

Fast Initial Decrease in Environmental Concentrations of OCs – A Result of Atmospheric Degradation? Part I.

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Introduction

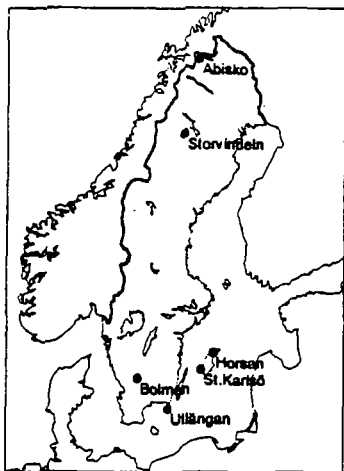
Recent reports show that concentrations of various OCs have decreased at a similar rate either the time trends were based on temporal trend studies from remote sub Arctic regions of Sweden or southern tempered regions of the Baltic (1, 2) and similar trends were also found irrespective we studied the trends in terrestrial, freshwater or marine environments. The theoretically well founded models for Global transport of volatile and semi volatile compounds presented by Wania and Mackay (3, 4, 5), predict that concentrations will decrease more slowly in remote cold areas than in tempered, southern areas after the discharges have ceased and faster decrease is expected in terrestrial environments than in marine. Recent field studies have also clearly shown that temperature is controlling the air concentrations of semi volatile compounds such as PCB (6, 7, 8). The similarities in temporal trends found in the Swedish material are thus different from the predicted differences. In our earlier reports we stressed the importance of more knowledge on the relative amount that actually will mobilise by evaporation at a specific time as well as the relative importance of non biotic as well as biotic environmental degradation processes for an understanding of environmental fate of various OCs (1, 2).

In this study, time trend series are used to investigate the rate of decrease the first 10 years after measures or other circumstances have reduced the output of various compounds to the environment. There is a risk that compounds like PCB will continue to leak out from the technosphere even after that production has been banned. Similarly, HCB, originally used as a pesticide, have been introduced to the environment also as a contaminant from the chlorine industry (9). Thus, both PCB and HCB seem unsuitable for studies concerning the speed at which concentrations decrease after dramatic reductions in use. Instead we have used the pesticide DDT and α -HCH, a major contaminant of the pesticide γ -HCH (Lindane). Pesticides are used at the fields and once there, they are mobilised, degraded or simply sedimentated to the soil. We could

not use Lindane since this product has been used in recent time in some western European countries. The major measures against DDT happened in the beginning of 1970s and consequently we have studied the trends for DDT starting in 1971. Our earlier studies have shown that α -HCH decrease dramatically during late 1980s and 1990s (1, 2) after the collapse of the Soviet Union and when the production of chemicals for agricultural use dropped in Russia (10). Thus our time trends for HCH start in 1987. The aromatic DDT and its degradation products DDD and DDE are less volatile than the aliphatic α -HCH. As a consequence of the differences in chemical properties between DDT and HCH, degradation mechanisms as well as transport rate may in many aspects differ between the two compounds.

Material and Methods

We have used samples from the Swedish Environmental Contaminant Monitoring Programme, where samples are annually collected since the end of the 1960s. Ten to twenty specimens of selected species have been analysed individually and the geometric annual mean value have been calculated. From the geometric mean values, log-linear regression analysis has been applied for the ten years following measures taken to stop the use or after production of other reasons have ceased. We have compared the calculated annual change in concentrations (percent). The sampling locations and the species have been selected to represent locally unpolluted environments in various parts of Sweden. All matrices used are biota samples and methods used for sampling, preparation and chemical analysis have been presented elsewhere (2). Following time series have been used: *guillemot eggs* from Stora Karlsö and *young herring* from Utlängan from the central southern parts of the seriously polluted Baltic, *pike* from Lake Bolmen and *roach* from Lake Horsan from Swedish fresh waters in the south, *pike* from Lake Storvindeln and



arctic char from Lake Abiskojaure from the northern remote sub Arctic part of Sweden, *reindeer* from Abisko, close to Lake Abiskojaure covering the period 1983-1995 earlier presented in Olsson et al. (1). Matrices and location of sampling sites, see Figure 1.

Figure 1. Sampling sites. Abisko - reindeer, Abiskojaure (close to Abisko) - arctic char, Storvindeln - pike, Horsan - roach, St. Karlsö - common guillemot, Bolmen - pike, Utlängan - herring.

Results and Discussions

The changes in concentrations of Σ DDT in 1971-1980 are very similar irrespective of the matrices and location. (Table 1). Furthermore, the change over time during the first ten years following the international measures taken to stop the use of DDT, is remarkably rapid, as much as around 20% per year. Also for α -HCH all time trends with a highly statistically significant annual decrease show a decrease of about 20%. The similarities in changes between the less volatile aromatic compound DDT and the more volatile aliphatic compound HCH in different matrices collected in different environments (marine, freshwater or terrestrial), both in tempered and in sub Arctic

regions is interesting.

In an earlier report we found indications that the major transport route to remote sub Arctic regions, explaining the present environmental concentrations, has not been via several continuous small steps, the 'grass-hopper' theory. From our time trend data it is more plausible that the transport has occurred as initial steps in the past (1, 2). Our data also support that the pesticide contamination of the Swedish environment largely is determined by long range transport. The found initial and fast decrease in concentrations both in tempered and sub Arctic areas found in the present report is unexpected. If volatility and dilution from hot spots to remote areas is explaining a decrease in concentration one would expect a difference in rate between the two substances since DDT and HCH have quite different volatility.

The common, fast, initial decrease might indicate a common environmental degradation process. If biological processes would explain the fast initial decrease, then the generally higher biological activity in tempered areas than in sub Arctic areas would result in a faster decrease in the south.

Table 1. Annual changes in concentrations of Σ DDT and α -HCH in various matrices from Sweden. The 95% Confidence Interval (C.I.) is indicated. N.a. indicates that the time interval is not covered by chemical analysis. All regression analysis are highly statistically significant ($p < 0.001$)*** except for the pike series of α -HCH ($p < 0.054$).

Area	SPECIES/Location	Σ DDT, %, C.I. 1971 - 1980	α -HCH, %, C.I. 1987 - 1996
Southern	Guillemot egg/ Stora Karlsö	-19 *** -25 - -12	-24 *** -33 - -15)
	Herring/ Utlången	-18 *** -26 - -11	-21*** -26 - -16
Southern	Roach/ Lake Horsan	N.a.	-21 *** -26 - -15
Southern	Pike/ Lake Bolmen	-20 *** -27 - -12	-10 -21 - 0.3
	Northern	Pike/ Lake Storvindeln	-19 *** -23 - -16
Northern	Arctic char/ Abiskojaure	N.a.	-18 *** -21 - -14
	Northern	Reindeer/ Abisko	N.a.

This cannot be found. The similarities in observed temporal trends may indicate a major degradation process in a medium where the contaminants appear in a fairly equal distribution. This could be in the atmosphere. Several reports in literature show that both UV radiation and chemical reactions with various radicals may explain the degradation of various OCs (11, 12, 13, 14). The present results indicate that we still need more appropriate information on atmospheric

degradation processes and their relative importance. One important question will be the identification of degradation products.

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