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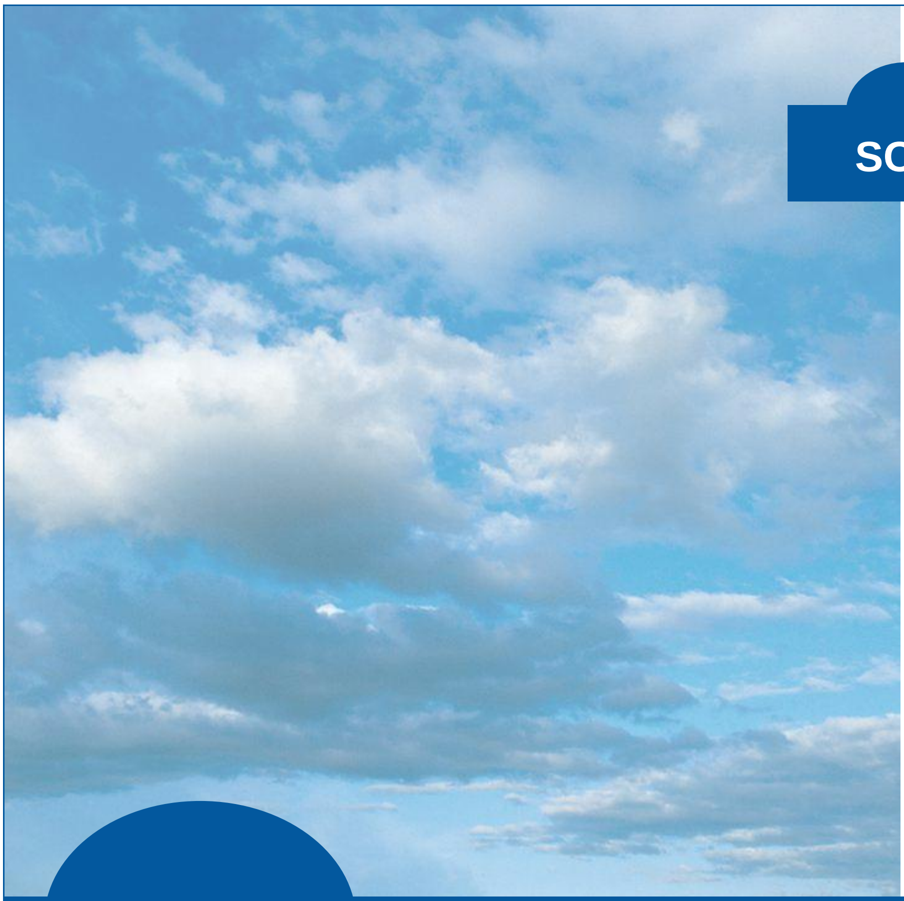

Hexachlorobenzene - Sources, environmental fate and risk characterisation

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Foreword

The Monitoring & Environmental Chemistry Working group (MECW) is a science group within Euro Chlor, the federation that represents the European chlor-alkali industry. The main objectives of the group are to identify both natural and anthropogenic sources of chlorinated substances, study their fate, gather information on the mechanisms of formation and degradation in the environment, and achieve a better knowledge of their persistence. The MECW often uses external specialists to assist in developing reports that review the state of existing knowledge of the different aspects mentioned above.

Dr Jonathan Barber is a Post Doctoral Research Associate at Lancaster University, UK. He has conducted research on persistent organic pollutants (POPs), including studying air-pollutant transfer kinetics, conducting air monitoring campaigns, developing new analytical methods for emerging chemicals and testing the genotoxic potential of chemicals at ‘low dose’ exposure levels using human cell cultures.

Dr Andrew Sweetman is a Post Doctoral Research Fellow also at Lancaster University. His recent research has involved the development and use of multi-media mathematical models for chemical fate and behaviour profiling and risk assessment on UK, European and global scales.

Kevin C. Jones is Professor of Environmental Chemistry and Ecotoxicology at Lancaster University. He leads a research group studying the environmental behaviour, fate and effects of organic pollutants and heavy metals in soils, inter-media pollutant transfers and the global cycling of persistent organic pollutants (POPs). His research also looks at long-term trends in environmental contamination. Prof. Jones has a special interest in chlorinated compounds, such as polychlorinated biphenyls (PCBs), polynuclear aromatic hydrocarbons (PAHs) and dioxins.

Regulated under several EU directives and even referred to as a banned substance in one of them, the behaviour and fate of hexachlorobenzene (HCB) is high on the political agenda. This Euro Chlor science dossier, *Hexachlorobenzene - Sources, environmental fate and risk characterisation*, identifies the sources of HCB pollution in the environment today. The paper looks at the chemical’s characteristics to explain the reasons for its distribution throughout the environment (in air, sea as well as soil), and why it is often found a long way from the point source. Also examined is the bioaccumulative behaviour of HCB and its presence in biota from fish to birds to human-beings.

This report argues that the increasing levels of HCB in the Arctic and the variation of HCB concentrations of background soils and sediments suggest that global presence of the substance is not evenly distributed as previously thought. On the overall decrease of HCB in the environment, the study concludes that levels will probably continue to decrease slowly in air and water, whereas levels in soil will diminish at a faster rate.

Summary

Hexachlorobenzene (HCB) was at one time used extensively as a seed dressing to prevent fungal disease on grains, but this use was discontinued in most countries in the 1970s. The amount of HCB currently used for this purpose is not known precisely. HCB continues to be released to the environment from several sources, including the use of some chlorinated pesticides, incomplete combustion, old disposal sites and as a by-product and waste material from manufacture of chlorinated solvents, chlorinated aromatics and chlorinated pesticides. Re-emission of ‘old’ HCB from soils is also thought to occur. It is believed that no single source dominates current emissions.

HCB is distributed throughout the environment because it is mobile and persistent, although slow photodegradation in air and microbial degradation in soil and sediment do occur. Low concentrations of HCB are present in ambient air (global average: 40 pg/m^3) and in water (tens of pg/l or less) in areas that are distant from point sources around the world. However, higher levels are still measured near point sources and in historically contaminated rivers. There is much evidence that HCB is subject to global re-distribution in the environment, and that cold condensation occurs.

HCB is bioaccumulative and has been detected in invertebrates, fish, reptiles, birds and mammals (including humans) distant from point sources, particularly in fatty tissues of organisms at higher trophic levels. The greatest losses of HCB in the environment probably occur from sediment and soil, with losses from water of less importance and losses from air insignificant. Levels of HCB in air and water will probably continue to decrease at a slow rate, whereas levels in soil will continue to decrease significantly.

1 Introduction

1.1 Aims and Scope of the Report

The overall objectives of Euro Chlor Science Dossiers are to:

- Identify the sources, both natural and anthropogenic, and fate of the substance being reviewed
- Derive an environmental budget, levels and possible trends in the environment.
- Discuss potential mechanisms of formation and degradation in the environment
- Evaluate the substance’s persistence in the environment and lifetime in the various environmental compartments.

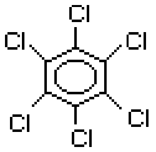
1.2 Legal Status

HCB is a priority substance in the UN-ECE Convention on Long Range Transport of Air Pollutants (CLRTAP) POPs protocol. HCB appears on List I of EU Council Directive 76/46/EEC, Annex 1A of the Third North Sea Conference, the OSPAR candidate list, the HELCOM priority list, and is referred to in Directive 76/769 EEC as a banned substance. HCB is regulated under several EU Directives: 76/769 on the marketing and use of certain dangerous substances; 79/117 which prohibits the marketing and use of certain dangerous plant protection products; 793/93 on the risk evaluation of existing chemicals; and 76/464, COMM(2000)47 and 99/61, all of which regulate discharges to the environment. It is also on the final list of 11 substances identified as "priority hazardous substances" under the proposed Water Framework Directive.

1.3 Background Information on HCB

1.3.1 Structure

HCB is a chlorinated aromatic hydrocarbon with the chemical formula C6Cl6. Its CAS registry number is 118-74-1.



1.3.2 Synonyms and Trade Names

HCB has been sold under the following synonyms and trade names: Amatin, Anticarie, Bunt-cure, Bunt-no-more, Co-op hexa, Granox NM, HCB, Julin's carbon chloride, No Bunt, No Bunt 40, No Bunt 80, No Bunt liquid, Pentachlorophenyl chloride, perchlorobenzene, Sanocide and Snieciotox.

1.3.3 Physicochemical Properties

Certain physical and chemical properties of HCB are listed in Table 1. At ambient temperature, HCB is a white crystalline solid that is virtually insoluble in water, but is soluble in ether, benzene and chloroform (NTP, 1994). It has a high octanol/water partition coefficient, low vapour pressure, moderate Henry's Law constant and low flammability. HCB is found almost exclusively in the gas phase (as is predicted by its vapour pressure), with less than 5% associated with particles in all seasons except winter, where levels are still less than 10% particle-bound (Cortes *et al*, 1998). HCB has a relatively high bioaccumulation potential because of moderately high lipophilicity (log $K_{OW} = 5.5$) and long half-life in biota (Niimi, 1987).

Table 1: Physical and chemical properties of hexachlorobenzene [a]

Property	**Value**
Relative molecular mass	284.79
Melting point (°C)	231
Boiling point (°C)	323-326 (sublimates)
Density (g/cm^3 at 20°C)	1.5691
Vapour pressure (Pa at 25°C)	0.0023
Log octanol/water partition coefficient	3.9-6.42 (5.5)
Water solubility (mg/litre at 25°C) (µg/l at 20 °C)	0.0062 58
Henry's Law Constant (calculated) [b] (Pa/mol per m^3)	131
Log octanol/carbon partition coefficient [c]	4.31
Conversion factors	1 ppm = 11.84 mg/m^3 1 mg/m^3 = 0.08 ppm

a From ATSDR (2002); Mackay *et al* (1992)
b The Henry's Law Constant has been calculated using the tabled values for aqueous solubility and vapour pressure
c Baker *et al* (2001)

The properties of HCB that are key to understanding its movement through the environment include vapour pressure, Henry’s law constant, and partition coefficients between octanol and environmental phases. All of these are strongly temperature dependent. Values for the three most important partition coefficients required for predictive modelling of HCB in the environment (octanol/water, air/water, and octanol/air partition coefficients), over a range of environmental temperatures, are listed in Table 2.

Table 2: Partition coefficients (octanol/water, air/water, octanol/air) of HCB at a range of environmental temperatures

Temperature (°C)	**Log K_{OW} [a]**	**Log K_{AW} [b]**	**Log K_{OA} [c]**
5	5.74	-2.47	7.83
15	5.06	-2.00	7.34
25	4.94	-1.64	6.88
35	4.79	-1.46	6.45

a Bahadur *et al* (1997)
b calculated from Tenulscher *et al* (1992)
c Harner and Mackay (1995)

2 Sources of HCB in the Environment

2.1 Industrial Production

Industrial synthesis of HCB is achieved through the chlorination of benzene at 150-200°C using a ferric chloride catalyst or from the distillation of residues from the production of perchloroethylene (Brooks and Hunt, 1984). HCB may also be synthesized by refluxing hexachlorocyclohexane (HCH) isomers with sulphuryl chloride or chlorosulphonic acid in the presence of a ferric chloride or aluminium catalyst (Brooks and Hunt, 1984). Technical grade HCB is available as a wettable powder, liquid and dust (NTP, 1994). Technical grade HCB contains about 98% HCB, 1.8% pentachlorobenzene and 0.2% 1,2,4,5-tetrachlorobenzene (IARC, 1979; IPCS, 1990), and it is known to contain a variety of impurities, including hepta- and octachlorodibenzofurans, octachlorodibenzo-*p*-dioxin and decachlorobiphenyl (Goldstein *et al*, 1978; IPCS, 1990).

HCB was introduced in 1945 in North America, and in 1948 in Canada when it was registered under the Pest Control Products Act. Estimates of global HCB production in the mid-1970s range from 1000 to 2000 tons (Courtney, 1979). Peak HCB production was in the late 1970s and early 1980s worldwide; production was about 10000 tons/year from 1978-1981 (Rippen and Frank, 1986).

Approximately 80% of HCB production in 1978 is believed to have been in Europe (Rippen and Frank, 1986). Production of HCB has declined as a result of restrictions on its use starting in the 1970s. In the US, 360000 kg/year was used around 1960 and an estimated 300 tonnes was produced by three manufacturers in the US in 1973 (IARC, 1979). US production in 1977 was 454 tonnes (SMOC Mexico, 1998). HCB was produced at a maximum rate of 3500 tonnes/year in Mexico in the 1970s, with a total of 39000 tonnes produced between 1970 and the end of production in 1991 (SMOC Mexico, 1998). The majority of this HCB was used within Mexico for agricultural purposes, with significant use continuing right up until it was banned in 1992 (SMOC Mexico, 1998).

Approximately 1500 tonnes of HCB were manufactured annually in Germany for the production of the rubber auxiliary, PCTP (IPCS, 1997), but this production was discontinued in 1993. In Germany HCB was produced at rates of approximately 4000 tonnes/year in 1974 and 2600 tonnes/year in 1976 (Rippen, 1989). No further centres of HCB manufacture in Europe or North America have been identified. In the Czech Republic, HCB production was terminated in Spolana Neratovice in 1968. India produced 42612 tonnes of technical grade HCB during 1995-97 (Ministry of Chemicals and Fertilisers, 2000). About 15390 tonnes of HCB were imported by Pakistan during 1970-92 and 12162 tonnes was used (1979-88). Bhutan reported stockpiles of insignificant quantity of HCB (approximately 118 kg) whereas 7.9 tonnes of HCB and 22.5 tonnes of mixed aldrin and HCB were reported from Nepal (UNEP, 2002c).

The amount released into the environment worldwide in the mid-1990s was estimated at 10-90 tons/year (Bailey, 2001).

2.2 HCB Uses

Historically, HCB had many uses in industry and agriculture. HCB was first introduced in 1945 as a fungicide for the seeds of onions, sorghum and crops such as wheat, barley, oats and rye. HCB was used extensively as a fungicide to control Bunt (*Tilletia caries*, *T. tritici* and *T. foetida*) in wheat, representing a major breakthrough for this disease.

HCB was applied as a dust that contained 10-40% active ingredient (FAO/WHO, 1970), and reported application rates range from 0.2oz of HCB per bushel of seed (US EPA, 1991) to 1-3oz of 30% HCB dust per bushel (330-990 ppm on the seed) (FAO/WHO, 1970). There is little information available about the amount of HCB that was used for this purpose, however. HCB was used briefly as a fungicide in small quantities in Australia and New Zealand in the 1960s and 1970s. In the 1960s in Australia, 12 million bushels of seed wheat were treated annually with HCB dust, requiring 200 tons of technical HCB (FAO/WHO, 1970). A smaller proportion of the total crop was probably treated in the US, Canada, UK and some other European countries, but there was extensive use in Turkey, Italy, Spain, Netherlands, Germany, France and some Eastern European countries (FAO/WHO, 1970). It has been reported that a company in Spain produced an estimated 150 tonnes of HCB annually (IARC, 1979), and this resulted in particularly high levels being detected in that area (Grimalt *et al*, 1994; see Section 5.2). Approximately 610 tonnes were used in the former USSR until HCB was banned for agricultural purposes in 1986. HCB was used in Canada as a seed dressing for several crops from 1948 to 1972.

Following the first evaluation of HCB at the 1969 FAO/WHO Joint Meeting, it was recommended that a suitable substitute be found for HCB as a seed fungicide (FAO/WHO, 1970). The use of HCB in such applications was discontinued in many countries in the 1970s because health concerns about its toxicity led to cancellation of the registrations of all pesticides that contained it as an active ingredient (US EPA, 2002).

In the US, the last registered use of the compound as a pesticide was voluntarily cancelled in 1984, and evidence indicates that it has not been commercially produced since the late 1970s. HCB was used in seed dressings in East Germany until 1984 (Manz *et al*, 2001). HCB was used as a pesticide in some countries; for example, HCB was still used in 1986 as a fungicide, seed-dressing and scabicide in sheep in Tunisia (Jemaa *et al*, 1986), and use of the material as a fungicide was terminated in Mexico in 1991 (SMOC, 1998). It is uncertain as to whether HCB is still used as a fungicide, but any such use would be at very low levels.

Historically, HCB has also been used as a wood-preserving agent, and in industry as a porosity-control agent in the manufacture of graphite anodes, as a peptizing agent in the production of nitroso and styrene rubber for tyres (Mumma and Lawless, 1975), in the production of pyrotechnics and tracer bullets for the US and Russian military (Shekhovtsov, 2002), as a fluxing agent in the manufacture of aluminium, and as a chemical intermediate in dye manufacturing (ATSDR, 2002). Quinlivan *et al* (1975) believed that use of HCB in pyrotechnics diminished in the early 1970s, and it is likely that some of the other applications have been discontinued, although little information is available.

There are no current commercial uses of HCB as an end product in the US (ATSDR, 2002), whereas in the Russian Federation, HCB is still used in pyrotechnical compounds (Shekhovtsov, 2002). In Canada, HCB has not been used commercially since 1972.

2.3 By-Product Emissions

Although HCB production has ceased in most countries, it is still being generated inadvertently as a by-product and/or impurity in several chemical processes, such as the manufacture of chlorinated solvents, chlorinated aromatics and pesticides (Jacoff *et al*, 1986). Past methods of disposal of these HCB wastes have included landfill, discharge to municipal sewage treatment plants, and incineration. For example, about 0.9 tonnes of HCB were released into the atmosphere in the UK in 1998, mostly from chlorothalonil use, plus manufacture of chlorinated solvents and secondary aluminium processing which can release HCB from the hexachloroethane used as a de-gassing agent (UK Environment Agency, 2004).

2.3.1 Chlorinated Solvent Production

Substantial quantities of HCB are contained in the wastes generated through the manufacture of chlorinated solvents. HCB is formed as a reaction by-product of thermal chlorination, oxychlorination, and pyrolysis operations in the manufacture of chlorinated solvents (mainly perchloroethylene (PCE), trichloroethylene (TCE), and carbon tetrachloride, Government of Canada, 1993). Although HCB is separated by distillation of the solvents, traces may remain. Manufacture of vinyl chloride monomer and volatile hydrocarbons is also known to produce HCB as a by-product. Separated HCB may be found in the distillation bottom fractions. The concentration of HCB in distillation bottoms in the 1980s was estimated to be 25%, 15% and 5%, respectively, for perchloroethylene, carbon tetrachloride and trichloroethylene (Jacoff *et al*, 1986).

Jacoff *et al* (1986) estimated that in the US from 1980 to 1983, approximately 4130 tonnes of HCB were generated annually as a waste product, and nearly 77% (3178 tonnes/year) of this was produced from the manufacture of PCE, TCE, and carbon tetrachloride. The remainder was produced by the chlorinated pesticide industry, of which only about 0.0065 tonnes/year was from the manufacture of chlorobenzene (Jacoff *et al*, 1986). Rippen and Frank (1986) estimated that more than 5000 tonnes of HCB were produced in Germany as a by-product of solvent manufacture in 1980.

Importantly, only a small fraction of the HCB generated as a by-product is released into the environment, depending on the process technology and waste-disposal practices employed. In the 1970s and 1980s, 60% and 81-99% respectively of by-product waste was incinerated, with the remainder going to landfill (Jacoff *et al*, 1986). It is likely that the proportion of HCB wastes disposed of by incineration has since increased further.

Landfilling was favoured in the 1970s because it was cheap and convenient, and the potential for HCB to leach into groundwater was not then fully understood (see Section 2.6). Incineration subsequently became the favoured disposal technique because it destroys almost all the HCB in the waste, and scrubbers collect the majority of the remainder (Mumma and Lawless, 1975). The proportion of HCB decomposed by thermal incineration is reported to be more than 99.97-99.99% in the US (Jacoff *et al*, 1986, Quinlivan *et al*, 1975). High temperature incineration (around 1300ºC) with a retention time of approximately 0.25 seconds is the recommended disposal technique because it is reported to destroy more than 99% of the chemical (IRPTC, 1985).

In 1977, about 300 tonnes of HCB were generated in Japan as a waste by-product in the production of perchloroethylene, almost all of which was incinerated (IARC, 1979). It was estimated that >5000 tonnes HCB/year were produced as a by-product during perchloroethylene production in the Federal Republic of Germany in 1980 (Rippen and Frank, 1986). It was estimated for the Czech Republic and Slovakia that 3-6 tonnes of HCB was released to the atmosphere in 1992, and 80% of this was from the incineration of PCE, TCE, and PVC (Vulykh and Putilina, 2000). Since these reports were published, HCB levels in products such as pesticides and solvents have decreased dramatically. When measurable, they are orders of magnitude below regulatory levels. While HCB could potentially also be a contaminant in the final product, it was not detected at a detection limit of 5 mg/litre in carbon tetrachloride or perchloroethylene in a study in Canada (UNEP, 1997). Analysis of production lots of tri- and perchloroethylene produced in Europe in 1996 failed to detect HCB at a detection limit of 2 µg/litre solvent (UNEP, 1997).

For over two decades, the chemical company Orica Australia Pty Ltd (formerly ICI Australia), produced a large quantity of HCB industrial waste as an unwanted by-product of the manufacture of chemical solvents (OZTOXICS, 2001). The chlorinated solvents, carbon tetrachloride and perchloroethylene were manufactured at Botany, Australia, from 1964 to 1991. This involved the reaction of propylene with chlorine, which produced both the desired commercial products and a range of unwanted organochlorine chemical residues. These residues were called the "heavy ends". From 1964 until 1977, the heavy ends were placed into drums and stored; in 1977 the Heavy Ends Treatment Plant was commissioned to reduce the amount of waste needing long-term storage. In this plant, HCB was crystallised and then separated from the liquid heavy end components, principally hexachlorobutadiene (HCBD) and hexachloroethane (HCE). The HCB crystals were then stored in drums while the HCBD and HCE were recycled back into the solvent production process. The drummed HCB waste from chlorinated solvent manufacture makes up the major component of the HCB waste at Botany. Orica holds the world’s largest stockpile of HCB in Botany: around 10500 tonnes of HCB waste, including 8300 tonnes of crystalline HCB waste stored in up to 60000 barrels. A further 1000 tonnes of HCB waste from the manufacture of ethylene dichloride (EDC) is kept in storage tanks. This material comprises solvent-laden low-grade polymers that have settled and partially polymerised during extended storage in concrete tanks. Orica is required under a Federal management plan to destroy the HCB stockpile. Orica proposes to use Geomelt technology, which involves putting the HCB waste mixed with soil into a crucible and applying high voltage to the mixture. This causes the mixture to melt. The HCB in the melt would be destroyed or vaporised and after the melt cools, a black, glass-like substance is left. The Geomelt process uses thermal oxidation (incineration) to treat any waste that is vaporised and not destroyed in the melt.

Emission factor estimates for chlorinated solvent production have been calculated (see Table 5). Using these factors, it has been estimated that 1-24g of HCB was emitted for each tonne of chlorinated solvents produced, equivalent to a 99.9% decomposition rate during incineration. About 300000 tonnes of solvents were used in North America in 1992. Thus, less than 1 kg of HCB is estimated to have been released annually from the use of chlorinated solvents in North America. Recent estimates for Europe from the European Chlorinated Solvent Association (ECSA, P.G. Johnson (1996) personal communication to IPCS) indicate that up to 4000 tonnes/year of HCB were made as a by-product during certain perchloroethylene production processes and that over 99% of this by- product was incinerated at high temperatures.

According to the US Toxic Chemical Release Inventory (TRI), releases of HCB from the ten largest processing facilities in 1990 were 460 kg, most of this to air, compared with almost 542 tonnes transferred offsite as waste. The TRI data are not comprehensive, since only certain types of facilities are required to report (ATSDR, 2002). In the 1996 NTI (National Toxics Inventory), emissions from the production of industrial organic chemicals represented 48% (2100 kg) of the total HCB emissions (US EPA, 2000). US industrial emissions of HCB inventoried by the TRI for the 2000 reporting year are as follows (US EPA, 2002): Total on-site releases were 11 tonnes (of these, releases to air were 650 kg, releases to water were 150 kg) and total off-site releases (transfers to disposal) were 5.9 tonnes. Total on- and off-site releases were 17 tonnes, of which approximately 800 kg could be expected to be transported within the environment.

Canadian industrial HCB emissions were inventoried by the NPRI for the 2000 reporting year and are available through the Query Site (NPRI Query, 2002)). Total on-site releases were 37.8 kg, transfers for disposal 10.45 kg, and transfers for recycling 0.25 kg.

2.3.2 Pesticide By-Product

Historically, HCB has been found as an impurity in several chlorinated pesticides, due to its formation as a by-product during the production process. Most current applications of such pesticides are dispersive, i.e. they are applied on or near the surface of the soil. HCB from this source will therefore be released to the environment, with between 8-80% of applied pesticides volatilising to the air (Nash and Gish, 1989). The waste streams from the production of pentachloronitrobenzene (PCNB), chlorothalonil (TPN), trimethyl 2,3,5,6-terephthalate (TCTP), picloram and dacthal contributed the bulk of HCB released from the pesticide industry in the 1970s and 1980s (Brooks and Hunt, 1984).

HCB was also generated as a waste by-product from the production of pentachlorophenol (PCP), atrazine, simazine, propazine and maleic hydrazide (Quinlivan *et al*, 1975; Mumma and Lawless, 1975). These pesticides contained HCB as an impurity in the final product, usually at levels of much less than 1% when appropriate procedures were used for the synthesis and purification stages (Tobin, 1986). When such procedures were not followed, the level of HCB could be much higher (for example, PCNB was reported to contain 1.8-11% HCB (Tobin, 1986)). Some examples of HCB concentrations in pesticides are shown in Table 3. In Japan, HCB was never registered as an agricultural chemical, but during the period from 1952-1972, 70 000 tonnes of HCB was produced as a raw material for PCP, and this was probably the major source of HCB in that country.

Table 3: HCB content rates in pesticides (UNEP, 2002c)

PCP	PCNB	TCTP	TPN	Picloram	Reference
-	-	10-14%	-	-	Wapensky (1969)
-	-	9% (1973) 8% (1974)	-	-	Burns *et al* (1974)
-	-	0.3% (1972)	-	-	Mumma *et al* (1975)
ND	0.5-2.0% (average 1.0%)	0.1–11% (average 7.7%)	ND	-	Saito *et al* (1976)
0.04%	-	-	-	-	Schewetz *et al* (1978)
-	1.8–11%	-	-	-	Sittig (1980)
-	0.1%	-	-	-	Nishimura *et al* (1980)
-	0.5% (1983) 0.1% (1988)	-	-	-	US EPA (1992)
<0.005%					ATSDR (2002)
	<500 ppm (0.05%)		18-26 ppm	8-50 ppm	Benazon (1999)
-	1.5%	-	-	-	FAO/WHO (1975)
-	<0.1%	-	-	-	FAO/WHO (1995)
0.4%	0.7%	-	-	-	Ando *et al* (1984)
Average 0.01%	<0.05%	<0.3%	<0.05%	<0.02%	Tobin (1986)
-	0.05%	0.07-0.3%	0.0018-0.0026%	-	Benazon (1999)

Owing to many voluntary and regulatory pressures, it is unlikely that such high levels of HCB are present in today's pesticide formulations. HCB levels in products such as pesticides have decreased dramatically as manufacturers have made process and raw material changes. For example, the maximum HCB concentration in PCNB was reported to be 27,000 ppm in a 1971 study, and 10,000 ppm in 1976. The HCB concentration in PCNB had dropped to 5000 ppm by 1983, 1000 ppm by 1988 and is presently less than 500 ppm. In the US, the EPA regulates the maximum concentration of HCB present in the pesticides as shown in Table 4.

Table 4: Regulatory levels for HCB as a contaminant in pesticides, and concentration used for emission factor estimates by Bailey (2001)

Product	Maximum HCB concentration	HCB concentration used by Bailey (2001)
Lindane		50 ppm
Dacthal (DCPA)	3000 ppm	1000 ppm
Pentachlorophenol	75 ppm	50 ppm
Atrazine	40 ppm	1 ppm
Simazine	40 ppm	1 ppm
Picloram	50 ppm	50 ppm
Pentachloronitrobenzene (PCNB)	500 ppm	500 ppm
Chlorothalonil	40 ppm	40 ppm

Current manufacturing methods produce materials with average HCB concentrations that are orders of magnitude below regulatory levels and may not even be detectable (Benazon, 1999). Regulatory levels should therefore not be used to calculate HCB release quantities from pesticide use. HCB may currently be found as an impurity during manufacture of the pesticides ametryn, atrazine, cyanazine, dacthal, dienochlor, dipropetryn, lindane, maleic hydrazide, mirex, PCP, picloram, prometon, prometryn, propazine, simazine and terbutryn (ATDSR, 2002). Bailey (2001) estimated the concentrations in the various pesticides in order to calculate global HCB emissions (see Table 4). By multiplying the concentrations by the usage of each pesticide, it was estimated that in the 1990s, 1270 kg/year of HCB entered the environment in the US, 580 kg/y in the UK (UK National Env. Tech. Centre, 2000), and 6463 kg/year worldwide (Bailey, 2001).

China and Russia still manufacture PCP from HCB by caustic soda hydrolysis, which utilises the waste α-HCH from lindane manufacture. This is believed to be the only current direct use of HCB as a chemical intermediate (Bailey, 2001). China has been listed as an exemption from the Stockholm Convention for production and use of HCB as an intermediate, and HCB is still being produced in a large chemical factory near Ya-Er Lake, in Hubei province, NE China (Kunisue *et al*, 2004).

2.4 Auxiliary Activity Releases

Small amounts of HCB are released from many processes where both carbon and chlorine are present at high temperatures. Thus, nearly all combustion processes and some metal processes emit small amounts of HCB. The measurement of very low emissions is difficult and emissions vary widely depending on many factors so that it is not possible to develop precise emission estimates at present. Production of these small quantities of HCB is not easily avoided since both carbon and chlorine are natural components of the environment. HCB has been reported in emissions from a number of industries, including paint manufacturers, coal and steel producers, pulp and paper mills, textile mills, pyrotechnics producers, aluminium smelters, soap producers and wood-preservation facilities (Quinlivan *et al*, 1975; Gilbertson, 1979; Alves and Chevalier, 1980), probably reflecting the use of products contaminated with HCB. Municipal and industrial wastewater facilities may also discharge HCB-contaminated effluents (King and Sherbin, 1986), due to inputs from industrial sources. HCB may also be emitted as a result of the use of HCB-containing ferric chloride in water treatment operations, for odour control and to facilitate settling of particles in the water (Benazon, 1999). The estimated emission factors for some of these processes are listed in Table 5.

Table 5: Emission factors for HCB

Sources of HCB emission to the atmosphere	**Emission factor**		
	Vulykh and Putilina (2000) (in µg/t)	Bailey (2001) (in µg/t)	Pacyna *et al* (2003) (in µg/t)
Combustion of organic fuel			
Coal	16	1.3-130	16
Wood	60	6-600	60
Diesel fuel			21 ng/vehicle/km
Leaded gasoline			0.87 ng/vehicle/km
Unleaded gasoline			0.024 ng/vehicle/km
Industrial processes			
Production of chlorinated hydrocarbons:		1 x 106	
Tetrachloromethane	(1-20) x 106		(1-20) x 106
Trichloroethane	(1-6) x 106		(1-6) x 106
Perchloroethane	(3-10) x 106		(3-10) x 106
Cement		17-1700	
Metals			
Iron sintering		150-15000	
Copper smelting	3.9 x 104	3.9x103-3.9x105	3.9 x 104
Magnesium smelting			
Secondary production of aluminium	5 x 106	8.6x108-4.3x109	

Sources of HCB emission to the atmosphere	**Emission factor**		
	Vulykh and Putilina (2000) (in µg/t)	Bailey (2001) (in µg/t)	Pacyna *et al* (2003) (in µg/t)
Waste incineration			
Solid municipal		2.9x103-2.9x105	10 x 103
Medical	19 x 103	2.9 x 103	19 x 103
Sewage sludge	5 x 106	470-47000	5 x 105
Industrial wastes:		1.9x103-1.9x105	
Tetrachloromethane	8 x 103		
Trichloroethylene	3 x 103		
Perchloroethylene	6 x 106		
Polyvinylchloride	5 x 106		
Agriculture			
Used as fungicide	5 x 1011		5 x 1011
Pesticide use		See Table 4	50-150 mg/ha/year
PCP use (wood protection)		50 x 106	10-30 mg/inhab/year

2.4.1 The Metals Industry

HCB is emitted to the atmosphere in flue gases generated by metallurgical industries (Bailey, 2001). A potential source of HCB is the electrolytic production of aluminium and magnesium. The process of degassing of molten aluminium with hexachloroethane (HCE) gas, to remove hydrogen gas prior to casting, occurs in some aluminium smelters (van der Most and Veldt, 1992). However, HCE gas can form HCB in the smelter, which may be released, with an emission factor of 5.2mg-HCB/tonne-aluminium (Westberg *et al*, 1997). Few, if any, aluminium smelters currently use the hexachloroethane degassing process, limiting this potential source of HCB release. The smelting of magnesium is also believed to produce HCB, by the following three processes: i) conversion of MgO/MgCO$_3$ to MgCl$_2$ by heating with coke in a pure Cl$_2$ atmosphere; ii) purification of molten MgCl$_2$ by transformation of residual MgO using gaseous HCl, mixed with graphite blades; and iii) electrolysis of molten MgCl$_2$ using anodes and bipolar electrodes made from graphite. There is an overall emission factor of 2.5-3.5g-HCB/tonne-Mg (Deutscher and Cathro, 2001).

The smelting and refining of secondary copper has also been suggested as a source of HCB. Aluminium plasma etching is performed in the production of microchips and other electronic components in the semiconductor industry. Under production conditions, the wastes from aluminium plasma etching using chlorine-containing etchants were found to contain HCB, among other chlorinated organic compounds (Schmidt *et al*, 1995). Potential HCB release from this process has not been evaluated (US EPA, 2000).
Métallurgie Magnola established a plant in 2000 that uses asbestos tailings for magnesium extraction in Québec. The Magnola plant transforms large quantities of magnesium-rich asbestos mining tailings (serpentine), into magnesium chloride and thence into magnesium metal (58000 tonnes/year) by electrolysis at about 650ºC (Bramley, 1998). The process creates a variety of POPs, including HCB, dioxins and PCBs, and a portion of the resulting POPs will be emitted to the atmosphere directly from the plant (notably 21.4 kg of HCB annually; Vachon, 1998). Much larger quantities of HCB will be dumped in the tailings lagoon; and filtered out and sent off-site, probably for incineration at Swan Hills, Alberta. Four experts consulted by the BAPE (Vachon, 1998) believe that the decanting pool will not have the required organic matter to neutralize the pollutants so that all the HCB will evaporate into the atmosphere. It has been estimated that 53.8 kg of HCB will be emitted from the tailings pond (Case, 2000), giving a total of 75.2 kg/year emissions for the plant. A fate model simulation indicates that Magnola's will increase the regional ambient HCB concentration by about 2%, which will cause a 2% increase in biota HCB concentrations (Case, 2000).

2.4.2 The Chlor-Alkali Industry

The chlor-alkali industry produces chlorine (Cl_2), hydrogen and caustic soda (NaOH) by electrolysis of purified and concentrated sodium chloride (NaCl). Processes using graphite anodes are known to produce HCB as a by-product (Quinlivan *et al*, 1975; Mumma and Lawless, 1975; Alves and Chevalier, 1980) owing to the reaction of chlorine with graphite anode materials such as carbon and oils. Depending on the purification procedures, the final products might also be contaminated with HCB. In most countries since the early 1970s, graphite anodes have been replaced by dimensionally stabilised anodes (DSA), which do not generate HCB (Brooks and Hunt, 1984). Although current HCB production from this source is negligible, unknown quantities of HCB may continue to be released from sites where wastes from these industries were landfilled.

2.4.3 Pulp and Paper Mills

Conditions in pulp and paper mills are not thought to favour production of HCB, contrary to previous reports (NCASI). The only indication of HCB release was contained in a single report from the Ontario Ministry of Energy and Environment, based on Municipal/Industrial Strategy for Abatement (MISA) monitoring data. Upon investigation, NCASI learned that the preliminary data upon which the estimates were based had been excluded from the MISA database after QA/QC review found sufficient irregularity in them to make it very doubtful that the mills were actually emitting the compounds.

2.5 Combustion-Related Sources

HCB can be emitted from incinerators as a result of incomplete thermal decomposition of HCB-contaminated industrial wastes, and incomplete thermal decomposition of a variety of chlorinated organics such as Kepone, mirex, chlorobenzenes, PCBs, PCP, PVC and mixtures of chlorinated solvents (Ahling *et al*, 1978; Dellinger *et al*, 1991). Formation of HCB has also been reported from municipal waste, medical waste and sewage sludge incinerators, as well as by cement and aggregate kilns (Benazon, 1999; Cohen *et al*, 1985). Historical improvements in combustion practices, designed to reduce emissions of all products of incomplete combustion (such as dioxins and furans) have also reduced HCB emissions because of similarities in formation mechanisms. Regulations based on ‘maximum achievable control technology’ (MACT) have been established to control hazardous air pollutant emissions from incinerators, medical waste incinerators, cement kilns, and lightweight aggregate kilns that burn hazardous wastes. However, the database of HCB emission factors is not complete enough to quantify the emission reductions.

2.5.1 Incineration

Incineration is an important source of HCB in the environment. HCB has been detected in emissions from the combustion of coal and hazardous waste. For example, total HCB releases from municipal incinerators in the US were roughly estimated to be between 310 and 977 kg/year in the early 1980s (Brooks and Hunt, 1984), and the total amount of HCB released from incineration in East Germany in 1982 was estimated to be between 0.8-4.4 tonnes (Rippen and Frank, 1986). The current practice of making emission estimates involves calculating them using measured emission factors, and the amount of waste burnt annually. Emission factors determined in laboratory experiments and by stack monitoring vary widely (Bailey, 2001; Kaba *et al*, 1992). Emission levels from incinerators are very site-specific, and therefore typical levels are difficult to estimate.

HCB emissions from incineration facilities vary widely depending on the type of furnace, collection method and incineration temperature; the measured values of HCB concentrations in flue gas range from 0.13 $\mu g/m^3$ to a few hundred $\mu g/m^3$. Kaba *et al* (1992) carried out research into HCB emission routes (see Table 6). The most important emission route was concluded to be flue gas, accounting for 62-97.9%, followed by 1.9-38% (42.2-2020 ng/g dw) in fly-ash, 0-2% (ND-28.7 ng/g dw) in incinerator ash, with negligible amounts found in the cooling water for the flue gas. Although the range of estimated HCB emission factors was wide, most were within 1-100 mg/t-waste. Multiplying these emission factors by 40 million tonnes/year, which was the annual amount of municipal waste incinerated in the 1990s in Japan, the total HCB emissions were estimated to be 40-4000 kg-HCB/year. HCB emissions originating from municipal solid waste incineration were estimated at 0.56-56 tonnes/year worldwide in the 1990s (Bailey, 2001).

Table 6: HCB emission factors derived from municipal solid waste incineration in Japan (Kaba *et al*, 1992)

HCB concentration in flue gas (µg/Nm^3)			HCB emission factor (mg/t-waste)		
Minimum	Maximum	Average	Minimum	Maximum	Average
0.03	3.2	1.3	1.0	51	19
0.16	12	2.9	0.80	90	24
0.36	14	7.9	3.6	200	76
0.05	0.09	0.063	0.32	0.61	0.44
0.04	3.5	1.6	0.29	25	12
0.82	110	38	5.7	770	270
6	110	59	90	1700	880
	-	11	-	-	77
-	-	0.11	-	-	0.77

2.5.2 Cement Production

Another controlled combustion process is cement production. Using data from a survey conducted by the Canadian Portland Cement Association, the HCB emission factor in cement production was estimated to be 0.17 mg-HCB/t-cement (Bailey, 2001). As a typical example, the production of cement in Japan has been 70 million-90 million tonnes/year since the late 1970s; thus, annual HCB emissions from cement production in Japan are approximately 10 kg of HCB.

2.5.3 Uncontrolled Combustion

Uncontrolled combustion processes, such as fires and open burning, increase the amount of HCB released to the environment. The global distribution of ‘dirty’ combustion processes will approximately mirror that of the population distribution, and this is greater in the northern hemisphere. Data on HCB emissions from combustion is quite variable. With a simulation of open burning in steel drums, Lemieux (1999) estimated HCB emissions to be 22-48 mg-HCB/t-waste, which is two orders of magnitude higher than the current emission factor for waste incineration. Gullett *et al* (2003) measured emission factors for HCB from residential fireplaces and woodstove appliances in San Francisco. Considerable differences were noted between appliance and fuel type (see Table 7), with values ranging from 13-990 µg-HCB/t-waste. Gullett *et al* (2003) estimated that annual HCB emissions from forest fires range from 0.5-12.1 kg, which is an insignificant amount in comparison with other emission sources.

Table 7: HCB emission factors for uncontrolled combustion (Gullett *et al*, 2003)

Wood	Appliance	Emission Factor (ng/kg fuel)
Oak	Woodstove	13
Oak	Fireplace	310
Pine	Fireplace	380
Artificial Log	Fireplace	990

2.6 Leaks from Landfill Sites and Stockpiled Pesticides

Although only a small proportion of the HCB-containing waste generated is currently landfilled, HCB may continue to leach to groundwater from previously landfilled HCB waste sites. The contribution of this route is uncertain, although HCB is not easily leached, and landfills containing HCB are now designed to prevent leachate losses into adjacent water systems (Brooks and Hunt, 1984). HCB emission into the atmosphere from landfills containing HCB wastes occurs from slow volatilisation and from displacement of the contaminated soil (Brooks and Hunt, 1984). If not properly managed, landfills are a potential source of HCB to the environment. This has been most apparent in the HCB releases to the St. Clair River from the Dow Chemical landfill in Sarnia, Ontario. HCB was found at concentrations as high as 400 mg/kg in the monitoring well of an uncontrolled hazardous waste site (Davis and Morgan, 1986). Soil and sediment samples from nearby areas were recorded at 20 mg/kg and 39.5 mg/kg respectively, indicating that significant leaching had occurred. Soil and sediment collected from another disposal site near Sorrento, Louisiana contained 62 mg/kg and 130 mg/kg, respectively (Laseter *et al*, 1976). Concentrations of 20 mg/kg and 53.1 mg/kg were recorded in soil from disposal sites at Crystal City, Texas, and Geismar, Louisiana, respectively.

There are also well known, highly contaminated sites and stockpiles that may be a significant environmental source of HCB in Brazil, such as Cubatão, where HCB was probably generated from perchloroethylene production and burning of other chlorinated residues (CETESB, 2001). do Nascimento *et al* (2004) recently investigated the levels of HCB in soil and water samples close to one of these disposal sites. A plume of HCB was detected in the vicinity of the dump, up to 500m away. Concentrations in the soil varied over six orders of magnitude from <200 pg/g in unaffected soil to 81 μg/g in ‘hotspots’ close to the disposal site.

The internationally suggested maximum limit for HCB in soil is 0.7 ng/g (US EPA-RCRA, 1987), and this level was exceeded in almost 50% of the samples analysed. HCB was also detected in water samples at concentrations ranging from <2-312 ng/l. Clear evidence was found that HCB migrates through the soil horizons, with the pollutant distribution pattern indicating travel in the direction of water flow. The dispersal of HCB was probably a result of complexation and transport with dissolved organic matter in the water moving through the soil (do Nascimento *et al*, 2004). The concentration of HCB in the deeper groundwater was below the detection limit, which suggests that the groundwater was not polluted.

2.7 Metabolism of Lindane

There have been reports that lindane (γ-HCH) is biologically converted into HCB in mammals (Gopalaswamy and Aiyar, 1986). After oral administration of lindane to rats, HCB was one of the metabolites reported in tissues (Seidler *et al*, 1975). There have also been reports of HCH conversion to HCB in plants (Kohli *et al*, 1976; Engst *et al*, 1977; Steinwandter and Schulter, 1978). Although HCH has been used much more recently than HCB and in large quantities (Li *et al*, 1998), this is unlikely to be a significant source of HCB to the environment, due to the slow rates of metabolic processing of chlorinated compounds.

2.8 Secondary Sources

A substantial portion of HCB measured in the atmosphere is thought to come from volatilisation of ‘old’ HCB in the soil from past contamination (Bailey, 2001). Volatilisation of HCB from the soil is a function of organic content, temperature and exposure to the atmosphere (Beall, 1976; Nash and Gish, 1989; Wang and Jones, 1994a). Wang and Jones (1994b) showed that 5.9-21% of HCB in soil was lost after 100 days, and similarly Nash and Gish (1989) showed that 8-80% of soil-applied HCB volatilised to the atmosphere. Beall (1976) studied the persistence of HCB aerially applied to grass and soil. Following application to the soil, there was rapid loss by volatilisation from the surface of the soil. HCB concentrations in the top 2 cm of soil found after 0.5, 1, 6.5, 13 and 19 months were 45.2, 24.4, 7.9, 4.7 and 3.4% of day 1 values, respectively. However, within the bulk of the soil, HCB was found at approximately 2% of the initial top 2 cm value, and was extremely persistent, with no significant change in the 2-4 cm layer after 19 months. There was an extremely rapid rate of loss of 89.5% of applied HCB from grass surfaces within the first two weeks, but after that there was a steadier decline in the rate of loss with less than 0.01% of the initial concentration detectable after 19 months. Franke *et al* (1996) estimated that emissions of HCB in Germany from soil were 10-50 tonnes/year, compared to estimated current primary emissions of only 200 kg/year.

Meijer *et al* (2003d) reported an estimated global background soil content of 1020 tonnes (see Section 4.5). Volatilisation of this amount of HCB is expected to be a first order process with a half-life of several years. As this historical HCB volatilises from soil, the atmospheric concentration of HCB should decrease to a much lower concentration in equilibrium with current atmospheric emissions. If the half-life for volatilisation of HCB adsorbed and absorbed onto soil organic matter is 20 years, about 3.45% of the HCB would be volatilised each year or 0.0345 year^{-1}*1020 t = 35 tonnes/year. Because the reservoir is becoming more dilute each year, the atmospheric concentration would be expected to decrease by 3-4%/year. If the half-life for volatilisation is only 10 years, then the amount volatilised would be greater, and the rate of decrease would be correspondingly higher.

2.9 Historical Changes in Importance of Emission Sources

It is believed that agricultural use of HCB dominated its emissions during the 1950s and 1960s. At its peak, thousands of tonnes of HCB were used each year. Unfortunately, details of the global production or usage pattern is not well understood for this peak period, and the amount used as a fungicide is unknown. Table 8 shows the available information on production and emission of HCB. Today, however, HCB is no longer used in agriculture. Its use as a fungicide was banned in most countries in the 1970s and 1980s (see Table 9). This has removed the largest single primary source of HCB in the environment, and therefore emissions of HCB fell sharply in the 1980s, followed by a steadier decline throughout the 1990s (see Table 11). Current emissions are 70-95% lower than in 1970. UK HCB emission estimates have decreased from 1.3 tonnes/year in 1990 to 0.3 tonnes/year in 2002 (DEFRA, 2004). However, over the same period, emissions in France remained relatively constant (CITEPA, 2004).

As discussed above, HCB is a by-product of the manufacture of industrial chemicals including carbon tetrachloride, PCE, TCE and PCBs, but this occurs much less in Europe and North America, due to changes in production processes and improvements in disposal practices. According to Pacyna *et al* (2003), the most important sources of HCB in European countries in 1993-95 were: i) application of contaminated pesticides (80%); ii) solvents and other product use (13%); iii) emission from other production processes (7%); and iv) waste treatment and disposal (<1%). In the UK, current HCB emissions are dominated by application of contaminated pesticides, with chlorothalonil and chlorthal-dimethyl use responsible for 92% of total emissions (Dore *et al*, 2003). The relative contribution of various sources to total emissions in Europe, the US, Canada and globally is shown in Table 10.

Currently, the principal sources of HCB to the Canadian environment are estimated to be application of HCB-contaminated pesticides, by-products from the manufacture and use of chlorinated solvents, waste incineration (including municipal solid waste) and long-range transport from other countries. It was estimated that long-range transport and total deposition of HCB to the Canadian environment totals approximately 510 kg/year, an amount similar to that from all other sources combined (Government of Canada, 1993). Emissions from chlorinated solvent production and pesticide application are the most significant sources in the UK and in 1996 were estimated to account for around 47% and 45%, respectively, of total emissions (UK National Env. Tech. Centre, 2000). Outside of Europe and North America, the main source of release of HCB into the environment is currently reported to be as a by-product of certain industrial processes, such as aluminium smelting, with some countries still using older technologies for production of PCE and vinyl chloride monomer (UNEP, 2002a).

Globally, the principal sources of HCB in the environment are estimated to be the manufacture of chlorinated solvents, the manufacture and application of HCB-contaminated pesticides, and inadequate incineration of chlorine-containing wastes (Bailey, 2001). The current global estimate of HCB emissions of 23 tonnes/year is only about one-tenth of what is estimated to be required to maintain observed atmospheric HCB concentrations, using Frank Wania's global distribution model ‘GLOBAL-POP’ (Wania and Mackay, 2000). There may be significant primary HCB emission sources that are currently unknown, but that is unlikely. It is possible that secondary emission of HCB from historically contaminated soil is responsible for this shortfall, and if so, this would make soils the largest current source of HCB emissions to the global environment.

Table 8: Summary of reported HCB production and emission data

Location	Year	Notes	Amount of HCB	Units	Reference
Global		Production	>100 000	tonnes	ATSDR (1997)
Global	mid-1970s	Production	1000-2000	t/yr	Courtney, 1979
Global	1978-81	Production	10000	t/yr	Rippen and Frank, 1986
Europe	1978-81	Production	8000	t/yr	Rippen and Frank, 1986
Global	mid-1990s	Emission estimate	22.7 (12-92)	t/yr	Bailey, 2001
Australia	1960s	Use	200	t/yr	FAO/WHO, 1970
US	1960	Use	360	t/yr	IARC, 1979
US	1973	Production	300	tonnes	IARC, 1979
US	1977	Production	454	tonnes	SMOC Mexico, 1998
US	1985	Emission estimate	18-29	t/yr	Carpenter *et al*, 1986
US	mid-1990s	Emission estimate	2.8	t/yr	Bailey, 2001
Canada	mid-1990s	Emission estimate	0.11	t/yr	Bailey, 2001
Mexico	1970-1996	Production	39000	tonnes	SMOC Mexico, 1998
Mexico	1970s	Production	3500	t/yr	SMOC Mexico, 1998
Europe	1970	Emission estimate	192	t/yr	Pacyna *et al*, 1999
Europe	1993/5	Emission estimate	23	t/yr	Pacyna *et al*, 1999
Europe	1970-93/5	Emission estimate	651	tonnes	Pacyna *et al*, 1999
Europe	1985	Industrial emissions to air	137.5	kg/yr	Euro Chlor
Europe	2002	Industry emissions to air	0.05	kg/yr	Euro Chlor
Europe	1985	Industry emissions to water	713	kg/yr	Euro Chlor
Europe	2002	Industry emissions to water	8.5	kg/yr	Euro Chlor
Germany	1974	Production	4000	t/yr	Rippen, 1989
Germany	1976	Production	2600	t/yr	Rippen, 1989
Germany	1980	Production for PCTP manufacture	5000	tonnes	Rippen and Frank, 1986
Germany	Until 1993	Production for PCTP manufacture	1500	t/yr	IPCS, 1997
Spain	1970s	Production	150	t/yr	IARC, 1979
UK	1990	Emissions	1.3	t/yr	DEFRA, 2004
UK	2002	Emissions	0.3	t/yr	DEFRA, 2004
France	1990	Emissions	1.7	t/yr	CITEPA, 2004
France	2002	Emissions	1.7	t/yr	CITEPA, 2004
USSR	Prior to 1986	Use	610	tonnes	UNEP, 2002c
India	1995-97	Production	42612	tonnes	Ministry of Chemicals & Fertilisers, 2000
Pakistan	1979-88	Use	12162	tonnes	UNEP, 2002c
Japan	1952-1972	Production for PCP manufacture	70000	tonnes	UNEP, 2002c
Japan	1977	Production for PCP manufacture	300	tonnes	IARC, 1979

Table 9: Dates of legislative action on HCB in different countries

Legislative action	**Country**
Never registered as pesticide	Iceland, Uruguay, Chile, Nicaragua, Costa Rica, Malaysia, Japan, Korea, Mongolia, Uzbekistan
Banned/withdrawn/restricted in 1960s	Argentina (r 1963), Hungary (b 1968)
Banned/withdrawn/restricted in 1970s	New Zealand (b 1972), Australia (b 1972), UK (b 1975), Canada (w 1976), Finland (w 1977), European Union (b 1979), Japan (b 1979)
Banned/withdrawn/restricted in 1980s	Sweden (w 1980), Czech Republic (w 1980), Poland (w 1980), Egypt (b 1981), USA (r 1984), East Germany (b 1984), Morocco (b 1984), Ecuador (b 1985), USSR (b 1986), Tunisia (w 1986), Switzerland (b 1986), Panama (b 1987), Brazil (r 1985), Singapore (r 1985), Norway (w 1987),
Banned/withdrawn/restricted in 1990s	Papua New Guinea (b 1990), Mexico (w 1991), Vietnam (b 1992), Paraguay (b 1993), Columbia (b 1993), Iceland (b 1996), Turkey (b 1997), Bosnia & Herzegovina (b 1997), Syria (b 1998), Malta (b 1998), Slovenia (b 1998), Jamaica (b 1999), Peru (b 1999),
Banned/withdrawn/restricted in 2000s	Argentina (b 2000), El Salvador (b 2000), Thailand (b 2001), Jordan (b 2001), Bolivia (b 2002), Chile (b 2002), Canada (b 2003)
Banned/withdrawn/restricted (no date available)	Denmark (b), Cambodia (b), Indonesia (b), Guatemala (b), Philippines (r)
Still used as an intermediate	China, Russia

b - banned; r – restricted

Table 10: Estimated relative contribution of different sources to total emissions in Europe, the US, Canada and globally

Source Category	**Annual emission (t/y) (percentage of total)**			
	Europe [1] (1990)	US [2] (mid-1990s)	Canada [2] (mid-1990s)	Global [2] (mid-1990s)
Fuel combustion	0.46 (6.3 %)	0.024 (0.9 %)	0.0005 (0.5 %)	0.843 (3.7 %)
Iron and steel industry	0.24 (3.3 %)	0.018 (0.6 %)	0.001 (0.9 %)	0.070 (0.3 %)
Non-ferrous metal industry	0.05 (0.7 %)	0.156 (5.6 %)	0.015 (14 %)	8.154 (36 %)
Organic chemical industry	0.56 (7.6 %)	0.399 (14 %)	0.0001 (0.09 %)	1.308 (5.8 %)
Other solvent use	0.21 (2.9 %)	0.0003 (0.01%)	0.000025 (0.02 %)	0.001 (0.004 %)
Waste incineration	0.07 (1 %)	0.917 (33 %)	0.054 (50 %)	5.862 (26 %)
Pesticide use	5.76 (78 %)	1.27 (46 %)	0.0337 (31 %)	6.463 (28 %)
Total	**7.35**	**2.785**	**0.108**	**22.703**

1 Berdowski *et al* (1997)
2 Bailey (2001)

Table 11: HCB emission estimates for 1970-1995 (in kg/year), showing significant decreases in emissions

Country	1970	1975	1980	1985	1990	1993-95	Change	Ref
Albania	249	249	165	170	88	55	*-78 %*	1
Armenia	163	164	111	110	55	50	*-69 %*	2
Austria	2097	2018	1959	240	81	81	*-96 %*	1
Azerbaijan	615	620	417	415	206	187	*-70 %*	2
Belarus	1762	1752	1161	1155	573	471	*-73 %*	1
Belgium	1500	1593	1226	418	73	75	*-95 %*	1
Bulgaria	1156	1176	795	789	398	300	*-74 %*	1
Czechoslovakia (former)	1500	1500	990	990	490	330	*-78%*	1
Denmark	3181	3166	3024	319	131	115	*-89 %*	1
Estonia	289	281	181	177	87	71	*-75 %*	1
Finland	3160	3132	2915	290	127	126	*-96 %*	1
France	25662	25621	23197	3776	1276	1285	*-95 %*	1
Georgia	519	523	353	351	175	158	*-70 %*	1
Germany	20486	20969	17857	5259	1654	1555	*-92 %*	1
Greece	4672	4628	4510	493	195	175	*-96 %*	1
Hungary	1341	1336	877	863	427	305	*-77 %*	1
Hungary					0.304	0.660		3
Iceland	85	96	15	16	7	7	*-92 %*	1
Ireland	1646	1485	1289	139	47	46	*-97 %*	1
Italy	20123	17274	15767	2792	839	795	*-96 %*	1
Kazakhstan	2431	2449	1649	1641	817	740	*-70 %*	2
Latvia	501	483	309	310	155	128	*-74 %*	1
Lithuania	631	644	437	431	213	176	*-72 %*	1
Luxembourg	83	77	72	10	3	3	*-96 %*	1
The Netherlands	1423	1470	1370	389	93	92	*-94 %*	1
Norway	1014	991	995	129	45	45	*-96 %*	1
Poland	3907	3901	2612	2634	1321	930	*-76 %*	1
Portugal	4740	4645	3651	416	159	145	*-97 %*	1
Republic of Moldova	383	390	269	278	144	100	*-74 %*	2
Romania	2847	2887	1940	1955	970	740	*-74 %*	1
Russian Federation	32365	32613	21971	21860	10868	9846	*-71 %*	1
Russian Federation					1.6	1.3		3
Russian Federation	36092	36369	24501	24376	12120	10980	*-70 %*	4
Slovenia	139	139	93	93	47	15	*-89 %*	2
Spain	25384	25848	23899	3084	1175	1172	*-95 %*	1
Spain					9227	6088		3
Sweden	3800	3720	3493	413	161	161	*-96 %*	1
Switzerland	633	648	727	185	59	56	*-91 %*	1
Ukraine	7759	7840	5292	5263	2619	2095	*-73 %*	1
United Kingdom	11071	10926	9707	2175	548	511	*-95 %*	1
United Kingdom					1267	1248		3
Yugoslavia (former)	2800	2800	1900	1900	940	310	*-89 %*	1
Europe Total (t/y)	**192**	**190**	**157**	**62**	**27**	**23**	***-88 %***	**1**
Europe Total (t/y)				**51**				**5**
Europe Total (t/y)					**7.35**			**6**
US Total (t/y)				**18-29**				**7**
US Total (t/y)					**2.3**			**8**
US Total (t/y)						**0.2-0.4**		**9**
Global Total (t/y)	**10 000-30 000**							**1**
Global Total (t/y)						**23**		**10**

[1] Pacyna *et al* (2003); [2] EMEP Report 4/2001; [3] Official emissions reported to UN ECE Secretariat; [4] Münch and Axenfeld (1999); [5] Axenfeld *et al* (1992); [6] Berdowski *et al* (1997); [7] Carpenter *et al* (1986); [8] US EPA TRI for 1990 (1998); [9] US EPA TRI for 1990-97 (1998); [10] Bailey (2001)

3 Environmental Loss Processes

Although HCB is very persistent, it does degrade at a slow rate in all environmental compartments. For modelling purposes, HCB was assigned approximate half-lives of 17000 hours (1.9 years) in air and 55000 hours (6.3 years) in water and sediment by Mackay *et al* (1992). These were 'default' values, assigned to HCB based on its categorisation as a 'non-degrading' chemical. The net residence time of HCB in air is significantly less than one year, and is based on physical translocation and not on chemical transformation (Ballschmiter and Wittlinger, 1991). Loss processes from the environment are discussed in more detail for various environmental compartments in the following sections.

3.1 Degradation in Air

HCB has a high activation energy of 24.3 kJ/mol due to steric considerations caused by the presence of a chlorine on each carbon atom (Brubaker and Hites, 1998). Thus, atmospheric degradation of HCB is extremely slow, and is not an efficient removal process. HCB may be removed from the atmosphere by photolysis and by chemical reaction with hydroxyl (OH) radicals. Howard (1991) estimated a half-life of HCB in air due to photo-oxidation ranging from 156 days to 4.2 years. Kwok and Atkinson (1995) estimated the rate constant for the gas-phase reaction of OH radicals with HCB to be 0.2×10^{-13} cm^3 s^{-1} using a structure-reactivity method, with a half-life ranging from 158 days to 4.3 years. Wania and Mackay (1995) predicted that the degradation half-life of HCB would vary in different regions, with half-lives of 0.63 years (230 days) in tropical/subtropical regions, 1.94 years (708 days) in temperate/boreal regions, and 6.28 years (2292 days) in polar regions. Brubaker and Hites (1998) reported an OH radical attack rate at $25^{0}C$ of 0.27×10^{-13} cm^3 s^{-1} from chamber studies. This leads to an estimated atmospheric lifetime (1/e) of 940 days (a half-life of 1.69 years). These results suggest that HCB can be transported great distances in the atmosphere before removal by degradation. In the troposphere, HCB is probably photochemically stable, but degradation by photo-dissociation by shorter-wavelength, higher energy ultraviolet light may be a mechanism for atmospheric degradation in the stratosphere (Parlar, 1978).

3.2 Degradation in Water

HCB may be removed from water by photolysis, but at a very slow rate, with a half-life of about 70 days (Mill and Haag, 1986). HCB in aqueous solution is degraded into pentachlorophenol, which is then degraded further. However, HCB in the water column rapidly sorbs to particulate matter, making it unavailable for photolysis (Schauerte *et al*, 1982). Thus, hydrolysis is not expected to be an important fate process. A half-life ranging from 2.7 to 5.7 years in surface water and 5.3 to 11.4 years in groundwater has been suggested, based on unacclimated aqueous aerobic biodegradation (Mackay *et al*, 1992; Howard, 1991). Hydrolysis is not expected to be an important fate process.

3.3 Degradation in Soils

HCB is persistent in soils because it is strongly sorbed to soil organic matter, which gives it a low bioavailability (Beurskens *et al*, 1993), and due to its completely substituted nature. The half-life for residence of HCB in soil has been estimated to be 970-2100 days (Griffin and Chou, 1981), with the major loss process from soil at the surface being volatilisation. In a study of treated soil stored under aerobic and anaerobic conditions in covered containers to retard volatilisation, no detectable loss of HCB occurred over the one-year experiment (Isensee *et al*, 1976). In another sturdy, up to 78% of soil-applied HCB could be recovered in an oxic soil after one year (Beall, 1976). Aerobic and anaerobic biodegradation are the major means of HCB removal at lower soil depths, with half-lives of 2.7-5.7 years (Beck and Hansen, 1974) and 10.6-22.9 years (Howard, 1991), respectively. Meijer *et al* (2001) reported a half-life of 11.7 years for sewage-sludge treated soils over the period 1968-1990. However, if only the data points after the 1980 peak are used, a half-life of 4-6 years is obtained. A major problem with these data is that measured 'disappearance' from soils includes both volatilisation and degradation. Whereas biodegradation completely removes

HCB from the environment, volatilisation leads to continued existence of HCB in a different environmental compartment.

Brahushi *et al* (2004) studied the anaerobic biodegradation of HCB in an arable soil. Several dechlorination pathways were detected, indicating the following main HCB transformation pathway: HCB → pentachlorobenzene (QCB) → 1,2,3,5-tetrachlorobenzene (TeCB) → 1,3,5-TCB → 1,3-dichlorobenzene (DCB), with 1,3,5-TCB as main intermediate dechlorination product. The other TeCB-, TCB- and DCB- isomers were also detected in low amounts, showing the presence of more than one dechlorination pathway. Sulphate-reducing or nitrate-reducing microorganisms rather than methanogenic bacteria were implicated as the dechlorinating organisms (Rosenbrock *et al*, 1997; Brahushi *et al*, 2004).

Bosma *et al* (1997) have suggested that complete degradation of HCB in soil or sediment cannot occur, because as the bacteria break down HCB and the availability of further HCB is reduced by mass transfer from the organic matter, a threshold concentration is reached below which no biotransformation is possible. Initial degradation rates are directly related to the aqueous concentration according to the Michaelis-Menten relationship (Pavlostathis and Pyrtula, 2000). Subsequently, retarded desorption occurs, and the effective diffusivity of HCB (mainly controlled by diffusion in the OM) controls the degradation progress. A point is then reached when the 'available' HCB concentration falls below that necessary to support the microbial population feeding on it.

3.4 Degradation in Sediment

Using fugacity modelling and published reaction rates, Mackay *et al* (1992) suggested that the half-life of HCB was greater than 6 years. Beurskens *et al* (1993) investigated microbial degradation of HCB in a sedimentation area of the River Rhine. They found that significant anaerobic dechlorination had occurred, with up to 80% of the HCB that had been deposited in the 1970s having been dechlorinated by 1988. They calculated a maximum half-life of HCB in sediment of seven years. HCB was transformed into the less toxic and more mobile 1,3,5-TCB, 1,2-DCB and 1,3-DCB. Chang *et al* (1998) investigated this anaerobic dechlorination pathway under laboratory conditions. Dechlorination was strongest in methanogenic conditions, followed by sulphate reducing conditions, but did not occur under denitrifying conditions. Biotransformation occurred along the metabolic decay route: HCB → QCB → 1,2,3,5-TeCB → 1,3,5-TCB + 1,2,4-TCB → 1,3-DCB (Chang *et al*, 1998). Zhao *et al* (2003) also investigated the anaerobic degradation of HCB in sediments, and observed a degradation rate of 0.035 month^{-1}, which increased to 0.088 month^{-1} when extra organic carbon was added to the sediment. These values correspond to half-lives of 1.7 and 0.7 years. A wide range of degradation rates have been reported in other studies, depending on experimental set-up, with values of 0.110 d^{-1} (Susarla *et al*, 1997), 0.021 d^{-1} (Jackson and Pardue, 1998), 0.0256 d^{-1} (Masunaga *et al*, 1996) and 0.0022 d^{-1} (Prytula and Pavlostathis, 1996) found by different authors. These correspond to half-lives of 6.3 days, 33 days, 27 days and 315 days, respectively. In the first of these experiments, after almost a year, 98% of the HCB was dechlorinated to 1,3,5-TCB and 1,4-DCB in sediment from a fresh water lake (Susarla *et al*, 1997). These half-lives are all considerably lower than the 6.3 year half-life estimated by Mackay *et al* (1992).

HCB has also been shown to degrade anaerobically in sewage sludge, with 1,3,5-trichlorobenzene (1,3,5-TCB) as the main product, along a similar pathway to that reported above (Fathepure *et al*, 1988). This process was examined in detail by Yuan *et al* (1999). Biotransformation occurred along a slightly different pathway of HCB→ QCB → 1,2,3,4-TeCB + 1,2,3,5- TeCB → 1,2,4-TCB + 1,2,3-TCB + 1,3,5-TCB → 1,2-DCB + 1,4-DCB. This means that TCB dechlorinated further to DCB, contrary to the findings of Fatherpure *et al* (1988). Highest-to-lowest dechlorination rates under three reducing conditions were reported as methanogenic conditions (0.30 mg/l/day) > sulphate-reducing conditions (0.23 mg/l/day) > denitrifying conditions (0.08 mg/l/day).

3.5 Degradation in Plants

HCB degradation in barley was found to be less than 0.8% per week (Trapp *et al*, 1990), with a similar rate measured in the soil. Other studies have observed biodegradation of HCB by plants, with the maximum reported rate only 0.1% per week (Mansour *et al*, 1986).

The low degradation rate in plants is unlikely to have significant effects on global HCB concentrations.

3.6 Degradation in Biota

HCB is a highly deactivated molecule and therefore it is metabolised at a very slow rate, albeit by a variety of different organisms (Mathews, 1986). The metabolism of HCB has been studied in a number of species using various routes of exposure, and has been subject to several reviews (Renner, 1988; Rietjens *et al*, 1997). HCB metabolism has been demonstrated in mussels, and both marine and freshwater fish (Ernst, 1986). In mammals, HCB is metabolised into less chlorinated benzenes, chlorinated phenols, and other minor metabolites. Metabolism occurs primarily in the liver, and occurs along three distinct pathways: i) oxidation giving rise to phenolic metabolites including pentachlorophenol (PCP), tetrachlorohydroquinone and tetrachlorobenzoquinone; ii) glutathione-conjugation leading to pentachlorobenzenethiol, pentachlorothioanisole and other sulphur-containing metabolites; and iii) reductive dechlorination yielding less chlorinated benzenes (Yang *et al*, 1978; Debets and Strik 1979, Renner, 1988). A total of 21 different metabolites of HCB have been identified in the urine of rats (Koss *et al*, 1986). Oxidative metabolism of HCB is thought to be catalysed by cytochrome P450 enzymes (predominantly by CYPIIIA1/2), epoxyhydratase, and glutathione transferases (van Ommen *et al*, 1989). In rodents, enzyme induction has been observed following short-term repeated dosing, including induction of isoforms of cytochrome p-450 (P-4501A1, P-4501A2, and P-4502B) and glutathiome-S-transferases (Renner, 1988). These studies have indicated that HCB is a mixed-type cytochrome P-450-inducing compound exhibiting Phenobarbital- and 3-methyl-cholanthrene-like inducing properties. In humans, pentachlorobenzenethiol (PCBT) is the main metabolite of HCB (To-Figueras *et al*, 2000) with PCP also excreted in urine in significant amounts (Gomez-Catalan *et al*, 1987; To-Figueras *et al*, 1997). Most HCB is eliminated unchanged by excretion primarily in the faeces (To-Figueras *et al*, 2000) and a small portion in the urine (To-Figueras *et al*, 1997). HCB levels in seals are considerably lower than in their dominant prey (Goerke *et al*, 2004; Muir *et al*, 1988; Ruus *et al*, 1999; Fisk *et al*, 2001). This suggests that seals have an exceptional capacity to eliminate HCB by biotransformation. This metabolic ability does not appear to be shared by other marine mammals.

4 Factors Controlling Transfer of HCB around the World

HCB is distributed throughout the environment because it is mobile and resistant to degradation. HCB has a high volatility and is a ‘multi-hop chemical’, that demonstrates long-range transport in a way that has been described as the ‘grasshopper effect’ (Wania and Mackay, 1995; 1996). With multi-hop chemicals, a compound re-enters the atmosphere after initial deposition to the earth’s surface, and continues over time to move through the environment in multiple hops. This means that the chemical is trapped in an organic phase without being degraded, and depending on the development of an equilibrium imbalance, it is then released back into the air or water, where it can travel long distances before being trapped again. Processes by which this can occur include volatilisation from the earth’s surface at temperatures warmer than during initial deposition; volatilisation from water surfaces into a formerly ‘dirty’ but now cleaner atmosphere; and re-suspension from sediments and subsequent volatilisation from water surfaces. The last process is common for lakes and coastal areas close to sources. Theses processes continue until the chemical is ultimately degraded. HCB undergoes the iterative process of deposition, remobilisation into the atmosphere and re-deposition. This ‘global distillation’ (Mackay and Wania 1995) and long-range transport have been given as a reason for the prevalence of HCB in polar regions (Meijer *et al*, 2003a). The distillation and condensation of HCB on top of cold mountains also occurs.

Barrie *et al* (1992) concluded that both the atmosphere and the ocean currents are the major transport mechanisms that must operate in conjunction with global distillation for the movement of HCB to occur. Generally, the atmosphere contains a relatively small amount of HCB compared to the total amount in other environmental compartments. The importance of the atmosphere is that it provides a significant mode of rapid transport of HCB from source areas to remote regions. The time for an air parcel to completely mix in the troposphere of the northern hemisphere is around six months. In contrast, transport times of water parcels are measured in years and decades (Plumb and Mahlman, 1987). In the troposphere, HCB is transported over long distances due to its persistence, but does undergo slow photolytic degradation (the half-life is approximately 80 days; Mill and Haag, 1986), or is removed from the air phase *via* atmospheric deposition to water and soil (Bidleman *et al.*, 1986; Ballschmiter and Wittlinger, 1991; Lane *et al*., 1992a, 1992b). The major transfer pathways between the atmosphere and the surface include: snow, particle deposition, particle re-suspension from the surface, gas exchange in both directions, and rain. Van Pul *et al* (1998) modelled the atmospheric residence time of POPs. Transport distances were calculated as the distance over which 50% of the chemical is removed. The value for HCB was 10^5 km. Due to this long atmospheric residence time it is distributed widely on national, regional and global scales.

4.1 Observed Occurrence of HCB in the Environment

The occurrence of HCB in an area is influenced by the nature and rate of emissions from sources locally, the processes by which HCB is transported to the area, removal processes, and the exchange of compounds between the various compartments. Significant quantities of HCB are present in the atmosphere, the surface oceans and the terrestrial environment. The volume of the accessible terrestrial environment of soils and vegetation is relatively small compared to that of the atmosphere and the oceans. However, the hydrophobic nature of HCB results in its preferential partitioning into soil and plant surfaces from water or air. This leads to the accumulation of HCB in biota, with significant bioconcentration in top predators, such as fish, marine and terrestrial mammals and birds. Although uptake into food-chains is critical from the viewpoint of human exposure and ecological effects, biota are of minor importance in the overall mass balance on global, hemispheric and regional scales. The possible exceptions to this are microbial conversion in soils and partitioning to planktonic organisms in fresh and marine waters, which may significantly influence the removal of HCB from the environment. Vegetation may also pay a key role in atmosphere-surface transport, especially in forested systems, due to its large surface area and its close coupling with the atmosphere. The ultimate fate of HCB in the environment depends on several factors including concentration; water solubility; biogeochemical processes taking place; adsorption to soils, suspended particulates and sediments; lipophilicity; and bioaccumulation in living organisms.

4.2 Air

There have been a large number of studies of the levels of HCB in the atmosphere (see Annex 1). The majority of these have been in Europe and North America, but reasonable global coverage has been achieved. It has been reported that there are some problems associated with collecting air samples for HCB analysis. Due to the high volatility of HCB, after quite small volumes of air, breakthrough can occur in the sampling device, where not all the HCB in the air entering the device is collected (Billings and Bidleman, 1980; Billings and Bidleman, 1983; Hoff *et al*, 1992). Consequently, a number of authors have reported HCB air concentrations that they believe to have been affected by breakthrough (Oehme and Manø, 1986; Oehme, 1991a; Hoff *et al*, 1992; McConnell *et al*, 1996; Halsall *et al* 1998; Romanic and Krauthacker, 2000; Qiu *et al*, 2004). In these cases actual air concentrations will be higher than reported. The efficiency of HCB capture by samplers decreases with increasing temperature. Hoff *et al* (1992), using a single PUF plug, got breakthrough in volumes over 250 m^3 at summer temperatures. Samplers using XAD in conjunction with PUF plugs are more efficient: Hippelein *et al* (1993) had no significant breakthrough at 2^oC in samples of 2200 m^3 of air.

Historically, the highest levels of HCB in the air were reported from places where HCB was used or produced in large amounts. Since the majority of these emissions occurred in the Northern Hemisphere, this is where highest atmospheric concentrations occurred. In North America highest concentrations were reported in the Great Lakes area, due to the concentration of industry and pesticide use there. Air samples taken inside a typical PCP manufacturing plant in the US in the mid-1980s had HCB concentrations ranging from <0.1-630 µg/m^3 (Marlow, 1986), indicating that workers at these plants had high occupational exposure to HCB. Other reports of up to 150 µg/m^3 inside an industrial plant (Currier *et al*, 1980) and 22 µg/m^3 inside a pesticide production facility (Davis and Morgan, 1986) have been made. Mann *et al* (1974) measured HCB levels ranging from 0.070 to 23.296 µg/m^3 near chlorinated solvent and pesticide manufacturing facilities. Similarly, Spigarelli *et al* (1986) detected HCB in air in the US at concentrations of up to 24 µg/m^3 at an on-site landfill of a manufacturer of PCE, carbon tetrachloride and chlorine. Levels of 1 µg/m^3 were detected 700m downwind of the incinerator. In Europe, high concentrations were reported in industrialised regions of countries such as Germany, Spain, and the Czech Republic. Popp *et al* (2000) found elevated levels of HCB in the atmosphere near a former chemical plant in Germany that produced technical HCH and lindane between 1951 and 1982, and also near the landfill site used for disposal of its waste. Elevated HCB levels were detected in Flix, Spain, a small village located near a chlorinated solvent manufacturing plant. Air levels of HCB were 100 times higher in Flix (35 ng/m^3) than in nearby Barcelona (0.3 ng/m^3) (Grimalt *et al*, 1994). High levels of up to 2.4 ng/m^3 have also been reported from agricultural areas of France (Chevreuil *et al*, 1996; Sanusi *et al*, 2000).

Due to the high long-range transport potential of HCB, it has also been measured in remote areas far from sources, such as the Arctic and the middle of the Pacific Ocean (Atlas and Giam, 1989). HCB is found at the highest concentration of any individual organochlorines (OCs) in Arctic air (de March *et al*, 1998). HCB has also been detected at a number of locations in the Southern Hemisphere, such as Australia (Iwata *et al* 1994b), New Zealand (Ministry for the Environment, New Zealand, 1998), and the Southern Ocean (Bidleman *et al*, 1993). However, it is not clear whether this HCB originates from the Northern or Southern Hemispheres, because it is not known how efficient the exchange of HCB between the two Hemispheres is. Current thinking is that the presence of the Inter-Tropical Convergence Zone will limit atmospheric transport between the two Hemispheres. Little inter-hemispheric mixing of air will occur due to the presence of Hadley cells over the Tropics (Levy, 1990). Indications are that background Southern Hemisphere air concentrations in general are lower than background Northern Hemisphere concentrations in the Atlantic (10 *vs* 40 pg/m^3; Jaward *et al*, 2004a), and that levels of HCB in the Antarctic were lower historically (60 pg/m^3, Bidleman *et al*, 1993) when compared with the Arctic (>100 pg/m^3).

As emissions of HCB from point sources have decreased, the variability in HCB air concentrations has also decreased. In fact, regionally, levels of HCB in the atmosphere are very uniform. Studies of HCB atmospheric concentrations at several sites within a region tend to show little variation (Ecker and Horak, 1994; Kaupp *et al*, 1996; Murayama *et al*, 2003). For example, in the early 1990s, average atmospheric HCB concentrations around the Great Lakes ranged from 74 pg/m^3 at Eagle Harbour to 82 pg/m^3 at Sleeping Bear Dunes and 84 pg/m^3 at Sturgeon Point (Cortes and Hites, 2000). Four sites in Alberta, Canada had similar, low concentrations (averages ranging from 38-45 pg/m^3 in 1999-2000) and all locations recorded a single high concentration value (range 350-670 pg/m^3) in mid-September, whose origin was unknown (Kumar, 2001). A passive sampling survey in 2002 using PUF (polyurethane foam) discs showed that HCB concentrations varied by less than a factor of six over the whole of Europe, with the highest concentration at a rural site in Germany (Jaward *et al*, 2004b). Similar results were found when HCB air concentrations were measured by using passive air samplers (SPMDs) along a latitudinal transect from the south of the UK to the north of Norway during 1998-2000 (Meijer *et al*, 2003a). Differences between highest and lowest sequestered HCB concentrations were only a factor of around three. Absolute amounts of HCB sequestered by the sampler increased with increasing latitude, and decreasing temperature, suggesting that this compound was “grasshopping” and undergoing “cold condensation”. Van Drooge *et al* (2004) found that HCB concentrations at remote mountain sites in the Pyrenees (Spain) and the High Tatras (Slovenia) varied only slightly temporally, and no correlation was found between air concentration and back trajectory or temperature.

Some locations do show relationships between HCB air concentration and back trajectory, indicating that source regions do still exist. For example, in the Lake Taihu Region of China, lower concentrations were found in air masses originating over the Pacific Ocean (23 pg/m^3) than those from Southern (54 pg/m^3) and Northern (70 pg/m^3) China (Qiu *et al*, 2004). Several studies have shown a relationship between temperature and air HCB concentration. This is important evidence for the movement of HCB around the environment by repeated hops, and recycling of residual HCB from soil and water. Ma *et al* (2003) studied the trends of HCB in air samples collected by the IADN network in the Great Lakes between 1992 and 1999. They found that higher winter concentrations occurred during El Niño events, with associated warmer winter temperatures, and lower concentrations occurred during La Niña events, with associated colder winter temperatures. These variations in HCB concentrations were not caused by variations in current emissions, and therefore must have been due to re-volatilisation of HCB from the soil contaminated from past use.

Kaupp *et al* (1996) observed a clear annual cycle in HCB air concentrations in Germany, with the mean urban concentration in summer being three times higher than in winter. Atmospheric HCB concentrations showed significant temperature dependence in the Great Lakes area (Cortes *et al*, 1998), with levels of HCB in air 2-3 times higher in the summer than the winter (Monosmith and Hermanson, 1996). Similarly, levels of HCB in air in the Canadian Arctic vary seasonally, showing a bimodal seasonal distribution (Hargrave *et al*, 1997), with maxima in February-May and July-August, separated by minimum values in June. Similar results were obtained in Japan (Murayama *et al*, 2003). Contrasting reports exist that levels of HCB at Alert, in the Canadian Arctic, and in the Great Lakes, were higher in the winter than the summer (Hoff *et al*, 1992; Halsall *et al*, 1998), but these were almost certainly an artefact of the sampling process, since breakthrough of HCB had occurred in their samples in the summer months. In contrast, HCB atmospheric concentrations at a rural site in the UK were found not to exhibit the significant short-term temperature-dependent diurnal cycling observed for other POPs (Lee *et al*, 2000).

Data from the studies that show temperature dependence in air concentration can be used to calculate enthalpies of air/surface phase change (ΔH – the energy necessary to overcome a chemical’s own attractive forces to convert Δ1 mol from the dissolved or liquid phase to the gas phase) for HCB. Reported ΔH values ranged from 5 kJ/mol at Eagle Harbour to 13 kJ/mol at Sturgeon point in the Great Lakes (Cortes *et al*, 1998), 36.6 kJ/mol in Germany (Kaupp *et al*, 1996), and 1.9 and 17 kJ/mol in the UK (Lee *et al*, 2000). These comparatively low ΔH values suggest that HCB air concentrations are only weakly temperature dependent, with other processes such as long-range transport or changes in wind direction strongly affecting air concentrations.

4.2.1 Deposition Processes

Emissions to the atmosphere are returned to the land or sea by wet and dry deposition. Wet deposition (deposition *via* rain or snowfall) is the primary mechanism for transport of HCB from the atmosphere to aquatic and terrestrial systems (Eisenreich and Strachan, 1992). Transfer from the atmosphere to the earth's surface in the polar regions are complicated by the presence of a seasonally varying intermediate media, namely snowpack and ice. The snowpack plays an important role in atmosphere-surface exchange. During the winter, as snow accumulates on the surface as snowpack, it stores contaminants gained during its formation in addition to those delivered by particle dry deposition and gaseous exchange. The latter component may be negative (surface loss) or positive (surface gain).

4.2.2 Rain

Wet deposition of HCB is more efficient at depositing HCB than dry deposition (van Drooge *et al*, 2001). The washout of HCB from air by rain can be calculated from equilibrium partitioning, using a washout ratio, W:

$$W = \frac{RT}{H}$$

where R is the gas constant, T is the temperature (in K), and H is the Henry's Law constant. The washout ratio (concentration in rainwater/concentration in air) for HCB has been calculated to be 3.7×10^2, with measured values of 1.5×10^3 observed (Eisenreich *et al*, 1981). There are few studies of HCB concentration in rainfall. The results from those that exist are shown in Annex 2.

As in air, levels of HCB in rain are higher near to sources, and vary depending on the back trajectory of the air mass. Levels of HCB were generally high in France, although still significantly higher in urban areas than in rural areas, which suggests a probable industrial emission route (Teil *et al*, 2004). Background levels of HCB in rain are approximately 15-30 pg/l. Carrera *et al* (2002) found in a study of deposition of HCB in remote high mountain lakes of Europe in 1996-98 that an order of magnitude more HCB was deposited in the Caledonian Mountains in Northern Norway than in the Alps or Pyrenees. This is evidence for preferential long-range transport distillation towards the polar regions. Higher deposition was measured at all sites during warmer periods, which is consistent with enhanced volatilisation at higher temperatures from source areas (Carrera *et al*, 2002).

4.2.3 Snow

The efficiency of scavenging HCB from the atmosphere differs for snow and rain. Snow is thought to be more effective at scavenging atmospheric particles and gases due to its high porosity and large specific surface area (Wania *et al*, 1998). Above 0°C, rain is a more efficient scavenger of HCB, but below 0°C, snow is increasingly efficient (Barrie *et al*, 1997). Since the scavenging efficiency of snow for HCB increases as the temperature decreases, the deposition of HCB in snow will therefore be greater at the poles. HCB is largely volatilised out of freshly fallen snow, with Villa *et al* (2003a) reporting losses of greater than 90% from snow after 65 days, over a period when temperatures remained below 0°C. Similar results were obtained by Donald *et al* (1999), with 84-88% volatilisation losses, and by Wania (1997), who predicted losses of up to 90%.

Concentrations of HCB in snow are low compared to those of other POPs (Gregor, 1989). HCB is fairly uniformly distributed in snow in the Arctic, and levels in the Arctic are similar to those at Lake Superior (see Annex 3). This is an indication of the ubiquity and uniformity of distribution of HCB in the global environment. Carrera *et al* (2000) found higher concentrations of HCB in snow on mountains in Northern Europe than in Southern Europe, providing further evidence of global distillation of HCB.

4.3 Water

Significant amounts of HCB can be found in water in rivers, lakes and seas (see Annex 4), and generally, river concentrations > estuary concentrations > lake concentrations > sea concentrations. In the aquatic environment, HCB may remain in solution, may undergo photolysis, may be adsorbed onto solid surfaces (sediments and biota), or may be reversibly transferred into the atmosphere by volatilisation. For example, the major HCB removal processes from Lake Ontario in 1982 were volatilisation from water to air (80%) and sedimentation following adsorption onto suspended particulates (15%, Oliver and Charlton, 1984).

Levels of HCB in rivers in remote areas are low, with values of 0.5-20 pg/l reported in New Zealand (Hermann, 1987) and 3-14 pg/l reported in Northern Ontario rivers (McCrea *et al*, 1984). In contrast, very high concentrations of HCB in rivers have been reported in industrial areas of Europe, the US, and South America as a result of inputs of effluents from HCB production plants and other industries, and run-off from agricultural fields. Slightly elevated (140 pg/l) levels of HCB were found in effluent from an urban wastewater treatment plant in Columbia, US (Petty *et al*, 2004) compared to the nearby Missouri River (12 pg/l). Levels of HCB in European rivers exceeded 100 ng/l in the 1970s (Greve, 1986), and in the St. Clair River, US, near the Dow Chemical outfall, were as high as 87 ng/l in 1985 and 75 ng/l in 1986 (Oliver and Kaiser, 1986). Levels of HCB in rivers in South America (mainly Brazil and Argentina, and to a lesser extent Ecuador, Chile and Uruguay) range from 1–14160 ng/l, with an average of 418 ng/l (or 1384 ng/l if extreme values are included, UNEP, 2002d). The average is raised by Brazilian data, which has the largest data set (25/31), with the highest concentrations (6000-14160 ng/l; CETESB, 1997). A study in Ecuador reports concentrations ranging from 9 to 14 ng/l in surface waters at the Guayas Ecosystem (Resabala *et al*, 2002).

In freshwater rivers, HCB is found more frequently in solution than in suspended particles, but in estuaries, which have a higher particle burden, the reverse is true (Greve, 1986). Wodarg *et al* (2004) reported a log organic carbon/water coefficient for HCB in the Baltic Sea of 6.0 ± 0.1. Quemerais *et al* (1994) reported that 23% of HCB in whole water samples collected from the St. Lawrence River was associated with the dissolved phase, while 77% was associated with the particle phase. Levels of HCB in estuaries also tend to be lower due to the increased sedimentation rate removing HCB from the water. Higher HCB concentrations are found in estuaries around the North Sea (up to 15000 pg/l) than in the Sea itself, and so clearly fluviatile transport plays a major role in the discharge of HCB into the North Sea (Ernst, 1986). In coastal waters, levels of HCB are higher in the vicinity of estuaries (Ernst, 1986) and contaminated harbours (Abassy *et al*, 2003). Studies have been carried out of the input of HCB into coastal waters, and these have established that riverine input or land-based sources are more dominant in the near shore environment, but atmospheric input is more important for the open ocean (GESAMP, 1985). The Gulf of Mexico was estimated to receive 145 kg/year HCB via suspended sediment particles of the Mississippi River (Rostad *et al*, 1999). In the North Sea, as distance from the coast increased, HCB concentrations decreased from hundreds of pg/l to 50 pg/l or less (Ernst, 1986).

Once a contaminant has been introduced into the marine environment, it can be dispersed by two groups of processes: advection and dilution. Advection transports contaminants from one place to another, while dilution reduces its concentration by mixing (Williams, 1979). Eventually, HCB is permanently deposited in the sea floor sediments, but only after cycling through the water, biota, sediments and ice, perhaps many times.

Levels of HCB in seawater are generally low, ranging from 700 pg/l in the German Bight in the North Sea, to 2 pg/l in the southern Atlantic Ocean (see Annex 4). HCB has been found to be one of the largest contributors to total organochlorines in seawater at high latitudes (Barrie *et al*, 1997), because it has a higher transport potential than other OCs. Levels of HCB in seawater in 1986-87 near the Canadian Archipelago were 22 pg/l (Hargrave *et al*, 1989), with lower values of 19 pg/l at Ice Island (Beaufort Sea, Hargrave *et al*, 1988). Levels were significantly lower (approx 9 pg/l) 175m below the surface than between 0-60m (approx 19 pg/l), possibly due to river input and exchange of the surface water with the North Atlantic and Bering Seas (Hargrave *et al*, 1988). Levels of HCB in seawater at Barrow Strait/Resolute Bay in 1992 in the Canadian Arctic averaged 15 pg/l (range 14-18 pg/l), and were relatively constant throughout the year (Hargrave *et al*, 1997). Similar values were found in 1993 in the Bering/Chukchi Seas (average 15 pg/l, range 12-17 pg/l) by Strachan *et al* (2001). Wodarg *et al* (2004) performed an extensive baseline survey of HCB concentrations in Baltic Sea water. Concentrations averaged 5 pg/l, and were very uniform throughout the Baltic Sea, with 95% confidence intervals for the 13 transect samples within ±10% of the mean (Wodarg *et al*, 2004). Sea water HCB concentrations from a cruise in the Eastern Atlantic in 1998 range from 4.3-7.7 pg/l in the Northern Hemisphere to 2.2-2.4 pg/l in the Southern Hemisphere (Booij, personal communication), again suggesting that the global redistribution of HCB may be limited by interhemispheric mixing.

In the late 1980s, levels of HCB in Arctic seawater were in an apparent condition of supersaturation in relation to air concentrations, leading to a resulting flux out of the sea (Cotham and Bidleman, 1991). More recently, Booij and van Drooge (2001) studied levels of HCB in air, seawater, and at the sea-air boundary during autumn and winter at a coastal location in the Western Wadden Sea using SPMDs. HCB concentrations in the atmosphere were about eight times higher than the predicted equilibrium concentration, so were oversaturated with respect to sea concentrations. Over the period of decreasing temperature from October 1996 to January 1997, HCB air concentrations decreased by a factor of 10, and water concentrations increased by a factor of two. This suggests that there was a net movement of HCB into the sea from the air in the winter. There was no difference in HCB concentration between the sea-surface micro-layer (top 5 mm or so) and the water column (3m depth), suggesting that HCB in the ocean surface is well mixed. Although the concentrations of HCB in seawater are low, the volume of the oceans is very large, and therefore oceans may be important sinks for HCB. Transport *via* ocean currents may thus be a significant transport mechanism for HCB between regions.

Several studies have reported HCB contamination in lakes. Lakes fall into two categories: those that have been directly contaminated by HCB emissions, and those that have received HCB as a result of long-range transport. In the former category, concentrations can be as high as in contaminated rivers, with concentrations of up to 100 ng/l reported in lakes in Egypt and Zimbabwe. In the latter category, concentrations are low, with concentrations ranging from <10 pg/l in remote mountain lakes (Vilanova *et al*, 2001) to 80 pg/l in Arctic lakes (Cotham and Bidleman, 1991). HCB concentrations of 22 pg/l have been reported in snow meltwater that feeds Arctic lakes (Gregor and Gummer, 1989). HCB levels in European remote mountain lakes exhibited a uniform geographic distribution, in contrast to currently used pesticides, which differed between the sites (Vilanova *et al*, 2001).

4.4 Sediments

The adsorption of HCB onto particulate matter and sediment is an important mechanism for its removal from the water column. Consequently, the sediment component of aquatic ecosystems can be a significant sink for HCB. Chemical or biological degradation is not considered to be important for the removal of HCB from water or sediments (Mansour *et al*, 1986; Mill and Haag, 1986; Oliver and Carey, 1986). Suspended particles entering slow moving waters, such as large water bodies and estuaries, settle out, and their associated burden is added to the existing sediment load. Strandberg *et al* (1998a) measured settling fluxes for HCB on particulate matter in the Baltic Sea, and from this estimated a total annual sedimentation of HCB in the Baltic Sea of 840 kg, with rates approximately three times higher in the Bothnia Sea than Bothnia Bay. Vertical fluxes of particle-bound HCB in the Mediterranean Sea were analysed in 1981 by Fowler *et al* (1986). HCB concentrations in suspended particles ranged from non-detectable to 7.3 ng/g dw. The vertical flux of HCB to a depth of 100m averaged 59 pg/cm^2/year, whereas the flux to sediments at 250m averaged 7.3 pg/cm^2/year (Fowler *et al*, 1986). This indicates that as much as 87% of the particle-associated HCB is consumed, dissolved or degraded before burial into sediments. A residence time of between 1.5 and 135 years was estimated for HCB in the Mediterranean coastal water column (Fowler *et al*, 1986).

Once in the sediments, HCB tends to accumulate and become trapped by overlying sediments (Oliver and Nicol, 1982). Although HCB is not readily leached from sediments, slow desorption does occur and may be an important and continuous source of HCB back into other environmental compartments, even if inputs to the system cease (Oliver *et al*, 1989). It has been shown that mineralogical particles have a low absorption capacity and a low retention capability for HCB in water (Wilken and Wirth, 1986), whereas organic particles have a much higher absorption capacity for HCB, and a low tendency to release HCB back into the water column. It has, however, been shown that HCB can be desorbed back into the water when the sediment is resuspended artificially in the laboratory (Booij *et al*, 2003). Desorption from sediments has been described by a three compartment model, consisting of a labile desorption compartment from which HCB can be readily and reversibly desorbed, and two entrapped, or 'irreversible' compartments, where desorption is hindered by organic matter (Kan *et al*, 1998; Kan *et al*, 2001). Historically contaminated sediments will have completed the rapid desorption phase, and any further desorption will therefore be 'slow'. Desorption of HCB in sediment from the two 'irreversible' compartments have been observed to have rate constants of 0.31 per day (a half-life of days) and 0.0021 per day (a half-life of years, Kan *et al*, 2001). The fractional mass of HCB in the first irreversible compartment is reported to be negligible (Kan *et al*, 2001), and therefore desorption of HCB from sediments can be considered to have a half-life of years. Cornelissen *et al* (1997) observed rate constants ranging from 0.0033-0.0054 per hour for the slow desorbing fraction, which made up greater than 90% of the mass of HCB in contaminated sediment from Delfzijl, The Netherlands. Ten Hulscher *et al* (2004) measured rate constants of very slow desorption of 0.00016-0.00019 h^{-1} at 10ºC and 0.00023-0.00027 h^{-1} at 20ºC for contaminated sediment from Lake Ketelmeer, The Netherlands. The activation energy of the slow desorption process was calculated to be 31 kJ mol^{-1} (Ten Hulscher *et al*, 2004).

Studies have reported HCB concentrations in sediments, from riverine, estuarine, lacustrine and marine environments (see Annex 5). Levels of HCB in sediment in areas remote from sources are low, with values of <100 pg/g reported from the Persian Gulf region (Al-Majed *et al*, 2000), <130 pg/g dw and <20 pg/g dw from Pacific islands of Vanuatu and Tonga (Harrison *et al*, 1996), and from <10–8240 pg/g dw in Fiji (Morrison *et al*, 1996). HCB in these areas would have been deposited following long-range transport. Levels of HCB in sediments from lakes in a North-South transect through Canada were significantly correlated with latitude when expressed on an organic matter basis, but not on a dry weight basis (Muir *et al*, 1995a). This is further evidence for long-range transport of HCB and cold condensation in polar regions.

Generally, HCB concentrations in source regions of the world average about 1 ng/g dw, with some 'hot spots' found close to sources (see Annex 5). In the Black Sea, levels of HCB in sediment are generally low (<0.2 ng/g dw), but with high values up to 23 ng/g dw found close to the River Danube in Romania (Fillmann *et al*, 2002). In the Caspian Sea, higher levels of HCB (up to 600 pg/g dw) were observed in the Azerbaijan sector than in Russian, Iranian or Kazakstan sectors, which had concentrations <100 pg/g dw (de Mora and Sheikholeslami, 2002).

HCB is widely distributed in sediments in the Western Mediterranean, at levels below 1 ng/g in the coastal shelf (Prats *et al*, 1992; Tolosa *et al*, 1995). Hot spots have been reported at the Gulf of Fos, the Rhone and the Ebro Deltas (30-39 and 19 ng/g dw respectively). HCB residues in sediments in the River Nile have been reported in the range of 3.8-4.5 ng/g dw (Yamashita *et al* 2000), with higher levels ranging from 5-60 ng/g dw in coastal bays (Abd-Allah and Abbas, 1994). HCB was found in shallow sediments of the Venice Lagoon (Italy) at concentrations ranging from 0.85-1.11 ng/g dw (Bernstein *et al*, 2002) and in river sediments at levels ranging from <0.1-4.8 ng/g dw. In the early 1990s, concentrations of HCB at Lake Baikal in the Russian Federation ranged from 5-160 pg/g dw in the lake sediments (Iwata *et al*, 1995). A recent study in the Philippines (Lee *et al*, 1997a) reported HCB levels in river sediments of 0.6-4.5 ng/g (mean 2.8 ng/g).

Some very high HCB sediment concentrations have been reported in industrial areas of Europe, the US, Asia and South America. Very high levels of HCB were detected in sediment and soil in the Ya-Er Lake area in China (Wu *et al*, 1997), where from 1962 to 1987 a factory that primarily produced HCB discharged large volumes of contaminated wastewater into this lake. High levels of HCB were found in sediment and clams collected in the Ca Long estuary in Vietnam, a densely populated rice producing area where HCH and lindane are still being used extensively (Nhan *et al*, 1999). Levels at this site were up to 150 times higher than in other coastal areas.

High levels of HCB were found in sediment (mean of 871 ng/g) offshore from the Malabar outfall in Sydney Australia, which contains a large proportion of industrial waste (Mortimer and Connell, 1995). Lee and Fang (1997) found that HCB concentrations were an order of magnitude higher in marine sediments near industrial and urban areas than in remote areas from Taiwan in 1995. Levels of HCB in sediments in South America range from 0.1-22 µg/kg dw (with an extreme value of 22200 µg/kg dw), with an average of 6 µg/kg dw (3701 µg/kg dw if high values are included, UNEP, 2002d). The ‘hot spots’ are principally from cities in Brazil (Del Grande *et al*, 2003).

The Great Lakes region is historically considered to be the dominant source region for HCB in North America. Spatial studies of HCB in Lake Erie sediments showed that the highest concentration was near the outflow of the Detroit River in both 1971 (60 ng/g dw) and 1997 (12 ng/g, Marvin *et al*, 2004). The majority of HCB entering Lake Erie has been attributed to waterborne inputs (Cohen *et al*, 1985). In The Netherlands, large amounts of HCB were drained in the period 1969-1986 into the Sea Harbour Channel from Delfzijl, due to industrial activities near the channel. This resulted in a relatively high HCB content in the channel sediment (up to 6000 ng/g). In 1994, a large dredging operation was carried out to clean-up this site, resulting in a 97% reduction of HCB content. The dredged material was disposed in an erosion area of the Ems-Dollard estuary, where it is being diluted with suspended matter and transported to other areas (Eggens and Bakker, 2001). Booij *et al* (2003) measured HCB in sediment around a wastewater discharge area in a harbour in the Ems Estuary, The Netherlands. Concentrations were up to six times higher in the immediate vicinity of the outfall than at a reference site, but dropped back to background levels 200 metres out into the estuary.

4.5 Soil

Several studies have reported soil HCB concentrations, both in contaminated industrial areas and remote global background sites (see Annex 6). Soils are natural sinks for persistent and lipophilic compounds such as HCB, which adsorb onto the organic carbon and remain relatively immobile in this reservoir (Mackay, 2001). The relationship between soil and air concentrations under equilibrium conditions is described by K_{SA}, the soil/air partition coefficient. Reported log K_{SA} values for HCB range from 7.27 (Meijer *et al*, 2003b) to 4.98 (Hippelein and McLachlan, 1998) to 5.68 and 6.35 (Meijer *et al*, 2003b). Thus, although the global volume of soil is considerably less than the volume of air, at equilibrium, soil will contain a much greater mass of HCB than air.

Soil is a typical ‘long-memory’ accumulating matrix, and therefore HCB inputs received in the past will persist with very little clearance and long half-lives. The concentrations in soil tend to reflect the baseline contamination of a region. Soils receive inputs of environmental pollutants *via* different pathways, including direct pesticide application, atmospheric deposition, application of sewage sludge or compost, spills, erosion from nearby contaminated areas and contaminated water irrigation. Therefore, urban areas exhibit higher concentrations than rural, at least for HCB released by industrial activities alone, but obviously higher levels of pesticide-derived HCB are expected in soils treated historically with these substances. An HCB ‘hot spot’ was reported for Canoas in Brazil, with soil concentrations of 6480 ng/g dw reported (CETESB, 1997). This high value is associated with its proximity to an organochlorine waste landfill site. Very high soil, sediment, and water HCB concentrations were observed at Bitterfeld, Germany, an industrial area with numerous unsealed chemical waste dumps, with soil concentrations averaging 3276 ng/g dw (Vrana *et al*, 2001). Very high soil HCB concentrations averaging 638 ng/g dw were reported at Lake Päijänne, Finland, around a lake contaminated by discharge from industry (Rantalainen *et al*, 1998). Concentrations of HCB in soil were significantly higher at one site near an OC producing factory (337 ng/g) than at other Romanian locations (0.3-1.9 ng/g, Covaci *et al*, 2001a). The reported reference value for HCB in soils is 2.5 ng/g dw, and this is exceeded significantly at these sites. (Manz *et al*, 2001).

Levels of HCB reported from remote sites are much lower, but the presence of HCB in these locations indicates how mobile it is in the environment. Concentrations of soil on mountains on Tenerife Island averaged >0.5 ng/g dw (Ribes *et al*, 2002), and in the Pyrenees averaged >1 ng/g dw. Buckland *et al* (1998) summarised the results of a detailed New Zealand study on ambient concentrations of HCB in soils. HCB was found commonly, on average at > 0.2 ng/g dw in urban, forest, agricultural soils, with urban soils containing maximum concentrations of 1.22 ng/g. Similar HCB soil concentrations averaging > 0.2 ng/g dw were reported in Australia (Cavanagh *et al*, 1999). Meijer *et al* (2003d) studied HCB concentrations in background surface (0-5 cm) soils collected in 1998 from 191 locations around the world. The global mean background soil HCB concentration was 0.68 ng/g dw. The lowest (0.010 ng/g dw) and highest (5.20 and 4.83 ng/g dw) concentrations were found in samples from Bear Island (Norway), south Norway and Russia, respectively.

HCB concentrations correlated significantly with soil organic matter (SOM) content. The highest concentrations were in the Northern Hemisphere in general and Europe in particular, between the latitudes 55° and 70°N. This corresponds with the location of the soils with the highest SOM content, associated with forests, peat bogs and grassland systems, and is also situated north of the areas where the highest global HCB emissions are likely to have occurred. This is indicative of the strong influence of SOM on HCB distribution, and again suggests that the HCB has been subject to the 'grasshopper effect', with soils with high organic matter (OM) content and lower temperatures providing important global sinks. This relationship suggests that HCB has approached air-soil equilibrium with both low and high OM soil, with an appreciable loss from low OM soils occurring, and significant retention in high OM soils (Gouin *et al*, 2004). The global burden of HCB in background soils was calculated to be 1020 t, of which 490 t was in the band 60-90°N, 410 t was in the band 30-60°N, and only 120 t was in the band 90°S – 30°N (Meijer *et al*, 2003d).

Ribes *et al* (2002) found that concentrations of HCB in mountain soils from Tenerife were significantly correlated with the total organic carbon content of the soil, and inversely correlated with atmospheric temperature, giving evidence of cold condensation. The distribution of HCB in the mountain soils suggests steady state/equilibrium conditions for HCB. Enthalpy of phase change (ΔH) for HCB was calculated as 157 kJ/mol, which is higher than most reported values. Meijer *et al* (2003c) found K_{SA} for HCB to be strongly temperature dependent, with ΔH values of 85.6 and 77.1 for two different soils. This compares with a ΔH value measured using K_{OA} of 55.7 (Shoeib and Harner, 2002). These relatively high ΔH values suggest that HCB readily undergoes air/soil exchange. Meijer *et al* (2003b) measured HCB air concentrations in an agricultural area of Canada at 3 cm above the soil and at 150 cm above the soil and found similar values at both heights (64 pg/m^3 *vs* 58 pg/m^3). They concluded that for HCB, the ambient air was at or close to equilibrium with the soil.

In soil, volatilisation is the major removal process at the surface (Kilzer *et al*, 1979; Griffin and Chou, 1981; Schwarzenbach *et al*, 1983; Nash and Gish, 1989), while slow aerobic (half-life of 2.7-5.7 years) and anaerobic biodegradation (half-life of 10.6-22.9 years) are the major removal processes at lower depths (Beck and Hansen, 1974; Howard, 1991). Volatilisation and degradation of HCB will be more rapid in the tropics than in the temperate regions (Wania and Mackay, 1995). HCB has been reported to volatilise from soil at a very slow rate (Sotiriou *et al*, 1994; Wilson and Meharg, 2003), with Wilson and Meharg (2003) reporting that less than 1% volatilisation occurred over a five-week experiment. However, in their experiment, they did not specifically sample surface soil layers, which are likely to have a higher volatilisation rate. Their results give a volatilisation half-life for the whole soil of approximately five years. In addition to the volatilisation of HCB from soil, another important aspect controlling its fate is its mobility in soil. Due to its relatively low water solubility and its high affinity for soil organic matter, leaching of HCB from soil by water is not believed to be a significant process.

4.6 Vegetation

There have been a limited number of studies that report HCB concentrations in vegetation (see Annex 7). Uptake of HCB into vegetation occurs through both gas phase deposition to aerial plant surfaces, and through uptake by roots from the soil, with little or no translocation occurring (Bacci and Gaggi, 1986; Schroll and Scheunert, 1992; Schroll *et a*l, 1994). McLachlan and Horstmann (1998) predict that for semi-volatile compounds with $7 < \log K_{OA} < 11$ and $\log K_{AW} > -6$ there is a forest filter effect whereby forests filter airborne POPs and transfer them to soil. HCB fits into this range of properties at temperatures below 20^{o}C (see Table 2). Plants have a reasonably low bioconcentration factor (BCF) for HCB in comparison with other POPs, with reported values for BCFm (mass/mass, leaf/air BCF in ng/g dry leaf/ng/g of air) ranging from 1300-1900 (Bacci *et al*, 1990; Calamari *et al*, 1991) and values for log BCF (volume/volume, leaf/air BCF in ng/l wet leaf/ ng/l of air) ranging from 5.62-6.92 (Morosini *et al*, 1993; Horstmann and McLachlan, 1998).

In soils that have received high concentrations of HCB from pesticide spraying or application of contaminated sewage sludge, root vegetables such as carrots can accumulate high concentrations of HCB (Wang and Jones, 1994b). Leafy parts of plants can intercept HCB that is volatilising from underlying contaminated soil and therefore also accumulate high concentrations of HCB. Thus, vegetation and vegetables grown on contaminated soil are a source of HCB in terrestrial food chains. Wilson and Meharg (2003) reported plant/soil BCF values (mass/mass, leaf/soil BCF) ranging from 0.12-0.33 µg/g wet leaf/ µg/g of dry soil for grass grown on contaminated soil.

Away from contaminated areas, deposition from the bulk atmosphere is the dominant pathway for plants, and therefore concentrations in these plants are reflective of the atmospheric concentration of HCB in an area. This has resulted in a number of studies that use vegetation as a passive sampler to monitor remote areas (Calamari *et al*, 1991; Jensen *et al*, 1992). Bacci *et al* (1986) measured HCB concentrations of 0.30–2.2 ng/g dw in lichen and moss samples collected from Port Lockroy, Galindez Island and Rothera Point, Antarctica. Weiss (2001) reported levels of HCB ranging from 0.5-0.9 ng/g dw in Norway Spruce needles from a remote region of Slovenia. HCB concentrations in pine needles analysed from the island of Tenerife in 1998 ranged from <0.01 to 0.59 ng/g dw (Villa *et al*, 2003b). The values from Tenerife are generally lower if compared with literature data of Europe (Calamari *et al*, 1994; Simonich and Hites, 1995), but an order of magnitude higher than levels found in mango leaves in Africa (Tremolada *et al*, 1993). There were no significant differences found in HCB concentration between sites at different altitudes on the island, whereas significant differences did occur for DDTs and HCHs. This suggests that the much more volatile HCB is at equilibrium over the entire island. In contrast, Davidson *et al* (2003, 2004) found that the concentrations of HCB in pine and spruce needles in mountains of western Canada were significantly correlated with temperature and altitude. This suggests that cold condensation is occurring in elevated area and is not exclusive to remote polar regions. They calculated an enthalpy of phase change (ΔH) value for HCB of 46 kJ/mol (Davidson *et al*, 2003).

The global occurrence of HCB in plant biomass was investigated by Calamari *et al* (1991). Samples from Africa were generally below detection limits (Bacci *et al*, 1988; Calamari *et al*, 1991). It was observed that polar areas had higher HCB concentrations than tropical areas, due to the volatility of HCB and its consequent movement towards the poles, with condensation in the colder regions. Their data indicated that the observed concentrations of HCB were the result of long-term distribution, regulated by global processes and physicochemical properties more than by direct contamination. In spatial studies using vegetation as a passive sampler, HCB has often been found to have a uniform distribution at the national (Sinkkonen *et al*, 1995a; Herrmann and Baumgartner, 1987) and regional levels (Jensen *et al*, 1992; Holoubek *et al*, 2000). Jensen *et al* (1992) found that in 1986, with a few exceptions, the concentrations of HCB in pine needles in a North/South transect through Europe were very similar. A similar study was carried out in 1989 by Strachan *et a*l (1994), and they too found that HCB was uniformly distributed throughout the sample area, with the exception of higher levels measured in central Germany. Hellström *et al* (2004) also found similar spatial trends in surveys in 1989 and 1990, with concentrations generally ranging from 0.5-4 ng/g dw across Europe, but with elevated levels of up to 10 ng/g dw in 6 of the 72 sites (in Estonia and Germany). Ockenden *et al* (1998) looked at spatial trends of HCB along a latitudinal transect in Norway in 1994 in two species of plants. Concentrations averaged 1150 pg/g dw in pine needles (range 680-2100 pg/g dw) and 3160 pg/g dw in lichens (range 1600-4800 pg/g dw). There was no relationship between HCB concentration and latitude or temperature. In contrast, Simonich and Hites (1995) have shown that a correlation exists between latitude and concentrations of HCB in tree bark samples. Howe *et al* (2004) also found that lipid normalised HCB concentrations in spruce needles in Alaska decreased with latitude, and with distance away from the coast.

Vegetation has been also used to produce emission maps around local point sources. Pine needles from the vicinity of a metal recycling facility in Finland showed elevated levels of HCB (Sinkkonen *et al*, 1995a) that were nearly an order of magnitude higher than the regional average (Sinkkonen *et al*, 1995a). Elevated levels were also found near a pulp and paper mill that carried out waste combustion (Sinkkonen *et al*, 1997). High levels of HCB were found in pine needles in the more industrialised East of China than other parts (Xu *et al*, 2004). Weissflog *et al* (1999) observed that levels of HCB in pine needles in former East Germany increased between 1992 and 1993 as a result of changes in emissions from metal plants and domestic stoves.

4.7 Biota

HCB is persistent in the environment and, because of its lipid solubility, it bioaccumulates in the lipid rich tissues of organisms (e.g. Gabrielsen *et al*, 1995). Accumulation of HCB in vegetation results in the incorporation of HCB into terrestrial food chains. Transfer within the food web leads to biomagnification: the successive increase in concentration of a chemical with increasing trophic levels. Bioconcentration factors (BCFs) of 3000 to >35000 have been reported, along with evidence for biomagnification along food chains (IPCS, 1997). Significant bioaccumulation of HCB in fish, marine mammals, birds, and other animals has been observed. In these species, HCB accumulates significantly in fatty tissues, including fat deposits and the liver, with virtually no biodegradation by the exposed organism. The low solubility of HCB in water and the ease of uptake by organisms result in high BCFs in aquatic organisms. BCFs for a range of aquatic organisms are shown in Annex 8. On a lipid weight basis, HCB levels in freshwater fish are generally higher than levels in terrestrial herbivores and are similar to or higher than levels in mammalian carnivores.

The bioconcentration of HCB in the aquatic environment is the result of both direct uptake from water, and from food chain transfers. For different species, the contribution of these two processes varies depending on how high up in the food chain they are. In small organisms such as phytoplankton, direct absorption of dissolved HCB through the ‘skin’ is a major pathway of uptake because of their high surface area to volume ratio (Chuchiolo *et al*, 2004). However, the primary route of initial entry into the aquatic food chain is through active uptake of HCB-contaminated particulate matter by zooplankton and other filter-feeders. At higher trophic levels, dietary uptake is progressively more important than direct absorption. It has been predicted that POPs with log K_{OW} = 5-7 will be subject to biomagnification (uptake through food consumption) rather than bioconcentration (uptake from water) in water-breathing animals (Mackay and Fraser, 2000). HCB is in this range of K_{OW} and experimental evidence does show that HCB is biomagnified in Antarctic food chains (Goerke *et al*, 2004) and high mountain lake food chains (Catalan *et al*, 2004). At the top levels of the aquatic food web are fish-eating species, such as humans, seabirds, seals and otters. Hop *et al* (2002) investigated the Barents Sea food chain and found that HCB concentrations decreased in the order glaucous gull > kittiwake and harp seal > ringed seal and guillemot > fish and spider crab > zooplankton and ice-amphipods. Biomagnification factors were orders of magnitude higher for homeotherms (seals and birds) than for poikilotherms (zooplankton, ice-fauna and fish).

4.7.1 Plankton and Shellfish

Plankton, shellfish and other invertebrates accumulate HCB from water and particles, and therefore are important sources of HCB in aquatic food chains. Shellfish make an important contribution to the diet of some fishing communities, and may therefore contribute significantly to their total daily intake. There have been studies of HCB concentrations in zooplankton and shellfish (see Annexes 9 and 10). The position of these invertebrates at the bottom of the food chain, coupled with their limited capacity to degrade HCB, means that concentrations in them closely reflect water concentrations. Thus, studies have been carried out using them to map the spatial distribution of contaminants on national and regional levels (see Annex 10). Levels of HCB in amphipods collected in seven lakes spanning a 1300m elevation gradient in Alberta, Canada were positively correlated with altitude and negatively correlated with temperature, indicating that distillation was taking place (Blais *et al*, 2003). This pattern was also observed in other POPs that have vapour pressure > 0.03 Pa at 20°C, but not in less volatile POPs. Levels of HCB in krill were found to be lower in the Antarctic than in the Arctic (Corsolini *et al*, 2001). HCB levels in polychaete worms from the English Channel and the German Bight had similar concentrations, illustrating the homogeneous input of HCB into the sea (Goerke and Weber, 1998). Data collected from the US National Oceanic and Atmospheric Administration’s (NOAA) Mussel Watch Program show that HCB concentrations in mussels from the Great Lakes were more than five times higher than in mussels along marine coasts of the US. Concentrations in mussels from Alaska, however, were no different from concentrations on the Pacific coast of the US (Mearns *et al*, 2002). Cleeman *et al* (2000a) showed that HCB concentrations were higher in mussels from East Greenland than in those from West Greenland. Geographical distribution of HCB levels in mussels in South-East Asia were reported by Monirith *et al* (2003) following a large scale survey covering several countries. Concentrations were generally uniform in this region (0.01-0.1 ng/g ww), with relatively high levels of up to 13 ng/g ww found in samples collected near industrialised cities such as Jiao Zhou Wan and Qing Dao City in China, Bombay in India and Tokyo Bay in Japan.

In a survey of mangrove oysters from along the Pacific Coast of Mexico in 1996, of 22 organochlorine compounds detected, HCB was the pesticide found most frequently (Páez-Osuna *et al*, 2002). High HCB concentrations were found in oysters from those lagoons that are bordered by extensive agricultural lands (911 ng/g and 183 ng/g), possibly indicating that HCB use is still occurring in these agricultural areas. Values were less than 100 ng/g at all other sites, with the lowest HCB concentrations being measured in areas with limited agricultural activities (1.1 and 2.0 ng/g, Páez-Osuna *et al*, 2002). High levels of HCB were measured by Connor *et al* (2001) in mussels in 1994 in the Mersey Estuary in the UK. This area is close to a number of industrial sources of HCB, in particular the UK chlor-alkali industry.

4.7.2 Fish

Fish accumulate chemical substances either directly from the surrounding environment or from their diet, with the largest proportion of HCB intake coming from the diet. Bioaccumulation is affected by factors such as concentrations in prey and food chain length, as well as traits of individual fish, such as growth rate and lipid content. Lipid concentration plays a pivotal role in the bioaccumulation of HCB, with a strong relationship observed between HCB concentration and lipid content. Thus, fatty fish species accumulate more than lean fish species. HCB accumulates in tissues with high lipid content (Niemi, 1983), such as the liver, and therefore this organ is often analysed to determine the HCB content of fish (see Annex 12). Contaminated fish are the primary source of HCB to most humans and aquatic wildlife. Consequently there have been a large number of studies of levels of HCB in fish (see Annex 11). Average HCB concentrations in fish caught around several different countries are as follows: the US 5.8 ng/g ww (Kuehl and Butterworth, 1994), Australia 4.2 ng/g ww (Kannan *et al*, 1995b), Cambodia 0.09 ng/g ww (Monirith *et al*, 1999), India 0.07 ng/g ww, Thailand 0.24 ng/g ww, Vietnam 0.05 ng/g ww, Indonesia 0.05 ng/g ww, and Papua New Guinea 0.03 ng/g ww (Kannan *et al*, 1995b). Generally, fish show higher concentrations in regions with previously high emissions of HCB.

Fish can occupy different positions in the food chain, and therefore the concentrations of HCB can vary greatly between fish species, for example between grazers and predators at the top of long food chains. Bioaccumulation is also lower in ‘fast-growing’ fish compared to ‘slow-growing’ fish. Consequently the use of different fish species to determine spatial and temporal trends is not recommended due to the high variability between species. Useful information, however, can be obtained using single species with broad geographical distributions. Studies have shown that HCB concentration differences occur between fish from different regions. For example, levels of HCB in silverfish in the Antarctic are lower than in the Arctic (Corsolini *et al*, 2001), and levels of HCB in fish from the South Atlantic are lower than those in the North Atlantic and North Pacific (Looser *et al*, 2000). Other authors have reported similar HCB concentrations in North Sea and Antarctic fish (Weber and Goerke, 1996). Studies have reported HCB concentration differences over relatively small spatial ranges. Levels of HCB in polar cod in Greenland have been shown to be significantly higher on the East Coast than on the West Coast (Cleemann *et al*, 2000a), due to the fact that these regions receive air masses from different parts of North America. Muir *et al* (1990b) showed that HCB occurred at higher concentrations in burbot livers at high latitude sites in Canada, which suggests that cold condensation is occurring. Voorspoels *et al* (2004) found that levels of HCB in fish and shellfish in the Belgian North Sea decreased with distance out to sea away from Antwerp, pointing to a higher degree of exposure further upstream.

Of the many species of fish caught for human consumption and oil extraction, cod liver and the oil extracted from it, is probably most highly affected by accumulation of HCB. The concentration of HCB was determined in cod-liver oil originating from the Icelandic North Atlantic and the eastern North Atlantic basin in the 1980s (Falandysz *et al*, 1994). Concentrations in the North Atlantic (110 ng/g lipid) were similar to those from the North Sea (130 ng/g lipid) but HCB concentrations were significantly higher in the Baltic. (Falandysz *et al*, 1994). The concentration of HCB in livers of fish from the North Sea were also closely comparable with those found in fish near the coast of southwest Greenland and in the North Atlantic Sea (Ernst, 1986), whereas HCB levels in fish livers from Tenerife were an order of magnitude lower. Levels of HCB in fish varied in different regions of the Baltic Sea, with fish from the southern part having the highest concentrations (280 ng/g lipid), which were almost double that found in fish from the western part (160 ng/g lipid, Falandysz *et al*, 1994).

Around the Swedish coast, levels of HCB in herring were highest around the Stockholm archipelago (30 ng/g lipid), with lower levels in herring from Bothnian Bay (20 ng/g lipid), and lowest levels of all (about 12 ng/g lipid) on the Swedish west coat, outside the Baltic (Bernes, 1998). Even within a much smaller area, the Gulf of Gdansk in the southern part of the Baltic Sea, there was variability in HCB concentrations in fish. Stickleback sampled from a site impacted by industry in the Gulf of Gdansk had elevated concentrations of HCB (40 ng/g lipid) compared with three other sites in the Gulf where concentrations ranged from 18-28 ng/g lipid (Falandysz *et al,* 1997). Herring and pike from the same site also contained elevated concentrations of 82 and 270 ng/g lipid, respectively, while they were between 18 and 33 ng/g lipid for the other sites (Strandberg *et al*, 1998b). Levels of HCB should be consistent throughout the Baltic Sea if aerial deposition is the only source. Clearly there are other routes by which HCB continues to enter the Baltic Sea.

Fish and invertebrate samples collected from deep-sea areas have been used to determine how well mixed HCB is between surface waters and deeper waters. Looser *et al* (2000) found that HCB concentrations were higher in deep-water fish than surface fish in the North Atlantic, the South Atlantic and the North Pacific, and suggested that this was caused by higher HCB pollution from the 1970s and 1980s having reached and contaminated the deep-sea environment. In contrast, Lee *et al* (1997b) found similar levels of HCB in coastal water and deep-sea organisms from the West Coast of Japan, and suggested that this implied that a continuous input of HCB was occurring in these waters. De Brito *et al* (2002c) found levels of HCB were higher in surface-water fish than deep-sea fish from off Tohoku, Northeast Japan. These observations suggest that concentrations of HCB in the water column decrease with depth in this region. Concentrations were also lower in deep-sea fish collected from the North Pacific near Japan than those collected in Japanese coastal waters (Takahashi *et al*, 2000). Migrant fishes that come up to the surface had higher HCB levels than those that remained in deep waters (Takahashi *et al*, 2000). HCB was only a minor contaminant in deep-sea fish from off the coast of Greenland (Berg *et al*, 1997) and in a Norwegian Fjord (Berg *et al*, 1998). Deep-sea organisms generally show no consistent trend between HCB content and prey-predator relationships (Takahashi *et al*, 1998; Berg *et al*, 1998).

4.7.3 Reptiles and Amphibians

There have been a few studies that reported concentrations in reptiles and amphibians. The results of these are shown in Annex 13. These have shown that HCB bioaccumulates in these animals, with highest levels located in the liver and eggs. However, these are not an important food item for human consumption, and levels detected are not likely to cause adverse effects to the animals themselves.

4.7.4 Aquatic Birds

A number of studies have investigated levels of HCB in birds (see Annex 14). There are a host of different water birds and birds of prey that consume fish as the main constituent of their diets. As a consequence, many of these species become exposed to contaminants accumulated in their prey. Removal of these pollutants from their tissues relies on metabolic degradation of compounds and excretion through urine and faeces. Removal from avian tissues can be very slow, often slower than the rate of uptake, leading to a gradual increase in tissue residues of toxic compounds through life. Therefore, older animals can have very high concentrations of these substances in their organs and tissues. Birds may remove HCB from their body when they lay eggs, and these can contain high levels of HCB (see Annex 15).

When an aquatic food chain involves piscivorous birds, final biomagnification turns out to be very large, and therefore birds that feed from the sea tend to have higher HCB concentrations than those that feed on land (Hoshi *et al*, 1998). Top avian predators of aquatic food chains, including raptors such as the osprey and bald eagle, prey on fish that are also predators, such as trout and salmon, whereas smaller fish-eating wildlife (such as kingfishers) tend to feed on the smaller forage fish. Birds that feed higher up the food chain have higher levels of HCB in their eggs than birds that feed lower down (Becker *et al*, 2001). Buckman *et al* (2004) looked at HCB levels in seven species of Arctic seabird. Concentrations varied between seabird species with the highest concentrations in the seabirds at the higher trophic levels. Concentrations were greater in fat than liver for all seabird species.

Predatory birds such as cormorants, eagles and guillemots provide a useful means of monitoring environmental contamination by organochlorine compounds. In Canada, the double-crested cormorant, a predator that eats fish, is used as a national indicator of persistent organochlorine levels because of its broad distribution across southern Canada, especially in areas of concentrated human activity (Yamashota *et al*, 1993). Fish-eating birds such as herring gulls (Pekarik and Weseloh, 1998), ospreys (Elliot *et al*, 2000), bald eagles (Elliot *et al*, 1996), and Caspian and common terns (Yamashota *et al*, 1993) are top predators in the Great Lakes ecosystem and these birds have been used to monitor contamination of HCB in the Great Lake region. Guillemot eggs have been used in the Swedish Contaminant Monitoring system because they provide measure of toxic pollutant levels in the Baltic Sea as a whole (Bignert *et al*, 1998; 2004). Guillemots obtain their food from numerous different populations of herring and other fish and the levels of contaminants in the eggs therefore provide a useful indication of those occurring in the Baltic fish populations. Glaucous gulls and kittiwakes have also been used for monitoring pollutants in Scandinavia (Savinova *et al*, 1995).

Comparisons can be made between bird populations both regionally and from different parts of the world. Levels of HCB in cormorants in the Aleutian Islands increase from East to West, indicating that long-range transport from Asia is probably the source of HCB to this area (Rocque and Winker, 2004). Further north, levels of HCB in common murre eggs are significantly higher in the Gulf of Alaska than in the Berring Strait (Pol *et al*, 2004). Braune (1993, 1994) found significantly higher levels of HCB in thick-billed murre eggs from low Arctic colonies than high Arctic colonies. Spatial trends in migratory birds are more complex, since their levels of HCB also reflect their overwintering grounds (Muir *et al*, 1999). Levels of HCB in eider ducks, for example, from the low and high Arctic are higher than those from the western Arctic, because the former overwinter in the ‘contaminated’ St. Lawrence Estuary, whereas the latter overwinter in less contaminated northern waters (Muir *et al*, 1999).

Levels of HCB in albatrosses in the Southern Hemisphere are an order of magnitude lower than levels in the Northern Hemisphere (Guruge *et al*, 2001). Donnewald *et al* (1979) found HCB concentrations in subcutaneous fat of penguins sampled at the Antarctic Peninsula ranging from 87-600 ng/g fat. Luke *et al* (1989) analysed eggs from 15 species of seabirds at Davis and Casey stations (1981-83) on the coast of Eastern Antarctica, and from Macquarie Island (1978-83). Levels of HCB were similar in all penguin eggs (10-20 ng/g fresh mass). Levels in Canadian Arctic seabirds were lower than European Arctic seabirds (Borgå *et al*, 2001). Levels of HCB in guillemots in East Greenland, are similar to levels in Iceland, the Barents Sea and the Canadian Arctic, but levels are lower in West Greenland (Vorkamp *et al*, 2003). An average of 470 ng/g lipid HCB has recently been reported for cormorant tissues in the Baltic Sea (Falandysz *et al*, 2000a). This is significantly lower than the residues found in white-tailed sea eagles, for which a maximum concentration of 9200 ng/g lipid was recorded for one individual from the Baltic south coast (Strandberg *et al*, 1998c). As with many other compounds analysed, eagles from coastal areas had very much higher residues of HCB in their tissues (3300 ng/g lipid) than did birds from inland areas (190 ng/g lipid), highlighting the increased exposure of birds living in a marine environment (Falandysz *et al*, 2000a). Becker *et al* (2001) reported the spatial distribution of HCB in common tern and oystercatcher eggs in the Wadden Sea in 2000. There were elevated levels of HCB at the Elbe estuary (>100 ng/g lipid) and the inner German Bight (10-20 ng/g lipid). The river Elbe discharges large amounts of HCB (60-250 kg) into the North Sea every year. Levels in the eggs of the Dutch and Danish parts of the Wadden Sea were on a similar and lower level of <10 ng/g lipid (Becker *et al*, 2001). HCB levels were measured in eggs of a variety of different bird species from Lake Baikal Russia (Lebedev *et al*, 1998). Very high concentrations were measured in species that wintered in China (up to 10700 ng/g dw), with much lower concentrations in resident birds (<500 ng/g dw), suggesting that significant sources of HCB exist in China. Levels of HCB in birds from southern India were low, despite more recent use of large quantities of technical HCH mixture in this area (Tanabe *et al*, 1998).

4.7.5 Marine Mammals

Marine mammals such as seals, whales and dolphins accumulate relatively high body burdens of HCB. Levels of HCB in marine mammals have been reported for decades throughout the world's ecosystems (see Annexes 16, 17 and 18). Marine mammals are susceptible to the accumulation of HCB in their tissues due to their high trophic level, high lipid content and longevity, and the passage of HCB from females to their young.

HCB accumulates in the thick layer of fatty blubber and, once retained in such fat-rich tissue, they are not easily eliminated. HCB may be released from fat, however, both during periods of starvation or weight loss and during lactation. Marine mammals transfer large amounts of fat soluble and persistent substances to their offspring during periods of lactation that may last from a couple of weeks (certain seal species) to two years (polar bear). The milk has a fat content ranging from 30-60%. Thus, marine mammal offspring are potentially exposed to a large amount of fat-soluble substances during periods of development and maturation of vital organs. HCB concentrations in females tend to be significantly lower than in males (Krahn *et al*, 1997; Gauthier *et al*, 1997; Cleemann *et al*, 2000b).

Concentration of HCB tends to increase with age in males, and decrease in the blubber of mature females, due to reproductive transfer to offspring during both gestation and lactation (Borrell *et al*, 1995). Levels of HCB differ tremendously between mammal types - for example between seals, whales and dolphins - but they also differ considerably between species. This is due to differences in diet, longevity, blubber content and metabolic activity. For example, harbour (common) and hooded seals have lower HCB concentrations than harp and grey seals (Hobbs *et al*, 2002). Harbour porpoises feed at high levels in the food chain and have a small body size.

The geographical distribution of HCB has been studied using levels in marine mammals. Overall, the Southern Hemisphere has a lower level of pollution. HCB levels in marine mammals off South America, South Africa and Australia are invariably low (Aguilar *et al*, 2002). However, even in a moderately polluted region like the Southern Hemisphere, certain restricted areas or localities, like river mouths or semi-enclosed water masses, may be especially contaminated as a result of point sources of pollution.

The HCB levels in a number of species of dolphins sampled in the North Pacific and Indian Ocean were higher in animals inhabiting cold and temperate waters than in those inhabiting tropical waters, a result that was perhaps reflective of atmospheric transport away from tropical areas towards the northern sinks (Prudente *et al*, 1997; Struntz *et al*, 2004).

Levels of HCB are comparatively high in Baltic Sea seals, reflecting the proximity to industrial and agricultural sources and the generally high degree of contamination of the Baltic Sea. Levels of HCB in harbour porpoises in the southern Baltic Sea are higher than those reported from Danish and Norwegian Waters (Strandberg *et al*, 1998c). Bruhn *et al* (1999) found that HCB levels in harbour seals decreased in the order Baltic Sea > North Sea > Arctic, with significant differences observed between the three areas. Levels of HCB in whales in the Arctic however, do not differ significantly from those in the St Lawrence River (Muir *et al*, 1990a).

4.7.6 Terrestrial Mammals and Birds

The most significant input of HCB in terrestrial food chains comes from the deposition of airborne HCB to vegetation. Bioaccumulation of HCB along terrestrial food chains occurs, with higher levels of HCB found in omnivorous mammals than herbivores, and in raptors than in omnivorous birds (Hoshi *et al*, 1998).

Several studies have looked at levels of HCB in terrestrial mammals and birds (see Annex 19). For most terrestrial birds and mammals in the Arctic, HCB is one of the most abundant POPs present (Thomas *et al*, 1992). The bioaccumulation of HCB in the lichen/caribou/wolf food chain in the Canadian Arctic has been well documented (Muir *et al*, 1997). Caribou have a winter diet of lichen, which bioaccumulate HCB from the air. HCB is one of the predominant OC contaminants in arctic air, and therefore HCB is also one of the predominant OCs observed in caribou, at levels in the range 20-130 ng/g lipid, which is higher than Total PCB levels (Muir *et al*, 1997). This suggests a direct air-plant animal pathway into the terrestrial food chain. Levels of HCB in caribou in the Canadian Arctic vary spatially, with levels in the west generally a factor of 2-5 lower than levels in the east.

Caribou are a staple source of food for people and wolves in the Arctic, and HCB has been shown to bioaccumulate from caribou to wolves. Levels of HCB were measured in the livers of wolves in 1998 and arctic foxes in 2000, with values of 31 ng/g lipid and 30.4 ng/g lipid reported, respectively (Hoekstra *et al*, 2003).

HCB levels in red foxes in Italy were higher in an agricultural area than in two urban areas (Corsolini *et al*, 2000), suggesting that agriculture was a dominant use for HCB in Italy. Levels of HCB in red foxes in Quebec were three times lower than those reported for Italy (de March *et al*, 1998). Concentrations of HCB in mammals are higher in adipose or liver tissue than in muscle, except in red foxes and sperm whales, where muscle has a higher concentration (Corsolini *et al*, 2000).

Polar bears are omnivores that consume terrestrial mammals and seals. They are therefore at the top of both terrestrial and aquatic food chains, and have very high HCB burdens. Geographical distribution of HCB in polar bear liver in the Canadian Arctic is uniform, with levels in high Arctic zones not significantly different from those in Hudson Bay (Norstrom *et al*, 1988). This result is consistent with the finding of lower geographical variation of HCB in air and seawater in the northern latitudes than in tropical areas (Iwata *et al*, 1993).

4.8 Foodstuffs

Because of its high octanol/water partition coefficient, HCB is associated chiefly with fatty tissues, and human intake is therefore governed largely by the proportion of fatty fish, animal fats and dairy products in the diet. Foods treated with HCB-contaminated pesticides also tend to have higher levels of HCB. Many of the species discussed in previous sections are consumed by people, and the rule that the higher the trophic level an organism feeds at, the higher the HCB consumption, applies also to humans. Highest dietary intake will thus result from consumption of top predators from aquatic food chains, such as whales and seals, which occurs in some aboriginal populations. High dietary exposure will also result from consumption of fish from contaminated areas such as the Baltic Sea (Sjödin *et al*, 2000).

Virtually all (>98%) of the estimated intake of HCB by members of the Canadian general population is *via* food, primarily from such dairy products as milk, butter and ice cream, and to a lesser extent from fresh meat and eggs and peanuts or peanut butter (Government of Canada, 1993). The daily intake of HCB in Canada in 1992 was estimated to be 0.0028 µg/kg bw/day for adults and 0.214 µg/kg bw/day for breast-fed infants. In Inuit people, who consume large quantities of marine mammal flesh and blubber, the daily intake in adults is estimated to be 0.092 µg/kg bw/day. Levels were mostly much less than 1 ng/g on a wet weight basis.

An extensive survey was carried out recently in Spain in 2000-2002 to investigate levels of HCB in food in Catalonia (Falcó *et al*, 2003). Levels in vegetables, tubers, fruits, pulses and milk were all > 0.02 ng/g ww. Higher levels between 0.02-0.4 ng/g ww were found in meat and meat products, oils and fats, eggs, yoghurt, seafood and most fish. The highest levels were recorded in sardines (0.8 ng/g ww) and cheese (1.7 ng/g ww).

Levels of HCB in various foodstuffs have also been reported for Vietnam (Kannan *et al*, 1992b), Australia (Kannan *et al*, 1994), Papua New Guinea (Kannan *et al*, 1994), China (Nakata *et al*, 2002) and the Solomon Islands (Kannan *et al*, 1994). Comparatively high levels of HCB have been reported in milk and dairy products in Argentina, with values of 7 ng/g (Maitre *et al*, 1994) and 0.85 ng/g (Villaamil, 2000), respectively. Similar concentrations of 6 ng/g were observed in butter from Chile (Ministry of Health. Institute of Public Health, Chile). Kalantzi *et al* (2001) carried out a survey of the levels of HCB in butter samples collected from around the world in 1998-99 (see Annex 20). The difference between the highest and lowest concentrations was a factor of 18, in contrast with DDT and HCHs, which varied by only three orders of magnitude. Some regional variation was apparent, with particularly high levels recorded in samples from the Czech Republic, Austria, China and Italy (Kalantzi *et al*, 2001).

Butter is clearly of varying importance as a source of HCB in the diet around the world. Levels of HCB in adipose in bovine and swine were significantly lower in swine in Northern Sweden than Southern Sweden (Glynn *et al*, 2000a).

Daily intake of HCB in food has fallen in many countries, with higher values reported in the 1980s for some industrialised countries than at present. For example, levels in the 1980s were: Finland, 0.014 µg/kg/day (Moilanen *et al*, 1986); The Netherlands, 0.014 µg/kg/day (Greve, 1986); Japan, 0.002 µg/kg/day; the US, 0.004 µg/kg/day; and the UK, 0.002 µg/kg/day (Burton and Bennett, 1987). In the 1990s, Vaz (1995) reported a dietary HCB intake in Sweden of 0.005 µg/kg/day, an intake of 0.002 µg/kg/day was reported in The Netherlands (Brussard *et al*, 1996), and an intake of 0.003 µg/kg/day was reported in Germany for 1995 (Wilhelm *et al*, 2002). Average daily intakes of HCB in Vietnam, Thailand and Papua New Guinea were 0.0014, 0.0018 and 0.0001 µg/kg/day (Kannan *et al*, 1992b; 1994), and in China was 0.0007 µg/kg/day (Nakata *et al*, 2002).

From the Spanish Food survey in Catalonia in 1988-90, average daily intakes of HCB averaged 0.0029 µg/kg/day (Urieta *et al*, 1996). Results from the Spanish Food Survey in 2000-2002 ranged between 0.0017 and 0.0024 µg HCB/kg body weight/day (Falcó *et al*, 2003). The daily intake of HCB by adults is currently estimated to range from 0.003-0.0004 µg/kg/day in various countries (IPCS, 1997). These levels are all well below the recommended safe levels (see Section 6.12), and are considered unlikely to result in any harmful effects.

5 Reported Human Exposure Levels

Exposure to HCB is believed to occur chiefly by absorption across the gastrointestinal tract following the ingestion of contaminated food. The inhalation pathway contributes to about two orders of magnitude less to intake than the ingestion pathway in a non-occupationally exposed population (Burton and Bennett, 1987). HCB is lipid-soluble, which permits passive diffusion from the intestinal contents into intestinal tissue and then to blood, and *vice versa*. The absorption of HCB from the intestine is generally very efficient, with Schlummer *et al* (1998) calculating that 85.4% of ingested HCB will be absorbed when the blood contains no HCB, with this percentage reduced by 0.2% per ng of HCB per g lipid in blood. This is the result of increased diffusion of HCB from the blood to the intestine rather than reduced absorption from the intestine to the blood (Moser and McLachlan, 2001).

Concentration of HCB in the blood is the primary determinant of the degree of absorption, and therefore older individuals with a high prior history of HCB accumulation tend to have lower net absorption. In the blood, HCB is adsorbed onto blood proteins and rapidly transported throughout the body (Mathews, 1986). HCB is therefore widely distributed in mammalian tissues, with preferential distribution to tissues and fluids with high fat content (ATSDR, 2002). In humans, concentrations of HCB tend to increase with age, indicating that bioaccumulation occurs (Bertram *et al*, 1986; Robinson *et al*, 1986; Schlummer *et al*, 1998).

Elimination from animals and humans takes place by excretion *via* faeces (To-Figueras *et al*, 2000), or by metabolism (see Section 3.6) followed by excretion of polar metabolites (and to a lesser extent HCB) in urine (To-Figueras *et al*, 1997). Elimination of HCB from the body following intake is generally slow, and is believed to occur primarily by desorption from the intestinal tract followed by excretion in faeces (Rozman, 1985; To-Figueras *et al*, 2000). Levels of HCB in urine are very low, because lipid-soluble chemicals such as HCB are adsorbed onto blood proteins and are therefore not included in the aqueous portion of plasma that is subject to glomerular filtration (Mathews, 1986). Excretion of more polar metabolites of HCB occurs *via* both urine and faeces. The elimination of HCB from an exposed population has been shown to be very slow (<0.05%/day) compared with to the burden stored in adipose deposits (To-Figueras *et al*, 2000) or 4-6.4% of the total blood level. This slow elimination results in an average estimated whole-body half-life of six years.

Another method of clearing HCB is by passing it onto the next generation *via* lipid rich eggs (Avrahami and Steele, 1972), or mother's milk, as has been shown in rats (Nakashima *et al*, 1997), sheep (Jan *et al*, 1999), seals (Espelend *et al*, 1997; Sørmo *et al*, 2003a) and humans (Peters *et al*, 1966; Jensen and Slorach, 1991; Hooper *et al*, 1997; Shechter *et al*, 1998; Czaja *et al*, 1999; Gladen *et al*, 1999). HCB can also be transferred *via* the placenta to the developing foetus (Siyali, 1974; Ando *et al*, 1985; Skaare *et al*, 1988; Lackmann *et al*, 1996; Sala *et al*, 2001a). In this manner, babies receive a large dose of HCB over a short period of time.

There is a dynamic equilibrium between the levels of HCB in blood and adipose tissue (Needham *et al*, 1990, Jensen and Slorach, 1991). At 'steady state', the levels in blood, muscle, adipose and milk are about the same if calculated on a lipid basis. The levels of HCB in adipose represent a reservoir that is built up during many years of exposure. The blood concentration will depend on mobilisation from the body stores and the current intake of HCB, with current intake most decisive. With lactating women, the situation is different because daily elimination is much greater than the daily uptake, and so adipose tissue supplies most of the HCB to the blood. Breast milk is enriched with HCB, relative to blood, and blood levels actually drop in lactating mothers (Greizerstein *et al*, 1999; Petzold *et al*, 1999). Thus, a high net absorption of HCB in infants might be expected when comparing their relatively low body burden of HCB to the high concentrations in the mother's milk. The levels of HCB in the serum of breast-fed infants are significantly higher than those in bottle-fed infants at six weeks of age (Lackmann *et al*, 2004). The mobilisation of fat, including the HCB in it, during lactation explains the common finding in monitoring of human (and other mammal) adipose tissue that the level of HCB is significantly lower in females than in males. Generally, the transfer of HCB *via* the milk is 10 to 20 more times important than the transfer across the placental membrane (Jensen and Slorach, 1991).

5.1 Background Human Exposure Levels

Owing to its persistence and lipophilicity, HCB is probably present at low levels in the fatty tissues of all members of the general population. Based on representative levels of HCB in air, water and food, the total intake of HCB by adults in the general population is estimated to be between 0.0004 and 0.003 ng/g body weight per day (ATSDR, 2002).

The intake of HCB by adults in the general population is predominantly from the diet (about 90%). In Germany, Schade and Heinzow (1998) showed that women who ate a healthy diet with low meat consumption and high fruit and vegetable intake had much lower levels of HCB in their milk compared to women who ate a lot of meat. Sjödin *et al* (2000) showed that the levels of HCB in Swedish and Latvian men were significantly correlated with the amount of fatty fish from the Baltic Sea that they consumed. A number of other studies have shown elevated HCB concentrations in people with a high consumption of fatty fish (Conde *et al*, 1993; Newsome *et al*, 1995; Albers *et al*, 1996; Kosatsky *et al*, 1999; Hanaoka *et al*, 2002), whereas other studies have found no difference (Fitzgerald *et al*, 2001).

Concentrations of HCB in human milk were three times higher in samples from northern Canada than samples from southern Canada (Newsome and Ryan, 1989). Studies from Greenland and the Canadian Arctic have shown high concentrations of HCB in the blood and fat of the Inuit, compared with Caucasians (Dewailly *et al*, 1993; 1995; 1996; 1999; Bjerregaard and Hansen, 2000). This is attributed to their subsistence hunting lifestyle, which results in high consumption of the meat and blubber of caribou, whales, polar bears and seals (Dewailly *et al*, 1993).

Mean levels in adipose tissue of the general human population in various countries range from tens to hundreds of ng/g ww (IPCS, 1997; see Annex 21). Mean levels in blood of the general human population in various countries are generally less than 1 ng/ml of whole blood (see Annex 22). HCB has also been measured in human follicular fluid (De Felip *et al*, 2004).

Owing to the presence of HCB in breast milk, mean intakes by nursing infants have been estimated to range from <0.018 to 5.1 ng/g body weight per day in various countries. In general, concentrations of HCB in breast milk in various countries or regions range widely, and appear to be related to the degree of industrialisation and/or urbanisation within the survey area (Harris *et al*, 2001; see Annex 23). Historically, areas with less industrialisation have significantly lower levels of HCB in the general population. Significantly higher HCB concentrations have been reported for women living in industrial areas (Quinsey *et al*, 1995), urban dwelling mothers (Horn *et al*, 1994), and mothers living in larger towns (Breivik and Bjerk, 1978).

Higher levels of HCB have also been observed in placental samples from a region of Slovakia associated with the organic chemical industry, than in other areas associated with the metallurgical industry, agriculture and mining (Reichrtova *et al*, 2001), and in blood samples from newborns in metropolitan areas of Germany than in rural areas (Lackmann *et al*, 2001). In China, higher levels of HCB in human milk were found in the North-East of China, near a currently operational HCB factory, than in the South of the country (Kunisue *et al*, 2004). Higher serum HCB concentrations were found in southern Sweden than in northern Sweden (Glynn *et al*, 2003). This may be because southern Sweden is more industrialised and has more agriculture.

In contrast, higher HCB levels were found in human milk in the North of Russia (within the Arctic circle) than in towns further south (Polder *et al*, 2003). In the Arctic, 2-6 times higher concentrations of HCB were found in human serum in Greenland than in other Arctic countries (Hansen, 1998). Higher levels of HCB have been observed in human milk samples from the more industrialised regions of Czechoslovakia (Schoula *et al*, 1996), Poland (Czaja *et al*, 1997b), and Jordan (Nasir *et al*, 1998) than other regions, and in urban regions of Mexico (Waliszewski *et al*, 1996) and Canada (Frank *et al*, 1988) in comparison with suburban regions. A study in 1992 found that HCB levels in the breast milk of women living in the industrialized Kola Peninsula of Russia were twice as high as those in Norway and the Netherlands (Polder *et al*, 1998).

5.2 Elevated (Occupational) Human Exposure

Poisoning incidents in Europe and the US involving HCB have illustrated the importance of diet as an exposure pathway. The most notorious example is the HCB exposure incident in Turkey (see Section 6.3). In another example of the potential for exposure *via* the food chain, cattle from an industrial area of Louisiana were quarantined because of high levels of HCB in their milk and fat (Courtney, 1979). Currently, the use of HCB has been banned throughout the world, and dietary exposure to high levels of HCB is much less likely.

There have been a number of reported cases of high HCB levels in occupationally exposed humans, as a result of inhaling high quantities of HCB (see Annexes 21-23). Burns *et al* (1974) found elevated levels of HCB in plasma of pesticide applicators who had sprayed HCB-contaminated dimethyl-1,2,3,5,6-tetrachloroterephthalate (DCPA) onto vegetables. Levels of HCB ranged from less than 1 to 310 ng/g and averaged 40 ng/g. These workers showed no signs of *porphyria* (a disorder involving skin lesions, caused by an abnormal metabolism of the haem biosynthesis pathway, see Section 6.3) or altered serum enzyme levels.

Burns and Miller (1975) found elevated levels of HCB in plasma of residents living or working in an area of Louisiana where transportation and disposal of chemical waste containing HCB took place. Air samples near a landfill site contained 16 µg/m^3 HCB (US EPA, 1975). Average plasma HCB levels in the general population were 2.4 ng/g, compared with 0.5 ng/g in controls. Blood samples from farm workers in the same area ranged from 14-223 ng/g (Burns and Miller, 1975). However, there was no evidence of cutaneous porphyria.

Four workers in Argentina whose occupation required the handling of HCB showed the prominent signs of *porphyria cutanea tarda*, and the urine of one worker contained increased amounts of both uroporphyrins and coproporphyrins (Mazzei and Mazzei, 1972). This worker had 0.38 ppm HCB in the blood. A year after leaving the job and receiving treatment, the skin lesions subsided, and the blood level declined to 0.268 ppm. Marked changes in kidney function including microproteinuria were observed in Czechoslovakian workers with high HCB levels following occupational inhalation exposure to HCB, originally at 2.1-10.8 mg/m^3 and then at 0.012-0.022 mg/m^3, from 1983 to 1990 (Richter *et al*, 1994).

Elevated HCB blood levels were determined over a four-year period in men employed in the manufacture of chlorinated solvents (Currier *et al*, 1980). Airborne HCB concentrations ranged from <1-13 ppb. Blood levels ranged from 5-1121 ng/g (mean 311 ng/g) in 1974, 30-990 ng/g (mean 312 ng/g) in 1975, 3-600 ng/g (mean 160 ng/g) in 1976, and 22-467 ng/g (mean 170 ng/g) in 1977. HCB blood levels were strongly correlated with the number of years worked in the chlorinated solvent plant. In another study of factory workers from an organochlorine manufacturing plant with HCB air concentrations of 0.035 ug/m^3, serum levels of up to 1616 ng/ml were observed, without any evidence of porphyria (Herrero *et al*, 1999).

Selden *et al* (1997) studied HCB levels in the blood of aluminium foundry workers who used hexachloroethane as a degassing agent for aluminium. Airborne HCB levels in the plant ranged from 0.066-11 ng/m^3. A significant increase of mean HCB concentration was found among the exposed subjects compared with the controls (63 *vs* 35 ng/g lipid). However, no significant difference in kidney, pancreas or liver function was found between the two groups, so although levels were high, there was no evidence of subclinical organ toxicity.

Santos *et al* (2003) studied blood HCB levels in people living on an open-air dump in Piloes, in the municipality of Cubatao, Sao Paolo, Brazil. The average blood level of HCB in Piloes was 4.66 µg/l, which was 155 times higher than the average for the control population living in the same municipality (0.03 µg/l). A significant association was found between dwelling near the dump and the presence of HCB in the blood. Volatilisation of HCB from the contaminated sediments of the Aral Sea in Uzbekistan has been shown to cause elevated HCB levels in maternal blood, umbilical blood and milk samples of local inhabitants (Ataniyazova *et al*, 2001).

Elevated HCB levels have repeatedly been detected among the inhabitants of Flix, Spain, a small village located near a chlorinated solvent manufacturing plant. Grimalt *et al* (1994) found the mean HCB level in serum obtained from village inhabitants was 26 ng/ml, whereas the mean level from a reference population in Barcelona was 4.8 ng/ml. This study produced evidence for elevated cancer incidence in the exposed population (see Section 1.3.3.8). Sala *et al* (1999a) studied the same population in 1994, and found average HCB levels of 36.7 ng/ml. Higher HCB levels were found in workers from the factory (54.6 ng/ml) than non-workers (9-14.9 ng/ml, Sala *et al*, 1999b). Levels in retired workers declined over time.

Another study of the same population showed HCB levels in serum ranged from 1.1-953 ng/ml, with averages of 72 ng/ml in males and 19 ng/ml in females (To-Figueras, 1997), with factory workers averaging 93.4 ng/ml, whilst the Barcelona control population averaged 4.1 ng/ml (range 0.7-9.7 ng/ml, To-Figueras *et al*, 1995). These high concentrations were associated with significantly altered levels of total thyroxine (T4) and γ-glutamyltransferase (GGT) in the blood, indicating that thyroid hormone status was being disrupted (Sala *et al*, 2001b). A later study of the same exposed population yielded HCB serum concentrations ranging from 2.4-1485 ng/ml, the highest HCB blood levels ever reported (To-Figueras *et al*, 2000).

6 Toxicity

Exposure to HCB through ingestion of food contaminated with low levels of the compound is probably the greatest source of exposure for the general population (see Section 5.1). The presence of HCB in tissues has been shown to have a number of adverse affects on animals and humans. HCB toxicity has been reviewed by Courtney (1979) and more recently by the Agency for Toxic Substances and Disease Registry (ATSDR, 2002). A summary of their findings, including recent developments, follows.

6.1 Acute Toxicity

HCB exhibits low acute toxicity in studies involving either oral or inhalation exposure of experimental animals. Oral LD_{50} values range from 1700 mg/kg in cats to between 3500 and 10000 mg/kg in rats, with values for mice, rabbits and guinea-pigs falling between these values (IARC, 1979). LC_{50} values for inhalation exposure range from 1600 mg/m^3 for cats to 4000 mg/m^3 for mice. Acute lethal and near lethal doses cause convulsions, tremors, weakness, ataxia and paralysis.

6.2 Subchronic Toxicity

Short-term repeated dosing experiments in animals have shown that HCB has hepatotoxic and neurological effects, such as altered body weight, cutaneous lesions, hepatomegaly, liver damage, tremors, and alterations in porphyrin or haeme metabolism (Courtney, 1979; Strik, 1986). It induces dioxin-like activity by hepatic microsomal enzymes such as isoforms of cytochrome P-450 (CYP1A1, CYP1A2) and glutathione-S-transferases (den Tonkelaar and van Esch, 1974), as well as phenobarbitol-like activity like induction of hepatic cytochrome P-450 (CYP2B).

HCB has also been shown to disrupt glucose metabolism in rats, by reducing levels of gluconeogenesis enzymes in blood plasma (Mazzetti *et al*, 2004). The induction of splenomegaly, and skin and lung lesions in rats were found to not be caused by metabolites of HCB from the P450 glutathione conjugation pathway, and must therefore be caused by HCB itself or as-yet-unidentified non-P450-generated metabolites (Michielsen *et al*, 2000).

A recent study of gene expression levels in rats after subchronic exposure to HCB using DNA microarrays confirmed known effects of HCB such as stimulatory effects on the immune system and induction of enzymes involved in drug metabolism, porphyria and the reproductive system (Ezendam *et al*, 2004). Increased transcription levels of markers for granulocytes and macrophytes were observed, and new findings included the up-regulation of genes encoding pro-inflammatory cytokines, antioxidants, acute phase proteins, mast cell markers, complement, chemokines, and cell adhesion markers. In general, the gene expression data provided evidence that HCB induces a systemic inflammatory response, accompanied by oxidative stress and an acute phase response (Ezendam *et al*, 2004).

6.3 HCB-Induced Porphyria

Porphyria is the most consistently identified effect following exposure of humans or animals to HCB. It was discovered that HCB induces porphyria in humans during an outbreak of *porphyria cutanea tarda* (PCT) in Turkey between 1955 and 1959 when several populations consumed bread made from HCB-contaminated flour. In this incident about 500 people were fatally poisoned, and about 4000 became sick. The ingested dose of HCB was estimated to be 0.7-2.9 mg/kg/day (Cam and Nigogosyan, 1963). Milk from exposed individuals contained 20-30 µg/g ww HCB with a maximum concentration of 700 µg/g ww (Peters *et al*, 1966). More than 95% of babies under two years old who had consumed highly contaminated maternal milk died within one year, compared to the mortality rate of about 14% in children who were not breast-fed (Peters, 1976).

Symptoms of the illness were neuritis, photosensitivity, fragile and scarred skin and increased excretion of porphyrins (Peters *et al*, 1982). The underlying cause of the illness was disruption of the balance between production and utilisation of various haemoproteins though an inhibition of porphyrin metabolism, specifically uroporphyrinogen decarboxylase.

The mean HCB milk concentration collected in the same area 20-25 years later was 0.23 µg/g ww, with a maximum concentration of 2.8 µg/g ww (Cripps *et al*, 1984). Alterations in porphyrin metabolism continued to be observed in a portion of the previously affected patients. It has been suggested that porphyria is induced by the production of a metabolite of HCB by CYP1A2 enzymes (den Besten *et al*, 1993; Sinclair *et al*, 2000).

6.4 Non-Hepatic Effects

Additional non-hepatic effects have been observed in animal studies involving repeated subchronic exposure to HCB, such as changes in calcium homeostasis and bone morphometry (Andrews *et al*, 1990). Indeed, Cripps *et al* (1984) observed a high incidence (65%) of osteoporosis in the follow-up study conducted 20-30 years after the inadvertent poisoning incident in Turkey, which supports these findings.

6.5 Genetic Toxicity

Studies investigating the genotoxicity of HCB have indicated that it exhibits weak or no genotoxic activity (IARC, 1979). HCB did not cause significant mutagenesis in a number of bacterial and animal studies (e.g. Haworth *et al*, 1983; Gorski *et a*l, 1986), but it did have a teratogenic effect on developing mice and rats, causing enlarged kidneys, hydronephrosis, hepatomegaly, and immune system deficiency (Ecobichon, 1996).

Dubois *et al* (1997) found that HCB-DNA adducts formed in rodent, bird and human hepatocytes exposed to HCB. Canonero *et al* (1997) found that HCB was mutagenic in human cells, producing a weak but significant increase in the frequency of micronuclei (but not DNA breaks) in human hepatocytes at levels that had no effects on rat and monkey hepatocytes. Salmon *et al* (2002) exposed human embryonic cells to the average newborn cord blood concentrations of Alaskan Arctic natives, and observed cell membrane damage, a short-term decrease in cell number, increased DNA strand breaks, and a long-term decrease in colony survival. This demonstrated that relevant environmental concentrations of HCB have genotoxic effects on cultured embryonic cells.

6.5.1 Developmental and Reproductive Toxicology

Studies of reproductive toxicity have indicated that the ovary and testis are target sites for HCB toxicity. Female animals dosed with a high dose of HCB develop altered ovarian steroid levels, as well as alterations in the pituitary hormones, FSH and prolactin (Alvarez *et al*, 2000). In addition to altering function, HCB induced structural changes have been observed in the ovary (Alvarez *et al*, 2000). Male reproduction is also altered by repeated exposure to HCB, with histological changes in the testes and reduced serum testosterone levels reported (Elissalde and Clarke, 1979).

HCB has been shown to cause significant toxicity in the offspring of exposed animals (Arnold *et al*, 1985). Developmental effects, such as altered neurobehavioral development, have been observed in exposed rats (Lilienthal *et al*, 1996). A significantly elevated incidence of stillbirths was observed in humans following the Turkish contaminated-bread incident (Peters *et al*, 1966), but no effect was observed at typical ‘background’ exposure levels (Leoni *et al*, 1986).

Schade and Heinzow (1998) found that high concentrations of HCB in human milk were correlated with a lower birth weight of the nursed child, and Ribas-Fito *et al* (2002) found high HCB umbilical cord concentrations were correlated with increased incidence of premature birth and low birth weight. Hardell *et al* (2003) found that levels of HCB were elevated in mothers of men with testicular cancer, and speculated that high levels experienced by the men in their mother’s womb may be responsible.

6.5.2 Immunotoxicity

A number of studies in experimental animals have shown that HCB affects the immune system. Generally, HCB exhibits immunosuppressive effects in mice and immunostimulatory effects in rats (Michielsen *et al*, 1999a; Vos, 1986). Alterations in the immune system in mice include a reduction in resistance to challenge by tumour cells, a depression of cytotoxic macrophage activity of the spleen, and an increased susceptibility to infection (Loose *et al*, 1981).

In the rat, immunostimulatory effects include elevated serum levels of IgM, peripheral neutrophilic, basophilic and mononuclear leukocytes, and increased spleen, and lymph node weights (Vos, 1986). Inflammation of the skin and alterations to the lungs has also been observed in exposed rats (Michielsen *et al*, 1999b). HCB-induced alterations to the immune system have also been reported in other animals such as rhesus monkeys and dogs.

In humans, evidence for immunotoxic effects of HCB comes from a study of workers in Brazil exposed to HCB while working in a chemical plant producing carbon tetrachloride and perchloroethylene (Queiroz *et al*, 1997, 1998). The study participants in the exposed group showed impaired functions of neutrophilic granulocytes compared to a control group. Increased serum IgM and IgG levels were also observed in these workers, although IgA levels were normal (Queiroz *et al*, 1998).

The levels of HCB in Canadian Inuit infants correlated with increased risk of otitis media in their first year of life, but not with other serum immune parameters (immunoglobulins, cytokines, lymphocyte activation markers, Dewailly *et al*, 2000). The mechanism of action of HCB has been shown to be different to the typical mechanism of binding to macromolecules, with subsequent T- and B-cell activation. Instead, some other complex T-cell mediating activity occurs (Michielsen *et al*, 1999a).

6.6 Neurotoxicity

HCB is reported to cause neurological effects in adult animals both in the presence and absence of evidence of histopathological alterations of neurological structures. At low doses, HCB produces little central nervous system toxicity, but at high doses central nervous system disorders can be the major symptom in the clinical profile. Observed effects include electrophysiological changes, tremors, seizures, muscular weakness, lethargy and unsteady gait, with the extent of effect or severity both dose- and time-dependent. HCB has also been reported to cause neurological effects in humans, with exposed individuals from Turkey reporting muscle weakness, paraesthesia, neuritis and myotonia (Cripps *et al*, 1984).

6.7 Chronic Toxicity

A variety of non-neoplastic effects have been observed in experimental animals resulting from chronic exposure to HCB. The effects observed at the lowest dose levels are primarily hepatic in nature, although effects on the kidney, spleen, and thyroid have also been observed (IPCS, 1997). Hepatic effects include centrilobular basophilic chromatogenesis of the liver, nephrosis and peliosis of the liver, and increases in hepatic lymphocytosis and fibrosis (Arnold *et al*, 1985). Effects associated with sub-chronic short-term exposure, such as porphyria and induction of mixed-function oxidase activity, have also been observed (Grant *et al*, 1974). There is evidence for a link between high serum HCB concentrations and elevated risk of diabetes amongst Swedish women (Glynn *et al*, 2003).

6.8 Carcinogenicity

Studies have been conducted to evaluate the carcinogenicity of HCB in animals, and these have shown significant increases in tumours in various tissues in mice, rats and hamsters. In hamsters, there is a significant dose-related increase in hepatomas, liver hemangioendotheliomas and thyroid alveolar adenomas on a diet of HCB-treated food (Cabral *et al*, 1977). Mice and rats also show a significant increase in the number of liver cell tumours (Cabral *et al*, 1979; Smith and Cabral, 1980), with rats also developing an increased number of kidney tumours (Arnold *et al*, 1985). As many of the organs affected are endocrine organs, it is possible that these tumours are caused by chronic endocrine disruption with compensatory cell proliferation.

Grimalt *et al* (1994) found a significant increase in the number of thyroid neoplasms and soft-tissue sarcoma in men working in an organochlorine compound manufacturing plant in Flix, Spain. Significantly higher levels of HCB were found in Germany, Canada and Sweden in women with breast cancer compared with patients without breast cancer (Güttes *et al*, 1998; Dewailly *et al*, 1994; Liljegren *et al*, 1998), but a number of other studies found no link (e.g. Peters *et al*, 1982; Cripps *et al*, 1984; Mendonca *et al*, 1999; Scheele *et al*, 1996; Zheng *et al*, 1999; Lopez-Carillo *et al*, 2002).

6.9 HCB Dioxin-like Activity

HCB is a compound that binds to the aryl hydrocarbon (Ah) receptor, results in 'dioxin-like effects' (induction of CYP1A1/2, hepatic porphyria and degenerative effects, changes in thyroid hormone levels and metabolism, and reproductive, developmental and immunological effects), and bioaccumulates. These are the three factors for evaluating dioxin-like chemicals using the toxic equivalency factor (TEF) concept, and thus van Birgelen (1998) proposed that HCB should also be evaluated in this way.

HCB binds to the Ah receptor in rats *in vitro* about 10000 times less than TCDD (Hahn *et al*, 1989), and is about 10000 times less potent than TCDD based on *in vitro* cytochrome P4501A induction (Linko *et al*, 1986) and porphyrin accumulation. As a result, van Birgelen proposed a dioxin-like TEF value of 0.0001 for HCB (van Birgelen, 1998). This could add 10-60% to the total TEQ in human milk in most countries, and up to 600% in a few countries such as Spain and the Czech Republic (van Birgelen, 1998).

However, although there is an overlap in the types of effects produced by HCB and TCDD, the evidence that the HCB toxicity is mediated *via* the Ah receptor is limited to *in vitro* studies and therefore inconclusive. Pohl *et al* (2001) agreed with the TEF value recommended for HCB, but pointed out that the occurrence of HCB with TCDD together at hazardous waste sites is very rare, and therefore toxicity of these two chemicals is best considered separately.

6.10 Health Guidelines

Based on studies of toxicity of HCB in animals, health guidelines have been developed which recommend the maximum daily human exposure to HCB that is likely to be without appreciable risk of adverse non-cancer health effects over a specified duration of exposure, and levels at which chronic exposure will cause toxic effects to specific organs (ATSDR, 1996; 2002). These 'minimum risk levels' (MRLs) and 'target-organ toxicity dose values' (TTDs) are listed below:

Acute (1-14 days) oral MRL (based on hepatic effects) = 8 µg/kg/day

Intermediate (15-364 d) oral MRL (based on reproductive effects) = 0.1 µg/kg/day

Chronic (> 365 days) oral MRL (based on developmental effects) = 0.05 µg/kg/day

$\text{TTD}_{\text{endocrine}}$ = 1 µg/kg.day

$\text{TTD}_{\text{immuno}}$ = 0.4 µg/kg/day

$\text{TTD}_{\text{neuro}}$ = 0.8 µg/kg/day

Values are calculated using reported animal 'lowest observed adverse effect level' (LOAEL) values and multiplying them by an uncertainty factor ranging from 100-1000 to take into account: a) extrapolating from animals to humans, b) protection of sensitive individuals, and c) using an LOAEL instead of a NOAEL (no observed adverse effect level).

The average daily intake of HCB in adults is typically below the chronic MRL level in most normal populations (see Table 3 and Section 4.8 on Foodstuffs). Average daily intakes for breast-fed infants have historically been much higher, with Jensen and Slorach (1991) reporting values of 0.3-6.5 for the 1970s and 1980s (see Table 3). For a breast-fed infant consuming 130 g milk/kg body weight/day, with a milk fat content of 3.5%, the milk HCB content needs to be below 11 µg/kg milk fat for a daily intake of less than 0.05 µg/kg/day.

Levels of HCB in human milk are falling, but still exceed 11 µg/kg milk (see Section 5.1), and therefore breast fed infants may exceed the chronic MRL level.

The US EPA has classified HCB as a *probable* human carcinogen, based on sufficient evidence of cancer in studies on animals, but insufficient evidence of cancer in exposed humans. IRIS (2001) presents an oral slope factor of 1.6 per (mg/kg)/day and an inhalation unit risk of 4.6×10^{-4} per (µg/m^3) for HCB based on hepatocellular carcinoma in orally exposed female rats.

7 Observations on the Environmental Fate of HCB

7.1 Geographical Trends in Observed Environmental HCB Concentrations

The large number of articles reporting HCB concentrations makes it possible to develop a picture of the spatial distribution of HCB in the environment. These geographical trends may provide an insight into the transport processes that redistribute POPs such as HCB.

HCB is widely distributed in the environment. The atmospheric concentrations are now fairly uniform, varying by less than an order of magnitude at background sites. This is evidence for its extreme atmospheric persistence and mixing through long-range atmospheric transport. However, the highest HCB air concentrations are still found in historical source regions, indicating that there are some ongoing primary or secondary emissions.

HCB comprises a smaller portion of the total organochlorines (OC) load in animals living in waters close to where OCs are used and released (for example, around the Baltic Sea), but in waters remote from the release site (for example, the open Atlantic Ocean), HCB constitutes a larger proportion of the total OC load (Muir *et al*, 2000; Sørmo *et al*, 2003b). HCB is also one of the dominant OC contaminants in Arctic air and water, comprising more than 20% of the total OC, but the relative fraction declines to less than 5% in polar bears, as other chemicals have higher bioaccumulation factors (Norstrom and Muir, 1994). HCB is generally the predominant POP in Antarctic animals (Goerke *et al*, 2004), and HCB concentrations in dolphins are lower in sub-tropical and tropical areas than in higher latitudes (Prudente *et al*, 1997). Indeed there is much evidence that HCB is subject to global re-distribution in the environment, and to cold condensation (Muir *et al*, 1995a; Blais *et al*, 2003).

The HCB levels reported in biota and humans give important information about the global distribution of HCB. Poor interhemispheric mixing, combined with a much lower HCB usage in the Southern Hemisphere means that levels of HCB are lower in the Southern Hemisphere than the Northern Hemisphere (Looser *et al*, 2000; Aguilar *et al*, 2002), and levels in the Antarctic are lower than in the Arctic (Corsolini *et al*, 2001). Within the Northern hemisphere there is considerable variability in HCB levels. Levels are higher in the European Arctic than the Canadian Arctic (Borgå *et al*, 2001), reflecting the fact that Europe was historically the largest user of HCB. Within Europe, levels are higher in the Baltic Sea than the North Sea and North Atlantic Ocean (Falandysz *et al*, 1994; Bruhn *et al*, 1999). HCB levels appear to be elevated in China, which is a acting as a source region of HCB for surrounding countries (Lebedev *et al*, 1998), in particular the Industrialised North East of the country, where HCB is still manufactured (Xu *et al*, 2004; Kunisue *et al*, 2004). Levels of HCB in Mexico are also elevated, reflecting the fact that HCB was used in agriculture there until fairly recently (Páez-Osuna *et al*, 2002). On the local level, highest concentrations are found near industrial areas (Quinsey *et al*, 1995; Polder *et al*, 1998) and large towns (Horn *et al*, 1994), and levels in the marine environment decrease with distance away from harbours or estuaries (Becker *et al*, 2001; Voorspoels *et al*, 2004).

The 'global distillation' phenomena (Mackay and Wania, 1995; Wania and Mackay, 1995) and long-range transport have been given as a reason for the prevalence of HCB in polar regions (Meijer *et al*, 2003a). The distillation and condensation of HCB on top of cold mountains also occurs (Carrera *et al*, 2002; Ribes *et al*, 2002; Davidson *et al*, 2003; 2004). However, global re-distribution is also a mechanism by which HCB 'finds' new soils of high OM content and global background soils show HCB is strongly correlated with SOM content. It is hypothesised that, over time, HCB will approach a 'global equilibrium', driven by repeated air-surface exchange, to 'find' environmental compartments such as high OM content soils and sediments, and colder regions, where it partitions or 'condenses'.

An important question is to what extent this has already happened, or may be continuing to happen now. This will depend on the timing and magnitude of the primary emissions 'pulse' to the atmosphere, the extent of ongoing (more diffusive) primary emissions, and the potential for soils where HCB has been used in the past to supply the atmosphere. These issues will be explored in the following sections.

7.2 Global Environmental Budget

Significant quantities of HCB are present in the atmosphere, the surface oceans and the terrestrial environment. The volume of the accessible terrestrial environment of soils and vegetation is relatively small (effective depth 5-30 cm) compared to that of the atmosphere (approximately 8000m deep) and the oceans (effective depth of mixed layer 100m). However, the hydrophobic nature of HCB results in its preferential partitioning into soil and plant surfaces from water or air. Using some of the data compiled here, the current global mass balance of HCB in air, soil, water, sediment and plankton is estimated in the subsequent sections.

7.2.1 Air

Annex 25 shows HCB background air concentrations from recent studies. These values have been used to calculate average HCB air concentrations of 55 pg/m^3 and 18 pg/m^3 for the Northern and Southern Hemispheres, respectively. Assuming an atmospheric height of 8000m, and surface areas of 2.55 x 10^8 km^2 for both the Northern and Southern Hemispheres, the mass of HCB in the air at any given time is estimated to be 112 t and 37 t for the two hemispheres, giving a total of 149 t.

7.2.2 Background Soil

Meijer *et al* (2003d) studied HCB concentrations in background surface (0-5 cm) soils collected in 1998 from 191 locations around the world. The global mean background soil HCB concentration was 0.68 ng/g dw (range 0.010-4.8 ng/g dw). The global burden of HCB in background soils was calculated to be 1020 t, of which 490 t was in the band 60-90°N, 410 t was in the band 30-60°N, and only 120 t was in the band 90°S-30°N (Meijer *et al*, 2003d). It is important to note that this calculation only includes the top 5 cm of soil. This may underestimate the mass of HCB in background soils.

7.2.3 Contaminated/Treated Soil

It has been suggested that the greatest atmospheric sources of HCB could be soils that were planted with treated seed. HCB was first introduced in 1945 as a fungicide for the seeds of onions, sorghum, and crops such as wheat, barley, oats and rye (see Section 2.2). HCB was applied as a dust that contained 10-40% active ingredient (FAO/WHO, 1970), and reported application rates range from 0.2 oz of HCB per bushel of seed (US EPA, 1991) to 1-3 ozs of 30% HCB dust per bushel (equivalent to 330-990 ppm on the seed, FAO/WHO, 1970). Importantly, the HCB was applied to the seed and was then planted into the soil, which makes it much less likely to evaporate than HCB that is sprayed on as a pesticide. However, there is limited information available about the amount of HCB that was used for this purpose.

To determine the amount of HCB currently present in directly contaminated (treated) soil, it is necessary to recreate the HCB-treated seed application history to work out the amount originally applied, and then use reported soil degradation rates to estimate the amount remaining now. The application history has been estimated assuming that HCB was used from 1945 until the date that it was banned in individual countries (see Table 3), in conjunction with the area of land that wheat (the main crop that HCB was used for treating) is grown on in each country (using FAO statistics for 2003). The assumptions were made that all seed was treated with HCB during this period, and that the area of land that wheat was grown on was the same as in 2003 in all years from 1948 until today. Countries for which we had no knowledge of the date when HCB was banned were excluded from the calculation. Figure 1 shows the history of seed treatment calculated in this fashion.

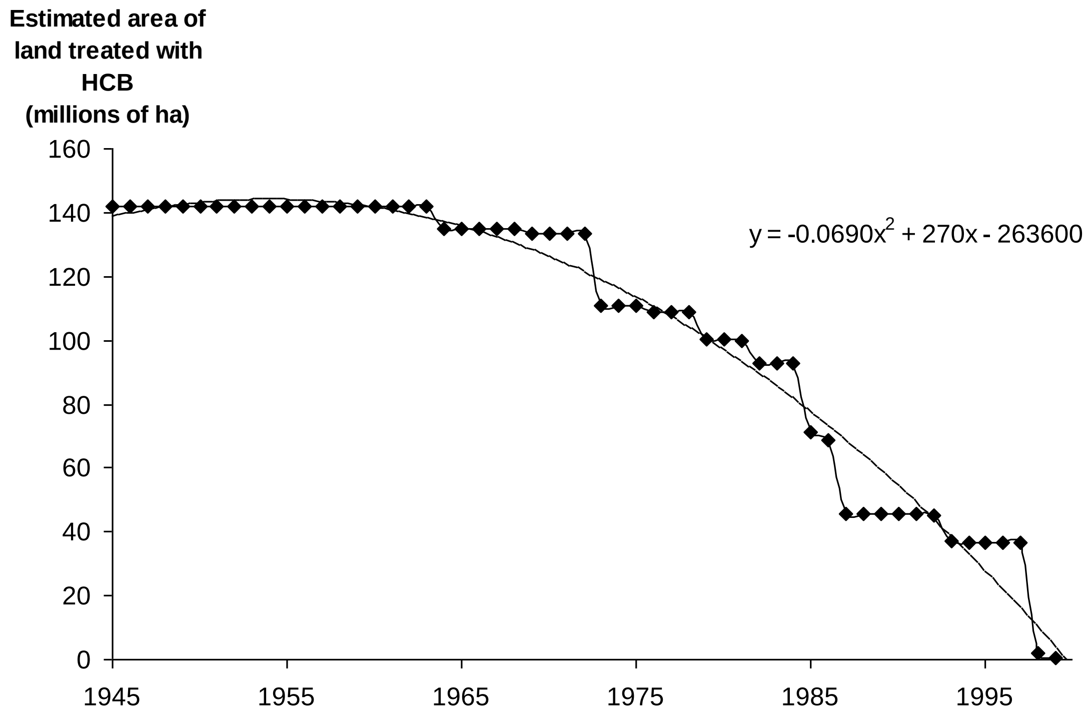


Figure 1: Estimated HCB wheat seed treatment history recreated using dates of approved use and FAO wheat cultivation land areas

This application history (scaled to a percentage of the maximum applied land area) has been used in conjunction with the global area of land with wheat grown on it (from FAO statistics for 1960-2000, with the years 1945-1959 assumed to be the same as 1960), a wheat seed application rate of two bushels per hectare, and a HCB application rate of 0.2 ounces per bushel (1 ounce = 28.38 g) to estimate the amount of HCB applied between 1945 and 2000.

The HCB present in soil has been subject to reduction each year using degradation half-lives of 2.7 years, 5.7 years (range from Beck and Hansen, 1974) and 11.7 years (Meijer *et al*, 2001) in order to find the cumulative amount of HCB in the soil. The temporal trend of estimated HCB concentrations in contaminated soil under these three degradation scenarios is shown in Figure 2.

The amount of HCB present in contaminated soil in the year 2000 would be 1170 t, 5410 t and 17000 t, respectively, for the scenarios with half-lives of 2.7 years, 5.7 years and 11.7 years. Clearly, the actual degradation rate for HCB in soil would have a large effect on the amount of HCB remaining in contaminated soil. The figures produced in this manner are probably overestimates because the more recent seed application rate for HCB is probably much less than the estimate used. However, the amount of HCB estimated to be applied to the soil annually in the mid 1970s matches the figures of 1000-2000 tonnes/year reported by Courtney (1979), and the total amount of HCB applied is very similar to the global estimate of 100000 t (ATSDR, 1996).

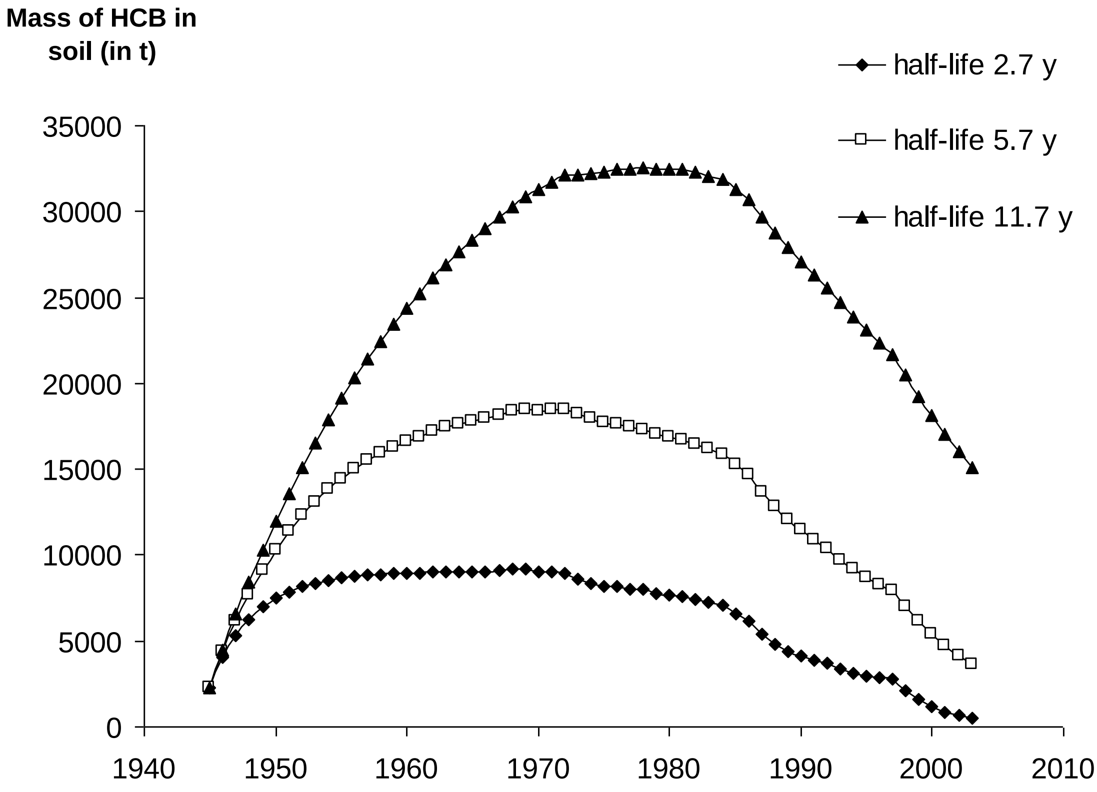


Figure 2: Estimated mass of HCB contained in treated agricultural soils after three degradation scenarios

7.2.4 Vegetation

Plants have a reasonably low bioconcentration factor (BCF) for HCB in comparison with other POPs, with reported values for BCFm (mass/mass, leaf/air BCF in ng/g dry leaf/ng/g of air) ranging from 1300-1900 (Bacci *et al*, 1990; Calamari *et al*, 1991) and values for log BCF (volume/volume, leaf/air BCF in ng/l wet leaf/ ng/l of air) ranging from 5.62-6.92 (Morosini *et al*, 1993; Horstmann and McLachlan, 1998). It has been suggested that the mass of a chemical in vegetation (in kg) can be approximated by the mass of lipid in the plant (in kg) * K_{OA} (the octanol/air partition coefficient) * concentration of the chemical in air (in kg/kg, Cousins and Mackay, 2001).

The mass of plant lipid can be estimated from the land area covered by vegetation (see Annex 26), the approximate phytomass in wet weight of foliage = area * 1 kg/m^2 (Cousins and Mackay, 2001), and plant lipid content is 1.2% on ww basis (Severinson *et al*, 1998, see Annex 26). Therefore the total mass of HCB in vegetation (in kg) is equal to the mass of plant lipid (1.559×10^{12} kg, from Annex 26) * K_{OA} (log K_{OA} at 15 °C is 7.34; Harner and Mackay, 1995) * C_{air} in kg/kg (40 pg/m^3 global average, density of air is 1.225 kg/m^3) = 1.11×10^6 kg = 1114 t.

7.2.5 Surface Seawater

Levels of HCB in seawater are generally low, ranging from 27 pg/l in the Eastern Mediterranean, to 2 pg/l in the southern Atlantic Ocean (see Annex 27). However, the volume of the oceans is very large, and therefore oceans may be important sinks for HCB. Assuming that the air and the surface waters are at equilibrium, the concentration of HCB in the surface waters (C_W) can be estimated from air concentrations (C_A) using the dimensionless Henry's law constant (K_{AW}) from the equation:

$$C_{\mathrm{W}} = \frac{C_{\mathrm{A}}}{K_{\mathrm{AW}}}$$

The sea surface water HCB concentration has been estimated using the temperature corrected K_{AW} values of Ten Hulscher *et al* (1992) and the estimates of Northern and Southern Hemisphere air concentrations made previously (see Annex 28). The average temperature of the ocean surface waters is about 17°C. The average temperature of the rest of the ocean water, which makes up the majority, is between 0-3°C (CRC Handbook, 1980).

The HCB concentrations calculated assuming equilibrium partitioning are in good agreement with the measured values (see Annex 27). The average HCB water concentrations of 12 pg/l and 2.3 pg/l for the Northern and Southern Hemispheres were therefore used to estimate the mass of HCB in the surface oceans.

Booij and van Drooge (2001) found no difference in HCB concentration between the sea-surface micro-layer (top 5mm or so) and the water column (3m depth) at a coastal location in the western Wadden Sea, and suggested that HCB in the ocean surface is well mixed. The ocean 'well mixed layer' continues to a depth of 100m on average (CRC Handbook, 1980). Using this value, total surface areas of 2.55 x 10^8 km^2 for both the Northern and Southern Hemispheres, and values of 61% and 81% for the percentage of each hemisphere covered by oceans, the mass of HCB in the surface ocean is calculated to be 187 t and 47.5 t for the Northern and Southern hemispheres, respectively, giving a total of 234 t.

7.2.6 Deep Seawater

There are very few reports about the concentration of HCB in water below the surface mixed layer. Levels of HCB in seawater 1986-87 near the Canadian Archipelago were significantly lower (approximately 9 pg/l) 175m below the surface than between 0-60m (approximately 19 pg/l), possibly due to river input and exchange of the surface water with the North Atlantic and Bering Seas (Hargrave *et al*, 1988). For this reason, we have assumed that the deep water HCB concentration is half the mixed layer concentration, or 6 pg/l in the Northern Hemisphere and 1.1 pg/l in the Southern Hemisphere.

Using an average ocean depth of 3794m (CRC Handbook, 1980) minus the surface mixed depth of 100m, surface areas of 2.55 x 10^8 km^2 for both the Northern and Southern Hemispheres, and values of 61% and 81% for the percentage of hemisphere covered by oceans, the mass of HCB in the deep ocean is calculated to be 3450 t and 839 t for the Northern and Southern Hemispheres, respectively, giving a total of 4289 t. This is a very uncertain estimate, due to the slow mixing time for the deep oceans, and is probably an overestimate.

7.2.7 Plankton

Dachs *et al* (2002) have produced a method for estimating the mass of POPs in plankton using water concentration, chlorophyll a concentration and a plankton bioaccumulation factor BCF_m. The chlorophyll a concentration is used as an approximation for phytoplankton biomass according to the equation: phytoplankton biomass (in mg/l ww) = 0.15 * (chlorophyll a conc. in µg/l)$^{1.2}$ (Kuusisto *et al*, 1998).

The average chlorophyll concentration is 8.8 mg/m^3 in the Northern Hemisphere and 8 mg/m^3 in the Southern Hemisphere (Dachs *et al*, 2002). Therefore the plankton biomass is 2.04 mg/l ww in the Northern Hemisphere and 1.82 mg/l ww in the Southern Hemisphere. The total plankton biomass (in g) can be calculated from (plankton biomass content in g/m^2) * (mixed depth in m) * (surface area of ocean in m^2) which gives 3.17 x 10^{10} t in the Northern Hemisphere and 3.76 x 10^{10} t in the Southern Hemisphere. The plankton HCB bioaccumulation factor BCF_m can be estimated with the equation:

$$\mathrm{Log\ BCF_m} = 1.085 * \mathrm{Log\ K_{OW}} - 3.770$$

(Del Vento and Dachs, 2002)

which, using a Log K_{OW} of 5.5 for HCB, gives a BCF_m of 158 m^3/kg. This compares reasonably well with a laboratory-determined BCF_m in algae of 740 m^3/kg (Isensee *et al*, 1976). The plankton HCB concentration (C_P) in ng/kg can be calculated from:

$$\mathrm{C_P} = \mathrm{BCF_M} * \mathrm{C_W}$$

(Del Vento and Dachs, 2002)

where C_W is the HCB water concentration in ng/m^3. This equation gives values for the plankton HCB concentration of 1900 ng/kg in the Northern Hemisphere and 363 ng/kg in the Southern Hemisphere. Thus, the total mass of HCB in plankton (calculated from plankton biomass * HCB concentration) is 60.1 t in the Northern Hemisphere and 13.7 t in the Southern Hemisphere, giving a total of 73.8 t globally.

7.2.8 Coastal Shelf Sediments

A complex inventory calculation for PCBs in coastal shelf sediments was recently performed by Jönsson *et al*, using a large number (2880) of sediment concentrations, and information on the surface area of the world's sediment basins (2003). This exercise was performed as part of the GLOBALSOC project that also produced the global soil inventory (Meijer *et al*, 2003d). A similar undertaking is not possible for HCB, however, because of the limited amount of HCB sediment data in the literature.

We have made a rough estimate by comparing the soil inventories for HCB and PCB-28, and then using the sediment inventory for PCB-28 to approximate for HCB. For PCB-28 the global soil inventory was 190 t (Meijer *et al*, 2003d), and the global coastal sediment inventory was 463 t, comprising local 2.0 t, regional 58 t and remote 403 t (Jönsson *et al*, 2003). Using the HCB global soil inventory of 190 t (Meijer *et al*, 2003d), and assuming a similar HCB:PCB-28 ratio for sediments, gives a global coastal sediment inventory of approximately 2500 t, comprising local 10.7 t, regional 311 t and remote 2163 t.

Using the areas of the world's sediment basins and a rearrangement of equation 1 from Jönsson *et al* (2003), the sediment HCB concentrations that this represents can be calculated. The results give an average HCB sediment concentration of 1.5 ng/g on a dry weight basis, comprising local 20.5 ng/g, regional 23.4 ng/g, and remote 1.3 ng/g. Looking at reported values in Annex 29, these concentrations are perhaps an order of magnitude too big, and therefore this inventory is likely to be a considerable overestimate.

7.2.9 Deep Ocean Sediment

The amount of HCB that is in deep ocean sediment can be estimated using the equations of Dachs *et al* (2002), which calculate the sinking rate of organic matter from the mixed surface layer into the deep ocean layer. The sinking organic matter flux, F_{OM} (in ng m^{-2}d^{-1}) is estimated from:

$F_{OM} = 1.8 * F_{OC}$ Dachs *et al* (2002)

with the flux of organic carbon, F_{OC} (in ng m^{-2}d^{-1}) estimated from chlorophyll concentration (in mg m^{-3}) according to:

$\text{Log } F_{OC} = 2.09 + 0.81 * \text{Log(Chl)}$ Dachs *et al* (2002)

Using chlorophyll concentrations of 8.8 mg m^{-3} for the Northern Hemisphere and 8 mg m^{-3} for the Southern Hemisphere (from Dachs *et al*, 2002) gives F_{OC} values of 720 ng m^{-2}d^{-1} for the Northern Hemisphere and 660 ng m^{-2}d^{-1} for SH. The F_{OM} is therefore calculated to be 1290 ng m^{-2}d^{-1} for the Northern Hemisphere and 1190 ng m^{-2}d^{-1} for the Southern Hemisphere. The flux of HCB out of the mixed surface layer into the deep ocean layer, F_{SINK} (in ng m^{-2}d^{-1}), is calculated from:

$F_{SINK} = F_{OM} * C_P$ Dachs *et al* (2002)

where CP is the particle concentration (in kg/kg). Using the plankton HCB concentration of 1900 ng kg^{-1} in the Northern Hemisphere and 363 ng kg^{-1} in the Southern Hemisphere calculated earlier, F_{SINK} is calculated to be 2.4 pg m^{-2}d^{-1} in the Northern Hemisphere and 0.43 pg m^{-2}d^{-1} in the Southern Hemisphere. The mass of HCB globally sinking out of the surface ocean layer into the deep sea, M_{SINK} (in ng/year) is calculated from:

$M_{SINK} = F_{SINK} * \text{Surface area of ocean (in m}^2\text{)} * 365$ Dachs *et al* (2002)

Therefore, using total surface areas of 2.55 x 10^8 km^2 for both Hemispheres, and values of 61% and 81% for the percentage of hemisphere covered by oceans, M_{SINK} is 138.9 kg y^{-1} in the Northern Hemisphere and 32.7 kg y^{-1} in the Southern Hemisphere. Assuming that all of this reaches the deep sea sediment layer, and that sediment accumulated HCB at the same rate between 1945 and 2000, the amount of HCB in deep sediment is 7.64 t in the Northern Hemisphere and 1.8 t in the Southern Hemisphere, or a total of 9.4 t. This may be a considerable underestimate, since the plankton concentrations will probably have been higher than the values used for most of the 55-year period.

On the other hand, assuming that all the HCB on particles leaving the mixed layer reaches the deep-sea sediment is likely to be incorrect. When vertical fluxes of particle-bound HCB in the Mediterranean Sea were analysed in 1981 by Fowler *et al* (1986), the vertical flux of HCB to a depth of 100m averaged 59 pg $cm^{-2}year^{-1}$, whereas the flux to sediments at 250m averaged 7.3 pg $cm^{-2}yr^{-1}$ (Fowler *et al*, 1986). This indicates that as much as 87% of the particle-associated HCB is consumed, re-dissolved or degraded before burial into sediments.

7.2.10 Dominant Environmental Sinks for HCB

The HCB inventory estimates for the various environmental compartments are summarised in Annex 30, which includes three values for contaminated soil based on the three different estimated half-lives used for the HCB. The total mass of HCB currently in the environment ranges from 10000-26000 t, depending on the estimate for contaminated soil. Interestingly, vegetation contains roughly the same amount of HCB as the top 5cm of background soil, which is in line with the similarities between Maximum Reservoir Capacity (MRC) of soil and vegetation noted by Dalla Valle *et al* (2004), although their calculation only included the top few millimetres of soil.

7.2.11 Multimedia model predictions of HCB distribution

The environmental fate of HCB was investigated by MacLeod and Mackay (1999) using a simple equilibrium criterion model (EQC) with a land area of 90% and a water area of 10%. When HCB is emitted to air, as most current emissions are, at steady state they estimated 27% is in air, 1% in water, 62% in soil and 10% in sediment. When HCB is emitted to soil, which historical agricultural emissions mostly were, at steady state 0.02% is in air, 0.006% in water, 99.9% in soil and 0.09% in sediment. Finally, when HCB is emitted to water, at steady state 1% is in air, 6% in water, 2% in soil and 91% in sediment. The estimates made above will reflect an unknown proportion of emissions to air, water and soil, over an area with much greater water coverage so these numbers are difficult to compare with each other. The EMEP-MSCE hemispheric POP model (with roughly 60% water and 40% land coverage), ran using the 1990 European emissions of Pacyna *et al*. (1999), predicts that after a year 25% of emitted HCB is in air, 44% in seawater, 20% in soil, 4% in vegetation and 7% in forest litter (Malenichev *et al*., 2002).

When the Pacyna historical emissions are used in a European model (with roughly 47% water and 53% land coverage) and ran to a steady state, 89 % of total HCB content in the environment ends up in seawater, 8 % in soil and 2.3 % in air (Shatalov *et al*., 2001). These predictions should be more realistic since they contain long-term emissions and larger water coverage. In comparison, the results from our calculation, taking the middle contaminated soil estimate (half-life of 5.7 years), predict that soil contains 44%, seawater 31%, sediment 17% and air 1% of the HCB currently in the environment. The results of this last model are clearly very different from the estimates obtained in the mass balance calculation, however the comparison between a European model and a global inventory is meaningless. Unfortunately, because of the way the mass balances in treated soil, vegetation and sediment have been calculated, it is not possible to alter the scale of our estimates down from a global to a European level, so a more appropriate comparison cannot be made.

7.3 Dominant loss processes for HCB in the environment

A summary of the range of different environmental degradation rates for different media from Section 3 is shown in Annex 31. It is difficult to know which values to assign to a medium as an ‘average’ degradation rate out of the reported rates.

For air, the measured hydroxyl-reaction degradation rate of Brubaker and Hites (1998) has been selected over estimated values for this property (i.e. those reported in Howard *et al*., 1991; Kwok and Atkinson, 1995). For soils, a wide range of half-lives has been reported, dependent on whether conditions are aerobic or anaerobic, and so a value at the high end of the aerobic range and low end of the anaerobic range has been chosen. For the moment the assumption is made that all of this loss is actual transformation to another chemical. For sediments there is a very big difference between values reported, with half-lives ranging from days to years, and so an intermediate value of 1.7 years has been selected. This is considerably lower than the 6.3 year half-life estimated by Mackay *et a*l (1992).

For surface water, a half-life of 4.2 years was chosen, which is central in the range reported by Howard *et al* (1991). It is possible that this half-life will lead to an overestimation of the degradation of HCB in water, since degradation is not considered to be important for the removal of HCB from water (Mill and Haag, 1986). For deep ocean water, an arbitrary value of twice the surface water half-life has been used, but this would be an over-estimate if photolysis were the only breakdown process in the water column. No degradation rates are available for HCB in vegetation. All loss processes will obviously be highly variable across the globe, depending on temperature and other environmental features.

As mentioned above, it is important to remember that the reported half-lives may not necessarily reflect actual degradation rates. This is because other processes such as volatilisation and desorption have not been excluded or, in the case of sediments, experiments may have been carried out with artificially high HCB sediment concentrations or using pre-adapted HCB degrading cultures.

These degradation rates have been combined with the estimated HCB inventory from the global mass balance section (using the contaminated soil value estimated using a half-life of 5.7 years) to estimate the amount of HCB that is being removed from the environment currently (see Annex 31). In the Annex, the ‘average’ mass of HCB degraded is based on the ‘average’ loss half-life, and the range is calculated using the same inventory but the full range of reported degradation rates.

The greatest losses occur from sediment and soil, with losses in water of less importance and losses in air insignificant. The total amount of HCB that will be lost in the year that the HCB inventory represents is estimated to be 2260 t (range 760-5300 t). This is considerably larger than Bailey’s global emission estimate of 23 tonnes/year (Bailey, 2001) and suggests that levels in the environment will continue to fall. However, we should point out that this calculation contains a number of assumptions and uncertainties. For example, the amount estimated in this document to be lost from soil by ‘degradation’ will however include a significant proportion that is actually volatilised, since these two phenomena are not distinguished in experiments that measure loss rates of HCB from soil. Whereas biodegradation completely removes HCB from the environment, volatilisation leads to continued existence of HCB in a different environmental compartment. This volatilisation from soil may have an important effect on levels and trends in these other compartments.

Using the environmental fate predictions for the environmental distribution of HCB a year after emission calculated by Malenichev (2002), it is possible to estimate the amount of HCB added to the different environmental compartments annually. Using this method, of the 23 t emitted, 25% (6 t) will remain in air, 20% (5 t) will deposit to soil and 44% (10 t) will deposit to water. Thus the amount of the annual emission that is added to air is less than the 60 t estimated to be degraded, and therefore re-emission of HCB from soil is needed to maintain contemporary air concentrations. The 10 t added to water is also less than the 39 t estimated to be degraded in surface water, and if this degradation rate is to be believed, levels here will also continue to decline. Importantly, the 10 t that is estimated to be added to soil is greatly exceeded by the amount ‘degraded’ (~800 t), suggesting that a significant decrease in soil concentrations should be observed.

7.4 Temporal Trends In Observed Environmental HCB Concentrations

Levels of HCB in the environment have declined significantly since emissions of HCB were reduced. The following section reviews the rates of decline that have been found in various media from many locations over the past 30 years.

7.4.1 Air

Peak concentrations at Spitsbergen in the Arctic were measured in the 1980s, when sampling first began (Pacyna and Oehme, 1988), and reported levels in 2001 are 75% lower (EMEP, 2003). In the Great Lakes, peak concentrations occurred in the late 1970s and early 1980s, with a significant decline observed by the Integrated Atmospheric

Deposition (IADN) Program between 1990 and 1995 (Cortes *et al*, 1998). The atmospheric half-lives for HCB calculated from Great Lakes air concentration data from the early 1990s ranged from 3.3 years at Sleeping Bear Dunes to 12 years at Eagle Harbour (Cortes *et al*, 1998).

Cortes *et al* (1998) estimated disappearance times for HCB in the Great Lakes using these atmospheric half-lives and the assumption of first order decay. Cortes and Hites (2000) improved the method of calculating these half-lives, and produced revised (longer) atmospheric half-life values ranging from 7.7 years at Sleeping Bear Dunes to 14 years at Eagle Harbour. More recently, Buehler *et al* (2004) reported on the IADN data up to the year 2001. They found that over this longer time period half-lives, calculated using a multiple linear regression model, were much longer ranging from 15 years at Sleeping Bear Dunes to 29 years at Eagle Harbour (Buehler *et al*., 2004). The Great Lakes region of North America was acting as a source of HCB to the atmosphere prior to 1998, but since then the net flux is believed to be from the atmosphere to this area (IADN, 1998). This trend suggests that following the reduction in HCB usage, air-water equilibrium is being approached for HCB in this region.

7.4.2 Rain

The Great Lakes rainfall concentration data show a significant trend of decreasing levels of HCB. HCB concentrations decreased in the Great Lakes region by 97% between 1978 and 1983 (Eisenreich *et al*, 1981; Strachan, 1985). Between 1988 and 1994, individual Lakes had differing trends, with rainfall levels in Lakes Superior and Erie decreasing by 31% and 63% respectively, whereas rainfall levels in Lake Ontario approximately doubled (Chan *et al*, 1994; Hoff *et al*, 1996).

7.4.3 Snow

Temporal trends for HCB have been studied using dated ice cores collected from the Lys glacier in the Swiss/Italian Alps in 1996 (Villa *et al*, 2003a). Concentrations were below the detection limit between the 1960s to late 1970s, after which concentrations increased, peaking in the mid-1980s, and then decreasing until the early 1990s. These cores probably reflected European rather than global trends. Donald *et al* (1999) looked at levels of HCB in a Greenland glacier as a means for determining historical snow deposition.

Concentrations of HCB in precipitation increased from 1959 to 1995 at a relatively constant rate, about 0.34 pg/l/year. The maximum concentration of 0.05 ng/l was recorded in snow from 1994. The mean concentration for the 1990s was 0.011 ng/l, suggesting that annual deposition was about 3.3 ng/m^2, a rate about 100 times lower than accumulation rates in the 1980s in peat bogs of eastern North America where the maxima occurred in 1970 (Rapaport and Eisenreich, 1988).

It is important to note that there are uncertainties over the reliability of snow/ice cores as a record of POPs deposition. However, trends for HCB in fresh snow from the Arctic also show increasing concentrations in recent times. Levels in the Agassiz Icecap, Ellesmere Island, Northern Canada increased by a factor of 10 between 1985-86 and 1989-90 (Barrie *et al*, 1997), and levels increased in other Arctic locations by factors of 3-8 between 1986-87 and 1994 (Gregor and Gummer, 1989; Barrie *et al*, 1997).

7.4.4 Water

Temporal trends for HCB in rivers show declines in a number of locations. However, levels of HCB in historically contaminated rivers remain high, despite the cessation of major emissions, which indicates that remobilisation of residual HCB from contaminated sediments is occurring. Concentrations of HCB in the Rhine River were historically very high, but fell by 90% between 1970 and 1984 from 140 ng/l to <10 ng/l (Greve, 1986), and by 99% between 1972 and 1990 from 100 ng/l to 1 ng/l (Beurskens *et al*, 1993). Levels of HCB in the Niagara River fell by over 50% between 1992 and 1998 (Williams *et al*, 2000) and levels of HCB in the nearby Great Lakes fell by 25-83% between 1980 and 1992 (Stevens and Nielson, 1989; Oliver and Nicol, 1982; Hoff *et al*, 1996). There are no data showing temporal trends for HCB in seawater.

7.4.5 Sediment

Fox *et al* (2001) looked at dated sediment cores from the Mersey estuary in the UK. HCB showed three peaks: a small peak beginning around 1900 corresponding with the beginning of chlorination of benzene on Merseyside, a sharp peak beginning around 1940 corresponding with the peak of HCB pesticide production on Merseyside, and a lower broad peak in the 1960s probably as a result of HCB by-product manufacture.

Levels of HCB in sediment have decreased since 1970 by more than 95% from the last peak to the present day (Fox *et al*, 2001). Temporal trends for HCB have also been studied in dated sediment cores from the central Venice lagoon (Marcomini *et al*, 1999). In one core, levels were declining with time, but in two others, cores showed a continued increase over time.

In the Great Lakes, levels of HCB in Lake Erie sediments decreased significantly over the period 1971-1997 from an average of 5.7 ng/g dry weight to 1.6 ng/g dry weight (Marvin *et al*, 2004b). Peak levels of HCB in sediment cores from Canadian mid latitude lakes consistently occurred in layers from the 1970s in all lakes studied (Muir *et al*, 1995a).

In contrast, peak levels in sediments from Lake Ontario date from the 1960s (Durham and Oliver, 1983; Eisenreich *et al*, 1989; Oliver *et al*, 1989), which corresponds with the period of peak chlorobenzene production in the US. Sedimentary core studies in the St. Lawrence River have shown a decline in HCB contamination (by a factor of 5-10) since its production was discontinued in the 1970s (Carignan *et al*, 1994).

In sediment cores from the Gulf of Alaska, HCB levels rose in the 1960s, reaching a peak in the 1970s, before beginning a gradual decline in the 1980s (Iwata *et al*, 1994a). HCB first appears in sediment cores from Greenland lakes in layers from the 1960s (Malmquist *et al*, 2003). Peak levels varied from the 1970s to 2000s in the different lakes, and are therefore later than peak levels reported in North America.

This is evidence for delayed deposition of HCB away from source areas. In Lake Manzala in Egypt, levels of HCB in sediment cores increased rapidly from the early 1960s to a maximum in the 1980s, whereas in Nile River sediments HCB concentrations continue to increase steadily to the present day (Yamashita *et al*, 1997).

7.4.6 Soil

Information on temporal trends for HCB in soils is scarce. From studies of dated peat cores from eastern North America, HCB levels showed a slow increase from 1915 to 1945, followed by a more pronounced increase to a peak in 1967. The concentrations then dropped off sharply by 50% by the mid-1970s, with levels then remaining fairly stable for the next decade (Rappaport and Eisenreich, 1988). These changes mirrored US chlorobenzene production levels.

In archived UK background soils, the peak concentration was measured over a long period from the late 1960s and early 1970s to the early 1980s (Meijer *et al*, 2001). Levels of HCB in these soils decreased by between 65% and 75% from the late 1980s to the early 1990s.

7.4.7 Vegetation

There are no reported incidences of vegetation being used to determine temporal trends for HCB in the environment.

7.4.8 Biota

7.4.8.i Shellfish

High levels of HCB were measured by Connor *et al* (2001) in mussels in the Mersey Estuary in the UK in 1994. This area is close to a number of industrial sources of HCB, in particular the UK chlor-alkali industry. However, levels fell by 70% between 1994 and 1998, probably as a result of legislation to reduce the discharges of HCB into the environment. Levels of HCB in Boston Harbour also decreased between 1992 and 2001 (Lefkovitz *et al*, 2002). Levels of HCB in mussels from the coast of Spain showed an order of magnitude decrease over the period 1980-92 (Sole *et al*, 1994). Similarly, Villeneuve *et al* (1999) reported that levels of HCB in mussels from the North-Western Mediterranean had fallen by more than 90% between 1973 and 1989.

7.4.8.ii Fish

Monitoring of fish in Lake Storvindeln and Abiskojaure in Northern Sweden has shown that declines in levels of HCB in pike and roach have occurred in these Arctic locations (Bignert *et al*, 1998; 2004). Significant decreases have also been observed in cod and herring from the West Coast of Sweden (Bignert *et al*, 2004). However, HCB levels in three species of Antarctic fish did not change significantly between 1987 and 1996, although slight declines were noted (Weber and Goerke, 2003).

Long term monitoring of HCB in fish has been carried out in the Baltic since the 1970s (Bignert *et al*, 1998; Falandysz, 1984; Falandysz *et al*, 1997). Kannan *et al* (1992a) looked at levels of HCB in cod liver oil from the southern part of the Baltic over the period 1971-1989. A slow, consistently downward trend was observed during the study period. The maximum concentration of 460 ng/g was recorded in 1973 and 1974, and this value was approximately three times higher than the value in 1989.

The slow rate of reduction indicates the high environmental stability of HCB and suggests ongoing emissions. Decreases were also observed throughout the early 1990s, but concentrations levelled off in the late 1990s (Bignert, 2002a). Some areas appear to have reached a steady state of HCB contamination, indicating a continuous input of this compound (Falandysz, 2000, Falandysz *et al,* 1994; Falandysz *et al,* 2000b; Sinkkonen and Paasivirta, 2000).

In contrast, de Boer (1989) found HCB concentrations to be stable in the North Sea over the period 1977-1987, and Haarich *et al* (2002) reported that between 1987 and 1999, levels actually increased in some areas of the North Sea, whereas other areas showed no change or recorded decreases. Increases tended to occur near estuaries of contaminated rivers such as the River Elbe (Marth *et al*, 2000), which suggests that historical contamination is being re-mobilised from river sediments and moving downstream into the North Sea.

7.4.8.iii Birds

Levels of HCB in herring gull eggs in the Great Lakes fell dramatically between 1974 and 2001, by between 90% and 98% (Weseloh *et al*, 2002). Decreases of 71%, 89% and 98%, respectively, were also observed in the St Lawrence, Niagara and Detroit Rivers (Weseloh *et al*, 2002). Levels of HCB in the Canadian Arctic in birds eggs also fell between 1987 and 1998, by between 40% and 65%, depending on bird species (Braune *et al*, 2001). Levels of HCB in common murre eggs from Alaska, however, remained stable between 1973-76 and 1999-2000, in contrast to other OCs that significantly decreased (Pol *et al*, 2004).

Results from the German Environmental Specimen Bank (ESB), which included 10 years of herring gull eggs collected in the North and Baltic Seas (Marth *et al*, 2000) showed that levels decreased at Heuwise in the Baltic Sea and Mellum in the North Sea, but increased at Trischem, another site in the North Sea, near the River Elbe. In another study, temporal trends of HCB levels in common tern and oystercatcher eggs over the period 1991-2000 showed an increase in the River Elbe and one other site influence by the River Rhine, whereas other sites in the Wadden Sea either showed no trend or a slight decrease (Becker *et al*, 2001). Discharges of HCB from the River Elbe into the Wadden Sea have also shown a recent increase (Becker *et al*, 2001).

Temporal trends over the longer period 1981-2000 show significant decreases at all Wadden Sea sites, with the predominant changes occurring in the late 1980s and early 1990s (Becker *et al*, 2001). Similarly, levels of HCB in guillemot eggs in the Baltic Sea fell at a rate of 9% per year between 1979 and 2000 (Bignert, 2002a). In the Barents Sea, levels of HCB in herring gull eggs fell by 60% between 1979 and 1983, but remained relatively constant between 1983 and 1993 (Barrett *et al*, 1996).

Significant declines were found, however, over the period 1983-93 in levels of HCB in six of the seabird species breeding in northern Norway (Savinova *et al*, 1995). HCB levels in black guillemots in Iceland decreased slowly between 1976 and 1996, with a $t_{1/2}$ of 16.1 years (Ólafsdóttir *et al*, 2005). It was hypothesised that in this area far away from sources, the observed levels reflected proximity to an equilibrium between the long-range transport and the natural breakdown rate of the chemicals.

7.4.8.iv Marine Mammals

Temporal trends for HCB contamination in marine mammals have been reported. Hobbs *et al* (2001) found that levels of HCB in blubber of fin whales fell by 60% between 1971 and 1991 of the coast of Eastern Canada. Levels of HCB in striped dolphins from the North-Western Pacific, near Japan, also decreased between 1978 and 1986 (Lognathan *et al*, 1990). Similarly, levels of HCB in harbour seals from Puget Sound, Washington, US, fell by 40% between 1984 and 1997 (Calambokidis *et al*, 1999). Although levels of HCB in ringed seals from Holman in the western Canadian Arctic remained relatively constant over the period 1972-81, HCB burdens fell by 40% to 50% between 1981 and 1991 (Addison and Smith, 1998).

These temporal trends are consistent with changes in the production and use pattern of HCB. However, there are many studies that show levels of HCB remaining unchanged in marine mammals. Levels of HCB in beluga whales from Hudson Bay remained unchanged between 1966 and 1986 (Muir *et al*, 1990a), and showed no trend in female belugas from the St. Lawrence Gulf from 1986 to 1994 (Muir *et al*, 1996b). Levels of HCB in harbour and grey seals in St. Lawrence Gulf also remained constant between 1989 and 1996 (Bernt *et al*, 1999), and levels in grey seal near Sable Island, North-Western Atlantic, did not change between 1984-94 (Addison and Stobo, 2001). Similarly, Muir *et al* (2000) found no significant change in HCB concentration in Greenland Walrus between 1978 and 1988, and levels of HCB were stable in minke whale from Antarctica between 1984-93 (Aono *et al*, 1997).

7.4.8.v Reptiles

Bishop *et al* (1996) used eggs from common snapping turtles to illustrate that levels of HCB in the Great Lakes fell by 50% between 1981 and 1991.

7.4.8.vi Terrestrial Animals and Foodstuffs

In most countries, the HCB concentration in food declined from the mid-1970s to the mid-1990s (IPCS, 1997). In Germany, levels of HCB in cows' milk peaked in 1974, with levels declining rapidly by 85% by 1978 (Nijhuis and Heeschen, 1986). Levels of HCB in adipose tissue in cattle and pigs from Sweden fell by 38% and 64% between 1991 and 1997 (Glynn *et al*, 2000a). Levels of HCB in polar bears decreased between the 1980s and 1990s, but levels remained constant from 1991-94 (Bernhoft *et al*, 1997).

7.4.9 Humans

Monitoring studies have been used to investigate temporal trends in HCB concentrations in human tissues. In Germany, levels of HCB increased in milk from 1975-80 by 250%, but then decreased by 30% by 1984 (Nijhuis and Heeschen, 1986). Levels decreased further to 40% of 1984 values by 1995 (Ott *et al*, 1999). A big decline in HCB levels in adipose tissue of German people was also observed, with concentrations decreasing by 90% between 1983 and 1998 (Lackmann, 2002).

Sweden has witnessed a clear decline in the levels of HCB detected in breast milk since use of HCB as a fungicide was banned in 1980, with levels in 1997 down by 95% from 1972 (Lundén and Norén, 1998; Norén and Meironyte, 2000). Norway experienced a similar decrease, with levels of HCB in breast milk dropping by 65% between the mid-1970s and early 1990s (Johansen *et al*, 1994). Similarly, the burden of HCB in adipose tissues of the population of the Netherlands decreased by 60% between 1975 and 1986 (Greve and van Zoonan, 1990). In the Czech Republic, HCB levels in human milk decreased by 65% between 1975 and 1983 (Uhnák *et al*, 1986), and by 50% from 1994 to 2000 (Čajka and Hajšlová, 2003).

In Japan, levels of HCB in adipose tissue fell by 50% between 1976 and 1981 (Mori *et al*, 1983). HCB levels have been monitored from 1980-1998 in human breast milk obtained from Osaka Prefecture, Japan (Konishi *et al*, 2001). Concentrations showed a slight increase at the beginning of the 1980s and then decreased by 75% by 1998.

In the US, levels of HCB in human adipose increased between 1974 and 1976, but then declined steadily between 1976 and 1983 (Kutz *et al*, 1991). In Canada, levels of HCB in milk decreased by 50-75% between 1975 and 1992 (Craan and Haines, 1998). Levels of

HCB in umbilical cord blood collected from new-borns in the St. Lawrence River Region of Canada fell at a rate of 12% a year between 1993 and 2000 (Dallaire *et al*, 2002).

Results of most studies on the levels of HCB in human tissues over time clearly indicate that exposure of the general population to HCB declined from the 1970s to the mid-1990s in many locations. However, an investigation in Changsha, China, showed that HCB concentration of human breast milk had increased by 60% over 10 years (Li, 2002).

7.4.10 Summary

The consistent downward trend in the environment over the past 20 years suggests historic deposits of HCB are being depleted and HCB is being removed from the environment. Temporal trends of HCB in the environment vary, dependent on time period measured, media studied and study location, with peaks occurring in the 1960s (Durham and Oliver, 1983; Rappaport and Eisenreich, 1988; Eisenreich *et al*, 1989; Oliver *et al*, 1989; Fox *et al*; 2001), 1970s (Nijhuis and Heeschen, 1986; Kutz *et al*, 1991; Kannan *et al*, 1992; Muir *et al*, 1995a; Barrett *et al*, 1996) and 1980s (Yamashita *et al*, 1997; Konishi *et al*, 2001; Villa *et al*, 2003b). The only places that do not have a consistent downward trend are in China, where HCB is still manufactured (Li, 2002), in certain polar locations (Aono *et al*, 1997; Bernt *et al*, 1999; Malmquist *et al*, 2003), and in estuaries of historical souce regions (Marth *et al*, 2000, Becker *et al*, 2001), which are presumably affected by catchment run-off and remobilisation of contaminated sediments. Marine mammals also often show no consistent upward or downward trends.

Wania and Mackay (1995) predicted that the degradation half-life of HCB would vary in different regions, with half-lives of 0.63 years (230 days) in tropical/subtropical regions, 1.94 years (708 days) in temperate/boreal regions, and 6.28 years (2292 days) in polar regions. There is insufficient data from any single medium to test whether the slope of the decline is steeper in lower latitudes compared with higher latitudes (Wania and Mackay, 1996).

However, the fact that sediment and ice cores and precipitation in the Arctic show an increase in HCB concentration, as opposed to the general decreasing trend suggests that the poles are still acting as sinks in the global movement of HCB (Donald *et al*, 1999; Malmquist *et al*, 2003; Barrie *et al*, 1997). Air concentrations have declined, but again there is insufficient evidence to suggest that this is strongly latitudinally dependent, probably because atmospheric mixing and advection ensure reasonable homogeneity in background air concentrations.

Loss half-lives have been calculated from a number of the temporal studies, and these are shown in Annex 32. The average half-life from all the studies in this Annex is nine years. In many of these studies, the half-lives are longer than ‘average’ degradation half-lives. This is probably because HCB was still being emitted during these periods and therefore at this time the amount being degraded will be countered by additional emissions to the environment.

As emissions have significantly decreased since the 1970s, loss half-lives should be shorter for more recent studies because the proportion of the loss rate taken up with degrading newly emitted chemicals will be less. This phenomenon is apparent from a number of the studies reported in Annex 32, which do have shorter half-lives for periods which begin more recently. Although HCB emissions have decreased throughout the world, the rate of decrease has slowed markedly since 1990. The average half-life of the seven studies whose period only covers the 1990s is five years.

8 Re-emission of HCB from Soil

As discussed in Sections 2.8 and 7.3, some of the HCB measured in the atmosphere is thought to come from volatilisation of ‘old’ HCB in the soil from past contamination (Bailey, 2001). The amount estimated as being lost from soil by ‘degradation’ in Section 7.2.3. may include a significant proportion that is volatilised, since these two phenomena are not easily distinguished in experiments that measure loss of HCB from soil.

It is possible to estimate reconstruct the trends of HCB re-emission from treated soil, using the trends of HCB concentration in treated soil shown in Figure 2, and assuming that a proportion of the amount previously assumed to be ‘degraded’ is actually volatilised. We have reconstructed these trends using different scenarios. Firstly we assigned an arbitrary percentage (50%) that we assumed to be volatilised, and looked at the different ‘degradation’, or in this case ‘loss’, rates (2.7 yrs, 4.7 years and 11.7 years). Secondly we selected a single ‘loss’ rate (half-life 5.7 years) and 3 different values for the percentage that is re-volatilised (30%, 50% and 70%). Figures 9 and 10 show the results of these calculations.

The timing of the peak of soil HCB emission varies according to half-life from 1970 to 1980, with the peak for the longest half-life scenario latest (see Figure 3). At its peak, the amount of HCB emitted from soil was in the hundreds to thousands of tonnes per year, which would have made it a significant source of HCB to the environment. However, the amount of HCB being emitted from contemporary soil is much lower now than at the peak period irrespective of what fraction is assumed to be volatilised (see Figure 4), so the size of this secondary source of HCB to the environment has possibly declined as much as primary emissions. Indeed, only a small amount of re-emission of HCB from soil to air is required to maintain contemporary air concentrations under the current primary emission scenario.

Figure 3. Estimated historical emissions of HCB from treated soil using three degradation rates that incorporate a fraction (50%) that is volatilised rather than degraded.

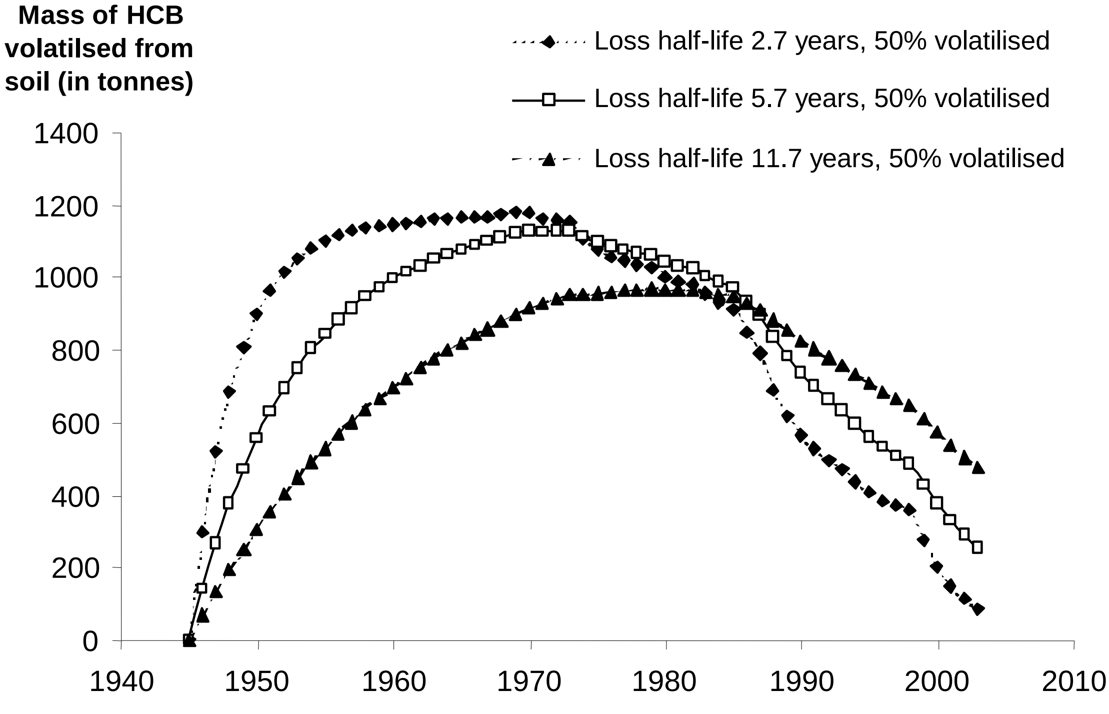
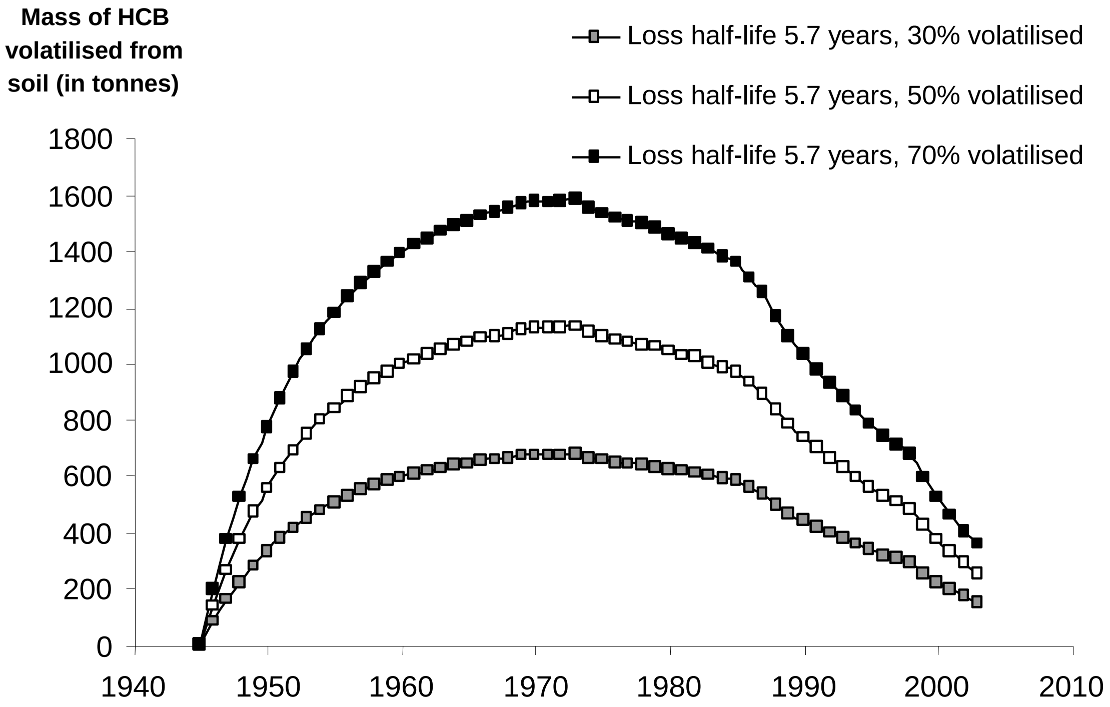


Figure 4. Estimated history of emission of HCB from treated soil using a ‘degradation’ half-life of 5.7 years, with a varying fraction (30%, 50% and 70%) that is volatilised rather than degraded.



9 Conclusions

There is much evidence that HCB is subject to global re-distribution in the environment, and to cold condensation (Muir *et al*, 1995a; Blais *et al*, 2003). It has been suggested that because of the general HCB decrease, and similar HCB concentrations in North Sea and Antarctic fish (Weber and Goerke, 1996), then the global distribution of HCB could be close to equilibrium (Aono *et al*, 1997).

However, the increasing levels of HCB in some Arctic media, and the wide variation of HCB concentrations of background soils and sediments, suggest that this is not the case. The combination of a mass balance calculation, existing multimedia fate models and reported degradation rates, suggests that the greatest losses of HCB in the environment occur from sediment and soil, with losses in water of less importance and losses in air insignificant. Levels of HCB in air and water will probably continue to decrease at a slow rate, whereas levels in soil will continue to decrease significantly, either by degradation or re-volatilisation.

10 References

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