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## Organic Contaminants in Moose (*Alces alces*) and Reindeer (*Rangifer tarandus*) in Sweden from the past twenty years.

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### Comments Concerning the National Swedish Contaminant Monitoring Programme in Terrestrial Biota

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## Sammanfattning

Föreliggande rapport är resultatet av ett uppdrag från Naturvårdsverket (Överenskommelse 221 0730) som syftar till att ta fram en aktuell bild av förekomsten av ett antal pesticider eller ofullständigt kända föroreningar i svensk terrestrisk natur där alltså osäkerhet om ämnenas spridning och ackumulation i landlevande organismvärlden råder. Som matris i detta fall har vävnader och organ av älg och ren från mellersta respektive norra Sverige valts. Matriserna finns tillgängliga i Miljöprovbanken vid Naturhistoriska riksmuseet

Valet av ämnen har väglets av tidigare resultat från övervakningen av föroreningar i svensk natur samt av resultat från screeningundersökningar av ofullständigt kända ämnen som tagits fram i Naturvårdsverkets regi.

Listan över analyserade ämnen innehåller enskilda substanser samt närbesläktade substanser tillhörande bl. a. följande grupper: klorbensen, HCB, oktaklorstyren, PCB, DDT, klordan, HCH, PBDE, dioxiner, furaner, endosulfan, heptaklor, pentaklorfenol och PAH.

Från Grimsö i Västmanland har vävnadsprover av årskalvar av älg insamlats till Miljöprovbanken och använts för analys av innehåll av miljögifter i denna studie. Med några få undantag är de provtagna individerna av hankön. Skillnader i halter av kadmium mellan könen har i tidigare studier visat sig icke existera. De utvalda proven av älg representerar perioden 1986-2005. Ett homogent av muskel har beretts per år under perioden baserat på 10 g av tio individer per år.

På samma sätt har prover av ren från sommarbetet S Abisko årligen insamlats i Rensjön NV Kiruna i samband med första höstslakten i mitten av september. Vävnadsprover har tagits av handjur av en ålder mellan 2 och 4 år, mestadels av 3-åriga djur. 1998 uppsköts den ordinarie slakten från första halvan av september till första halvan av november, vilket bör uppmärksammas vid tolkningen av analysresultaten. På samma sätt sköts slakten 2004 fram till mitten av december. Det förhållandet att renen förflyttas avsevärda sträckor under höstvandringen, från sommarbetet i de västra delarna av fjällkedjan till det mellansvenska skogslandet under vintern medför en ändring av föda, vilket kan inverka på exponering och bioackumulation av de studerade substanserna. Ju längre avvikelse från normal

slakttid desto längre exponering via lavar som är stapelföda i skogslandet under vintern och som innehåller högre koncentrationer av många ackumulerade ämnen.

För den aktuella studien valdes hanrenar av tre års ålder härrörande från Gabna, Lævas and Girjas samebyar vilka bildar ett sammanhängande område i norra Lappland. Provserien omfattar perioden 1987-2006. Ett homogent per år beredd baserat på 12 g muskel från tio individer per år.

Många av de analyserade ämnena förekommer i koncentrationer under detektionsnivån (LOD) under alla eller de flesta av de undersökta åren (Tabell 2). Halterna funna i älg och ren är i många fall lägre än de nivåer som observerats i fisk från såväl limniska som marina områden. Som exempel har PBDE och HBCD inte ens detekterats i vare sig älg eller ren men finns närvarande i både insjöfisk och havsfisk (Bignert et al 2008).

Penta- och hexaklorbensen, oktaklorstyren, polyklorerade bifenyler,  $\alpha$ - och  $\beta$ -hexaklorcyklohexan och polycykliska aromatiska kolväten förekommer samtliga i halter över LOD under samtliga eller större delen av åren under den undersökta perioden. Koncentrationerna av dessa ämnen har sjunkit med undantag för PAH där koncentrationerna är ungefär desamma under hela perioden. Den genomsnittliga årliga nedgången ligger mellan 3% och 10% med undantag för  $\alpha$ -HCH som visar den mest uttalade minskningen med 22% i älg och 17% i ren. Dessa resultat är samstämmiga med vad som observerats i biota från insjöar och marina områden i Sverige (Bignert et al. 2008, Bignert 2001).

De sjunkande halterna i landanknuten biota under de gångna 20 åren är sannolikt orsakade av regleringar och ansträngningar för att minimera utsläpp av kända persistenta, bioackumulerande och toxiska ämnen till miljön.

Ämnen hörande till PAHerna är huvudsakligen oavsiktligt spridda genom ofullständig förbränning av organiska ämnen. Källorna är många och varierande och till följd därav svåra att åtgärda med förbättringar. Detta är troligtvis förklaringen till avsaknaden av nedgående tendens för PAHer.

Koncentrationerna av PFCs i både älg och ren var i allmänhet låga. Samtliga analyserade prover hade koncentrationer under LOQ. Närvaro av PFOSA, PFOS och PFOA kunde emellertid detekteras vissa år men inte i nivåer tillräckligt höga för att kvantifieras.

## Summary

From the Grimsö area (see Figure 1.), tissue samples of male moose calves were selected for analyses with some few exceptions that were from female calves. Completion of male samples with samples of females was acceptable after studies of the relation level/sex for cadmium in kidney, which showed no difference according to sex. Specimens of calves of moose from Grimsö were selected from the period 1986-2005. One homogenate of muscle per year was prepared based on aliquots of 10 g of ten individual moose per year.

Specimens of male reindeer, three years old from Gabna, Lævas and Girjas Sámi Villages were selected from the period 1987-2006. The villages constitute a coherent area in northern Lapland. The reindeer spend the summer in the mountain area S Abisko. One homogenate of muscle per year was prepared based on aliquots of 12 g of ten individual reindeer per year. The slaughtering and sampling of reindeer was carried out between the end of August and mid-October, mostly in the mid of September. The age of the males from which samples were taken ranged mainly between 2 and 4 years. In 1998, the ordinary slaughter in the actual district was postponed from early September to early November, which should be noted when evaluating the time trends of levels. In 2004 the slaughter was delayed until mid-December. The fact that reindeer move from summer areas in the mountains to wintering areas in the forest means a change of diet, which might influence upon the exposition of contaminants via food. A later date for slaughtering means a longer foraging on winter diets that might contain higher concentrations of the actual compounds.

Many of the analysed compounds showed concentrations below the level of detection (LOD) for all or most of the investigated years (Table 2).

The concentrations found in moose and reindeer are in many cases lower than levels observed in fish from both fresh water and marine environments. As an example, PBDEs and HBCD are not even detected in either moose or reindeer but are present in both freshwater and marine fish (Bignert *et al.* 2008).

Penta- and hexa-chlorobenzene, octachlorostyrene, polychlorinated biphenyls,  $\alpha$ - and  $\beta$ -hexachloro-cyclohexane and polycyclic aromatic hydrocarbons were all over LOD for

the whole or the greater part of the investigated time period. The concentrations of these contaminants have decreased except for PAHs where the concentrations are about the same during the whole period. The average annual decrease is between 3% and 10% with exception for  $\alpha$ -HCH showing the most pronounced decrease of 17% in reindeer. These results are in compliance with what is observed in the Swedish marine and freshwater environments (Bignert *et al.* 2008, Bignert 2001).

Decreasing concentrations during the recent 20 years are most likely due to regulations and efforts taken to minimize the contribution of known persistent, bioaccumable and toxic organic compounds.

PAHs are mainly unintentionally emitted during incomplete combustion of organic material, the sources are numerous and diverse and thereby hard to take measures against. This is probably the reason for the lack of decreasing trends for PAHs.

Concentrations of PFCs in both moose and reindeer muscle were generally low. All samples had concentrations below LOQ for the analysed substances. However, concentrations of PFOSA, PFOS and PFOA could be detected some years but not in concentrations high enough to be quantified.

## Aim

This report has been carried out on request of and in cooperation with the Swedish Environmental Protection Agency (SEPA) to be part of the National Swedish Contaminant Monitoring Programme in Terrestrial Biota. The aim is to reveal the state of the art of contamination in terrestrial environments by screening two well-known matrices – moose (*Alces alces*) and reindeer (*Rangifer tarandus*) – according to some chlorinated, brominated and fluorinated compounds. The aim is also to follow any changes in concentration over time during the period 1986-2006.

## Organisation

The analysed material is collected by the Swedish Museum of Natural History, Dept of Contaminant Research (DCR) and stored in the Environmental Specimen Bank (ESB) in cooperation with the Swedish EPA as part of the National Swedish Monitoring Programmes. Chemical analyses have been carried out in cooperation with Eva Brorström-Lundén och Mikael Remberger, IVL Swedish Environmental Research Institute Ltd. Sara Danielsson, Tjelvar Odsjö and Anders Bignert at the DCR have analysed and evaluated the results and prepared the report, partly in cooperation with the chemists.

The study has been carried out also in cooperation with Britta Hedlund, Jonas Rodhe and Axel Hullberg, SEPA who gave the financial support to the study.

## Introduction

National and regional monitoring of pollution of contaminants in the Swedish environment comprises studies of the body burden of bio-accumulated substances in biota from terrestrial and freshwater reference areas in the Swedish mainland and from the surrounding seas and coastal areas (Odsjö and Olsson 1979a,b, Bernes 1985). Primarily, monitoring of pollutants aims at studying long-term changes of concentrations in the environment (trend monitoring) as well as spatial variation. Trend monitoring is considered as an important instrument for studies of the general bioaccumulation due to national and international use as well as for measures against use of different pollutants in order to minimise pollution of nature. By use of data from a net of localities, the transport and geographical distribution of contaminants is possible to study.

As part of the terrestrial contaminant monitoring programme, specimens of muscle, liver and kidney of moose have been collected since 1980 from Grimsö district in the Örebro county (T) in south-central Sweden (see Figure 1).

Moose, with a diet dominated by twigs and leaves of trees and shrubs (Cederlund *et al.* 1980), was chosen in the monitoring programme as a representative of biota in the Swedish forest areas. Since the moose is distributed almost all over the country, it was considered as an ideal matrix also for studies of spatial distribution of environmental pollution and bioaccumulation, which was the reason for a later on extended collection of samples in 1996-2006.

In the mountainous area of north-western Sweden, reindeer (*Rangifer tarandus*) is chosen as a representative indicator for the fauna living in that part of the country. Samples of reindeer have continuously been collected in three districts since the early 1980s. Later, the Declaration on the Protection of the Arctic environment established an Arctic Monitoring and Assessment Programme (AMAP) to monitor levels and assess effects of anthropogenic pollutants in components of the Arctic environment. The Programme recommends that collection of baseline data for heavy metals and radionuclides in caribou/reindeer should be mandatory for participating states due to the importance of that species in the diets of northern native people (AMAP 1993). The current material of reindeer from northern and

central Lapland partly satisfies the Swedish participation in the AMAP programme (see Figure 1).

Beside annual studies of an established cluster of contaminants in different matrices of the programmes, special studies are regularly carried out in order to screen new and/or poorly investigated substances judged as presumptive contaminants. The present study can be regarded as a screening study to reveal new threats of contaminants in remote or background areas of Sweden.

Monitoring studies of contaminants in terrestrial environments have earlier been carried out by use of moose and reindeer. The real time and retrospective studies comprised DDT- and PCB-substances, metals and trace elements (Odsjö *et al.* 2007a,b). Time trends of HCHs and HCB in reindeer have earlier been studied in a series from 1983-1995 (Odsjö *et al.* 1998) as well as for analyses of *e.g.* radiocesium in a study of effects of the fallout of Cs-132 from the Chernobyl accident in 1986 (Forberg *et al.* 1992). The present study is a retrospective time-trend study based on material collected in 1986-2006. Based on results from this study, decision may be taken to go further with long time retrospective trend studies.

## Material

### Specimen collection

#### *Moose.*

From the start of the collection in the Grimsö area (Figure 1), samples of liver, kidney and muscle have been collected from approximately 45-50 individuals annually during the hunting season in the autumn and, with special permit also in the winter and spring. Samples were taken from all individuals shot in the area during hunting despite age and sex. This was done to make it possible to select the most appropriate and homogeneous material for contaminant monitoring according to influence of biological variables (*e.g.* age, sex, etc.) on the concentrations. The samples were extracted at the slaughter, prepared in laboratory and stored in a temperature of -30°C until analysis. Individual age was determined by tooth sectioning after slaughter. Calves and, certain seasons also males were initially well represented in the material. However, the age structure of the material has changed considerably during the period, which may have consequences in future for the choice of appropriate and homogeneous material from a

smaller and spatial concentrated population like that in the Grimsö area. According to the extended hunting period and date of collection, selection of individual calves for analyses was restricted to the period October 1 - April 30 each hunting season. The selection of specimens started with the earliest shot animals each season. No significant variation in levels of Cd according to date of collection during the hunting season was revealed in an earlier study (Odsjö 2001).

From the Grimsö area, tissue samples of male calves were selected for analyses with some few exceptions that were from female calves. Completion of male samples with samples of females was acceptable after studies of the relation level/sex for cadmium in kidney, which showed no difference according to sex. Further tests and discussion of selection criteria of matrices of moose are reported elsewhere (Odsjö 2001).

#### *Reindeer.*

The herbivorous reindeer spend the summer time in the westernmost part of the high mountain areas S Abisko. Summer diets include grasses, sedges, twigs, leaves and mushrooms. During autumn they migrate eastwards to winter grounds in the central coniferous forest areas of the country, where they primarily feed on lichens, which are noted for their ability to accumulate nutrients and contaminants from the air. Winter diets also include sedges and twigs.

Since the start of the monitoring programme, collection of specimens of muscle, liver, kidney and left under jaw with teeth (for age determination) from at least 50 male reindeer has been carried out annually at regular slaughtering in each of three selected districts. In 1983-1986, the muscle samples were taken from the mandibles. After that, routines were changed and muscle samples were taken from the front leg tibia. The change of muscle samples led us to exclude mandible muscle in the present study since they were not comparable. Directly after collection in field, the specimens were frozen at a temperature of about -20°C (some at -80°C) and were then shipped frozen to the laboratory.

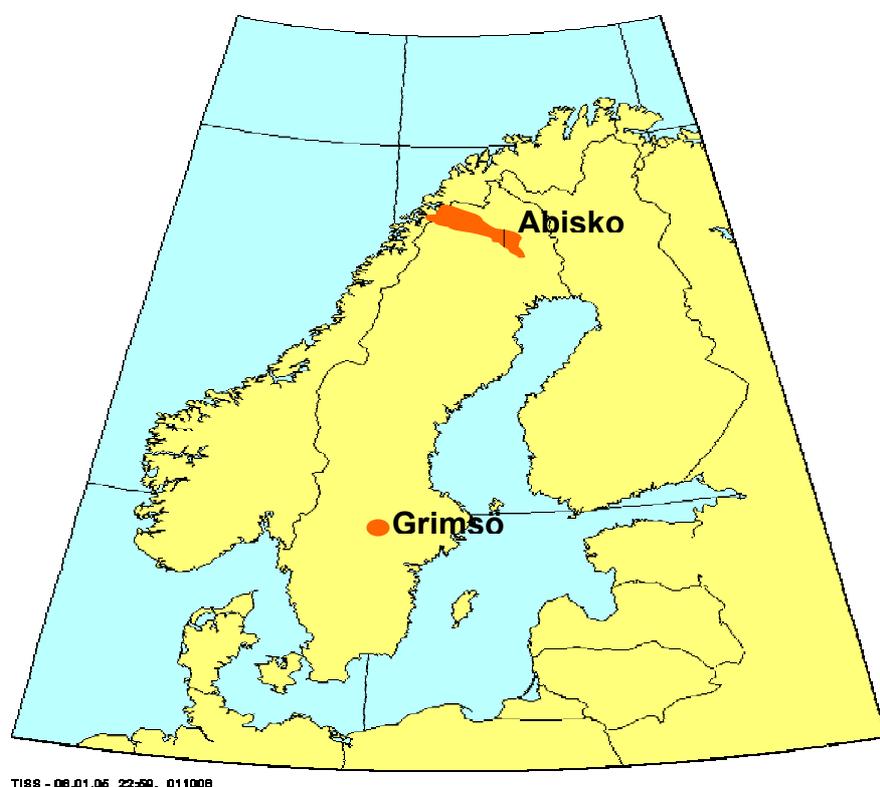
The slaughtering was carried out between the end of August and mid-October, mostly in the mid of September. The age of the males from which samples were taken ranged mainly between 2 and 4 years. Most specimens were 3 years old. Ages were determined in the lab after slaughter. In 1998, the ordinary slaughter in the northernmost district was postponed from early September to early November, which should be noted when evaluating the time trends of levels. In 2004 the slaughter was delayed

until mid-December. The fact that reindeer move from summer areas in the mountains to wintering areas in the forest means a change of diet, which might influence upon the exposition of contaminants via food. A later date for slaughtering means a longer foraging on winter diets that might contain higher concentrations of the studied compounds.

### Localities

Specimens of muscle, liver and kidney of moose have been collected since 1980 in Grimsö, a reference area in the monitoring programme and a coherent hunting district in the Örebro county (T) in south-central Sweden (Figure 1).

Collection of specimens of reindeer have annually been carried out in three districts along the Swedish, easternmost mountain chain. In the present study specimens were chosen of reindeer from the northernmost district, Gabna, Lævas and Girjas Sámi Villages. The district, reaches from the Swedish/Norwegian border eastwards to the central forest areas in the northern part of Lapland (Figure 1) and is referred to as Abisko (summer stay area) in this report.



**Figure 1.** Map showing the collection localities. Samples of reindeer are collected in the Abisko district and samples of moose in the Grimsö area.

### Tissue samples

Specimens of male calves of moose from Grimsö were selected from the period 1986-2005. One homogenate of muscle per year was prepared based on aliquots of 10 g of ten individual moose per year.

Specimens of male reindeer, three years old from Gabna, Lævas and Girjas Sámi Villages were selected from the period 1987-2006. One homogenate of muscle per year was prepared based on aliquots of 12 g of ten individual reindeer per year.

## Statistical treatment and graphical presentation

### Trend detection

One of the main purposes of the present investigation is to detect trends.

The slope of the line in the presented graphs describes the annual change. A slope of 5% implies that the concentration is halved in 14 years whereas 10% corresponds to a similar reduction in 7 years and 2% in 35 years. See Table 1 below.

**Table 1.** The approximate number of years required to double or half the initial concentration assuming a continuous annual change of 1, 2, 3, 4, 5, 7, 10, 12, 15 or 20% a year.

	1%	2%	3%	4%	5%	7%	10%	12%	15%	20%
Increase	70	35	24	18	14	10	7	6	5	4
Decrease	69	35	23	17	14	10	7	6	4	3

### Legend to the plots

The analytical results from many of the investigated elements are displayed in figures. A separate plot represents each matrix. The plot displays the concentration in the annual homogenate.

The overall geometric mean value for the time series is depicted as a horizontal, thin, dashed line.

The trend is presented by one regression line (plotted if  $p < 0.05$ , two-sided regression analysis); Ten years is often too short a period to statistically detect a trend unless it is of considerable magnitude.

The log-linear regression lines fitted through the geometric mean concentrations follow smooth exponential functions.

Each plot has a header with specimen, locality name and analysed compound. Below the header of each plot the results from several statistical calculations are reported:

**n(tot)** = The first line reports the total number of analyses included together with the number of years (  $n(\text{yrs})=$  ). Note that values below the limit of detection are included in this number.

**m** = The overall geometric mean value together with its 95% confidence interval is reported on the second line of the plot (N.B. d.f.= n of years - 1).

**slope** = reports the slope, expressed as the annual change together with its 95% confidence interval.

**SD(lr)** = reports the square root of the residual variance around the regression line, as a measure of between-year variation, together with the lowest detectable change in the current time series with a power of 80%, one-sided test,  $\alpha=0.05$ . The last figure on this line is the estimated number of years required to detect an annual change of 5% with a power of 80%, one-sided test,  $\alpha=0.05$ .

**power** = reports the power to detect a log-linear trend in the time series (Nicholson & Fryer, 1991). The first figure represents the power to detect an annual change of 5% with the number of years in the current time series. The second figure is the power estimated as if the slope were 5% a year and the number of years were ten. The third figure is the lowest detectable change for a ten-year period with the current between year variation at a power of 80%.

**y(06)** = reports the concentration estimated from the regression line for the last year together with a 95% confidence interval, e.g.  $y(06)=2.51(1.92,3.27)$  is the estimated concentration of year 2006 where the residual variance around the regression line is used to calculate the confidence interval. Provided that the regression line is relevant to describe the trend, the residual variance might be more appropriate than the within-year variance in this respect.

**r<sup>2</sup>** = reports the coefficient of determination (r<sup>2</sup>) together with a p-value for a two-sided test (H<sub>0</sub>: slope = 0) i.e. a significant value is interpreted as a true change, provided that the assumptions of the regression analysis is fulfilled.

## Analysed compounds

The analyses carried out comprise compounds and groups of compounds that are known to be persistent and accumulating in biota. Some substances are known to be toxic and are widely spread in nature (PBT compounds). Results from earlier screening studies enables some to be studied further in trend studies. Analysed groups and congeners are listed in Table 2.

**Table 2.** Compounds analysed in muscle of moose and reindeer.

<b>Group</b>	<b>Name/Congener</b>
Chlorobenzene	1,2,3-Trichlorobenzene
	1,2,4-Trichlorobenzene
	1,3,5-Trichlorobenzene
	1,2,3,4-Tetrachlorobenzene
	1,2,3,5-Tetrachlorobenzene
	1,2,4,5-Tetrachlorobenzene
	Pentachlorobenzene
	Hexachlorobenzene
HCBD	Hexachlorobutadiene
Octachlorostyrene	Octachlorostyrene
PCB	PCB 28
	PCB 52
	PCB 101
	PCB 118
	PCB 153
	PCB 138
	PCB 180

DDT	DDT
	DDE
	DDD
Chlordane	$\gamma$ -Chlordane
	$\alpha$ -Chlordane
	Transnonachlor
HCHs	$\alpha$ -hexachlorocyclohexane
	$\beta$ -hexachlorocyclohexane
	$\gamma$ -hexachlorocyclohexane
PBDE Polybrominated diphenyl ethers	PBDE 47
	PBDE 100
	PBDE 99
	PBDE 85
	PBDE 209 (Deca)
HBCD	Hexabromocyclododecane
Tributylphosphate	Tri-iso-butylphosphate, TIBP
	Tri-n-butylphosphate, TBP
Polychlorinated dibenzo-p-dioxins (PCPP)	2378-TCDD
	12378-PeCDD
	123478-HxCDD
	123678-HxCDD
	123789-HxCDD
	1234678-HpCDD
	OCDD
Endosulfan	$\alpha$ -Endosulfan
	$\beta$ -Endosulfan
	Endosulfansulphate
Heptachlor	Heptachlor
	Heptachlor-exo-epoxide
	Heptachlor-endo-epoxide
	Aldrin
	Endrin

	Dieldrin
Chlorophenols	Pentachlorophenol
Polychlorinated dibenzofuranes (PCDF)	2378-TCDF
	12378/12348-PeCDF
	23478-PeCDF
	123478/123479-HxCDF
	123678-HxCDF
	123789-HxCDF
	234678-HxCDF
	1234678-HpCDF
	1234789-HpCDF
	OCDF
PAH	Naphthalene
	Acenaphthene
	Fluorene
	Phenanthrene
	Anthracene
	Fluoranthene
	Pyrene
	Benzo(a)anthracene
	Chrysene
	Benzo(b)fluoranthene
	Benzo(k)fluoranthene
	Benzo(a)pyrene
	Dibenso(a,h)anthracene
	Benso(g,h,i)perylene
	Indeno(1,2,3-cd)pyrene
Nonortho-PCB	33'44'-TeCB (PCB-77)
	344'5-TeCB (PCB-81)
	33'44'5-PeCB (PCB-126)

Sum PCDD/PCDF	TE(Nordic)
	i-TE
	TE(WHO)
Sum TE-PCB	TE(WHO)
PFC	6:2 FTS
	PFOSA
	PFBS
	PFHxS
	PFOS
	PFDcS
	PFBA
	PFHxA
	PFHpA
	PFOA
	PFNA
	PFDcA
	PFUnA

Analysis of PAH, pentachlorophenol, endosulfan, tributylphosphate, tri-iso-butylphosphate, heptachlor, aldrin, heptachlorepoxyd, endrin and dieldrin.

### Extraction

Sample of muscle from elk or reindeer was homogenised in pre-heated sodium sulphate. Recovery standards for the different analytes were added. The homogenate was acidified with formic acid and subsequently extracted four times with dichloromethane. The pooled extract was concentrated by the aid of a RotoVap and finally with a gentle stream of pre-cleaned (activated carbon) nitrogen. Formic acid and other polar compounds were removed by shaking the extract with water. The final extract was dried over sodium sulphate and used for analysis of a broad spectrum of compounds. A part of the extract was submitted to gravimetric analysis to estimate lipid content.

### Fractionation of the extract in different substance classes

In a first step lipids were separated from the extract by partition between hexane and acetonitril (ACN). The hexan phase was discarded and the ACN-extract containing all

the target analytes was diluted with water and extracted with a mixture of hexane:MTBE. The obtained extract, containing all the target analytes, was further fractionated into three fractions on a silica column. Fraction 1 (F-1) contained polycyclic aromatic hydrocarbons (PAH) and the pesticides (heptachlor, aldrin, heptachlorepoxyd, endrin and dieldrin). Fraction 2 (F-2) contained tri-butyl phosphate (TBP), tri-*iso*-butyl phosphate (TIBP) and endosulfan. Fraction 3 (F-3) contained pentachlorophenol (PCP). F-1 was first analysed for pesticides using gas chromatography equipped with an electron capture detector (GC-ECD) and thereafter for PAH using a high performance liquid chromatograph with a fluorescence detector (HPLC-FD). F-2 and F-3 were analysed with gas chromatography connected to a mass selective detector (GC-MS).

### Analysis of PCB, chlorobenzenes, PBDEs, HBCD, octachlorstyrene, DDT, and HCHs.

#### **Extraction**

The sample was homogenized in sodium sulphate and solvent extracted in an ultrasonic bath with acetone. Water was added to the acetone extract and the mixture was extracted with pentan:diethyl ether twice. The extract was dried over sodium sulphate and concentrated. The extract, dissolved in pentane, was first purified by treatment with concentrated sulphuric acid. In a second step the sulphuric acid treated extracts was chromatographed on acidic aluminium oxide prior to GC-ECD analysis. Three fractions were collected from the column. In fraction 1 polychlorinated biphenyl's (PCB), octachlorstyrene (OCS) and chlorobenzene were collected. Fraction 2 contained DDT and hexachloro cyclohexanes ( $\alpha$ -,  $\beta$ -,  $\gamma$ -HCH) and all polybrominated diphenyl ethers (PBDE) but the fully brominated PBDE-209. Finally, fraction 3 contained PBDE-209 and hexacabromocyclododecane (HBCD).

PCB, DDT, HCH and the congeners PBDE-47 to PBDE-154 were analysed on a 30 m capillary column (0.25 mm ID and 0.25  $\mu$ m phase) on a GC equipped with ECD. The thermal labile compounds HBCD and BDE-209 were chromatographed on a short column (15 m, 0.25 mm ID; 0.12  $\mu$ m phase) with a thin phase to minimize the run time and thereby the thermal degradation.

Chlorobenzene was analysed on GC-MS in negative chemical ionisation mode (NCI).

### Analysis of polychlorinated dibenzo-p-dioxins (PCDD) and dibenzofurans (PCDF)

<sup>13</sup>C-labeled 2,3,7,8-chloro substituted PCDD and PCDF congeners were added to the sample and mixed well in a homogeniser. The sample was extracted in a Soxhlet apparatus with toluene. The extract was concentrated and subjected to lipid reduction on a column containing a multifunctional phase of silica in conjunction with a column containing activated carbon. The final clean-up of the extract was performed on silica coated with aluminium oxide. Prior to analysis by GC high resolution mass spectrometry (res> 1000) in electron impact ionisation mode (EI) an injection standard was added (method NILU-O-1 accredited after ISO/IEC-17025).

### Analysis of perfluorinated substances

Homogenized tissue was prepared using an Ultra-Turrax. The homogenised sample was extracted with acetonitrile. Internal standards were added prior to sample clean-up. The clean up was performed with graphitized carbon and the final extract was analysed by HPLC electrospray ionization quadrupole time-of-flight mass spectrometry (ESI-QTOF).

For further details of the analytical method see Kallenborn *et al.* (2004).

## Results

The concentrations of many of the analysed compounds were below the level of detection (LOD) right through the time series. For some other compounds we found detectable concentration for some years in an irregular pattern. The LOD and the occurrence of quantifiable and non-quantifiable concentrations is given in Table 3, below.

**Table 3.** Compounds for which concentrations in muscle were found to be below the LOD ng/g wet weight. Detectable Concentrations = DC

Group	Name/Congener	Below LOD Moose	Below LOD Reindeer
Chlorobenzene	1,2,3-Trichlorobenzene	<0.1	<0.1
	1,2,4-Trichlorobenzene	<0.1	<0.1
	1,3,5-Trichlorobenzene	<0.1	<0.1
	1,2,3,4-Tetrachlorobenzene	<0.1	<0.1
	1,2,3,5-Tetrachlorobenzene	<0.1	<0.1
	1,2,4,5-Tetrachlorobenzene	<0.1	<0.1
	Pentachlorobenzene	DC	DC
	Hexachlorobenzene	DC	DC
HCBD	Hexachlorobutadiene	<0.1	<0.1
Octachlorostyrene	Octachlorostyrene	DC	DC
PCB	PCB 28	<0.01	<0.01
	PCB 52	<0.01	<0.01
	PCB 101	<0.01	<0.01
	PCB 118	DC	<0.01/DC <sup>6</sup>
	PCB 153	DC	DC
	PCB 138	DC	DC
	PCB 180	DC	DC
DDT	DDT	<0.01	<0.01
	DDE	DC	<0.01/DC <sup>3</sup>
	DDD	<0.01	<0.01
Chlordane	γ-Chlordane	<0.01	<0.01
	α-Chlordane	<0.01	<0.01
	Transnonachlor	<0.01	<0.01
HCHs	α-hexachlorocyclohexane	<0.01/DC <sup>1</sup>	<0.01/DC <sup>7</sup>
	β-hexachlorocyclohexane	DC	DC

	$\gamma$ -hexachlorocyclohexane	<0.01/DC <sup>2</sup>	<0.01/DC <sup>8</sup>
PBDE Polybrominated diphenyl ethers	PBDE 47	<0.01/DC <sup>3</sup>	<0.01
	PBDE 100	<0.01/DC <sup>4</sup>	<0.01
	PBDE 99	<0.01/DC <sup>3</sup>	<0.01
	PBDE 85	<0.01	<0.01
	PBDE 209 (Deca)	<0.1	<0.1
	HBCD	Hexabromocyclododecane	<0.1
Tributylphosphate	Tri-iso-butylphosphate, TIBP	<2/DC <sup>9</sup>	<0.2
	Tri-n-butylphosphate, TBP	<2	<1.5/DC <sup>3</sup>
Polychlorinated dibenzo-p-dioxins (PCPP)	2378-TCDD	<0.01/DC	<0.01
	12378-PeCDD	<0.02/DC	<0.01
	123478-HxCDD	<0.02	<0.01
	123678-HxCDD	<0.02	<0.01
	123789-HxCDD	<0.02	<0.01
	1234678-HpCDD	<0.02/DC	<0.01/DC
	OCDD	DC	DC
Endosulfan	$\alpha$ -Endosulfan	<0.2	<0.6
	$\beta$ -Endosulfan	<0.2	<0.4
	Endosulfansulphate	<0.2	<0.2
Heptachlor	Heptachlor	<0.2	<2
	Heptachlor-exo-epoxide	<0.2	<2
	Heptachlor-endo-epoxide	<0.3	<3
	Aldrin	<0.2	<1.4
	Endrin	<0.6	<7.5
	Dieldrin	<0.4	<2.8
Chlorophenols	Pentachlorophenol	<0.05	<0.1
Polychlorinated dibenzofuranes	2378-TCDF	DC	<0.01/DC

(PCDF)			
	12378/12348-PeCDF	<0.01/DC	<0.01
	23478-PeCDF	<0.01/DC	<0.01
	123478/123479-HxCDF	DC	<0.01/DC
	123678-HxCDF	DC	<0.01/DC
	123789-HxCDF	<0.02/DC	<0.01
	234678-HxCDF	<0.01/DC	<0.01/DC
	1234678-HpCDF	DC	DC
	1234789-HpCDF	<0.01/DC	<0.01/DC
	OCDF	DC	<0.02/DC
PAH	Naphthalene	<0.1	--
	Acenaphthene	<0.01	<0.01
	Fluorene	<0.005/DC <sup>1</sup>	<0.04/DC <sup>7</sup>
	Phenanthrene	<0.02/DC <sup>7</sup>	<0.02/DC <sup>12</sup>
	Anthracene	<0.001/DC <sup>10</sup>	<0.003/DC <sup>13</sup>
	Fluoranthene	<0.01/DC <sup>6</sup>	<0.01/DC <sup>14</sup>
	Pyrene	<0.01/DC <sup>11</sup>	<0.01/DC <sup>15</sup>
	Benzo(a)anthracene	<0.01/DC <sup>3</sup>	<0.01/DC <sup>13</sup>
	Chrysene	<0.01/DC <sup>4</sup>	<0.01/DC <sup>13</sup>
	Benzo(b)fluoranthene	<0.006/DC <sup>3</sup>	<0.01/DC <sup>13</sup>
	Benzo(k)fluoranthene	<0.01/DC <sup>4</sup>	<0.004/DC <sup>13</sup>
	Benzo(a)pyrene	<0.005/DC <sup>4</sup>	<0.004/DC <sup>3</sup>
	Dibenso(a,h)anthracene	<0.005/DC <sup>4</sup>	<0.004/DC <sup>4</sup>
	Benso(g,h,i)perylene	<0.01/DC <sup>4</sup>	<0.01/DC <sup>3</sup>
	Indeno(1,2,3-cd)pyrene	<0.01/DC <sup>4</sup>	<0.008/DC <sup>3</sup>
Nonortho-PCB	33'44'-TeCB (PCB-77)	DC	DC
	344'5-TeCB (PCB-81)	DC	DC
	33'44'5-PeCB (PCB-126)	DC	DC
Sum PCDD/PCDF	TE(Nordic)	DC	DC
	i-TE	DC	DC
	TE(WHO)	DC	DC
Sum TE-PCB	TE(WHO)	DC	DC

PFC	6:2 FTS	<0.39-0.80 <sup>16</sup>	<0.32-1.07 <sup>16</sup>
	PFOSA	<0.06 <sup>9</sup>	<0.06 <sup>3</sup>
	PFBS	<0.05-0.23 <sup>16</sup>	<0.04-0.40 <sup>16</sup>
	PFHxS	<0.02-0.05 <sup>4</sup>	<0.02-0.06 <sup>4</sup>
	PFOS	<0.10 <sup>17</sup>	<0.10 <sup>11</sup>
	PFDCS	<0.02-0.04 <sup>16</sup>	<0.02-0.04 <sup>16</sup>
	PFBA	<0.05-0.09 <sup>4</sup>	<0.05-0.10 <sup>16</sup>
	PFHxA	<0.17-0.58 <sup>16</sup>	<0.14-1.65 <sup>16</sup>
	PFHpA	<0.15-0.40 <sup>16</sup>	<0.17-0.41 <sup>16</sup>
	PFOA	<0.17-0.51 <sup>4</sup>	<0.16-0.34 <sup>16</sup>
	PFNA	<0.17-0.36 <sup>16</sup>	<0.17-0.38 <sup>16</sup>
	PFDCA	<0.67-1.42 <sup>16</sup>	<0.53-1.80 <sup>16</sup>
	PFUnA	<0.76 <sup>16</sup>	<0.76 <sup>16</sup>

<sup>1/</sup> 14 years above LOD

<sup>2/</sup> 5 years above LOD

<sup>3/</sup> 2 years above LOD

<sup>4/</sup> 1 year above LOD

<sup>5/</sup> Not analysed

<sup>6/</sup> 16 years above LOD

<sup>7/</sup> 17 years above LOD

<sup>8/</sup> 10 years above LOD

<sup>9/</sup> 3 years above LOD

<sup>10/</sup> 8 years above LOD

<sup>11/</sup> 15 years above LOD

<sup>12/</sup> 18 years above LOD

<sup>13/</sup> 4 years above LOD

<sup>14/</sup> 12 years above LOD

<sup>15/</sup> 13 years above LOD

<sup>16/</sup> 0 years above LOD

<sup>17/</sup> 7 years above LOD

## Chlorobenzene

Chlorobenzene is a group of aromatic organic compounds. They are colorless, flammable liquids. They are common solvents and a widely used intermediate in the manufacture of other chemicals e.g. many pesticides. Tri- and tetra-chlorobenzenes were all below the LOD.

### *Pentachlorobenzene (PeCIBz)*

Pentachlorobenzene is a crystalline compound used e.g. for manufacturing other chemicals used for defeating pests (fungicide). In addition, it has been and is currently used as a fire retardant. Pentachlorobenzene is also produced unintentionally under incomplete combustion conditions. Pentachlorobenzene has a relative high bioaccumulation potential due to its high lipophilicity (log Kow=5) and a long half-life time in biota (Howard, 1991).

As in the case of HCB the concentrations of PeCIBz were above LOD for all years in the time series and tend to decrease significantly ( $p < 0.001$ ) in muscle of moose with 9.4% annually but not for reindeer (Figure 2 and 3). The concentrations of PeCIBz are not abnormally high in 1998 and 2004 as is the case for HCB, due probably in that case to contaminated winter diet.

### *Hexachlorobenzene (HCB)*

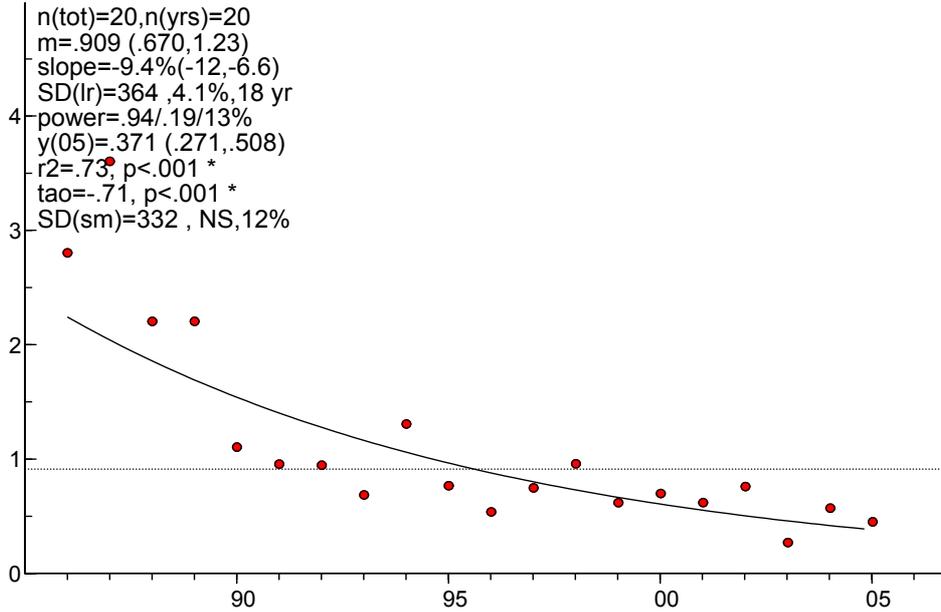
Hexachlorobenzene is a fungicide formerly used as a seed dressing agent, especially on wheat to control the fungal diseases. It has been banned globally under the Stockholm Convention on persistent organic pollutants. Hexachlorobenzene is also produced unintentionally under incomplete combustions conditions.

Hexachlorobenzene has a relative high bioaccumulation potential due to its high lipophilicity ( $\log K_{ow} = 5.5$ ) and a long half-life time in biota (Niimi 1987)

Concentrations of HCB were above LOD all years in the time series for both moose and reindeer. The concentrations in moose tend to decrease significantly with 6.7% annually ( $p < 0.001$ ) but not for reindeer (Figure 4 and 5). The concentrations of HCB in reindeer from 1998 and 2004 were abnormally high compared to the other years in the time series, which may be a result of late slaughter and two to three month of feeding on more contaminated winter food (e.g. lichens). If we on acceptable reasons exclude the data from these two years the statistics show a significant annual decrease of 3.3% ( $p < 0.006$ ).

Moose, Grimso

Pentachlorobenzene ng/g lw

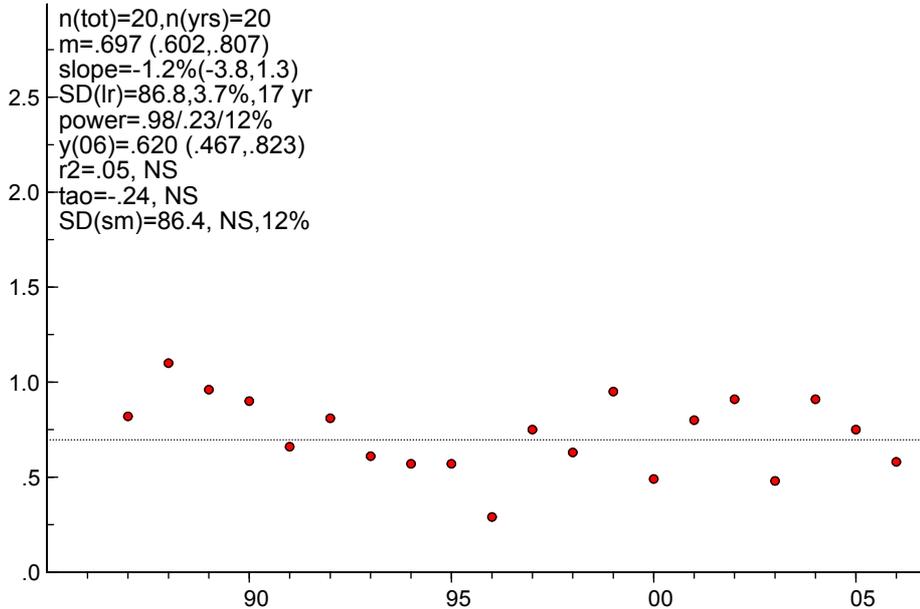


NRM, Dep. of Contaminant Research 08.08.21 13:10, ClBz\_alg

**Figure 2. Levels of pentachlorobenzene (ng/g, lipid weight) in muscle of moose from Grimsö. - - - - - indicates the overall geometric mean level over the period 1986-2005.**

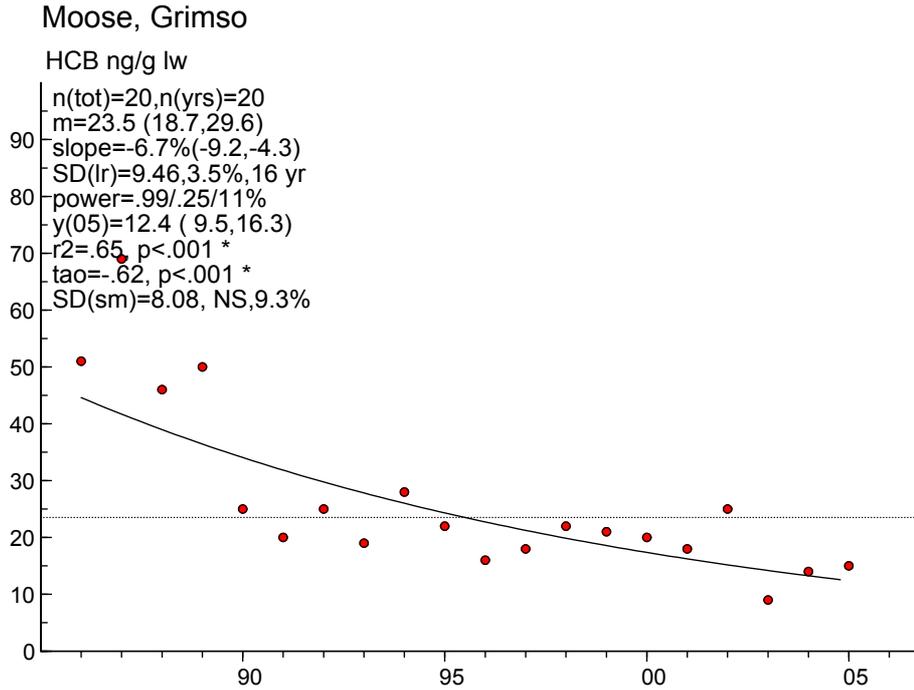
Reindeer, Abisko

Pentachlorobenzene ng/g lw



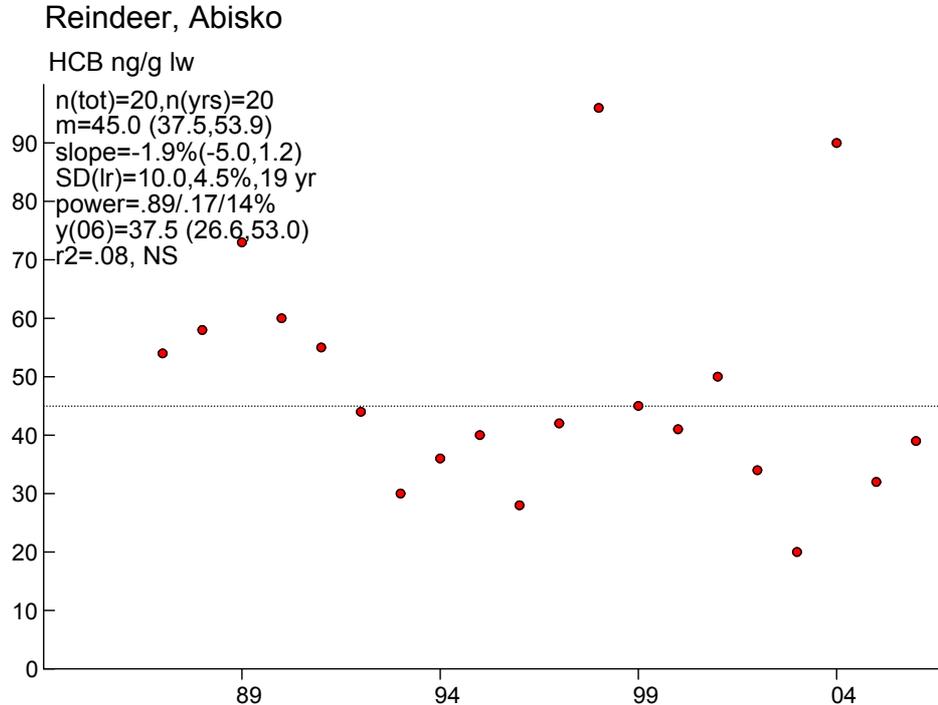
NRM, Dep. of Contaminant Research 08.08.21 13:17, ClBz\_ren

**Figure 3. Levels of pentachlorobenzene (ng/g, lipid weight) in muscle of reindeer from Abisko. - - - - - indicates the overall geometric mean level over the period 1987-2006.**



NRM, Dep. of Contaminant Research 08.08.21 12:55, HCB\_alg

**Figure 4. Levels of HCB (ng/g, lipid weight) in muscle of moose from Grimsö.**  
 - - - - - indicates the overall geometric mean level over the period 1986-2005.



NRM, Dep. of Contaminant Research 08.08.21 13:06, HCB\_ren

**Figure 5. Levels of HCB (ng/g, lipid weight) in muscle of reindeer from Abisko.**  
 - - - - - indicates the overall geometric mean level over the period 1987-2006.

### Hexachlorobutadiene HCB

HCB is used in a variety of industrial contexts as solvent, hydraulic oil, in manufacturing of rubber and production of chlorine and as a fungicide in cultivation of grapes. HCB is mainly produced as a by-product in production of chlorinated compounds like tri- and tetrachloroethane and tetrachloromethane (Botta 1996). HCB was recovered from this by-product. The commercial production of HCB was terminated in the 70<sup>th</sup>.

Concentrations in both moose and reindeer were below the LOD in all analysed years.

### Octachlorostyrene

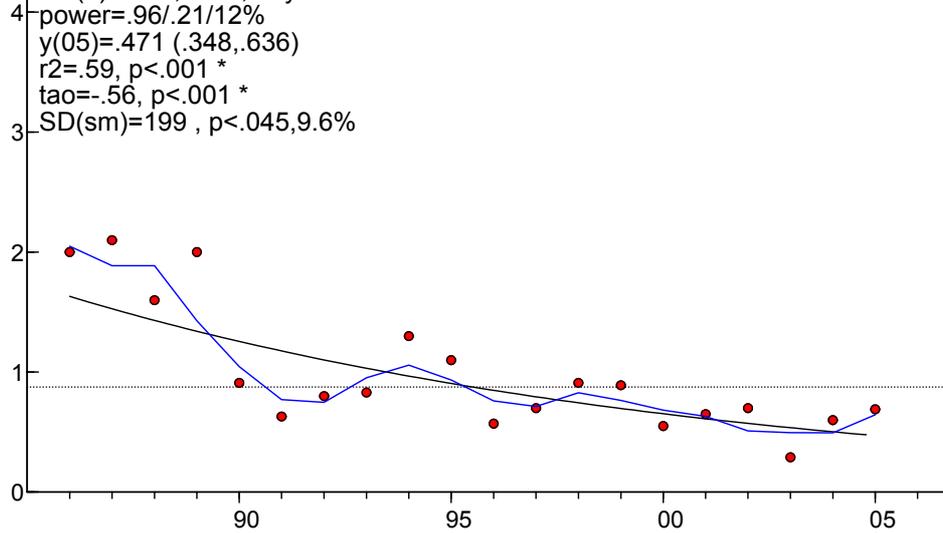
Octachlorostyrene is a persistent, bioaccumulative, and toxic halogenated aromatic compound. It is not commercially manufactured, but has been reported to be an inadvertent by-product of processes that combine carbon and chlorine, under elevated temperatures.

Concentrations of octachlorostyrene were above LOD all years in the time series for both moose and reindeer. The concentrations in both moose and reindeer tend to decrease significantly with 6.5% annually ( $p < 0.001$ ) for moose and with 3.5% annually ( $p < 0.003$ ) for reindeer (Figure 6 and 7). The concentrations in reindeer from 1998 and 2004 tend to be somewhat elevated most likely due to the later date for slaughter those years and a longer foraging on winter diets.

Moose, Grimso

Octachlorostyrene ng/g lw

n(tot)=20,n(yrs)=20  
 m=.876 (.692,1.11)  
 slope=-6.5%(-9.2,-3.8)  
 SD(lr)=251,3.9%,17 yr  
 power=.96/.21/12%  
 y(05)=.471 (.348,.636)  
 r2=.59, p<.001 \*  
 tao=-.56, p<.001 \*  
 SD(sm)=199, p<.045,9.6%



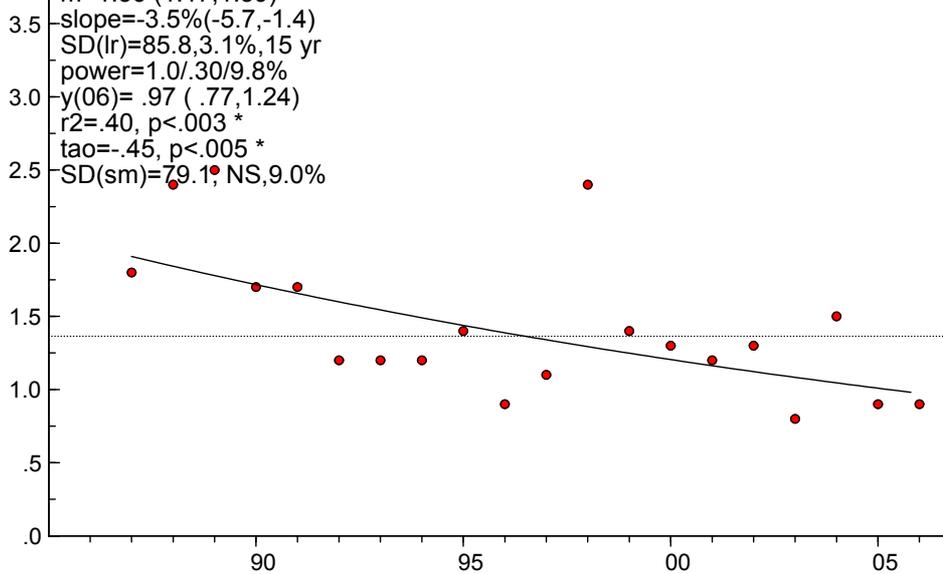
NRM, Dep. of Contaminant Research 08.08.21 13:18, OCLS\_alg

**Figure 6. Levels of octachlorostyrene (ng/g, lipid weight) in muscle of moose from Grimso. - - - - indicates the overall geometric mean level over the period 1986-2005.**

Reindeer, Abisko

Octachlorostyrene ng/g lw

n(tot)=20,n(yrs)=20  
 m=1.36 (1.17,1.59)  
 slope=-3.5%(-5.7,-1.4)  
 SD(lr)=85.8,3.1%,15 yr  
 power=1.0/.30/9.8%  
 y(06)=.97 (.77,1.24)  
 r2=.40, p<.003 \*  
 tao=-.45, p<.005 \*  
 SD(sm)=79.1, NS,9.0%



NRM, Dep. of Contaminant Research 08.08.21 13:20, OCLS\_ren

**Figure 7. Levels of octachlorostyrene (ng/g, lipid weight) in muscle of reindeer from Abisko. - - - - indicates the overall geometric mean level over the period 1987-2006.**

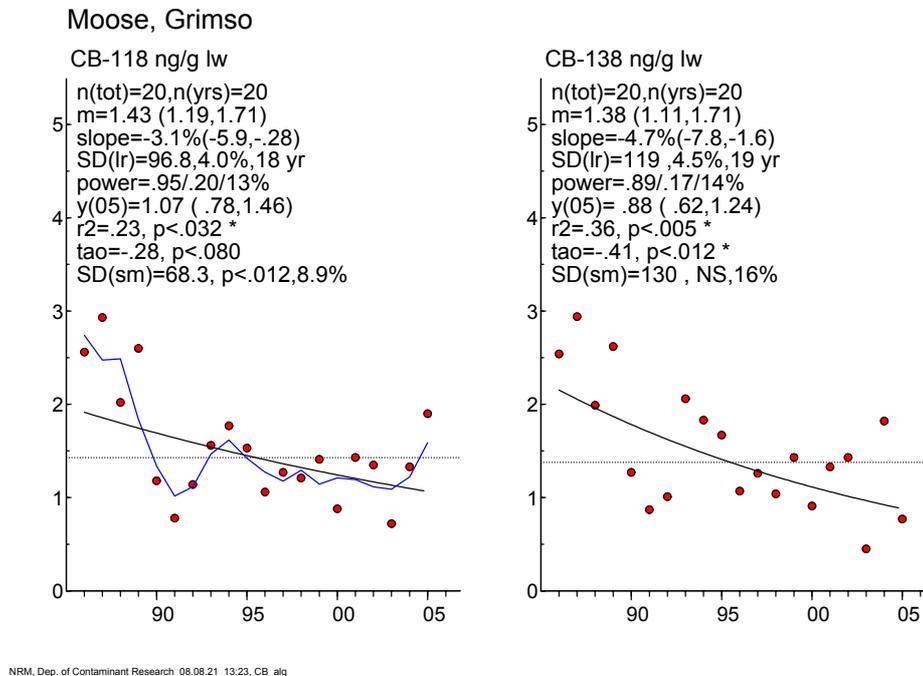
## Polychlorinated biphenyls, PCBs

These compounds are used in industry as heat exchange fluids, in electric transformers and capacitors, and as additives in paint, carbonless copy paper, sealants and plastics.

The concentrations of CB-28, CB-52 and CB-101 were all below the LOD in both moose and reindeer while CB-118, CB-153, CB 138 and CB-180 were found in almost all analysed samples included in this study (Table 2).

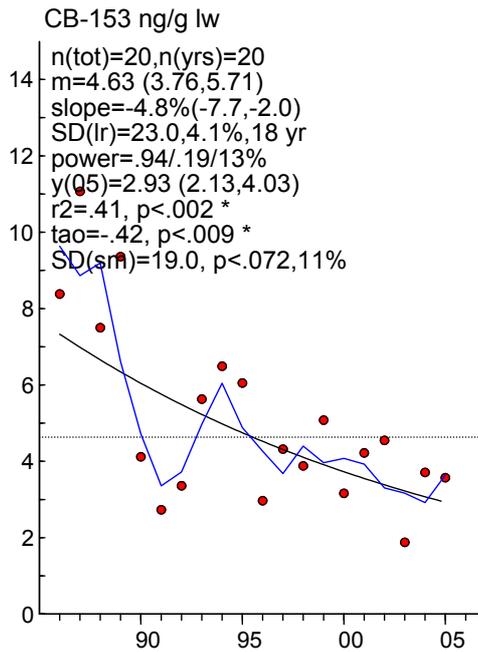
The concentrations of CB-118, CB-153, CB 138 and CB-180 in muscle of moose all decrease significantly during the period with 3.1%, 4.7%, 4.8% and 3.9%, respectively (Figure 8, 9, 10 and 11).

The concentrations of CB-118 in reindeer do not tend to decrease significantly during the period, probably partly due to the high concentrations found in samples collected late in 1998 (Figure 12). However, if the sample from 1998 is excluded from the trend analysis there is a significant decrease of CB-118 with an annual rate of 2.9% ( $p < 0.027$ ). The concentrations of CB-138 decrease with 4.2% annually ( $p < 0.007$ ) and CB-153 with 4.5% ( $p < 0.002$ ) and CB-180 with 3.4% annually ( $p < 0.009$ ). (Figure 13, 14 and 15).

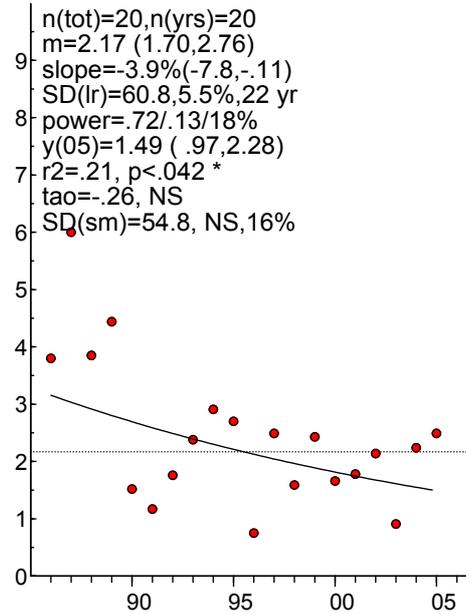


**Figure 8 (left).** Levels of CB-118 (ng/g, lipid w.) in muscle of moose from Grimsö.  
**Figure 9 (right).** Levels of CB-138 (ng/g, lipid w.) in muscle of moose from Grimsö  
 - - - - indicates the overall geometric mean level over the period 1986-2005.

Moose, Grimsö



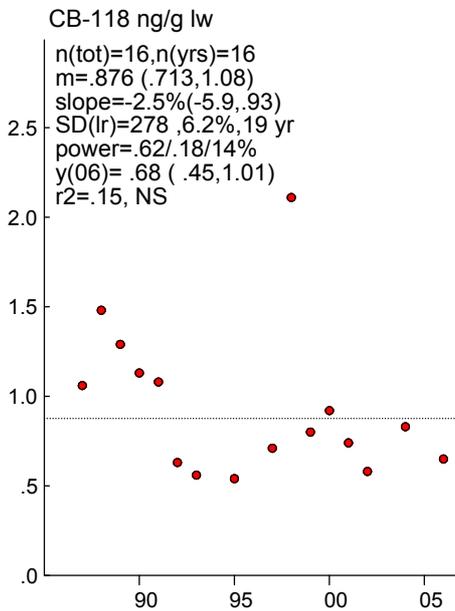
CB-180 ng/g lw



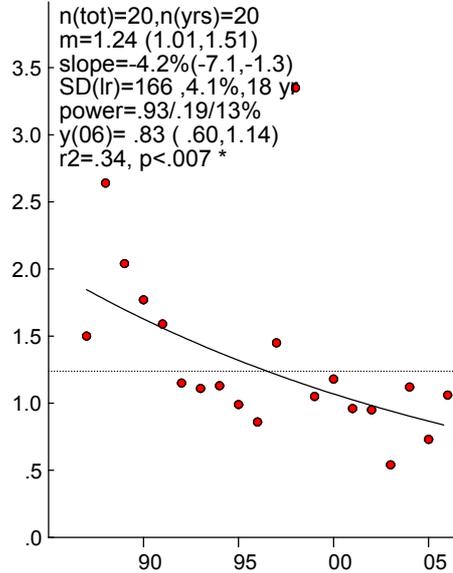
NRM, Dep. of Contaminant Research 08.08.21 13:33, CB2\_alg

**Figure 10 (left). Levels of CB-153 (ng/g, lipid w.) in muscle of moose from Grimsö**  
**Figure 11 (right). Levels of CB-180 (ng/g, lip. w.) in muscle of moose from Grimsö**  
 - - - - indicates the overall geometric mean level over the period 1986-2005.

Reindeer, Abisko



CB-138 ng/g lw



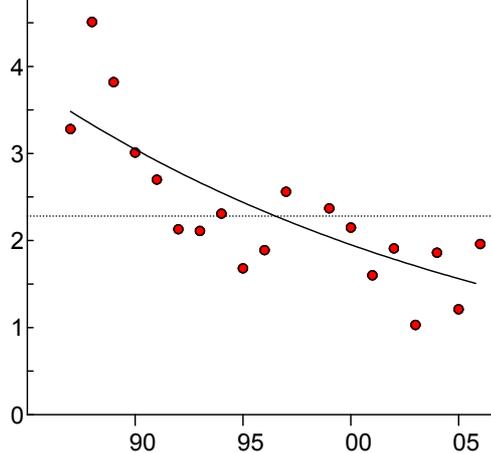
NRM, Dep. of Contaminant Research 08.08.21 13:29, CB2

**Figure 12 (left). Levels of CB-118 (ng/g, lip. w.) in muscle of reindeer from Abisko**  
**Figure 13 (right). Levels of CB-138 (ng/g, lip. w.) in muscle of moose from Grimsö**  
 - - - - indicates the overall geometric mean level over the period 1987-2006.

## Reindeer, Abisko

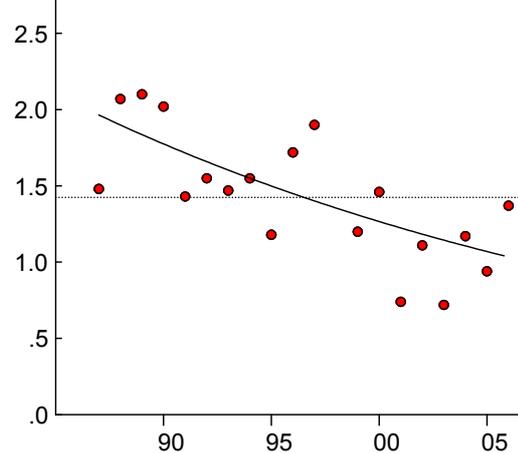
CB-153 ng/g lw

n(tot)=20,n(yrs)=20  
 m=2.28 (1.89,2.76)  
 slope=-4.5%(-7.0,-1.9)  
 SD(lr)=38.1,3.7%,17 yr  
 power=.98/.23/12%  
 y(06)=1.49 (1.12,1.99)  
 r<sup>2</sup>=.43, p<.002 \*



CB-180 ng/g lw

n(tot)=20,n(yrs)=20  
 m=1.43 (1.21,1.68)  
 slope=-3.4%(-5.8,-.96)  
 SD(lr)=83.9,3.5%,16 yr  
 power=.99/.25/11%  
 y(06)=1.03 (.79,1.35)  
 r<sup>2</sup>=.32, p<.009 \*



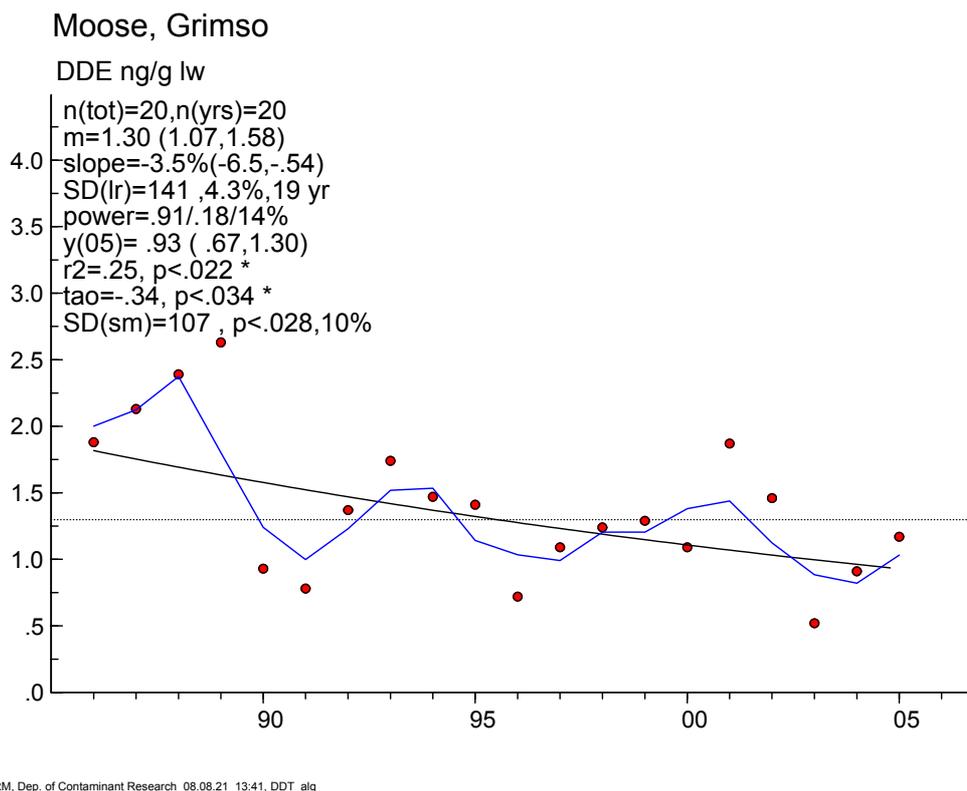
NRM, Dep. of Contaminant Research 08.08.21 13:37, CB1-1

**Figure 14 (left). Levels of CB-153 (ng/g, lip. w.) in muscle of reindeer from Abisko**  
**Figure 15 (right). Levels of CB-180 (ng/g, lip. w.) in muscle of moose from Grimsö**  
 - - - - indicates the overall geometric mean level over the period 1987-2006.

DDTs

DDT is probably the best known of the POPs. DDT was widely used from the 1940s up to the beginning of the 1970s to protect man from malaria, typhus, and other diseases spread by insects. It continues to be applied against mosquitoes in several countries to control malaria and most insects in agriculture, orchards, etc.

The concentrations of DDT and its metabolite DDD were all years under the LOD in both moose and reindeer. The concentrations of DDE were above the LOD in moose and reveal a significant decrease ( $p < 0.022$ ) for the period with 3.5% annually (Figure 16). Concentrations in reindeer were below quantification in all but the years 1989 and 1991 (Table 2).



**Figure 16. Levels of DDE (ng/g, lipid weight) in muscle of moose from Grimsö.**  
 - - - - - indicates the overall geometric mean level over the period 1986-2005.

### Chlordane

Chlordane is a chemical used as a pesticide in many countries mostly in the US up to the total ban there in 1988. Technical chlordane is a mixture of two isomers called alpha-chlordane and gamma-chlordane and many by-products from production. Chlordane has not been used in Sweden.

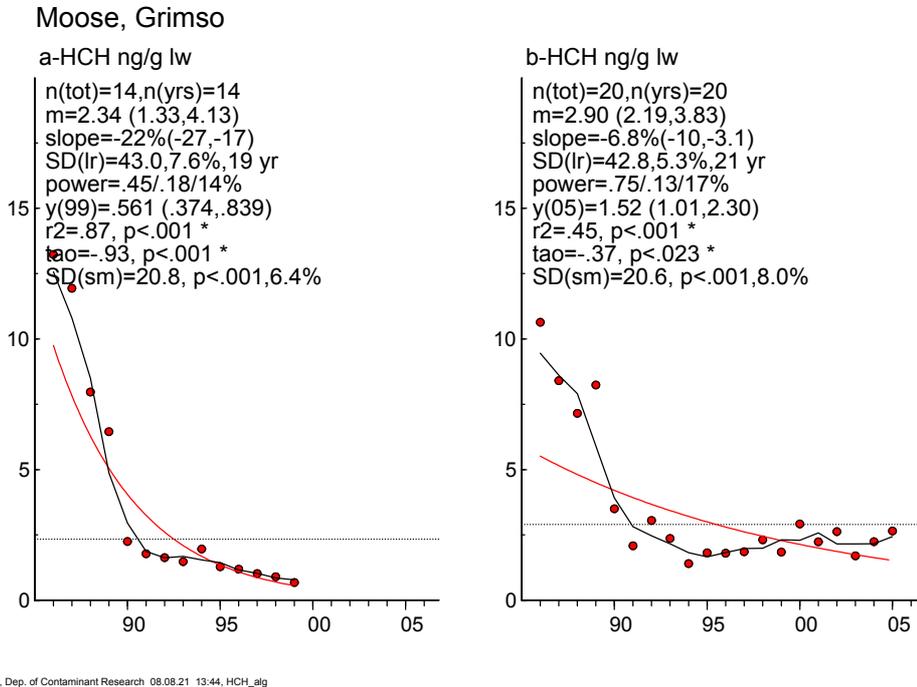
Concentrations of chlordane congeners and transnonachlor were all below the LOD in both moose and reindeer (Table 2).

### Hexachlorocyclohexanes, HCHs

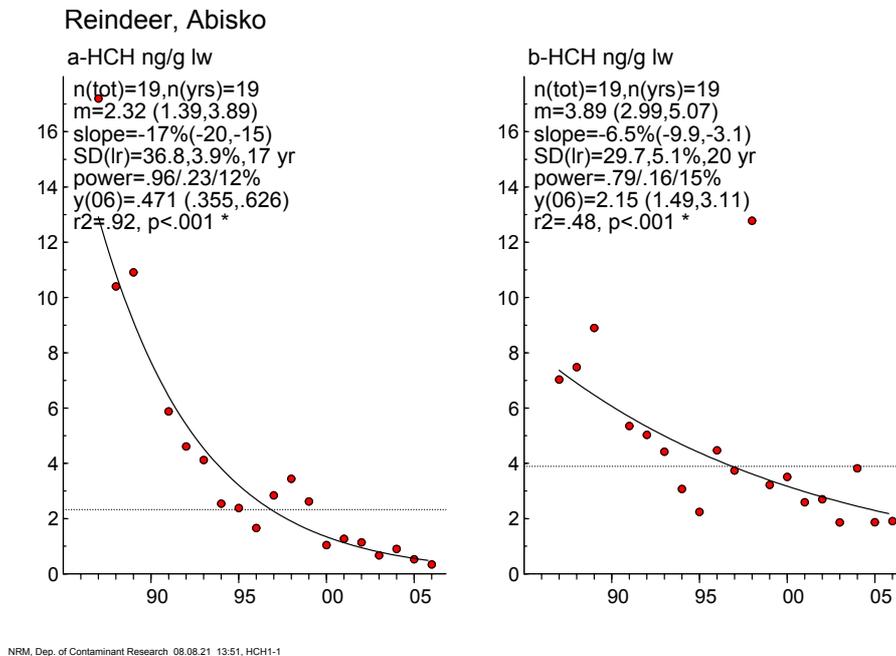
Technical grade HCH consists of 65-70%  $\alpha$ -HCH, 7-10%  $\beta$ -HCH, 14-15%  $\gamma$ -HCH and approximately 10% of other isomers and diverse compounds. Technical lindane contains more than 90% of  $\gamma$ -HCH, the substance with insecticidal effect. Lindane is one of the most widely used organochlorine insecticides for agricultural purpose.

The concentrations of  $\alpha$ -HCH were most of the years above the LOD (Table 2) and a statistical analysis is reported for both moose and reindeer. In moose the concentrations decreased with 22% annually during the investigated period ( $p < 0.001$ )

and in reindeer with 17% ( $p < 0.001$ ) (Figure 17 and 19). The concentrations of  $\beta$ -HCH were above the LOD (Table 2) all through the time series. In moose the concentrations decreased with 6.8% annually and in reindeer with 6.5% ( $p < 0.001$ ) (Figure 18 and 20).



**Figure 17 (left). Levels of  $\alpha$ -HCH (ng/g, lip. w.) in muscle of moose from Grimsö**  
**Figure 18 (right). Levels of  $\beta$ -HCH (ng/g, lip. w.) in muscle of moose from Grimsö**  
 - - - - indicates the overall geometric mean level over the period 1986-2005.



**Figure 19 (left). Levels of  $\alpha$ -HCH (ng/g, lip. w.) in muscle of reindeer from Abisko**  
**Figure 20 (right). Levels of  $\beta$ -HCH (ng/g, lip.w.) in muscle of reindeer from Abisko**  
 - - - - indicates the overall geometric mean level over the period 1986-2005.

### Polybrominated diphenyl ethers, PBDEs

PBDEs are organic compounds that are used as a flame retardants. PBDEs have been used in a variety of products as e.g. building materials, electronics, furniture, motor vehicles, plastics, polyurethane foams, and textiles. PBDEs are classified according to the average number of bromine atoms in the molecule.

For most PBDE congeners the concentrations in both moose and reindeer were below the LOD (Table 2) why no statistical analysis is performed.

### Hexabromocyclododecane, HBCD

HBCD is a brominated flame retardant. Its primary application is in extruded (XPS) and expanded (EPS) polystyrene foam that is used as thermal insulation in the building industry. HBCD is also used in furniture, automobile interior textiles, car cushions and insulation

The concentrations of HBCD in both moose and reindeer were below the LOD (Table 2) why no statistical analysis is performed.

### Tributylphosphate

Tributylphosphate is used in chemical manufacturing as additive chemicals. Tributylphosphate (TBD) and tri-iso-butylphosphate (TIBP) are colourless viscous liquids with a density slightly less than water. Mixtures of TBD and TIBP are used in aircraft hydraulic fluids wherein the amount of tributylphosphates ranges from about 35 to about 50 weight percent based on the total weight. Other applications are construction materials, paint, plaques, varnishes and different consumer preparations (SPIN).

Most annual concentrations of TIBP and TBP were below the LOD in both moose and reindeer (Table 2) why no statistical analysis is performed.

### Endosulfan

Endosulfan is an organochlorine insecticide used in most types of cultivations. It is an endocrine disruptor, and it is highly acutely toxic. It is banned in many countries among them the EU while it is still used in some countries.

Concentrations of  $\alpha$ -Endosulfan,  $\beta$ -Endosulfan and Endosulfansulphate were below the LOD in both moose and reindeer (Table 2) why no statistical analysis is performed.

### Heptachlor

Heptachlor is an organochlorine compound primarily used to control soil insects and termites. It has also been used more widely to control insects on cotton, grasshoppers, other crop pests, and malaria-carrying mosquitoes.

The concentrations of all compounds in this group (Heptachlor, Heptachlor-exo-epoxide, Heptachlor-endo-epoxide, Aldrin, Endrin and Dieldrin) were below the LOD (Table2) in both moose and reindeer why no statistical analysis is performed.

### Chlorophenols

Chlorophenols are a group of chemicals in which chlorines (between one and five molecules) have been added to phenol. Chlorophenols with at least two chlorines either have been used as pesticides or converted into pesticides. It has also been produced in minor quantities when wastewater or drinking water is disinfected with chlorine, if certain contaminants are present in the raw water. They are also produced during the bleaching of wood pulp with chlorine when paper is being produced.

The concentrations of Pentachlorophenol were below the LOD (Table2) in both moose and reindeer why no statistical analysis is performed.

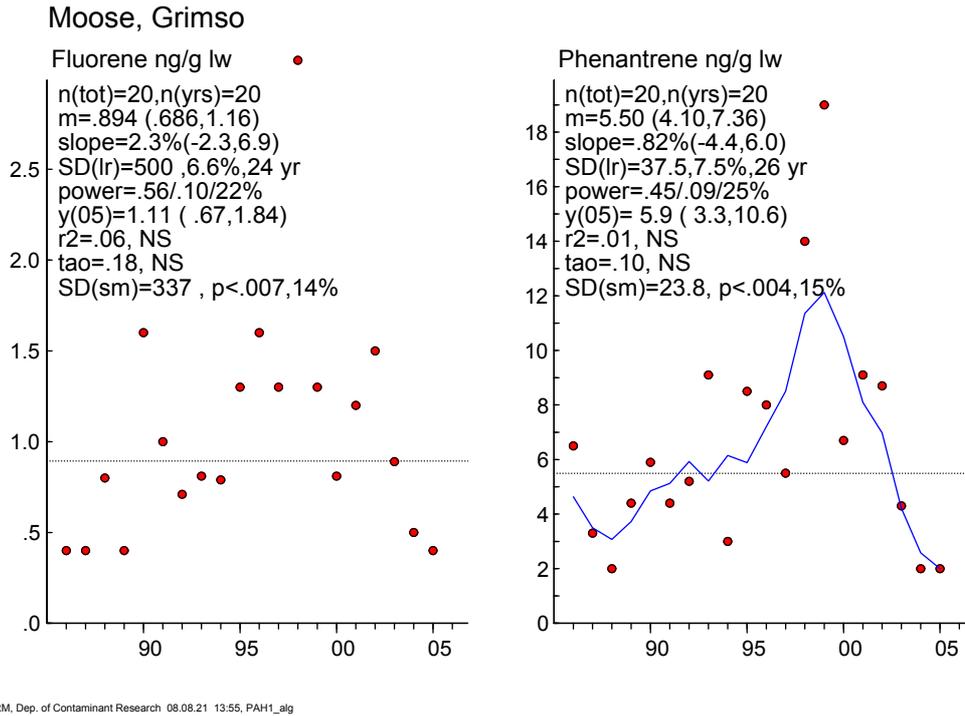
### Polycyclic Aromatic Hydrocarbons, PAH

PAH is a group of many hundred compounds with two or more benzene rings. They occur in aromatic oil products (HA-oils). Many of the PAHs are harmful for health and environment. Most of the compounds are persistent, bioaccumulation and carcinogenic.

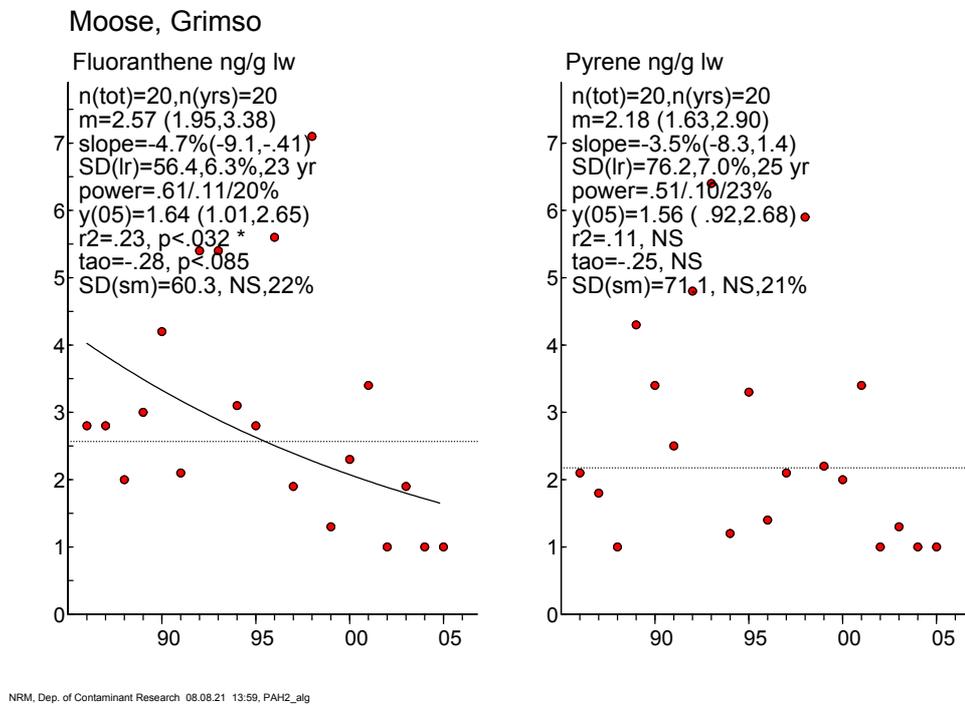
Concentrations of many of compounds in this group were below the LOD (Table 2).

Some of them, however, show levels which were possible to quantify and for which statistical analyses were performed. The compounds that were present in highest

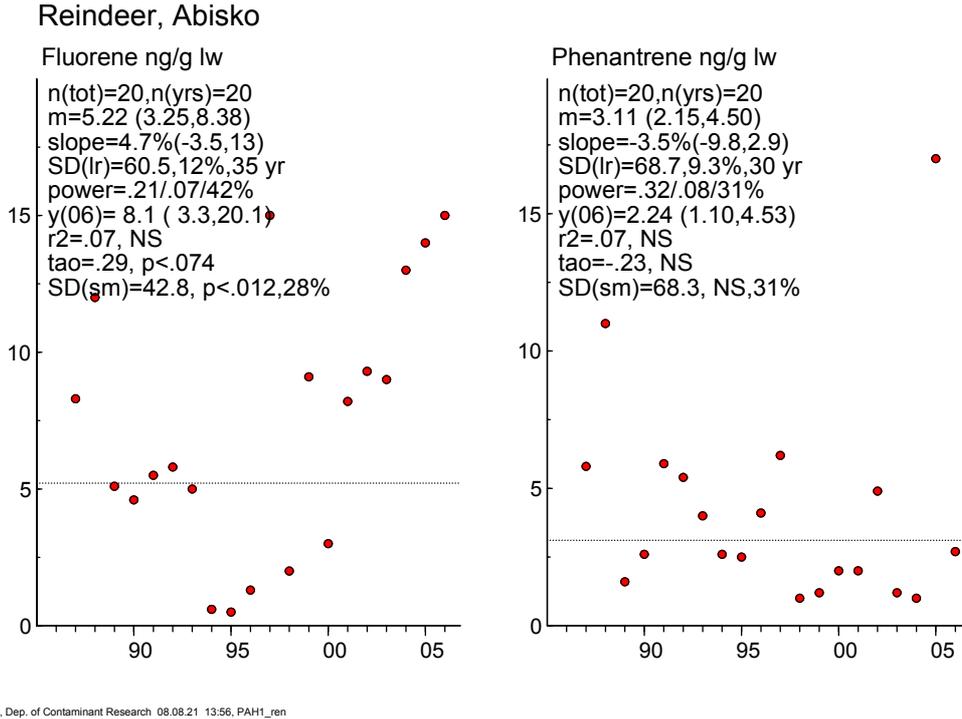
extension of the PAHs were Fluorene, Phenantrene, Fluoranthene and Pyrene. The concentrations of Fluorene, Phenantrene and Pyrene vary considerably over the period in both moose and reindeer and do not change significantly in any trend over the period in the two matrices (Figure 21, 22, 24, 25, 26, 28). However, in moose the concentrations of Fluoranthene have decreased (Figure 23) with an annual rate of 4.7% ( $p < 0.032$ ). The concentrations of Fluoranthene in reindeer are in many years below the LOD and no decreasing trend can be observed (Figure 27).



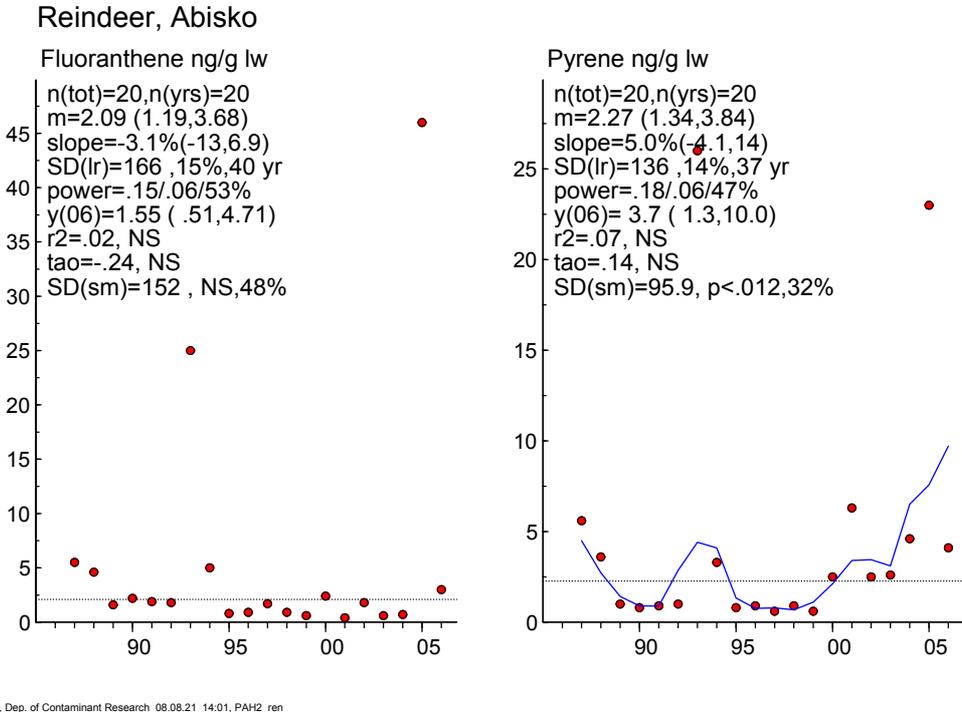
**Figure 21 (left). Levels of fluorene (ng/g, lip. w.) in muscle of moose from Grimsö**  
**Figure 22 (right). Levels of phenanthrene (ng/g, lipid w.) in muscle of moose from Grimsö**  
 - - - - indicates the overall geometric mean level over the period 1986-2005.



**Figure 23 (left). Levels of fluoranthene (ng/g, lipid weight) in muscle of moose from Grimsö.**  
**Figure 24 (right). Levels of pyrene (ng/g, lip. w.) in muscle of moose from Grimsö.**  
 - - - - indicates the overall geometric mean level over the period 1986-2005



**Figure 25 (left). Levels of fluorene (ng/g, l. w.) in muscle of reindeer from Abisko**  
**Figure 26 (right). Levels of phenantrene (ng/g, lipid w.) in muscle of reindeer from Abisko.**  
 - - - - indicates the mean overall geometric level over the period 1987-2006.



**Figure 27 (left). Levels of fluoranthene (ng/g, l. w.) in muscle of reindeer from Abisko.**  
**Figure 28 (right). Levels of pyrene (ng/g, l. w.) in muscle of reindeer from Abisko.**  
 - - - - indicates the overall geometric mean level over the period 1986-2005

### PCDD, PCDF and nonortho-PCB

PCDD are chemicals produced unintentionally due to incomplete combustion, as well as during the manufacture of certain pesticides and other chemicals. In addition, certain kinds of metal recycling and pulp and paper bleaching can release PCDD. PCDD have also been found in automobile exhaust, tobacco smoke and wood and coal smoke.

PCDF are also produced unintentionally from the same processes that release PCDD, and they are also found in commercial mixtures of PCBs.

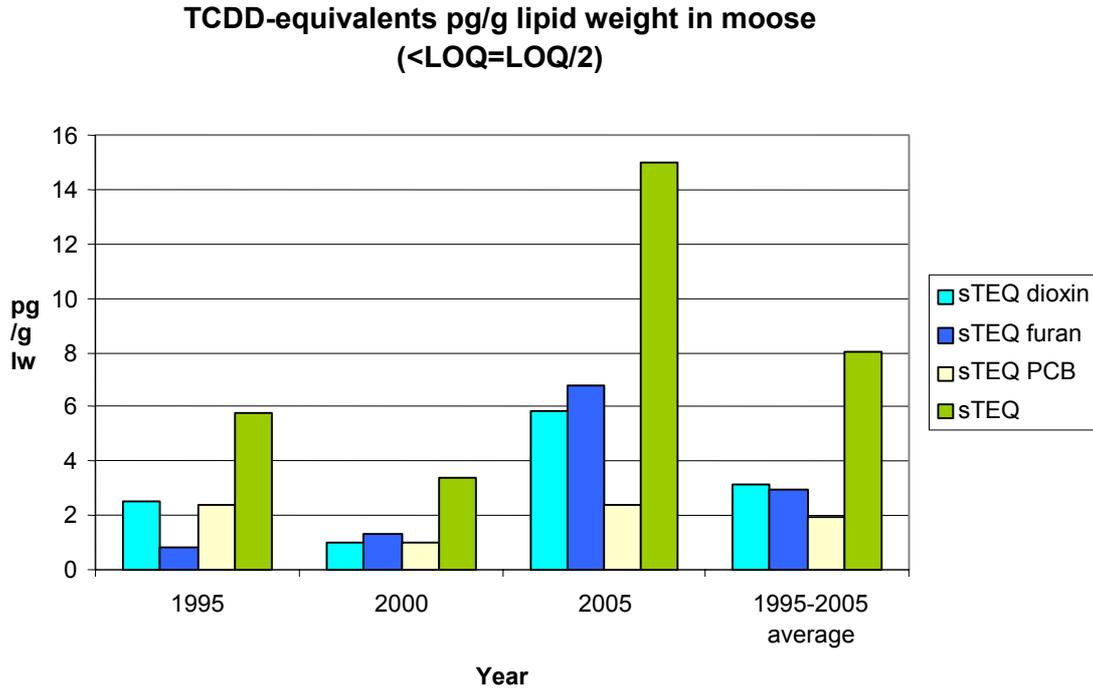
The concentrations of PCDD and PCDF are in general very low in the analysed samples of moose and reindeer, in many cases below the limit of detection. Concentrations of nonortho-PCBs are found in detectable concentrations in both moose and reindeer in all samples.

The figures (Figure 29 and 30) below show concentrations of dioxins, furans and nonortho-PCBs expressed as TCDD-equivalents. The concentrations of the summarised TCDD-equivalents (including PCDD, PCDF and nonortho-PCBs) in moose are between 3.4 and 15.0 pg/g lipid weight (which represents 0.04-0.14 pg/g wet weight) and with a mean concentration 8.1 pg/g l.w.

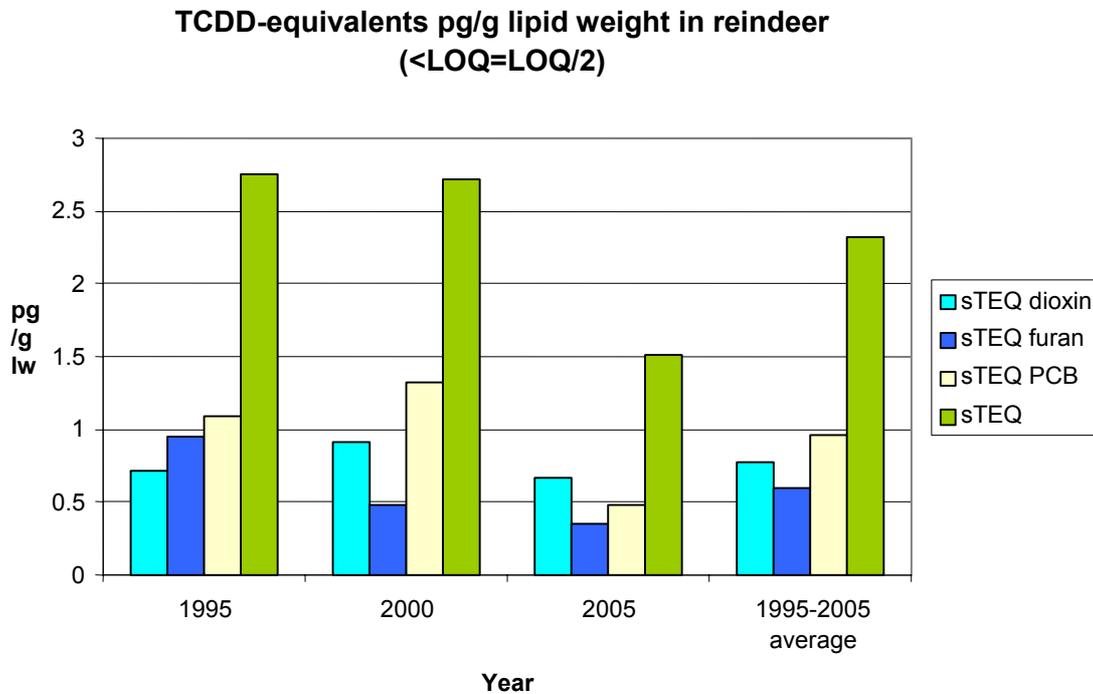
Concentrations of almost all PCDD congeners in reindeer are below LOD, it is only OCDD that is found in all three samples. The summarised concentrations of TCDD-equivalents in reindeer are between 1.5-2.8 pg/g l.w in the analysed years with a mean value of the three years on 2.3 pg/g l.w.

The limit for TCDD-equivalents in beef set by the European Union is 4.5 pg/g l.w.

PCDD, PCDF and nonortho-PCBs have only been analysed in one pooled sample of each moose and reindeer in three years 1995, 2000 and 2005. This material is thereby too small to draw any statistical conclusions of possible differences between years or changes over time.



**Figure 29. TCDD-equivalents (pg/g lipid weight) in muscle of moose from Grimsö in three different years. ( $<LOQ=LOQ/2$ ).**



**Figure 30. TCDD-equivalents (pg/g lipid weight) in muscle of reindeer from Abisko in three different years. ( $<LOQ=LOQ/2$ ).**

### Perfluorinated compounds PFCs

Perfluorinated compounds are a group of substances characterized by carbon chains with all hydrogen atoms replaced by fluorine atoms. PFCs have been produced since the beginning of the 1950s and have been used in a variety of different products. PFCs are used as emulsifiers, as protection in food packing-paper, leather and textiles, in cleaning products and in fire fighting foams.

Concentrations of PFCs in both moose and reindeer muscle were generally low. All samples had concentrations below LOQ for the analysed substances. However, concentrations of PFOSA, PFOS and PFOA could be detected some years but not in concentrations high enough to be quantified. Concentrations of PFOSA were found in moose three years and in reindeer two years. PFOS was found in seven years for moose and 15 for reindeer and for PFOA detectable concentrations was only found in moose one year.

One plausible cause to the low levels found may be that levels tend to be low in muscle in relation to levels in liver tissue. That remains to be proved.

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