194	2358.0	1841.1	679.1	3207.0	1274.9	1292.2
CB-209	651.5	942.5	126.5	998.6	169.3	282.9

Ring No.:	3050526	3050526	3050142	3050142	3050556	
Reg. No.:	01-1613	01-1614	01-1606-1	01-1606-2	01-1645	
Batch No.:	#02-17	#02-17	#02-17	#02-17	#02-17	
Year of sampling:	1998	1999	2000	2000	2001	
Place of sampling:	Upernaviarsuk	Upernaviarsuk	Igaliko	Igaliko	Egaluit	
Sample weight (g):	6.76	7.4561	7.5774	7.4622	7.022	
Fat content (%):	7.6171	3.8854	2.7293	2.8214	6.4711	
Dry matter (% weight):	33.1614	21.707	28.907	23.4434	24.5998	
Substance	Concentration in ng/g wet weight					
CB-28	2.25	3.27	4.65	4.05	11.49	
CB-31	1.12	2.36	1.58	1.32	2.45	
CB-44	0.52	0.55	2.09	1.96	2.16	
CB-49	< 0.08	< 0.08	< 0.08	0.35	< 0.08	
CB-52	0.36	0.36	0.37	0.36	0.40	
CB-99	35.41	66.16	40.20	40.04	154.87	
CB-101	0.68	0.96	1.29	0.64	2.16	
CB-105	16.91	26.44	26.74	25.40	60.79	
CB-110	0.78	0.84	0.50	0.30	1.19	
CB-118	87.45	141.60	94.11	91.21	273.25	
CB-128	23.69	41.72	28.88	27.96	68.08	
CB-138	184.01	305.81	161.57	158.55	423.01	
CB-149	1.16	1.95	10.65	9.86	8.60	
CB-151	4.27	0.27	1.88	2.23	0.59	
CB-153	501.79	881.58	387.26	378.27	853.21	
CB-156	24.99	42.47	20.04	19.11	38.10	
CB-170	105.95	183.30	71.94	70.90	109.53	
CB-180	417.64	725.12	271.99	271.70	337.02	
CB-187	111.03	175.82	110.83	107.52	266.25	
CB-194	108.81	166.90	54.95	54.05	59.90	
CB-209	11.41	15.28	13.52	13.45	20.22	
	Concentration in ng/g fat weight					
CB-28	29.5	84.2	170.4	143.5	177.6	
CB-31	14.7	60.7	57.9	46.8	37.9	
CB-44	6.8	14.2	76.6	69.5	33.4	
CB-49	< 1.1	< 2.1	2.9	12.4	< 1.2	
CB-52	4.7	9.3	13.6	12.8	6.2	
CB-99	464.9	1702.8	1472.9	1419.2	2393.3	
CB-101	8.9	24.7	47.3	22.7	33.4	
CB-105	222.0	680.5	979.7	900.3	939.4	
CB-110	10.2	21.6	18.3	10.6	18.4	
CB-118	1148.1	3644.4	3448.1	3232.8	4222.6	
CB-128	311.0	1073.8	1058.1	991.0	1052.1	
CB-138	2415.7	7870.7	5919.8	5619.6	6536.9	
CB-149	15.2	50.2	390.4	349.4	132.9	
CB-151	56.0	6.9	68.8	78.9	9.1	
CB-153	6587.7	22689.6	14189.1	13407.2	13184.9	
CB-156	328.1	1093.1	734.3	677.2	588.8	
CB-170	1391.0	4717.6	2635.7	2513.1	1692.6	
CB-180	5483.0	18662.8	9965.4	9629.9	5208.0	
CB-187	1457.7	4525.3	4060.6	3811.0	4114.4	
CB-194	1428.5	4295.5	2013.3	1915.8	925.7	
CB-209	149.7	393.2	495.5	476.9	312.4	

DDT and degradation products. Toxaphene. Chlordane-related compounds and Hexachlorobenzene

Ring No.:		3050122	3050142		3050526	3050541
Reg. No.:	01-1597	01-1601	01-1607	01-1609	01-1616	01-1618
Batch No.:	#03-16	#03-16	#03-16	#03-16	#03-16	#03-16
Year of sampling:	1988	1992	2000	1988	2000	1990
Place of sampling:	Morten	Eqaluit	Sdr. Igaliko	Igaliko	Upernaviarsuk	Skyggesø
Sample weight (g):	3.04	3.34	3.56	3.52	3.05	3.54
Fat content (%):	20.31	9.31	3.58	22.42	4.63	n.a.
Dry matter (% weight):	38.73	18.25	13.34	45.16	17.54	21.09
Substance	Concentration in ng/g wet weight					
alfa-HCH	n.a.	2.80	0.91		1.25	2.10
beta-HCH	n.a.	11.48	9.50		8.13	14.70
gamma-HCH	n.a.	1.53	0.34		0.51	0.81
HCB	66.77	37.05	28.08		43.98	424.99
o'p-DDE	n.a.	99.92	19.25		42.07	45.06
o'p-DDT	< 4.15	17.29	< 0.42		< 0.42	< 0.42
p'p'-DDD	16.44	11.72	4.55		2.08	16.43
p'p-DDE	9119.00	6753.89	1355.48		2154.78	3531.04
p'p-DDT	6.57	70.91	17.39		44.66	56.13
CHB-26	53.21	20.50	3.93	345.09	32.18	40.17
CHB-40	9.44	1.48	2.39	79.15	6.28	10.85
CHB-41	6.78	6.11	1.30	28.16	7.32	6.45
CHB-44	39.45	24.68	3.26	133.61	22.17	25.31
CHB-50	83.42	34.59	3.93	451.98	50.54	56.18
CHB-62	< 2.05	4.98	< 1.76	22.63	13.64	4.54
oxychlordan	262.37	259.44	42.73	1193.27	90.51	105.27
trans-chlordan	0.29	0.31	< 0.09	3.06	0.12	0.16
cis-chlordan	1.53	0.58	0.18	4.09	0.36	0.94
trans-nonachlor	106.84	153.74	19.84	704.40	69.20	76.14
cis-nonachlor	55.27	111.39	15.95	362.22	22.85	51.03
	Concentration in ng/g fat weight					
alfa-HCH		30.0	25.4		27.0	
beta-HCH		123.3	265.3		175.5	
gamma-HCH		16.4	9.6		11.1	
HCB	328.8	398.0	784.2		949.9	
o'p-DDE		1073.2	537.8		908.5	
o'p-DDT	< 20.4	185.8	< 11.7		< 9.1	
p'p'-DDD	80.9	125.9	127.0		44.8	
p'p-DDE	44899.1	72544.5	37862.5		46539.5	
p'p-DDT	32.3	761.7	485.7		964.5	
CHB-26	262.0	220.2	109.7	1539.2	694.9	
CHB-40	46.5	15.9	66.8	353.0	135.6	
CHB-41	33.4	65.6	36.4	125.6	158.1	
CHB-44	194.2	265.0	91.1	595.9	478.9	
CHB-50	410.7	371.5	109.8	2016.0	1091.7	
CHB-62	< 10.1	53.5	< 49.1	101.0	294.7	
oxychlordan	1291.8	2786.7	1193.5	5322.4	1954.9	
trans-chlordan	1.4	3.3	< 2.5	13.6	2.6	
cis-chlordan	7.5	6.2	5.1	18.2	7.7	
trans-nonachlor	526.0	1651.4	554.2	3141.8	1494.5	
cis-nonachlor	272.1	1196.4	445.4	1615.6	493.6	

Ring No.:	3050541	3050541				
Reg. No.:	01-1620	01-1622	01-1630	01-1631	01-1633	02-1789-1
Batch No.:	#03-16	#03-16	#03-16	#03-16	#03-16	#03-16
Year of sampling:	1992	1992	1994	1994	1994	1999
Place of sampling:	Skyggesø	Skyggesø	Havnen	Havnen	Havnen	Upernaviarsuk
Sample weight (g):	3.62	3.45	3.61	3.67	3.57	3.71
Fat content (%):	6.84	5.63	6.74	7.92	5.73	3.98
Dry matter (% weight):	17.74	15.58	17.87	20.85	15.54	16.04
Substance	Concentration in ng/g wet weight					
alfa-HCH	1.41	1.13	0.87	0.90	1.01	0.76
beta-HCH	8.69	6.44	3.86	3.26	2.70	2.53
gamma-HCH	0.76	0.63	0.81	0.92	0.55	0.38
HCB	34.95	27.66	15.37	19.34	13.66	17.56
o'p-DDE	65.47	51.47	11.38	13.82	10.49	33.91
o'p-DDT	< 0.42	< 0.42	< 0.42	< 0.42	< 0.42	< 0.42
p'p'-DDD	6.06	8.08	2.93	6.28	3.14	< 0.21
p'p-DDE	2507.37	2062.07	771.31	1012.89	779.15	1014.48
p'p-DDT	42.55	12.97	7.27	9.48	6.39	21.33
CHB-26	33.82	24.37	3.16	3.14	1.06	12.36
CHB-40	4.72	3.97	1.95	2.51	1.30	2.91
CHB-41	6.97	5.56	0.68	0.64	0.16	4.45
CHB-44	28.55	22.81	2.15	2.08	1.71	12.06
CHB-50	53.92	37.61	1.19	0.60	0.26	28.62
CHB-62	11.23	2.37	< 1.73	< 1.70	< 1.75	7.42
oxychlordan	93.12	69.45	45.59	56.22	40.80	60.39
trans-chlordan	0.16	0.13	< 0.09	< 0.09	< 0.09	< 0.09
cis-chlordan	0.60	0.38	< 0.09	< 0.09	< 0.09	0.15
trans-nonachlor	79.13	61.14	7.25	8.88	6.33	59.58
cis-nonachlor	32.73	25.06	2.83	3.20	2.57	17.12
	Concentration in ng/g fat weight					
alfa-HCH	20.6	20.0	12.9	11.4	17.6	19.0
beta-HCH	127.0	114.4	57.3	41.2	47.2	63.7
gamma-HCH	11.1	11.2	12.0	11.6	9.7	9.6
HCB	511.0	491.2	228.0	244.2	238.3	441.2
o'p-DDE	957.2	914.1	168.8	174.5	183.1	852.1
o'p-DDT	< 6.1	< 7.5	< 6.2	< 5.3	< 7.3	< 10.6
p'p'-DDD	88.7	143.6	43.5	79.3	54.8	< 5.3
p'p-DDE	36657.4	36626.4	11443.8	12789.0	13597.8	25489.3
p'p-DDT	622.1	230.4	107.9	119.7	111.5	535.9
CHB-26	494.5	432.8	46.9	39.6	18.5	310.6
CHB-40	69.0	70.6	28.9	31.7	22.7	73.0
CHB-41	101.9	98.7	10.1	8.0	2.8	111.8
CHB-44	417.4	405.2	31.9	26.2	29.9	303.0
CHB-50	788.3	668.0	17.7	7.6	4.6	719.1
CHB-62	164.2	42.1	< 25.7	21.5	< 30.6	186.3
oxychlordan	1361.4	1233.5	676.5	709.8	712.0	1517.3
trans-chlordan	2.4	2.4	< 1.3	< 1.1	< 1.6	< 2.2
cis-chlordan	8.8	6.8	< 1.3	< 1.1	< 1.5	3.7
trans-nonachlor	1156.9	1086.0	107.6	112.1	110.5	1497.0
cis-nonachlor	478.5	445.2	42.0	40.4	44.9	430.1

Ring No.:		3050501	3050541	3050502		3050122
Reg. No.:	02-1789-2	01-1610	01-1617	01-1611	01-1629	01-1602
Batch No.:	#03-16	#03-14	#03-14	#03-14	#03-14	#03-14
Year of sampling:	1999	1988	1989	1991	1991	1994
Place of sampling:	Upernaviarsuk	Igaliko	Skyggesø	Bagerfalken	Hosp.dal	Eqaluit
Sample weight (g):	3.38	3.40	3.41	3.52	3.30	3.36
Fat content (%):	3.93	18.93	10.69	6.84	5.15	7.48
Dry matter (% weight):	16.27	35.29	22.42	18.97	17.45	22.36
Substance	Concentration in ng/g wet weight					
alfa-HCH	0.78	12.40	2.60	2.13	3.41	4.05
beta-HCH	2.27	56.91	14.80	6.63	4.18	6.03
gamma-HCH	0.36	21.27	1.09	7.69	5.54	0.79
HCB	17.23	742.14	17.70	12.61	23.93	26.76
o'p-DDE	34.03	306.08	17.28	34.40	27.87	98.17
o'p-DDT	< 0.42	21.38	< 0.21	1.55	1.94	2.68
p'p'-DDD	< 0.21	10.89	4.92	10.71	11.93	24.26
p'p-DDE	1224.05	6004.10	2408.25	3180.37	4369.87	8518.84
p'p-DDT	20.74	120.43	12.59	76.20	54.24	97.72
CHB-26	12.84	278.10	8.39	34.01	15.87	12.97
CHB-40	2.81	59.25	3.79	19.41	5.36	1.46
CHB-41	4.60	23.76	3.17	13.65	3.95	4.14
CHB-44	11.83	85.05	9.08	31.19	11.39	20.26
CHB-50	28.02	372.52	13.40	51.11	25.44	18.68
CHB-62	7.28	28.45	< 1.83	9.24	3.22	3.00
oxychlordan	59.62	1030.09	52.68	59.74	69.76	214.46
trans-chlordan	< 0.10	2.52	0.51	0.08	0.13	0.19
cis-chlordan	0.15	2.89	0.57	0.76	0.63	0.49
trans-nonachlor	58.36	610.70	15.02	42.79	33.52	94.68
cis-nonachlor	17.45	325.14	7.68	30.07	21.06	61.78
	Concentration in ng/g fat weight					
alfa-HCH	20.0	65.5	24.3	31.1	66.2	54.1
beta-HCH	57.7	300.6	138.5	97.0	81.1	80.7
gamma-HCH	9.2	112.3	10.2	112.4	107.6	10.6
HCB	438.5	3920.5	165.5	184.4	464.7	357.8
o'p-DDE	866.0	1616.9	161.7	502.9	541.2	1312.4
o'p-DDT	< 10.7	113.0	< 2.0	22.7	37.6	35.9
p'p'-DDD	< 5.3	57.5	46.0	156.6	231.7	324.3
p'p-DDE	31146.2	31717.4	22528.1	46496.6	84851.9	113888.2
p'p-DDT	527.8	636.2	117.7	1114.0	1053.3	1306.4
CHB-26	326.7	1469.1	78.5	497.3	308.2	173.3
CHB-40	71.4	313.0	35.4	283.7	104.0	19.5
CHB-41	117.0	125.5	29.7	199.6	76.7	55.4
CHB-44	300.9	449.3	84.9	455.9	221.3	270.9
CHB-50	713.0	1967.9	125.4	747.2	494.0	249.7
CHB-62	185.2	150.3	< 17.1	135.1	62.5	40.2
oxychlordan	1517.0	5441.6	492.8	873.4	1354.5	2867.1
trans-chlordan	< 2.4	13.3	4.8	1.1	2.6	2.6
cis-chlordan	3.8	15.2	5.3	11.2	12.2	6.6
trans-nonachlor	1485.0	3226.1	140.5	625.6	650.9	1265.8
cis-nonachlor	444.1	1717.6	71.8	439.6	409.0	826.0

Ring No.:	98764543	3050142		3050526	3050491	3050563
Reg. No.:	02-1785	01-1604	01-1626	01-1615	02-1788	02-1787-1
Batch No.:	#03-14	#03-14	#03-14	#03-14	#03-14	#03-14
Year of sampling:	1995	1998	1999	2000	2002	2002
Place of sampling:	Lejren	Sdr.Igaliko	Lejren	Upernaviarsuk	Enoraq	Qanisartut
Sample weight (g):	3.52	3.45	3.38	3.42	3.34	3.47
Fat content (%):	6.30	5.80	6.02	3.94	6.87	5.27
Dry matter (% weight):	18.96	34.20	17.26	13.65	20.05	15.91
Substance	Concentration in ng/g wet weight					
alfa-HCH	2.54	3.72	1.26	1.79	2.65	0.88
beta-HCH	8.61	7.95	3.04	5.94	2.11	3.42
gamma-HCH	0.58	0.47	0.25	0.65	1.16	0.80
HCB	43.46	94.21	14.67	46.84	22.49	15.83
o'p-DDE	44.67	54.04	10.20	39.17	60.80	10.44
o'p-DDT	2.36	0.50	0.21	1.08	0.91	1.33
p'p'-DDD	15.23	60.73	78.06	5.10	7.08	6.32
p'p-DDE	4858.12	2391.55	1219.15	2332.16	4413.67	9077.83
p'p-DDT	27.58	9.14	3.77	20.87	17.17	24.76
CHB-26	8.02	0.83	0.87	24.05	13.87	31.35
CHB-40	3.52	2.11	1.18	6.52	1.88	5.74
CHB-41	2.49	n.a.	0.13	5.82	5.48	10.06
CHB-44	7.02	3.13	0.77	17.00	23.70	18.02
CHB-50	10.70	< 0.18	< 0.19	38.32	21.68	50.23
CHB-62	< 1.77	< 1.81	< 1.85	4.89	< 1.87	2.73
oxychlordan	216.03	123.80	56.83	90.45	165.68	27.82
trans-chlordan	0.19	0.50	0.04	0.10	0.11	0.04
cis-chlordan	0.22	5.95	0.04	0.21	0.36	0.09
trans-nonachlor	101.49	113.45	15.62	64.35	96.12	24.04
cis-nonachlor	56.10	97.38	4.67	19.94	45.44	11.68
	Concentration in ng/g fat weight					
alfa-HCH	40.3	64.1	20.9	45.5	38.5	16.7
beta-HCH	136.6	137.0	50.5	150.7	30.7	64.8
gamma-HCH	9.2	8.1	4.1	16.5	16.9	15.1
HCB	689.9	1624.3	243.7	1188.9	327.4	300.3
o'p-DDE	709.1	931.8	169.5	994.1	885.0	198.2
o'p-DDT	37.5	8.6	< 3.5	27.5	13.2	25.2
p'p'-DDD	241.8	1047.0	1296.7	129.4	103.0	119.8
p'p-DDE	77113.0	41233.6	20251.6	59191.9	64245.5	172254.7
p'p-DDT	437.8	157.5	62.6	529.8	249.9	469.9
CHB-26	127.2	14.4	14.5	610.4	201.9	594.9
CHB-40	55.9	36.4	19.6	165.5	27.4	108.8
CHB-41	39.5		2.1	147.7	79.8	190.8
CHB-44	111.4	54.0	12.8	431.6	345.0	342.0
CHB-50	169.8	< 3.1	< 3.1	972.5	315.6	953.2
CHB-62	< 28.2	< 31.2	< 30.8	124.1	< 27.2	51.7
oxychlordan	3429.1	2134.5	944.1	2295.7	2411.7	528.0
trans-chlordan	3.0	8.7	0.6	2.4	1.6	< 0.7
cis-chlordan	3.5	102.6	0.6	5.4	5.3	1.8
trans-nonachlor	1610.9	1956.0	259.5	1633.2	1399.1	456.2
cis-nonachlor	890.5	1678.9	77.5	506.0	661.5	221.7

Ring No.:	3050563				362867	3050501
Reg. No.:	02-1787-2	03-0542-1	03-0542-2	03-0543	01-1593	01-1608
Batch No.:	#03-14	#03-19	#03-19	#03-19	#02-17	#02-17
Year of sampling:	2002	2003	2003	2003	1986	1987
Place of sampling:	Qanisartut	Skyggesø	Skyggesø	Enoraq	Igaliko	Igaliko
Sample weight (g):	2.99	3.58	3.72	3.57	6.6544	5.9662
Fat content (%):	5.27	6.04	5.89	4.02	6.956	9.4145
Dry matter (% weight):	15.88	18.18	18.16	20.38	19.7007	28.9606
Substance	Concentration in ng/g wet weight					
alfa-HCH	0.67	0.33	0.28	0.64	2.48	7.25
beta-HCH	4.11	1.08	1.63	4.36	20.59	41.76
gamma-HCH	0.72	41.56	38.91	1.09	0.44	6.60
HCB	15.24	11.52	10.62	26.64	26.15	460.79
o'p-DDE	9.50	8.05	7.66	1.34	95.52	172.92
o'p-DDT	0.50	n.a.	n.a.	n.a.	1.29	12.69
p'p'-DDD	6.78	0.76	0.78	6.52	644.06	11.01
p'p-DDE	9086.29	1016.64	912.76	3518.54	7095.20	3452.92
p'p-DDT	23.45	13.21	12.90	32.84	65.30	35.03
CHB-26	30.71	2.72	2.59	13.55	0.86	163.53
CHB-40	5.77	1.32	1.22	1.03	1.65	30.17
CHB-41	9.70	1.36	1.26	3.60	0.27	13.67
CHB-44	17.95	2.76	2.47	25.47	< 0.09	43.16
CHB-50	50.93	4.27	3.98	15.10	< 0.09	245.97
CHB-62	2.77	2.07	< 1.68	2.63	< 0.47	< 0.52
oxychlordan	26.32	33.87	31.77	176.13	228.50	592.50
trans-chlordan	< 0.04	0.04	0.02	0.22	0.86	1.48
cis-chlordan	0.11	< 0.09	< 0.08	n.a.	1.28	1.91
trans-nonachlor	23.57	10.15	9.72	137.70	112.63	357.64
cis-nonachlor	11.23	5.33	4.93	76.54	53.37	183.87
	Concentration in ng/g fat weight					
alfa-HCH	12.7	5.4	4.8	15.9	35.6	77.0
beta-HCH	78.1	17.8	27.6	108.5	296.0	443.6
gamma-HCH	13.6	687.6	660.2	27.2	6.4	70.1
HCB	289.1	190.7	180.1	663.3	376.0	4894.5
o'p-DDE	180.2	133.2	130.0	33.4	1373.2	1836.7
o'p-DDT	9.5				18.5	134.8
p'p'-DDD	128.7	12.6	13.3	162.2	9259.0	117.0
p'p-DDE	172415.4	16820.1	15486.0	87600.1	102001.2	36676.6
p'p-DDT	445.1	218.5	218.8	817.6	938.8	372.1
CHB-26	582.8	45.0	43.9	337.4	12.4	1737.0
CHB-40	109.6	21.9	20.8	25.6	23.7	320.5
CHB-41	184.1	22.6	21.5	89.7	3.8	145.2
CHB-44	340.6	45.7	41.8	634.2	< 1.3	458.4
CHB-50	966.4	70.7	67.6	375.9	< 1.3	2612.7
CHB-62	52.5	34.2	< 28.5	65.6	< 6.8	< 5.5
oxychlordan	499.4	560.5	539.1	4385.1	3284.9	6293.5
trans-chlordan	< 0.8	0.7	0.3	5.4	12.4	15.8
cis-chlordan	2.1	< 1.4	< 1.4		18.5	20.3
trans-nonachlor	447.3	167.9	164.8	3428.4	1619.2	3798.8
cis-nonachlor	213.0	88.2	83.7	1905.7	767.2	1953.0

Ring No.:	3050543	3050541	3050107	3050541		3050142
Reg. No.:	01-1623	01-1619	01-1598	01-1621	01-1632	01-1603
Batch No.:	#02-17	#02-17	#02-17	#02-17	#02-17	#02-17
Year of sampling:	1990	1990	1991	1992	1994	1995
Place of sampling:	Igaliko	Skyggesø	Kirkeruin	Skyggesø	Havnen	Igaliko
Sample weight (g):	7.1024	6.7776	6.7367	7.0974	6.8119	6.9215
Fat content (%):	6.3047	6.3009	7.5179	4.8213	3.5919	6.5446
Dry matter (% weight):	22.8597	25.9584	27.5136	23.2656	21.0737	45.4613
Substance	Concentration in ng/g wet weight					
alfa-HCH	4.95	1.51	1.10	1.16	0.78	4.58
beta-HCH	17.29	17.16	6.58	11.21	4.35	8.56
gamma-HCH	2.45	0.67	0.70	0.68	0.75	1.28
HCB	207.33	444.61	13.04	33.69	12.78	17.74
o'p-DDE	78.76	38.31	42.95	62.64	9.55	36.53
o'p-DDT	2.48	1.21	0.98	1.02	< 0.11	1.29
p'p'-DDD	60.15	8.39	12.73	13.89	4.08	2.69
p'p-DDE	5660.37	3473.51	1886.47	2496.44	754.20	1944.52
p'p-DDT	20.65	44.37	21.78	12.41	2.26	10.75
CHB-26	8.27	31.13	10.40	29.47	2.12	6.51
CHB-40	13.78	7.40	2.30	4.76	1.92	2.23
CHB-41	3.08	4.52	1.84	6.38	0.42	3.32
CHB-44	8.73	10.92	6.38	24.62	1.11	4.14
CHB-50	1.89	4.84	15.52	47.36	0.28	10.21
CHB-62	< 0.44	6.69	2.34	1.77	0.46	1.02
oxychlordan	157.90	79.14	213.53	79.38	36.33	115.33
trans-chlordan	0.65	< 0.19	< 0.19	< 0.18	< 0.19	< 0.19
cis-chlordan	4.20	0.99	0.82	0.66	0.06	0.38
trans-nonachlor	103.79	61.18	72.79	78.43	6.34	36.75
cis-nonachlor	93.07	41.28	25.07	31.37	2.47	18.87
	Concentration in ng/g fat weight					
alfa-HCH	78.5	23.9	14.6	24.1	21.6	70.0
beta-HCH	274.2	272.4	87.5	232.4	121.0	130.9
gamma-HCH	38.9	10.7	9.4	14.0	20.8	19.5
HCB	3288.4	7056.3	173.4	698.8	355.9	271.0
o'p-DDE	1249.2	608.0	571.3	1299.3	265.8	558.2
o'p-DDT	39.3	19.2	13.0	21.1	3.1	19.7
p'p'-DDD	954.1	133.1	169.4	288.0	113.6	41.0
p'p-DDE	89780.2	55127.2	25093.0	51779.4	20997.3	29711.8
p'p-DDT	327.5	704.2	289.7	257.3	62.8	164.2
CHB-26	131.2	494.1	138.3	611.1	59.1	99.5
CHB-40	218.5	117.5	30.6	98.6	53.5	34.1
CHB-41	48.9	71.7	24.4	132.4	11.6	50.8
CHB-44	138.5	173.3	84.8	510.7	30.8	63.3
CHB-50	30.0	76.8	206.4	982.3	7.8	156.0
CHB-62	< 7.0	106.2	31.2	36.7	12.8	15.5
oxychlordan	2504.6	1256.0	2840.3	1646.4	1011.5	1762.2
trans-chlordan	10.3	< 3.0	< 2.5	< 3.7	< 5.3	< 2.9
cis-chlordan	66.6	15.7	10.9	13.7	1.7	5.8
trans-nonachlor	1646.2	970.9	968.2	1626.7	176.4	561.6
cis-nonachlor	1476.2	655.1	333.4	650.6	68.7	288.4

Ring No.:	3050526	3050526	3050142	3050142	3050556	
Reg. No.:	01-1613	01-1614	01-1606-1	01-1606-2	01-1645	
Batch No.:	#02-17	#02-17	#02-17	#02-17	#02-17	
Year of sampling:	1998	1999	2000	2000	2001	
Place of sampling:	Upernaviarsuk	Upernaviarsuk	Igaliko	Igaliko	Egaluit	
Sample weight (g):	6.76	7.4561	7.5774	7.4622	7.022	
Fat content (%):	7.6171	3.8854	2.7293	2.8214	6.4711	
Dry matter (% weight):	33.1614	21.707	28.907	23.4434	24.5998	
Substance	Concentration in ng/g wet weight					
alfa-HCH	0.49	0.57	0.49	0.47	0.26	
beta-HCH	4.67	5.61	9.75	10.21	8.59	
gamma-HCH	0.39	0.53	0.27	0.31	< 0.22	
HCB	24.04	17.09	21.56	20.84	10.83	
o'p-DDE	12.02	39.15	13.55	14.06	32.39	
o'p-DDT	0.67	1.17	< 0.11	< 0.11	1.29	
p'p'-DDD	1.34	1.61	5.02	5.29	1.96	
p'p-DDE	686.85	1144.09	1109.92	1070.98	1287.45	
p'p-DDT	10.95	12.55	5.96	6.68	10.74	
CHB-26	6.74	12.13	2.93	2.87	1.49	
CHB-40	2.11	2.99	1.96	2.02	0.35	
CHB-41	2.42	4.80	1.02	1.09	0.72	
CHB-44	5.68	10.07	1.13	1.56	3.32	
CHB-50	14.71	23.30	2.32	2.47	3.15	
CHB-62	2.28	5.49	< 0.41	< 0.42	< 0.45	
oxychlordan	27.22	62.20	30.73	31.16	76.74	
trans-chlordan	< 0.19	0.18	< 0.17	< 0.17	< 0.18	
cis-chlordan	0.13	0.39	0.09	0.10	0.08	
trans-nonachlor	22.17	65.65	15.94	16.60	28.50	
cis-nonachlor	7.00	18.09	10.73	11.67	5.66	
	Concentration in ng/g fat weight					
alfa-HCH	6.4	14.6	17.8	16.6	4.0	
beta-HCH	61.3	144.3	357.3	362.0	132.7	
gamma-HCH	5.1	13.5	10.1	11.0	< 3.4	
HCB	315.6	439.9	790.0	738.7	167.4	
o'p-DDE	157.8	1007.7	496.5	498.2	500.5	
o'p-DDT	8.8	30.1		< 3.9	20.0	
p'p'-DDD	17.6	41.5	183.8	187.3	30.3	
p'p-DDE	9017.2	29445.9	40666.8	37959.1	19895.4	
p'p-DDT	143.8	323.0	218.4	236.9	166.0	
CHB-26	88.5	312.1	107.3	101.7	23.1	
CHB-40	27.7	77.0	71.7	71.7	5.5	
CHB-41	31.8	123.5	37.4	38.5	11.1	
CHB-44	74.6	259.1	41.4	55.5	51.2	
CHB-50	193.1	599.8	85.1	87.6	48.7	
CHB-62	29.9	141.2	< 15.0	< 14.9	< 7.0	
oxychlordan	357.3	1600.8	1126.0	1104.3	1185.9	
trans-chlordan	< 2.5	4.6	< 6.2	< 6.0	< 2.8	
cis-chlordan	1.7	10.1	3.3	3.5	1.2	
trans-nonachlor	291.1	1689.7	583.9	588.3	440.4	
cis-nonachlor	91.9	465.6	393.1	413.5	87.4	

Appendix 4

Eggshell thickness

Reg. No.	Eggshell thickness (mm)
01-1593	0.276
01-1598	0.285
01-1601	0.312
01-1602	0.308
01-1603	0.292
01-1604	0.273
01-1606-1	0.297
01-1606-2	0.297
01-1607	0.322
01-1608	0.301
01-1609	0.307
01-1610	0.293
01-1611	0.345
01-1613	0.307
01-1614	0.299
01-1615	0.308
01-1616	0.294
01-1617	0.328
01-1618	0.283
01-1619	0.294

Reg. No.	Eggshell thickness (mm)
01-1620	0.308
01-1621	0.290
01-1622	0.290
01-1623	0.337
01-1626	0.323
01-1629	0.291
01-1630	0.317
01-1631	0.309
01-1632	0.340
01-1633	0.331
01-1645	0.351
02-1785	0.303
02-1787-1	0.287
02-1787-2	0.287
02-1788	0.307
02-1789-1	0.301
02-1789-2	0.301
03-0542-1	0.340
03-0542-2	0.340
03-0543	0.285

Appendix 5

Long-term changes in eggshell thickness of the Peregrine Falcon *Falco peregrinus tundrius* in Greenland

Abstract

Thickness of eggshell fragments and whole eggs form the Peregrine Falcon *Falco peregrinus* collected in South Greenland between 1981 and 2003 were measured and compared to shell thickness of pre-DDT eggs, also collected in Greenland. Linear regression on fragment data yields a weak but significant increase in the average thickness of eggshells over the period, corresponding to a change in eggshell thinning from 12.8% in 1981 to 8.2% thinning in 2003. When the sample was grouped into two equal time periods, a significant difference in mean shell thickness could also be detected.

Introduction

The effects of persistent organic pollutants (POP) on the eggshell thickness and breeding success in high-trophic level birds have been widely documented. Especially DDT and its metabolites have been identified as a key group of POPs responsible for the widespread reduction in breeding success and subsequent population decline in the Peregrine Falcon *Falco peregrinus* (Hickey 1969, Hickey & Andersson 1968, Newton 1979, Peakall et al. 1975, Peakall et al. 1976, Peakall & Kiff 1979, Ratcliffe 1970, Ratcliffe 1980, Walker et al. 1973).

The Greenlandic Peregrine Falcon (*F.p. tundrius*) population has been the subject of long-term studies in West Greenland since 1972 (e.g. Burnham & Mattox 1984, Mattox and Seegar 1988, Restani and Mattox 2000) and South Greenland since 1981 (Falk & Møller 1986, 1988). The Greenland population is roughly estimated at 500-1000 pairs (Falk & Møller 1988), and based on autumn migration counts in the eastern US (Titus & Fuller 1990) there is slight evidence for population increase in the Arctic.

It is thus to be expected that a population of peregrines recovering from the effects of POP-exposure (such as the Greenlandic) will show an increase in eggshell thickness over time.

Eggshell thickness analyses have often relied on analysis of whole addled eggs. However, these eggs are relatively rare occurrences, are not present in the most successful nests (all eggs hatch), and may potentially be biased since very thin eggs would break (Odsjö 1982). By using eggshell fragments from all nests a more representative and comprehensive survey of the wild population can be obtained, and based on the intra- and inter-clutch variation in shell thickness, Falk & Møller (1990) recommended that studies of eggshell thinning based on fragments should include as many nests as possible.

Methods

Study area and sampling

The study area covers the inner parts of the three southernmost municipalities of Southwest Greenland, Nanortalik, Qaqortoq and Narsaq, approx within 60° – 61° N and 45° – 46° W (Fig. 1). The area is low Arctic, with tundra vegetation and willow and birch shrub in the warm, sub-arctic areas far from the cool outer coast. Sheep grazes a substantial part of the area.

Field surveys of the Peregrine Falcon population have been conducted in the study area annually between 1981 and 2003. Active nests were visited at least once post-haching, and, when conditions allowed, the nest scrape carefully searched for eggshell fragments deriving from the hatched eggs. In addition, any whole-addled eggs were collected for contaminant analysis (to be published elsewhere).

Measurements and analyses

The shell fragments were measured with a computer-connected Mitutyuo Digital Micrometer (type 293-521-30) with a small stainless steel ball glued to the rotating jaw in order to fit in the inner curved surface of the eggshell fragments.

Each fragment was scrutinised to determine whether any membrane was still adhering to the inner surface. Measurements were performed only on (parts of) fragments without any membrane because on fragments it is difficult to be certain if both membranes (shell membrane and egg membrane) are present.

Samples from a total of 93 clutches were measured and provided from 3 to 91 membrane-free measurements. However, since eggshell thickness varies within the egg there is a risk that too few samples may bias results. Hence, we chose to include only 79 clutches that provided 20 or more measurable fragments – the same threshold selected by Odsjö (1982) in a study of Swedish Ospreys – and assumed they represented the thickness of the entire clutch.

32 whole, addled eggs were also collected and kept frozen. When opened in the laboratory for contaminant studies, the eggs were cut along the equator and the empty half shells washed with water before being left to dry for 3 months at room temperature. The half shells were measured along the equator with a modified Mitutyo Micrometer (type 147-301) – the same device used to measure 16 Greenlandic pre-DDT clutches (48 eggs) in the collection at Zoological Museum, Copenhagen (Falk & Møller 1990). There was no significant difference in measurements taken with the two different tools. The egg opening method left parts of the shell with membranes intact and other parts without membranes. Consequently, most measurements include membranes but some are without. To be able to compare measurements with and without membranes, a membrane factor was independently determined by measuring adjacent points with and without membranes on 26 whole eggs (the cut halves): 0.071 mm (n = 83, S.D. = 0.013). This compares well to the 0.07 mm used by Nygård (1983) and 0.069 mm reported by Court et al. 1990).

The sample unit is 'nest-year', i.e. a mean shell thickness value for each nest and year.

Results

Mean shell thickness for whole eggs was 0.307 mm (S.D. = 0.0215) while mean thickness of fragments was 0.302 mm (S.D. = 0.0206). The difference was not significant (T-test, P=0.2608).

During the period 1981-2003 there was a weak but significant (P=0.02532, N=79) increase in the average thickness of eggshell fragments (Fig. 2a & b); the slope of the regression shows an average increase of 0.21% per year. This would correspond to a change in eggshell thinning from 12.8% in 1981 to 8.2% thinning in 2003 when compared to pre-DDT eggs collected in Greenland (0.336 mm, 48 eggs from 16 clutches, Falk & Møller 1990).

A similar trend could not be detected in the whole egg sample (1986-2003, P=0.776, N=28).

When the combined sample (whole eggs and fragments) were grouped into two equal time periods a significant difference in mean shell thickness could be detected (1981-1992: 0.299 mm; 1993-2003: 0.307 mm; T-test, P=0,0260).

Discussion

Although the embryo may extract minerals from the eggshell, it has been shown that while shell density (measured as shell index, Ratcliffe 1967) is affected, the shell thickness *per se* does not change significantly during incubation (Bennett 1995, Bunck 1985). State of incubation is thus not considered in this study.

Walker et al. (1973) compared shell thickness of material from 9 eggs (2 whole eggs and fragments from 7 eggs) collected in

West Greenland to 42 pre-DDT eggs from Greenland and measured a 14% thinning. Similarly, in a previous assessment based on our samples from 1981-1985 (Falk and Møller, 1988) and the same collection of Greenlandic pre-DDT eggs we arrived at 14% eggshell thinning. The current measurements from the same period, treated isolated, show a 13% shell thinning, which is in good agreement with the previous results.

In an early review Peakall and Kiff (1988) reported post-DDT eggshell thinning in peregrine populations from 30 different parts of the world ranging from below 5% and up to 25%. Among these, northern migrating populations had eggshell thinning values between 13% and 23% in the 1960's and 70's (Berger et al. 1970, Burnham & Mattox 1984, Cade et al. 1971, Nelson & Myres 1975, Nygård 1983, Odsjö & Lindberg 1977, Peakall et al. 1975, White & Cade 1977). More recent studies of eggshell thinning in arctic migrating peregrine populations show values of 10,6% (Alaska, 1991-95, Ambrose et al. 2000) and 15% (Arctic Canada, 1990-1994, Johnstone et al. 1996). No time trend could be detected in any of these studies.

If we assume the trend to be linear, the regression line based on eggshell fragment data can be extended backwards for a rough assessment of when/if the thinning exceeded the critical empirical "threshold" of about 17% (Peakall & Kiff 1988). The shell thinning might have been near or above the critical limit in the 1950'es only – probably too short after DDT became widespread (introduced 1947) to have had a marked effect on the Greenlandic Peregrine population as supported by evidence of a strong population since the 1970's (Burnham & Mattox 1984). This is despite the fact that the Arctic subspecies in Greenland migrates through and/or to areas (Latin America) where phasing out of the pesticides has been slower than in North America, or where a renewed use has been deemed necessary to fight Malaria.

To our knowledge, this is the first time an increase in eggshell thickness over time has been detected in a Peregrine population. However, Nygård (1999) observed a slight increase in shell thickness of eggs from Norwegian Merlins (*Falco columbarius*) when comparing eggs from the 1990's (8-11% reduction) to eggs from the 1960's and 1970's (15% reduction).

Acknowlegdements

The field samples have been collected during more that two decades, with financial support from public funds (Danish Natural Science Reearch Council (SNF) and Commision for Scientific Research in Greenland (KVUG)) and private funds (Inge og Skjold Burnes Fond, Dronning Margrethe og Prins Henriks Fond, Aase og Jørgen Münters Fond, K.A. Rohde og Hustrus Legat, Bodil Petersen Fonden, Kronprins Frederiks Fond, Det Kongelige Grønlandsfond, Folketingets Grønlandsfond, Tipsmidlerne, Lions Klubbernes Grønlandsfond, The Peregrine Fund). Fieldwork was carried out by tens of dedicated birders

on a voluntary basis. Especially we will thank Kaj Nielsen, Qaqortoq, for providing our boat, maintenance and various logistic support every year, and shifting managers and staff at Fjeldstationen, Narsarsuaq, for allowing us to use the station as an informal base. The Danish Defence Command provided aerial support to/from Greenland, and for several years the Ice Patrol has assisted with ad hoc helicopter flights. Danish Environmental Protection Agency, Ministry of Environment, has supported the analyses.

Figure legends

Figure 1. Map of sampling sites in South Greenland spanning from outer coast to inland areas; all nest sites sampled in this study are located inside the hatched area. White areas with dashed line edge are ice/glaciers.

Figure 2. Eggshell thickness vs. sampling year of Peregrine Falcon eggs from South Greenland based on: a) eggshell fragments 1981 – 2003, and b) whole addled eggs.

Figure 3. Shell thickness of Peregrine eggs from Greenland; pre-DTT Peregrine eggs (1881 – 1930) from museum collections, and recent samples from South Greenland (this study) with filled circles representing whole eggs, and open circles indicating fragments. The reference lines indicate pre-DTT mean thickness (0.336 mm), and the approximate empiric "17% threshold" (0.279 mm) for population declines (Peakall and Kiff 1988). The regression line of the recent change in thickness is extrapolated backwards.

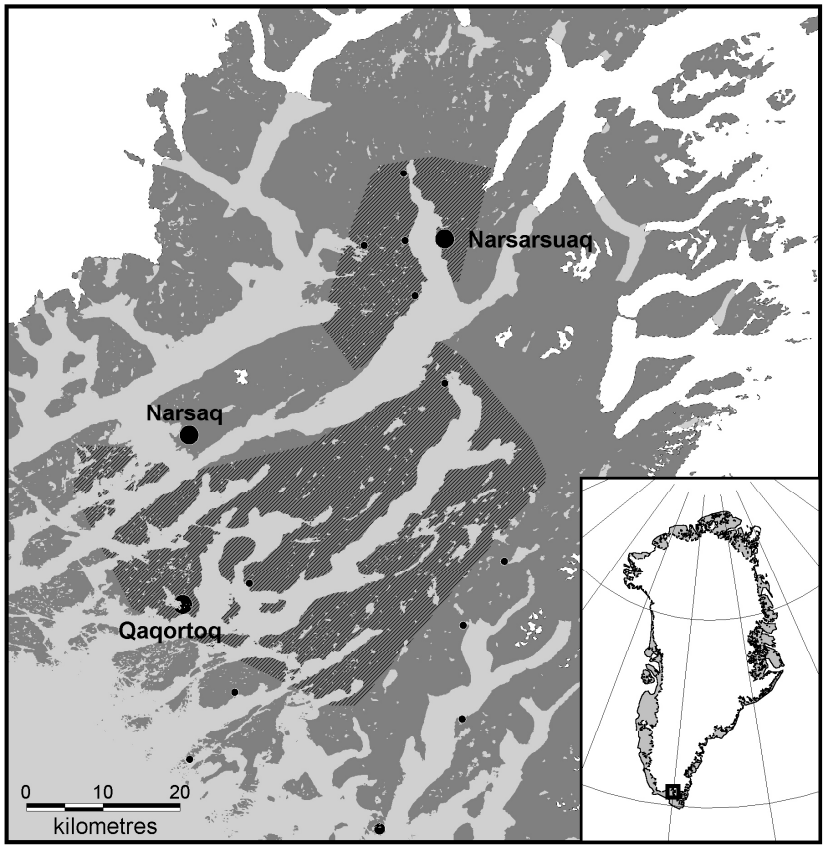

Figure 1.

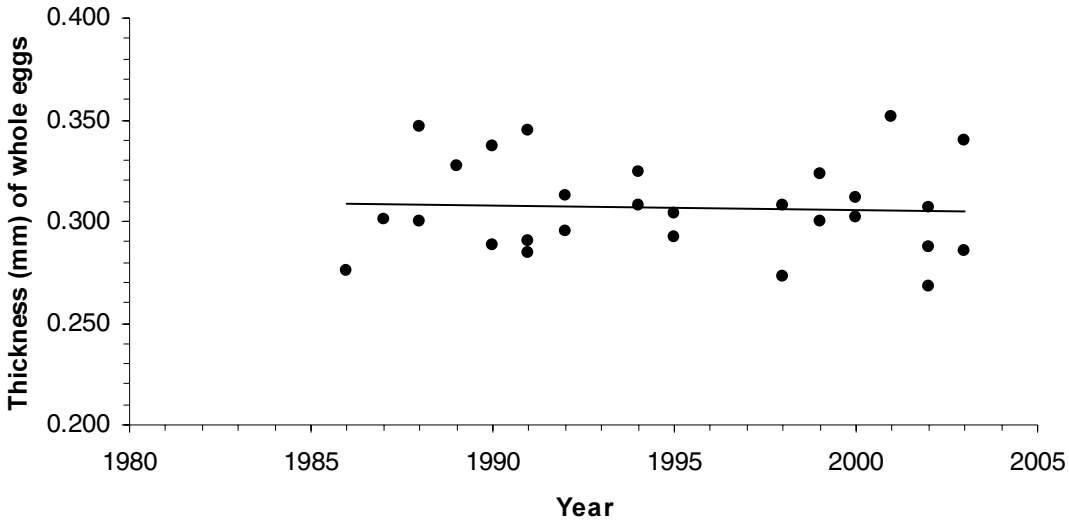

Figure 2a

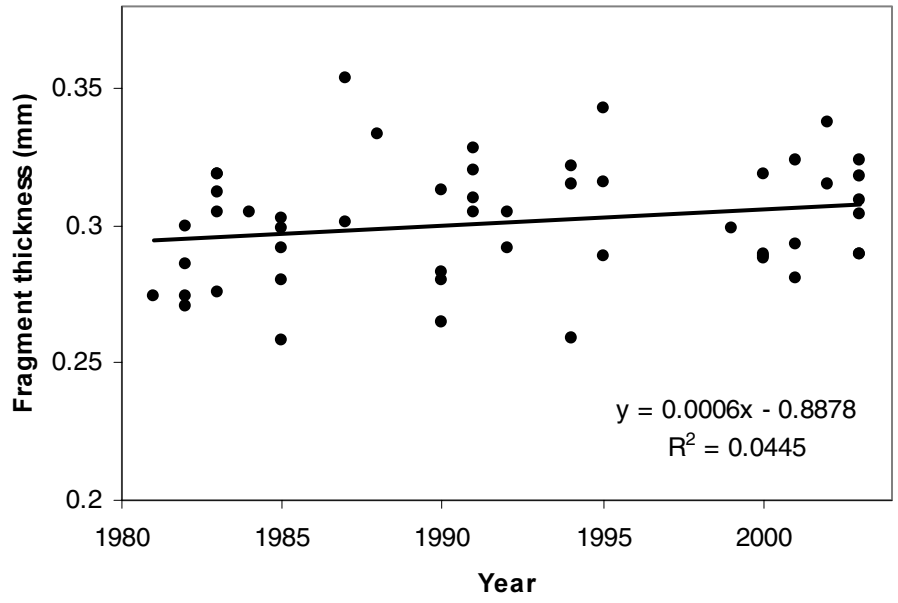

Figure 2b

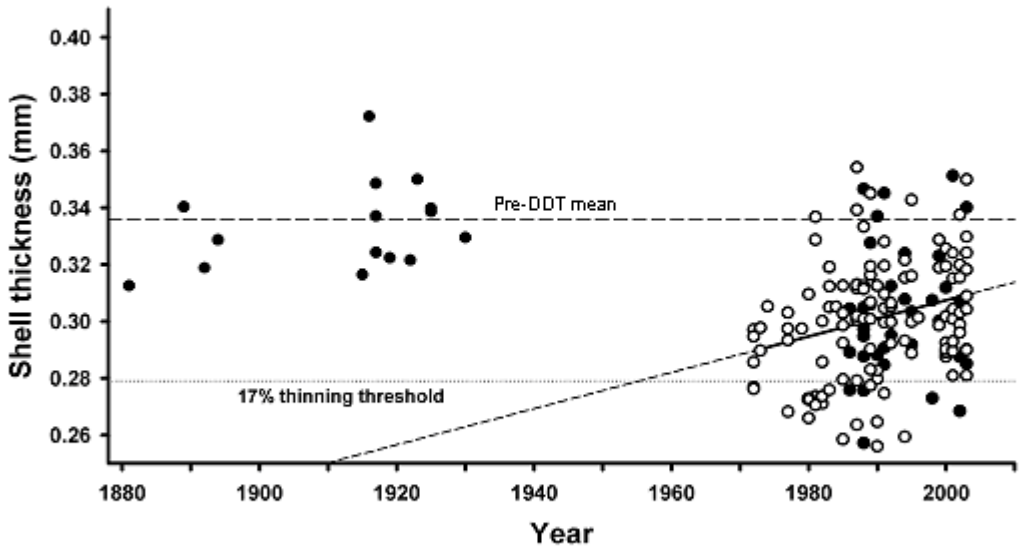

Figure 3.

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Appendix 6

Correlation matrix (Pearson correlation coefficient)

	CB-28	CB-31	CB-44	CB-49	CB-52	CB-99
CB-28	1.00	0.70	0.82	0.79	0.57	0.84
CB-31	0.70	1.00	0.72	0.71	0.59	0.86
CB-44	0.82	0.72	1.00	0.91	0.78	0.81
CB-49	0.79	0.71	0.91	1.00	0.92	0.82
CB-52	0.57	0.59	0.78	0.92	1.00	0.70
CB-99	0.84	0.86	0.81	0.82	0.70	1.00
CB-101	0.40	0.48	0.54	0.86	0.71	0.51
CB-105	0.84	0.85	0.83	0.81	0.69	0.98
CB-110	0.76	0.77	0.71	0.86	0.73	0.87
CB-118	0.79	0.86	0.79	0.81	0.70	0.99
CB-128	0.78	0.85	0.78	0.78	0.66	0.97
CB-138	0.77	0.84	0.74	0.78	0.68	0.97
CB-149	0.69	0.66	0.82	0.83	0.74	0.75
CB-151	0.28	0.11	0.30	0.44	0.39	0.35
CB-153	0.73	0.81	0.69	0.76	0.66	0.94
CB-156	0.66	0.84	0.68	0.72	0.61	0.92
CB-170	0.66	0.81	0.67	0.73	0.63	0.90
CB-180	0.62	0.78	0.65	0.73	0.64	0.86
CB-187	0.76	0.85	0.75	0.76	0.67	0.96
CB-194	0.54	0.77	0.60	0.70	0.59	0.81
CB-209	0.67	0.73	0.69	0.62	0.57	0.78
alfa-HCH	0.45	0.58	0.67	0.78	0.80	0.61
beta-HCH	0.45	0.55	0.60	0.43	0.47	0.53
gamma-HCH	0.33	-0.10	0.11	0.02	-0.03	0.12
HCB	0.61	0.57	0.60	0.63	0.64	0.65
o'p-DDE	0.45	0.64	0.52	0.56	0.54	0.62
o'p-DDT	0.59	0.70	0.38	0.37	0.34	0.59
p'p'-DDD	0.26	0.43	0.52	0.40	0.59	0.32
p'p-DDE	0.47	0.54	0.54	0.51	0.61	0.50
p'p-DDT	0.44	0.64	0.40	0.41	0.43	0.54
CHB-26	0.38	0.49	0.27	0.39	0.32	0.43
CHB-40	0.29	0.32	0.35	0.45	0.46	0.30
CHB-41	0.43	0.48	0.35	0.46	0.34	0.45
CHB-44	0.49	0.69	0.39	0.53	0.40	0.59
CHB-50	0.42	0.55	0.39	0.49	0.41	0.46
CHB-62	0.11	0.46	0.24	0.41	0.29	0.25
oxychlordan	0.63	0.84	0.66	0.66	0.66	0.85
trans-chlordan	0.47	0.67	0.55	0.55	0.57	0.62
cis-chlordan	0.51	0.49	0.58	0.48	0.56	0.52
trans-nonachlor	0.68	0.89	0.68	0.72	0.66	0.85
cis-nonachlor	0.75	0.88	0.77	0.77	0.72	0.86
BDE-17	0.73	0.78	0.75	0.72	0.24	0.59
BDE-28	0.56	0.12	0.39	0.54	0.60	0.44
BDE-49	0.41	0.57	0.42	0.46	0.31	0.53
BDE-47	0.75	0.65	0.65	0.71	0.53	0.76
BDE-66	0.58	0.42	0.73	0.84	0.84	0.70
BDE-100	0.61	0.58	0.56	0.57	0.36	0.67
BDE-99	0.61	0.54	0.50	0.51	0.28	0.62
BDE-85	-0.21	0.08	-0.03	-0.07	0.07	0.03
BDE-154	0.47	0.69	0.56	0.53	0.53	0.70
BDE-153	0.52	0.55	0.52	0.43	0.36	0.58
BDE-183	0.52	0.24	0.43	0.21	0.17	0.32
BDE-209	0.28	0.20	0.27	0.05	0.06	0.34
Shell Tickn.	-0.15	-0.37	-0.26	-0.17	-0.32	-0.33
HBCD	-0.18	-0.23	-0.26	-0.47	-0.57	-0.24
Me-TBBP-A	0.12	0.18	-0.02	-0.17	-0.38	0.18

	CB-101	CB-105	CB-110	CB-118	CB-128	CB-138
CB-28	0.40	0.84	0.76	0.79	0.78	0.77
CB-31	0.48	0.85	0.77	0.86	0.85	0.84
CB-44	0.54	0.83	0.71	0.79	0.78	0.74
CB-49	0.86	0.81	0.86	0.81	0.78	0.78
CB-52	0.71	0.69	0.73	0.70	0.66	0.68
CB-99	0.51	0.98	0.87	0.99	0.97	0.97
CB-101	1.00	0.53	0.74	0.54	0.50	0.53
CB-105	0.53	1.00	0.86	0.98	0.98	0.95
CB-110	0.74	0.86	1.00	0.86	0.83	0.88
CB-118	0.54	0.98	0.86	1.00	0.98	0.98
CB-128	0.50	0.98	0.83	0.98	1.00	0.97
CB-138	0.53	0.95	0.88	0.98	0.97	1.00
CB-149	0.71	0.82	0.71	0.76	0.78	0.71
CB-151	0.26	0.42	0.38	0.38	0.41	0.37
CB-153	0.53	0.92	0.85	0.96	0.95	0.98
CB-156	0.49	0.91	0.83	0.95	0.95	0.97
CB-170	0.46	0.89	0.81	0.92	0.94	0.95
CB-180	0.46	0.85	0.80	0.89	0.90	0.93
CB-187	0.51	0.95	0.84	0.96	0.98	0.97
CB-194	0.44	0.80	0.74	0.84	0.87	0.87
CB-209	0.42	0.81	0.62	0.78	0.84	0.75
alfa-HCH	0.59	0.62	0.65	0.61	0.59	0.61
beta-HCH	0.00	0.54	0.31	0.50	0.47	0.44
gamma-HCH	0.11	0.13	0.26	0.09	0.09	0.12
HCB	0.36	0.67	0.66	0.62	0.59	0.64
o'p-DDE	0.23	0.59	0.55	0.59	0.58	0.61
o'p-DDT	0.35	0.54	0.62	0.57	0.50	0.55
p'p'-DDD	0.32	0.34	0.21	0.31	0.33	0.31
p'p-DDE	0.48	0.55	0.44	0.50	0.53	0.45
p'p-DDT	0.48	0.55	0.62	0.55	0.54	0.55
CHB-26	0.24	0.41	0.50	0.41	0.40	0.44
CHB-40	0.27	0.31	0.39	0.27	0.26	0.31
CHB-41	0.28	0.48	0.50	0.44	0.45	0.41
CHB-44	0.36	0.58	0.61	0.58	0.59	0.60
CHB-50	0.29	0.44	0.50	0.44	0.42	0.43
CHB-62	0.47	0.27	0.43	0.30	0.27	0.35
oxychlordan	0.47	0.80	0.76	0.85	0.80	0.85
trans-chlordan	0.26	0.61	0.45	0.59	0.50	0.52
cis-chlordan	0.32	0.53	0.53	0.44	0.48	0.48
trans-nonachlor	0.43	0.82	0.76	0.84	0.82	0.83
cis-nonachlor	0.49	0.86	0.79	0.83	0.83	0.81
BDE-17	0.01	0.72	-0.09	0.58	0.83	0.48
BDE-28	0.41	0.52	0.38	0.36	0.52	0.43
BDE-49	0.45	0.61	0.46	0.61	0.60	0.54
BDE-47	0.47	0.77	0.72	0.75	0.78	0.76
BDE-66	0.73	0.74	0.71	0.72	0.66	0.66
BDE-100	0.39	0.70	0.55	0.70	0.73	0.66
BDE-99	0.34	0.64	0.52	0.64	0.67	0.62
BDE-85	0.24	0.06	-0.03	0.12	0.20	0.17
BDE-154	0.36	0.71	0.54	0.75	0.76	0.76
BDE-153	0.38	0.65	0.44	0.65	0.68	0.60
BDE-183	0.28	0.40	0.33	0.33	0.37	0.30
BDE-209	-0.02	0.38	0.08	0.38	0.43	0.34
Shell Tickn.	-0.10	-0.34	-0.25	-0.33	-0.34	-0.33
HBCD	-0.37	-0.26	-0.33	-0.28	-0.24	-0.18
Me-TBBP-A	-0.24	0.23	0.09	0.20	0.26	0.20

	CB-149	CB-151	CB-153	CB-156	CB-170	CB-180
CB-28	0.69	0.28	0.73	0.66	0.66	0.62
CB-31	0.66	0.11	0.81	0.84	0.81	0.78
CB-44	0.82	0.30	0.69	0.68	0.67	0.65
CB-49	0.83	0.44	0.76	0.72	0.73	0.73
CB-52	0.74	0.39	0.66	0.61	0.63	0.64
CB-99	0.75	0.35	0.94	0.92	0.90	0.86
CB-101	0.71	0.26	0.53	0.49	0.46	0.46
CB-105	0.82	0.42	0.92	0.91	0.89	0.85
CB-110	0.71	0.38	0.85	0.83	0.81	0.80
CB-118	0.76	0.38	0.96	0.95	0.92	0.89
CB-128	0.78	0.41	0.95	0.95	0.94	0.90
CB-138	0.71	0.37	0.98	0.97	0.95	0.93
CB-149	1.00	0.49	0.68	0.65	0.63	0.60
CB-151	0.49	1.00	0.38	0.34	0.37	0.35
CB-153	0.68	0.38	1.00	0.98	0.97	0.96
CB-156	0.65	0.34	0.98	1.00	0.98	0.97
CB-170	0.63	0.37	0.97	0.98	1.00	0.99
CB-180	0.60	0.35	0.96	0.97	0.99	1.00
CB-187	0.75	0.41	0.94	0.93	0.92	0.88
CB-194	0.56	0.37	0.92	0.94	0.97	0.98
CB-209	0.78	0.46	0.72	0.70	0.73	0.67
alfa-HCH	0.57	0.27	0.57	0.58	0.55	0.57
beta-HCH	0.43	0.13	0.41	0.45	0.42	0.43
gamma-HCH	0.09	0.20	0.12	0.08	0.10	0.09
HCB	0.58	0.45	0.62	0.61	0.59	0.61
o'p-DDE	0.37	0.05	0.56	0.59	0.57	0.58
o'p-DDT	0.32	-0.05	0.53	0.56	0.46	0.43
p'p'-DDD	0.42	-0.16	0.26	0.27	0.28	0.28
p'p-DDE	0.67	0.16	0.41	0.41	0.42	0.36
p'p-DDT	0.54	0.19	0.51	0.51	0.48	0.45
CHB-26	0.28	0.24	0.45	0.48	0.48	0.48
CHB-40	0.31	0.20	0.32	0.36	0.36	0.41
CHB-41	0.41	0.33	0.42	0.45	0.46	0.45
CHB-44	0.41	0.11	0.59	0.63	0.63	0.62
CHB-50	0.26	0.01	0.45	0.48	0.51	0.51
CHB-62	0.29	0.07	0.41	0.46	0.46	0.51
oxychlordan	0.55	0.05	0.83	0.82	0.77	0.76
trans-chlordan	0.38	-0.42	0.48	0.52	0.44	0.46
cis-chlordan	0.42	-0.04	0.41	0.42	0.45	0.46
trans-nonachlor	0.61	0.15	0.82	0.82	0.82	0.80
cis-nonachlor	0.74	0.27	0.78	0.78	0.78	0.76
BDE-17	0.14	-0.89	0.42	0.52	0.59	0.44
BDE-28	0.80	0.75	0.40	0.27	0.36	0.32
BDE-49	0.56	0.22	0.60	0.60	0.58	0.58
BDE-47	0.64	0.42	0.77	0.71	0.74	0.72
BDE-66	0.75	0.41	0.61	0.51	0.47	0.46
BDE-100	0.62	0.39	0.69	0.64	0.66	0.62
BDE-99	0.53	0.36	0.64	0.60	0.62	0.59
BDE-85	0.14	0.17	0.27	0.25	0.36	0.37
BDE-154	0.49	0.11	0.79	0.80	0.81	0.81
BDE-153	0.63	0.36	0.64	0.60	0.63	0.60
BDE-183	0.48	0.40	0.32	0.23	0.28	0.26
BDE-209	0.30	0.36	0.37	0.34	0.38	0.34
Shell Tickn.	-0.20	-0.10	-0.30	-0.36	-0.37	-0.36
HBCD	-0.34	-0.43	-0.22	-0.10	-0.16	-0.15
Me-TBBP-A	-0.03	-0.05	0.19	0.32	0.25	0.23

	CB-187	CB-194	CB-209	alfa-HCH	beta-HCH	gamma-HCH
CB-28	0.76	0.54	0.67	0.45	0.45	0.33
CB-31	0.85	0.77	0.73	0.58	0.55	-0.10
CB-44	0.75	0.60	0.69	0.67	0.60	0.11
CB-49	0.76	0.70	0.62	0.78	0.43	0.02
CB-52	0.67	0.59	0.57	0.80	0.47	-0.03
CB-99	0.96	0.81	0.78	0.61	0.53	0.12
CB-101	0.51	0.44	0.42	0.59	0.00	0.11
CB-105	0.95	0.80	0.81	0.62	0.54	0.13
CB-110	0.84	0.74	0.62	0.65	0.31	0.26
CB-118	0.96	0.84	0.78	0.61	0.50	0.09
CB-128	0.98	0.87	0.84	0.59	0.47	0.09
CB-138	0.97	0.87	0.75	0.61	0.44	0.12
CB-149	0.75	0.56	0.78	0.57	0.43	0.09
CB-151	0.41	0.37	0.46	0.27	0.13	0.20
CB-153	0.94	0.92	0.72	0.57	0.41	0.12
CB-156	0.93	0.94	0.70	0.58	0.45	0.08
CB-170	0.92	0.97	0.73	0.55	0.42	0.10
CB-180	0.88	0.98	0.67	0.57	0.43	0.09
CB-187	1.00	0.86	0.86	0.58	0.42	0.08
CB-194	0.86	1.00	0.71	0.51	0.36	0.03
CB-209	0.86	0.71	1.00	0.42	0.33	-0.04
alfa-HCH	0.58	0.51	0.42	1.00	0.49	-0.09
beta-HCH	0.42	0.36	0.33	0.49	1.00	-0.24
gamma-HCH	0.08	0.03	-0.04	-0.09	-0.24	1.00
HCB	0.56	0.50	0.42	0.57	0.68	0.01
o'p-DDE	0.55	0.52	0.38	0.61	0.54	-0.13
o'p-DDT	0.49	0.35	0.09	0.43	0.47	0.55
p'p'-DDD	0.33	0.25	0.37	0.54	0.40	-0.40
p'p-DDE	0.55	0.35	0.68	0.50	0.33	-0.06
p'p-DDT	0.56	0.44	0.55	0.36	0.27	0.16
CHB-26	0.44	0.47	0.37	0.32	0.31	0.17
CHB-40	0.25	0.35	0.13	0.52	0.49	0.20
CHB-41	0.45	0.46	0.50	0.32	0.25	0.19
CHB-44	0.63	0.64	0.61	0.38	0.28	0.09
CHB-50	0.42	0.52	0.42	0.34	0.25	0.06
CHB-62	0.24	0.52	0.10	0.13	0.28	-0.05
oxychlordan	0.81	0.68	0.54	0.66	0.51	-0.02
trans-chlordan	0.44	0.36	0.24	0.70	0.87	-0.47
cis-chlordan	0.47	0.38	0.38	0.68	0.39	0.30
trans-nonachlor	0.82	0.76	0.70	0.57	0.54	-0.07
cis-nonachlor	0.83	0.71	0.76	0.67	0.58	0.01
BDE-17	0.82	0.48	-0.43	0.80	0.63	0.43
BDE-28	0.59	0.26	0.60	0.40	0.51	0.30
BDE-49	0.49	0.58	0.45	0.27	0.21	-0.02
BDE-47	0.76	0.69	0.73	0.39	0.17	0.12
BDE-66	0.60	0.35	0.48	0.69	0.17	0.40
BDE-100	0.69	0.65	0.74	0.26	0.10	0.00
BDE-99	0.64	0.61	0.68	0.19	0.02	0.06
BDE-85	0.22	0.46	0.38	-0.05	-0.48	-0.18
BDE-154	0.73	0.81	0.58	0.48	0.42	-0.10
BDE-153	0.64	0.64	0.76	0.19	0.17	-0.06
BDE-183	0.36	0.27	0.48	0.04	-0.07	0.49
BDE-209	0.42	0.37	0.54	0.13	0.03	0.09
Shell Tickn.	-0.34	-0.36	-0.40	-0.35	-0.32	0.40
HBCD	-0.27	-0.19	-0.40	-0.41	-0.04	0.20
Me-TBBP-A	0.17	0.28	0.07	-0.09	0.22	0.32

	HCB	o'p-DDE	o'p-DDT	p'p'-DDD	p'p-DDE	p'p-DDT
CB-28	0.61	0.45	0.59	0.26	0.47	0.44
CB-31	0.57	0.64	0.70	0.43	0.54	0.64
CB-44	0.60	0.52	0.38	0.52	0.54	0.40
CB-49	0.63	0.56	0.37	0.40	0.51	0.41
CB-52	0.64	0.54	0.34	0.59	0.61	0.43
CB-99	0.65	0.62	0.59	0.32	0.50	0.54
CB-101	0.36	0.23	0.35	0.32	0.48	0.48
CB-105	0.67	0.59	0.54	0.34	0.55	0.55
CB-110	0.66	0.55	0.62	0.21	0.44	0.62
CB-118	0.62	0.59	0.57	0.31	0.50	0.55
CB-128	0.59	0.58	0.50	0.33	0.53	0.54
CB-138	0.64	0.61	0.55	0.31	0.45	0.55
CB-149	0.58	0.37	0.32	0.42	0.67	0.54
CB-151	0.45	0.05	-0.05	-0.16	0.16	0.19
CB-153	0.62	0.56	0.53	0.26	0.41	0.51
CB-156	0.61	0.59	0.56	0.27	0.41	0.51
CB-170	0.59	0.57	0.46	0.28	0.42	0.48
CB-180	0.61	0.58	0.43	0.28	0.36	0.45
CB-187	0.56	0.55	0.49	0.33	0.55	0.56
CB-194	0.50	0.52	0.35	0.25	0.35	0.44
CB-209	0.42	0.38	0.09	0.37	0.68	0.55
alfa-HCH	0.57	0.61	0.43	0.54	0.50	0.36
beta-HCH	0.68	0.54	0.47	0.40	0.33	0.27
gamma-HCH	0.01	-0.13	0.55	-0.40	-0.06	0.16
HCB	1.00	0.50	0.39	0.27	0.30	0.32
o'p-DDE	0.50	1.00	0.56	0.37	0.31	0.41
o'p-DDT	0.39	0.56	1.00	-0.06	0.13	0.44
p'p'-DDD	0.27	0.37	-0.06	1.00	0.54	0.23
p'p-DDE	0.30	0.31	0.13	0.54	1.00	0.60
p'p-DDT	0.32	0.41	0.44	0.23	0.60	1.00
CHB-26	0.46	0.36	0.47	-0.17	0.44	0.51
CHB-40	0.64	0.41	0.29	0.12	0.34	0.37
CHB-41	0.40	0.37	0.23	-0.17	0.51	0.58
CHB-44	0.36	0.45	0.44	0.08	0.64	0.70
CHB-50	0.28	0.46	0.31	0.15	0.50	0.56
CHB-62	0.44	0.33	0.36	0.02	0.02	0.58
oxychlordan	0.56	0.62	0.60	0.43	0.41	0.44
trans-chlordan	0.73	0.45	0.28	0.59	0.38	0.18
cis-chlordan	0.56	0.62	0.15	0.55	0.18	0.20
trans-nonachlor	0.63	0.67	0.58	0.41	0.58	0.61
cis-nonachlor	0.69	0.63	0.56	0.46	0.66	0.63
BDE-17	-0.53	0.67	0.49	0.48	0.19	-0.96
BDE-28	0.83	0.03	0.25	-0.24	0.30	-0.09
BDE-49	0.45	0.22	0.17	0.12	0.09	0.28
BDE-47	0.49	0.44	0.20	0.19	0.43	0.46
BDE-66	0.55	0.41	0.18	0.34	0.41	0.36
BDE-100	0.25	0.32	0.04	0.12	0.41	0.44
BDE-99	0.22	0.29	0.04	0.04	0.34	0.35
BDE-85	-0.23	-0.27	-0.67	0.08	0.13	0.05
BDE-154	0.39	0.48	0.18	0.54	0.38	0.45
BDE-153	0.25	0.19	-0.11	0.25	0.47	0.50
BDE-183	0.13	-0.05	0.14	-0.06	0.17	0.20
BDE-209	-0.01	-0.02	-0.22	0.03	0.18	-0.01
Shell Tickn.	-0.34	-0.27	0.21	-0.40	-0.46	-0.35
HBCD	-0.09	-0.38	0.24	-0.34	-0.49	-0.13
Me-TBBP-A	0.01	0.05	0.27	-0.32	-0.20	0.19

	CHB-26	CHB-40	CHB-41	CHB-44	CHB-50	CHB-62
CB-28	0.38	0.29	0.43	0.49	0.42	0.11
CB-31	0.49	0.32	0.48	0.69	0.55	0.46
CB-44	0.27	0.35	0.35	0.39	0.39	0.24
CB-49	0.39	0.45	0.46	0.53	0.49	0.41
CB-52	0.32	0.46	0.34	0.40	0.41	0.29
CB-99	0.43	0.30	0.45	0.59	0.46	0.25
CB-101	0.24	0.27	0.28	0.36	0.29	0.47
CB-105	0.41	0.31	0.48	0.58	0.44	0.27
CB-110	0.50	0.39	0.50	0.61	0.50	0.43
CB-118	0.41	0.27	0.44	0.58	0.44	0.30
CB-128	0.40	0.26	0.45	0.59	0.42	0.27
CB-138	0.44	0.31	0.41	0.60	0.43	0.35
CB-149	0.28	0.31	0.41	0.41	0.26	0.29
CB-151	0.24	0.20	0.33	0.11	0.01	0.07
CB-153	0.45	0.32	0.42	0.59	0.45	0.41
CB-156	0.48	0.36	0.45	0.63	0.48	0.46
CB-170	0.48	0.36	0.46	0.63	0.51	0.46
CB-180	0.48	0.41	0.45	0.62	0.51	0.51
CB-187	0.44	0.25	0.45	0.63	0.42	0.24
CB-194	0.47	0.35	0.46	0.64	0.52	0.52
CB-209	0.37	0.13	0.50	0.61	0.42	0.10
alfa-HCH	0.32	0.52	0.32	0.38	0.34	0.13
beta-HCH	0.31	0.49	0.25	0.28	0.25	0.28
gamma-HCH	0.17	0.20	0.19	0.09	0.06	-0.05
HCB	0.46	0.64	0.40	0.36	0.28	0.44
o'p-DDE	0.36	0.41	0.37	0.45	0.46	0.33
o'p-DDT	0.47	0.29	0.23	0.44	0.31	0.36
p'p'-DDD	-0.17	0.12	-0.17	0.08	0.15	0.02
p'p-DDE	0.44	0.34	0.51	0.64	0.50	0.02
p'p-DDT	0.51	0.37	0.58	0.70	0.56	0.58
CHB-26	1.00	0.76	0.92	0.91	0.89	0.61
CHB-40	0.76	1.00	0.66	0.58	0.54	0.58
CHB-41	0.92	0.66	1.00	0.91	0.91	0.61
CHB-44	0.91	0.58	0.91	1.00	0.89	0.60
CHB-50	0.89	0.54	0.91	0.89	1.00	0.54
CHB-62	0.61	0.58	0.61	0.60	0.54	1.00
oxychlordan	0.38	0.30	0.29	0.51	0.45	0.24
trans-chlordan	0.20	0.42	0.10	0.38	0.43	0.29
cis-chlordan	0.06	0.37	0.16	0.24	0.26	0.22
trans-nonachlor	0.60	0.46	0.63	0.76	0.74	0.50
cis-nonachlor	0.56	0.50	0.63	0.71	0.68	0.37
BDE-17	-0.04	0.57	0.10	0.16	0.16	-0.29
BDE-28	0.35	0.57	0.50	0.00	-0.47	0.45
BDE-49	0.04	0.13	0.34	0.19	0.11	0.55
BDE-47	0.42	0.25	0.59	0.56	0.55	0.38
BDE-66	0.02	0.32	0.41	0.08	0.17	0.36
BDE-100	0.25	0.01	0.50	0.44	0.42	0.19
BDE-99	0.26	0.00	0.50	0.44	0.40	0.20
BDE-85	-0.15	-0.25	0.21	0.09	0.10	0.08
BDE-154	0.11	0.18	0.13	0.51	0.40	0.50
BDE-153	0.10	-0.04	0.32	0.37	0.30	0.27
BDE-183	-0.07	-0.15	0.14	0.06	-0.03	-0.04
BDE-209	-0.08	-0.21	0.09	0.08	-0.01	-0.31
Shell Tickn.	-0.30	-0.22	-0.37	-0.41	-0.54	0.13
HBCD	0.10	0.20	-0.18	-0.08	-0.07	0.10
Me-TBBP-A	0.19	0.14	0.21	0.22	-0.03	0.26

	oxychlordan	trans-chlordan	cis-chlordan	trans-nonachlor	cis-nonachlor	BDE-17
CB-28	0.63	0.47	0.51	0.68	0.75	0.73
CB-31	0.84	0.67	0.49	0.89	0.88	0.78
CB-44	0.66	0.55	0.58	0.68	0.77	0.75
CB-49	0.66	0.55	0.48	0.72	0.77	0.72
CB-52	0.66	0.57	0.56	0.66	0.72	0.24
CB-99	0.85	0.62	0.52	0.85	0.86	0.59
CB-101	0.47	0.26	0.32	0.43	0.49	0.01
CB-105	0.80	0.61	0.53	0.82	0.86	0.72
CB-110	0.76	0.45	0.53	0.76	0.79	-0.09
CB-118	0.85	0.59	0.44	0.84	0.83	0.58
CB-128	0.80	0.50	0.48	0.82	0.83	0.83
CB-138	0.85	0.52	0.48	0.83	0.81	0.48
CB-149	0.55	0.38	0.42	0.61	0.74	0.14
CB-151	0.05	-0.42	-0.04	0.15	0.27	-0.89
CB-153	0.83	0.48	0.41	0.82	0.78	0.42
CB-156	0.82	0.52	0.42	0.82	0.78	0.52
CB-170	0.77	0.44	0.45	0.82	0.78	0.59
CB-180	0.76	0.46	0.46	0.80	0.76	0.44
CB-187	0.81	0.44	0.47	0.82	0.83	0.82
CB-194	0.68	0.36	0.38	0.76	0.71	0.48
CB-209	0.54	0.24	0.38	0.70	0.76	-0.43
alfa-HCH	0.66	0.70	0.68	0.57	0.67	0.80
beta-HCH	0.51	0.87	0.39	0.54	0.58	0.63
gamma-HCH	-0.02	-0.47	0.30	-0.07	0.01	0.43
HCB	0.56	0.73	0.56	0.63	0.69	-0.53
o'p-DDE	0.62	0.45	0.62	0.67	0.63	0.67
o'p-DDT	0.60	0.28	0.15	0.58	0.56	0.49
p'p'-DDD	0.43	0.59	0.55	0.41	0.46	0.48
p'p-DDE	0.41	0.38	0.18	0.58	0.66	0.19
p'p-DDT	0.44	0.18	0.20	0.61	0.63	-0.96
CHB-26	0.38	0.20	0.06	0.60	0.56	-0.04
CHB-40	0.30	0.42	0.37	0.46	0.50	0.57
CHB-41	0.29	0.10	0.16	0.63	0.63	0.10
CHB-44	0.51	0.38	0.24	0.76	0.71	0.16
CHB-50	0.45	0.43	0.26	0.74	0.68	0.16
CHB-62	0.24	0.29	0.22	0.50	0.37	-0.29
oxychlordan	1.00	0.74	0.56	0.86	0.83	0.53
trans-chlordan	0.74	1.00	0.64	0.72	0.74	0.06
cis-chlordan	0.56	0.64	1.00	0.61	0.74	0.57
trans-nonachlor	0.86	0.72	0.61	1.00	0.95	0.52
cis-nonachlor	0.83	0.74	0.74	0.95	1.00	0.72
BDE-17	0.53	0.06	0.57	0.52	0.72	1.00
BDE-28	-0.38	-0.31	0.58	-0.01	0.37	-0.02
BDE-49	0.43	0.42	0.35	0.62	0.58	0.81
BDE-47	0.59	0.11	0.41	0.77	0.76	0.64
BDE-66	0.48	0.44	0.57	0.59	0.69	0.92
BDE-100	0.43	-0.09	0.08	0.62	0.59	0.60
BDE-99	0.40	-0.20	0.08	0.58	0.55	0.65
BDE-85	-0.07	-0.29	0.00	0.13	0.09	1.00
BDE-154	0.66	0.53	0.37	0.66	0.63	0.73
BDE-153	0.33	0.01	0.02	0.50	0.50	-0.19
BDE-183	0.14	-0.48	0.34	0.14	0.29	0.01
BDE-209	0.17	-0.19	-0.02	0.17	0.20	0.18
SkalTyk	-0.38	-0.54	-0.27	-0.53	-0.51	0.60
HBCD	-0.28	0.22	0.21	-0.29	-0.33	0.87
Me-TBBP-A	0.00	-0.02	0.07	0.01	-0.01	0.97

	BDE-28	BDE-49	BDE-47	BDE-66	BDE-100	BDE-99
CB-28	0.56	0.41	0.75	0.58	0.61	0.61
CB-31	0.12	0.57	0.65	0.42	0.58	0.54
CB-44	0.39	0.42	0.65	0.73	0.56	0.50
CB-49	0.54	0.46	0.71	0.84	0.57	0.51
CB-52	0.60	0.31	0.53	0.84	0.36	0.28
CB-99	0.44	0.53	0.76	0.70	0.67	0.62
CB-101	0.41	0.45	0.47	0.73	0.39	0.34
CB-105	0.52	0.61	0.77	0.74	0.70	0.64
CB-110	0.38	0.46	0.72	0.71	0.55	0.52
CB-118	0.36	0.61	0.75	0.72	0.70	0.64
CB-128	0.52	0.60	0.78	0.66	0.73	0.67
CB-138	0.43	0.54	0.76	0.66	0.66	0.62
CB-149	0.80	0.56	0.64	0.75	0.62	0.53
CB-151	0.75	0.22	0.42	0.41	0.39	0.36
CB-153	0.40	0.60	0.77	0.61	0.69	0.64
CB-156	0.27	0.60	0.71	0.51	0.64	0.60
CB-170	0.36	0.58	0.74	0.47	0.66	0.62
CB-180	0.32	0.58	0.72	0.46	0.62	0.59
CB-187	0.59	0.49	0.76	0.60	0.69	0.64
CB-194	0.26	0.58	0.69	0.35	0.65	0.61
CB-209	0.60	0.45	0.73	0.48	0.74	0.68
alfa-HCH	0.40	0.27	0.39	0.69	0.26	0.19
beta-HCH	0.51	0.21	0.17	0.17	0.10	0.02
gamma-HCH	0.30	-0.02	0.12	0.40	0.00	0.06
HCB	0.83	0.45	0.49	0.55	0.25	0.22
o'p-DDE	0.03	0.22	0.44	0.41	0.32	0.29
o'p-DDT	0.25	0.17	0.20	0.18	0.04	0.04
p'p'-DDD	-0.24	0.12	0.19	0.34	0.12	0.04
p'p-DDE	0.30	0.09	0.43	0.41	0.41	0.34
p'p-DDT	-0.09	0.28	0.46	0.36	0.44	0.35
CHB-26	0.35	0.04	0.42	0.02	0.25	0.26
CHB-40	0.57	0.13	0.25	0.32	0.01	0.00
CHB-41	0.50	0.34	0.59	0.41	0.50	0.50
CHB-44	0.00	0.19	0.56	0.08	0.44	0.44
CHB-50	-0.47	0.11	0.55	0.17	0.42	0.40
CHB-62	0.45	0.55	0.38	0.36	0.19	0.20
oxychlordan	-0.38	0.43	0.59	0.48	0.43	0.40
trans-chlordan	-0.31	0.42	0.11	0.44	-0.09	-0.20
cis-chlordan	0.58	0.35	0.41	0.57	0.08	0.08
trans-nonachlor	-0.01	0.62	0.77	0.59	0.62	0.58
cis-nonachlor	0.37	0.58	0.76	0.69	0.59	0.55
BDE-17	-0.02	0.81	0.64	0.92	0.60	0.65
BDE-28	1.00	0.66	0.69	0.66	0.45	0.40
BDE-49	0.66	1.00	0.69	0.68	0.72	0.66
BDE-47	0.69	0.69	1.00	0.68	0.89	0.91
BDE-66	0.66	0.68	0.68	1.00	0.48	0.39
BDE-100	0.45	0.72	0.89	0.48	1.00	0.98
BDE-99	0.40	0.66	0.91	0.39	0.98	1.00
BDE-85	0.80	0.62	0.66	0.27	0.76	0.82
BDE-154	-0.24	0.54	0.53	0.35	0.50	0.44
BDE-153	0.10	0.68	0.71	0.46	0.85	0.79
BDE-183	0.84	0.54	0.49	0.60	0.51	0.53
BDE-209	0.12	0.29	0.40	0.06	0.55	0.54
Shell Tickn.	0.68	-0.18	-0.34	-0.15	-0.29	-0.25
HBCD	-0.97	-0.12	-0.36	-0.30	-0.38	-0.36
Me-TBBP-A	0.17	0.29	-0.05	-0.24	0.10	0.09

	BDE-85	BDE-154	BDE-153	BDE-183	BDE-209	Shell Tickn.
CB-28	-0.21	0.47	0.52	0.52	0.28	-0.15
CB-31	0.08	0.69	0.55	0.24	0.20	-0.37
CB-44	-0.03	0.56	0.52	0.43	0.27	-0.26
CB-49	-0.07	0.53	0.43	0.21	0.05	-0.17
CB-52	0.07	0.53	0.36	0.17	0.06	-0.32
CB-99	0.03	0.70	0.58	0.32	0.34	-0.33
CB-101	0.24	0.36	0.38	0.28	-0.02	-0.10
CB-105	0.06	0.71	0.65	0.40	0.38	-0.34
CB-110	-0.03	0.54	0.44	0.33	0.08	-0.25
CB-118	0.12	0.75	0.65	0.33	0.38	-0.33
CB-128	0.20	0.76	0.68	0.37	0.43	-0.34
CB-138	0.17	0.76	0.60	0.30	0.34	-0.33
CB-149	0.14	0.49	0.63	0.48	0.30	-0.20
CB-151	0.17	0.11	0.36	0.40	0.36	-0.10
CB-153	0.27	0.79	0.64	0.32	0.37	-0.30
CB-156	0.25	0.80	0.60	0.23	0.34	-0.36
CB-170	0.36	0.81	0.63	0.28	0.38	-0.37
CB-180	0.37	0.81	0.60	0.26	0.34	-0.36
CB-187	0.22	0.73	0.64	0.36	0.42	-0.34
CB-194	0.46	0.81	0.64	0.27	0.37	-0.36
CB-209	0.38	0.58	0.76	0.48	0.54	-0.40
alfa-HCH	-0.05	0.48	0.19	0.04	0.13	-0.35
beta-HCH	-0.48	0.42	0.17	-0.07	0.03	-0.32
gamma-HCH	-0.18	-0.10	-0.06	0.49	0.09	0.40
HCB	-0.23	0.39	0.25	0.13	-0.01	-0.34
o'p-DDE	-0.27	0.48	0.19	-0.05	-0.02	-0.27
o'p-DDT	-0.67	0.18	-0.11	0.14	-0.22	0.21
p'p'-DDD	0.08	0.54	0.25	-0.06	0.03	-0.40
p'p-DDE	0.13	0.38	0.47	0.17	0.18	-0.46
p'p-DDT	0.05	0.45	0.50	0.20	-0.01	-0.35
CHB-26	-0.15	0.11	0.10	-0.07	-0.08	-0.30
CHB-40	-0.25	0.18	-0.04	-0.15	-0.21	-0.22
CHB-41	0.21	0.13	0.32	0.14	0.09	-0.37
CHB-44	0.09	0.51	0.37	0.06	0.08	-0.41
CHB-50	0.10	0.40	0.30	-0.03	-0.01	-0.54
CHB-62	0.08	0.50	0.27	-0.04	-0.31	0.13
oxychlordan	-0.07	0.66	0.33	0.14	0.17	-0.38
trans-chlordan	-0.29	0.53	0.01	-0.48	-0.19	-0.54
cis-chlordan	0.00	0.37	0.02	0.34	-0.02	-0.27
trans-nonachlor	0.13	0.66	0.50	0.14	0.17	-0.53
cis-nonachlor	0.09	0.63	0.50	0.29	0.20	-0.51
BDE-17	1.00	0.73	-0.19	0.01	0.18	0.60
BDE-28	0.80	-0.24	0.10	0.84	0.12	0.68
BDE-49	0.62	0.54	0.68	0.54	0.29	-0.18
BDE-47	0.66	0.53	0.71	0.49	0.40	-0.34
BDE-66	0.27	0.35	0.46	0.60	0.06	-0.15
BDE-100	0.76	0.50	0.85	0.51	0.55	-0.29
BDE-99	0.82	0.44	0.79	0.53	0.54	-0.25
BDE-85	1.00	0.48	0.78	0.39	0.60	-0.38
BDE-154	0.48	1.00	0.68	0.24	0.38	-0.38
BDE-153	0.78	0.68	1.00	0.59	0.63	-0.35
BDE-183	0.39	0.24	0.59	1.00	0.54	0.13
BDE-209	0.60	0.38	0.63	0.54	1.00	-0.15
Shell Tickn.	-0.38	-0.38	-0.35	0.13	-0.15	1.00
HBCD	-0.02	-0.25	-0.35	-0.56	-0.28	-0.22
Me-TBBP-A	-0.39	0.13	0.06	0.04	0.09	0.08

	HBCD	Me-TBBP-A
CB-28	-0.18	0.12
CB-31	-0.23	0.18
CB-44	-0.26	-0.02
CB-49	-0.47	-0.17
CB-52	-0.57	-0.38
CB-99	-0.24	0.18
CB-101	-0.37	-0.24
CB-105	-0.26	0.23
CB-110	-0.33	0.09
CB-118	-0.28	0.20
CB-128	-0.24	0.26
CB-138	-0.18	0.20
CB-149	-0.34	-0.03
CB-151	-0.43	-0.05
CB-153	-0.22	0.19
CB-156	-0.10	0.32
CB-170	-0.16	0.25
CB-180	-0.15	0.23
CB-187	-0.27	0.17
CB-194	-0.19	0.28
CB-209	-0.40	0.07
alfa-HCH	-0.41	-0.09
beta-HCH	-0.04	0.22
gamma-HCH	0.20	0.32
HCB	-0.09	0.01
o'p-DDE	-0.38	0.05
o'p-DDT	0.24	0.27
p'p'-DDD	-0.34	-0.32
p'p-DDE	-0.49	-0.20
p'p-DDT	-0.13	0.19
CHB-26	0.10	0.19
CHB-40	0.20	0.14
CHB-41	-0.18	0.21
CHB-44	-0.08	0.22
CHB-50	-0.07	-0.03
CHB-62	0.10	0.26
oxychlordan	-0.28	0.00
trans-chlordan	0.22	-0.02
cis-chlordan	0.21	0.07
trans-nonachlor	-0.29	0.01
cis-nonachlor	-0.33	-0.01
BDE-17	0.87	0.97
BDE-28	-0.97	0.17
BDE-49	-0.12	0.29
BDE-47	-0.36	-0.05
BDE-66	-0.30	-0.24
BDE-100	-0.38	0.10
BDE-99	-0.36	0.09
BDE-85	-0.02	-0.39
BDE-154	-0.25	0.13
BDE-153	-0.35	0.06
BDE-183	-0.56	0.04
BDE-209	-0.28	0.09
SkalTyk	-0.22	0.08
HBCD	1.00	0.67
Me-TBBP-A	0.67	1.00

Appendix 7

Multivariate data analysis

Principal Component Analysis (PCA) of chemical profile data from eggs collected in the years 1986 to 2003

Brief description of data

A total of 54 compounds have been measured for in each of 42 samples of eggs. The chemical profile data consists of 21 PCBs, HCB, 3 HCHs, 5 chlorinated pesticides, HBCD and Me-TBBP-A, 6 toxaphene congeners, 5 chlordanes and 12 PBDEs. In the following data analysis the chemical profile data make up 54 *explanatory variables*. The *objects* are the individual egg samples, which are denoted by the year of sampling. The 42 eggs and the 54 compounds form a 42X54 data matrix (X matrix). The concentrations measured in each egg in relation to the lipid content were used as basis. The values were ln-transformed in order to improve the approximation of normality. The raw data set can be seen in Appendix 3.

Brief description of the PCA methodology

To get an overview of the correlation patterns between compounds as well as information related to the chemical profile measurements of concentrations in the individual egg, a principal component analysis (PCA) was performed. The PCA approach is an attempt to explain the structure of the variation in the X-data by projection of the original multi dimensional variables into a lower dimensional hyperspace (cf. Figure 1) spanned by few numbers of dimensions denoted principal components.

In the actual case, 42 eggs are analysed for 54 compounds. Each egg defines a data point so the number of compounds is equivalent to the number of dimensions. This forms a 42x54 X-data matrix of single number elements ($x_{n,k}$), where n is the specific egg number and k is the specific compound number. The $x_{n,k}$ value could be a ln-transformed lipid concentration value taken form Appendix 3, however, in order to avoid dominance from trivial relations the following preprocessing of data was performed prior to PCA analysis. Firstly a scaling of every compound is done using the mean of all eggs in order to eliminate the trivial fact that some compounds are found in higher concentrations than other compounds. In this way all $x_{n,k}$ values will have values around unity. Secondly the fact that the variability between eggs for some compounds is larger than for others is defined as trivial in this investigation. So, every $x_{n,k}$ value is scaled using the standard deviation for the specific compound. This scaling will force the standard deviation to be unity for every compound.

The final data matrix is decomposed into the following bilinear form:

$$\mathbf{X} = \sum_{i=1}^{I} \left(\mathbf{t}_{i,n} * \mathbf{p}_{i,k}^{\mathrm{T}} \right) + \mathbf{E}_i$$

where *I* is the number of dimension in the hyperspace, $\mathbf{t}_{i,n}$ is the score vectors of the individual eggs in the hyper-planes spanned by the a number of principal components described by the loading vectors $\mathbf{p}_{i,k}$. The score vector is a linear combination of columns of **X** such that $\boldsymbol{t}$ is maximised. The $\boldsymbol{t}$ vector is the same for all the compounds in a specific direction, i.e. specific for the eggs. The corresponding $\boldsymbol{p}$ vector can be understood as expressing the orientation of the obtained model plane inside the k-dimensional variable space. The direction of a given principal component, $\boldsymbol{p}_{i,k}$ in relation to the original variables x_1, x_2, and x_3 is calculated as the cosine of the angles between the three original variables and $\boldsymbol{p}_{i,k}$. The calculated values indicate how the original variables "load" into (=contribute to) $\boldsymbol{p}_{i,k}$. So this equation describes how the n'th egg in the i´th dimension ($\boldsymbol{t}_{i,n}$) is related the k chemicals of the i'th principal component. The projection of X onto a hyper-plane is illustrated in Figure 1 below:

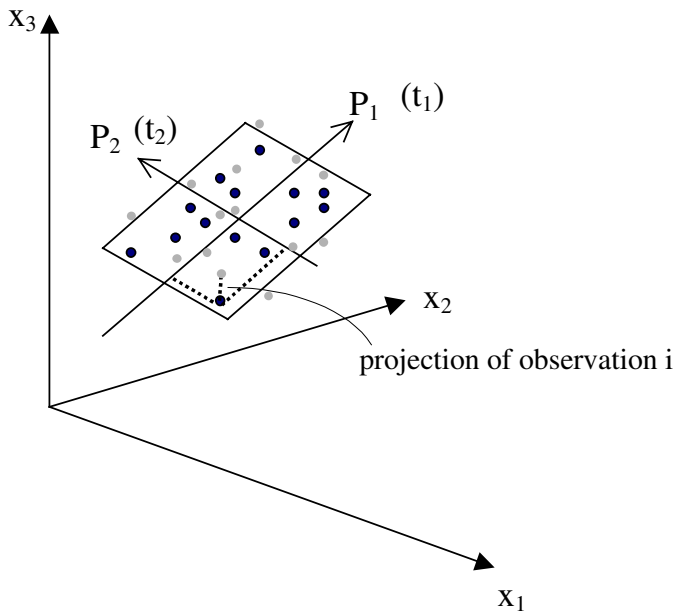


Figure 1. Geometrical representation of a two dimensional PC-model. The two PCs define a plane that can be seen as a two dimensional "window" (=hyperplane) into the multivariate chemical profile descriptor space.

PCA is a projection method that provides an approximation of the matrix X in terms of two smaller matrices **T** and **P** which may be represented as in Figure 2:

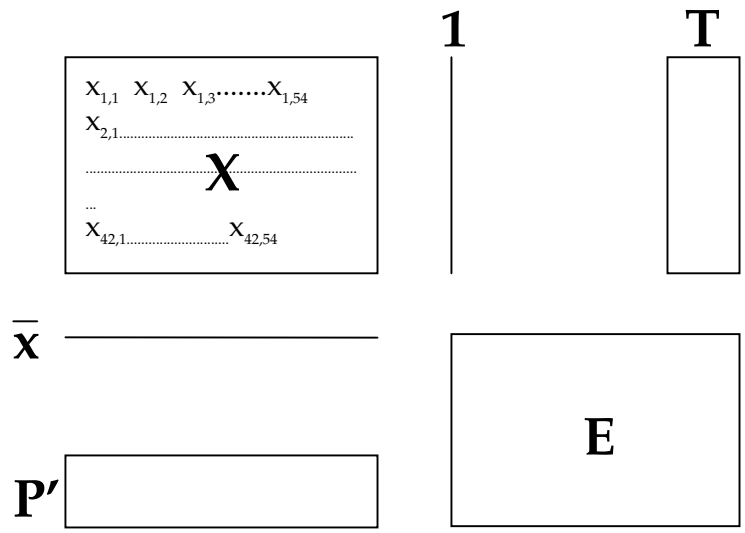


Figure 2. Matrix representation of the decomposition of **X,** (**X**=**1***$\bar{\mathbf{x}}$ + **T***$\mathbf{P}'$ + **E**). **X** is the original 42x54 data matrix, **1** is here a column-vector with element one in all positions, $\bar{\mathbf{x}}$ is a row-vector comprising the average object value for each variable and **E** is the residual matrix, i.e. the part of the data that is not explained by the PC model. The matrices **T** and $\mathbf{P}'$ extract the essential information and patterns from **X**.

The latent variables are orthogonal and are linear combinations of the original variables, and can thus be regarded as the principal properties of the characterized subsystem. By plotting the columns in **T**, a picture of the dominant "object pattern" of X is obtained and, analogously, plotting the rows of $\mathbf{P}'$ shows the complementary "variable pattern". Further information on the mathematical and geometrical properties of PCA and PLS can be found in Höskuldsson, A. (1996) and Eriksson, L. (1999), respectively.

In this case study, objects are the egg samples denoted by the year of sampling, whereas variables are the individual compounds measured for in each egg sample. Thus, in the following analysis, plotting the columns in **T** will provide information regarding dominant egg sample patterns, and plotting the rows of $\mathbf{P}'$ will show the complementary chemical compound pattern.

Data analysis

The purpose is: 1) to get a picture of the correlation patterns between compounds and/or chemical classes and 2) to see if there are interesting patterns in egg samples, e.g. any visible time trends in sample years explained by the chemical contamination profile variables.

Variables include all the above mentioned chemicals. Objects are the years of the sampling ranging from 1986 to 2003 (exclusive year 1993, 1996 and 1997 where no measurements were made). Each year occurs, as many times as a sample has been measured within that year. In the following PCA plots, each egg sample has 1) a sample-number, 2) a ring number and 3) a batch-number. The numbers refers to 1) the exact time, place of sampling, 2) the mother bird of the eggs and 3) the quality assurance of the laboratory chemical profile analysis and quantification.

In Figure 3 a score plot (corresponding to the "window" or the two-dimensional hyperplane in Figure 1) spanned by $\mathbf{t}_2$ and $\mathbf{t}_1$ is shown. $\mathbf{t}_2$ and $\mathbf{t}_1$ represent the concentrations in eggs. The objects are assigned by the year of sampling followed by a letter corresponding to individual batch of analysis and a number corresponding to different ring numbers.

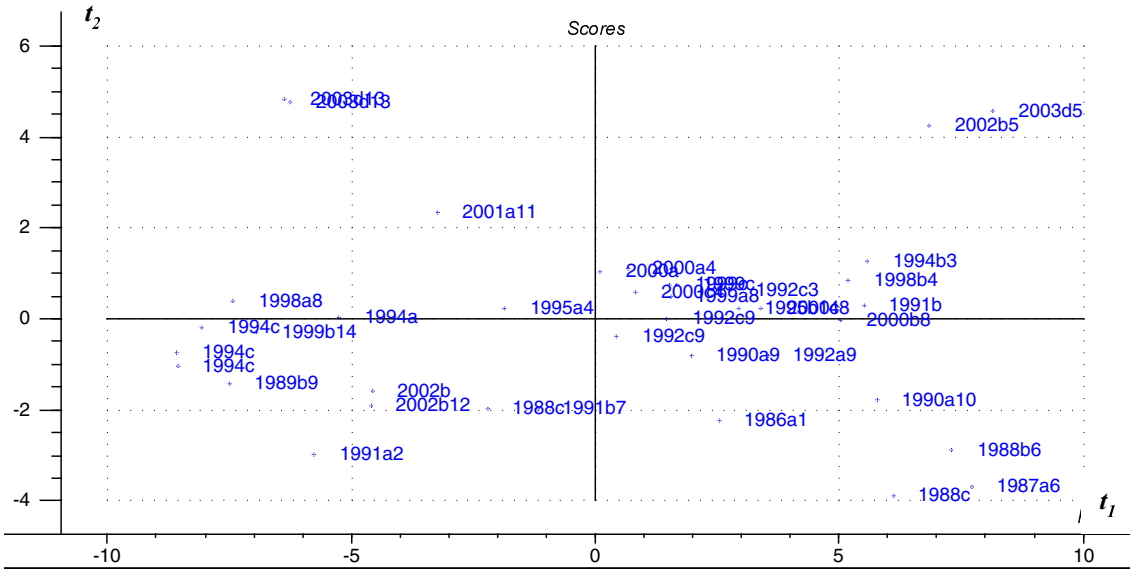


Figure 3. Score plot of t_2 versus t_1 , showing the "scores" or the position of the objects in the hyperplane. By looking at the score plot it seems difficult to identify any patterns in the egg sample scores indicating the presence of a time trend. There may be a tendency for early years to have negative score values in t_2 and of late years, to have positive score values in t_2.

The majority of the egg samples have positive score values in t_1. For t_2 it can be seen that late years are related to higher positive numbers (i.e. score values) while early years more relates to negative score values. This indicates a time trend being presents in t_2; however, this component only explains 9 % of the total variation. The time trend is thus relatively weak as concluded also in the following PLS-regressions.

Some egg samples have high leverage, i.e. high influence on the estimated planes and may be outliers. These samples are, e.g., the 2002 and 2003 samples having high positive score values in t_2 and high negative and positive score values in t_1, respectively. At this point no samples are excluded, as we are interested in identifying original variables responsible for the inhomogeneous spanning of the score plot.

The complementary loading plot in Figure 4 shows patterns in inter-correlation between compound variables, as well as importance of the original variables in the principal components p_1 and p_2. The first principal component, p_1, explains 53 % of the variation in the X-space, while p_2 explains 9 % of the X-variance. Samples with high positive score values in t_1 have high contents of chemicals with high positive loadings in p_1, whereas samples with high negative score values have lowest contamination with chemical variables with high positive

loading in p_1. There is a high degree of intercorrelation between chemicals in the principal component p_1. This is seen in Figure 4, where the majority of chemical variables have loading values above 0.1 in p_1. For these chemical variables the contamination level is increasing in egg samples going from the left to the right in the score plot in Figure 3.

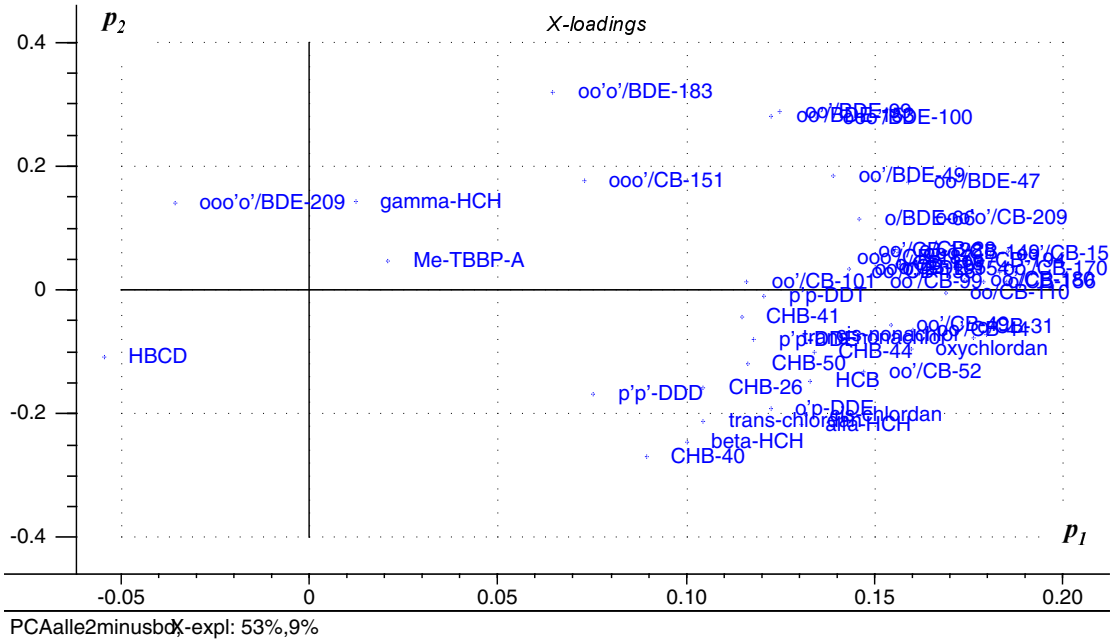


Figure 4. Loading plot of p_1 versus p_2 showing visible groupings of intercorrelated variables. The first principal component, p_1, explains 50 % of the variance, whereas p_2 explains 9 % of the variance in the X-space. The tendency for a time trend to be present in t_2 explained by p_2 suggests that the BDEs are the only group of chemicals showing an increasing time trend, together with CB-151. Most of the PCBs show an insignificant or negative time trend (having close to zero loading values in p_2). The chlordanes, toxaphene congeners and DDT and degradation products have negative loadings of varying importance in t_2. o´,p-DDE, trans- and cis-chlodane, alfa- and beta-HCH and CBH-40 have highest negative loading in t_2. BDE-17, -28, -85, o,p´-DDT, trans-chlordane and CHB-62 have a frequency of measurements below the detection limit which is above 50% and have, with exception of trans-chlordane, therefore been excluded from the analysis.

The BDEs, are positioned in the upper right of the loading plot having high positive loadings in both p_1 and p_2. The chlordanes are overlapping with the PCBs having high positive loading in p_1, while the toxaphene congeners, CHB-26, -44 and -50 have less positive loading in p_1 together with p,p'-DDT, the DDEs, beta and alfa-HCH, two chlordanes and CB-101. The majority of chemicals within the compound classed are grouped together. The loading values of the PCBs, the chlordanes and the toxaphene congeners in the direction of p_2 is overlapping to some extent, while the BDEs clearly have high positive loading values separating them from the remaining compound classes in p_2. Few compounds have low or negative loading in p_1. The egg samples with highest negative score values in t_1 seems to differ from egg samples with high positive score values in t_1 by having a higher contamination by the few compound with low or negative loading values in p_1.

Due to the tendency for clustering of chemical variables into chemical classes, we need to analyse the individual chemical classes in separate models to obtain more information of the relative importance of single compounds within each compound group. Furthermore we want to investigate the presence of any correlation between chemical profile contamination in individual egg sample and the measured eggshell thickness. To investigate if the chemical profile variable can explain the variation in eggshell thickness Partial Least Square Regression (PLS-Regression) were performed on several data subsets.

Very shortly PLS is a projection method as PCA, but in this case the decomposition of the X-matrix is done so that T and P extract information from X, that are relevant for explaining the variation in Y, the eggshell thickness.

Evaluation of model performances of Partial Least Square Regression (PLS-R) models for estimating the eggshell thickness

In the PLS-Regressions both X- and Y-variables are weighted so that only the relative differences among the variance of the X-variables and the relative differences among the variance of the Y-variables influence the model. The weighting by division using the standard deviation, i.e. multiplying by $1/S_{dev}$, is called standardization and is used to give all the variables the same variance. In this way, all the variables are given the same chance to influence the estimation of the components. Furthermore data have been centred, i.e. the mean of each variable has been subtracted each x-value, so that every variable has mean zero and variance equal to unity.

In the first PLS-Regression all chemical variables have been included. This was done in order to be able to compare the percent variance of X which is usable for explaining the variation in Y, the eggshell thickness, compared to the X-variance explained in the PCA (cf. Figure 3 and 4), which were 59 %. The results of the PLS-regression based on all chemicals are shown in Figure 5.

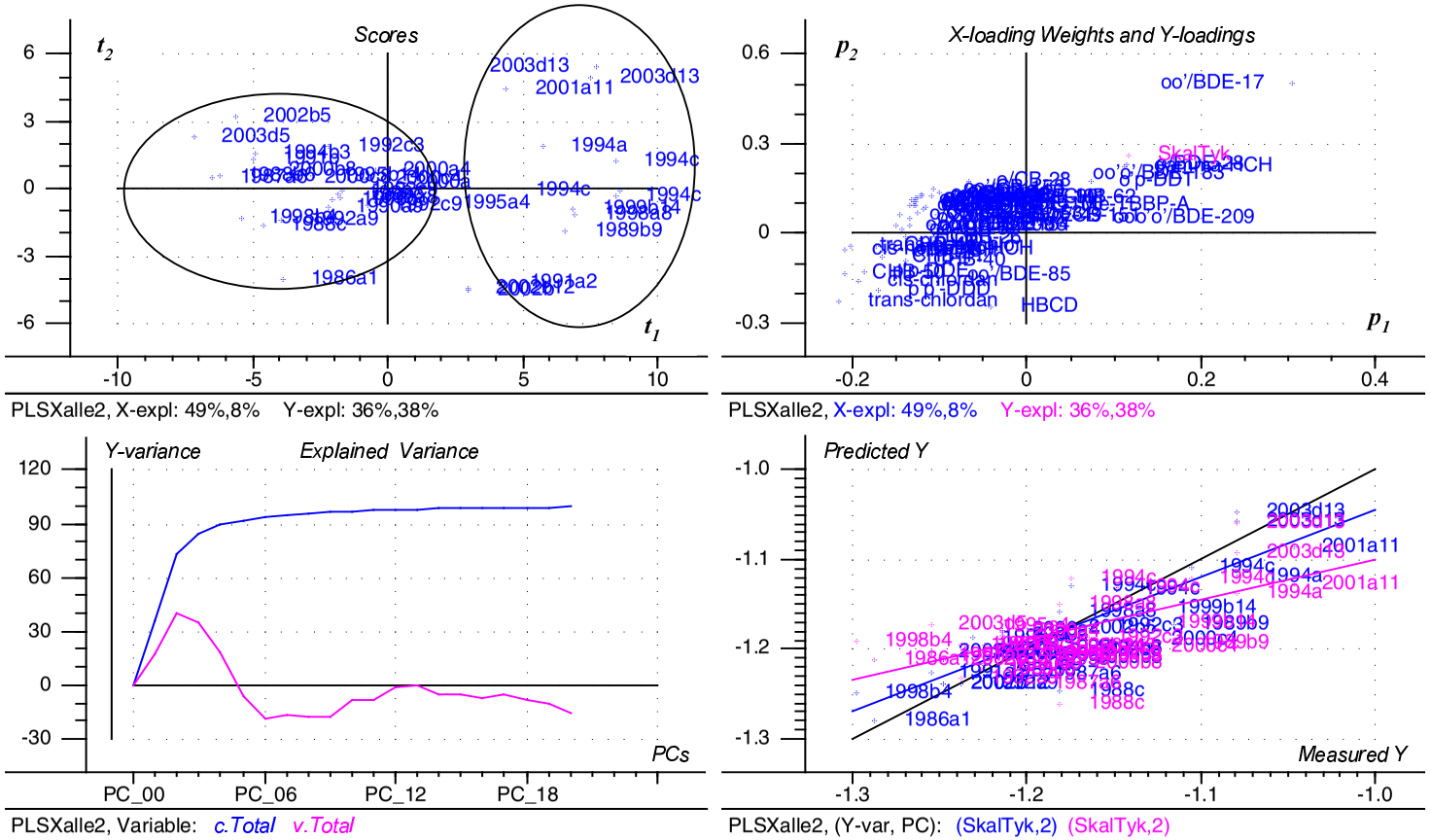


Figure 5. In the upper left corner, the score plot analogous to Figure 3, is shown except that the X-variance has been extracted to increase explainability with respect to the eggshell thickness. The appearance of two clusters in the score plot in the PCA (cf. Figure 3) are also observed in the PLS score plot. To the upper right the X-loading weights and Y-loading is shown. The lower right figure shows predicted versus measured eggshell thickness values; the blue numbers and regression line are predicted versus measured eggshell thickness by calibration of the model, whereas the pink numbers are the estimated versus predicted eggshell thickness values by cross-validation using the leave-one-out method. Lastly the black line is the target line of R^2 equal to one (Höskuldsson, 1996). BDE-17, -85 and trans-chlordane were not excluded from the model, even though there was a high frequency of missing data. Their inclusion does not change the correlation patterns to any significant degree. The model is a two-component model as seen from the explained Y-variance by cross-validation, which decreases by inclusion of a third PC as seen from the lower left figure (pink curve).

Figure 5 does not allow for details in variable intercorrelations and patterns in score values to be discovered. But overall it is easy to see the appearance of two groups in the score plot of t_2 versus t_1. The group to the left has relatively higher contamination with the majority of chemical variables having negative loading weights in the upper right loading weight plot, which are all inversely correlated to the eggshell thickness. Few chemical compound variables have positive loading weights and the most extreme example is BDE-17, which is non detectable in many egg samples. The egg samples with positive score values are identified as having a lower contamination level compared to the samples positioned to the left in the score plot. The compound variables with high negative loading weights in p_1 and p_2 are the chemicals for which the inverse correlation to the eggshell thickness is most pronounced. These chemicals are trans- and cis-chlordane, the toxaphene congener CHB-50, p,p´-DDD and p,p´-DDE. The mother compound p,p´-DDT is also present in the third quadrant but has only a slightly negative loading weight in p_2. This is opposite to the o,p-configurations of DDT and its degradation products, which have positive loading weight in p_2 and in the case of o,p-DDT also in p_1 (cf. Appendix 8 concerning in the influence on molecular flexibility and the degree of inverse relation between exposure concentration and eggshell thickness). Toxaphene congeners, chlordanes in addition to HCB, alpha- and beta-HCH and p,p´-configurations of DDT and its degradations products dominate the third quadrant, so they have the highest degree of inverse relation to the eggshell thickness in both p_1 and p_2.

The appearance of two groups in the score plot of t_2 versus t_1 is also seen from the score plot in the PCA shown in Figure 3, even though the grouping is less distinct. The presence of two distinct groups in the score plot reveals that two different regression models are needed for the two groups.

For the sake of comparison and continuity, two strategies were chosen. One is that the clustering of objects into two distinct clusters are due to variables of low or no explainability regarding eggshell thickness, i.e. the groups are eliminated by eliminating bad descriptors and outliers. The second strategy is to accept the presence of two groups and use different regression models for each of the groups. PLS-Regressions were performed on three sets of egg samples, all objects, the right cluster and the left cluster of objects. Furthermore, PLS-Regressions were performed using all chemical variables, all chemicals variables excluding one chemical class at a time and using a single chemical class as original explanatory variables at a time.

The results of PLS-R models, ignoring the presence of two egg sample groups in the score plot (Figure 5, upper left plot), are shown in Table 1 to 3. The results of PLS-R models based on the left side group of objects are given in Table 4 to 6, and model results based on the right side egg sample group is given in Table 7 to 9.

Summary of model performance and results of PLS-Regressions based on all samples

Table 1. Model performance parameters for models based on all egg samples.

model*	n	n PC's	q^2	r^2	RMSEP	RMSEC
PLS_all_without_BDEs	39	2	0,445	0,769	0,0584	0,0403
PLS_all_without Toxaphenes	39	4	0,527	0,914	0,0554	0,0256
PLS_all_without PCBs	39	1	0,448	0,592	0,057	0,0509
PLS_all_without HBCD and Me-TBBP-A	39	2	0,522	0,803	0,0547	0,0376
PLS_all_without Chlordanes	39	7	0,612	0,965	0,0512	0,0167
PLS_all	37	4	0,611	0,920	0,0486	0,0235
PLS_Toxaphenes	35	1	0,602	0,671	0,0444	0,041
PLS_PCB	38	1	0,288	0,441	0,0618	0,0573
PLS_Chlordanes	36	1	0,599	0,673	0,0486	0,0448
PLS_BDE	32	3	0,782	0,892	0,0367	0,0261

*n is the number of samples, n PC is the number of principal components included in the model, r^2 is the correlation coefficient which expresses the fraction of the variance in the eggshell thickness explained by the models, q^2 the cross-validated correlation coefficient, which expresses the fraction of predicted variance according to the leave-one-out method (Höskuldsson, 1996; CAMO ASA, 1998). For each of the left-out eggshell thickness measurements, predictions are made, and the root mean square error of predictions, RMSEP, is calculated through the expression:

$$RMSEP = \sqrt{\frac{\sum (y_i - \hat{y}_i)^2}{I}}$$

Both the root mean square error of calibration (RMSEC) and prediction (RMSEP) are given in Table 1 above.

There are several stopping-criteria that one can use for determining how many principal components that should be included in the models. One way is to compare change in the correlation coefficients r^2 and q^2 as a function of the number of principal components. The correlation coefficients of predicted versus measured eggshell thickness values, i.e. based on the calibrated (r^2) and the cross-validated (q^2) models, display entirely different behaviour as function of increasing model complexity, i.e. number of PCs. The calibration correlation coefficient, r^2, is inflationary and approaches unity as model complexity increases. The goodness of predictions, q^2, is not inflationary and will not approach one with increasing complexity. Another stopping- criteria is to look at the explanatory capacity of the calibrated and cross-validated principal components, respectively, as illustrated in Figure 5; the lower left Figure. In Figure 6, the r^2 and q^2 as function of the number of PC's for the models, given in Table 1, is shown.

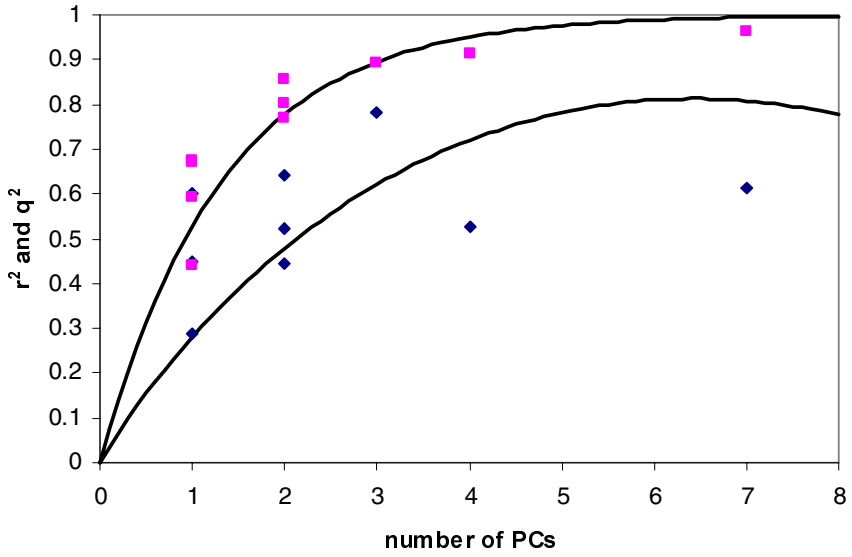


Figure 6. The upper points represent the r^2 values and the lower points the q^2 values as function of number of principal components in the individual models.

An optimal balance between fit and prediction ability is obtained by increasing the number of PCs in the model until q^2 begins to decrease or the residual Y-variance by validation increases. Therefore, the robustness of the models is reflected in the difference between r^2 and q^2. This difference is highest for the PLS models based on all chemicals leaving one chemical class out in each of the models. The robustness is highest for the model based on all chemicals except the PCBs (PLS_all_without PCBs), decreasing as follows: PLS_all, PLS_allwithout HBCD and Me-TBBP-A, PLS_all_without_BDEs, PLS_all_without Chlordanes, PLS_all_without Toxaphenes. The model performance data of the PLS models based on one chemical class only shows that the PLS- Regression based on the PCBs alone has lowest robustness, whereas the remaining three models have higher robustness in terms of the r^2-q^2 difference. The PLS model based on the BDEs alone has highest r^2 and q^2 and lowest root mean square of error on calibration, RMSEC, as well as predictions, RMSEP, compared to the other PLS models based on a single chemical class as original explanatory variables.

Table 2. Explained variance in the first two principal components and total explained variance of the individual models based on all samples.

	Explained X- and Y-variance in PC1, PC2 and total											
	X-variance by calibration			*X-variance by cross-validation*			*Y-variance by calibration*			*Y-variance by validation*		
Model*	PC_1	PC_2	$\sum PC_{cal}$	PC_1	PC_2	$\sum PC_{val}$	PC_1	PC_2	$\sum PC_{cal}$	PC_1	PC_2	$\sum PC_{val}$
PLS_all_without_BDEs	55	6	61	50	3	52	24	35	59	16	7	22
PLS_all_without Toxaphenes	51	7	70	47	3	56	24	37	84	20	4	29
PLS_all_without PCBs	38		38	25		25	35		35	19		19
PLS_all_without HBCD and Me-TBBP-A	50	7	57	44	3	48	27	38	64	22	10	32
PLS_all_without Chlordanes	48	7	78	40	2	53	26	39	93	16	5	41
PLS_all	52	8	68	46	4	52	34	28	84	26	7	38
PLS_Toxaphenes	76		76	69		69	45		45	42		42
PLS_PCB	62		62	57		57	19		19	11		11
PLS_Chlordanes	79		79	69		69	45		45	42		42
PLS_BDE	33	34	74	20	29	50	52	17	80	14	34	60

**PC1, PC2 and ∑PCcal under "X-variance by calibration" is the percent X-variance, (= t_1, t_2 and total X-variance) used for explaining the variation in Y by calibration*

PC1, PC2 and ∑PCval under "X-variance by validation" is the percent X-variance, (= t_1, t_2 and total X-variance) used for explaining the variation in Y by validation

PC1, PC2 and ∑Pccal under "Y-variance by calibration" is the percent Y-variance, (= p_1, p_2 and total Y-variance) explained by t_1, t_2 and percent X-variance used for explaining Y in total – by calibration

PC1, PC2 and ∑PCval under "Y-variance by validation" is the percent Y-variance, (=p_1, p_2 and total Y-variance) explained by t_1, t_2 and percent X-variance used for explaining Y in total – by validation

As seen from column three under the "Y-variance by calibration" the total explained Y-variance increases with increasing model complexity, i.e. number of latent variables or PCs, included in the models. However, the explained Y-variance by validation reveals what is also seen from Figure 6 that the model robustness decreases as well. The best model at this stage seems to be the PLS-R model based on the BDEs alone, which uses a total of 74 % of the X-variance to explain 80 % of the Y-variance.

To verify any presence of time trend, the score values of the PLS regression models were used as explanatory variables in simple linear regressions against year and eggshell thickness respectively. The results are given below.

Table 3. Results on Linear Regression of Principal Components as function of year and eggshell, respectively.

	PCs as function of year						*PCs as function of eggshell thickness*					
	t_1			t_2			t_1			T_2		
Model	slope	R^2	P(slope)	slope	R^2	P(slope)	slope	R^2	P(slope)	slope	R^2	P(slope)
LR_all_without_BDEs	0,27	0,09	0,98	0,06	0,03	0,88	34,54	0,24	1,00	15,47	0,39	1,00
LR_all_without Toxaphenes	0,17	0,03	0,87	0,08	0,04	0,91	37,49	0,24	1,00	18,38	0,39	1,00
LR_all_without PCBs	0,05	0,01	0,71				30,04	0,35	1,00			
LR_all_without HBCD and Me-TBBP-A	0,19	0,04	0,89	0,07	0,03	0,88	40,20	0,27	1,00	19,05	0,41	1,00
LR_all_without Chlordanes	0,13	0,02	0,82	0,05	0,02	0,82	36,96	0,26	1,00	19,42	0,43	1,00
LR_all	0,23	0,06	0,93	0,15	0,15	0,99	47,36	0,34	1,00	19,27	0,33	1,00
LR_Toxaphenes	0,09	0,05	0,91				24,92	0,45	1,00			
LR_PCB	0,19	0,06	0,93				27,05	0,19	1,00			
LR_Chlordanes	141,8	0,12	0,99				22200	0,45	1,00			
LR_BDE	-6,87	0,00	0,54	148,78	0,13	0,99	20534	0,52	1,00	15261	0,20	1,00

To verify if the presence of a time trend is inherently related to eggshell thickness, the principal components of the PLS-R models were fitted to year of sampling as well as eggshell thickness. The regression of the principal components as function of year and eggshell thickness, respectively, shows that there is a stronger correlation to the eggshell thickness compared to the year of sampling, i.e. Table 3 shows a relatively weak temporal influence but a more strong relation to the eggshell thickness. The correlation between sample score values and eggshell thickness is strongest for the model based on the BDEs alone, i.e. having a R^2 value of 0,52. In this model all of the BDEs are inversely related to the eggshell thickness, except for BDE-28, -66, -183 and -209, which are the compounds also showing positive or insignificant Pearson´s correlation coefficient when correlated one-by-one to the eggshell thickness (cf. Appendix 6). No egg sample score values in $\boldsymbol{t}_1$ or $\boldsymbol{t}_2$ as function of time is significant (cf. the R^2 values given in Table 3, column 3 and 6).

The regression of PCA principal components, which is not optimized with regard to eggshell thickness, does not improve the correlation to year of sampling.

Summary of model performance and results of PLS-Regressions based on the left side samples in the score plot in Figure 5

Table 4. Model performance parameters for models based on the left side egg samples.

model*	n	n PC's	q^2	r^2	RMSEP	RMSEC
PLS_all_without_BDEs	20	5	0,433	0,962	0,033	0,01
PLS_all_without Toxaphenes	20	1	0,218	0,581	0,023	0,019
PLS_all_without PCBs	18	2	0,542	0,863	0,019	0,011
PLS_all_without HBCD and Me-TBBP-A	17	4	0,57	0,934	0,019	0,008
PLS_all_without Chlordanes	11	3	0,669	0,988	0,014	0,003
PLS_all	17	5	0,682	0,971	0,017	0,005
PLS_Toxaphenes	17	3	0,528	0,787	0,024	0,017
PLS_PCB	10	2	0,518	0,944	0,016	0,006
PLS_Chlordanes	23	2	0,608	0,791	0,027	0,021
PLS_BDE	15	1	0,203	0,645	0,021	0,016

**cf. explanation of model performance parameters in Table 1*

For the first six models based on all chemical variables, excluding one chemical class, the robustness is highest for the model based on all chemicals (PLS_all). The robustness decreasing as follows: PLS_all, PLS_all_without Chlordanes , PLS_without_PCBs, PLS_all_without Toxaphenes, PLS_allwithout HBCD and Me-TBBP-A and PLS_all_without_BDEs. The model performance data of the PLS models based on one chemical class only shows that the PLS- Regression based on the Chordanes and Toxaphenes are better than for PCBs and BDEs, respectively. The model based on the BDEs alone has lowest robustness.

Table 5. Explained variance in the first two principal components and total explained variance of the individual models based on the left side samples.

	Explained X- and Y-variance in PC1, PC2 and total											
	X-variance by calibration			*X-variance by cross-validation*			*Y-variance by calibration*			*Y-variance by validation*		
Model*	PC_1	PC_2	$\sum PC_{cal}$	PC_1	PC_2	$\sum PC_{val}$	PC_1	PC_2	$\sum PC_{cal}$	PC_1	PC_2	$\sum PC_{val}$
PLS_all_without_BDEs	18	29	**72**	7	16	***38***	60	8	**92**	-15	-3	***18***
PLS_all_without Toxaphenes	31		**31**	19		***19***	34		**34**	5		***5***
PLS_all_without PCBs	19	31	**49**	1	13	***14***	63	11	**74**	15	22	***37***
PLS_all_without HBCD and Me-TBBP-A	20	25	**69**	8	15	***36***	52	21	**87**	7	0	***35***
PLS_all_without Chlordanes	30	22	**72**	8	4	***27***	68	28	**97**	27	23	***53***
PLS_all	32	15	**75**	18	2	***28***	47	34	**94**	23	11	***51***
PLS_Toxaphenes	68	17	**90**	50	12	***62***	28	24	**62**	13	7	***30***
PLS_PCB	41	17	**58**	22	5	***26***	63	26	**89**	24	12	***37***
PLS_Chlordanes	61	26	**87**	44	18	***63***	35	27	**63**	23	27	***50***
PLS_BDE	28		**28**	0.5		***0.5***	42		**42**	3		***3***

*cf. explanation given beneath Table 2

In general the robustness of models based on the left side group is lower than for the model based on all samples. This is also seen by comparing the validation (bold, italic numbers) and calibration (bold numbers) X-variance used for explaining the Y-variance, i.e. high differences in explained Y-variance by calibration compared to validation (bold and italic numbers) is seen from Table 5.

Table 6. Results on Linear Regression of Principal Components as function of year and eggshell, respectively.

	PCs as function of year						*PCs as function of eggshell thickness*					
	t_1			t_2			t_1			t_2		
Model	Slope	R^2	P(slope)	slope	R^2	P(slope)	slope	R^2	P(slope)	slope	R^2	P(slope)
LR_all_without_BDEs	0,05	0,01	0,66	0,37	**0,27**	1,00	51	0,60	1,00	22	0,06	0,86
LR_all_without Toxaphenes	-0,01	0,00	0,52				80	0,34	1,00			
LR_all_without PCBs	-0,01	0,00	0,55	0,32	**0,32**	1,00	65	0,63	1,00	34	0,08	0,90
LR_all_without HBCD and Me-TBBP-A	0,11	0,04	0,80	0,38	**0,34**	1,00	88	0,52	1,00	61	0,19	0,98
LR_all_without Chlordanes	-0,04	0,00	0,57	0,59	**0,75**	1,00	150	0,68	1,00	93	0,28	0,98
LR_all	-0,02	0,00	0,55	0,30	**0,29**	1,00	116	0,47	1,00	64	0,33	1,00
LR_Toxaphenes	0,05	0,02	0,71	0,03	0,02	0,71	37	0,28	0,99	15	0,17	0,97
LR_PCB	-0,29	0,11	0,87	0,04	0,00	0,59	137	0,64	1,00	55	0,25	0,96
LR_Chlordanes	108,19	0,16	0,98	1,32	0,00	0,51	20475	0,27	1,00	11638	0,16	0,98
LR_BDE	0,10	0,10	0,91				49	0,42	1,00			

The regression of the principal components, i.e. egg sample score values, as function of year and eggshell thickness, respectively, shows that there is a stronger correlation to the eggshell thickness in t_1 than in t_2. For the models based on all chemicals and all chemicals leaving one group out there are small, and only in one case a relatively high correlation between egg sample score values in t_2 and year. In general, the correlation coefficients of t_2, based on all chemical groups and leaving one group out, as a function of year (bold number) is significantly better than seen in any of the other models.

For the BDEs only a one component model could be obtained for the left side objects. All results and comparisons between chemical groups should still be evaluated with care as the number of chemicals varies from few to many between chemicals classes, and the number of objects in the lefts and right side object groups varies as well. These circumstances may indeed influence the robustness of the models, which varies to a high degree.

Overall there is an inherent time trend present in t_2 in models based on all chemical groups, and all chemical groups leaving one group out at a time in the left side group of objects. The time trend and eggshell thickness is not covarying as the correlation to eggshell thickness are present in t_1 of the models, whereas the time trend are present in t_2, which shows clearly from column 6 and 9 in Table 6.

Summary of model performance and results of PLS-Regressions based on the right side samples in the score plot in Figure 5

Table 7. Model performance parameters for models based on the right side egg samples.

model*	n	n PC's	q^2	r^2	RMSEP	RMSEC
PLS_all_without_BDEs	13	10	0,885	0,999	0,0327	0,00272
PLS_all_without Toxaphenes	15	4	0,673	0,994	0,0547	0,0079
PLS_all_without PCBs	11	2	0,871	0,962	0,0296	0,0163
PLS_all_without HBCD and Me-TBBP-A	13	8	0,894	0,999	0,0309	0,038
PLS_all_without Chlordanes	11	2	0,854	0,971	0,0346	0,0105
PLS_all	13	8	0,938	0,999	0,0258	0,0036
PLS_Toxaphenes	13	5	0,765	0,947	0,0474	0,0202
PLS_PCB	14	8	0,856	0,999	0,0362	0,0035
PLS_Chlordanes	12	2	0,43	0,726	0,0605	0,0436
PLS_BDE	14	3	0,780	0,919	0.0426	0.0257

**cf. explanation Table 1*

For the first six models based on all chemical variables, exclusive one chemical class, the robustness is highest for the model based on all chemicals (PLS_all). The robustness decreased as follows: PLS_all, PLS_without_PCBs, PLS_allwithout HBCD and Me-TBBP-A, PLS_all_without_BDEs, PLS_all_without Chlordanes and PLS_all_without Toxaphenes. The model performance data of the PLS models based on one chemical class at time shows that robustness is highest for the model based on the PCBs and decreases as follows: BDEs, Toxaphenes and Chlordanes.

Table 8. Explained variance in the first two principal components and total explained variance of the individual models based on the right side samples.

	Explained X- and Y-variance in PC1, PC2 and total											
	X-variance by calibration			*X-variance by cross-validation*			*Y-variance by calibration*			*Y-variance by validation*		
Model*	PC_1	PC_2	$\sum PC_{cal}$	PC_1	PC_2	$\sum PC_{val}$	PC_1	PC_2	$\sum PC_{cal}$	PC_1	PC_2	$\sum PC_{val}$
PLS_all_without_BDEs	31	16	**47**	5	1	***6***	78	15	**93**	54	7	***61***
PLS_all_without Toxaphenes	27	18	**45**	11	8	***20***	57	33	**90**	3	20	***23***
PLS_all_without PCBs	40	30	**69**	20	10	***30***	81	12	**93**	70	9	***80***
PLS_all_without HBCD and Me-TBBP-A	30	17	**47**	7	8	***15***	78	12	**90**	51	11	***63***
PLS_all_without Chlordanes	27	24	**51**	8	8	***17***	86	9	**94**	58	19	***77***
PLS_all	30	16	**46**	4	6	***11***	81	12	**93**	59	10	***69***
PLS_Toxaphenes	66	21	**87**	34	26	***61***	42	15	**57**	22	15	***37***
PLS_PCB	37	16	**54**	17	11	***28***	53	27	**80**	28	3	***31***
PLS_Chlordanes	51	23	**74**	12	-2	***9***	42	10	**52**	16	7	***23***
PLS_BDE	61	18	**86**	50	8	***58***	43	35	**84**	28	34	***64***

In general the total explained Y-variance by calibration and the X-variance used for explaining Y in the models based on the right side group is high compared to the left side group (Table 5) as well as the models where all objects are included (Table 3). In the case of the model based on BDEs alone, 86 % calibration X-variance is used for explaining 84 % of the Y variance. Still the robustness is varying as seen from the validated X and Y-variance (bold italic numbers) compared to the calibrated models (bold numbers). The PLS_BDE model has high robustness and well-balanced and high explanatory as well as explained Y-variance both in the calibrated and validated model.

Table 9. Results on Linear Regression of Principal Components as function of year and eggshell, respectively.

	PC as function of year						*PC as function of eggshell thickness*					
	t_1			t_2			t_1			t_2		
Model	slope	R^2	P(slope)	slope	R^2	P(slope)	slope	R^2	P(slope)	slope	R^2	P(slope)
LR_all_without_BDEs	0,27	0,09	0,98	0,06	0,03	0,88	35	0,24	1,00	15	0,39	1,00
LR_all_without Toxaphenes	0,16	0,07	0,85	-0,12	0,04	0,78	31	0,57	1,00	24	0,32	1,00
LR_all_without PCBs	-0,21	0,11	0,88	0,30	0,35	0,99	43	0,81	1,00	13	0,12	0,89
LR_all_without HBCD and Me-TBBP-A	186	0,06	0,82	-139	0,07	0,83	45069	0,78	1,00	11195	0,09	0,88
LR_all_without Chlordanes	0,03	0,00	0,56	0,11	0,03	0,73	48	0,86	1,00	13	0,08	0,83
LR_all	0,19	0,05	0,81	-0,13	0,06	0,81	50	0,81	1,00	12	0,10	0,89
LR_Toxaphenes	0,06	0,02	0,68	0,01	0,00	0,58	19	0,42	1,00	5	0,14	0,93
LR_PCB	126	0,04	0,78	9,96	0,00	0,54	31783	0,54	1,00	13434	0,31	0,99
LR_Chlordanes	-0,14	0,21	0,96	0,13	0,15	0,93	14	0,42	1,00	7	0,09	0,86
LR_BDE	0,07	0,04	0,76	-0.06	0,08	0,86	20	0,43	1.00	10	0,34	1,00

The regression of the principal components as function of year does not verify any patterns in score values of the egg samples indicating the presence of a time trend for the right side groups of objects. The regression of the principal components as function of eggshell thickness, however, shows that a more or less pronounced correlation to eggshell thickness is present in the first principal component, t_1, for all models.

The third dimension of the PLS models

In t_3 of all of the PLS-R models (which includes a third dimension; cf. Table 1) based on all objects, the BDEs are inversely related to the eggshell thickness as shown in Figure 7. The inverse relation between the BDEs and the eggshell thickness are accompanied by a time trend which is positive for the BDEs and negative for the eggshell thickness; a variance pattern of 5 % X-variance explaining 15% Y-variance in the case of the PLS-all model in Table 1.

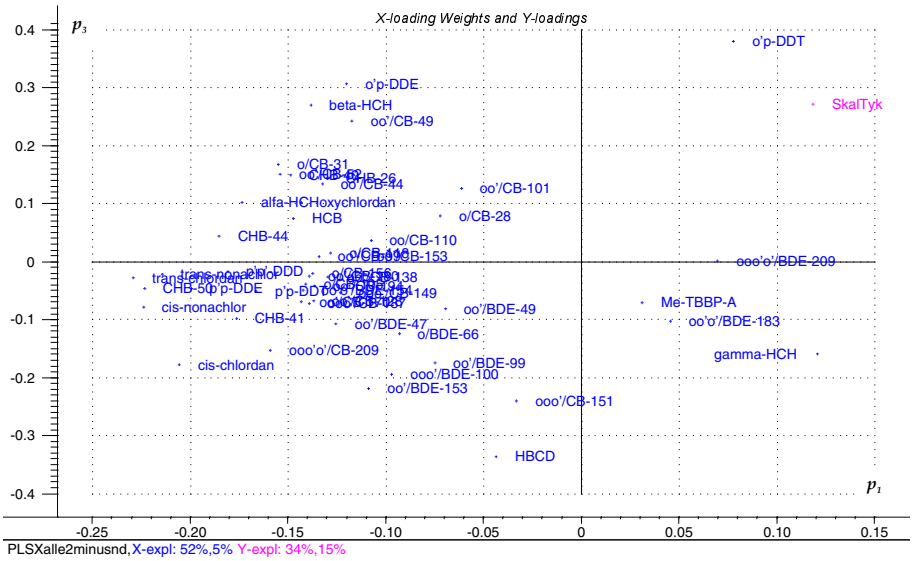


Figure 7. The loading plot of p_3 versus p_1 shows the BDEs as being the only group of chemicals, in addition to HBCD, having significant negative loading values in p_3. The third PC uses only 5 % X-variance to explain 15 % variance in eggshell thickness.

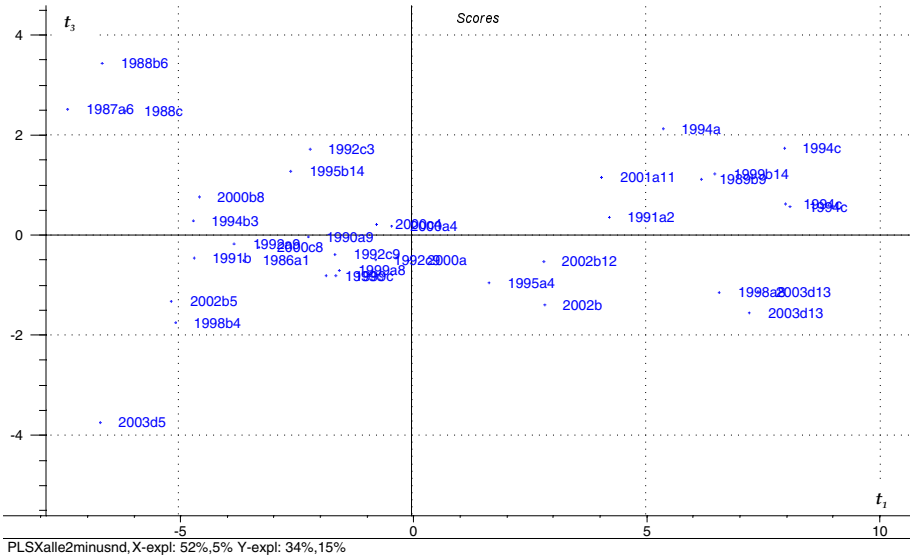


Figure 8. The score plot of t_3 versus t_1 showing a high frequency of egg samples of earliest years to have positive score values in t_3, whereas late egg sample years have negative score values in t_3. The pattern in t_3 of the score plot verifies the positive time trend for the BDEs as concentration profiles of the BDEs are increasing from top towards bottom in the score plot.

The time trend pattern in the score plot in Figure 8 was verified by linear regressing of t_3 as function of time which resulted in a correlation coefficient of 0.34 and a negative slope of –0.17, i.e. earliest year have highest score values. The correlation coefficient t_3 as function of eggshell thickness was only 0,1 in agreement with the overall insignificant time trend for the eggshell thickness (cf. Figure 10 in the main report).

Key results

Principal Component Analysis (PCA) and Partial Least Square Regression (PLS-R) showed that the egg samples seem to be divided into groups of egg samples (two clusters).

A minor group of egg samples (cluster of objects) includes no time trend, while a major group of egg samples includes a time trend.

The explanatory variables in the PLS-R models used in this appendix are so-called principal components and the overall results are as follows:

The first dimension of the models described an inverse relation between the majority of chemicals and the eggshell thickness. This dimension includes no time trend. The PCBs have high importance in this dimension.

The second dimension includes a time trend explained by an increasing eggshell thickness inversely correlated to the p,p´-configuration of DDT and its degradation products, the chlordanes and toxaphene congeners, i.e. the eggshell shows patterns of increasing time trend, whereas the concentration of chemicals of high importance are decreasing in time.

The p,p-configuration of DDE and DDD is the most significantly original variables inversely correlated to the eggshell thickness as these chemicals have high importance in the first as well as the second dimension of the model planes.

The third dimension includes a time trend pattern of decreasing eggshell thickness explained by an increase in the BDEs, i.e. the BDEs show increasing time trend and are inverse correlated to the eggshell thickness in this dimension.

References

Höskuldsson, A. (1996). *Prediction Methods in Science and Technology.* Thor Publishing, Denmark, p. 405.

Eriksson, L., Johansson, E., Kettaneh-Wold, N., Wold, S. (1999). *Multi- and Megavariate Data Analysis using Projection Methods (PCA & PLS),* UMETRICS AB, p. 490.

Appendix 8

Rank correlation for shell thickness, concentration and molecular flexibility

Ranking of substances and correlation to eggshell thickness

The lipid concentration level may be a potential factor governing the potential toxic effect responsible for the thinning of eggshells. Both the hydrophobicity and general level of environmental contamination govern the concentration level in the lipid. The correlation, in form of the Pearson correlation coefficient, is taken from Appendix 4, between the eggshell thickness and the mean lipid concentration. This correlation is shown as a function of the mean concentration level in figure 1. The mean lipid concentration level is calculated based on the ln transformed data assuming a ln-Normal distribution.

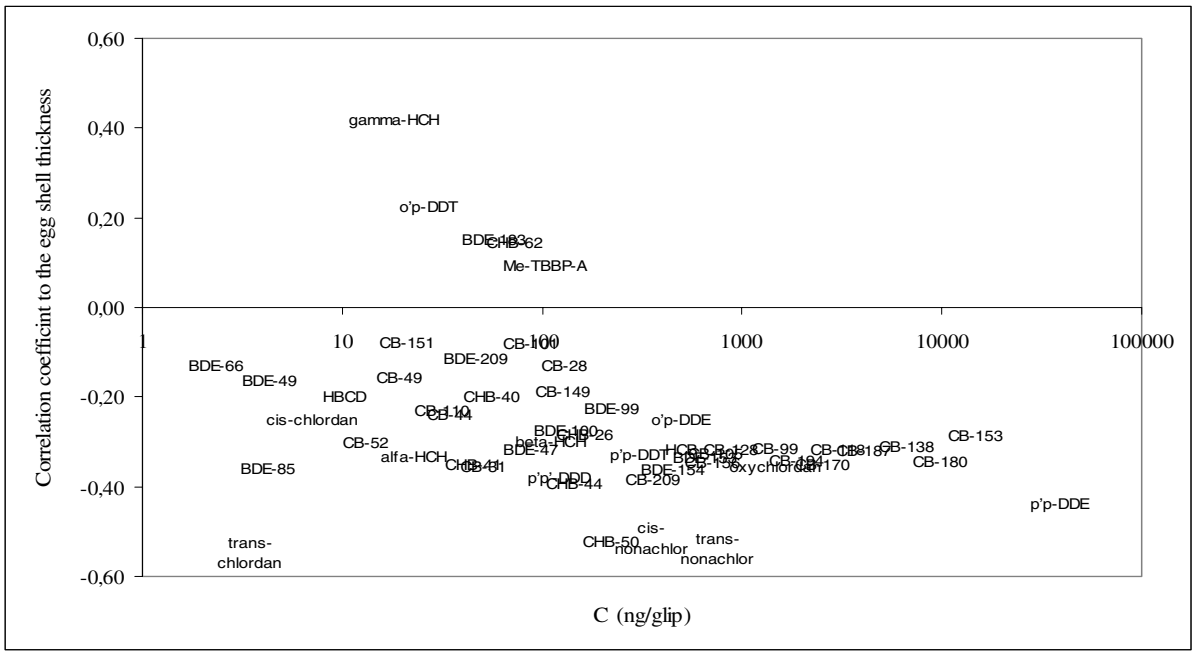


Figure 1. The correlation on the y-axis is measured by the Pearsons Coefficient between the lipid concentration level (ln transformed data) and the egg shell thickness for single egg measurements.

In Figure 1, a general agreement seems to exist for most substances that there is a negative correlation with eggshell thickness when the concentration level exceeds approximately 100 ng/g lipid. Below this concentration level, the toxic influence from the substances can not be strongly identified. However, there may still be some substances in the lower concentration range that do have toxic influence, e.g. transchlordan. This is evidence for a general negative influence on eggshell thickness at higher contamination levels.

Not only the contamination level, but also, e.g., the flexibility of the molecule can have some influence on the toxicity (Thomsen and Carlsen, 2002). For a flexible molecule there will be a higher risk that the

structure can fit into a specific receptor-site and induce a harmful effect. The basic structure of the molecules in this analysis is two connected benzene rings. If this connection is flexible then the planes of the benzene rings will change the angle over time due to the Brownian vibrations of the molecules. But, the presence of substituents in the orto position on the benzene rings will induce lower flexibility due to increased angle strain. So, an increasing number of ortho-substituents may induce lower toxicity if the hypothesis about the flexible molecule as being most toxic is true. It is possible to have maximal two ortho-substituents on each benzene ring, so, the possible numbers of ortho-substituents are 0,1,2,3,4. If the toxicity is increasing by increasing molecular flexibility then rank of substances in relation to increasing toxicity will follow the inverse rank of the number of ortho-substituents. In the following all substances having the two-ring structure are selected yielding a set of 38 substances. The data is shown in Table 1.

Table 1. Data for the substances having two-ring structure. The concentration is a mean value estimate based on ln-transformed data. The id number is used rankings shown in the following figures. Negative Pearson coefficients relate to substances where an increasing concentration leads to thinner eggshells.

id	Substance	Mean Concentration (ng/glip)	Number of substituents	Pearson Correlation
1	p'p'-DDD	123	0	-0,40
2	p'p-DDE	38652	0	-0,46
3	p'p-DDT	306	0	-0,35
4	CB-28	129	1	-0,15
5	CB-31	50	1	-0,37
6	CB-105	732	1	-0,34
7	CB-118	3016	1	-0,33
8	CB-156	714	1	-0,36
9	o'p-DDE	488	1	-0,27
10	o'p-DDT	27	1	0,21
11	BDE-47	88	1	-0,33
12	BDE-99	222	1	-0,25
13	CB-44	34	2	-0,26
14	CB-49	19	2	-0,17
15	CB-52	13	2	-0,32
16	CB-99	1460	2	-0,33
17	CB-101	87	2	-0,10
18	CB-110	31	2	-0,25
19	CB-128	871	2	-0,34
20	CB-138	6621	2	-0,33
21	CB-153	14611	2	-0,30
22	CB-170	2542	2	-0,37
23	CB-180	9718	2	-0,36
24	CB-194	1876	2	-0,36
25	BDE-49	4	2	-0,18
26	BDE-66	2	2	-0,15
27	BDE-100	130	2	-0,29
28	BDE-154	453	2	-0,38
29	BDE-153	653	2	-0,35
30	BDE-209	46	2	-0,13
31	CB-149	126	3	-0,20
32	CB-151	21	3	-0,10
33	CB-187	4095	3	-0,34
34	BDE-85	4	3	-0,38
35	BDE-183	57	3	0,13
36	HBCD	10	3	-0,22
37	CB-209	356	4	-0,40
38	Me-TBBP-A	104	4	0,08

In the following a ranking method is applied which has been developed by Sørensen et al., (2003) using software described in Sørensen et al., (2004). Two rankings are made: (1) (*set 1*): Inverse ranking of the Pearson Coefficient, where the substances having the strongest influence for eggshell thinning are ranked at the top. (2) (*set 2*): Ranking of the mean concentration **and** the inverse ranking of the number of ortho-substituents in **one** ranking plot, where substances have high concentration **and** a low number of substituents are ranked at the top. If there exists a toxicity effect on the eggshells due to both high concentration and low number of substituents then there will be a correlation between *set 1* and *set2* ranking. The ranking of the two sets is shown in Figure 2 including the principle for the rank correlation where every rank between two substances in one set is compared with the corresponding rank between the same two objects in the other set.

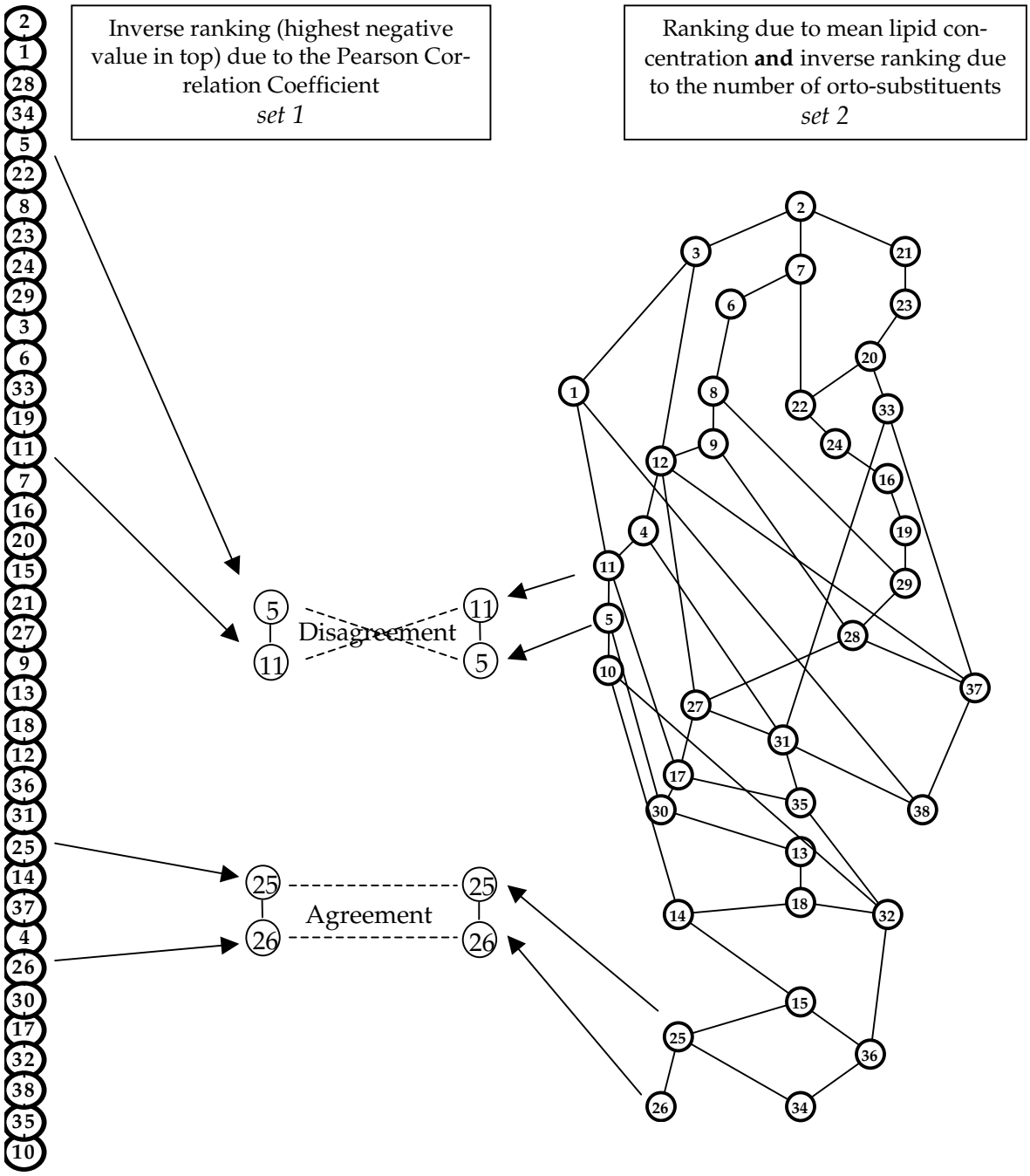


Figure 2. Rank correlation between *set 1* and *set 2*, showing an example of respectively an agreement and a disagreement between the two ranking sets.

The ranking in *set 2* is not complete when two parameters together are used for the ranking. A ranking is therefore only true when both parameters agree about the ranking forming a partial order. This partial order is shown in Figure 2 for *set 2* using a Hasse diagram, where connecting lines are made between substances for which a ranking can be made.

The results from the rank correlation are shown in Table 2, where the correlation parameter is defined as:

$$T(0,0) = \frac{\sum Agreements}{\sum Agreements + \sum disagreements}$$

Thus the value: *T(0,0)*=0.5 tells that there are the same number of agreements and disagreements and thus no correlation between the two sets. A value larger than 0.5 tells that there are a positive correlation and values below 0.5 indicates a negative correlation between the two sets. The value of significance in Table 2 is the probability for the value not to be just a random sample from non-correlated sets and thus the probability for rejecting the Ho hypothesis. Different versions of set 2 are tested in Table 2 in order to investigate the relevance of each of the single parameters: Lipid concentration (Analysis no. 2) and number of substituents (Analysis no. 3).

The Pearson correlation coefficient and both the lipid concentration and the number of substituents are seen to have positive and rather significant correlations in Table 2 especially when they are used together. This clearly indicates that both the contamination level and the number of substituents have negative effects on the eggshell thickness.

Table 2. For all substances selected in Table 1.

Analysis no.	Set 1	Set 2	Number of agreements	Number of disagreements	T(0,0)	Significans
1	Pearson Corr (Inverse ranking)	Lipid Conc Number of ortho-sub (Inverse ranking)	380	127	0.75	> 1.000
2	Pearson Corr (Inverse ranking)	Lipid Conc	475	227	0.68	0.999
3	Pearson Corr (Inverse ranking)	Number of ortho-sub (Inverse ranking)	340	155	0.69	0.995

A more detailed interpretation of the correlation between *set 1* and *set 2* is possible using the ranking of both *set 1* and *set 2* simultaneously where all parameters (Inverse Pearson corr., lipid concentration and inverse number of substituents) are used in one ranking. The result is shown in Figure 3 as a Hasse diagram, where the id 2 (p'p-DDE) is shown to be a strong top candidate. This tells that no other molecules having the two-ring structure have higher negative effect on the eggshell thickness than id 2 and that this is strongly supported by a high lipid concentration and by an absence of ortho-substituents on the phenyl rings. However, not all substances are well ranked in Figure 3. E.g. the id 34 (BDE-85) is only ranked below two substances (ids 1, 2 and 28 respectively). In order to investigate this more closely another ranking is needed in the form of the conflict ranking as discussed in Sørensen et al., (2003) and this ranking is shown in Figure 4.

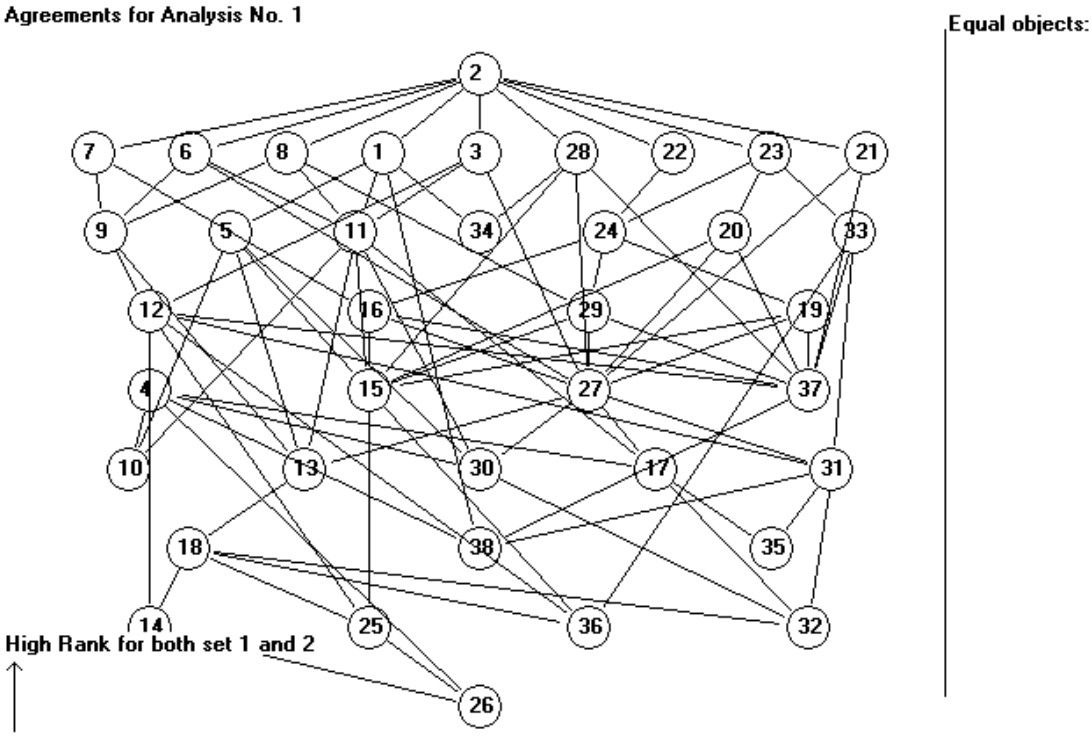


Figure 3. Ranking using: Inverse Pearson corr., lipid concentration and inverse number of substituents) together.

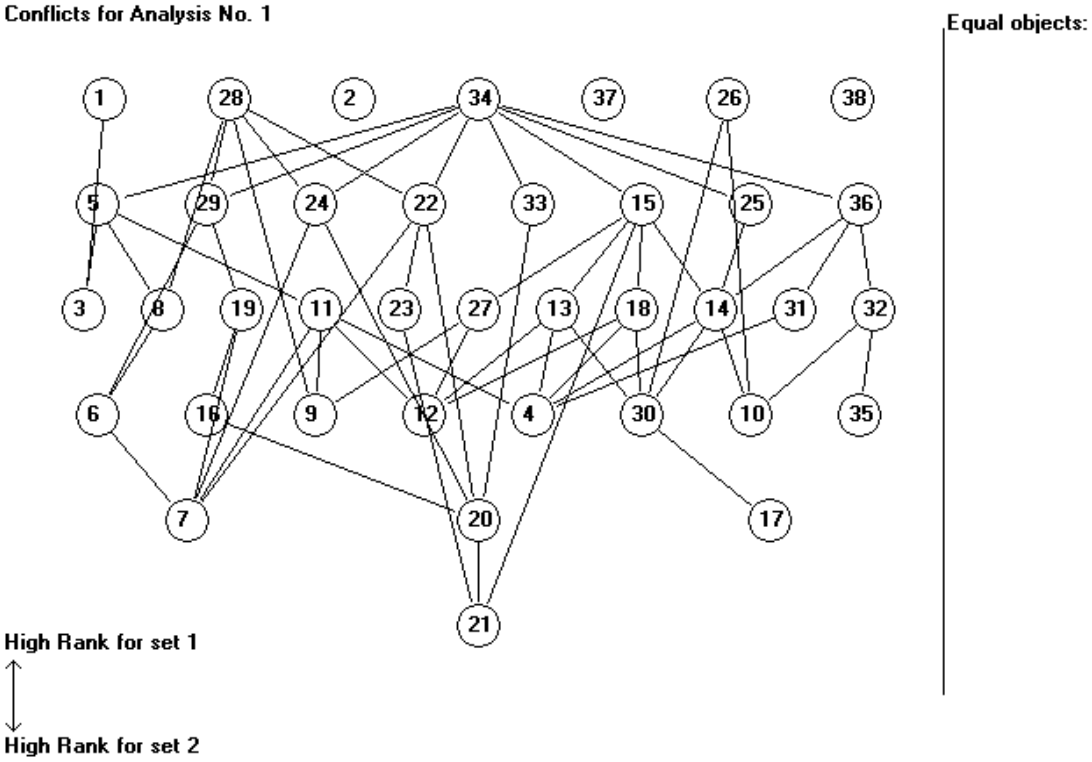


Figure 4. Ranking using the attributes Inverse Pearson corr., inverse lipid concentration and number of substituents together.

In Figure 4 the rankings of the parameters for *set 2* are turned upside-down and thus the inverse rank of the lipid concentration and number of substituents are used. The rank of *set 1* is kept unchanged in Figure 4. Thus the rankings in the Figure 3 and 4 are complementary

to some extent. Figure 3 shows all the ranking in agreement with the hypothesis that there is a correlation between set 1 and 2 and Figure 4 shows the ranking being in contradiction with the hypothesis. High ranked substances in Figure 4 are important because they are predicted by *set 2* to be less toxic than the eggshell measurements show so they are in this way false negatives. The id 34 (BDE-85), which was seen to have a low number of rankings in Figure 3 is seen in Figure 4 to have many rankings. Furthermore, the id 34 is ranked upwards in Figure 4, telling that this substances is a strong false negative, where the toxicity is higher than predicted by *set 2*. This will be investigated in more detail in the following.

In Table 3, the analysis in Table 2 is repeated but the substance id 34 is withdrawn from the data set. By comparing Analysis no. 1 (from Table 2) and 4 it is seen that the number of agreements is only decreased by 3, but the number of disagreements has decreased by 29. Thus both the *T(0,0)* value and the significance have increased. The analysis no. 5 and 6 show improvements compared to analysis 2 and 3. This indicates that both the rank of concentration level and the rank using the inverse number of ortho-substituents improves when the id 34 is withdrawn.

Table 3. For all substances selected in Table 1 excluding BDE 209 (id 34)

Analysis no.	Set 1	Set 2	Number of agreements	Number of disagreements	T(0,0)	Significans
4	Pearson Corr (Inverse ranking)	Lipid Conc Number of orto-sub (Inverse ranking)	377	96	0.80	> 0.999
5	Pearson Corr (Inverse ranking)	Lipid Conc	471	195	0.71	> 0.999
6	Pearson Corr (Inverse ranking)	Number of orto-sub (Inverse ranking)	335	128	0.72	0.999

References

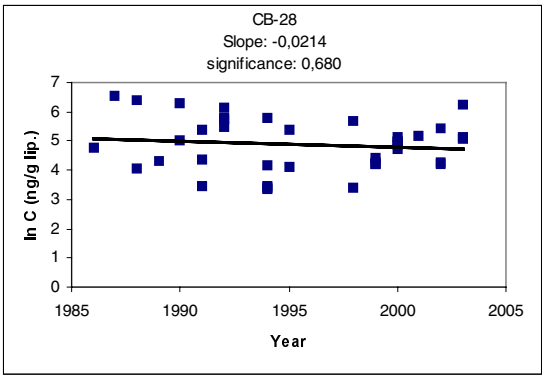
Thomsen, M. & Carlsen, L. 2002: Evaluation of empirical versus non-empirical descriptors. SAR and QSAR in Environmental Research 13, 525-540.

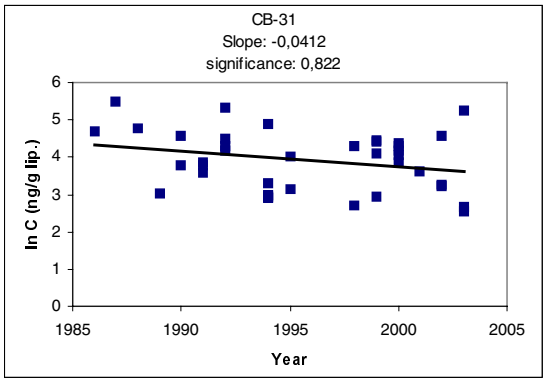
Sørensen, P.B., Brüggemann, R., Thomsen, M. & Lerche, D.B., 2004: Application of multidimensional rank-correlation. Submitted October 2004 (for more information contact Peter B. Sørensen (pbs@dmu.dk))

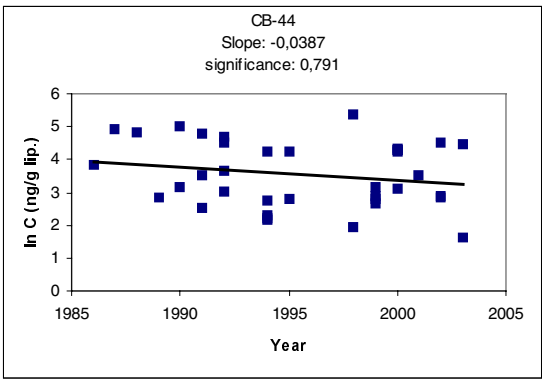
Sørensen, P.B., Brüggemann, R., Carlsen, L., Mogensen, B.B., Kreuger, J. & Pudenz, S., 2003: Analysis of monitoring data of pesticide residues in surface waters using partial order ranking theory - Data interpretation and model development, Environmental Toxicology and Chemistry, Vol 22, No. 3, pp. 661-670.

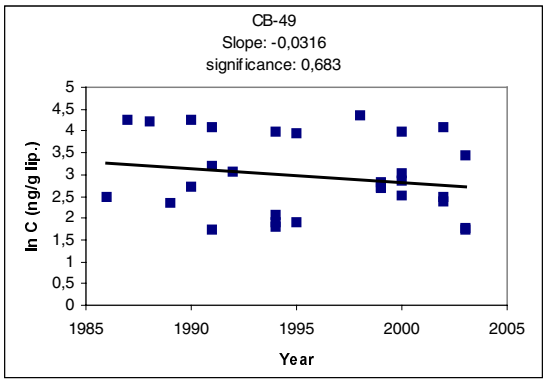
Appendix 9

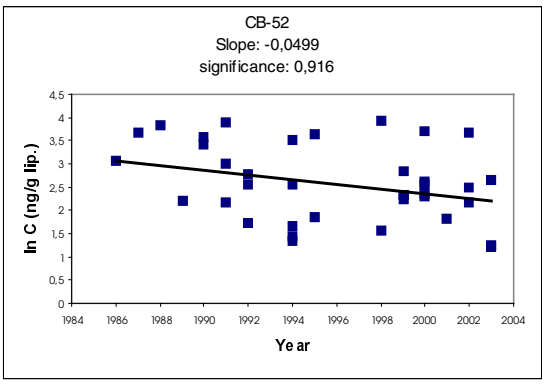
Time trend of the single chemicals

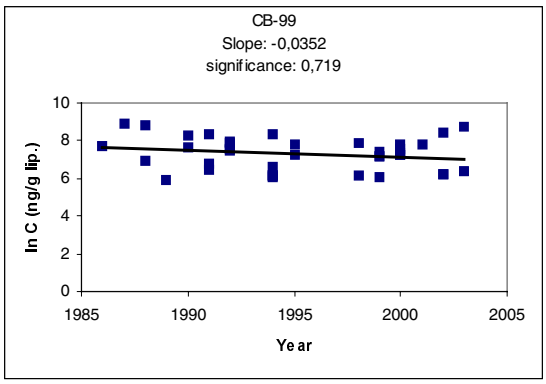


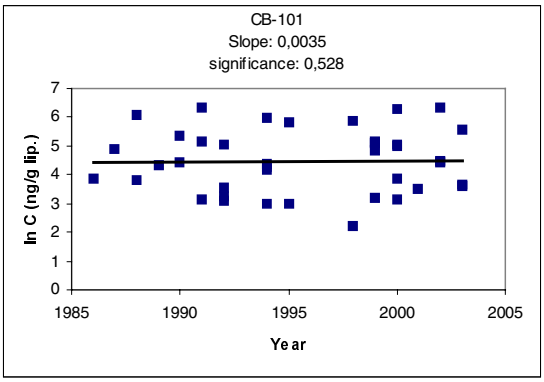








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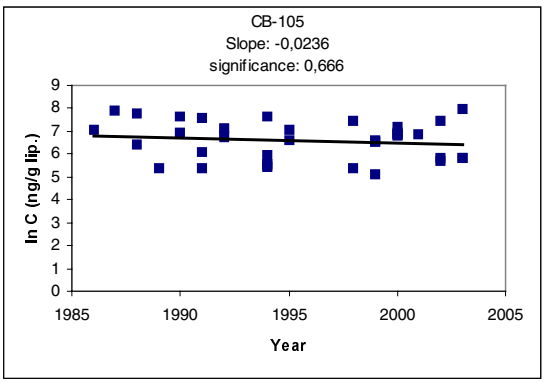
Year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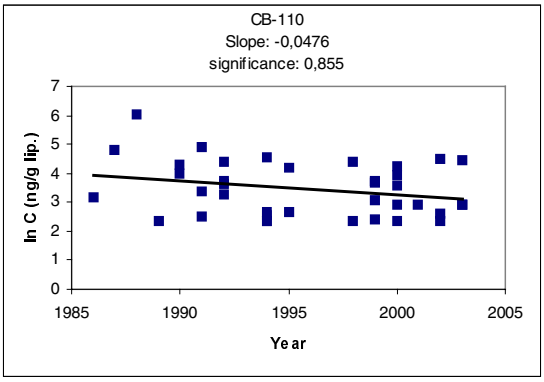
Year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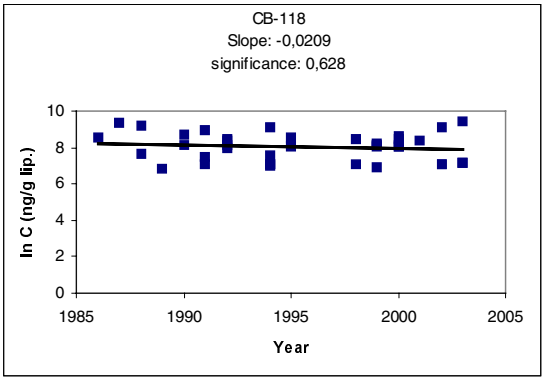
Year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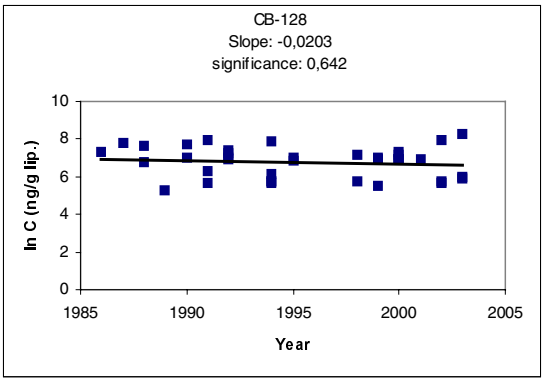
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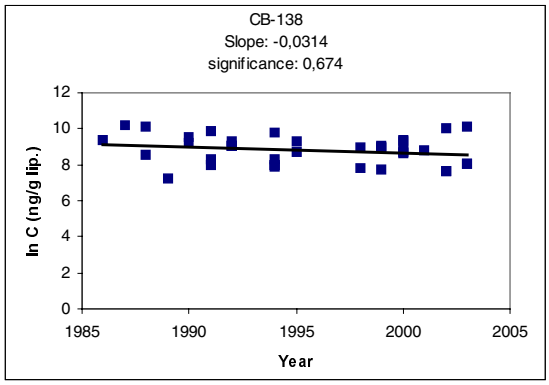
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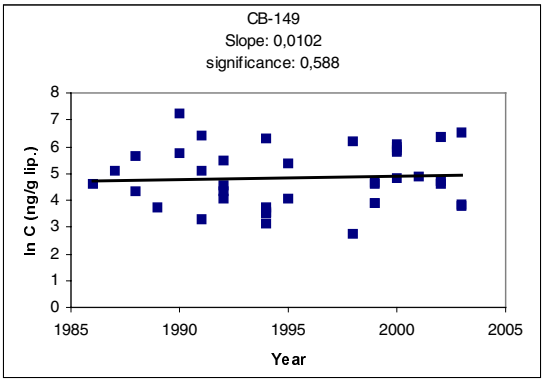
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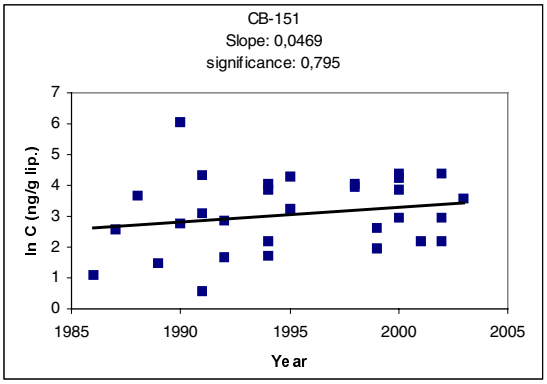
Year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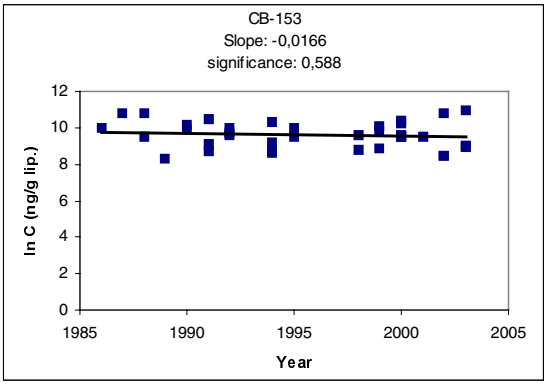
Year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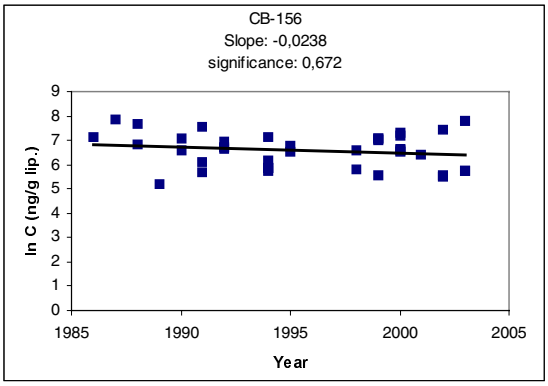
Year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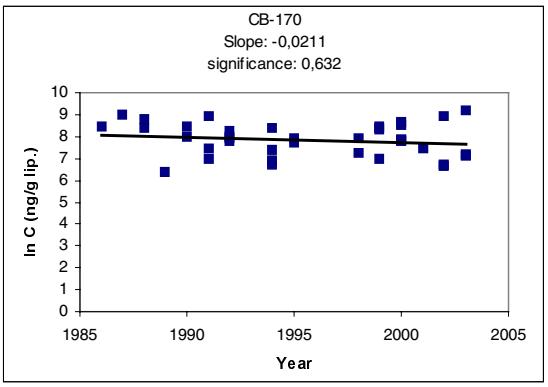
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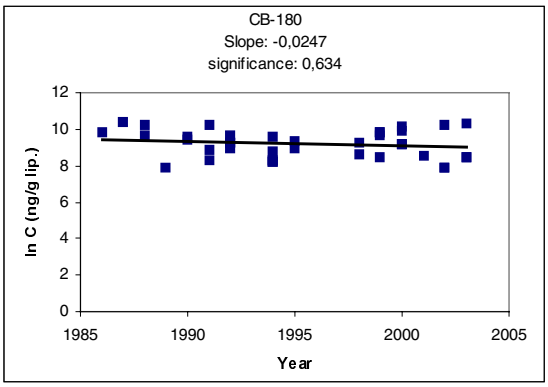
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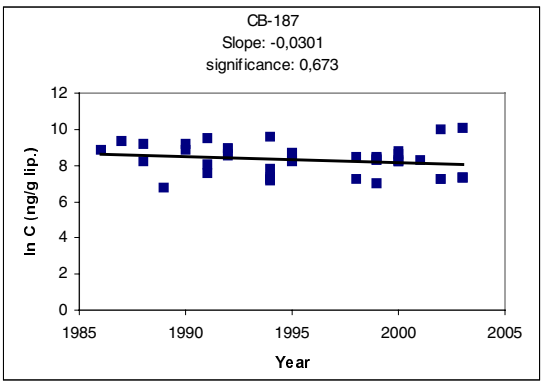
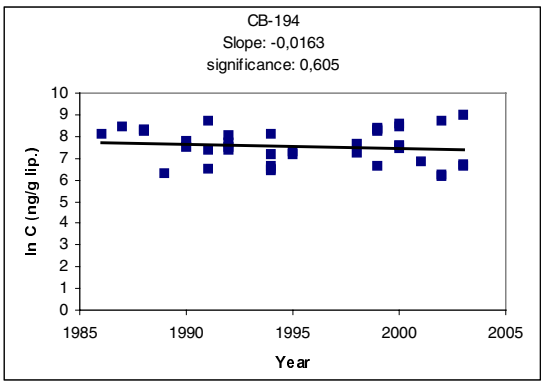
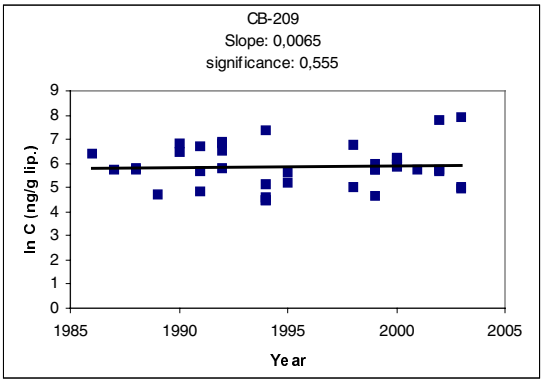
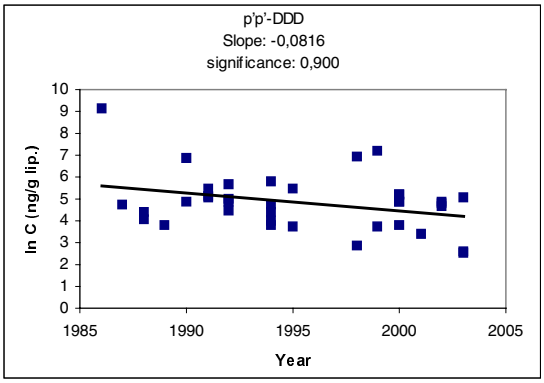
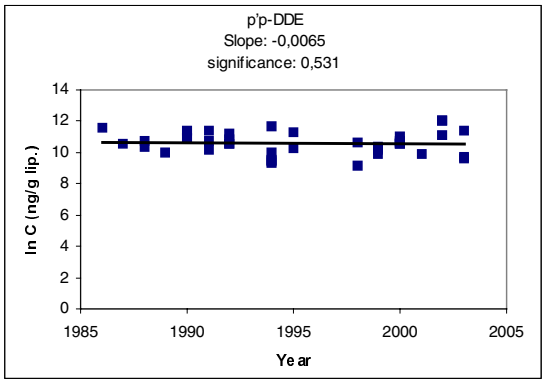
ln C (ng/g lip.)
Year

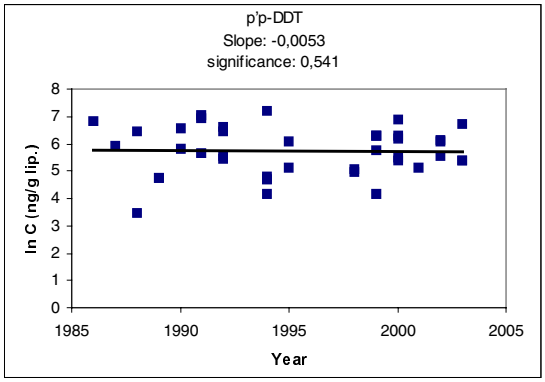

CB-187
Slope: -0,0301
significance: 0,673
ln C (ng/g lip.)
Year

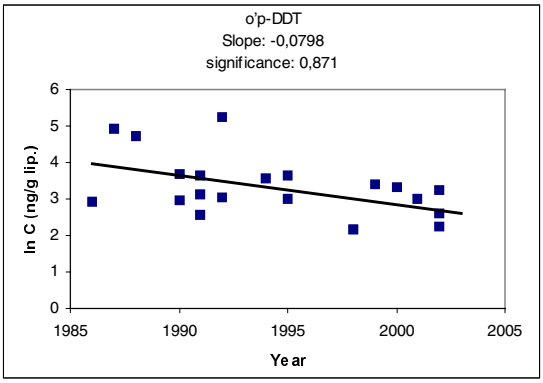

CB-194
Slope: -0,0163
significance: 0,605
ln C (ng/g lip.)
Year

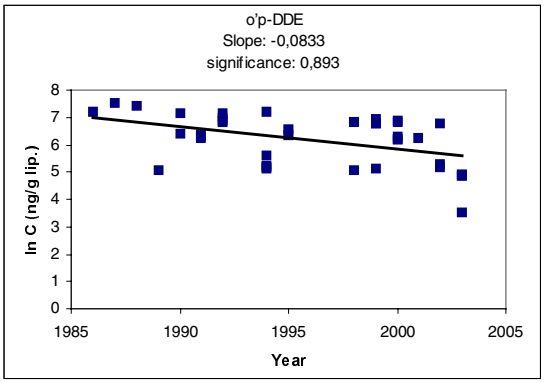

CB-209
Slope: 0,0065
significance: 0,555
ln C (ng/g lip.)
Year

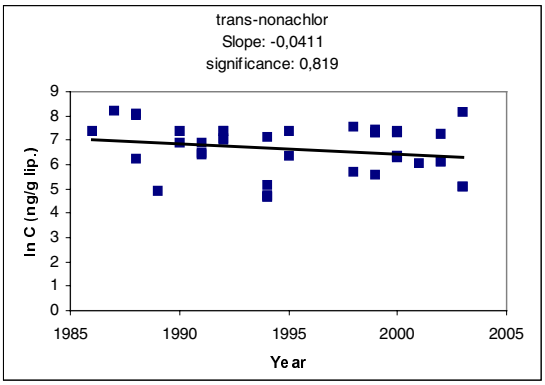

p'p'-DDD
Slope: -0,0816
significance: 0,900
ln C (ng/g lip.)
Year

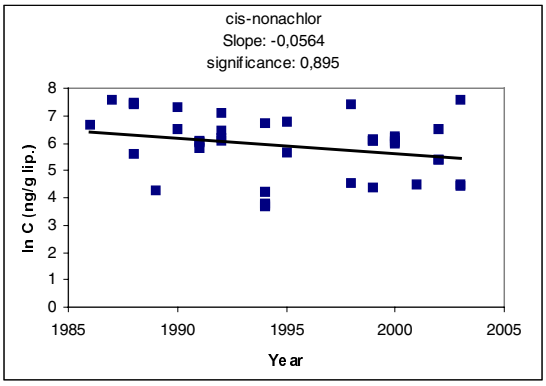

p'p-DDE
Slope: -0,0065
significance: 0,531
ln C (ng/g lip.)
Year

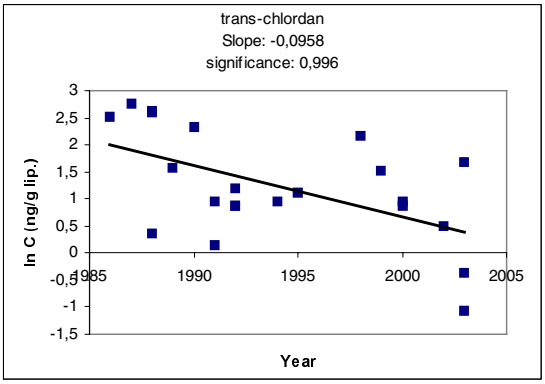

p'p-DDT
Slope: -0,0053
significance: 0,541
ln C (ng/g lip.)
Year

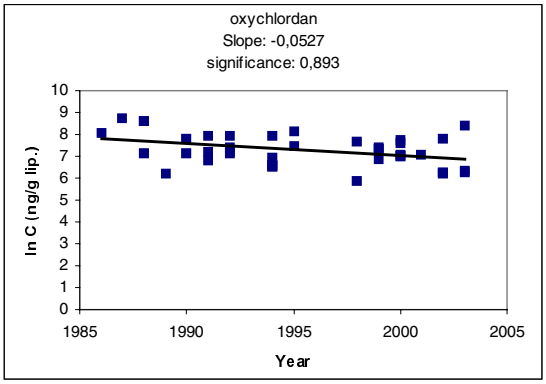

o'p-DDT
Slope: -0,0798
significance: 0,871
ln C (ng/g lip.)
Year

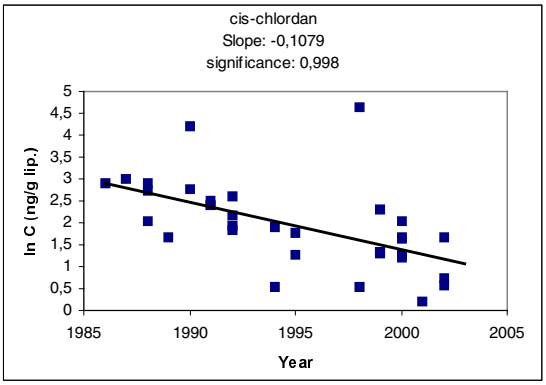

o'p-DDE
Slope: -0,0833
significance: 0,893
ln C (ng/g lip.)
Year

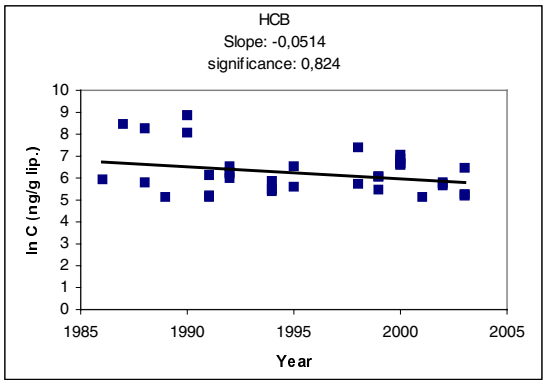

trans-nonachlor
Slope: -0,0411
significance: 0,819
ln C (ng/g lip.)
Year

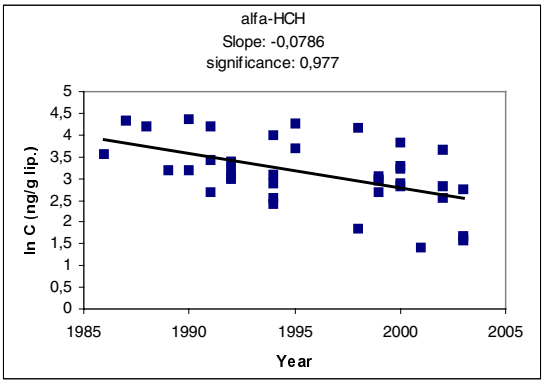

cis-nonachlor
Slope: -0,0564
significance: 0,895
ln C (ng/g lip.)
Year

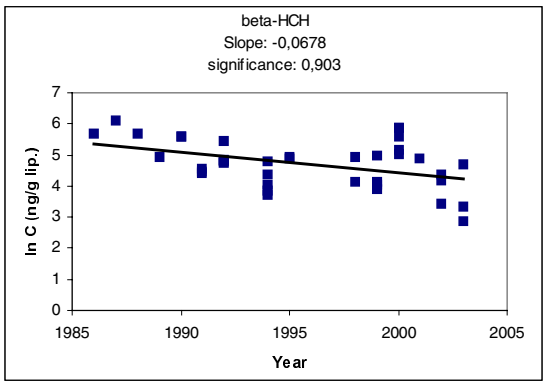

trans-chlordan
Slope: -0,0958
significance: 0,996
ln C (ng/g lip.)
Year

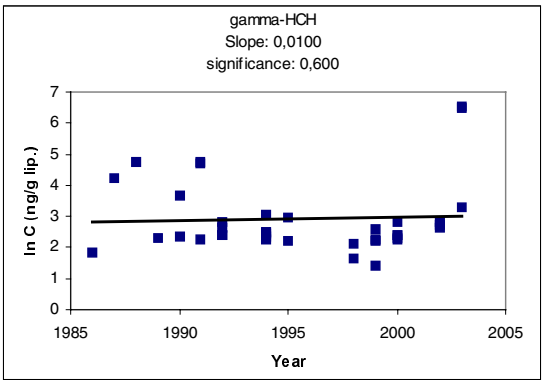

oxychlordan
Slope: -0,0527
significance: 0,893
ln C (ng/g lip.)
Year

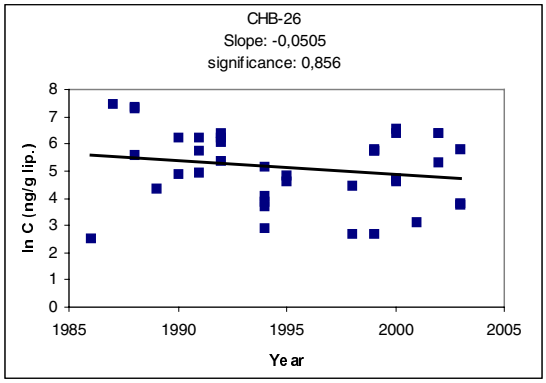

cis-chlordan
Slope: -0,1079
significance: 0,998
ln C (ng/g lip.)
Year

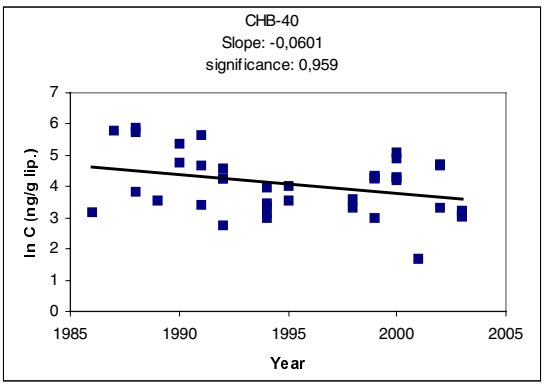

HCB
Slope: -0,0514
significance: 0,824
ln C (ng/g lip.)
Year

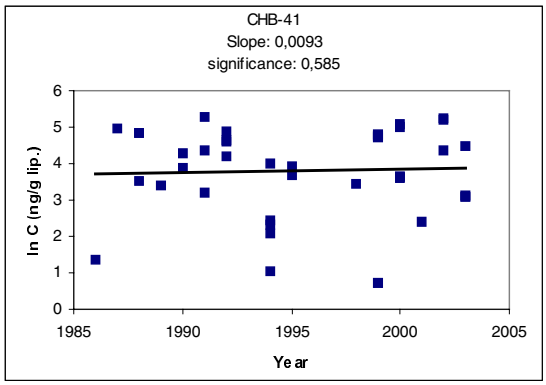

alfa-HCH
Slope: -0,0786
significance: 0,977
ln C (ng/g lip.)
Year

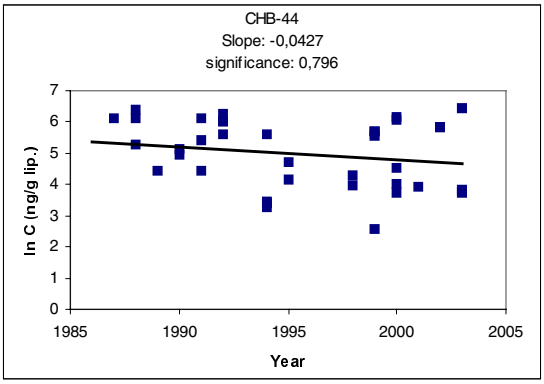

beta-HCH
Slope: -0,0678
significance: 0,903
ln C (ng/g lip.)
Year

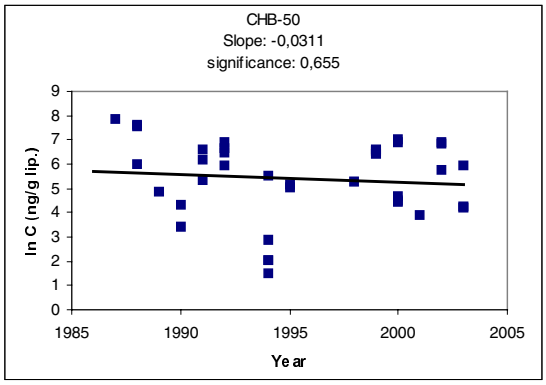

gamma-HCH
Slope: 0,0100
significance: 0,600
ln C (ng/g lip.)
Year

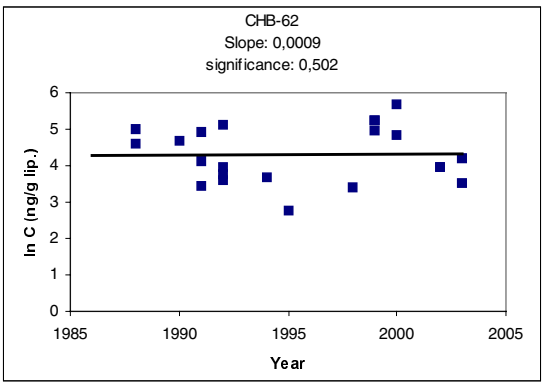

CHB-26
Slope: -0,0505
significance: 0,856
ln C (ng/g lip.)
Year

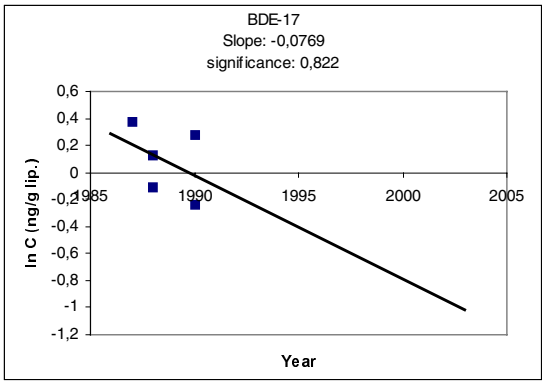

CHB-40
Slope: -0,0601
significance: 0,959
ln C (ng/g lip.)
Year

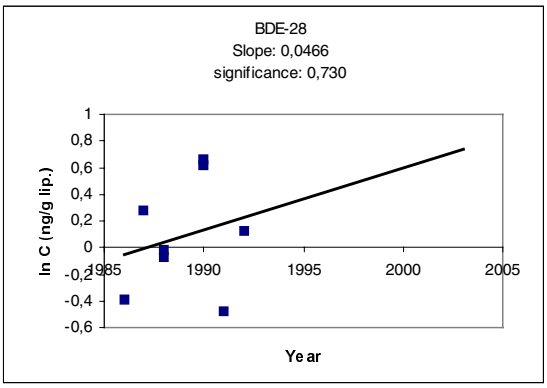

CHB-41
Slope: 0,0093
significance: 0,585
ln C (ng/g lip.)
Year

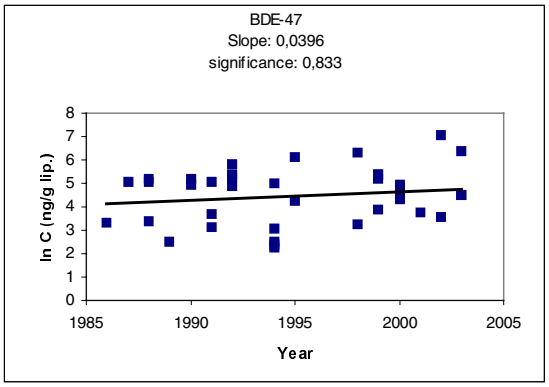

CHB-44
Slope: -0,0427
significance: 0,796
ln C (ng/g lip.)
Year

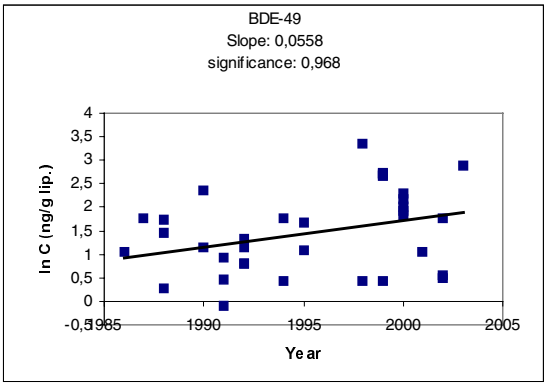

CHB-50
Slope: -0,0311
significance: 0,655
ln C (ng/g lip.)
Year

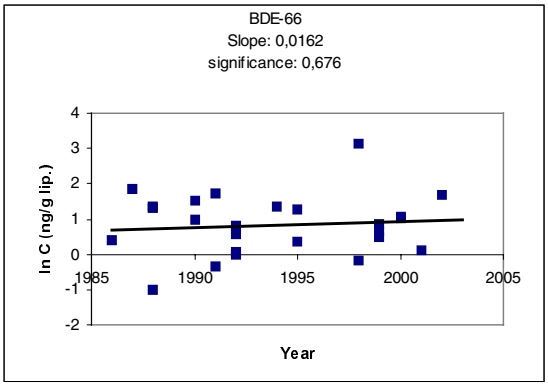

CHB-62
Slope: 0,0009
significance: 0,502
ln C (ng/g lip.)
Year

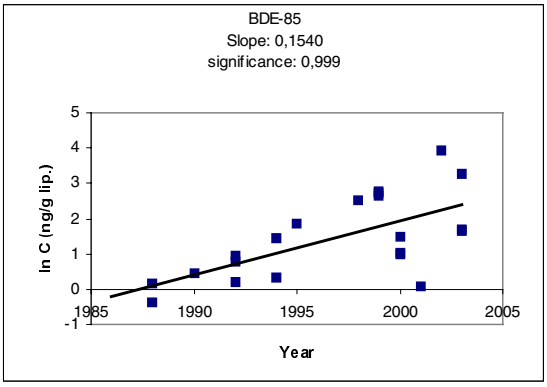

BDE-17
Slope: -0,0769
significance: 0,822
ln C (ng/g lip.)
Year


BDE-28
Slope: 0,0466
significance: 0,730
ln C (ng/g lip.)
Year


BDE-47
Slope: 0,0396
significance: 0,833
ln C (ng/g lip.)
Year


BDE-49
Slope: 0,0558
significance: 0,968
ln C (ng/g lip.)
Year


BDE-66
Slope: 0,0162
significance: 0,676
ln C (ng/g lip.)
Year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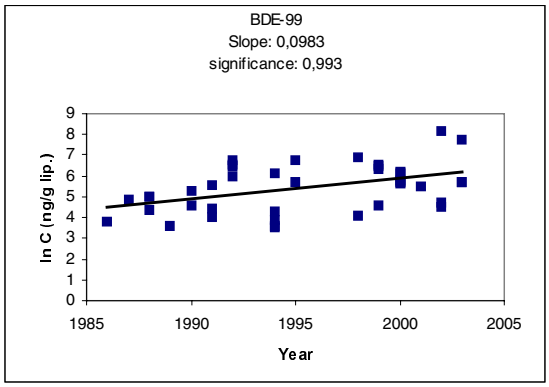
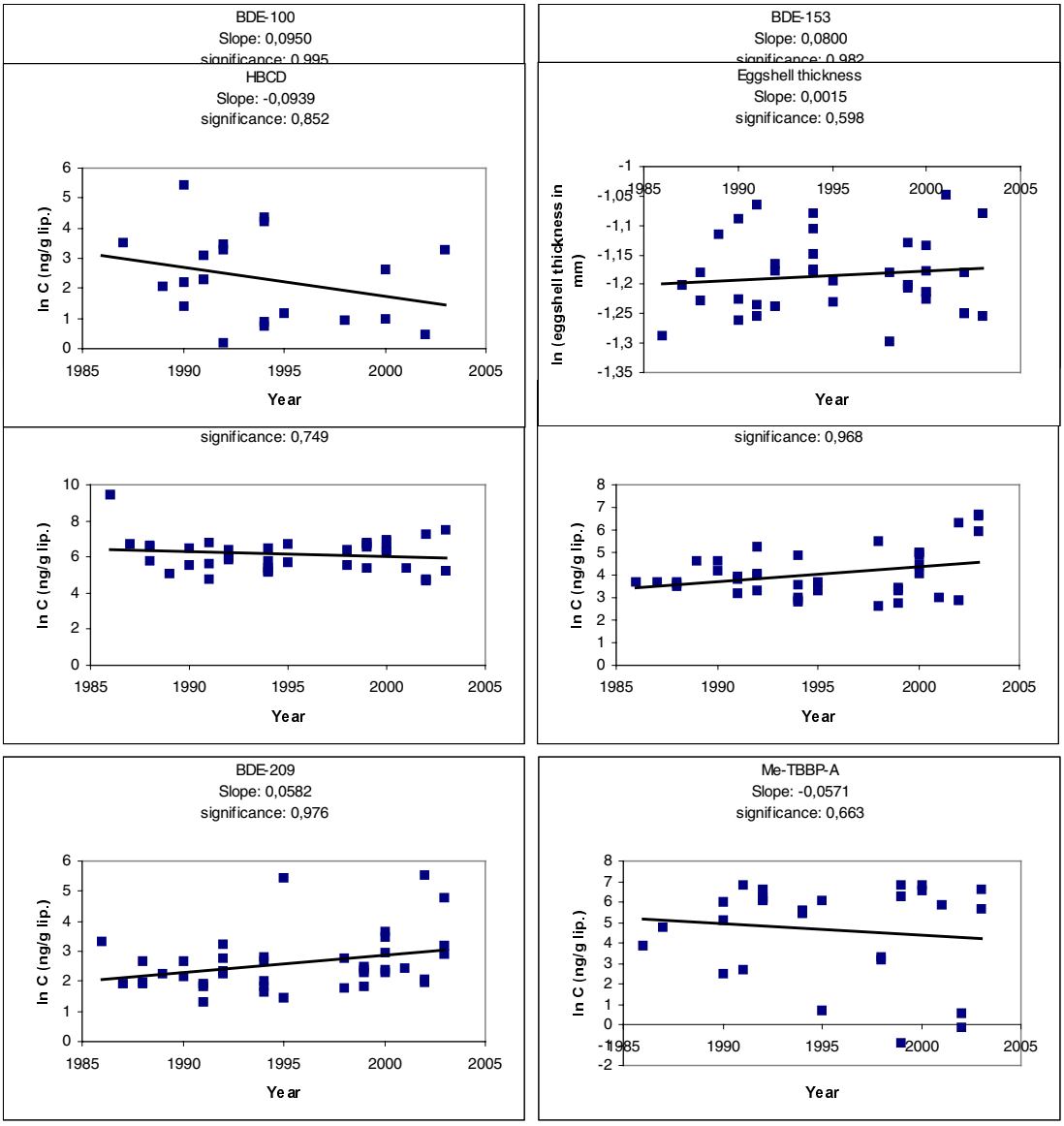
Slope: 0,1540
significance: 0,999
ln C (ng/g lip.)
Year


BDE-99
Slope: 0,0983
significance: 0,993
ln C (ng/g lip.)
Year


BDE-100
Slope: 0,0950
significance: 0,995
BDE-153
Slope: 0,0800
significance: 0,982
HBCD
Slope: -0,0939
significance: 0,852
Eggshell thickness
Slope: 0,0015
significance: 0,598
significance: 0,749
significance: 0,968
BDE-209
Slope: 0,0582
significance: 0,976
Me-TBBP-A
Slope: -0,0571
significance: 0,663
ln C (ng/g lip.)
ln (eggshell thickness in mm)
Year

National Environmental Research Institute

The National Environmental Research Institute, NERI, is a research institute of the Ministry of the Environment. In Danish, NERI is called Danmarks Miljøundersøgelser (DMU).
NERI's tasks are primarily to conduct research, collect data, and give advice on problems related to the environment and nature.

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Publications:
NERI publishes professional reports, technical instructions, and the annual report in Danish.
A R&D projects' catalogue is available in an electronic version on the World Wide Web.
Included in the annual report is a list of the publications from the current year.

NERI Technical Reports

2003

Nr. 481: Polybrominated Diphenyl Ethers (PBDEs) in Sewage Sludge and Wastewater. Method Development and validation. By Christensen, J.H. et al. 28 pp. (electronic)

2004

Nr. 482: Background Studies in Nuussuaq and Disko, West Greenland. By Boertmann, D. (ed.) 57 pp. (electronic)
Nr. 483: A Model Set-Up for an Oxygen and Nutrient Flux Model for Århus Bay (Denmark). By Fossing, H. et al. 65 pp., 100,00 DDK.
Nr. 484: Satellitsporing af marsvin i danske og tilstødende farvande. Af Teilmann, J. et al. 86 s. (elektronisk)
Nr. 485: Odense Fjord. Scenarier for reduktion af næringsstoffer. Af Nielsen, K. et al. 274 s. (elektronisk)
Nr. 486: Dioxin in Danish Soil. A Field Study of Selected Urban and Rural Locations. The Danish Dioxin Monitoring Programme I. By Vikelsøe, J. (electronic)
Nr. 487: Effekt på akvatiske miljøer af randzoner langs målsatte vandløb. Pesticidhandlingsplan II. Af Ravn, H.W. & Friberg, N. 43 s. (elektronisk)
Nr. 488: Tools to assess the conservation status of marine habitats in special areas of conservation. Phase 1: Identification of potential indicators and available data. By Dahl, K. et al. 94 pp., 100,00 DKK
Nr. 489: Overvågning af bæver Castor fiber i Flynder å, 1999-2003. Af Elmeros, M., Berthelsen, J.P. & Madsen, A.B. 92 s. (elektronisk)
Nr. 490: Reservatnetværk for trækkende vandfugle. En gennemgang af udvalgte arters antal og fordeling i Danmark 1994-2001. Af Clausen, P. et al. 142 s. , 150,00 kr.
Nr. 491: Vildtudbyttet i Danmark i jagtsæsonen 2002/2003. Af Asferg, T. 24 s. (elektronisk)
Nr. 492: Contaminants in the traditional Greenland diet. By Johansen, P. et al. 72 pp. (electronic)
Nr. 493: Environmental Oil Spill Sensitivity Atlas for the South Greenland Coasatl Zone. By Mosbech, A. et al. 611 pp. (electronic)
Nr. 494: Environmental Oil Spill Sensitivity Atlas for the West Greenland (68°-72°N) Coasatl Zone. By Mosbech, A. et al. 798 pp. (electronic)
Nr. 495: NOVANA. Det nationale program for overvågning af vandmiljøet og naturen. Programbeskrivelse - del 1. Af Danmarks Miljøundersøgelser. 45 s., 60,00 kr.
Nr. 496: Velfærdsøkonomiske forvridningsomkostninger ved finansiering af offentlige projekter. Af Møller, F. & Jensen, D.B. 136 s. (elektronisk)
Nr. 497: Air Quality Monitoring Programme. Annual Summary for 2003. By Kemp, K. & Palmgren, F. 36 pp. (electronic)
Nr. 498: Analyse af højt NO_2 niveau i København og prognose for 2010. Af Berkowicz, R. et al. 30 s. (elektronisk)
Nr. 499: Anvendelse af Vandrammedirektivet i danske vandløb. Af Baattrup-Pedersen, A. et al. 145 s. (elektronisk)
Nr. 500: Aquatic Environment 2003. State and Trends - technical summary. By Andersen, J.M. et al. 50 pp. , 100,00 DDK
Nr. 501: EUDANA - EUtrofiering af Dansk Natur. Videnbehov, modeller og perspektiver. Af Bak, J.L. & Ejrnæs, R. 49 s. (elektronisk)
Nr. 502: Samfundsøkonomiske analyser af ammoniakbufferzoner. Udredning for Skov- og Naturstyrelsen. Af Schou, J.S., Gyldenkærne, S. & Bak, J.L. 36 s. (elektronisk)
Nr. 503: Luftforurening fra trafik, industri og landbrug i Frederiksborg Amt. Af Hertel, O. et al. 88 s. (elektronisk)
Nr. 504: Vingeindsamling fra jagtsæsonen 2003/04 i Danmark. Af Clausager, I. 70 s. (elektronisk)
Nr. 505: Effekt af virkemidler på kvælstofudvaskning fra landbrugsarealer. Eksempel fra oplandet til Mariager Fjord. Thorsen, M. 56 s. (elektronisk)
Nr. 506: Genindvandring af bundfauna efter iltsvindet 2002 i de indre danske farvande. Af Hansen, J.L.S., Josejson, A.B. & Petersen, T.M. 61 s. (elektronisk)
Nr. 507: Sundhedseffekter af luftforurening - beregningspriser. Af Andersen, M.S. et al. 83 s. (elektronisk)

The project studied the long-term time trend of brominated flame retardants, poly-chlorinated biphenyls (PCBs) and organochlorine pesticides in peregrine falcon (Falco peregrinus) eggs. Furthermore, possible effects of the contamination on the eggshell thickness were investigated using multivariate statistical methods. The contamination profile of the eggs was dominated by PCBs and organochlorine pesticides, but the polybrominated diphenyl ethers (PBDEs), including the fully brominated congener BDE-209, were also found in all eggs analysed. All compound groups were found at very high concentrations, reaching median summed concentrations of 55 µg/g lw for PCBs. Indications of an increase in PBDE concentrations during the last 17 years were found, while concentrations of organochlorine compounds seemed to decrease or remain constant. The correlation coefficient between the concentration and the eggshell thickness was negative, indicating a negative influence of the contaminants on the eggshell thickness. Thus, it has not been possible to identify remarkable improvement in the ecotoxicological pressure on the peregrine falcons during the pe-riod of investigation.

National Environmental Research Institute
Ministry of the Environment

ISBN 87-7772-832-7
ISSN 1600-0048