

SEASONAL VARIATIONS IN THE UPTAKE OF AIRBORNE GASEOUS AND PARTICLE BOUND ORGANOCHLORINE COMPOUNDS IN SCOTS PINE NEEDLES

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Introduction

During recent years the uptake mechanisms of airborne persistent organic pollutants (POPs) into plants have received much attention. A hydrophobic epicuticular wax that will sorb hydrophobic compounds, such as the POPs, from the surrounding air covers the green parts of higher plants. Thus, plants play an important role in the deposition of hydrophobic compounds to terrestrial ecosystems, affecting both how much is deposited and acting as a buffer for the global fractionation of POPs¹⁻³. This is especially important in the evergreen boreal coniferous forests in the northern Temperate Zone due to the high degree of forestation and high leaf area index of conifers (leaf area index is a dimensionless parameter representing the area of leaf surface over unit area of ground). However, the mechanisms behind the sequestering of airborne POPs by plants are still not well understood. Modeling attempts suggest that boreal forests will significantly influence the fate only of POPs that have physicochemical properties only within a narrow range and that the influence of the forests on relatively volatile POPs, such as the hexachlorocyclohexanes (HCHs) is small³.

To understand the role plants play for the deposition of POPs to terrestrial ecosystems and in the global distribution of POPs, it is necessary to understand the sorption/revolatilization mechanisms for POPs in the plant/air system. It is also necessary to understand these mechanisms to evaluate plants as monitors of the geographical distribution of POPs. Although conclusions from models based on laboratory experiments have been applied directly to field studies^{3,4}, validation of the models using long time-series of field observations has not been performed.

To obtain basic information on how plants, especially conifers, sorb POPs from the surrounding air we have performed a thirty-month study where pine needles have been sampled continuously throughout the year simultaneously with air sampling. We here present data on the accumulation of selected compounds in the epicuticular wax of Scots pine needles during two and a half years, the whole life span of a generation of needles.

Methods and Materials

Scots pine (*Pinus sylvestris*) needles were collected off the Stockholm university campus on the northern outskirts of Stockholm. The methods used are described in full by Kylin et al.^{5,6}.

Results and Discussion

Air concentrations could only be obtained for the HCHs. The levels and seasonal variations are similar to what was found by Haugen et al.⁷ during more or less the same period of time. The air concentrations of α -HCH are more or less constant during the whole year (Figure 1), while the concentrations of γ -HCH show clearly elevated levels during spring and summer during the spraying season in Southern Europe (Figure 2).

Although the concentration profiles in the air of α - and γ -HCH show clear differences, the concentrations in the needles of these compounds show remarkable similarities. Both compounds accumulate in the needles during spring and summer, while very little concentration change takes place during autumn and winter. All the most volatile of the analytes in this study, the HCHs, HCB, CB-28 and CB-54, show the same type of seasonal changes. These compounds are more or less always entirely in the gas phase with only a small proportion bound to particles⁸.

It is significant that the highest accumulation rate coincides with the highest temperatures. It has been shown that this also coincides with the time when the needles contain the highest levels of hydrophobic compounds⁹. The drops in needle concentrations in late winter/early spring are due to the accumulation of starch in the needles, which affects the dry weight determination¹⁰. It is also interesting that the only case in which we can see any revolatilization from the needles in this long term study is for γ -HCH after the elevated air concentrations in spring have gone down.

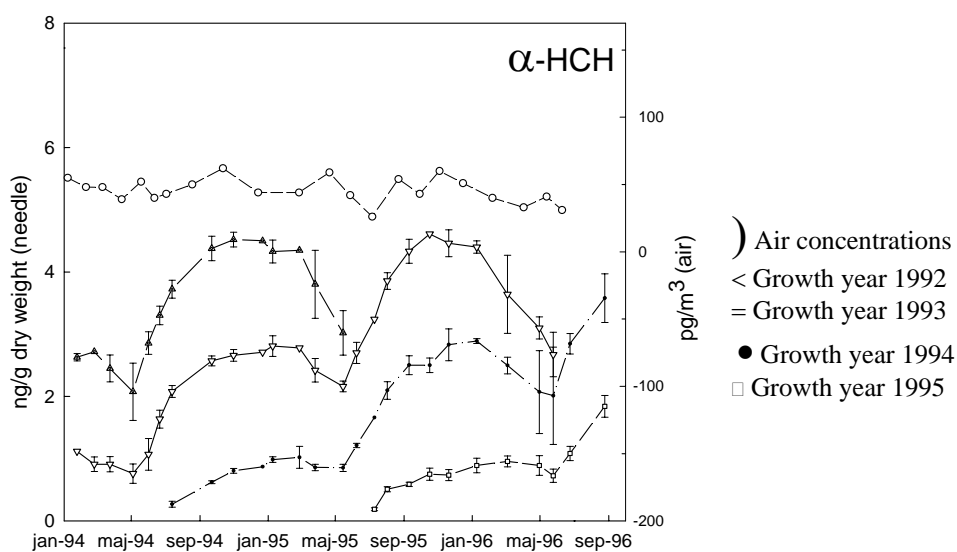


Figure 1: Concentrations of α -HCH in air and Scots pine needle wax. Mean values ($n = 2$ to 5) and standard deviations.

The behavior of more particle bound compounds such as CB-170 and CB-180 show a totally different seasonal behavior (Figure 3). The concentrations in the needles of these compounds show a prominent peak during the winter months with very little total accumulation.

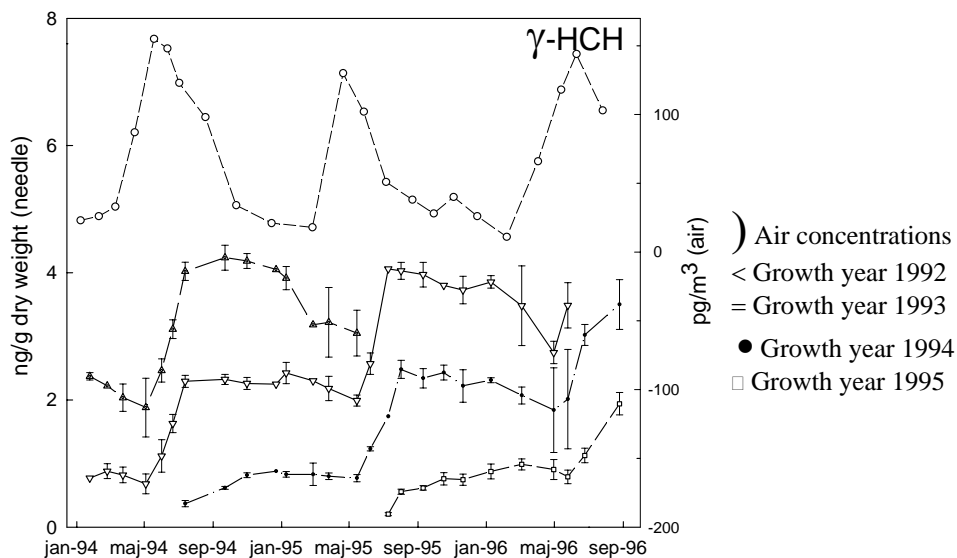


Figure 2: Concentrations of γ -HCH in air and Scots pine needle wax. Mean values ($n = 2$ to 5) and standard deviations.

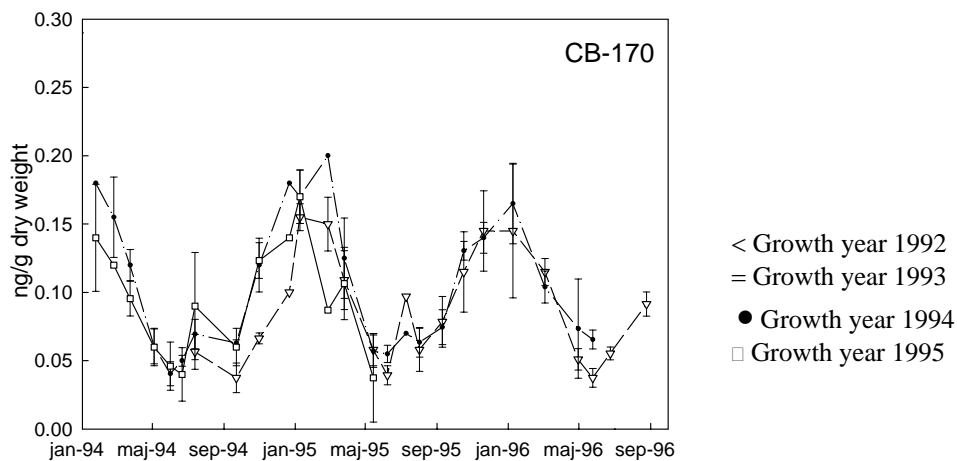


Figure 3: Seasonal concentration pattern of CB-170 in Scots pine needle wax. Mean values ($n = 2$ to 3) and standard deviations.

The seasonal pattern for compounds that have particle binding characteristics intermediate between the two previously mentioned groups, DDT, DDE, CB-138 and CB-157, show an intermediate seasonal concentration pattern. They show a general accumulation throughout the life

span of the needles, presumably sorption from the gas phase, overlaid by a prominent winter peak associated with particle deposition (Figure 4).

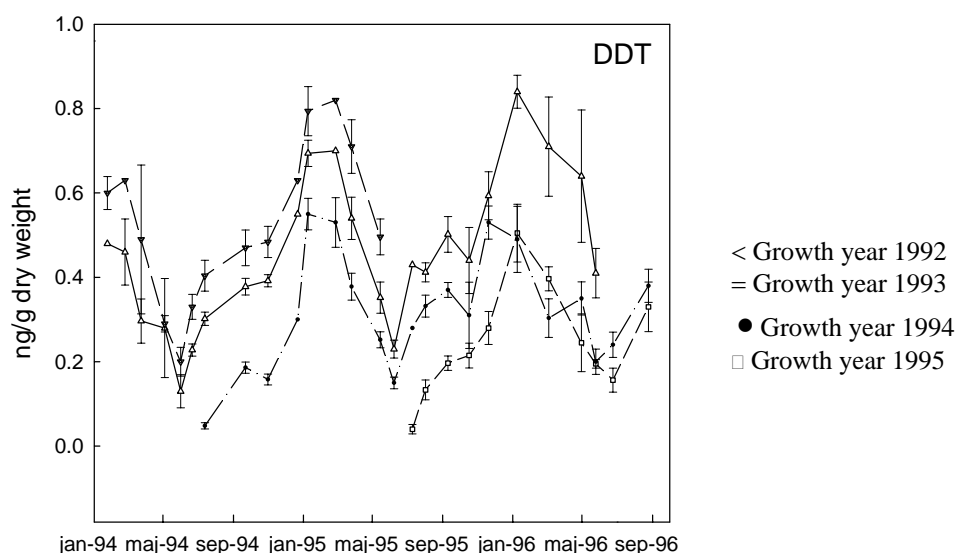


Figure 4: Seasonal concentration pattern of p,p'-DDT in Scots pine needles. Mean values (n = 2 to 5) and standard deviation.

This long-term study clearly shows that the uptake of organic pollutants in pine needles is not governed by the physicochemical properties of the compounds only. It is significant that it is the most volatile compounds that show the most prominent accumulation in the needles and that this accumulation continues throughout the life span of the needles. There must be biological factors that influence the sorption of compounds in the gas phase so that these behave in an opposite way to what is expected from physicochemical properties.

References

1. Wania F; Mackay D (1996) *Environ Sci Technol*, 30, 390A-396A
2. McLachlan M; Horstmann M (1998) *Environ Sci Technol*, 32, 413-420
3. Wania F; MacLachlan M (2001) *Environ Sci Technol*, 35, 582-590
4. Calamari D, Bacci E, Focardi S, Gaggi C, Morosini M, Vighi M (1991) *Environ Sci Technol*, 25, 1489-1495
5. Kylin H, Nordstrand E, Sjödin A, Jensen S (1996) *Fresenius J Anal Chem*, 356, 62-69
6. Kylin H, Sjödin A (2003) *Environ Sci Technol*, *in press*
7. Haugen J-E, Wania F, Ritter N, Schlabach M (1998) *Environ Sci Technol*, 32, 217-224
8. Bidleman T; Wideqvist U; Jansson B; Söderlund R (1987) *Atmos Environ*, 21, 641-654
9. Kylin H, Söder K, Undeman A, Franich R (2002) *Bull Environ Contam Toxicol* 68, 155-160
10. Flower -Ellis J G K, Olsson L (1993) *Studia Forestalia Suecica*, No 190, pp 1-19