

GLOBAL MULTICOMPARTMENTAL CYCLING OF *g*-HCH (LINDANE) - A MODEL STUDY

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Introduction

The long-range transport of long-lived (persistent) substances which are distributed over several environmental media (multicompartmental substances) is thought to be determined by winds and on long time scales also by oceanic currents as well as by the multicompartmental cycling. Due to the complexity of this type of cycling large-scale modelling studies are pursued since only recently.^{1,2} Its understanding is needed for chemicals regulation and risk assessment under regional and global convention processes.

Here we report on the global cycling of insecticide *g*-hexachlorocyclohexane (*g*-HCH, lindane), which is expectedly considerably partitioning to the hydrosphere and undergoes long-range transport, e.g. into the Arctic.^{3,4}

Materials and Methods

For the study of the long-lived semivolatile organic compounds (SOCs), we use a 3D, fully dynamical multicompartment model which is based on an atmospheric general circulation model, ECHAM5, including a preliminary version of the dynamic aerosol model HAM.⁵ The oceans, soils and vegetation are represented as mixed surface layers.² Degradation takes place in all compartments following first-order kinetics. 3D distributions of the hydroxyl radical are considered in the atmosphere. It is assumed that no decay takes place for molecules adsorbed to aerosol particles (adsorption described according to Junge) or dissolved in cloudwater. A doubling of the rate constant per 10 K increase is assumed for the ground compartments to account for the temperature dependency of biodegradation. Via seasonal movement of the mixing depth, lindane deposited to the ocean can undergo a second final sink. Suspended particulate matter in ocean is neglected.

We report on the results of simulations (months-years) in the (multicompartment) chemistry-climate mode, i.e. under conditions of present-day climate. Spatial and temporal model resolutions were 2.75°x2.75°, 19 levels (1000-10 hPa) and 30 min, respectively. The spatial and temporal patterns of substances' entry into the model world mimic the global usage in agriculture in 1980 and are based on FAO data (on a country basis, distributed with resolution 1°x1° within the countries scaled with the crop density as a surrogate; Fig. 1).⁶ 80% of lindane were applied to the vegetation, 20% to soil, throughout the year, not only during non-precipitation day-light hours. In successive years same amounts were entried.

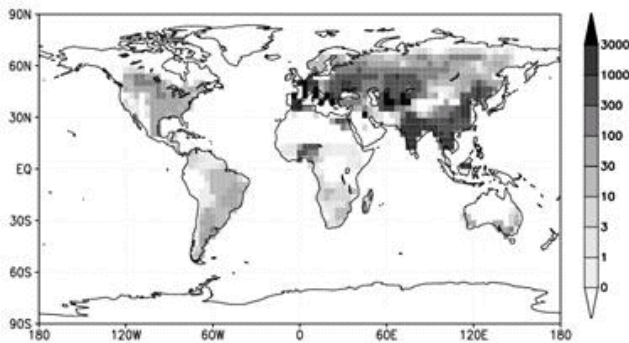


Fig. 1: Global application distribution of *g*-HCH ($\text{t a}^{-1} \text{gridcell}^{-1}$).

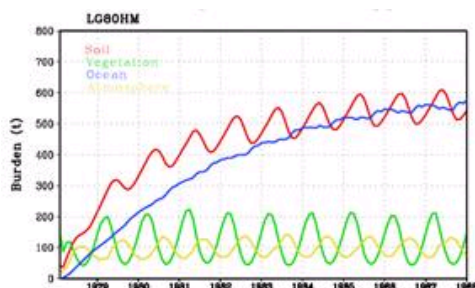
Results and Discussion

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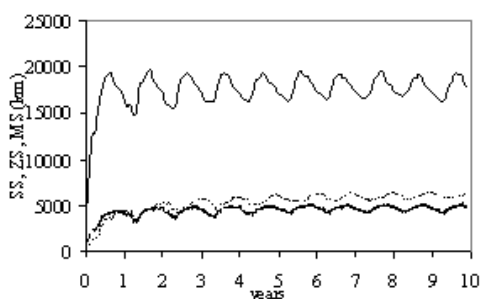
With different characteristic times the compartments approach equilibrium with regard to continuous emissions of lindane (Fig. 1a). While the ocean continues to take up strongly, the global soils have reached equilibrium after 6 years, the vegetation surfaces and the atmosphere within 2 years. After 9 years the oceans have absorbed almost half (42%) of the global inventory of the substance. A large fraction remains stored in the soils (42%), each ca. 8% in vegetation and in the atmosphere. During the first 9 years more than 6 times the burden of these compartments had been 'buried' in the deep sea. The total environmental lifetime, P_{overall} , levels off at around 9 months. The atmospheric burden is concentrated in the boundary layer, i.e. 50% between 900 and 700 hPa. A second maximum, ca. 10%, is found around 200 hPa.

By the seasonally varying atmospheric transports the substance is spreading geographically, in particular also towards the high latitudes (Fig. 1b). This tendency is illustrated by the long-range transport indicators meridional (MS), zonal (ZS) and spatial spreading (SS), which measure the extension of the 5 and 95%ile fringes of the geographic distribution of the substance in east-west (MS), north-south (ZS) or provide a means for the growth of the area covered by the 'cloud' (SS) since entry of the substance into the model world.⁷ While the spreading from the application areas in east and west direction as well as by area is in quasi-steady state from the second year onwards, the ZS is growing from ca. 5000 km in the 2nd year over 6470 km (6th year) to 6670 km in the 10th year. This reflects the tendency of accumulation of lindane in high latitudes in the model world.

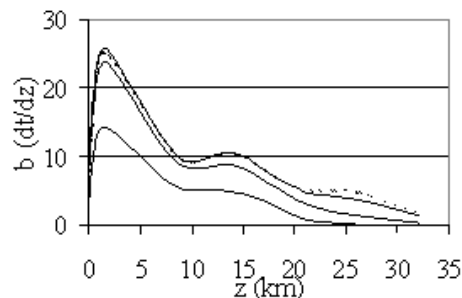
Such an accumulation has been observed in the field and the 'global distillation' and 'global fractionation' hypotheses had been put forward for explanation, basically implying that it is the substance properties which on the long term determine global distribution rather than geospheric spatial and temporal patterns.^{8,9}



a.



b.



c.

Fig. 2: Filling of the model world by lindane (a.) temporally (global total environmental burdens) and (b.) geographically (from top to down: meridional, zonal and spatial spreadings of the total environmental burden, see text), (c.) vertically in the atmosphere (from top to down: global atmospheric burden in the 7th, 4th, 2nd and 1st year upon entry).

By a pair of model experiments with lindane being allowed to re-volatilise in one, but not in the other model world, we separate the two modes of transport, i.e. upon initial emission from vegetation or soil (singlehopping transport mode) vs. upon second or later volatilisation from vegetation, soil or ocean (multihopping transport mode or grasshopping).¹⁰ The resulting geographic distributions of *g*-HCH in the atmosphere are shown in Fig. 3. The grasshopper effect dominates transport within the atmospheric boundary layer and in the vicinity of the application areas. Larger fractions of the re-volatilized substance reside aloft, namely 12% around 200 hPa a 3rd maximum in the vertical distribution around 30 hPa. For the 10th year of the substance's cycling in the environment, the model predicts higher spreading tendencies for the singlehopping transport mode than for the multihopping mode, e.g. 5270 vs. 4870 km for ZS. Less *g*-HCH is predicted to be deposited in the Arctic upon re-volatilisation than upon initial emission, i.e. 2040 vs. 2720 t, but vice versa with regard to deposition in the Antarctic, namely 85 vs. 11 t. Re-volatilization (the grasshopper effect) contributes to accumulation in the polar regions. However, the model experiment predicts an accumulation of lindane in the Arctic and Antarctic even without the grasshopper effect, i.e. solely due to atmospheric transports upon initial emission.

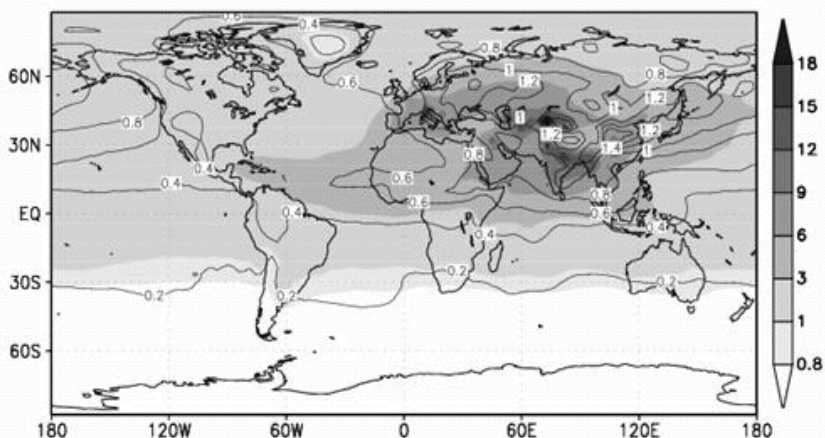


Fig. 3. Atmospheric burden ($\mu\text{g m}^{-2}$) of *g*-HCH transported by multi-hopping or single-hopping (shades) and multi-hopping only (isolines). Mean of the 10th simulated year.

Considerable uncertainties remain in the understanding of the large-scale environmental cycling of lindane. E.g. long-

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range transport is sensitive to the degradation rate adopted for the particle-bound state (0 in this study). Moreover, long-range transport in the ocean was neglected in the model study and an artificial, i.e. non-historic emission scenario was assumed.

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