NATIONAL INSTITUTE OF PUBLIC HEALTH AND ENVIRONMENTAL PROTECTION
BILTHOVEN
THE NETHERLANDS

Report no. 758473011

INTEGRATED CRITERIA DOCUMENT HEXACHLOROCYCLOHEXANES

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October 1988

This investigation has been carried out on behalf and for account of the Directorate-General for Environmental Protection, Direction Toxic Substances and Risk-Management.
This document is the English edition of Basisdocument Hexachloorcyclohexanen, Rep. no. 758473004, July 1987

Mailing list integrated criteria document Hexachlorocyclohexanen

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Title	Integrated criteria document hexachlorocyclohexanes
Keywords	Criteria document, hexachlorocyclohexanes, HCH, air quality, water quality, soil quality, standards, monitoring methods, measurement strategies, sources, emissions, production, dispersion, transformation, environmental concentrations, human toxicity, ecotoxicity, control techniques, cost of control, business consequences, risk analysis
Summary	This report contains a systematic survey and a critical evaluation of the most important data on the priority substance HCH on behalf of the environmental effect oriented policy
Date of publication	October 1988
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CONTENT

SUMMARY

This document contains data on the hexachlorocyclohexanes (HCHs) concerning sources and distribution patterns (soil, water, air, biota), the risks that present exposure levels present for man, (parts of) ecosystems and materials, and the technical possibilities and economic consequences with regard to the reduction of these risks. This information serves as a scientific basis for the formulation of the effect-oriented standard setting policy.

HCH, a man-made organochlorine compound, has 8 stereo-isomers, of which the γ-HCH is the bestknown. This compound is used as an insecticide. Attention is drawn to HCHs for two reasons. Firstly because of the possible risks related to their occurrence in the environment as a result of production and dumping in the past; secondly because of the potential risks the current use entails.

As to the 8 HCH-isomers, only for α-, β- and γ-HCH is there sufficient information on the occurrence and the effects to assess a risk. The risks presented by the other isomers are not estimated greater than those related to α-, β- and γ-HCH because the exposure levels are lower.

With regard to the risks of oral exposure for man, for α-HCH an γ-HCH tolerable daily intake (ADI), respectively, of 1 $\mu g.kg^{-1}$ body weight is proposed. Because of the accumulative properties of β-HCH chronic exposure to this isomer is considered undesirable; therefore no tolerable daily intake for chronic exposure is given. The present oral exposure of the population in The Netherlands is below the proposed limit values, whereas β-HCH is mostly (in more than 90% of the cases) absent in the daily diet and there is a wide margin between the tentative no-effect level for β-HCH (20 $\mu g.kg^{-1}$ body weight per day) and the maximally observed uptake through food (2 μg per day). It is concluded that the present oral exposure level does not present a risk for the population.

In respect to inhalatory exposure, only data on the effects of lindane (γ-HCH) are available. On the basis of limited data a no-effect level of 100 $\mu g.m^{-3}$ has been derived. This level is more than a factor of 100,000 higher than the large-scale annual average concentration in The Netherlands.

Taking into account the often lower concentrations at which the other isomers are present in the atmosphere, the risk of the inhalatory exposure level on the national scale are considered negligible. In contrast to this, indoor air concentrations may be increased for weeks or months after the use of lindane as a timber preservative applied on large surfaces to the extent that the margin between these levels (some tens of $\mu g.m^{-3}$) and the no-effect level is estimated too small to exclude a certain risk. The risk related to the application of lindane as an insecticide for parasite control in houses is not known.

In view of the persistent character of HCH and its function it is obvious that this group of compounds presents a certain risk for both aquatic and terrestric ecosystems.
The ecotoxicological acceptable level for α-, β- and γ-HCH in the fresh-water environment is considered 5, 1 and 0.1 $\mu g.l^{-1}$ respectively; for α- and γ-HCH in the marine environment this level is 1 and 0.002 $\mu g.l^{-1}$, respectively. In the fresh water of The Netherlands β- and γ-HCH reach concentration levels that are about the same (β-HCH) or above (γ-HCH) these acceptable levels only locally (harbours, ditches in agricultural areas etc.), whereas in the coastal waters the γ-HCH content exceeds the ecotoxicologically acceptable level. The toxicological limit values are above the present environmental standard for fresh water (<0.05 $\mu g.l^{-1}$); the latter is frequently exceeded in polder ditches and in systems of reservoirs for superfluous polder water and, incidentally, in the large rivers. The reference values for soil (10 $\mu g.kg^{-1}$) are frequently exceeded in the sediments and in sewage sludge.
As a result of insufficient data in respect to the terrestric ecosystems, a provisional limit value has been proposed only for lindane: 10 $\mu g.kg^{-1}$ soil, which corresponds to the present reference value. This value is exceeded locally to a great extent. Data on the contents in agricultural areas where lindane is applied are lacking. Estimates, however, indicate a base content of 80-250 $\mu g.kg^{-1}$. Resulting from the persistent character of lindane, the limit value even in natural reserves in The Netherlands is approximated and sometimes exceeded.

At several locations in The Netherlands the soil and the groundwater are heavily polluted with HCH as a result of activities, mostly related to the production of HCH in the past (in The Netherlands the production is stopped

in 1954). There are several possible methods for cleaning up (cost appr. Dfl. 200-400 per m^{-3}); the choice to actually proceed to action as well as the method to be applied depend on the use and the nature of the area, the possibilities of dumping the soil elsewhere or of modifying the destination, as well as the acceptibility of the risks and the political situation. It is considered desirable to start the cleaning if there are (unacceptable) risks of further distribution from the polluted locations. In that case it is recommended to take measures aimed at promoting the capacity for self-purification of the soil.

With regard to the present use of HCH it is striking that there are strong indications that it has increased by appr. 50% in the past 10 years, whereas the policy has been aimed at the reduction of the application of lindane-containing products. At present the emissions to soil and coastal water result in a load that is beyond the toxicologically acceptable level. These emissions may only be reduced by limiting or prohibiting its use. It is recommended to stimulate the use of alternative products or, in the case of side-effects occurring, to force this by limiting or terminating the permits. These aspects should be considered with in the framework of the permission policy under the Act on Pesticides.

Regarding the gaps in knowledge, further investigation is recommended into (a) the concentration in indoor air during domestic application of lindane, (b) the chronic toxicity of lindane to marine organisms and (c) the present concentrations in agricultural areas where lindane is applied.

INTRODUCTION

The environmental policy in The Netherlands is in the first place aimed at achieving and maintaining an environmental quality, which guarantees the health and well-being of people and the preservation of animals, plants, goods and patterns of utilization in a general sense (IMP Milieubeheer 1986 - 1990). Adequate knowledge lacking it will, however, not be possible for some time to fully define the general environmental quality aimed at. Therefore, attention is concentrated first on factors which may present great risks, such as substances hazardous to the environment. A selection has been made of the many substances that are important because of emission or usage and a priority list has been drawn up. For most priority substances socalled Integrated Criteria Documents are written.

Arranged by substances or groups of substances, Integrated Criteria Documents contain data on sources and distribution patterns (soil, water, air, biota), the risks of present concentrations for man, (parts of) ecosystems and materials, and the technical and economic possibilities of reducing these risks. This information serves as a scientific basis for the formulation of the effect-oriented environmental policy. Environmental quality requirements and a general term of reference for the emission reductions per type of source may result from this.

In this document attention is given to the various stereo-isomers of hexachlorocyclohexane (HCH), of which γ-HCH (lindane) is the best known. At present this organochlorine compound is especially used to control soil insects. On the one hand HCH is a historical problem: at several locations in The Netherlands the soil has been greatly polluted with HCH by the production in the fifties. On the other hand the present application may present a certain risk, especially because of the persistency of this group of substances in environment of the Netherlands.

The document has been drawn up by the National Institute of Public Health and Environmental Protection (RIVM), the Dutch Organization for Applied Scientific Research (TNO), the Institute for Environmental Studies (IvM) and the Economic and Social Institute (ESI), under the auspices of the RIVM.

For various parts, such as control techniques and their costs, valuable information has been obtained from several chemical concerns. This appreciated cooperation has been achieved through the Department Environment and Environmental Planning of the Council of the Dutch Employers' Union VNO and NCW. The content of this document has been checked by a Review Committee of the RIVM. Support in the preparation of this document was rendered by a Counselling group consisting of staff members of the Ministry of Public Housing, Physical Planning and the Environment, The Department of Inland Waterways (RIZA) and the Ministry of Agriculture and Fisheries.

1. PROPERTIES AND EXISTING STANDARDS

1.1. PROPERTIES

1.1.1. Structural formula and molecular structure

Structural formula: $C_6H_6Cl_6$ Molecular weight: 290.85

1,2,3,4,5,6-Hexachlorocyclohexane may occur in mixtures up to 8 different isomers. The HCH-isomers are distinguished by the position of the chlorine atoms on the cyclohexane ring. This may be indicated by "α" and "β" or by "a" (axial) and "e" (equatorial), as shown in figure 1.1.

Figure 1.1. HCH: α- and β-bonds (left) and axial (a) and equatorial (e) bonds (right)

The α-bonds are turned upwards, the β-bonds downwards. The axial bonds are at right angles to the ring level, the equatorial bonds are situated at the same level of the ring. The equatorial position being the most favourable for a relatively large substituent like chlorine, the molecule will take the formation at which as much chlorine atoms as possible are situated equatorially. The position of the chlorine atoms and the preferred conformations are summarized in table 1.1.
Of all HCH-isomers only α-HCH can be split into (+) and (-) antipodes.

The registration numbers are given in table 1.2.

Table 1.1. Position of the chlorine atoms with the preferred conformation for each of the various HCH-isomers (Kirk-Othmer, 1979)

Isomer	*Position of the Cl-atoms*	*Preferred conformation **
α-HCH	*1α,2α,3β,4α,5β,6β*	*a,a,e,e,e,e*
β-HCH	*1α,2β,3α,4β,5α,6β*	*e,e,e,e,e,e*
γ-HCH	*1α,2α,3β,4α,5α,6β*	*a,a,a,e,e,e/e,e,e,a,a,a*
δ-HCH	*1α,2α,3α,4β,5α,6β*	*a,e,e,e,e,e*
ε-HCH	*1α,2α,3α,4β,5β,6β*	*a,e,e,a,e,e*
ζ-HCH	*1α,2α,3α,4α,5α,6α*	*a,e,a,e,a,e/e,a,e,a,e,a*
η-HCH	*1α,2α,3α,4α,5β,6β*	*a,a,e,a,e,e/e,e,a,e,a,a*
θ-HCH	*1α,2α,3α,4α,5α,6β*	*a,e,a,e,e,e*

** the two conformations for γ-, ζ- and η-HCH are equivalent as in both cases 3 Cl-atoms are in the equatorial position*

1.1.2. Registration numbers

Table 1.2. Registration numbers (NIOSH, 1983)

Isomers	*Chemical Abstracts Service (CAS) reg. no.*	*Registry of Toxic Effects of Chemical Substances (RTECS) reg. no.*
HCH mixture	-	*GV 4950000*
α-HCH	*319-84-6*	*GV 3500000*
β-HCH	*319-85-7*	*GV 4375000*
γ-HCH	*58-89-9*	*GV 4900000*
δ-HCH	*319-86-8*	*GV 4550000*
ε-HCH	*6108-10-7*	
θ-HCH	*6108-11-8*	
η-HCH	*6108-12-9*	
ζ-HCH	*6108-13-0*	
HCH-in general	*608-73-1*	*GV 3150000*

1.1.3. Nomenclature and common synonyms

Hexachloorcyclohexaan (Dutch), hexachlorocyclohexane (IUPAC, English), Hexachlorcyclohexan (German), hexachlorocyclohexane (French); abbreviation HCH (less common: HCCH). It is advisable not to use the older name benzenehexachloride (abbreviation: BHC); in the Anglo-Saxon literature, however, the abbreviation BHC is still being used. In this document the term HCH without further description always to refers 1,2,3,4,5,6-hexachlorocyclohexane; γ-HCH is also known as lindane (IUPAC, English, French: lindane; German: Lindan) or, less commonly used, gammaxane (German: Gammexan).

Lindane is sold as an insecticide in many formulations under a large number of trade names: in The Netherlands alone appr. 50 lindane-containing trade preparations have been admitted (Gewasbeschermingsgids, 1985); in the Chemical Abstracts more than 80 trade names are mentioned. According to the EC-directive 84/491/EEC lindane should contain at least 99% γ-HCH . The other HCH-isomers have no generally accepted trivial names. Technical HCH contains appr. 15% lindane, furthermore appr. 65% α-HCH, 10% β-HCH and 10% other isomers and pollutants. Enriched HCH contains appr. 45% lindane, 25% α-HCH and 4% β-HCH, fillers, other isomers and pollutants.

1.1.4. Physical properties

Physical properties are reasonably wellknown for α-HCH trough δ-HCH only. The summary presented in table 1.2. is composed of data obtained from DFG (1983), Kirk-Othmer (1979), Patty (1981), OHM/TADS (1985), TDS (1985), Balson (1947), Spencer and Cliath (1979), Slade (1945), Kurihara et al. (1972), Canton et al. (1982) and Veith et al. (1979). It is noted that some values from the literature vary considerably.

Table 1.3. Summary of a number of physical properties of HCH-isomers

Parameter	*HCH-isomer*			
	α	*β*	*γ*	*δ*
Melting point (^{o}C)	*159.2*	*311.7(subl.)*	*112.9*	*140.8*
Boiling point (^{o}C/mmHg)	*288/760*	*60/0.58*	*323/760*	*60/0.34*
Vapour pressure (mmHg/20^{o}C)	2.54×10^{-5}	2.85×10^{-7}	9.4×10^{-6}	1.7×10^{-5}
Solubility in water (mg.l^{-1} 20^{o})	*1.5*	*0.2*	*7.3*	*9*
IR-spectrum (Sadtler)	*9699*	*9698*	*11843*	*9696*
max (nm)	*125.8*	*134.6*	*132.2*	*118.1*
K_{ow}	*3.81*	*3.72*	*3.85*	-
Henry constant:				
- atm.m^{-3}.mol^{-1}	6.4×10^{-6}	5.0×10^{-7}	4.9×10^{-7}	7.3×10^{-7}
- no dimension	2.7×10^{-4}	2.0×10^{-5}	2.0×10^{-5}	3.0×10^{-5}

1.1.5. Chemical properties (Kirk-Othmer, 1979)

The raw reaction product has a characteristic pungent musty odour, pure γ-HCH only has a weak odour.

At room temperature all HCH-isomers are stable; β-HCH is the most stable because of its thermodynamically favourable e,e,e,e,e,e-conformation. HCHs

hardly react with acids, if at all; in an alkaline environment dehydrochlorination occurs when heated, first to pentachlorocyclohexane, finally to trichlorobenzenes. The relative amounts of trichlorobenzene-isomers formed depend on the initial product and this may be used as a confirmation of a reaction at the residue level (PAM, 1985; OVR, 1985).
Under the influence of UV, γ-HCH may partly be converted to small amounts of α-HCH, but usually it is rapidly degraded (DFG, 1983). Formation of β-HCH from γ-HCH has not been observed.
During the usual production processes 5 isomers are formed; in the environment only α-, β- and γ-HCH occur.

1.2. EXISTING STANDARDS AND GUIDELINES

With regard to definitions of the concepts used in the framework of the effect-oriented standard setting policy, reference is made to the appendix to this document. In The Netherlands and abroad standards and guidelines have been determined with respect to soil, water and air.
In The Netherlands requirements are also made for lindane with respect to chemical waste, food, cosmetics, animal fodder and usage.

1.2.1. Soil

In the "Leidraad Bodemsanering" (VROM, 1983) a framework is given for the evaluation of the concentration levels in soil. The indicative guidelines mentioned should not be considered as standards, but as a method for assessment. The following distinction is made: A-reference value, B- critical value for further investigations and C- critical value for clean-up investigations. The concentrations are considered in connection with the function of the soil and the local pollution conditions (table 1.4.).

In accordance with the Discussion Paper on Soil Quality (1986) for β- and γ-HCH a level of 10 $\mu g.kg^{-1}$ dry substance is considered as a tentative reference value for a multifunctional soil, based on a standard soil with an organic matter content of 10%. For all halogenated hydrocarbons combined a value of 20 $\mu g.kg^{-1}$ dry substance is being applied.

Table 1.4. Testing scheme for the evaluation of the concentration levels of lindane in soil in the framework of soil sanitation

Occurrence in	*Soil ($mg.kg^{-1}$ d.s.)*			*Groundwater ($\mu g.l^{-1}$)*		
Component/level	*A*	*B*	*C*	*A*	*B*	*C*
Pesticides:						
organochloro (individually)	*0.1*	*0.5*	*5*	*0.05*	*0.2*	*1*
organochloro (total)	*0.1*	*1*	*10*	*0.1*	*0.5*	*2*
in general (total)	*0.1*	*2*	*20*	*0.1*	*1*	*5*

According to the RIKILT (1985) caution should already be taken with grazing at levels as low as 0.05 mg β-HCH per kg of soil. The limit at which it is still acceptable to cultivate plants of which the vegetative parts are to be consumed by man, is 0.5 mg β-HCH per kg of soil (sandy soils).
The policy is directed at the restriction or banning of the application of compounds considered persistent, whenever substitutes which are not persistent and no more hazardous in other respects are available. The Ministry of Agriculture and Fisheries (LAC, 1986) applies signal values for substances hazardous to the environment (such as HCH) present in polluted soils (table 1.5.).

Table 1.5. Reference and signal values for β-HCH in sandy soil (4% humus) at optimal pH and a manuring ($mg.kg^{-1}$ dry substance) according to LAC (1986)

Reference value	*0.01 (detection limit)*
Signal value	
- grassland	*> 0.01*
- arable farming land with fodder crops	*0.02*
- other arable land, incl. horticultural land (food)	*0.2*
- horticultural land (flowers)	*-*

1.2.2. Water

In tables 1.6. and 1.7. the quality requirements for surface water and tolerable levels for waste water, respectively, are given.

Table 1.6. Quality requirements for surface water in $\mu g.l^{-1}$

Country	*Type/use*	*Max.conc.*	*Remarks*	*Ref.*
The Netherlands	*-drinking water*	*<0.05*	*α-HCH*	*1*
		<0.05	*lindane*	*1*
	-water for shell-fish	*harmless*	*hal.hydrocarbons*	*1*
	-basic quality	*≤0.01*	*α-HCH, lindane*	*2*
EEC	*-drinking water,*			
	simple purification	*1*	*pesticides*	*3*
	normal purification	*2.5*	*total*	
	thorough purification	*5*		
	-inland waters	*0.1*	*HCH at discharge*	*4*
	-estuaries and territorial marine waters	*0.02*	*HCH*	
USA	*-fresh water*	*0.08*	*lindane, 24 hr average*	*5*
		2.0	*lindane, max. concentr.*	
	-salt water	*0.16*	*lindane, max. concentr.*	
Countries on river Rhine	*-drinking water*	*0.1*	*organochl.pest., natural purific.*	*6*
		0.5	*chem.physical purification*	

1: Staatsblad (1983)
2: IMP-Water (1985-1989)
3: EEC (1975)
4: EEC (1984)
5: US Federal Register (1980)
6: IAWR (1986)

Table 1.7. Limit values and the effective dates from which these levels are to be observed for HCH containing waste water in the EEC (1984), both for emissions per ton of product and concentrations in waste water

Branch of industry*	Measuring unit	Limit values to be observed as from : ***** 1-4-1986	1-10-1988
- Company for HCH production	g HCH per ton HCH produced **	3	2
	mg HCH per liter discharged waste water***	3	2
- Company for lindane extraction	g HCH per ton HCH treated **	15	4
	mg HCH per liter discharged waste water ***	8	2
- Company for both HCH production and lindane extraction	g HCH per ton HCH produced **	16	5
	mg HCH per liter discharged waste water ***	6	2

* These limit values also concern waste water possibly resulting from further processing of lindane at the same location. If necessary, the EEC will determine appropiate measures and limit values for HCH processing branches not mentioned in this table, especially with reference to lindane processing industrial companies producing products for the protection of plants, timber and cables. Up to that time, the member states determine the emission standards themselves, taking into consideration the best available technical means.

** The values are expressed in terms of weight (monthly average)

*** The values are expressed in terms of concentrations (monthly average of the HCH-concentration, weighted on the basis of the discharge of the waste water)

****Limit values applicable to the total quantity of HCH in all HCH-containing discharges of water at the site of the industrial plant

1.2.3. Air

In tables 1.8. and 1.9. air quality requirements are presented for work places and outdoor air respectively.

Table 1.8. MAC-values for lindane (8 hr time weighted average, TWA, at the work place) in mg.m^{-3}

Country	Concentration	Reference
The Netherlands	0.5	Nat. MAC-list (1986)
USA	0.5	NIOSH/OSHA (1981)
FRG	0.5	TNO (1977)
DRR	0.2	
USSR	0.05	

Table 1.9. Air quality standards for outdoor air in mg.m^{-3}

Country	Concentration	Remarks	Reference
USSR	0.03	daily average	Izmerov (1973)
	0.03	"maximal" value; measuring time max. 20 minutes	

1.2.4. Food and drinking water

In tables 1.10. and 1.11. the permissible residue content in food and the quality requirements for drinking water, respectively, are presented.

Table 1.10. Maximal permissible HCH-content in food (Staatscourant, 1986)

Product	Concentration in μg.kg^{-1}		
	α	β	γ
Milk	4	2	8
Meat *	300	100	2,000
Cacao *	150	150	1,000
Cereals	100	20	500
(remaining) Vegetables and fruit	50	50	1,000
Eggs *	500	200	1,000
Tea (α and β)	100		100
Coffee	100	100	1,000
Poultry meat *	300	100	700
Tropical seeds	100	100	
Rabbit *	300	100	2,000
Stone fruit, grapes and tomatoes			500
Whole meal			50
Leaf vegetables			2,000
Winter carrots, carrots			100
Potatoes			10
Remainder **	0(10)	0(10)	0(10)

* calculated in terms of fat content

** no residues should be present; at a level of 10 μg.kg^{-1} this requirement is considered to be met

A committee of the Health Council proposed an ADI-value for lindane of 5 $\mu g.kg^{-1}$ body weight. In the USA a maximum acceptable level is proposed for the intake of lindane through polluted water of 18.6 $ng.l^{-1}$ (with a risk of cancer of 1 to 10^6 individuals) , for aquatic organisms 62.5 $ng.l^{-1}$ (EPA, 1981) and for drinking water 26.4 $ng.l^{-1}$ (US-EPA, 1984). IMPR (1985) mentions an ADI for lindane of 10 $\mu g.kg^{-1}$ body weight but states that for HCH insufficient information is available to determine an ADI.

Table 1.11. Quality requirements for drinking water ($\mu g.l^{-1}$)

Country	*Max. conc.*	*Remarks*	*References*
The Netherlands	*0.1*	*pest., per compound*	*Waterl.besluit (1984)*
	0.5	*pest., total*	
EEC	*0.1*	*pest., per compound*	*EEC (1980)*
	0.5	*pest., total*	
USA	*0.21*	*lindane, 24 hr average*	*WDSHS (1984)*
	2.9	*lindane, max. conc.*	
	0.054	*lindane, in preparation*	
	0.004	*lindane, acceptable level*	*WDSHS (1983)*

1.2.5. Storage and transport

The International Regulations on Railtransported Dangerous Goods (RID), the European Agreement concerning Roadtransported Dangerous Goods (ADR), the Regulations on Rhine-transported Dangerous Substances (ADNR) and the Intergovernmental Advisory Maritime Organization (IMCO) place HCH in class of risk 1 (Staatsuitgeverij, 1978). Each organization specifies the measures to be applied and the labels to be used in storage and transportation of this substance.
The Principal Inspectorate for Environmental Hygiene of the Ministry for Housing, Planning and Environmental Control has given guidelines with respect to permit allowances and nuisance act regulations for storage depots of pesticides or their active components.

1.2.6. Remainder

- Chemical Substances Act: organic halogenated compounds are considered to be chemical waste at concentrations higher than 5 $g.kg^{-1}$ (Staatsblad 435, 1977).

- Substances decree on Cosmetics: cosmetics are not permitted to contain 1,2,3,4,5,6-hexachlorocyclohexane.
- Medicines Act: the use is regulated (e.g. for the control of head-lice).
- Regulation for fodder (Besluit VVR, 1975): α-HCH is permitted in fodder for ruminants up to 30 $\mu g.kg^{-1}$ and in feed for fatting-chickens up to 80 $\mu g.kg^{-1}$; for the total of HCH these levels are 200 and 500 $\mu g.kg^{-1}$, respectively.
- Permitted use: lindane may be used for the control of insects indoors or outdoors, except for the following applications: in winter on consumption crops under glass; on dairy cattle or on poultry; on mushrooms. Applications on carrots and tuberous plants are permitted before sowing only. For the use on other livestock about 45 additional products are permitted.

2. PRODUCTION, APPLICATIONS, SOURCES AND EMISSIONS

2.1. PRODUCTION IN EUROPE

HCH is synthetized on a large-scale by the reaction of benzene with chlorine under the influence of light. During this process a mixture of HCH-isomers is obtained, as well as a number of side products like hexachlorobenzene (HCB). The composition (in %) of the raw reaction product is given in table 2.1.

Table 2.1. Composition of the raw reaction product HCH (in %)

HCH-isomer					*Other compounds*	*References*
α	β	γ	δ	ε		
65-70	*7-10*	*14-15*	*7*	*1-2*	*1-2*	*CIEL (1982)*
64-65	*7-10*	*9-14*	*8-9*	*1-4*	*2*	*OHM/TADS (1985)*
65-70	*6- 8*	*12-15*	*2-5*		*5*	*TDB (1985)*
55-70	*5-14*	*10-18*	*6-10*	*3-4*		*Kirk-Othmer (1979)*

In the past the raw reaction product was directly applied in insecticide formulations; currently in most countries only the use of lindane is permitted. The product provided under this name contains only 0.2-0.5% α-HCH. Also in The Netherlands the permitted use as a pesticide is restricted to products on the basis of lindane. In The Netherlands, however, products are formulated for export from technical or enriched HCH. Although enriched HCH is not produced in Western-Europe (De Bruin, 1979), it may be imported from other countries. Production figures on HCH are extremely rare. When data are available it is often not clear whether the figures concern technical HCH or lindane. Therefore, the data in table 2.2. are mostly based on indications and estimates rather than on solid figures (CID-TNO, 1985; De Bruin, 1979; Van de Waerdt, 1983).

In the United States as well as in Europe various production installations have been closed since 1950. In The Netherlands the production was stopped in 1954. Large production companies exist for instance in India and Turkey,

although the production in also these countries is decreasing to some extent (CID-TNO, 1985). In Western-Europe there are still 6 production companies; in Austria, France (2), West-Germany and Spain (2) (SRI, 1984). Further production companies exist in East-Germany, Poland, Czechoslovakia, Yugoslavia and the USSR (De Bruin, 1979; CID-TNO, 1985). Data on capacity and production are very seldom available. Furthermore, pesticides based on lindane are formulated at many locations. De Bruin (1979) estimates the number of formulating companies within the EEC in 1979 at appr. 100. In table 2.2. an estimate is presented of the production in various countries. The data are based on information obtained in the year mentioned, extrapolated to 1983.

Table 2.2. Production of technical HCH and lindane in various countries in 1983 in ktons per year

Country	*Technical HCH*	*Lindane*	*Base year*
West-Germany	*15*	*1.5*	*1977*
France	*28*	*3.0*	*1977*
Austria	*1*	*0.1*	-
Spain	*10*	*1.0*	*1984*
Eastern-Europe	*10*	*1.0*	-
USSR	*20*	*2.0*	*1984*
Turkey	*3*	*0.1*	*1977*
India	*30*	*3.0*	*1981*
Pakistan	*1*	*0.1*	*1979*
South-America	*20*	*2.0*	*1979*
Other countries	*12*	*1.4*	-
World	*150*	*15.2*	-

2.2. IMPORT AND EXPORT IN THE NETHERLANDS

Data on the import and export are based on mostly limited information from customs declarations (CBS, 1981-1984). The term "hexachlorocyclohexanes" may refer to lindane, technical HCH or enriched HCH. Assuming that mainly one product is traded per country, it is possible to indicate the various amounts on the basis of the price of the products. In the period 1981-1984 the prices per country appear to amount to either more than Dfl.20,- per kg (pure lindane), or less than Dfl.10,- per kg (technical HCH). Considering the sharp distinction in price levels the assumption that one country produces only one product seems reasonable. The results of this calculation are given in table 2.3.

Table 2.3. Import surplus of hexachlorocyclohexanes in tons per year

Products	*1981*	*1982*	*1983*	*1984*
Price more than Dfl. 20 per kg	*14*	*49*	*66*	*106*
Price less than Dfl. 10 per kg	*22*	*30*	*33*	-

Apparantly, in 1984 no technical HCH was imported. However, according to the CBS the impure product is present in 1985 again (Boon, 1985). The import and export of formulated products cannot be deduced from the import and export statistics, as these products are classified under collective terms such as insecticides, pesticides in small packages etc. An attempt by the CBS to obtain more detailed information by testing a sample of customs declaration forms did not lead to any results (Boon, 1985).
Production statistics of the pesticide industry form another source of information (CBS, 1978-1983), although the problem arises that not all producers are included under the heading "pesticides industry" in the "Standaard Bedrijfsindeling". In comparing the purchase figures from the production statistics with the import data, the effect of companies having possibly been missed appears to be small (table 2.4.).
Considering the rather extensive use of HCH in developing countries (Van de Waerdt, 1983) the import of foodstuffs could be another source of HCH. This especially applies to mixed feed consisting for 75% of imported substances, among which many residual compounds and derivates of vegetable products. However, as measuring results indicate that in 1983 and 1984 only appr. 40 kg HCH was imported in this manner (mainly in mixed fodder), the contribution made by import is negligible.

Table 2.4. Comparison of the import of HCH and the purchase and sale by the pesticides industry in tons per year (CBS, 1981-1983)

	1978	*1979*	*1980*	*1981*	*1982*	*1983*
Import	*205*	*201*	*155*	*181*	*86*	*120*
Purchase	*200*	*200*	*170*	*180*	*90*	*130*
Sales (formulated)	*1200*	*500*	*500*	*600*	*500*	*500*
*Average content * (%)*	*17*	*40*	*34*	*30*	*18*	*26*

** the content of the product sold most at present amounts to 21%*

2.3. APPLICATIONS IN THE NETHERLANDS

2.3.1. Application in agriculture and cattle breeding

- Treatment of soil

The most important application is the control of soil insects like leatherjackets and wireworms. The products are deposited in the form of spraypowder, liquid or pellets and subsequently mixed with the soil. About 30 products are permitted. Such soil treatments are applied especially for the culture of maize and beet. The treatment of soil with HCH is also applied in greenhouse horticulture to control wood-lice, carrot centipedes, etc. The quantities used in 1984 are estimated at 20 tons, of which 0.3 ton in the greenhouse horticulture (Mulder, 1985; Nollen, 1985).

- Treatment of seed

Pretreatment with lindane-containing products may prevent the seeds of especially beet, maize, corn and flax from being affected by soil insects. The products are applied in the form of a powder, sometimes in combination with fungicides. About 5 products are permitted. The quantities used in 1984 are estimated at appr. 2 tons (Mulder, 1985).

- Other applications in agriculture and horticulture

The use of lindane-containing products to control leaf insects has been reduced considerably. In greenhouses and bulb-sheds, smoke-products and spraypowders are still used to control inaccessable insects. The quantities used in 1984 are estimated at appr. 0.5 ton (Mulder, 1985).

- Parasite control in cattle

With respect to the application in cattle (e.g. sheep) the estimated quantities used in 1984 amount to appr. 2 tons (Kommery, 1985; Mulder, 1985).

2.3.2. Other applications

- Control of crawling insects in buildings

In accordance with the statistics on pest control (CBS, 1981-1984) in 1981 appr. 100 kg was used by municipalities. Besides a large number of private

companies are active in this sector. The quantities used by these companies are estimated at appr. 300 kg per year (Bosman, 1985). Hence, the total use is appr. 0.4 ton per year.

- Control of wood-insects in buildings

Both municipalities and private companies use lindane-containing products to control woodworms and long-horned beetles in buildings. The estimated quantities used amounted maximally 2 tons in 1984 (Bosman, 1985).

- Wood preservation

The use of lindane-containing products for the preservation of wood has decreased strongly, especially since meranti, currently very much in use, does not need preservation (Van der Drift, 1985). The amounts used in 1984 have been estimated at 1.8 tons.

- Domestic use

More than 30 lindane-containing products are permitted for the control of insects in and around the house. The products are applied in the form of liquids or powders, often by means of spray-cans or strips. The amount of lindane concerned is estimated at appr. 0.1-0.2 ton per year (Bosman, 1985).

Data concerning the amounts turned over in The Netherlands by the members of the Dutch Foundation for Phytopharmacy (NEFYTO, 1985) are available over a few years. These data, completed with data on 1977 (De Bruin, 1979), are given in table 2.5.

Table 2.5. Pesticides sold by the NEFYTO-members in The Netherlands: quantities of lindane used in tons per year

Year	*Tons*
1974	*19*
1975	*21*
1976	*25*
1977	*24*
1984	*27*
1985	*28.5*

In spite of attemps to reduce the use of lindane-containing products, the quantities turned over appear to have increased by appr. 50% in the past ten years.
The NEFYTO-members account for 80-90% of the sales in the overall pesticide market. For individual compounds or specific applications this percentage may be much lower, this being the case with HCH for wood preservation and domestic use. The total amount of HCH turned over in The Netherlands is therefore estimated at appr. 29 tons in 1984.
Details on the distribution of this amount over the various applications have not been identified by the NEFYTO. An estimate of this distribution is presented in table 2.6.

Table 2.6. Distribution of the use of HCH over the various applications in 1984 (estimated quantities)

Application	*Tons*
Treatment of soils	*20.0*
Treatment of seeds	*2.0*
Leaf applications and agricultural space treatments	*0.5*
Control of parasites in cattle	*2.0*
Control of crawling insects in buildings	*0.4*
Control of wood-insects in buildings	*2.0*
Wood preservation	*1.8*
Domestic use	*0.2*
Remainder	*0.1*
Total	*29.0*

2.4. EMISSIONS

In this document an emission is defined as the desirable or undesirable introduction of a substance into the environment, irrespective of the function of the substance.
Production of HCH in The Netherlands has not taken place since the fifties. At several locations the soil is still contaminated with HCH-containing waste as a result of production in the past; this will be discussed in detail in section 4.2.
In 1984 a total of 106 tons of lindane was processed for the manufacture of pesticides, of which, according to De Bruin (1979), appr. 0.1% reaches the water at the formulating plant. Without control measures this should result

in an emission of 100 kg into the water. The emission is generally effectively controlled by adsorption methods and therefore the residual emission may be considered insignificant. However, this process leads to solid wastes which so as far as is known- are carried off to the Incineration Plant Rijnmond (AVR) in most cases. Charges that have failed and contaminated packing materials are also transported to the AVR. As the emissions into the air are controlled by absolute filters, the residual emissions in these cases are quite insignificant.

By application of lindane-containing pesticides appr. 29 tons enter the environment each year (table 2.6.), of which 22 tons enter the soil directly in the treatment of soils and seeds. As a result of spray mist drift and condensation emissions may also occur into air, surface water or, through sewage, into sewage sludge. As a result of fumigation emissions occur into the air .

The control of ectoparasites in cattle results in emission into soil by dripping (30%) and into air by evaporation. Emissions into water and sewage sludge also take place at locations at which scrubbing-sewers and spraying-grounds drain into a sewer. Leftovers from dips (used for sheep) lead to emission into the soil (directly, or indirectly via the manure-pit). Discharges into surface waters are not permitted.

Wood preservation and some of the domestic uses mainly cause emissions into the air. The destruction of treated timber may also result in emissions into the air. The amounts emitted cannot be estimated.

According to the Foundation of Applied Research for Waste Water Treatment (STORA, 1985), both α-HCH and γ-HCH are present in domestic waste water. The total amount is appr. 200 kg, of which 180 kg is lindane (Feenstra and Van der Most, 1985). HCH was consistently found in another investigation at six water treatment plants; the efficiency of the purification varied from 14 to 79% (average value: 38%; RIZA, 1986).

The estimated emissions to the various environmental compartments are summarized in table 2.7.

Trans-frontier emissions occur through water and air. The total trans-frontier flux of lindane through the surface water amounts to appr. 2.2 tons. Of this amount appr. 1.8 tons were supplied via the Rhine (averaged over the period 1980-1983; IMP, 1985). The measured concentrations in the river Meuse are in the same order of magnitude and, based on the ratios between the discharge levels, the import via the Meuse is estimated at appr. 0.2 ton per year (Faasen, 1985).

Considering the nature of the applications the contribution of transfrontier emissions is relatively small. The contribution from Belgium and West-Germany is estimated at 0.04 and 0.03 ng.m^{-3}, respectively, whereas that from the rest of Western-Europe is even smaller (chapter 3).

Table 2.7. Estimated HCH emissions to the various environmental compartments in tons per year (1984)

Cause	*Soil*	*Water*	*Air*	*Waste*
Formulation of pesticides	-	+	+	+
Treatment of soils, greenhouse horticulture excluded	*19.7*	+	+	-
Greenhouse horticulture	*0.3*	-	+	-
Treatment of seed	*2.0*	-	+	-
Application to leaves	*0.2*	+	*0.3*	+
Control of parasites in cattle	*1.5*	+	*0.5*	+
Control of crawling insects in buildings	-	-	*0.4*	-
Control of wood-insects in buildings	-	*0.1*	*1.9*	-
Wood preservation	*1.8*	-	+	+
Domestic uses	-	*0.1*	*0.1*	+
Other uses	-	-	*0.1*	-
Total	*25.5*	*0.2*	*3.3*	+

+ : possible source of probably limited extent

2.5. SUMMARY AND CONCLUSIONS

In The Netherlands HCH has not been produced since 1954. In spite of the endeavour to reduce the use of lindane-containing products, there are strong indications that this use has increased by appr. 50% over the past ten years. The application as a pesticide is the most important source of contamination of the environment. In 1984 appr. 29 tons of HCH was used, at which mainly emission into the soil occurred (25.5 tons); the remaining emissions into water and air amounted to appr. 0.2 and 3.3 tons, respectively. Most of the lindane is emitted into soil by the treatment of soils for the control of soil insects, in particular in connection with the cultivation of crops such as maize and beet, as in greenhouse horticulture.

3. DISTRIBUTION AND TRANSFORMATION

3.1. BEHAVIOUR IN SOIL

3.1.1. Distribution

In Germany (Hamm and Sprendlingen) HCH appears to be dispersed to more remote soils being blown about from (open) chemical dumping grounds. In Hamm this mainly concerned β-HCH at levels of 0.17-0.5 $mg.kg^{-1}$ in plough furrows. At a depth of 25-50 cm 0.04 $mg.kg^{-1}$ was found; in deeper parts no residues were found, or only traces. From this observation Kampe (1980) concluded that transportation of β-HCH to groundwater is unlikely, explaining this on the basis of low water solubility and anaerobic degradation. In Hessen (Weiterstadt) however, β- and δ-HCH were also found in deeper layers (40 cm)(Kampe, 1980).
Schrimpff (1985) investigated immissions by the deposition of lindane and α-HCH in Bavaria. In some summers, after a very dry spring, concentration peaks in the groundwater were found very soon (appr. 1-2 months) after the peaks in the precipitaion rates. This may be explained by the occurrence of cracks in the soil.

3.1.2. Adsorption

In table 3.1. the various Freundlich relationships, found for the adsorption-isotherm of lindane to sediments and soils, are presented. Some adsorption parameters measured for α- and β-HCH are given in table 3.2.
From these tables the adsorption coefficient K appears to be related to the organic carbon content. The parameter 1/n fluctuates around 1. Table 3.1. may serve as a guide for the quantification of the adsorption of lindane to the soils in The Netherlands, with the variability in the results being indicative for the inaccuracy.
The linear dependency of the adsorption of the organic matter content does not hold at higher organic carbon contents. According to Wahid and Sethunathan (1979) this applies to an organic matter contents in the soil of 4% and more.

Table 3.1. Freundlich constants for γ-HCH (materials ranging from submerged sediments to soil)

The parameters K and 1/n are applicable to the relationship $S = K.c^{1/n}$, in which: S = adsorption ($mg.kg^{-1}$)
K = adsorption constant
c = concentration in pore water ($mg.l^{-1}$)

Organic C (%)	K	1/n	References
0 (sand)	0.3	1	Kay and Elrick (1967)
0 (silt)	16.5	1	Kay and Elrick (1967)
0 (quartz)	0		Wirth (1985)
0 (kaolite)	0		Wirth (1985)
0 (illite)	26	0.90	Wirth (1985)
0 (Na-bentonite)	88	1.02	Wirth (1985)
0.35	7.9	0.80	Hamaker and Thompson (1972)
0.5	10	1	Kay and Elrick (1967)
1	2000	1.3	Weil et al. (1973)
1.2	8.9	0.90	Wahid and Sethunathan(1979)
1.3	11	1	Uchrin and Katz (1985)
1.5	24	1	Kay and Elrick (1967)
1.7	17	1	Kay and Elrick (1967)
1.9	22.7	1	Kay and Elrick (1967)
2.0	25.1	0.80	Wahid and Sethunathan(1979)
2.2	20.4	1	Kay and Elrick (1967)
2.6	13	1	Uchrin and Katz (1985)
3.5	45.7	1	Hamaker and Thompson (1972)
3.8	31.6	0.80	Wahid and Sethunathan(1979)
5.3	446.7	1.10	Wahid and Sethunathan(1979)
6.2	36.3	1	Kay and Elrick (1967)
12.8	331	0.98	Hamaker and Thompson (1972)
18	295	0.92	Hamaker and Thompson (1972)
38.3	368	1	Kay and Elrick (1967)
43	608	1.01	Wirth (1985)

Table 3.2. Freundlich-constants for α-HCH and β-HCH (Wahid and Sethunathan, 1979)

Organic C (%)	α-HCH		β-HCH	
	K	1/n	K	1/n
1.2	28.2	1.60	5.1	0.82
2.0	52.5	1.26	39.8	1.20
3.8	70.8	0.94	79.4	0.80
5.3	501.2	1.16	158.5	0.80

In the literature there is no agreement on the remobilization c.q. desorption of lindane, when "clean" water is exposed to polluted soil. Bowman et al. (1965), Adams and Li (1971), Bouchier and Lee (1972) and Wirth (1985) found a considerable desorption, even for a material such as peat (50-100%). Others (Wahid and Sethunathan, 1979) observed a very strong adsorption hysteresis and consider the remobilization of lindane negligible.
Generally, the adsorption of HCH-isomers is rather strong. With respect to applications in agriculture this had led to the assumption that lindane poses no threat to the groundwater. At the high concentrations occurring in local soil pollution, however, washing out of a small percentage of the initial amount may lead to unacceptable concentrations in groundwater. This is most likely in highly permeable soils with a low organic carbon content (sandy soils deficient in humus). In a soil deficient in humus with an organic content of 1%, for which a K-value of 10 may be assumed for lindane, a pollution front moves downwards with the nett deposition at an average velocity of 2.5 cm per year. At higher or lower organic carbon contents this velocity varies proportionally. In view of the transformation rates (see 3.1.2.), the probability of lindane reaching the groundwater with a velocity of 2.5 cm a year is very slight. For instance, calculations indicate that at a thickness of the unsaturated zone of 50 cm and a DT50 of 500 days (see table 3.3.), 1/160 % of the dose will reach the groundwater; at a DT50 of 1,000 days at most 1% would reach the groundwater.

3.1.3. Transformation

- Transformation rate

With respect to the data mentioned below, only those of Kampe (1980), Doelman et al. (1985) and Doelman and Zehnder (1986) concern cases of soil pollution. All the other studies are aimed at the behaviour of lindane after agricultural applications.
Although transformation in the soil is mainly microbiological, in alkaline soils abiotic transformation takes place as well (Portmann, 1979).
It is striking that the degradation curves found in the literature often comprise a linear part (the degradation rate is constant), taking a more exponential character only at relatively low HCH concentrations.

In submerged soils the transformation rate is higher than in aerated soils (Khonen et al., 1975), however, recent studies in The Netherlands on polluted soil from "Twente" indicate the opposite (see below).
Several investigators isolated *Clostridium* -species from submerged rice fields and used these species to study the degradation of HCH (e.g. Ohisa and Yamaguchi, 1979). The latter stated that in rice soils *Clostridium* -species are the main HCH-degraders. These organisms are strictly anaerobic and their occurrence in aerated soils is quantitatively low. However, in these soils the HCH-degrading facultative anaerobic organisms like enterobacteria or especially *Bacillus* -species, that also live in anaerobic conditions, are present in large numbers. These species, however, are only capable of degrading HCH in anaerobic conditions. It is known that in aerobic soils micro-anaerobic zones occur, for example pores filled with stagnant water in which HCH-degradation may take place (Haider, 1980).

α-HCH

Kampe (1980) assumed that the half-life figures for α-HCH and γ-HCH are the same. Doelman et al. (1985) carried out laboratory studies on the degradation under anaerobic and aerobic conditions in a polluted Dutch sandy loam with 6.5% organic matter, at concentrations of appr. 5,000 mg.kg^{-1}. They found constant degradation rates of 10 and 14 mg.kg^{-1} per day, respectively. At lower concentrations (appr. 4,000 mg.kg^{-1}) the average transformation rate appeared to be higher (24 mg.kg^{-1} per day), both under aerobic and anaerobic conditions. The transformation was ascribed to microbial processes.
According to the authors, the higher persistence of HCH-isomers at locations of soil pollution in The Netherlands (chiefly dumping grounds) is caused by the poor availability of HCH (in clods and the like) to microorganisms.
From current studies on HCH polluted soil by Doelman and Zehnder (1986) α-HCH appears to degrade considerably better in aerobic conditions (aerated) than in anaerobic conditions (non-aerated slurry), both in the laboratory and in soil in green-houses. In a slurry (containing 7-9 mg O_2.l^{-1}) and in aerated humid soil at appr. 15°C in a green-house, a decrease in concentration of 80 and 40%, respectively, was found within 30 days (initial concentration 350-400 mg.kg^{-1}). The decrease was less at lower oxygen contents. In addition, β-HCH appeared not to be degradable in the mixture. The difference with respect to the oxygen levels found and those

from the literature may be explained by the higher concentration levels used (derived from local soil pollutions in Twente) as compared to concentrations resulting from agricultural applications. The degradation products may be toxic at high concentrations, however, the products formed in aerobic conditions are less toxic.
Assuming the degradation process to be of a first order reaction, T1/2-values for α-HCH of 125 and 48 days may be deduced from laboratory studies by MacRae et al. (1984) in aerobic and anaerobic conditions, respectively (soil from Japan with 4.0% organic C).

β-HCH

MacRae et al. (1984) carried out laboratory studies on the transformation of β-HCH in a soil from Japan with 4% organic C, under aerobic and anaerobic conditions. From the transformation rates found, half-life figures of 91 and 122 days, respectively, may be deduced (dosage 20 mg β-HCH per g of soil). Kampe (1980) derived a DT50 of 8 years from the relative HCH-isomers composition of residues in a 10 year old dump of HCH waste. Korte (1980) stated that the high β-HCH contents (relative to α-HCH and Γ-HCH) in human adipose tissue, urine and blood are not primarily caused by applications of technical HCH as a pesticide before the seventies, but mainly because α-HCH is eliminated more rapidly from the soil than β-HCH. The studies by Suzuki et al. (1975) clearly indicate this strong persistence of β-HCH, although it strongly depends on the type of soil and, for example, for α-, β-, γ- and δ.HCH, appeared to be almost negligible in a loamy soil with 10.5% organic matter.
As mentioned before, the lowerr biodegradability of the β- isomer in relation to the α-isomer has also been observed by Doelman.

γ-HCH

Table 3.3. gives a summary of the literature data on the half-life of lindane in the soil. Some of these half-life times have been calculated from the data in the articles referred to; in these cases a first order degradation process is assumed.

Table 3.3. Degradation rate of lindane in various types of soil (characterized by the organic carbon content), expressed as DT50 or DT95 (time required to degrade 50% and 95 %, respectively, of the initial concentration)

DT50 (days)	DT95 (days)	Conditions (temp.)	OC-content (%)	References
720		aerobic		Menzie (1972)
	1080-3600	aerobic		Edwards (1966)
Field 88-1146		aerobic	0.28-23	Rao & Davidson (1982)
415		aerobic (22°C)	0.5	Kampe (1980)
106		aerobic (22°C)	2.9	Kampe (1980)
Lab 276		aerobic	3.0	Rao & Davidson (1982)
Lab 174		anaerobic (23°C)	3.0	Rao & Davidson (1982)
Lab 14		anaerobic (35°C)	4.0	MacRae et al. (1984)
Lab ∞		aerobic (35°C)	4.0	MacRae et al. (1984)
Lab 12		anaerobic (25°C)		Kohnen et al. (1975)
Lab 100		aerobic (25°C)		Kohnen et al. (1975)

The DT50-values obtained from the various investigations vary considerably, this being caused by differences in type of soil and, possibly, by differences in temperature. Assuming that lindane is not washed out below the level of the plough furrows (appr. 20 cm), a half-life of 350 days for lindane means that one year after the application 50% of the dose is still present. Annual application of lindane will finally result in the presence of a basic residue level 1 time the annual dose. At a half-life of 175 days this ultimate level is 1/3 of the annual dose.

δ-HCH

MacRae et al. (1984) found half-life times of 62 and ∞ days under aerobic and anaerobic conditions, respectively, using the same type of soil as in their studies on β-HCH.

- Degradation products

It is not known to what extent the groundwater is threatened by the metabolites of lindane and the other HCH-isomers. An overview of the adsorption coefficients and half-life times of the metabolites (tables 3.4. and 3.5.) is not available. It is known, however, that under aerobic conditions chlorobenzenes are not degradable, or very slowly (Bouwer, 1982). Table 3.4. presents the transformation products obtained under aerobic conditions. As shown in the table, γ-pentachlorocyclohexene dominates. Table

3.5. presents the transformation products obtained under anaerobic conditions; under these conditions Cl^- is formed (MacRae et al., 1969; Haider and Jagnow, 1975).

Besides Kohnen et al. (table 3.5.), other authors also reported the formation of α-HCH from γ-HCH: Engst et al. (1979) with *Pseudomonas aeruginosa* in a mixed culture under anaerobic conditions and Vonk and Quirijns (1979) did so for two types of soil, under exclusively anaerobic conditions. In the latter study *E. coli* was responsible for the transformation. Eichler (1980) doubts whether such conditions occur in the natural environment. He assumes that a possible isomerization is of very minor importance and he supposes that when residues of α-, β- or γ-HCH are found, these are not caused by the use of lindane in agriculture but the result of past applications of technical HCH. A similar conclusion is drawn by Haider (1980). Koss et al. (1980) proved the potential transformation of γ-HCH into α-HCH with entero-bacteria, but they are unable the indicate the environmental relevance of this finding.

Therefore, the persistence of the HCH-isomers in the soil is high, both in aerobic and anaerobic conditions. Although the half-life times vary considerably, they are generally more than half a year. In cases of local soil pollution with high concentrations, the persistence is likely to be much higher than the values mentioned in section 3.1.2. This is probably caused by the poor accessibility of HCH to the micro-organisms and by the fact that in these cases the concentrations reach toxic levels.

Table 3.4. Transformation products of HCH-isomers under aerobic conditions

Compound	Products	Remarks	References
γ-HCH	γ-pentachlorocyclohexene	percolated soil + batch humid soil	Yule et al. (1967)
γ-HCH	γ-pentachlorocyclohexene	laboratory	Vonk and Quirijns (1979)
γ-HCH	2,3,4-tetrachlorophenol, pentachlorophenol, 1,2,3-trichlorobenzene, tetrachlorobenzenes, pentachlorobenzene, hexachlorobenzene, γ-pentachlorocyclohexene	lysimeters; over grown; 14% total residues in soil, of which 1% metabolites	Korte (1980)
All isomers	γ-pentachlorocyclohexene, chlorobenzenes, chlorophenoles	laboratory	Haider (1980)

Table 3.5. Transformation products of HCH-isomers under anaerobic conditions

Compound	Products	Remarks	References
γ-HCH	γ-3,4,5,6-tetrachlorohexane, γ-pentachlorocyclohexene	submerged soil in trace amounts	Tsukano and Kobayashi (1972)
γ-HCH	γ-3,4,5,6-tetrachlorocyclohexane,	submerged sandy	Mathur and
	γ-chlorocyclohexene,	loam	Saha (1977)
	1,2,4-trichlorobenzene, 1,2,3,5-tetrachlorobenzene 1,2,4,5-tetrachlorobenzene 1,2,3,4-tetrachlorobenzene	in small amounts	
γ-HCH	γ-pentachlorocyclohexene α-HCH (< 1% of the dose)	submerged grey-brown podzolic on loess	Kohnen et al. (1974)
γ-HCH	γ-tetrachlorocyclohexene	laboratory	Vonk and Quirijns (1979)
All isomers	tetrachlorocyclohexene-isomers	laboratory	Haider (1980)

3.2. BEHAVIOUR IN SURFACE WATER

Most data on the behaviour of HCH in surface water concern lindane.

3.2.1. Distribution

Evaporation and adsorption to solid particles are important processes. Reverse processes also play a part: deposition from the air (see 3.3.2.) and remobilization from silt and sediment.

- Evaporation

Büscher et al. (1964) demonstrated that aeration of aqueous solutions of lindane results in a loss of 10% in three days. Bowman et al. (1964) found losses of 30% from test solutions within 20 hours. These losses were ascribed to a co-distillation process as they were greater than could be explained by evaporation alone (the Henry constant amounts to 4.93×10^{-7} $atm.m^3.mol^{-1}$ at 25 °C according to MacKay and Walkoff (1973) corresponding to a dimensionless Henry constant 2.05×10^{-5}). On theoretical (thermodynamical) grounds MacKay and Walkoff (1973) confirmed that evaporation of water is an important process in the loss of HCH. The results of calculations are consistent with experimental values. Lichtenstein and Schulz (1970) found a 16.5% HCH-loss from a non-aerated aqueous solution in 24 hr at 30 °C.

- Adsorption

HCH can adsorb to particles: the more organic carbon the solid material contains the higher the adsorption (Portmann, 1979). Freitag et al. (1985) studied the accumulation of HCH in active sludge. After 5 days they found an accumulation factor of 1,200 for β-HCH and 800 for γ-HCH. With respect to data on the adsorption of HCH, reference is made to the behaviour in soil (section 3.1.2.).

3.2.2. Transformation

- Abiotic transformation

It is obvious that an insecticide such as γ-HCH should be fairly stable to meet its requirements in a number of residual applications. The compound is relatively little reactive with respect to heat and daylight. In alkaline

environments, however, the compound is unstable and decomposes to trichlorobenzenes and hydrochloric acid (Ullmann, 1972), resulting in a more rapid degradation in the usually weak alkaline sea water (pH 8.3) (Portmann, 1979).
The photochemical transformation of α- and γ-HCH has been studied by Malaiyandi et al. (1982): in pure water and in fulvic acid solutions both isomers are transformed to a limited extent under the influence of direct sunlight. Furthermore the authors found indications for some transformation from γ-HCH to α-HCH.

- Biotic transformation

Bacteria and other organisms may degrade HCH. Various metabolites have been identified, such as pentachlorocyclohexane, tetra- and trichlorobenzenes and chlorophenols (Portmann, 1979).
Freitag et al. (1985) determined the biodegradation of β- and γ-HCH and found less than 0.1% mineralization within 5 days. Engst et al. (1979) found metabolization and isomerization of γ-HCH to α-HCH in a mixed culture with *Pseudomonas aeruginosa* being the main component. Haider (1975) observed a rapid transformation: 90% Cl-free metabolites within 4 days by anaerobic bacteria. It may be concluded that while in anaerobic conditions the compound biodegrades rapidly, in aerobic conditions biodegradation is limited.

3.3. BEHAVIOUR IN AIR

3.3.1. Distribution

Lindane is used as an insecticide in the cultivation of various plants, for the treatment of soil and seeds as well as for the spraying of the crops. As to the latter application, strong emissions take place into the air which gradually decrease, lasting for several hours to several days. Consequently, concentrations may occur in the air nearby (leeward) a treated field, initially amounting to some dozens of $\mu g.m^{-3}$, however, according to calculations, decreasing after the short lasting emission.
HCHs emitted into the soil and water will evaporate slowly from these compartments. This is related with the low saturation pressure and high adsorption, respectively (Verschueren, 1977). For this reason the distribution is only calculated on the basis of the emissions into air,

which has been estimated at 3.3 tons per year in The Netherlands (see table 2.7.). Since this emission originates from scattered sources it results in a concentration contribution at a national scale. From calculations according to Van Egmond and Huygen (1979) and Van Hout et al. (1983) it has been estimated for scattered sources that the HCH concentration resulting from emissions in The Netherlands is appr. 0.13 $ng.m^{-3}$. The contribution of the sources abroad was estimated, assuming residence times characteristic for HCH of 1 and 10 days, respectively, . Data on the HCH emissions i. surrounding countries are lacking; therefore the emissions per inhabitant were assumed to be equivalent to those in The Netherlands. The contributions from Belgium and West-Germany were estimated at 0.04 and 0.03 $ng.m^{-3}$, respectively, whereas the contribution from the rest of Western-Europe will probably be even smaller. On this basis the large-scale concentrations of HCHs in The Netherlands are estimated at appr. 0.2 $ng.m^{-3}$, this being about one third of the measured values (chapter 4).

3.3.2. Deposition

- Dry deposition

No data have been found in the literature on measurements of the dry deposition rate of HCH (Eisenreich et al., 1981; Bidleman and Christensen, 1982). The deposition rate (v_d) depends on the nature of the surface above which the organic components are present. For surface water the v_d = 0.04-0.14 $cm.s^{-1}$. For The Netherlands with its vegetated surfaces and its generally humid climate, a value of 0.1-0.3 $cm.s^{-1}$ seems representative. For the process of dry deposition the removal constant $k_v = v_d/h$, in which h is the height of the mixing layer. At h = 1000 m, $k_v = 10^{-6} - 3x10^{-6}$ s^{-1}; the half-life for dry deposition is 2-8 days. From this it may be derived that the annual flux to soil and water in The Netherlands amounts to 0.5-1.5 tons at an outdoor air concentration of 0.4 $ng.m^{-3}$. However, this should be considered a rough estimate: in view of the high emissions in the past it is possible that locally a nett flux takes place from the soil.

- Wet deposition

From measurements by the KNMI/RIVM it follows that throughout the year HCH is present in rainwater in concentrations of 10-140 $ng.l^{-1}$ (measuring location De Bilt). In table 3.6. a summary is given of the amounts of precipitation and the HCH concentrations in rainwater, observed in 1983.

Table 3.6. Amounts of precipitation and HCH concentrations in rainwater in 1983 (KNMI/RIVM, 1983)

	Range	Average
Precipitation (mm per month)	*25 - 121*	*76*
α-HCH concentration ($\mu g.l^{-1}$)	*<0.01 - 0.02*	*0.01*
γ-HCH concentration ($\mu g.l^{-1}$)	*0.01 - 0.18*	*0.06*
Amount of α- and γ-HCH ($\mu g.m^{-2}$ per month)	*0.4 - 22.4*	*5*

The differences among the locations Witteveen, De Bilt and Vlissingen are relatively small: the wet deposition amounted to 11.9, 15.8 and 20.9 $\mu g.m^{-2}$, respectively, in the first half of 1983. In the second half the wet deposition appeared to be considerably less (10 $\mu g.m^{-2}$ as compared to 50 $\mu g.m^{-2}$). Measurements indicate that the total amount of HCH removed in 1983 by wet deposition amounts to 2.5 tons. In view of the measuring data of 1985 there may be tendency to decrease.

The wash-out may also be estimated by the method given by Eisenreich et al. (1981). The flux $F = (J \cdot X_a)/H$, in which H is the Henry constant, J the amount of rainfall and X_a the outdoor air concentration at an average height of 1000 m. At an average temperature of 11 °C (representative for The Netherlands) The Henry constant is 5.0×10^{-6}, as calculated from the extrapolated value $P_v = 2.84 \times 10^{-4} P_a$ and an estimated solubility of 7 $g.m^{-3}$. Hence, at a surface of 41×10^9 m^2 (The Netherlands) 2,500 kg is deposited. This (calculated) result is consistent with the flux determined on the basis of measurements (table 3.6.).

3.3.3. Transformation

Considering its low concentrations in air and limited presence of HCH in air as compared to other compartments, the total transformation in air is relatively insignificant. For HCH no data are given on the reactivity in relation to the hydroxyl radical. However, it may be expected that HCH is little reactive. At a k_{OH} = 200-500 $ppm^{-1}.min^{-1}$ (estimated on the basis of structural characteristics) and a $[OH^{\bullet}]$ of 2×10^{-8} ppm, T1/2 = 50-120 days. In the literature photolytical transformations of HCH are mentioned: α-HCH may arise from all forms by (photo)isomerisation (Hamada et al., 1981; Malaiyandi and Shah, 1984; Malaiyandi et al., 1982). This transformation explains why the α-isomer is found especially in areas far from sources, such as Norway and Sweden (Villeneuve and Holm, 1984; Carlberg et al.,

1983). However, this statement is made with some reservation since during the processing of the analytic procedure a loss of the volatile γ-HCH may occur.

3.4. BEHAVIOUR IN BIOTA

In water the bioaccumulation factor amounts to 100-1000. Foodchain effects are expected to be limited. The equilibrium levels are rapidly reached (a few days to two weeks).
In soil organisms the accumulation/elimination process is slower (equilibrium levels are reached within 5 weeks) and there is hardly any bioaccumulation. In plants lindane is mainly absorbed through the roots; after reaching the equilibrium levels the highest concentrations are found in the roots (see also chapter 5).
Further attention is given to the transfer to cattle through soil and plants. Taking the MacKay-model as a starting-point (MacKay et al., 1985), the transfer may be calculated in two ways:

1. The MacKay-model gives a concentration in the soil. Transfer factors (to be discussed) enable an estimate of the concentration in cows, assuming that the absorption by cows does not affect HCH-concentrations in soil in any substantial way.
2. A biota compartment is incorporated in the MacKay-model. Thus, biota represents the population of The Netherlands. This second method requires on investigation into the exact coefficients for transportation and degradation.

In this document only the first method will be discussed. The absorption of HCH, by cows is the result of the consumption of soil, grass or straw and, possibly, of treatments with lindane-containing ointments. The HCH-concentrations in grass and straw from summer wheat are determined by multiplying the HCH-concentration in the soil by a transfer factor. Both concentrations are expressed on the basis of dry substance. The highest concentrations are found in fatty tissues. The concentrations in meat and milk are also determined by means of transfer factors. All factors mentioned have been derived from a paper by the RIKILT (1985). The factor for the transfer from the soil to plants appears to be dependent on the type of soil, the content of organic matter in the soil and the nature of the crop. The transfer factor from plants to meat and milk is dependent on the type of HCH: α-, β- or γ-HCH. Since the highest factor for the transfer from

plants to animals is found for β-HCH and the standard for β-HCH is the most rigid one (RIKILT, 1985), β-HCH could be used for further calculations. However, since β-HCH-concentrations are available from only a number of heavily polluted areas, not to be considered representative for the situation in The Netherlands, and since the emission of β-HCH is only an unkown fraction of γ-HCH, calculations have been based on γ-HCH concentrations, using the transfer factors for β-HCH to estimate transfers to the biota. Although such a procedure does not reflect reality, it will definitely not underestimate the actual load.

Two examples are presented:

- saturated peaty soil of peat with an organic carbon content of 10%;
- dry sandy soil with an organic carbon content of 1%.

These two types of soil may be considered as extremes. The following distribution has been used in calculating the mass percentage of dry substance in the soil:

1 dm^3	0.3 dm^3	density:
soil	water/air	air: appr. 0
		water: appr. 1 $kg.dm^{-3}$
	0.7 dm^3	sandy soil: appr. 2.7 $kg.dm^{-3}$
	solid matter	peaty soil : appr. 0.9 $kg.dm^{-3}$

This leads to the following mass percentages dry substance/total mass:

$$\text{dry sandy soil: } \frac{0.7 \times 2.7}{0.7 \times 2.7 + 0.3 \times 0} = 100\%$$

$$\text{humid peaty soil: } \frac{0.7 \times 0.9}{0.7 \times 0.9 + 0.3 \times 1} = 68\%$$

The transfer factor from soil to grass has been assumed to be 0.05. The factors from grass to meat and milk are assumed to be respectively 15 and 10. The daily consumption of cows is estimated 15 kg of grass (on the basis of dry substance) and an additional 0.5 kg of soil. For sandy soils and peaty soils the densities have been put at respectively 1.89 and 0.93 $kg.l^{-1}$. The degradation of HCH in the soil has been put at 0.1% per day. The results are presented table 3.7.

Table 3.7. Concentrations of HCH (in mg.kg^{-1}) calculated using the MacKay model and the daily amounts of HCH (in mg) absorbed by cows

	Humid peaty soil	*Dry sandy soil*
Concentration in soil:		
- on the basis of humid soil	1.27×10^{-2}	6.16×10^{-3}
- on the basis of dry matter	1.87×10^{-2}	6.16×10^{-3}
Concentration in grass	9.34×10^{-4}	3.08×10^{-5}
Daily absorption	2.34×10^{-2}	7.70×10^{-3}
Maximally tolerable daily absorption	7.5×10^{-2}	7.5×10^{-2}

From table 3.7. it may be concluded that even under the most unfavourable circumstances (humid peaty soil) the standard is not exceeded on a national scale. The actual situation is even better, the national average β-HCH-concentrations in the soil probably being one order of magnitude below the lindane concentrations (see chapter 4). The tolerable level may only be exceeded locally in highly polluted areas.

3.5. MULTICOMPARTMENTAL DISTRIBUTION

The national distribution of HCHs over the air, water, soil and sediment compartments may be estimated on the basis of emission data, using the fugacity model developed by MacKay et al. (1985). As a model for the distribution on a regional level is still under preparation it cannot be applied here. From common model calculations the degradation in soil appears to be the dominant removal mechanism; most of the HCH ends up in soil and water and the degradation rate in water is low compared to that in soil.

3.6. SUMMARY AND CONCLUSIONS

With respect to the large-scale distribution of HCH over the environmental compartments, model calculations indicate that most of the HCH emissions enter the soil compartment and that the degradation in soil is the main removal mechanism by which HCHs are eliminated from the environment. Reference is made to table 2.7. for an estimate of the distribution in the environment at the various applications: the major part of the HCH will be found on and in the soil, with the possible exception of the relatively limited emission during leaf applications, parasite control in cattle,

domestic use and insect control in buildings. However, the degradation in soil progresses slowly and results mainly in metabolites that are rather persistent themselves.

Generally, adsorption to soil is rather high. The probability that HCHs reach the groundwater is slight; less than 1% of the soil lood reaches the groundwater. Only in cases of local soil pollution as a result of dumping with HCH concentrations at wich biodegradation is inhibited, may wash-out lead to high concentration levels in the groundwater. The highest probability is in sandy soils deficient in humus. Considering its persistence, the accumulation of lindane in the soil will amount to maximally 1/3 to 1 of the annual dose. However, measured data on actual concentrations in agricultural soils in The Netherlands are lacking.

HCH hardly bioaccumulates in soil organisms. In plants lindane is mainly absorbed by the roots, the highest concentration levels also being found here after equilibrium. On the basis of model calculations on the transfer of HCH from soil and vegetation to cattle it may be concluded that the standard currently in force is not exceeded, with the possible exception of highly polluted localities as a result of dumpings.

In surface water adsorption to solid particles and evaporation play an important part in determining the behaviour of HCHs. Both the biotic and abiotic transformation are limited. In water organisms bioaccumulation occurs (a factor of 100-1000), at which equilibrium is rapidly reached (a few days to two weeks). Biomagnification is insignificant.

During spraying of vegetation shortlasting emissions into the air occur. After treatment the concentrations decrease rapidly as a result of transport; data on degradation are lacking. The dry deposition is estimated at 0.5-1.5 tons per year (however, locally this depostion could be negative); the wet deposition at appr. 2 tons per year.

4. CONCENTRATIONS IN THE ENVIRONMENT AND EXPOSURE LEVELS

4.1. BACKGROUND CONCENTRATIONS

No natural sources of HCHs are known. All concentrations measured, including those in background areas, are ascribed to the production and use of lindane. Other isomers may occur as manufacturing pollutants or they may be formed by photo-isomerization after applications with lindane. The concentration levels in background areas depend on the distance from areas where lindane is used. Areas may be considered background areas when they are far from agricultural areas (oceans, deserts, polar regions). Data on the concentrations in these areas are summarized in table 4.1. (air) and table 4.2. (sea water). Besides the level of ϵ-HCH mentioned in table 4.2., Tanabe et al. (1982) found the following concentration ranges and averages for respectively α-, β- and γ-HCH in the West Pacific, Indian and Antarctic Ocean in the period of November 1980 to March 1981 : 0.02-3.4 (0.5), 0.005-1.0 (0.15) and 0.16-3.7 (1.0). The concentrations appear to be related to the location and are highest in the Indonesian Archipelago. The α/γ ratio increases considerably above 20^0 SL. The lowest concentrations ($\leq$ 0.1 ng.m^{-3}) are found in the South Pole (Tanabe et al., 1983). In comparison, the concentrations at some locations in the NW Pacific and the East Arabic Sea are high (Tanabe and Tatsukawa, 1980) and therefore should no longer be considered as background concentrations.

The following may be observed with respect to the measurements. The results of Atlas and Giam (1981) and Bidleman and Leonard (1982) are based on samples taken in accordance with the Bidleman and Olney method (reference given in ch. 6). According to Bidleman et al. (see ch. 6) this method is not applicable to HCH. As a result the concentrations measured are far too low (a factor 5 for samples > 600 m^3), and they should therefore be regarded as minimum values.

Table 4.1. Concentrations of HCH-isomers in outdoor air (in ng.m^{-3}) at various locations in the world (mostly background areas)

Area	Sampling rate ($m^3.min^{-1}$)	Sampling volume (m^3)	N α/γ	α-HCH conc.range(average) ($ng.m^{-3}$)	γ-HCH conc.range(average) ($ng.m^{-3}$)	α+γ HCH conc.range(aver.) ($ng.m^{-3}$)	References
Enewatak atol (North-Pacific)	0.5 0.03	1,000-2,000 75-150	9/17	0.075- 0.57 (0.25)	0.006- 0.02 (0.015)		Atlas and Gias (1981)
Arabic Sea,	0.35-0.5	600	10	0.03 - 0.92 (0.28)	0.02 - 0.22 (0.07)		Bidleman and
Persian Gulf, Red Sea			11	0.09 - 1.8	0.01 - 0.67		Leonard (1982)
Lillestrum	0.67	1,000-2,000	4	0.35 - 1,23 (0.875)	0.15 - 0.41 (0.32)		Oehme and Stray (1982)
Bearisland	0.67	1,000-2,000	5	0.22 - 0.52 (0.37)	0.015- 0.04 (0.03)		Oehne and
Bearisland	0.67	1,000-2,000	5/4	0.185- 0.27 (0.22)	0.01 - 0.02 (0.01)		Manu (1984)
Spitsbergen	0.67	1,000-2,000	4	0.71 - 1.29 (0.92)	0.025- 0.05 (0.04)		
Spitsbergen	0.67	1,000-2,000	5	0.16 - 0.27 (0.22)	0.01 - 0.02 (0.01)		
N.W. Pacific	0.007	10-20	10			0.4 -13.8 (6.0)	Tanabe and
N.W. Pacific	0.007	10-20	4			1.8 - 3.8 (2.7)	Tatsukawa
N.W. Pacific	0.007	10-20	4			1.0 - 6.0 (2.7)	(1980)
N.W. Pacific	0.007	10-20	14			0.4 - 1.8 (1.0)	
N.W. Pacific	0.007	10-20	7			1.0 - 7.6 (3.7)	
N.W. Pacific and Bering Sea	0.007	10-20	10			0.5 - 7.2 (1.1)	
N. Pacific	0.007	10-20	21			0.2 - 1.2 (0.6)	
E. China Sea	0.007	10-20	3			0.2 - 1.2 (0.7)	
S. China Sea	0.007	10-20	10			0.2 - 1.5 (0.4)	
Bay of Bengal	0.007	10-20	6			0.1 - 1.0 (0.4)	
Arabic Sea	0.007	10-20	8			0.2 - 3.3 (1.3)	
E. Arabic Sea	0.007	10-20	4			2.0 -10.0 (6.9)	
W.Pacific, Ind. Ocean, Antartic Ocean	0.026	1,000	27	0.02 - 0.66 (0.26)	0.03 - 0.68 (0.21)	0.0- 0.13*(0.003) (*β-HCH)	Tanabe et al (1982)
Syowa Station, The Antartic	0.026	1,000	13			0.04- 0.13 (0.09)	Tanabe et al (1983)
The Antartic, 4 stations	0.026	1,000	4			0.04- 0.17 (0.10)	

4.2. OCCURRENCE IN SOIL

4.2.1. Nature reserves

In The Netherlands Edelman (1984) analysed the upper 10 cm of the soil from 38 nature reserves for α- and γ-HCH. The distribution of the contents is given in table 4.3.

Table 4.2. Concentration of all HCH-isomers (Σ-HCH) (in $ng.l^{-1}$) in sea water (Tanabe et al. 1980, 1983)

Area	Period	Number of samples	Concentration Σ-HCH range	Average concentration
NW Pacific	July/Aug. 1976	7	4.9-22.6	10.6
NW Pacific	Jan/Feb. 1978	6	0.8- 1.9	1.3
NW Pacific	June/July 1979	3	3.7- 5.5	4.3
NW Pacific	July 1979	5	6.2-14.2	9.1
Bering Sea	July 1979	7	3.2- 4.4	3.9
E. and S. China Sea	January 1977	3	1.3- 3.4	2.2
Bay of Bengal	October 1976	3	0.7- 1.9	1.2
Arabic Sea	Nov./Dec. 1976	3	0.2- 1.3	1.0
the Antartic (under "fast ice")	July/Oct. 1981 January 1982	3	0.2- 0.6	0.45
(under packice border)	Jan./Febr. 1981	3	0.3- 0.9	0.5

Table 4.3. Frequency table of the α- and γ-HCH contents (in $\mu g.kg^{-1}$ of soil) of 96 soil samples taken from 38 natural reserves (Edelman, 1984)

Concentration range	<1	1-10	10-20	20-40	40-60	60-80
α-HCH	94	0	0	1	1	0
γ-HCH	59	21	9	5	0	2

Relatively high levels for α-HCH were found in the "Lettelberter Petten" (Groningen) and in "Wormer- and Jisperveld" (North Holland); for lindane in the "Lettelbertel Petten" and in the "Korenburgerveen" (Achterhoek). In

view of diffuse pollution levels in samples from nature reserves Edelman (1984) argues that these should no longer be considered as background levels.

4.2.2. Agricultural areas

Measuring data on HCH contents in agricultural areas are not available. However, as indicated in sections 3.1.2. and 3.1.3., the concentrations in groundwater and the topsoil may be estimated. In case of a sandy soil deficient in humus and a half-life of 500 days it was estimated that appr. 1/160 % of the dose will wash out. When maize is cultivated on this type of soil and lindane is applied every year for soil treatment (dose 612-735 $g.ha^{-1}$), the concentrations in the upper groundwater below these parcels will amount to appr. 15 $ng.l^{-1}$ under stationary conditions. As it may be assumed that in practice the wash-out is less, the concentration in the groundwater should be less than 15 $ng.l^{-1}$, not having taken into consideration the higher dosage levels applied in the past. In soils where the wash-out is negligible (humous soils) the accumulation leads to a basic level of the lindane residue of maximally 1/3 to 1 of the annual average dose. In case of the dose for maize mentioned above, at which the compound is assumed to be homogeneously mixed with the soil in the plough furrows (top 20 cm), a basic content of lindane is attained of 80-250 $\mu g.kg^{-1}$ (present every year one year after dosing).

4.2.3. Dumping grounds

In The Netherlands the soil is polluted with HCH at various locations, the cause usually being the production of HCH in the fifties. The concentrations found range to a few thousand mg HCH per kg of dry soil. Two areas where soil pollution with HCH is highest are Twente, with many industrial sites within a large area, and the DAGRA premises in Bunschoten.

- Twente

The Department of Public Works of the province of Overijssel (1985) states that the presence of HCH in the soil in the municipalities of Hengelo and Enschede is caused by both the dumping of chemical waste (the filling up of verge ditches, the levelling up of areas with "black soil" originating from the premises of C.T. Stork Chemical Industries Ltd., diggings which have

been filled with industrial waste, the dumping ground of the same concern) and the dispersal of chemical waste from dumping grounds and chimney losses at the chemical industry. The source of these pollutions is the former C.T. Stork Chemical Industries Ltd.

In the beginning of the eighties 152 locations were identified in this region for further studies on HCH. A rough survey in each location indicated that many parcels were polluted with HCH. In a small number of parcels very high HCH concentrations were found up to a depth of more than 220 cm below ground level, mostly α- and β-isomers. It is likely that at some places HCH will be present at even greater depth, up to the ground water. This could also be deduced from HCH concentrations measured in the "Twentekanaal". Pollution of the area bordering on the canal has a distinct bearing on the water quality of the canal, as followed from the decrease in HCH concentration after the HCH waste was removed from this area. Apart soil, at one parcel wash-out also contained HCH. Besides HCH, other chlorinated hydrocarbons and heavy metals were found in the soil.

The DAGRA-premises

On the premises of the former DAGRA company at Bunschoten, where flea-powder was produced in the years 1945-1949 and formulated in 1949-1952, soil, mud in ditches and groundwater are highly polluted with HCH. The HCHs must have reached the soil by the burying of rejected products in clay holes in the area, through the air by dispersal from an open storage depot and by spilling during production, storage and handling.

Table 4.4. shows the concentrations of three HCH-isomers found in soil, silt, groundwater and surface water. It should be stressed that this table shows the highest concentrations found.

Table 4.4. Highest HCH concentrations found in the DAGRA area (RAUW, 1984)

Pollution	*Soil*	*Silt*	*Groundwater*	*Surface water*
	(in mg.kg^{-1})		*(in µg.l^{-1})*	
α-HCH	*900,000*	*240*	*1.3*	*2*
β-HCH	*40,000*	*6*	*0.002*	*0.3*
γ-HCH	*55,000*	*60*	*0.56*	*0.2*

In particular the α-isomer was detected in high concentrations locally; β- and γ-HCH occurred in relatively lower concentrations, in most cases at the same locations and in a more or less fixed ratio to α-HCH. Pollution was also found at a depth of 100-180 cm below ground level.
Pollution of groundwater is restricted to the vicinity of the former company building; horizontal transportation appeared to be limited.
In this case numerous of other chlorinated hydrocarbons were found as well.

4.2.4. Sewage sludge

Water quality administrations periodically monitor the occurrence of pesticides in sewage sludge. In table 4.5., average concentrations and highest and lowest values found are given for each area.

Table 4.5. HCH contents in sewage sludge in 1981 per management area (in μg per kg d.m.; Fieggen, 1981)

Area	*α-HCH*			*β-HCH*			*γ-HCH*		
	min.	*aver.*	*max.*	*min.*	*aver.*	*max.*	*min.*	*aver.*	*max.*
Province of Utrecht	-	5	10	2	40	1100	1	10	55
Province of Groningen	-	5	7	1	50	90	2	10	13
Regge and Dinkel	2	7	15	21	30	85	2	8	10
Province of Zeeland	-	-	-	-	-	-	-	15	-
Veluwe	1	7	15	3	40	85	1	40	85
East-Brabant	3	5	7	-	-	-	1	40	100
West-Brabant	2	10	30	3	30	275	1	30	200
Riverine District	1	7	35	3	40	75	2	10	46
Eastern Gelderland	3	10	30	-	-	-	3	20	80
Province of Limburg	1	8	10	-	-	-	1	10	40
Hollands Isl. & Holmes	1	8	10	1	50	150	1	20	210
Kennemerl.& W.Friesland	10	70	190	120	150	160	10	40	95
City of Amsterdam	-	-	-	-	-	-	10	40	70
Amstel & Gooiland	5	50	70	-	-	-	20	50	100
Delfland	-	10	-	-	-	-	10	20	35
Province of Drenthe	5	30	80	-	-	-	10	30	1600
Province of Friesland	6	20	40	-	-	-	20	30	25
West-Overijssel	1	5	7	-	-	-	2	10	100

The incidental occurrence of high concentration levels of the α- and β-isomers is striking. In one case this phenomenon was related to a discharge of polluted groundwater in the measuring period; usually the cause is not

known. Furthermore, β-HCH is incidentally measured at concentrations surpassing the levels for both other isomers (β-HCH is generally less biodegradable than the other isomers; see chapter 3). It is concluded that the number of localities with low concentrations is increasing. However, the problem of incidentally very high levels remains unabated. Incidentally high levels also occur among the limited preliminary figures for 1985.
Fieggen (1981) compared the results of the measurements carried out in 1975, 1978 and 1981. The results of these comparisons are presented in table 4.6. for lindane.

Table 4.6. Comparison of the contents of lindane in sewage sludge in 1975/1976, 1978 and 1981 (in μg per kg d.m.)

	1975/1976	*1978*	*1981*
Number of samples	*143*	*202*	*386*
Median value	*10*	*20*	*< 1*
Mean value	*25*	*43*	*12*
95-Percentile	*100*	*180*	*40*
Maximum value	*440*	*780*	*1600*

4.3. OCCURRENCE IN SURFACE WATER

4.3.1. State-managed waters

The Department of Public Works has since 1973 frequently determined the α- and γ-HCH levels in the rivers Rhine, Meuse and West-Scheldt (Dijkzeul, 1982a, 1982b; Van der Kooij, 1982; Rijkswaterstaat, RIVM, 1985). In figure 4.1. the results from the sampling stations Lobith, Eijsden and Schaar van Ouden Doel are given as annual averages. As to the Rhine, a sharp decline is apparent until 1975. After that, the load with lindane has remained more or less stable, whereas the contents of α-HCH have decreased further. In the Meuse and the West-Scheldt the levels of lindane show a more capricious course, whereas the α-HCH levels are low in these rivers as well (average value $< 0.01\ \mu g.l^{-1}$).
Since 1983 the contents of β-HCH are also being determined at the sampling stations. Generally the levels found are $< 0.001\ \mu g.l^{-1}$. The concentrations at the frontier are usually higher than those at locations situated further

downstream. At Lobith as well as at Eijsden and Schaar van Ouden Doel the basic quality standard (see chapter 1) is often exceeded.

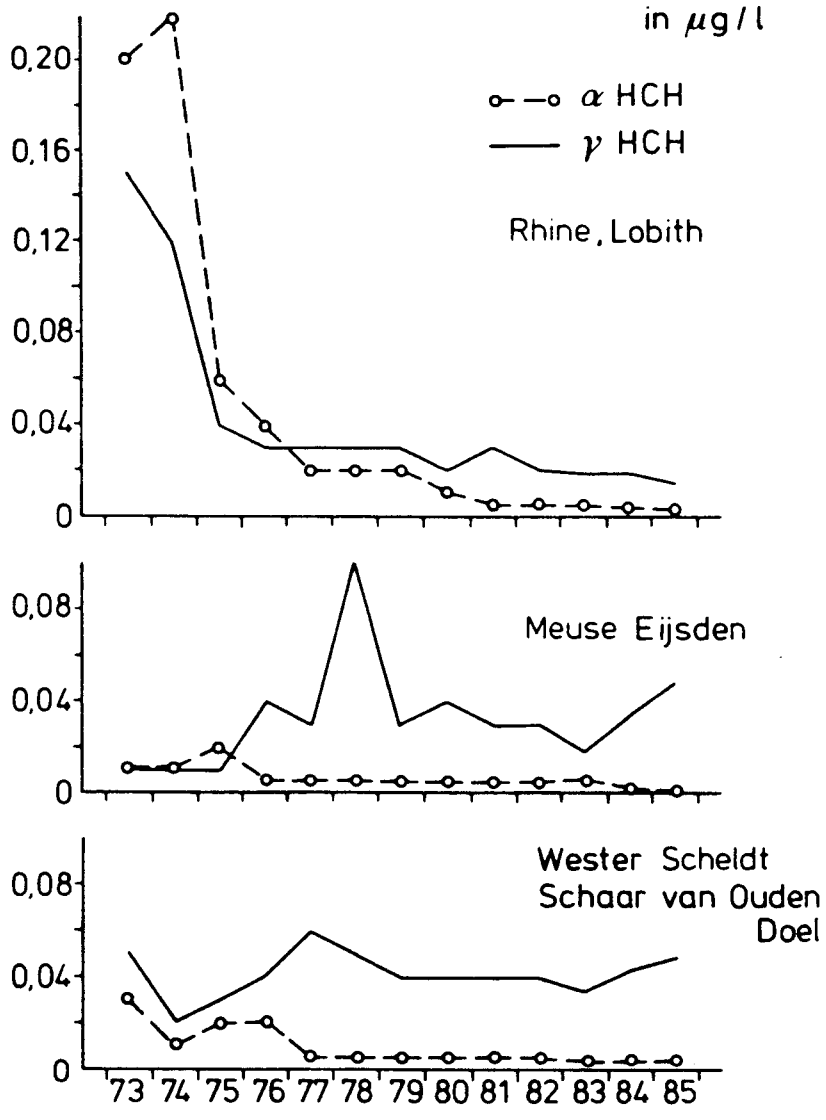

Figure 4.1. Concentrations of α- and γ-HCH in the rivers Rhine, Meuse and West-Scheldt (annual mean values, $\mu g.l^{-1}$)

4.3.2. Non state-managed waters

Systematic measurements of organic micropollutants over a period of several years are only available for a limited number of areas (Feenstra and Van der Most, 1985). For a few years, the RIVM has analysed smaller surface waters in agricultural areas several times per year. The concentrations of α-HCH were usually below 0.03 $\mu g.l^{-1}$. For the greater part this applies equally to the γ-HCH concentrations. However, for this compound a number of peak levels between 0.1 and 0.47 $\mu g.l^{-1}$ were found as well (Wammes et al., 1983, 1985, 1986).

The data mentioned below are mainly derived from measurements in "Rijnland" and in the area of Hollands Islands and Holmes, complemented with results from some incidental measurements from other areas. It may be stated that the α-HCH concentrations are usually between 0.001 and 0.01 $\mu g.l^{-1}$ with peak levels of up to 0.002-0.03 $\mu g.l^{-1}$. The number of such peaks, probably the result of injudicious uses of HCH, tends to decrease.

The non-systematic measurements are often carried out at locations at which higher concentration levels might occur as a result of specific applications (greenhouse horticulture) or the presence of dumping grounds. Hence, in these cases the concentrations found are often rather high (sometimes 16 $\mu g.l^{-1}$ in Delfland), in the polder-waters and reservoir systems in Rijnland α-HCH levels often exceeding 0.01 $\mu g.l^{-1}$. In the Twente Canal the α-, β- and γ-isomers have been found incidentally at levels of 0.1-1.0 $\mu g.l^{-1}$, probably related to the HCH-containing dumping grounds in this area (Laseur, 1985).

4.3.3. Underwater soils

The data on underwater soils are chiefly based on inventorial studies on state-managed waters by the Laboratory for Watercourses (Kerkdijk, 1985, 1986). A summary of the results for the most important waters are presented in table 4.7.

Table 4.7. Median values of HCH concentrations in underwater soil in state-managed waters in $\mu g.kg^{-1}$ d.m. (Kerkdijk, 1985, 1986)

No.	Water	Median Values		
		α-HCH	β-HCH	γ-HCH
1	Rhine and its branches	10	26	15
2	Meuse and its branches	4	6	18
3	Harbours of Rotterdam	213	214	342
4	New Meuse, Old Meuse, New Waterway	*	*	245
5	Lake IJssel and its border lakes	10	9	35
6	Hollandse IJssel	54	*	35
7	North Sea Canal, North-Holland Canal, Gent-Terneuzen Canal	*	*	*
8	West-Scheldt	*	*	240

Remarks:
1,2,4 : Qualitative values
3 : Especially "Dokhaven", "First Petroleumhaven" and "Chemiehaven"
6 : Samples especially taken in the vicinity of dumping grounds
7 : Elevated concentrations in these sediments might be expected

Furthermore, elevated concentration levels were found at some locations in the "Zuid-Willemsvaart", near the sluices of "Haringvliet" and at a few dock yards. However, as the measuring programme carried out has been rather too limited it is not possible to draw definite conclusions.
For the non state-managed waters it may be assumed that the contents in waters mainly fed by the Rhine are in the same order of magnitude as those in the Rhine. Locally very high concentration levels may occur resulting from injudicious use or from wash-outs from dumping grounds.

4.3.4. Estuaries and seawater

- Meuse-Rhine estuary

Duinker and Hillebrand (1979) studied the behaviour of a number of organochlorine micropollutants in the Meuse-Rhine estuary. In table 4.8. and in figure 4.2. the results are given of the analyses of samples taken in December 1974. These results show that at least part of the HCH-isomers is present in solution and that the concentrations in coastal waters are lower than those in rivers, being in the range of some nanograms per litre (HCH passing a Whatman GF/C-filter was considered to be dissolved).

Table 4.8. Concentrations of HCH-isomers in solution and in suspension in the Meuse-Rhine estuary (Duinker and Hillebrand, 1979)

HCH-isomer	Dissolved		Suspended	
	average ($ng.l^{-1}$)	range ($ng.l^{-1}$)	range ($ng.l^{-1}$)	range ($ng.g^{-1}$)
α-HCH	20	12-25	0- 6	<20-100
β-HCH	6	2-10	<1- 3	< 6- 75
γ-HCH	20	10-30	1-20	<20-2000

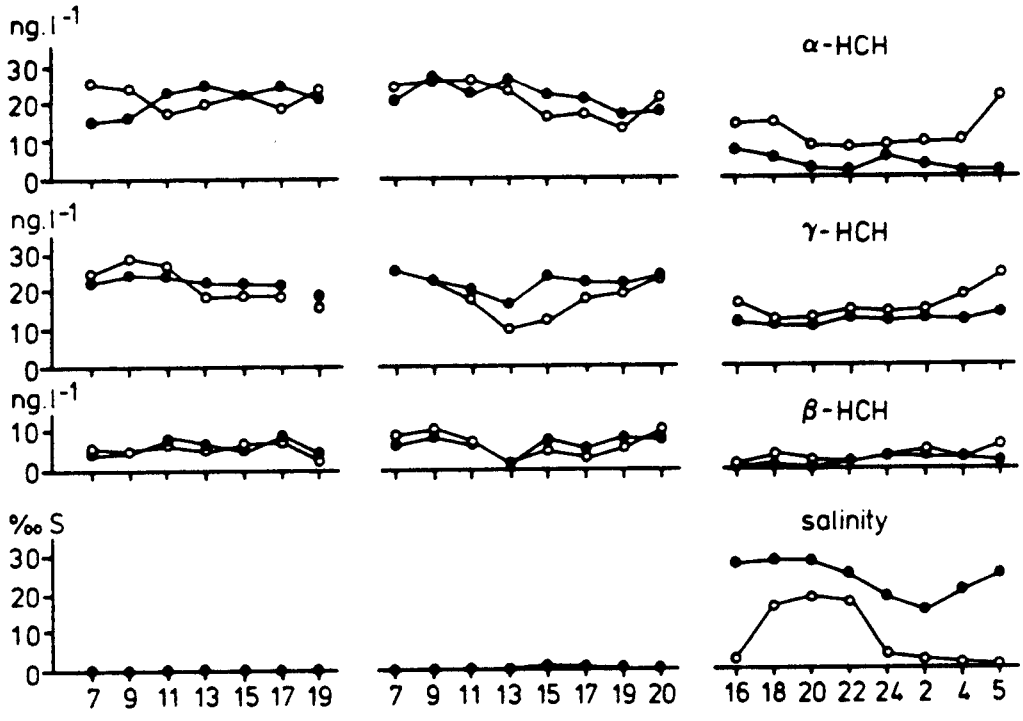

Fig. 4.2. Concentrations of HCH-isomers in solution and salinities (o/oo) at three locations, at the surface (o) and at the bottom (•). Samples were taken in December 1974 (Duinker and Hillebrand, 1979)

- Wadden Sea and the mouth of the Eems

The measuring results of a similar study at two locations in the mouth of the Eems in the summer of 1967 (Duinker et al., 1985) are presented in figure 4.3. The major part of the HCH appears to be dissolved.

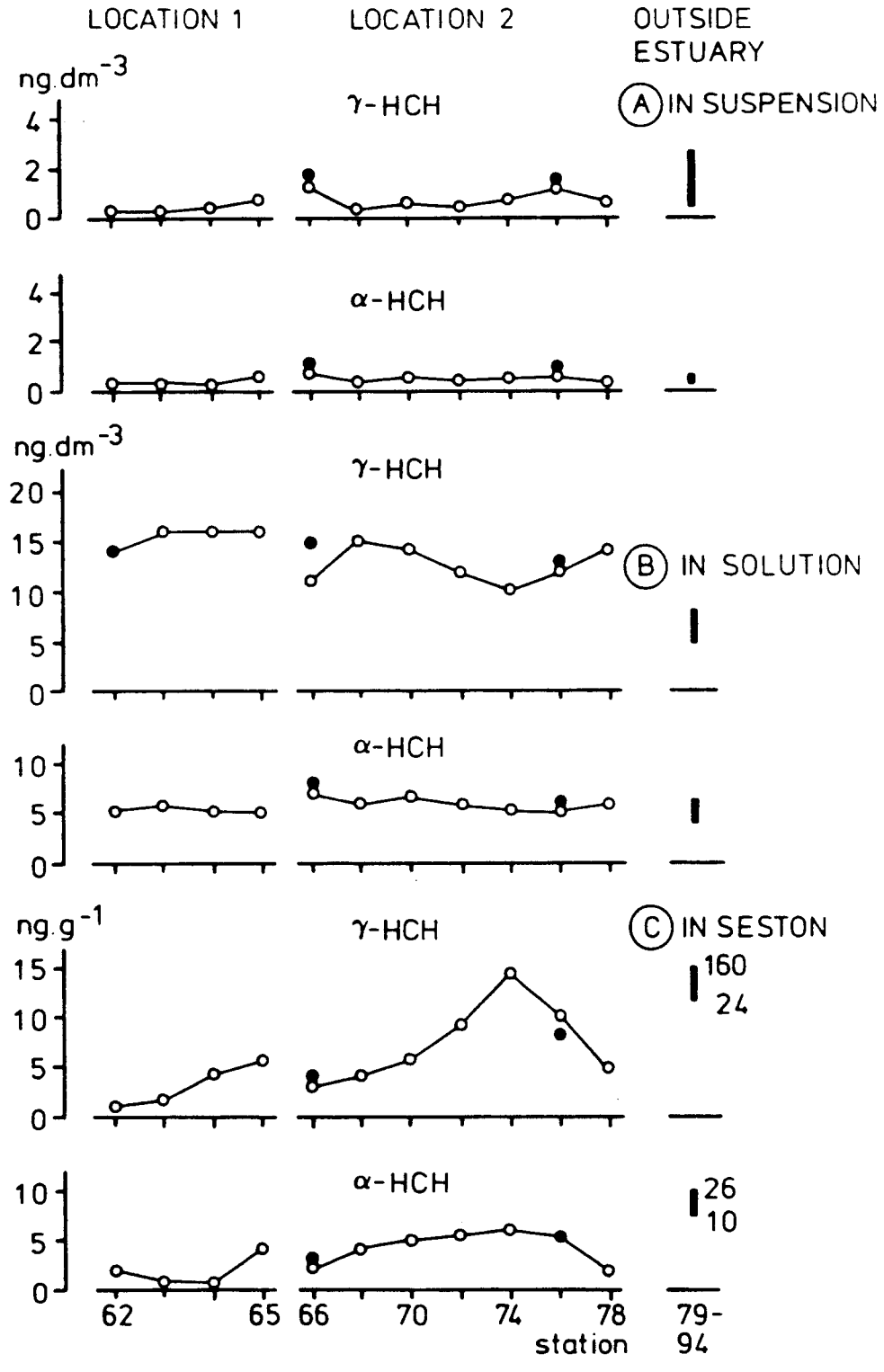

Fig. 4.3. HCH-concentrations in suspension, in solution and in the seston in the mouth of the Eems (Duinker et al., 1985)

According to investigations carried out by the RIZA (1984) in 1982 in the Wadden Sea and the Eems-Dollard estuary, lindane was found in the underwater soil at nearly all locations, while the contents increased from the western part of the Wadden Sea to the Eems-Dollard estuary. At the foot of the dike of the "Balgzand" an elevated level was found, whereas at some distance from the dike no lindane was demonstrated.
In contrast with lindane, β-HCH is present in detectable levels only in the Wadden Sea, the highest contents being measured at the Frisian Flats (0.15-0.55 $\mu g.kg^{-1}$ d.m.).

- Weser and Elbe

In the same way Duinker et al. (1982) studied the estuaries of the Weser and the Elbe, the results being comparable to those obtained for the Rhine and the Eems.

- Coastal water in The Netherlands

In 1981 some measurements were carried out (RIZA, 1983), as presented in table 4.9.

Table 4.9. HCH contents in coastal waters in The Netherlands in 1981, dissolved (D: in $ng.l^{-1}$) and bound to particles (P: in $ng.g^{-1}$)

HCH-isomer	*Flushing*		*10 km w of Walcheren*		*70 km w of Noordwijk*		*Marsdiep*		*70 km nnw of Rotterdam*	
	D	*P*	*D*	*P*	*D*	*P*	*D*	*P*	*D*	*P*
α-HCH	*1.0*	*5.3*	*1.0*	-	*0.9*	-	*1.6*	-	*1.1*	-
γ-HCH	*4.6*	*8.7*	*3.2*	*3.9*	*0.9*	-	*3.1*	-	*1.0*	*11.4*

- = not demonstrated

4.4. OCCURRENCE IN AIR

4.4.1. Indoor air

- Occupational exposure

The most important application of HCH in The Netherlands is the control of soil and leaf insects. Dependent on the spraying method a higher or lower

level of occupational exposure to HCH will take place. This relates to both indoor and outdoor air concentrations.

Baunok (1984) studied the course of the lindane concentration over time on a lawn treated with lindane by hand spraying. At a height of 50 cm above ground level, the initial concentration amounted to appr. 5 $\mu g.m^{-3}$. In the field Opekar et al. (1974) found that the average concentrations varied from 0 to 4 $\mu g.m^{-3}$. Similar values have been found during the preparation of lindane emulsions and the subsequent spraying of conifers (Kolmodin-Hedman and Swensson, 1979). According to these authors the time-weighted average concentration (TWA) during digging up activities amounts to 5-180 $\mu g.m^{-3}$ 7 days after spraying; the TWA decreased to 40 $\mu g.m^{-3}$ maximally after 11 days.

Treatment with HCH may prevent the sowing-seed from being affected. Grey et al. (1983) examined two seed companies at which sowing-seed was pretreated with a lindane-maneb powder. Assuming a respiration rate of 1.2 m^3 per hour the TWA amounted to 300 and 450 $\mu g.m^{-3}$ for the first and second company, respectively.

The formulation and the packaging of HCH-containing pesticide powders and liquids may often lead to exposure of the employees. Based on calculations Elguindi and Rajhans (1979) mention concentrations of 450-1,300 $\mu g.m^{-3}$.

All above-mentioned data are from foreign studies; data for the situation in The Netherlands are not available.

- Non-occupational exposure

In six homes in The Netherlands built on former dumping grounds the average lindane concentration was 0.006 $\mu g.m^{-3}$ (0.001-0.014 $\mu g.m^{-3}$), whereas in the space beneath the floor lindane was below the detection limit (< 0.001 $\mu g.m^{-3}$ in this case). During this study the outdoor air concentration amounted to 0.0003-0.0004 $\mu g.m^{-3}$, which may be considered normal for The Netherlands (see section 4.4.2.). In a similar study, also carried out in homes situated on former dumping grounds, the outdoor air concentration levels as well as the air concentrations in the space beneath the floor were elevated; the highest indoor air concentration observed amounted to 0.09 $\mu g.m^{-3}$ (Thijsse, 1986).

A steep concentration increase occurs in homes when large surfaces have been treated with lindane-containing products to control wood-worms or long-horned beetles. It is evident that the maximum concentration is reached in 4 weeks, after which the concentration gradually decreases. In

two spaces that were investigated the peak levels amounted to 61 and 51 $\mu g.m^{-3}$ and after 10 weeks the concentrations decreased to 24 and 8 $\mu g.m^{-3}$, respectively (Dobbs and Williams, 1983). In a simulation experiment Dobbs et al. (1979) also found an initial increase of the lindane concentration. Comparable results were obtained by the Food-Inspection Departments in The Netherlands: after indoor application of lindane for wood preservation concentration levels of appr. 50 $\mu g.m^{-3}$ are commom (with peak levels of more than 100 $\mu g.m^{-3}$) and persist for weeks or months. Hence, after a single treatment the lindane concentrations in the home remain elevated during a very long period. Five months after treatment of a cellar, Zimmerli and Zimmermann (1979) found a concentration of 6 $\mu g.m^{-3}$ (4-10 $\mu g.m^{-3}$). Dobbs and Williams (1983) reported lindane concentrations in homes for a period of 1-10 years after application. For attics the average concentration for this period was 2.5 $\mu g.m^{-3}$ (0.08-11 $\mu g.m^{-3}$), for the other spaces appr. 0.6 $\mu g.m^{-3}$, with a range of 0.01-2.9 $\mu g.m^{-3}$. Treatment of only one space in a home results in elevated concentrations in the other, non-treated spaces. Dependent on the location of these spaces in relation to the treated space, the concentrations vary from 0.01 to 0.61 $\mu g.m^{-3}$ (Dobbs and Williams, 1983).
There are various HCH-containing products on the market for the control of insects in and around the home as well as for the control of parasites in domestic animals. The influence of these products on the indoor air concentrations is not known.

4.4.2. Outdoor air

In table 4.10. the data on the concentrations measured above the continent are given. The results of the measurements by Diederen et al. (1981) (published by Guicherit and Schulting, 1985) are included as well. The information on the HCH concentrations in The Netherlands is limited to two series of measurements. The first was carried out in Delft, already broadly outlined in table 4.10. and, per season, in table 4.11. Furthermore, in the framework of investigations into the background concentrations of trace elements and compounds (Thijsse and Huygen, 1985), measurements over a period of 3 days were analyzed. According to this study the respective general pollution levels during these days were low, average and high. The results for HCH are presented in table 4.12. Based on these data the

average concentration level of HCH in The Netherlands is estimated at appr. 0.5 $ng.m^{-3}$.

Table 4.10. Average concentrations of HCH-isomers (in $ng.m^{-3}$) at several locations on the continent of Europe and in the United States of America

Area	Period	Sampling discharge ($m^3.min^{-1}$)	volume (m^3)	α-HCH	γ-HCH	α+γ-HCH	Ref.
Columbia	Apr. 1980	0.35-0.5	1400			0.75	1
	Oct. 1980	0.35-0.5	600			1.5	1
Denver	Jan. 1980	0.35-0.5	750			0.25	1
			760			0.2	1
			1460			0.4	1
New Bedford	June 1980	0.35-0.5	360-760			1.0	1
Berlin	1970/71	1.33	2400	0.28 (0.02-1.80)	0.14 (0.01-1.35)		2
Ruhr area	1976/77	?	?	2.1 (0.15-7.4)	0.77 (0.10-3.7)		3
Delft	1979/80	0.07	100	0.25 (max. 1.2)	0.36 (max. 3.4)		4

1) Billings and Bidleman (1983): The concentrations measured for samples larger than appr. 750 m^3 are probably 10-20% too low.
2) Herzel and Lahmann (1972): As only glass fibre filters are used, the measured concentrations are too low (also according to the authors): possibly a factor 50
3) Selenka and Eckrich (1983): Reference is made to 1)
4) Guicherit and Schulting (1985): The fact that absorption resin was not used may have resulted in a 20% loss by break-throughs (Burghardt, pers. comm.)

Table 4.11. Concentrations of α- and γ-HCH (in $ng.m^{-3}$) in Delft (Diederen et al., 1981); β- and δ-HCH levels were below the detection limit (2-3 $pg.m^{-3}$)

Season	Number of samples	α-HCH average (stand. dev.)	γ-HCH average (stand. dev.)
Summer	29	0.29 (0.30)	0.46 (0.69)
Winter	26	0.20 (0.11)	0.25 (0.34)
Whole year	55	0.25 (0.23)	0.36 (0.56)

Table 4.12. Concentrations of lindane (in ng.m^{-3}) at three locations in The Netherlands (Thijsse and Huygen, 1985)

Location	*General level of air pollution*		
	Low	*Average*	*High*
Witteveen (Dr)	*0.01*	*0.2*	*0.7*
Bilthoven	*0.01*	*0.9*	*0.4*
Vlaardingen	*0.06*	*0.4*	*0.9*

In 1974-1982 rainwater (De Bilt) contained <10-340 ng.l^{-1} (Hofstee, pers. comm.).

4.5. OCCURRENCE IN FOOD AND DRINKING WATER

Several reviews are available on the occurrence of HCH in foodstuffs in The Netherlands (De Vos et al., 1984; Staarink and Hakkenbrak, 1984; CCRX, 1983). In table 4.13. a selection is given.

In 201 24-hr food intakes 1,2 and 3 μg (median values) was found for α-, β- and γ-HCH, respectively. In each food intake the contents varied from 0-17, 0-65 and 0-16 μg per day, respectively (Staarink and Hakkenbrak, 1982). De Vos et al. (1984) analyzed the pesticide content of 12 groups of such as cereal products, potatoes, vegetables and fruit, oil and fats, meat, fish and dairy products, in addition to beverages and drinking water. For α-, β- and γ-HCH the average daily intake for each of these groups appeared to be less than 1 μg per person . For the total diet the mean (and maximum) intakes amounted to: 1 (4), <1 (9) and 2 (5) μg per day, respectively. According to De Vos et al. (1984) the figures for total diets are slightly lower than those found in a few other countries. Recent studies (Greve et al., 1987) over the years 1984 and 1985 resulted in median values lower than 1 μg.kg^{-1} for the three isomers, the maximum levels being 1, 2 and 4 μg.kg^{-1}, respectively.

The main source of HCH in food is animal fat. However, examination of the fat from poultry, pork and beef revealed that only a low percentage of the samples contained more than 50 μg γ-HCH.kg^{-1}; 60-90% less than 10 μg.kg^{-1} (CCRX, 1983). In table 4.14. more recent data are presented on the maximum contents in animal fat and eggs (RIKILT, 1986).

Table 4.13. HCH contents in foods (median and maximum values); fat-containing products are based on the fat fraction (in $\mu g.kg^{-1}$)

Product	α-HCH		β-HCH		γ-HCH	
	median	max.	median	max.	median	max.
Wheat, rye, barley, rice		10-50				10-20
Spices, herbs		1500		600		360
Tea, cocoa						100-200
Margarine	20	60			30	130
Milk	<20	90	<20	200	<20	850
Cheese	20	150		70	20	160
Pork	<10	70			<10	30
Beef	<10	70		<20	<10	80
Meat-products	<10	30	<10	90	20	500
Poultry	<10		<20		<10	710
Eggs	<10	<20	<20	230	<10	400
Eel	10	50	<10	20	20	640
Haddock liver	90	1000	40	60	30	60
Mackerel					10	30
Shrimps		10		10		10

Table 4.14. Highest HCH contents (in $\mu g.kg^{-1}$) found in animal fat and eggs (RIKILT, 1986)

Product	Number of determinations	α-HCH	β-HCH	γ-HCH
Fat from beef	47	10	<20	1700
Fat from pork	49	<10	<20	3200
Fat from veal	47	<10	<20	790
Fat from sheep	24	<10	<20	160
Fat from poultry	29	110	80	60
Eggs	33	40	190	20

For all products mentioned the respective median contents were <10, <20 and 10 $\mu g.kg^{-1}$. In table 4.15. the frequency distribution of the γ-HCH contents in the 229 samples examined is given.

Table 4.15. Frequency distribution of γ-HCH contents of animal fat and eggs (calculated from RIKILT, 1986)

γ-HCH content ($\mu g.kg^{-1}$)	Number	Percentage
<10	195	85
10-100	27	12
100-1000	5	2
>1000	2	1

The maximum acceptable concentration of γ-HCH (2 $mg.kg^{-1}$) was exceeded only once, in a sample of pork fat.
According to investigations into organochlorine pesticide contamination of milk and dairy products, carried out in 1985 by the institutes for dairy produce control, these products contained 20-30 μg of total HCH per kg (per kg fat), of which appr. 5 μg lindane per kg.
In studies on fish products from various fishing-grounds (RIKILT, 1985), <1-5 μg α-HCH; <2 μg β-HCH and <1-8 μg γ-HCH per kg of product was found (n=10).
CCRX (1983) gives no information on the occurrence of HCH in drinking water. The present document only refers to the total dietary studies by De Vos et al. (1984) mentioned above, in which the contribution of drinking water has also been taken into account. In a review of drinking water studies in the USA the highest concentration of lindane found was 0.1 $\mu g.l^{-1}$ (IARC, 1979).

4.6. EXPOSURE LEVELS

Generally, the contribution from food is the most important and the other exposure routes are considered negligible. In this case the most common one, the individual daily intake for each of the three isomers (α-, β- and γ-) amounts to appr. 1 μg. This amount may be increased by consuming relatively large amounts of animal fat.
In 4.4.1. a strong increase was mentioned in the concentration in homes following treatments of large surfaces with lindane-containing wood preservation products, the levels remaining elevated for a very long period of time (1-10 years).
Assuming an average indoor concentration of 1 $\mu g.m^{-3}$, a respiration rate of 12 m^3 per day, a 75/25 ratio for staying indoors and outdoors respectively, a limited proportion of the Netherlands population is probably exposed to a daily amount of more than 10 μg lindane (assuming 100% absorption). The influence (probably lasting for a shorter period of time) of HCH-containing insecticides used in homes and for parasite control on the indoor air concentration is not known and deserves further attention.
During the formulation and the packaging of HCH-containing pesticide powders and liquids, as well as during the application of these as

insecticides the occupational exposure may be increased considerably. However, no published data are available for the situation in The Netherlands.
With regard to aquatic ecosystems, the exposure levels may be equated to the concentrations in surface water (fresh water: <0.01 (α); <0.001 (β) and 0.01-0.04 (γ) $\mu g.l^{-1}$ and marine water: 0.001-0.002 $\mu g.l^{-1}$).
With respect to terrestrial ecosystems the exposure levels may vary greatly among species to species, depending on habits and feeding patterns.

4.7. SUMMARY AND CONCLUSIONS

In table 4.16. a summary is given of what is known about the HCH concentrations and contents found in the various environmental compartments and in food. Data on isomers other than α-, β- and γ-HCH are either lacking or limited.
In soils and in groundwater of nature reserves the concentrations are generally low. As a result of agricultural applications the levels may be increased by a factor 10 or more. At dumping grounds the contents may be very high: 10^4 times higher.
In sewage sludge the contents show a tendency to decrease. However, high values occur incidentally. The relatively high levels found for β-HCH in such cases are striking.
Over the past 10 years, HCH concentrations in the larger rivers have been rather constant. In some waterways, in harbours and near dumping grounds the contents in the sediment have increased significantly. In smaller waters near dumping grounds and in agricultural areas higher concentrations may occur. In respect of the latter, it should be noted that the number of out-laying peak-levels, that are probably due to the injudicious use of HCH, shows a tendency to decrease. With regard to the levels in estuaries, recent data are not available: 13-20 years old data indicate that an important part of the HCH is dissolved. The lowest concentrations are found in the open sea; the levels are higher near the coast because of the increased effect of instreaming polluted fresh water.

Table 4.16. Summary of levels of HCH concentrations and contents in the environmental compartments and in foods in The Netherlands

Matrices	α-HCH	β-HCH	γ-HCH	Unit
Soil				
- soil material				
nature reserves (average)	< 1		1-10	$\mu g.kg^{-1}$
agricultural area (estimated)			80-250	$\mu g.kg^{-1}$
dumping grounds (maximum)		10-100	10-100	$g.kg^{-1}$
- sewage sludge (average)	5-70	30-150	8-50	$\mu g.kg$
- ground water				
near dumping grounds (max.)	1.3	0.002	0.56	$\mu g.l^{-1}$
agricultural area (estimated)			<0.015	$\mu g.l^{-1}$
Surface water				
- fresh water				
*large waters (average)	<0.01	<0.001	0.04	$\mu g.l^{-1}$
sediments (median)	10-50	6-26	15-240	$\mu g.kg^{-1}$
*locally (harbours)(average)	0.1-2	0.1-1	0.1-1	$\mu g.l^{-1}$
sediments (average)	200-400	200-400	200-400	$\mu g.kg^{-1}$
*small waters (average)	<0.03		<0.03	$\mu g.l^{-1}$
(maximum)	<2	<1	>0.01	$\mu g.l^{-1}$
- marine water				
* estuarine mixing zone				
dissolved	0.001-0.02	0.02	0.001-0.02	$\mu g.l^{-1}$
suspended	<0.001-0.005	<0.001-0.003	<0.001-0.02	$\mu g.l^{-1}$
* coastal water				
dissolved (average)	0.001		0.004	$\mu g.l^{-1}$
suspended	<0.001-0.005		<0.001-0.01	$\mu g.l^{-1}$
* North Sea				
dissolved	≤0.001		≤0.001	$\mu g.l^{-1}$
Air				
- indoor air				
* homes built on dumping grounds (average):			0.006	$\mu g.m^{-3}$
* homes after treatment for insect control (wood-worms etc.):				
. the first few weeks to months			40-60	$\mu g.m^{-3}$
. after 1-10 years			0.5-2.5	$\mu g.m^{-3}$
- outdoor air				
nationally (average)	0.0003	n.d.	0.0002-0.0005	$\mu g.m^{-3}$
Food				
- total diet (median)	< 1	<1	<1	$\mu g.day^{-1}$

The annual average outdoor air concentrations are ≤ 0.5 $ng.m^{-3}$. In homes built on dumping grounds or in homes treated with HCH-containing pesticides the respective indoor air concentrations may be a factor 10 or ≥100 higher. For each of the three isomers the population exposure level is appr. 1 μg per person per day. This level is determined mainly by the contents and the

composition of the daily food intake; the contribution made by the inhalatory route is considered negligible. This particular route may raise exposure levels to appr. 10 μg of lindane per person per day for people living in homes treated with HCH-containing products. The effect of domestic uses of HCH on respective indoor air concentrations and, consequently, on exposure levels of man is not known.

5. EFFECTS

This chapter contains a summary of data on the toxicological effects of HCH. More detailed information may be found in the appendix (Janssen et al., 1987).

5.1. ALPHA-HEXACHLOROCYCLOHEXANE

5.1.1. Human toxicity

- Chemobiokinetics and metabolism

Experimental data indicate that α-HCH stimulates its own degradation during permanent exposure. The clearance rate from fatty tissues in rats seems to be sex-dependent. Continuous accumulation of α-HCH has not been demonstrated in either of the sexes. Nevertheless, substantial deposition in fat may occur. The presence of α-HCH in sucklings after oral treatment of the mother animals suggests the occurrence of excretion in milk.

- Animal experiments

In test animals α-HCH exhibits a slight to moderate acute toxicity. Acute toxicity data are limited to oral studies. In a 90-day study on rats the principal effects were leucocytopenia and liver changes, indicating enzyme induction. These effects were evident at 50 and 250 $mg.kg^{-1}$ feed, only slight at 10 $mg.kg^{-1}$ feed and absent at 2 $mg.kg^{-1}$ feed. Another short-term test showed that α-HCH causes liver enzyme induction in rats at all dosage levels tested of which 5 $mg.kg^{-1}$ feed was the lowest.
In carcinogenicity studies in mice and rats hyperplasia, nodules and tumours developed successively in the liver at high dosage levels only. DNA-binding studies in mice and the available mutagenicity studies failed to provide evidence for a genotoxic action of α-HCH. Consequently the compound cannot be qualified as an initiating carcinogen.

- Observations in man

There are no data on the effects of α-HCH in humans. In The Netherlands the compound is frequently found in human tissues; in the past few years the median levels in both blood and fat are below the detection limit. Generally the compound is not detectable in mother's milk.

5.1.2. Ecotoxicity

Most data concern the effects on aquatic species; only very little information has been found with respect to the effects on other species in the environment.

- Uptake, metabolism, bioaccumulation and elimination

No reliable data are available on the metabolism in aquatic organisms and birds.

In fresh as well as in salt-water the majority of the BCF values (biological concentration factors) ranges from 100 to 1000. According to the classification system of Canton and Slooff (1979) α-HCH may be considered moderately bioaccumulative. In a foodchain study only a slight increase in the equilibrium level was observed: no clear biomagnification could be detected. Both the accumulation and elimination are rapid processes (15 min - 72 hrs).

In birds the average accumulation factor is 2 (fat and eggs). The half-life is less than 10-14 days. No data are available on the bioaccumulation and metabolism in terrestrial organisms (invertebrates and plants).

- Toxicity to aquatic organisms and ecosystems

According to the classification system of Canton and Slooff (1979) α-HCH may be considered as very toxic to crustaceans and fish in short-term toxicity tests in fresh and salt water (L(E)C50 < 1 $mg.l^{-1}$), whereas, within the solubility limits, no discernable effects have been found on algae in fresh and salt water. Based on the results of long-term tests with freshwater crustaceans and fish, α-HCH may be considered as moderately toxic (NOEC [No-Observed-Effect-Concentration] 0.01-0.1 $mg.l^{-1}$) and slightly toxic (NOEC 0.1-1 $mg.l^{-1}$), respectively. No reliable data on long-term effects to salt-water organisms are available.

- Toxicity to terrestrial organisms and ecosystems

There are no data on the toxicity of α-HCH to terrestrial organisms (invertebrates and plants). For mammals the acute oral LD50 is 500-4674 $mg.kg^{-1}$ body weight.

5.2. BETA-HEXACHLOROCYCLOHEXANE

5.2.1. Human toxicity

- Chemobiokinetics and metabolism

In all species studied, including man, β-HCH accumulates in fat during continuous exposure. Upon cessation of the exposure the compound is eliminated slowly from the body.

- Animal experiments

The acute toxicity of β-HCH is low. Toxicity data are limited to oral studies. In short-term toxicity studies on rats white blood abnormalities, liver hypertrophy, kidney damage and, at high dosage levels only, hormone-like changes were the most prominent effects. In vitro studies confirmed the oestrogenic action of β-HCH. In reproduction studies on rats the compound caused infertility, an effect for which, in view of the preliminary nature of the studies, a no-effect-level could not be determined. The tentative no-effect-level derived from short-term toxicity studies on rats is 0.4 $mg.kg^{-1}$ feed. The results of additional reproduction studies may necessitate adjustment of this level in future.

Results of available carcinogenicity studies in mice show some similarity with those for α-HCH; liver tumours occurred at high dosage levels only. This effect did not occur in rats. The results from the few mutagenicity experiments carried out do not provide a sufficient basis for the drawing of definite conclusions on the mutagenic activity of β-HCH. Thus, based on the available experimental data on carcinogenicity and mutagenicity the compound is not considered to be an initiating carcinogen.

- Observations in man

In The Netherlands β-HCH is found in human tissues. During the period 1975-1982 the concentrations in fat and blood do not show a downward trend. In a recent series of measurements β-HCH was found in all samples at a median concentration of 100 $\mu g.kg^{-1}$ (maximum 300 $\mu g.kg^{-1}$) in terms of fat content.

5.2.2. Ecotoxicity

The few data available concern the toxicological aspects of β-HCH on aquatic organisms and chickens.

- Uptake, metabolism, bioaccumulation and elimination

No relevant data on metabolism are available. Both in fresh and in salt-water the majority of the BCF values ranges from 100 to 1000. Based on the classification proposal of Canton and Slooff (1979) β-HCH may be considered as moderately bioaccumulative. The equilibrium levels are reached within 10 days. In marine molluscs a half-life of 4 days was observed.
The average BCFs for eggs and bird fat are 13 and 15, respectively. The half-life is 6 to 8 weeks.

- Toxicity for aquatic organisms and ecosystems

Except for a test on *Tetrahymena* (EC50=1.2 $mg.l^{-1}$) only marginal effects have been found in short-term experiments on freshwater organisms close to the solubility limit of β-HCH. Based on the proposal of Canton and Slooff (1979) β-HCH is slightly toxic (NOEC 0.1-1 $mg.l^{-1}$) to crustaceans and moderately toxic (NOEC 0.01-0.1 $mg.l^{-1}$) to fish in long-term freshwater tests. β-HCH exerts an oestrogenic action on male and female fish. No information was found on the toxicity of β-HCH to salt-water species. Tentative results from studies on an aquatic microcosm indicate that the effects of β-HCH may be attributed to an anomaly of the community metabolism.

- Toxicity to terrestrial organisms and ecosystems

No data are available on the toxicity of β-HCH to terrestrial organisms (invertebrates and plants). For birds a short-term no-effect-level of 625 $mg.kg^{-1}$ feed has been derived. For mammals the acute oral LD50 is ≥ 1500 $mg.kg^{-1}$ body weight. A tentative short-term oral no-effect-level in rats of 0.4 $mg.kg^{-1}$ feed (0.02 $mg.kg^{-1}$ body weight) has been established.

5.3. GAMMA-HEXACHLOROCYCLOHEXANE

5.3.1. Human toxicity

- Chemobiokinetics and metabolism

After an oral dosage absorption is almost complete; there is evidence that dermal absorption is considerable. Upon uptake in the body of rats γ-HCH is, to some extent, stored in fat and elimination is fairly rapid. In humans accumulation in fat was not found to occur.

- Animal experiments

γ-HCH is moderately toxic after single applications. The short-term oral toxicity has been studied in rats and dogs. In both species indications of liver enzyme induction were found and, predominantly in males, effects on kidneys were observed. In one 90-day oral study kidney and liver damage was observed at dosage levels of 4 $mg.kg^{-1}$ and higher in the feed, whereas the only effect in female animals at 0.8 $mg.kg^{-1}$ was an increased liver cytochrome P-450 concentration (indicative of liver enzyme induction). In the other 90-day studies liver effects (including enzyme induction) and kidney changes occurred at levels of 10 $mg.kg^{-1}$ feed or higher, however, not at 2 $mg.kg^{-1}$ feed. In additional short-term studies in rats on liver enzyme induction dosage levels in the feed of up to 2 $mg.kg^{-1}$ produced no effects. Therefore it is concluded that the toxicological no-effect-level in rats for short-term administration is 2 $mg.kg^{-1}$ feed. Effects on liver and kidney also occurred in a semi-chronic inhalation study on rats at dosage levels of 0.5 and 5 $mg.m^{-3}$, administered for 6 hrs per day for 90 days, whereas 0.1 $mg.m^{-3}$ produced no effects.

No adequate chronic toxicity data are available. The scarce oral studies available show results which are consistent with the toxicological profile of the compound, derived from the short-term tests.

The results from the carcinogenicity studies were in line with those found in the studies with α- and β-HCH: in mice liver tumours occurred at high dosages only, whereas in the studies on rats this type of tumour was not observed. Additional short-term in vivo tests in mice did not indicate any genotoxic action of γ-HCH. The mutagenic activity was examined in a variety of test systems with different endpoints. Plants, bacteria, yeasts, *Drosophila* , mammalian and human cells in vitro as well as whole mammals were used for testing. γ-HCH did not induce mutations in any of the systems

examined, however, some cytogenic damage was observed in mammalian as well as in human cells in vitro. γ-HCH is considered a mitotic poison inducing C-mitosis, polyploidy and chromosome aberrations. The increase of liver tumours in mice is not considered sufficient evidence for carcinogenicity. It is concluded that there is insufficient evidence to consider γ-HCH an initiating agent.
In the teratogenicity tests performed no teratogenic effects were observed. In one 3-generation reproduction study a marginal effect on liver weight occurred at 25 $mg.kg^{-1}$ feed, the lowest dosage level tested. There were no effects on reproduction.

- Observations in man

No human studies on chronic effects of γ-HCH have been carried out. In The Netherlands γ-HCH is frequently found in human tissues. Given the rapid excretion of the compound the presence in human tissues may be assumed to reflect recent exposure. Recently the median concentration found in samples of human tissues were below the detection limit. Occasionally, γ-HCH is found in mother's milk: a series of measurements recently carried out yielded an average concentration in whole milk of 10 $\mu g.kg^{-1}$ (median: <10; 90%-value: 20).

5.3.2. Ecotoxicity

Ulmann (1972), Portmann (1979) and Wilhelm (1984, 1985) reviewed the ecotoxicological aspects of γ-HCH.
In reporting the literature data a distinction has been made between γ-HCH and lindane, because some authors specifically mention γ-HCH as a test substance, whereas others carried out the experiments with lindane (composition not specified). Most data concern aquatic and soil organisms while only a few data are available on the effects on birds, higher plants and honey bees. When abundant information on specific subjects was available only the relevant articles were reviewed.

- Uptake, metabolism, bioaccumulation and elimination

In plants γ-HCH is metabolized by dehydrogenation, dehydrochlorination, dehydration, oxidation and conjugation resulting in numerous metabolites which have been identified. Although no solid data are available on the

metabolic pathways in aquatic and soil organisms or birds, it may be assumed that in these organisms dehydrochlorination is equally important.
Both in fresh and in salt water the majority of the BCF-values range from 100 to 1000; based on the proposal of Canton and Slooff (1979), γ-HCH may be considered moderately bioaccumulative. The biomagnification-values are in the same order of magnitude. The equilibrium levels are reached within 4 to 5 days (fish) and 10 to 14 days (annelids). In salt water complete elimination takes place within one week.
In plants the uptake of γ-HCH mainly occurs through the roots. Following dispersal most of the γ-HCH and its metabolites is found in roots and a small amount, or none, in green leaves or fruit. In soil organisms the uptake of lindane at first increases slowly (equilibrium level within 5 weeks) but later on it declines. No distinct bioaccumulation has been observed.
In birds lindane is readily dispersed (especially towards adipose issues and eggs). The average accumulation ratio is 2. Its elimination takes place within 3 to 14 days.

- Toxicity to aquatic organisms and ecosystems

In accordance with the classification proposed by Canton and Slooff (1979) γ-HCH may be considered as very toxic to crustaceans, amphibians and fish in short-term freshwater toxicity tests (L(E)C50 < 1 $mg.l^{-1}$), as well as to crustaceans and fish in long-term freshwater toxicity tests (NOEC < 0.01 $mg.l^{-1}$). Algae, protozoa and molluscs are less susceptible.
In acute tests with salt-water organisms a large variance in LC50 values for crustaceans is found, varying from very toxic (< 1 $mg.l^{-1}$) up to slightly toxic (10 to 100 $mg.l^{-1}$). γ-HCH is very toxic to salt-water fish as well (LC50 < 1 $mg.l^{-1}$), whereas molluscs and annelids are less susceptible (slightly toxic: 10 to 1000 $mg.l^{-1}$). Long-term toxicity data on salt-water organisms are lacking.

- Toxicity to terrestrial organisms and ecosystems

Lindane (1 $mg.kg^{-1}$) has no chronic adverse effects on soil micro-organisms. Lindane is moderately toxic to earthworms in short-term (LC50: 50-136 $mg.kg^{-1}$) and in long-term studies (NOLC > 30 $mg.kg^{-1}$). Increase in body weight and fertility were affected at soil concentrations of 12 $mg.kg^{-1}$. Arthropods are more susceptible, 0.28 to 2.6 $mg.kg^{-1}$ soil causing 50 to 100% mortality in both short-term and long-term studies. Lindane proved to

be ovicidal to carabids. In field studies soil concentrations ≥ 1.8 $mg.kg^{-1}$ caused a protracted reduction in the arthropod fauna. γ-HCH is very toxic to honey bees. Lindane is moderately toxic to birds in acute and short-term studies with LD50 and LC50 values being 50-600 $mg.kg^{-1}$ body weight and 425-5000 $mg.kg^{-1}$ feed, respectively. In long-term studies a no-effect level of 10 $mg.kg^{-1}$ feed was observed. Teratogenic effects were observed upon immersion of eggs in solutions containing lindane or lindane in oil at concentrations corresponding with dosages of 58 and 8.2-9.5 $kg.ha^{-1}$, respectively. Such effects were not observed after administration of lindane in bird feed. γ-HCH is moderately toxic to mammals with acute oral and dermal LD50-values of 70-480 and 50-500 $mg.kg^{-1}$ body weight, respectively. The inhalatory LC50 for rats is > 603 $mg.m^{-3}$. The short-term oral no-effect level in rats is 2 $mg.kg^{-1}$ feed (0.1 $mg.kg^{-1}$ body weight); the short-term inhalatory no-effect level in rats is 0.1 $mg.m^{-3}$.

5.4. DELTA-HEXACHLOROCYCLOHEXANE

5.4.1. Human toxicity

- Chemobiokinetcs and metabolism

The available data on chemobiokinetics and metabolism are fragmentary. The gastro-intestinal absorption appears to be high. It is not clear to what extent accumulation in fat occurs.

- Animal experiments

In animal experiments δ-HCH is slightly to moderately toxic after acute oral application. No adequate short-term or long-term studies are available. Carcinogenicity was tested in two limited studies on mice and rats; both tests were negative. There are no data on mutagenicity. Reproduction studies are also lacking.

- Observations in man

Data on the occurrence of δ-HCH in human tissues in The Netherlands are not available.

5.4.2. Ecotoxicity

Only a few data are available on the ecotoxicological aspects of δ-HCH. Toxicological effects with respect to aquatic organisms other than micro-organisms and molluscs have not been reported. As for mammalian wildlife reference is made to the data obtained from laboratory animal studies.
From the only study available on bioaccumulation in a freshwater fish species and a salt-water mollusc species, δ-HCH may be considered as moderately accumulative (BCF's 100-1000) in accordance with the classification of Canton and Slooff (1979). The equilibrium levels are reached within 4 days. Rapid elimination has been observed in freshwater fish (T1/2 < 2 days) whereas the elimination in molluscs is very slow.
No data are available on the toxicity to algae, crustaceans and fish. The only EC50-value available (a protozoa species) indicates that δ-HCH is very toxic to this organism in an acute test (< 1 $mg.l^{-1}$).
No data are available on the bioaccumulation, metabolism and ecotoxicity of δ-HCH in terrestrial organisms (invertebrates and plants). For mammals the acute oral LD50 is 750-1000 $mg.kg^{-1}$ body weight.

5.5. OTHER ISOMERS

No data are available on the human toxicity and ecotoxicity of the other HCH-isomers.

5.6. MIXTURES OF ISOMERS

Most studies were performed with the HCH-isomer mixture of technical HCH. The composition of technical HCH shows considerable variation, the concentrations ranging from 21 to 71% for α-HCH, from 2.1 to 10% for β-HCH, from 13 to 39% for γ-HCH, from 3.5 to 23% δ-HCH, and from 2 to 15% for others.

5.6.1. Human toxicity

- Animal experiments

The animal test data on mixtures of HCH-isomers do not provide evidence for a synergistic action of the isomers.

- Observations in man

Humans, occupationally exposed to high technical HCH concentrations, exhibited (slight) aberrations in white blood parameters and blood hormone levels. However, no data on exposure to lower concentration levels are available.

5.6.2. Ecotoxicity

Few literature data are available on the ecotoxicity of technical HCH. Only those relevant articles in which the composition of the technical HCH used was mentioned have been reviewed.

No reliable data on the metabolism of technical HCH in aquatic organisms, plants and terrestrial organisms are available. Based on the classification system of Canton and Slooff (1979) technical HCH may be considered as moderately bioaccumulative in salt-water species (100-1000). Elimination is completed within one week.

In short-term tests technical HCH is moderately toxic to freshwater fish and amphibians (LC50: 1-10 $mg.l^{-1}$) and slightly toxic to algae (EC50: 10-100 $mg.l^{-1}$) based on the same classification proposal. However, the results of the two short-term tests indicate that the compound is very toxic to salt-water organisms (LC50 < 1 $mg.l^{-1}$). No data from long-term tests are available.

At a soil content of 10 $mg.kg^{-1}$ or more plant growth may be affected, especially on light (sandy) soils. In field studies no residual effects on plant growth have been observed. The micro-arthropod fauna is strongly affected at dosages of 13.6 $kg.ha^{-1}$, while lycosid spiders are killed after soil applications of 30-60 $kg.ha^{-1}$ (food chain effect).

Technical HCH has a moderate acute toxicity in birds (LD50: appr. 600 $mg.kg^{-1}$ body weight). In a long-term study the no-effect level was 84 $mg.kg^{-1}$ feed.

For mammals the acute oral LD50 is 600-1250 $mg.kg^{-1}$ body weight.

5.7. EFFECTS ON MATERIALS

All HCH-isomers are stable when exposed to air, strong acids, light and heat (up to 180 ^{o}C). However, except β-HCH, they are sensitive to dehydrochlorination by bases at room temperature. At high temperatures and especially in direct contact with open fire HCHs decompose during which process the highly corrosive hydrochloric acid is formed. Therefore, effects may be expected on electric contact materials (Huck and Duerrwaechter, 1980), heat exchangers and flues in incinerators (Hama and Curley, 1965). The effects just mentioned have been observed for various chlorinated hydrocarbons at concentrations down to 25 ppm in inlet air of heating installations (Hama and Curley, 1965).
However, no effects have been reported on materials as a result of the presence of HCH in the vapour phase at common indoor and outdoor air concentration levels.

5.8. GLOBAL EFFECTS

By its persistence in the environment and its relative volatility HCH will be transported over long distances in water, but especially in air. As a result HCH is found in mosses, lichens, plants and animals far from anthropogenic sources. The significance of this world-wide dispersion is not known.

5.9. CONCLUSIONS

5.9.1. Humans

Experimental data on the metabolism and toxicity are available for α-, β-, γ- and δ-HCH and for the isomer mixture of technical HCH.
HCH-isomers are readily absorbed in the gastro-intestinal tract: for α-, β-, γ- and δ-HCH generally values of more than 90% of the dosages are reported. For γ-HCH substantial absorption through the skin was found for humans. After absorption the highest concentrations are present in adipose tissues. The extent of accumulation in these tissues may be as $\beta > \alpha > \gamma$ HCH and is apparently higher in females than in males. For β-HCH no

equilibrium is reached in humans between intake and excretion during an exposure period of 10 years or more.
The exact metabolic pathway of most HCH-isomers is not known. For the γ-isomer an almost complete metabolic model has been proposed. Animal experiments show that 2,4,6-trichlorophenol is the major metabolite for α-, β- and δ-HCH. In addition biotransformation of γ-HCH yields substantial quantities of 2,4,5-trichlorophenol and 2,3,4,6-tetrachlorophenol.

The toxicity of γ-HCH was investigated extensively whereas for the other isomers less data were available. The acute toxicity of HCH-isomers is generally slight to moderate. γ-HCH is the most toxic of the isomers after single application. In subacute dietary studies all isomers tested induced the following effects on the liver: hepatomegaly, induction of cytochrome P-450 and liver enzyme activities and indications of centrilobular SER-proliferation. The activity of the various isomers with regard to these effects did not show any consistent pattern in toxicity studies, which is probably due to methodological differences.
For β-HCH an estrogenic action was demonstrated in both in vitro and in vivo tests. In addition, preliminary findings indicate that low-level exposure may induce infertility and reduced viability in young animals. Changes in the kidney were observed after exposure to γ-HCH. These changes are characterized by an accumulation of hyaline droplets in the epithelium of the proximal tubules, predominantly occurring in male rats. The effects found with technical or artificial mixtures are consistent with those observed for the individual isomers without evidence of a synergistic action among the isomers.
In carcinogenicity studies with individual isomers as well as with (technical) mixtures induction of liver hyperplasia, nodules and/or carcinoma were found at dosage levels equal to or above those inducing the above-mentioned liver changes in short-term studies. γ-HCH is the only isomer tested extensively for mutagenic properties. In a wide variety of test systems with different end points no mutations were found. Limited tests for γ- and β-HCH are negative with respect to mutagenicity and/or binding to macromolecules.
Based on the available in vivo and in vitro experimental data none of the HCH-isomers qualifies as an initiating carcinogen. Therefore a non-threshold approach is not appropriate. The available toxicity data, and the

resulting no-effect levels, may be adopted as the basis for the allocation of acceptable exposure levels.

For α-HCH the 90-day oral no-effect level in rats is 2 $mg.kg^{-1}$ feed (appr. 0.1 $mg.kg^{-1}$ body weight). Using a safety factor of 100, a tolerable daily intake of 0.001 $mg.kg^{-1}$ body weight may be calculated. An acceptable daily intake (ADI) is not determined, α-HCH being considered an undesirable pollution. No experimental data on the inhalatory toxicity of α-HCH are available.

Of all HCH-isomers β-HCH has the strongest accumulative properties. The available data indicate that in animals as well as in humans no equilibrium reached between intake and elimination is during continuous exposure. The tentative short-term oral no-effect level is 0.4 $mg.kg^{-1}$ feed (appr. 0.02 $mg.kg^{-1}$ body weight). The results of future research on the effects of β-HCH on reproduction may necessitate adjustment of this level. Long-term uptake of β-HCH should be avoided on account of its accumulative properties. No experimental data on the inhalatory toxicity of β-HCH are available.

The short-term oral no-effect level for γ-HCH in rats is 2 $mg.kg^{-1}$ feed (appr. 0.1 $mg.kg^{-1}$ body weight). With a safety factor of 100 this results in an acceptable daily intake (ADI) for the oral route of 0.001 $mg.kg^{-1}$ body weight. The short-term inhalatory no-effect level in rats is 0.1 $mg.m^{-3}$. It should be noted that this level was found in a study in which there might have been some oral intake as well due to deposition of the compound tested in the exposure chambers. Other inhalatory studies not being available, however, 0.1 $mg.m^{-3}$ is used for the evaluation of the toxicological ory risk of the presence of γ-HCH in ambient air.

For δ-HCH the available data are insufficient for an adequate toxicological evaluation. The same applies for the other HCH-isomers. Generally the population is more likely to be exposed to mixtures of HCH isomers than to pure isomers. In this respect is is noteworthy that the available studies with isomer-mixtures (mostly technical HCH) did not provide evidence for synergism among the isomers.

5.9.2. Ecosystems

Aquatic life

At present there are no generally accepted methods for the extrapolation of results from laboratory single-species toxicity studies to ecosystems in the field. For the time being two different theoretical procedures will be used to calculate "safe levels" for the aquatic or terrestrial environment. In this way information may be gathered on the possible usefulness of these extrapolation methods, however, the results obtained should be considered as being indicative only. The results are given in table 5.1.

Table 5.1. Review of calculated "safe" concentrations of α-, β- and γ-HCH and technical HCH in freshwater and marine aquatic ecosystems according to the methods by Slooff et al. (1986) and Kooyman (1985a,b) and the toxicological limit values that may be derived from these data

Method	α-HCH	β-HCH	γ-HCH	Techn. HCH
	fresh/salt	fresh/salt	fresh/salt	fresh/salt
Slooff et al. (1986)				
-relation LC50 acute/NOECss				
NOECss *	40/27	-/-	0.21/0.0097	-/0.019
with uncertainty factor	1.6/1.1	-/-	0.008/0.0004	-/0.007
-relation LC50 acute/NOECeco				
NOECeco **	223/161	-/-	2.5/0.067	-/0.118
with uncertainty factor	2.6/1.9	-/-	0.030/0.0008	-/0.0014
-relation NOECss/NOECeco				
NOECeco	551/-	198/-	8.34/-	-/-
with uncertainty factor	16/-	6/-	0.25/-	-/-
Kooyman (1985a,b)				
-safe LC50	2.8/-	-/-	0.03@/0.0003@	-/-
-safe chronic value	-/-	0.14@/-	0.07@/-	-/-
Proposed "safe" value	5/1	1/-	0.1/0.002	-/0.001

* "No-observed-effect level" for single species

** Ibid for ecosystems

@ Based on the most susceptible species (crustaceans and fish); if based on all test results: 0.0015 $\mu g.l^{-1}$ (β-HCH, chronic/freshwater) and 0.004 $\mu g.l^{-1}$ (γ-HCH, acute/freshwater). In calculating the safe chronic value for γ-HCH on the slightly susceptible Lymnaea were excluded; when all data were used a safe chronic value of 0.00001 $\mu g.l^{-1}$ was calculated. For the water calculations of the "safe" concentrations for γ-HCH in salt water the data for the slightly susceptible molluscs and annelids were not taken into account; a value of 0.0000016 $\mu g.l^{-1}$ was calculated when these data were included.

- α-HCH

Because of the relatively small number of chronic studies a "safe" level of 5 $\mu g.l^{-1}$ for fresh water of is proposed (between 3 and 16 $\mu g.l^{-1}$). For salt water only the first two relations as proposed by Slooff et al. (1986) may be used because no data on chronic toxicity were available and only a few on acute toxicity. A "safe" level of 1 $\mu g.l^{-1}$ for salt-water is proposed.

- β-HCH

Because of the fact that within the solubility limits of β-HCH in fresh water in short-term tests only some behavioural effects on fish were observed, the third relation as proposed by Slooff et al. (1986) and the second method as proposed by Kooyman (1985b) may be used to extrapolate a "safe" level.

For fresh water 1 $\mu g.l^{-1}$ (between 0.14 and 6 $\mu g.l^{-1}$) may be proposed as "safe" level, because of the relatively small number of chronic studies.

- γ-HCH

The extrapolation methods proposed by Slooff et al. (1986) and Kooyman (1985a,b) have been applied to the results from the fresh-water toxicity studies:

A "safe" level of 0.1 $\mu g.l^{-1}$ for fresh water (between 0.07 and 0.25 $\mu g.l^{-1}$) is proposed because many data on chronic toxicity to susceptible species were available.

With respect to salt-water organisms only few acute toxicity data are available. Therefore only the first two relations as proposed by Slooff et al. (1986) and the first proposal of Kooyman (1985a) can be used to calculate a "safe" level. Because no chronic toxicity data are available and in acute tests γ-HCH proved to be about 20-40 times more toxic in salt water than in fresh water, a tentativel "safe" level of 0.002 $\mu g.l^{-1}$ (between 0.0025 and 0.005 $\mu g.l^{-1}$) is proposed.

- Technical HCH

Since there are insufficient data on toxicity to freshwater organisms no "safe" level for the freshwater ecosystem can be calculated.

Because only a few acute toxicity data on susceptible salt-water species are available, only the first two relations as proposed by Slooff et al. (1986) may be used to extrapolate a "safe" level, which is tentatively put at 0.001 $\mu g.l^{-1}$.

Terrestric life

- γ-HCH

From the data on soil organisms arthropods showed to be the most susceptible. Based on the toxicity values for these species a tentative concentration of 0.01 $mg.kg^{-1}$ soil is proposed, being a concentration which is unlikely to harm the terrestrial ecosystem.

6. MONITORING METHODS AND MEASURING STRATEGIES

6.1. MONITORING METHODS

Although an extensive literature exists on the analysis of organochlorine pesticides, few papers deal with the HCH-isomers only. The residue analysis has rapidly undergone substantial development and therefore many publications are either out of date or even unreliable. The "normalized" methods published are obsolete, especially in the USA. Therefore, a selection from the literature has been made, taking into account the practical experience at the various research institutes in The Netherlands.

For a general introduction reference is made to the biannual literature reviews in the April editions of Analytical Chemistry, a compendium of regulations made by TNO (TNO, 1980), the frequent publications in the J. Assoc. Off. Anal. Chemists and the review articles by Bayona et al. (1983), Onuska (1984), Cairns et al. (1983), Keith et al. (1983) and Stan and Goebel (1983). Beyermann (1984) presents an excellent review on the organic micro compounds in general.

The analysis of organochlorine pesticides is usually performed as follows. First, the compounds are extracted from the sample with an organic solvent. In addition to the pesticides a number of other compounds are extracted as well. These substances interfere with the analysis in two ways: in the gaschromatographic final analysis the pesticide peaks are masked and the GC-column rapidly becomes contaminated. Therefore, a so-called clean up is carried out subsequent to the extraction, in most cases with liquid chromatography over either silicagel, aluminium oxide, "Florisil" or an organic polymer. The organochlorine pesticides appear in the eluate and the interfering compounds must remain behind in the adsorption column or, at least, they must be separated. In this way most of the pesticides are separated from the PCBs as well. The eluate or its fractions are analyzed chromatographically.

Nowadays the GC-determination is fully automated. As yet, this has not not been achieved for the clean-up although attempts are being made (Gretch and Rosen, 1984).

The success of any method of analysis strongly depends on the nature of the sample material taken (the matrices). This is most pronounced in the analysis of biota, soil material and sewage sludge.

6.1.1. Sample preparation

- The required sample size

In table 6.1. the relationship between the analytical detection limit desired, the sensitivity of the detectors and the required sample size is given.

Table 6.1. Relationship between detection limit, sensitivity of GC detectors and sample size (smallest detectable amount with ECD 0.5 pg; with MSD (SIM) 5 pg; recovery 50%)

Matrix	*Concentration to be determined*	*Required sample volume/weight*			
		*inj. of 1% of an extract **		*inj. by thermic desorption*	
		ECD	*MSD*	*ECD*	*MSD*
Biota	*1 ng.kg^{-1}*	*0.1 kg*	*1 kg*	*after application of*	
Soil	*1 µg.kg^{-1}*	*0.1 g*	*1 g*	*purge and trap only*	
Silt	*1 mg.kg^{-1}*	*0.1 mg*	*1 mg*		
Water	*1 pg.l^{-1}*	*100 l*	*1000 l*	*after application of*	
	1 ng.l^{-1}	*100 ml*	*1 l*	*purge and trap only*	
	1 µg.l^{-1}	*100 µl*	*1 ml*		
Air	*1 pg.m^{-3}*	*100 m^3***	*1000 m^3*	*1 m^3*	*10 m^3*
	1 ng.m^{-3}	*100 l*	*1000 l*	*1 l*	*10 l*
	1 µg.m^{-3}	*100 ml*	*1000 ml*	*1 ml*	*10 ml*
	1 mg.m^{-3}	*100 µl*	*1000 µl*	*1 µl*	*10 µl*

* *Injection volume: 1 µl. However, much may be gained by injecting much larger quantities of an extract, up to 0.25 ml (Zlatkis et al., 1984).*

** *Some investigators take larger samples, of up to 4000 m^3.*

- Isolation of HCH from the sample

In general, HCH is isolated from a sample by extraction with organic solvents (or a mixture) (Van Steinwandter, 1985). Attention is further drawn to two additional methods.

The HCH-isomers are the most volatile of the organochlorine pesticides known. At room temperature, 1 m^3 of air saturated with lindane vapour contains appr. 150 µg of the compound. For the HCH-isomers purge and trap methods seem to offer possibilities, although no publications on this matter have been found. On the other hand, the extractive steam distillation

method has been published. The STORA-report (1980,1984) on the analysis of sewage sludge reveals moderate results obtained with this method, probably due to the unnecessarily complicated equipment used. The method gives high recoveries and clean extracts as long as the equipment described in the TNO Test Book (TNO, 1977) is used. Santa Maria et al. (1986) obtained good results with samples of water, fish, vegetables and fruit. In most cases the extracts obtained using this method only require a simple clean-up.
HCH-isomers easily decompose in an alkaline environment (see chapter 3). Therefore, (weak) alkaline samples (for example sea water) should be rendered weakly acid before applying extractive steam distillation.
Photochemical transformations of HCH-isomers are mentioned in the literature, for example the conversion of γ-HCH to α-HCH (see chapter 3). Whether these play a role during the storage of samples and extracts or during the analysis has not been subject to investigation.
In the literature no mention has been made of thermic isomerization.

- The clean-up

Saponification, either alone or in combination with treatment with concentrated sulphuric acid, is often applied for the clean-up. Alkaline saponification cannot be applied, as HCH decomposes in an alkaline environment. However, treatment with strong sulphuric acid is sustained well.
The liquid chromatographic purification of the extracts obtained takes place over aluminium oxide, silicagel, Florisil, and/or by gel permeation chromatography (Sephadex, Bio-Beads). The prescriptions based on the three first-mentioned adsorbents (there are hundreds of them) are not easily transferable; generally they have to be tested, adapted and continuously controlled in the laboratory. Not all of the HCH-isomers may reach the same eluate fraction.
The clean-up has two advantages: many substances that interfere with the analysis are removed and contamination of the capillar column is significantly reduced. The latter advantage remains even if one could refrain from a further clean-up of the extract by using a MSD. In the latter case, samples obtained by extractive steam distillation are by far superior as they no longer contain any fats.
Finally, attention is drawn to the enzymatic exclusion of biota. This rather simple technique is applied relatively less frequently despite the advantage it provides by facilitating the preparation of clear solutions

from homogenized biological material, which are better suited for further processing (Putman-Groenheyde, 1984).

- Reduction of the extract volumes

The relatively high volatility of the HCH-isomers may lead to losses during the reduction of extracts to smaller volumes by evaporation. Dry-damping is fundamentally wrong, unless an extract still contains a considerable amount of fat. Analytical procedures with an evaporation step that secures good recoveries for PCBs and DDT for example, may not be satisfactory for HCH-isomers. In such cases the results from HCH analyses should be interpreted with some caution.

6.1.2. Analytical methods

- Thin layer chromatography

The literature on the TLC of HCH-isomers has not been subject to exhaustive studies as this technique is little applied in the analysis of residues. At least six out of the eight HCH-isomers may be separated by TLC (Thielemann, 1976). In the Stahl's manual (1962) examples are given. The method has been used as a clean-up method in the determination of GC in air (Abott et al., 1966). Using micrc-TLC, 20 ng of lindane is still detectable (Visweswariah et al., 1971). In ecotoxicological research the most important application of TLC is the separation of radio-actively labelled isomers (Grundey and Kraus, 1976).

- Liquid chromatography

A rudimentary form of liquid chromatography is used during the clean-up of HCH-containing extracts over silicagel or other substrates. HPLC is another possibility (Bombaugh et al., 1970). On the basis of results obtained with TLC, it is likely that HPLC may be utilized for the separation of the various HCH-isomers.

- Gas chromatography

Although capillar columns have a higher separative capacity, packed columns may be used to separate HCH-isomers. Examples of this are given by Waliszewski et al. (1980a,b) and by Steinwandter (1980). Other examples may be given in which the stationary phase, the length and the temperature of the column and other variables have been adapted to the specific separation

problem, even though these are rarely focused on HCH-isomers (Onuska, 1984; Sandra, 1985; Riply and Braun, 1983 and Stan and Goebel, 1983). Increasingly, fused silica columns are being used.
The smallest amount of a HCH-isomer detectable with an ECD on a routine basis varies from 0.1 to 5 pg; in special cases 10 fg can be detected (Wells, 1983).
The sensitivity of the various chlorine specific GC-detectors is usually at the nanogram level, however, with the ECD and the MSD it is possible to work in the sub-picogram and low-picogram ranges respectively.
Thermic isomerization of the HCH-isomers has not been described; at high temperatures it is unlikely that injection will cause disturbances. As stated before, the final GC-determination may be fully automated.

- GC-MS

The use of two different GC-columns for a more precize identification is too laborious for routine residue analysis and, in addition, insufficient. Therefore, the MS-detection in the SIM-mode is being applied increasingly (Cairns et al., 1983; Hargesheimer, 1984; Lopez-Avila et al., 1985; Suzuki, 1983).
Although the fragmentation patterns of the different HCH-isomers are rather similar, according to Suzuki (1983) they show slight mutual differences. The following mass fragments are the most important ones for the GC-MS in the SIM-mode: m/e 109 and 111; 181, 183 and 185; 217, 219 , 221 (and 223). Under normal conditions, 5-10 pg may still be measured effectively with electron ionization.
GC-MS with negative chemical ionization (NCI) for the determination of HCH-isomers in air has been described by Oehme and Stray (1982). These authors achieved a detection limit of a few $pg.m^{-3}$. However, the number of mass spectrometers with which the NCI technique is applied for these purposes is limited.

6.1.3. The determination in soil

A general introduction into the problems associated with soil analysis is given by Baggen et al. (1983).

- The sampling

Recently (RIVM, 1986) a workshop was held on Sampling and Analysis in Soil Pollution studies, where a "draft tentative practical guidelines for sampling and analysis" was elaborated upon: An updated version has meanwhile been published, in which the results of the workshop have been integrated (DHV, 1986).

- The extraction of the samples

Most investigators dry the soil material by exposure to air at room temperature before extraction; others mix the soil material in a mortar with anhydrous sodium sulphate. Two methods are used for the extraction. One group of scientists use Soxhlet extraction as this method is believed to give the highest yield. This extraction method uses several organic solvents (acetone, dichloromethane) and a slightly polar solvent (hexane, toluene). Other researchers perform batch extraction, sometimes accelerated up by means of ultra sound. Hexane-acetone mixtures are often used for this purpose (DHV, 1985), but acetonitril (Suzuki, 1984) or toluene (Gast, 1985) may be used as well. Reference is made to Suzuki (1984), Müller et al. (1985) and RIZA (1985). Studies on the extraction of soil for the analysis of organochlorine pesticides have been carried out by Chiba (1969), Woolson (1974), Sporstol et al. (1983) and Steinwandter (1985).

- The clean-up and further analysis

See section 6.1.2. and the references given in section 6.1.6. With respect to the removal of sulphur from the extracts: see the STORA-report (1980-1984) including the comments given in section 6.1.6.

- Validation and quality control

Especially with regard to soil and sediment the validation and quality control form unsolved problems. The addition of spikes to the samples is of doubtful value. At best, it assists in assessing the reliability of the analytic procedures following extraction. As a consequence, one has to be satisfied with the extraction method that yields the highest results, without obtaining the percentage of the total amount of HCH recovered. An alternative is to render the soil matrix completely soluble, for example by the use of HF. This has been attempted for the analysis of fly-ash, but not for soil or sediment.

6.1.4. The determination in water

- The sampling

Sampling bottles should be made of glass or aluminium (Weil and Quentin, 1970; Gaul, 1986). Samples of up to 10 l can be taken with sampling bottles especially designed for this purpose (Stadler and Schomaker, 1977). In open sea, where the HCH concentrations are among the lowest, care should be taken to exclude the micro surface layer from the sample.
Several methods and materials for pumping up water have been described (Dawson and Riley, 1977; Weber and Ernst, 1983; Van de Meent et al., 1985). No attention has been paid to the possible effects of pumping procedures on the sample: these are assumed to be non-existent.
According to Weil and Quentin (1970) samples may be stored for two weeks, in either light or darkness, at 5 ^{o}C or 20 ^{o}C. However, this concerned fresh water only and no mention was made of the pH.
The prescription of the RIZA (1985) only mentions that samples should be stored in a refrigerator.

The micro-surface layer

In the past few years much research has been conducted on the so-called micro-surface layer at sea. In this layer the concentration of organochlorine pesticides may be 10^{5}-10^{8} times higher than in the water below. There is, however, no agreement on the definition of this layer or on the most suitable technique for sampling it (Baumann Ofstad et al., 1979; Van Vleet et al., 1980; Carlson, 1982).

The extraction of water samples

In order to minimize losses and contamination, the extraction should be carried out in the sampling bottle. As a result of the high partition coefficient of the HCH-isomers in the n-hexane/water system (appr. 10,000) this is quite feasible: high recoveries are obtained by one-off extractions with relatively small amounts of extraction liquid (Stadler and Schomaker, 1977; TNO, 1977). As little as one or two ml of n-hexane or cyclohexane is sufficient for a one litre sample. By the addition of water after extraction, such a small amount of hexane may be pressed into a small tube in order to be manipulated further with an injector. After extraction the empty

sampling bottle should be rinsed with the extraction liquid, even though HCH is hardly adsorbed on glass surfaces (Weil and Quentin, 1970,1971). According to the method of Stadler et al. (1977) HCH is obviously extracted from the water sample including the suspended particles it may contain. The usefulness of prior filtration and the related problems are still being discussed.
Several systems have been described for the manipulation of larger water samples: for continuous extraction (Duinker, 1984) and for passing such samples through an adsorption column (Duinker, 1984; Neu, 1985). The latter method poses problems as HCH-isomers bound to small particles may pass an adsorption column. Finally, attention is drawn to the possibility of applying extractive steam distillation.

- Further analysis of the extract

Reference is made to Waliszewski et al. (1980), Wegman and Greve (1980), Tenabe et al. (1983), Suzuki (1983), Steinwandter (1985) and the prescription by the RIZA (1985).

- Validation and quality control

It may be important to enrich ("spike") water samples with a suitable internal standard immediately after sampling. For this purpose, one may use an HCH-isomer usually not found in water samples such as δ-HCH. Evendently, C^{13}-marked HCH-isomers may be applied when the final determination takes place with GC-MSD.

6.1.5. The determination in air

Reference is made to Tanabe et al. (1982,1983), Oehme and Stray (1982), Bidleman (1985), Bidleman et al. (1981) and Billings and Bidleman (1983).

- The sampling

In view of the usually low concentrations sampling is combined with preconcentration. To this end, a sufficient amount of air is sucked through an adsorbent. Although many adsorbents have been described, 50% of the publications concern poly urethane foam (PUF), while in 25% of them XAD-resins are mentioned (Bidleman, 1985). Measuring results obtained from the analysis of dust particles and aerosols collected on filters with high volume samplers yield values that are much too low (Weil et al., 1973);

adsorbents are essential for proper sampling. The break-through volume should be determined in all instances (Burdick et al., 1981) and the effects of disturbing variables should be known.

Poly urethane foam has several advantages. Since this material has the lowest flow resistance, it may lend itself best for the high discharges required to take large samples (up to 4,000 m^3) for the detection of HCH in low concentrations. Moreover, the material is cheap and it is not easily spilled. The high sampling discharge leads to larger-sized adsorption cartridges: up to 10 cm in diameter and about the same length. Because of the volatility of the HCH-isomers, the sample volume is limited to 250 m^3 (Billings and Bidleman, 1983), or with smaller PUF-cartridges even to 50 m^3 (Burghardt, 1986). For the processing of large samples, combining PUF with XAD-2-resin may be instrumental in avoiding break-throughs. Smaller adsorbtion tubes will suffice for the higher concentration ranges (work places). The cartridge with PUF and/or XAD-resin should be preceded by a glass fibre filter, on which the particle fraction is collected. Extracts from filters and adsorbents are combined.

Adsorption cartridges loaded with HCH-isomers may be stored in a deepfreeze for at least one month. Billings and Bidleman (1983) mention storage periods of 8 months at room temperature and of three years in a deepfreeze. Long storage increases negative results.

An automated sampling device has been described by Gerhart et al. (1980). The literature provides many variations on this theme. For instance a portable sampler has been described Lewis et al. (1982)

- <u>The use of gas chromatography</u>

The HCHs adsorbed are recovered from the adsorbents by extraction with an organic solvent. The extract is concentrated by careful evaporation. Sometimes the extract can be analysed directly. Often, however, a simple clean-up is required first. Thermical desorption has not (yet) been applied for introducing samples of organochlorine pesticides into a gas chromatograph.

- <u>Validation and quality control</u>

The combination of sampling on PUF-cartridges, extraction, clean-up and GC-analysis (MSD and ECD) has been evaluated in a multicentre study in which 9 American laboratories participated (Bidleman, 1981). The relative standard deviation amounted to 50-60%.

The best currently available method for testing the analytical procedure is to add a known quantity of (if necessary marked) HCH to the filter or at the anterior end of the adsorption cartridge subsequently passing clean air through the system with a volume consistent with the sample size. The analysis follows as usual (Oehme and Stray, 1982). In practice, however, the break-through volume may be less than in the testing of clean air. The application of this method to every sample and the use of C^{13}-marked HCH-isomers may facilitate a continuous quality control and quarentee for the use of a GC with an MSD in the SIM-mode for the analysis.

6.1.6. The determination in biota

The various HCH-isomers occur in biota in concentrations in the range of 0.1 $\mu g.kg^{-1}$ in total organisms, i.e. up to 20 $mg.kg^{-1}$ in fatty tissues (see chapter 4).
The concentration of HCH in organisms may be used as a measure of environmental pollution with HCH. The advantage is that some kind of integrated picture of the pollution is obtained on the basis of HCH concentrations in the organisms which, because they are higher than those in the environment, are more readily determined. To this end, molluscs are often used for fresh and salt-water environments (De Kock and Marquenie, 1981; Satsmadjis et al., 1983; Mouvet et al., 1985 and Green et al., 1985); and mosses for outdoor air (Thomas and Herrmann, 1980; Oehme et al., 1985 and Gaggi et al., 1985).

- The taking, storage and homogenization of samples

The great diversity of biota renders statements on sampling techniques futile.
If samples need to be stored for a relatively limited period of time (a few weeks), storage in a deepfreeze is usually sufficient. If samples need to be stored for much longer periods (specimen banking) storage at -80°C is to be preferred. The necessity and justification of these statements need to be proven by testing. Norstrom and Won (1985) performed storage tests with chicken and herring-gull egg homogenates at -18 to -28 °C. The latter did not result in any changes in the composition of a number of organohalogenated pesticides when tested after three years. Large losses of HCH, however, appeared to result from freeze-drying. According to Kawar et al. (1973) dairy products containing lindane may be stored at -26 °C for at

least 4 months; cheese even at +7 °C. According to Shaw and Wilson (1981), α- and γ-HCH decompose between few days and several weeks in freeze-dried egg and in extracted egg white kept at room temperature.
It may be concluded with caution that in most (but not all) cases deepfrozen storage is possible for 3 years . Samples that are defrosted in order to take sub-samples for analysis, need to be homogenized again.

- The isolation from biota

Two methods are mentioned here: extraction with organic solvents, which is most commonly applied, and extractive steam distillation, which seems to gain ground. A proposal for the simplification of the extraction procedure is given by Steinwandter (1985a); in yet another paper Steinwandter (1985b) argues that the Soxhlet-extraction leaves much to be desired and that batch-extraction with a mixture of hexane-acetone is to be preferred. It is advantageous to apply an enzymatic dissolving method before extractive steam distillation (Putman-Groenheyde, 1984). This technique leads to very clean extracts that need only little clean-up. HCH isomer yields, however, have not been determined (TNO, 1977).

- The clean-up

Reference is made to 6.1.1. After comparing several clean-up methods for fish-oil Sperling et al. (1985) concluded that a liquid/liquid partition in the dimethylformamide/hexane system, followed by a reversed-phase separation (C18), results in the cleanest extracts, suitable for GC-MS. Contardi et al. (1985) describe a simplified extraction from fish tissues with 9.5-13 M sulphuric acid. According to the authors, the results were superior to than those obtained with Soxhlet-extraction.

- Procedures commonly applied in The Netherlands

The prescriptions for the analysis drawn up by the RIKILT (Tuinstra, 1981) are used in a number of laboratories in The Netherlands. In all instances the analysis is completed by means of automatic GC-ECD on a WCOT-column.

- Validation and quality control

The recovery of the analytic procedure may be controlled through the use of live organisms after having absorbed C^{14}-labelled HCH (Putman and Groenheyde, 1984). Internal standards need to be determined as well. Standards are equally required to test the stability of the GC-system. The results of several ring tests on the determination of HCH-isomers and other compounds in various materials have been published (McFarren and Lishca, 1971; Aspila et al., 1977; Eder et al., 1981; Burse et al., 1983; Ault and Spurgeon, 1984; Ribick et al., 1981 and Sawayer, 1985).

6.1.7. The determination in sewage sludge

In practice, several problems have arisen during the determination in sewage sludge. These were examined by the STORA and a prescription for the analysis has been proposed (STORA, 1980, 1984). In the relevant document, however, it is not explicitly proven that HCH-isomers survive the procedure used for the removal of elementary sulphur. In a more or less comparable procedure losses have been observed (Mattson et al., 1976). Although compounds added to silt may be recovered well, it has not yet been proven that compounds reaching sewage in a "natural" way can be extracted with the same efficiency. This problem will not be discussed further; the situation is similar in respect of soil, biota and fly-ash.
Two articles not mentioned in the STORA-report, are of importance: those of McIntyre et al. (1980) and Erickson and Pellizzari (1977).

6.2. MEASURING STRATEGIES

6.2.1. Soil

Immission of HCH into the soil takes mainly place on and in the top layer. With regard to the distribution over the soil compartments the HCH-isomers appear to be bound mainly to the solid phase. As a consequence, the wash-out of HCH-isomers to groundwater will be limited, while, as a result of low degradation rates, accumulation will occur in the top soil. Studies on soil pollution in Twente and Bunschoten indicate that residues do, however, reach ground- and surface water. Therefore, as a first step, sampling and analysis always should take place in the top 1-2 m below ground level. To

obtain bio-available fractions the soil material should be extracted with non-polar and polar organic extraction fluids.
At local dumping grounds HCH may have reached the groundwater as a result of, for instance, supersaturation of the binding capacity of the soil and/or short-cuts in the flow pathways like cracks. Sampling and analysis of the groundwater is highly essential in case positive (> 0.01-0.05 mg.kg^{-1}) contents are found in the top soil. The necessity for analysis of the top soil may be confirmed by a comparison of contents measured with the reference values as was done in Twente and Bunschoten (chapter 1), or with the toxicological limit value for terrestrial ecosystems (chapter 5). The latter values may locally be exceeded by a factor of more than 10^6. A similar reasoning is applicable to groundwater studies. In such studies attention should also be given to the possible metabolites, like those mentioned in chapter 3.

6.2.2. Water

The range of the maximum HCH concentrations in the state-regulated surface waters is just below the no-effect levels determined for long-term effects on aquatic organisms; the limit value for γ-HCH is often exceeded in coastal waters. Therefore, continuous monitoring of state-regulated waters and coastal waters is required. A measuring frequency of once a month at strategically important locations, as applied by the Department of Public Works, is considered reasonable.
In agricultural areas the levels found show considerable peaks as a result of fluctuations in the use of lindane. Samples taken at random in areas where the product is applied need to be examined to facilitate the inspection of its use.
The HCH concentration range is a factor 10 lower than would be expected on the basis of the contents in underwater soils and the water-silt partition coefficient. This suggests that underwater soils may act as sources of HCH for surface water. Therefore, the analysis of state-regulated waters should preferably be performed on the water itself rather than on underwater soils.

6.2.3. Air

A national measuring strategy for HCH in air is of no importance since the average concentration level of HCH in air (< 1 $ng.m^{-3}$) is far below the level at which toxicological effects may be expected.
HCH is mainly emitted into the air by the spraying of crops; after spraying the concentration of γ-HCH may increase up to some tens of $\mu g.m^{-3}$ in leeward situations for short periods of time (from a few hours to a few days). One may consider a measuring strategy of leeward sampling at locations at a distance varying 10 m to 1 km from the treated fields during short periods (one week maximally). Taking the fairly uniform application and the resulting equally uniform emission patterns into account, measurements may be restricted to a small number of representative fields.
The domestical use of lindane, which may lead to an increase of the indoor air concentration, may deserve further attention. Considering the various possible applications and the different emission patterns resulting from these, specific measuring strategies should be followed.

6.2.4. Food

The present intake of HCH through food is below the acceptable c.q. tolerable levels proposed in chapter 5. Since there are no reasons to assume that the contents in foodstuffs will increase, a measuring strategy for food is not considered necessary.

6.3. SUMMARY AND CONCLUSIONS

The available analytical methods for the determination of HCH-isomers are sufficiently selective and sensitive; table 6.1. presents an overview of the detection limits. Taking the possible contaminations into account it is recommended to perform screening tests in addition to the specific determinations. Problems remain in respect of the quality guarantee in the analysis of soil samples, but this applies equally to the analysis of other organic micro-pollutants. The measuring results presented in chapter 4 are mostly derived from studies that were primarily focussed on the determination of PCBs, DDT etc.. As the conditions for analyzing the HCH-isomers

were therefore often not optimal, the published measuring results should be handled with due caution in theses instances.

On a national scale, only the continuous monitoring of state-regulated waters (coastal waters included) is relevant; the measuring frequency of once a month at strategically important locations, as presently applied, is considered adequate.

On a local scale (dumping grounds, agricultural areas) the measurement of all environmental compartments may be desirable. If positive contents (>0-.01-0.05 $mg.kg^{-1}$) are found in the top soil and if the soil has a low binding capacity (< 2% organic material), the complementary analysis of groundwater is recommended. In agricultural areas where HCH is applied surface waters should be analyzed at random for purposes of inspection. Further investigation is recommended with respect to HCH concentrations in indoor air during and after various domestic applications.

7. EMISSION CONTROL

7.1. SOIL

With regard to the soil (ground water included) the following situations should be distinguished:

- Existing polluted locations; in particular the rehabilitation of locations that are heavily polluted by activities in the past, like the dumping of HCH-containing waste and the production and the formulation of HCH-containing pesticides. As a result of such pollutions the groundwater may be polluted with HCH as well by wash-out.

- Pollution by current applications of lindane; in particular the measures to reduce the emissions to the soil.

In addition to this lindane-containing waste matter may result from the present use which mostly enters the soil. The following sections describe the possible measures to reduce these emission.

7.1.1. Measures to rehabilitate polluted locations

The possible methods to clean historically polluted locations are still being studied. In the framework of the Interim Act on Soil Rehabilitation the clean up is planned to start within a few years. At this moment there are two options for the reconstruction of HCH polluted soil:

- To limit the dispersion of HCH to the environment c.q. threatened objects such as groundwater and/or vegetation and cattle. This may be achieved by a civil engineering or hydrogeological isolation and by constructing a clean root layer. If necessary, the use of the location may be adapted.

- To remove the polluted soil and to transport it to a (temporary or permanent) controlled storage depot and to fill up the location with clean soil from elsewhere.

In fact, in using these methods the soil remains polluted. Methods to purify the soil are not (yet) available; a number, however, are being studied:

- microbiological treatment by landfarming;
- microbiological treatment in a slurry reactor;
- thermal purification;
- extractive purification, in which the pollution is concentrated to a relatively small volume.

With respect to the microbiological treatment of soil Doelman and Slange (1984, 1985) demonstrated that the α-HCH content of appr. 5,000 $mg.kg^{-1}$ may be reduced to appr. 3,000 $mg.kg^{-1}$ (anaerobic treatment) or to 2500 $mg.kg^{-1}$ (aerobic treatment) within 20 weeks. (see section 3.1.3.).
Borkent-Verhagen et al. (1986) indicated that sandy soils can be purified to 0.1 $mg.kg^{-1}$ within half an hour at temperatures of 350°C and higher. For clay and peaty-soil more intensive thermal treatments are necessary. The treatment of waste gases liberated from the soil should take place under very strict conditions since there is a possibility of chlorinated dibenzodioxine being formed.
With respect to extractive purification (public) information is not yet available.
No statements can be made as to the chance of the purification techniques mentioned.

If HCH occurs only in the humus layer at concentrations that are not too high, the use of the capacity for self purification of the soil may be considered. Suitable methods are:

- To discontinue utilization of the soil for one or more years or to change the use into one which is less sensitive or vulnerable (possibly after removal).
- To take measures that stimulate the degradation of HCH. According to e.g. Ferreira and Rashu (1981) this may be done by creating anaerobic conditions in the soil, for example by irrigation or by the addition of compost or undecomposed organic matter (> 10 tons per hectare).
- To take measures that make the HCH less bioavailable, for example by the addition of organic matter.

It is obvious that combinations of these methods may be applied and that during the reconstruction no lindane-containing products may be used.

Presently there is little experience with these methods in The Netherlands. These in situ methods are considered less suitable for the reconstruction of localities polluted in the past, though, since the availability of HCH for biodegradation might be limited. For a final answer, however, further studies should be done.

The costs of the various methods for reconstruction are given in table 7.1. The following should be noted, however:

- The choice of a certain method for reconstruction at a location does not solely depend on the accompanying costs, but also on factors such as the possibility of dumping the soil somewhere else, the usage and the nature of the terrain, the urgency and the local political situation.
- During reconstruction works a certain amount of pulluted groundwater will often be liberated wich should be treated as well. Reference is made to section 7.1.4.
- The costs of purification cannot be presented since purification techniques for HCH-polluted soil are not yet in practice. It may be assumed that the costs range from Dfl. 200 to 400 per m^3 of soil (PWS, 1985; Roozeboom, 1986; Koen, 1986).
- The costs mentioned do not include the possible costs of expropriation, compensations, costs of supervision, research costs, purification of groundwater etc., unless mentioned otherwise. In practice the additional costs sometimes even exceed the basic costs.
- The costs mentioned apply to locations varying in size from appr. 0.1 to 10 ha.

Table 7.1. Rough costs for the reconstruction of HCH-polluted soil (PWS, 1985; Wicker, 1985; Roozeboom, 1985; Koen, 1986)

Method	Rough costs * (in Dfl.,excl. VAT)	Remarks
1] Discontinuation of utilization and planning of woods or parks		
- agricultural area	20-30 per m^2	Total costs, compensation for buildings excluded
- urban area	≥ 100 per m^2	
2] Hydrogeologial isolation application of clean root layers	50-75 per m^2	Dfl. 2-10 extra per m^3 is required for groundwater purification (Dfl. 2-20. per m^2 annually)
3] Like [2], incl. sheet-piling	50-100 per m^2 of additional sheet-piling	At vulnerable locations a sheet pile wall is sometimes required; sometimes savings on groundwater purification up to appr. 50% are possible
4] Removal and transportation of polluted soil; application of clean soil (see 5, 6 and 7)	0-80 per m^2	Possibly higher costs when removal is complex; Dfl. 10-20 extra per m^3 per year if temporary storage is required
5] Like [4] and transportation to (IBC) dumping grounds	20-80 per m^3 extra	
6] Like [4] and transportation to dumping grounds abroad	250-350 per m^3 extra	The possibilities of transportation abroad have decreased
7] Like [4] and purification of the soil:	not yet known, probably Dfl. 200-400 per m^3 extra	
- microbiologically		
- thermally		
- extraction		
8] In situ microbiological treatment (addition of organic matter, ploughing, spraying); temporary discontinuation of utilization	≤ 10 per m^3 annually	Only applicable to superficial pollution (<0.5 m) in agricultural areas; duration of treatment not known; less suitable for β-HCH and pollutions from the past

* The costs may vary considerably per location and additional costs are not included, unless stated otherwise

7.1.2. Groundwater

Pollution of groundwater with HCH may result from the HCH load of the soil, for example as a result of the dumping of HCH-containing waste in the past. In some cases it may be necessary to purify the groundwater. The techniques that may be used are discussed in section 7.2.1.

Van Luin and Warner (1985) described the purification of HCH-containing groundwater on a practical scale (coagulation, flocculation, flock-separator and active carbon adsorption with a capacity of 40 $m^3.h^{-1}$). The HCH concentrations of the groundwater to be purified amounted to 400-600 $\mu g.l^{-1}$; the discharge requirement was 1 $\mu g.l^{-1}$, being the sum of all the isomers. Since about 90% of the HCH was dissolved (probably partly complexated by the presence of humic acids), the purification efficiency of the flocculation treatment was low, only 10%. This result is in accordance with those obtained by other research workers e.g. Robeck et al. (1965). In contrast to this, excellent results were obtained using the right type of active carbon (initially the presence of other contaminants was not taken into consideration in the choice of carbon). In several cases the HCH-concentration of the purified water was < 0.05 $\mu g.l^{-1}$. Weekly one of the carbon filters used was replaced.

According to the report the costs of the groundwater purification are Dfl. 1.50-2.50 per m^3 at a capacity of 40 $m^3.h^{-1}$. For smaller installations with a capacity of 1-5 $m^3.h^{-1}$ (for areas between 5,000 and 20,000 m^2) the basic costs of groundwater purification will amount to Dfl. 5-10 per m^3.

7.1.3. Measures for emission reduction

- Application in agriculture and horticulture

In principle emission control is only feasible by prohibiting the present applications of lindane. Alternative products are available for the application of HCH on leaves as for the control of Colorado beetles, plant lice and trips (in 1984 appr. 0.2 tons of lindane was used for this application). Hence, the lindane emissions resulting from the use in agriculture and horticulture may be reduced by only 1% to appr. 22 tons per year. At present substitutes have also been admitted to control parasites in cattle (sheep and pigs), however, in The Netherlands hardly any experience has been gained with these compounds, moreover these products (when on the market) are about three times more expensive than lindane. A banning on the use of lindane in this application leads to a reduction of the emission into the soil of 0.5 tons per year (appr. 2%).

- Other applications

The other current applications of lindane (tables 2.6. and 2.7.) comprise appr. 6.5 tons of HCH annually, part of which (more than 2 tons) enters the soil. In this respect wood preservation and parasite control in cattle are the main sources of soil pollution.

The pyrethroids are a good alternative for lindane in wood preservation, although still insufficient experience has been gained with respect to the long-term effectivity of these products. Substitution in this application results in an emission reduction into the soil of appr. 1.8 ton per year (more than 7%). Good (but more expensive) substitute pesticides are available for the other applications, but substitution does not result in a detectable emission reduction to the soil compartment.

- Waste matter

Lindane, possibly present in compost of vegetative remains, sewage and in other residual matter, will mainly be degraded. The remainder will enter the soil surface, where the degradation is continued.

7.2. WATER

7.2.1. General

Several methods have been tested to remove HCH (especially lindane) from water. The following techniques may be mentioned: chemical coagualtion-flocculation (Thébault et al., 1981), chemical oxidation with chlorine, potassium permanganate and ozone (Robeck et al., 1965), adsorption to active carbon (Robeck et al., 1965), ion exchangers and other adsorption resins like styrol divinyl (DVB), co-polimerization products (Wolf and Lindau, 1977), magnetite (MacRae, 1985), argano-clay (Beall, 1983) and to combustion ashes (Winkler and Mueller, 1979), reverse osmosis (Chian et al., 1975; Malaiyandi et al., 1980) and irridation (Vollner et al., 1975).

Generally adsorption to active carbon appears to yield the best results. The experiments by Robeck et al. (1965), in which two serial columns with a carbon volume of appr. 55 l were used, indicated that pesticides -among which lindane- could be removed to a detection level of 0.01 $\mu g.l^{-1}$. However, no report on the adsorption capacity was made since during the experiments the point of saturation was not reached. Good results with the removal of lindane

from drinking water were also found by Moergeli (1972) by applying active carbon adsorption in a column with a carbon layer of 70 cm minimally and at a linear rate of 6.5-15 $m.h^{-1}$. Such results were not achieved when using rapid sand filtration and ozonization.
In the literature no data on the process and design for adsorption to active carbon were found. The choice of the type of active carbon seems to be extremly important. For each case a choice should be made based on experimental studies. In doing so, representative (waste) water should be used to prevent other organic matter from being adsorbed.
Research performed by Winkler and Mueller (1979) and Birr et al. (1983) shows that lindane is removed well also in aerobic biological purification. Removal percentages of 28-85% (load 0.15-0.26 kg BOD per kg dry matter per day) and 50-99% (load 0.6-1.1 kg BOD per kg dry matter per day) were found. Another reference (DBW/RIZA, 1986) mentions a removal of 14-80% at an influent concentration of 0.1 $\mu g.l^{-1}$ lindane. The removal of lindane is mainly based on adsorption to active silt flocks. It is assumed that lindane adsorbed to sewage sludge to be degraded anaerobically in the sludge fermentation tank.

7.2.2. Applications in agriculture, horticulture and cattle-breeding

In table 2.7. (chapter 2) it was indicated that the application of lindane in agriculture and cattle-breeding only leads to small emissions into water provided the instructions in the admission decree are followed. Parasite control in cattle is still a relatively important (potential) source of emissions into water. To reduce these emissions it is possible in principle to install a simple carbon filter system at the farm. In the past this has also been done for aqueous solutions of mercury-containing products which were used to fumigate flower-bulbs and seed-potatoes. Considering the rather poor results with the "farm method" in the fumigation of seed-potatoes a switch was made to collecting the fumigant liquids and processing them centrally. Such a method may also be chosen for lindane-containing water resulting from parasite control. Obviously this requires good management and motivated users of the lindane-containing pesticides. Another possible method to reduce the emission is to limit or ban the use of lindane-containing pesticides for these applications. Considerations in this matter should take place in the framework of the admission policy under the Act on Pesticides.

7.2.3. Other applications

Lindane originating from applications to control wood affecting insects in buildings and from domestic use is likely to occur mainly diffuse; it will partly enter the sewer with domestic waste water and rainwater. This municipal waste water is mostly purified mechanically biologically in a waste water treatment plant. In sections 3.2.1. and 7.2.1. it is indicated that lindane is partly removed in these plants and consequently ends up in the sewage sludge, resulting in pollution of the sludge, which may cause problems in the sales (for example to agriculture). However, considering the low concentrations that are found in practice according to DBW/RIZA (1986) (< 0.05 $mg.kg^{-1}$ d.m.) this will not be the case.
For the applications mentioned good substitutes are available on the market. A restriction of the use of lindane-containing products for these applications will lead to a maximum reduction of the emission into the water of 0.2 tons per year, by which, however, the diffuse load of surface water would be reduced to a large extent.

7.3. AIR

Emissions into air result almost only from activities mentioned in sections 7.1. and 7.2. Through the air the HCH will rapidly enter the water and the soil (see deposition, 3.3.2.). The measures to reduce the emissions of HCH have been described in the sections concerned.

7.4. SUMMARY AND CONCLUSIONS

When the soil has been polluted with HCH-containing (waste) matter as a result of dumping, only two possible methods for reconstruction are available at present: isolation of the location and removal of the soil followed by temporary (controlled) storage somewhere else. In the latter case the basic costs are between Dfl. 50 amd 100 per m^3, to be increased by Dfl. 5-15 per m^3 annually for the costs of control. Final transport to a dumping ground abroad amounts to Dfl. 300 per m^3. Several purification methods are still being studied; the costs of which are expected to be between Dfl. 250 and Dfl. 450 per m^3.

For polluted groundwater limited experience is available with a purification on the basis of adsorption to active carbon. The purification requirements can be met. In this case the costs are Dfl. 5-10 per m^3 for areas varying from 0.5 to 2 ha (1-5 m^3 per hour).
Current emissions may be prevented especially by the restriction or banning of the use of lindane as a pesticide. Considerations in this matter should take place in the framework of the admission policy under the Act on Pesticides.

8. BUSINESS ECONOMIC CONSEQUENCES OF EMISSION REDUCTION

The business economic consequences of the proposed measures to reduce the emissions will be discussed in this chapter. The sectors that may be affected are: formulating companies, agriculture and horticulture, and cattle breeding. In the following sections these three sectors will be discussed. Subsequently the remaining applications will be touched upon.

8.1. FORMULATING COMPANIES

The formulating companies may be confronted with the measures in three different ways: measures to reduce the emissions, decreasing market for HCH compounds and costs of reconstruction, if these are passed on to the companies and the costs of cleaning up if these are passed on to the companies.

- Emission reducing measures

In chapter 2 the present environmental provisions were considered sufficient. Therefore additional measures were not considered essential. Consequently there are no business economic consequences.

- Decreasing market

In 1983 the sales of lindane containing-compounds involved a value of Dfl. 3.7 million, which is less than 1% of the total turnover of the formulating companies in that year (Dfl. 602 million). Less than 20% of the Dfl. 3.7 million (less than Dfl. 1 million), was sold in The Netherlands. The rest was exported. Banning of all lindane use in The Netherlands with the exception of soil and seed treatment would decrease the sales by 6-7 tons of HCH (appr. 25 tons of HCH-compounds). This could cause a decrease of the turnover of appr. Dfl. 200,000. It is not possible to indicate to what extent a banning in The Netherlands would affect the export. Assuming all HCH-compounds sold in The Netherlands to be processed by Dutch formulating companies, their total turnover might decrease by less than 0.1 %. This effect is negligible. In addition the demand for substitutes would increase, being three times more expensive than lindane (Engelhard, 1986). On balance a partial banning of lindane may even increase the turnover of the formulating companies. Similar measures abroad would initially affect

the export to some extent, but on balance they will hardly affect the turnover of formulating companies.

- Costs of reconstruction

The costs of total rehabilitation of soil polluted with HCH, may, in Overijssel alone, mount up to Dfl. 70 million (140,000 m^3 of soil at Dfl. 500 per m^3). If the (former) producer were to be held responsible for this he would be confronted with extremaly high costs. However, it is still not certain whether and to what extent a cleaning up will take place (see 7-.1.1.). Furthermore the spread of the costs has not been determined yet; this will be considered both politically and legally. Finally it is incorrect to compare a large incidental burden as an inheritance from the past to the costs and yields belonging to normal business management. Therefore the business economic consequences of the measures will not be discussed extensively.

8.2. AGRICULTURE AND HORTICULTURE

The agricultural and horticultural companies may be confronted with measures to restrict or to ban the use of lindane-containing products. For that reason additional restrictive measures are not proposed. Hence, there are no business economic consequences.

In the application of lindane to control leaf insects, which has already been reduced considerably, appr. 0.5 tons of lindane is involved at a value of appr. Dfl. 15,000 per year. The product value of the potato culture, an important field of application, amounts to appr. Dfl. 1,500 million per year, including an added value of appr. Dfl. 300 million (NMB, 1986). Even if the present costs spent on HCH is tripled, this item would still be far below the 0.01% of the sales and added value. The economic consequences of substitution seem negligible.

At present appr. 20 tons of lindane per year is used in soil treatment, to a value of appr. Dfl. 600,000. If suitable substitutes would become available in the near future, these costs might be tripled to Dfl. 1.8 million after banning lindane. An important application is in sugar beet culture. The sales in this sector over the past 5 years are about Dfl. 750 million (LEI/CBS, 1986), including an added value of appr. Dfl. 150 million (NMB, 1986). If the additional costs are entirely on the account of this sector, this would imply an increase in costs by appr. 0.2% of the sales

(0.8% of the added value). This increase though not negligible, will not influence the management policy substantially. Apart from this the consequences will occur more diffuse since lindane is used for soil treatment in other sectors (maize, horticulture) as well. Therefore it is expected that termination of the use of lindane for soil treatment when substitutes are available will not affect the companies concerned.

8.3. CATTLE BREEDING

Appr. 2 tons of lindane are used annually for parasite control. In the case of lindane being be banned the costs for especially the sheep breeding, presently appr. Dfl. 65,000, could be tripled. The turnover in sheep breeding being Dfl. 160 million per year, this is an increase in costs in the order of 0.1%. An alternative with a limited chance of success is to collect the rinse water and to process this centrally (see 7.2.). Irrespective of the organizational problems such a system would become rapidly more expensive than a total banning, just because of the undoubtedly high transport costs. As in agriculture and horticulture the increase in costs is so slight that the business economic consequences are to be neglicted.

8.4. OTHER APPLICATIONS

In the other applications appr. 4.5 tons of HCH are involved, with a market-value of Dfl. 200,000 per year. The additional costs of a banning would partly weigh upon households, which involves no direct business economic consequences. For the rest the amounts of money are that small and in addition spread over so many government agencies and companies, that noticeable effects are not to be expected.

8.5. SUMMARY AND CONCLUSIONS

As a result of measures to restrict the use of lindane formulating companies may be confronted with emission reducing measures, decreasing sales and/or costs for cleaning. Additional restrictive measures are not considered not essential, whereas on the balance the turnover of the companies is hardly expected to be affected. In view of the uncertainty as to the

extent of the cleaning as well as in the ratio of distribution of the related costs, the business economic consequences are not pursued further. Companies in agriculture and horticulture may be confronted with measures to restrict or to ban the use of lindane-containing products. Restrictive measures have not been proposed here; in case of substitutes being available, banning of the use of lindane would be unlikely to have noticeable consequences for the companies. The same applies to the use in cattle breeding and to the other applications. This matter should be considered within the framework of the admission policy under the Act on Pesticides.

9. EVALUATION

9.1. RISKS AND GROUPS AT RISK

For only three of the eight stereo-isomers of HCH (α-, β- and γ-HCH) there are sufficient data to determine a risk. Considering the expected lower exposure levels of the remaining isomers, the related risks are not estimated to be higher than those related to α-, β- or γ-HCH.

9.1.1. Risks for man

With respect to the oral exposure to α-, β- and γ-HCH the following is recommended. A tolerable daily intake of 1 $\mu g.kg^{-1}$ body weight is proposed for α-HCH (this compound is legally impermissible). As to β-HCH it is concluded that permanent exposure to this compound is not desirable, because of its accumulative properties. Therefore a tolerable dose at chronic exposure as suggested for α-HCH cannot be given. For γ-HCH an acceptable daily intake (ADI) of 1 $\mu g.kg^{-1}$ body weight is proposed. The most recent data on oral exposure to the three isomers indicate there is no toxicological risk, the actual intake of both α- and γ-HCH being far below the proposed limit values. β-HCH usually appears to be absent in the daily diet (> 90% of the cases); besides there is a wide margin between the tentative no-effect level (20 $\mu g.kg^{-1}$ body weight per day) and the maximally observed intake through the food (most recent measurement: 2 μg per day). Therefore long-term oral exposure is expected only in exceptional cases, whereas the occurrence of direct toxic effects resulting from exceptionally high intake levels is unlikely. Hence, there will also be no toxicological risk worth speaking of with respect to β-HCH.

As to the inhalatory route of exposure, only toxicity data on lindane are available. On the basis of limited data a no-effect level of 100 $\mu g.m^{-3}$ was deduced. The large-scale annual average in The Netherlands amounts to appr. 0.5 $ng.m^{-3}$, so there is a safety margin of more than 10^5 between this exposure level and the level at which harmful effects may occur. Therefore there is no risk for the general population. Considering the usually lower concentrations at which α-HCH, and certainly the other isomers, are present in the atmosphere, the nation-wide risk of the other isomers is considered nil.

Apart from a small group of occupationally exposed workers, an unknown number of persons are exposed by the domestic use of lindane as a pesticide. The data on the exposure levels occurring in these situations concern mostly indoor air concentrations after the use of lindane as a wood preservative. After such applications the concentrations remain elevated for a long periods (order of magnitude: a few to some tens of $\mu g.m^{-3}$). The margin between these concentrations and the no-effect level for lindane is too narrow to consider them as being safe. The effects of the use of lindane-containing insecticides in and around the homes to control parasites on the indoor air concentrations are probably more short-lasting but not known and deserve further attention.

9.1.2. Risks for ecosystems

Considering the application of HCH it is evident that especially the active components (γ-HCH, technical HCH) pose a risk to certain groups of species in both aquatic and terrestrial ecosystems. In this respect it should be noted that, although lindane is used as an insecticide, species other than insects are susceptable to these organochlorine compounds as well.
A comparison of the concentrations in inland and marine waters (table 4-.16.) with the toxicologically safe levels (chapter 5) indicates that with respect to the fresh water the β- and γ-HCH concentrations reach (β-HCH) or exceed (γ-HCH) the toxicological limit values only locally (some harbours, channels, small waters in agricultural areas) and with respect to the marine environment the γ-HCH content of the coastal water exceeds the ecotoxicologically acceptable concentration. The pollution in this coastal water is mainly the result of the inflow of HCH-containing fresh water; exceeding of the toxicologically safe level is the result of the higher toxicity to salt-water species in comparison with freshwater species.
The toxicologically safe levels mentioned are above the present quality criteria for the fresh surface waters in The Netherlands (section 1.2.2.). These quality requirements are frequently exceeded in polder waters and their reservoirs; in the large rivers they are exceeded incidentally. No environmental quality requirements have been formulated for the marine environment (yet), where problems may arise in the coastal area by lindane.
It is noted that the median values of HCHs in the submerged soils as well as the average contents in sewage sludge frequently exceed the reference value for soil in many cases.

With respect to terrestrial ecosystems a tentative limit value of 10 $\mu g.kg^{-1}$ of soil was proposed, corresponding to the reference value given in the discussion paper on soil quality (section 1.2.1.). Locally, as in dumping grounds, this value is exceeded excessively. In agricultural areas where lindane-containing products are applied a basic content of lindane in the soil of 80-250 $\mu g.kg^{-1}$ is assumed, by which the tentative limit value c.q. reference value is exceeded. As a result of the persistence of HCH this level is approximated and incidentally exceeded even in nature reserves.

9.1.3. Other risks

As a result of their persistence HCHs are transported over large distances, resulting in pollution on a global scale. As yet the risks related to this event seem to be small, considering the present background levels.
The present exposure levels will not cause any damage to materials.

9.2. CONCLUSIONS AND RECOMMENDATIONS

9.2.1. Environmental pollution in the past

At several locations in The Netherlands the soil and the groundwater is heavily polluted with HCH as a result of activities, often related to the production of HCH in the fifties. The load resulting from this exceeds the criteria and guidelines mentioned in chapter 1; at some localities the soil may even be qualified as chemical waste. The possibilities for cleaning these historically polluted areas are presently still being studied. There are several methods, the costs of which amount to appr. Dfl. 200-400 per m^3 of soil. The choice to reconstruct or not, as well as the choice of the methods to be used, depends on the use and the nature of the area, the possibilities of dumping the soil elsewhere or changing its use, as well as the acceptability of the risks and the political situation. In principle measures are desirable if there is a risk for further distribution from these localities. In that case measures are recommended which are aimed at the stimulation of the self-purification capacity of the soil.

9.2.2. Environmental pollution resulting from present use

Although the environmental pollution with HCHs has sharply decreased after the closing down of production units in The Netherlands more than 30 years ago, there are strong indications that the use has increased by appr. 50% in the past 10 years. This, in spite of the policy to reduce the use of lindane-containing products. This emission into the environment results in a load of the soil and, by the persistence of the HCHs, of the coastal waters at a level that seems to be above the toxicologically acceptable level. The emission of HCHs can only be reduced by limiting or banning their use. Considering the small economic consequences on company level as well, it is recommended to stimulate the use of substitute products, or, when the occurring side effects give rise to act, to enforce this through limiting or terminating the admissions. When no substitutes are available, the development of these compounds should be stimulated.

With respect to public health the indoor environment deserves further attention in relation to the domestic use, the occurring exposure levels being insufficiently known. Further studies into this matter are recommended.

With respect to the ecosystems the acute toxicity data strongly indicate that lindane is more toxic to marine species than to freshwater species. In order to determine whether or not the present environmental requirements for fresh surfacewater are sufficiently protective to the aquatic life in the coastal water of The Netherlands, further chronic toxicity studies with salt-water organisms are recommended.

DEFINITIONS RELATED TO EFFECT ORIENTED STANDARD SETTING

GENERAL

Environmental standard

General rule describing the condition in which the environment should be, specified in terms of space and time. Dependent on the degree of obligation one may distinguish:

Tolerable level

A quality level which should at least be achieved or maintained and which should be observed by the authorities concerned. Therefore exceeding of this level is not permitted except in case of force majeure or if strict application stands in the way of an optimally integral efficiency for the environment (risks unacceptable).

Acceptable level

A quality level that should be present now or after a certain period of time, in certain areas or otherwise. This requirement offers the possibility of giving relatively unpolluted areas more protection or of stimulating the authorities to further emission reduction (risks acceptable).

Desirable level

A quality level at which no effects of environmental pollution that are considered to be harmful to man, animals, plants and materials are to be expected. This level is the ultimate quality aimed at (risks negligible).

Limit value

Recommended value for a desirable level or other environmental quality requirement, based on scientific data only.

SPECIFIC TERMS WITH RESPECT TO THE SOIL

Reference value

A quality level indicating the margin between a multi-functional soil and a non-multi-functional soil.

A, B, C- criteria

Quality levels that are used in a testing framewwork for soil sanitation and reconstruction: A is the reference value (comparable to the background concentration); B is the testing value for further investigation and C is the testing value for sanitation and reconstruction studies.

Signal value

Indicative level in the soil, above which problems in agriculture may be expected and which serves as a signal to perform (further) investigation into the significance of the pollution for agriculture.

Action limit value

Concentration level in the agricultural product itself, below the interim criterium, which induces further studies in order to eliminate this load by taking measures at the sources.

SPECIFIC TERMS WITH RESPECT TO SURFACE WATER

Basic quality

Minimum quality level which should be met in fresh surface water in The Netherlands to enable a general ecological base, existing of a set of 35 criteria.
(Furthermore there are criteria for specific functions such as drinking water preparation, swimming water and fishing water).

SPECIFIC TERMS WITH RESPECT TO AIR

Deposition objective

Amount of a compound or group of compounds, which is maximally allowed to deposit from the atmosphere per area and per period of time.

MAC-value (TLV)

Maximum accepted concentration at the working place of a gas, vapour, haze or dust, which is not detrimental to the health of both the employees and their posterity in general, at repeated exposure and during a long period, even during the total period of labour.

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10.8. REFERENCES CHAPTER 8

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