Temporal Trends of Organochlorines in Northern Europe, 1967-1995 Support Long Ranged Transport but not the 'Grass-hopper Effect'

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ABSTRACT

The Swedish Contaminant Monitoring Programme comprises time series from more than 40 different locations and matrices. The biota samples are carefully selected to provide comparable samples between years and are collected in locally unpolluted areas. This report compares time trends of sDDT, HCHs and HCB in 6 series representing herbivorous mammals, predatory birds, pelagic and predatory fishes covering environments in southern Baltic areas as well as northern arctic part of Sweden. The studied period (1967-1995) covers a period of extensive use of organochlorines (OCs), followed by a period of international legislation and measures to protect the environment and a period of recover. The time trends show a remarkable conformity with respect to changes over time and the annual change in concentrations were similar irrespective of whether the time trends represent southern highly polluted areas or arctic remote, low contaminated areas. The findings do not indicate that the 'Grass-hopper' effect, so far, has played an important role.

INTRODUCTION

The Baltic was earlier severely polluted by OCs[1)]. Temporal monitoring of contaminants has been conducted in Sweden since 1967 and this report presents only some of the time trends that are available [2)]. The Baltic receives pollutants from rivers as well as from airborne fallout. To study the influence of the latter, the selected time series includes series on birds and fish from the highly polluted Baltic and fish from lakes in the southern as well as the remote arctic region of Sweden where also arctic herbivorous reindeers is studied.

MATERIAL AND METHODS

Following matrices have been collected and analysed:

More or less residential young **herring** (*Clupea harengus*) give representative and comparable samples between years and geographical areas in the southern Baltic. **Guillemot** (*Uria aalge*) feeds on pelagic fish and stays all the year in the Baltic proper. Eggs have been collected. Stationary **pike** (*Esox lucius*) similarly offers comparability between years in lakes of northern and southern Sweden. Resident **arctic char** (*Salvelinus alpinus*) has been collected in a small lake 200 km north of the

Polar Circle in an area lacking local industries and human settlements in the drainage area. In this area, Abisko; three year old male **reindeers** (*Rangifer tarandus*),were collected.

Dorso-lateral muscle of fish, leg muscle of reindeer and homogenised guillemot egg content were analysed. The annual herring sample consists of 20-25 specimens analysed individually. Ten individuals per year were analysed of guillemot eggs, pike and arctic char. The annual reindeer samples comprised at least 10 pooled specimens a year. To investigate the previous concentrations of HCHs and HCB old samples of pike (1968-1986) from lake Storvindeln, stored as extracts in sealed glass ampoules, have been analysed as pooled samples comprising 10 individuals per year. To avoid impact of seasonal variations on contaminant concentrations the samples have been collected at the same time of the year throughout the study period. Sampling locations are indicated in the figure.

The extraction procedure and the fat determination was essentially the same during the entire study, based on the method published by Jensen et al.[3]. All analytical results are expressed on a lipid weight basis. During the study period the gas chromatographic technique has improved. The most important change has been the transition from packed column to capillary column GC. To check the comparability of data obtained by packed column during the late 1960s and early 1970s with samples analysed in the middle of 1980s, samples from the Swedish Environmental Specimen Bank have been re-analysed. When the capillary column was introduced in 1988 and the GC routine was changed the results obtained by the two methods were compared by parallel analysis of a number of samples of the different matrices and locations presented in this study in order to calculate species and location specific transformation ratios. The sDDT ratios were found to be very close to 1. The concentrations of HCHs and HCB are all analysed by capillary column and long time trend studies of these components are all based on recent analysis of material stored either as extracts or as tissue samples in the Environmental Specimen Bank.

Simple log-linear regression analysis based on the annual geometric mean values has been carried out in order to detect time trends.

RESULTS AND DISCUSSIONS

All results discussed here are presented in the Table and Figure. The concentrations of **sDDT** decrease significantly and concordantly over time in all time with a good agreement in the annual change of about 11% a year series, except in the reindeer serie where the concentrations are exremely low. This significant decline still continues. During the entire study period concentrations of sDDT are higher in pike in the south than in remote northern areas. Since the 1980s the concentrations of α-HCH, ß-HCH and γ-HCH are determined. In the Baltic, concentrations of **α-HCH** decrease significantly over time by about 20% a year. In the northern lakes, the more than 25 years long time series of pike from Lake Storvindeln as well as in the shorter 14 year time trend in Lake Abiskojaure, a significant and fast decline over time for α-HCH can be seen. Concentrations of α-HCH in reindeer decline at a rate very similar to what is found in arctic char. Thus, it seems to be only small if any differences in declining rates between terrestrial and freshwater environments and no obvious differences between southern locations and remote northern areas. No convincing difference in concentrations of HCHs was found between north and south that could indicate increasing concentrations to the north. **HCB** concentrations decline significantly in almost all presented time series. In the lakes no difference in concentrations of HCB was found between south and north.

Technical HCH, contains large quantities of α-HCH. Since the 1970s HCH has been replaced by pure γ-HCH in most western countries[4]. However, McConnell et al.[5] found indications on a recent use of technical HCHs in the Asian part of Russia. Li et al.[4] report a continuous use of technical HCH throughout the 1980s in various parts of the world. Earlier a decline has been found in air samples from the Beaufort Sea area[6]. The fast decline was almost identical to what we have found in biota

samples from southern and northern Scandinavia. This indicates a drop in releases from eastern sources. A substantial drop in the usage of pesticides in former Soviet Union after a sudden decline of the Russian economy has been reported[7]. Most of the usages of pesticides was located in the southern part of the country[7]. This indicate the importance of long range airborne transport.

The earlier Swedish use of DDT was very limited. The similar declining rates of sDDT found in the more than 20 years long time series from the marine as well as the freshwater environments located in quite different parts of Sweden indicate a common airborne pollution.

Wania and Mackay[8, 9, 10] have suggested a model for global transport of volatile and semi volatile compounds from lower to higher latitudes. The model suggests that concentrations of some persistent organic pollutants will decrease over time more rapidly in temperated agricultural and urbanised areas than in cold remote areas. The transport to higher latitudes is suggested to occur not only as a single step at the first release to the environment but also as a series of continuos steps (grass-hopper effect). Volatile compounds like HCHs, HCB but also less volatile like DDT would reach the maximum concentration in remote subpolar and polar regions than in warmer areas. Furthermore, the slope of the decline would be steeper in lower latitudes than in higher. Thus the model predicts that the annual decline in concentrations would be faster in the southern Baltic or lake Bolmen than in the remote northern lakes Storvindeln and Abiskojaure or in reindeers from arctic parts of Sweden. This is obviously not the case, whether we study DDT, HCH or HCB. The similarities in decline in southern as well as northern Scandinavia cannot be explained by local pollution in the north or by the vicinity to European sources since the concentrations found in Swedish Arctic fauna are similar to those found in arctic areas of North America[11, 12, 13]

Climate differ substantially between the southern and northern Sweden. The mean air temperature in the studied part of the Swedish Arctic is -1.5°C and the corresponding temperature in southern Sweden is 7.8°C[14]. In the north, ice and snow cover the lakes and ground for 9 to 10 months, in the south often less than 2 months. The southern Baltic is only occasionally ice covered. Locations in this study range from 57°00'N to 68°15'N i.e. more than 1300 km.

For the fairly volatile substances HCH and HCB there seems to be a discrepancy in the long time series on pike from lake Storvindeln starting during the late 1960s. There is an extensive random variation in the concentrations of α-HCH during the 1970s and early 1980s followed by a rather smooth decline. HCB does not seem to decline in the same smooth way in recent years. The difference between the two compounds in variations over time might indicate different sources for the pesticides HCHs and HCB. Since the 1970s, HCB has rarely been used as a pesticide but large amounts have been released from the chemical industry during the 1980s[15]. If the grass-hopper mechanism is the important explanation for transport of volatile compounds to the north, it is difficult to understand the extensive between-year variation in α-HCH concentrations in remote arctic lakes during a period of world-wide usage. This variation speaks in favour for a one-step transport from agricultural areas to Arctic regions controlled by the prevailing air transport and outfall of the specific year. Furthermore, the extensive variation between years makes it questionable to use, occasionally collected, single year samples for description of time trends or spatial variations not only in locally polluted areas but also in remote Arctic areas.

That the global fractionation as well as the grass hopper effect are working seems from a theoretical point of view well supported. There are also recent reports indicating an ongoing evaporation of PCBs[16, 17, 18]. However, our time trend series show that the most important transport of pollutants happened in the past and the grass-hopper effect does not play a very important role since the changes in concentrations over time are more or less identical in remote arctic regions of Sweden as well as in the most severely polluted European environments in the south. To our understanding there are some questions that call for concern in future. It seems important to estimate how much of the total amount of compounds, stored in soil, sediments, vegetation and biota, that will be mobilised by

evaporation to air at a specific time. Secondly, it is important to get more knowledge on the relative importance of abiotic and biotic degradation processes in the environment. Finally, still these compounds are released into the environment at an unknown rate either as pesticides or from the technosphere. Thus we have not yet been able to study the environmental processes at a real post-contamination period. However, our results indicate that we would expect lower environmental concentrations in future both in the Arctic and the rest of the world.

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Time trends for sDDT, α-HCH and HCB in various matrices and locations. Following data are given: time interval studied, the coefficient of determination (r^2) for the regression analysis, the mean concentrations in (**ng/g**, lipid weight) including their 95% confidence interval at the end (M_{end}) of the time period studied as well as the annual change (%). * indicate significant changes over time, $p<0.05$.

SPECIES, TISSUE/ LOCATION, TIME	**sDDT** r^2	M_{end} +/- CI	ANNUAL CHANGE %	**α-HCH** r^2	M_{end} +/- CI	ANNUAL CHANGE %	**HCB** r^2	M_{end} +/- CI	ANNUAL CHANGE %
Guillemot/egg									
Stora Karlsö	0.92*	25 000	-11	0.85*	16	-23	0.84*	1200	-9.2
1969-1995		20 000-32 000	-13 - -10		10-25	-31 - -14		940-1600	-13 - -5.6
Herring/muscle									
Karlskrona	0.92*	1 100	-12	0.96*	28	-17	0.84*	67	-13
1972-1995		900-1 400	-13 - -9.9		24-34	-20 - -14		49-92	-19 - -6.6
Pike/muscle									
Bolmen	0.81*	400	-9.9	0.39	9.0	-9.4	0.06	12	-3.9
1967-1995		290-550	-12 - -8.1		5.4-15	-20 - -2.4		6.5-23	-17 - +12
Pike/muscle									
Storvindeln	0.91*	68	-12	0.68*	6.8	-9.6	0.69*	11	-5.8
1968-1995		53-88	-13 - -10		4.3-11	-12 - -7.0		8.4-14	-7.3 - -4.3
Arctic Char/muscle									
Abiskojaure	0.66*	20	-11	0.91*	4.0	-18	0.64*	17	-6.0
1981-1995		13-30	-16 - -6.7		3.0-5.4	-21 - -14		14 - 22	-9.2 - -2.7
Reindeer/muscle									
Abisko	0.00	1.0	-1.2	0.93*	3.6	-21%	0.77*	10	-8.7
1983-1994		0.41-2.6	-15 - +15		2.6-4.7	-24 - -18		8-13	-12 - -5.4

The figure shows concentrations of sDDT, αHCH and HCB in guillemot egg from St Karlsö, central Baltic; herring from Karlskrona, southern Baltic; pike from Lake Bolmen and Lake Storvindeln in southern and northern Sweden respectively; arctic char from Abiskojaure in the northernmost Sweden and reindeer from Abisko in the vicinity of Abiskojaure. All concentrations given in µg/g lipid weight. Note that the time scale (x-axis) differ. Geographical position of sampling locations and polar circle are indicated.