

**SOURCE ACTIVITY TITLE: SOURCES OF HEXACHLOROBENZENE EMISSIONS**

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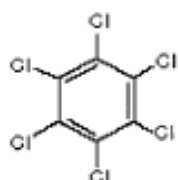
**Introduction**

This model chapter is aimed at summarizing information relevant to hexachlorobenzene emission for the purposes of its successful inventory.

**1.GENERAL****1.1. Definitions**

Hexachlorobenzene is a chlorinated hydrocarbon industrial chemical or by-product of the chemical synthesis. The general formula is  $C_6Cl_6$ . CAS number – 118-74-1.

Industrial production of HCB is based on the reaction of benzene chlorination at the temperature of about  $200^{\circ}C$  at the atmospheric pressure and the presence of ferrous oxide as a catalyst. Catalytic chlorination of isomers of hexachlorocyclohexane (HCH) is an alternative process. In this case HCB release is 80-95 % of the theoretical one without additional application of chlorine.

**1.2. Properties**

Hexachlorobenzene is a white crystalline substance with molecular mass 284.7840, with specific off-flavour, melting point  $231^{\circ}C$ , boiling point –  $326^{\circ}C$ . It is very soluble in many organic solvents and practically non-soluble in water.

Hexachlorobenzene is a very persistent environmental chemical due to its chemical stability and resistance to biodegradation. HCB life-time in the atmosphere is defined by the rate of interaction with hydroxyl-radical and it is equal 2.7–6 years depending on the geographical latitude. Such long life-time in atmosphere allows transboundary transmission of HCB. In water HCB binds to sediments and suspended matter. In soil HCB binds strongly and generally does not leach to water (life-time is about 6 years). Transport to ground water is slow (life-time is about 5.3–11.4 years), but varies with the organic make-up of the soil, as HCB tends to bind more strongly to soils with high organic content. HCB accumulates in both terrestrial and aquatic food chains. It accumulates in fat tissues of fish and marine animals, it can also accumulate in lichens, wheat, grass and vegetables (PBT National Action Plan..., 2000).

According to the classification of the International Agency on Cancer Research (IACR) it refers to carcinogen group 2B (2b).

**1.3. Hexachlorobenzene use**

HCB was widely used (from the 1940s to the late 1970s) as a fungicide on grain seeds such as wheat. In the USA the last registered use as a pesticide was voluntarily canceled in 1984. In the USSR HCB was used till 1990 as Hexathiuram and Hammahexane in agriculture, in forestry and for municipal purposes; in the period of 1990–1996 in Russia only remains of these products were

allowed for application. After 1996 in Russia no product containing HCB was allowed to use as pesticide.

HCB was also used to make fireworks, ammunition, and synthetic rubber (Toxicological Profile..., 2000).

## **2. SOURCES OF HCB EMISSION**

The main source of HCB emission is agriculture if HCB is used in this sector.

Hexachlorobenzene is currently emitted as by-product or impurity in the manufacture of several chlorinated solvents (tetrachloroethylene, trichloroethylene, carbon tetrachloride); other chlorinated compounds (vinyl chloride); and several pesticides, including pentachloronitrobenzene (PCNB), tetrachloroisophthalonitrile (chlorothalonil), 4-amino-3,5,6-trichloropicolinic acid (picloram), pentachlorophenol (PCP), and dimethyltetrachloroterephthalate (DCPA or Dacthal); and was also produced as by-product for production of atrazine, propazine, simazine, and mirex. The emissions of HCB in Czechia and Slovakia were 3–6 tones (1992), 80 % of HCB was released owing to burning of tetrachloroethylene, trichloroethylene and polyvinyl chloride (Zero Toxics..., 2001).

On the whole, processes resulting in dioxins/furans formation lead also to HCB emissions. Thus, pulp and paper production (with chlorine use for bleaching) results in HCB release; HCB is also contained in wastes of chloralkali production, wood impregnation processes (Leger, 1992). HCB is emitted into the air during wastes incineration.

According to (Supplementary Guide..., 2000), the list of HCB sources to be taken into account for emission inventory includes: wastes and sewage sludge incineration, base metals smelting (this refers to copper, lead, nickel and zinc), smelting of secondary lead and aluminum, sintering process, electric arc furnaces in steel manufacturing, production of magnesium, cement and chlorinated organic solvents or chlorinated monomers as well as combustion of fossil fuel.

The electrolytic magnesium production is a significant source of HCB emissions. Evidently that the production of other commercial metals based on electrolysis of molten chloride (such as titanium and nickel) serves as a source of HCB (Zero Toxics..., 2001).

## **3. CONTRIBUTION TO EMISSION**

According to (Berdowski et al., 1997), in 1990 in Europe the share of agriculture (pesticide use) made up to about 72 % total emissions of HCB (in some countries this figure made up to 100 %). Production of chlorinated products and electric steel making provided about 10 % HCB emissions.

According to (Pacyna et al, 1999), in 1993 – 1995 in Europe 80 % of total HCB was released from the agricultural sector; among other sources the solvents and other products use predominate (13 %). In addition, about 7 % was attributed to emissions from production processes and less than 1 % from waste treatment and disposal.

Data of HCB national inventory in the USA (PBT National Action Plan..., 2000) show, that emissions in 1996 from the production of industrial inorganic chemicals (silicone products) represent the largest source (48 % of total HCB emissions). The application of surface coatings to both the inside and outside of metal cans is reported to account for 21 % of total HCB emissions. About 7 % total HCB emissions accompanied processes of chlorine production, 11 % – pesticide production, 3 % – various processes of organic chemistry and HCl production.

According to results of HCB inventory in Belarus for 2003 main contributors to total HCB emission are stationary fuel combustion (60 %) and metal smelting (39 %).

#### 4. ABATEMENT

Taking into consideration heterogeneity of HCB pathways (direct application, in-stack emission etc.) prevention of HCB emissions into the environment can be made using strategies. They include:

- a) safe application of HCB (especially as pesticide) and HCB-polluted products;
- b) safe disposal of HCB wastes (including old pesticides storage and disposal);
- c) complete destruction of HCB wastes (vast list of technological measures aimed at high level of destruction of HCB);
- d) HCB emission control from thermal processes. Due to comparatively low levels in emissions from the most of processes special measures for HCB control are usually not proposed. HCB emissions can be controlled generally like other chlorinated organic compounds in emissions, for instance dioxins/furans and PCBs: regulation of time of combustion, combustion temperature, temperature in cleaning devices, sorbents application for waste gases cleaning etc.

#### 5. METHODOLOGY

A simpler methodology of emission estimation is based on the calculation using aggregated emission factors and production statistics by the formula:

Hexachlorobenzene emission = emission factor x output production (fuel or waste combusted, pesticides applied).

A more reliable methodology is based on emission factors for specific processes, taking into account data on the type and efficiency of control equipment, and the content of HCB in materials, fuels and wastes.

However these traditional methodologies can be applied mainly for in-stack HCB emission and pesticides application. For some other sources, including reservoirs, open burning simple emission factors can be proposed, but they are very uncertain and in most cases demand statistical data which is hardly available.

The detailed methodology to estimate emissions of HCB is based on measurements of a certain source or selected sources from a source category. For reservoirs emission modeling is to be applied, accounting content of HCB in substrate, properties of soils and surfaces, climate conditions etc.

#### 6. ACTIVITY STATISTICS

Data on the HCB usage as pesticide, industrial production and fuel combustion where HCB is emitted as by-product is available from national statistical reports. Data on scrap reclamation, wastes incineration and disposal, use of products potentially polluted with HCB (solvents) can be obtained from industry associations or enterprises. This information usually is satisfactory to estimate emissions with the use of the simpler estimation methodology. However, in most cases, no data are available from the statistics on the level of products pollution with HCB and on HCB-contaminated reservoirs. In such cases special studies are necessary or some expert estimates to be made.

The application of the detailed estimation methodology may be complicated unless the statistical data are available directly from a given source of emission.

Data on HCB content in soils, bottom sediments and wastes, wastes amount and density, air temperature, precipitation, etc. are necessary to evaluate HCB emissions from contaminated soils

and other substrates. At present, many of these parameters are out of public knowledge, since field studies and measurements available are rather limited.

## 7. POINT SOURCE CRITERIA

Chlorinated solvents production plants, secondary aluminium smelters, waste incinerators, sinter plants, magnesium plants should be regarded as point sources of HCB if plant specific data are available.

As a default approach solvent use can be considered as an area source.

## 8. EMISSION FACTORS

This chapter gives emission factors by source categories. It includes mainly tables with a short description. Application of HCB as pesticide are considered in the Chapter Use of the Pesticides (Agriculture).

### Fuel Combustion

Hexachlorobenzene emissions from fuel combustion are practically not studied. It is considered, that HCB is synthesizing from carbon and chlorine during high temperature processes, however the lack of measurements is evident. More often the presence of HCB is reported in emissions for coal, firewood and wastes combustion. Since HCB concentrations are rather low, the accuracy of measurements needs to be checked and specified. The content of HCB is defined by a large number of factors that is why it is rather problematic to receive accurate data. However, modernization of fuel combustion units and waste gas control systems contributes to HCB emission reduction (CGLI'S First..., 1998).

HCB emissions for coal and firewood combustion (with no source category) were taken into account for HCB emission inventory in Europe in the frames of the project POPCYCLING-Baltic (Pacyna et al., 1999). During European inventory 1990 (Berdowski et al., 1997) only emissions from residual oil combustion were taken into consideration.

Table 8.1 gives HCB emission factors available from literature.

**Table 8.1 HCB emission factors for fuel combustion, µg/t**

Fuel	Pacyna et al., 1999; Estimates of Mercury..., 2001
Hard coal	16
Firewood	60

### Sinter production

Sinter plant being a significant dioxin source, can be, too, a source of hexachlorobenzene emissions. Recommended emission factor is 32 µg/t (see Chapter B331 in the Guidebook).

### Magnesium production

Another potentially major source of HCB formation and release is the electrolytic production of magnesium. On a world-wide scale magnesium production makes up to about 1 million tons. Chlorine-based magnesium production processes are known to generate and release relatively large quantities of organochlorines, notably dioxins/furans and HCB (Bramley, 1998; Zero Toxics ..., 2001; Supplementary Guide..., 2000).

According to (Bramley, 1998), approximately between one-quarter and one-third of world magnesium production uses chemical reduction, and two-thirds to three-quarters uses electrolysis of  $MgCl_2$  (chlorine-based processes).

There are three principal processes which result HCB emission: 1) conversion of  $MgO/MgCO_3$  to  $MgCl_2$  by heating with coke in a pure  $Cl_2$  atmosphere; 2) purification of molten  $MgCl_2$  by transformation of residual  $MgO$  using gaseous  $HCl$ , mixed with graphite blades; and 3) electrolysis of molten  $MgCl_2$  using anodes and bipolar electrodes made from graphite. The first two stages can be applied at some plants, while the third one is common to all chlorine-based processes.

In accordance with (Bramley, 1998) the minimum values of HCB emission factors make up to 90-170 mg/t, maximum – 700–3000 mg/t. For one plant HCB emission factor made up to 300 mg/t.

### **Secondary aluminum production**

Secondary aluminum refers to aluminum-bearing scrap or aluminum-bearing materials. Secondary aluminum smelting involves two processes – pre-cleaning and smelting – both of which may produce emissions of dioxins/furans and HCB.

Scrap and aluminum raw material is made in reverberatory double-hearth and electric induction furnaces. Covering fluxes (300–350 kg/t metal) consisting of sylvinit and waste electrolyte of magnesium production are introduced into liquid bath during smelting in reverberatory furnaces. If necessary, refining fluxes containing sodium and potassium chloride, cryolite and fluorite are used.

The recommended HCB emission factor for secondary aluminum production is 5 g/t (see Chapter B3310 in the Guidebook).

### **Non-ferrous casting**

Non-ferrous metal melting is conducted in electric and gas-fired (combustion) furnaces of various designs. Hexachlorobenzene emissions are stipulated by hexachloroethane application at the metal refining stage. According to (Calculation of contaminants..., 2002) emission factors are 1.0 – 1.2 kg/t for melting of aluminium alloy in resistor furnace; 0.8 – 1.2 in crucible furnace and 1.2 kg/t in gas furnace.

The recommended factor for HCB emission evaluation is 0.5 g/t.

### **Cement production**

Cement production can be a HCB emission source. The emission factor is 11  $\mu$ g/t cement (see Chapter B331 in the Guidebook).

### **Chemical industry**

HCB is released into various components of the environment, products and wastes in the course of chlorine-bearing production processes.

On the whole, HCB emissions were not much taken into consideration for chlorine-bearing production processes. Only in the recent decade a few studies were made and evidence of HCB synthesizing in chemical industry have emerged, although emission factors are not developed for many processes. In accordance with (Supplementary Guide..., 2000), the emissions are suggested to calculate using measured concentrations in waste gases: in case the value obtained equals or exceeds 6  $ng/m^3$ , a plant should report data on HCB emissions.

### **Chlorinated solvents production**

Chlorinated solvents are a significant source of HCB emission into the environment, here, their emissions take place both during their production stage and during their application and utilization.

The list of chlorinated solvents comprises tetrachloromethane (carbon tetrachloride), perchloroethylene (tetrachloroethylene) and trichloroethylene.

Solvent production is a large-scale process. Thus, in the former USSR the production of carbon tetrachloride in the late 1980s made up to about 100 thousand tons, and about the same amount of trichloroethylene.

HCB emission factors have been developed for chlorinated solvents production and used for some POPs inventories (Table 8.2).

**Table 8.2 HCB emission factors for chlorinated solvent production (Duiser & Veldt, 1989)**

Solvent	Emission factor, g/t
Tetrachloromethane	1-20
Trichloroethylene	1-6
Tetrachloroethylene	3-10

### Vinyl chloride production

PVC is the most widely used polymer. The world production of vinyl chloride makes up to 34 % total chlorine production, more than 30 % chlorine produced is used in its process stages. In Russia in 1988 about 700 thousand tons of vinyl chloride were produced (Fedorov, 1993), in 1990 – 445.7 thousand tons, in the Ukraine – 265 thousand tons.

Vinyl chloride is produced by different methods: ethyne hydrochlorination in vapor phase on a catalyst, combined method of ethyne and ethylene (or 1,2-dichloroethane), combined method of straight-run naphtha and balanced method of ethylene. In all cases high temperature processes that result in HCB formation are required.

Distillation residues (20–110 kg per ton production) representing a mixture of ethane and ethylene polychlorides and resinous substances, as well as tri- and perchloroethylene are used for vinyl chloride production. Secondary distillation residues are incinerated that also result in HCB release.

HCB emission factors have not been developed for this process.

### Mobile sources

Mobile sources are considered to contribute to HCB emissions. Only rather old data for the US situation are available. The emission factors used are the following (Cohen et al., 1995): diesel fuel – 21 ng/veh.km; leaded gasoline – 0.87 ng/veh.km; unleaded gasoline – 0.024 ng/veh.km.

### Solvent use

The application of chlorine-containing solvents is one of HCB sources, since HCB can be present as trace pollutants in chlorinated solvents (up to 2 µg/kg). According to evaluations (CGLI's First..., 1998), in 1992 in North America about 300 thousand tons solvents were used, and about 1 kg HCB was emitted into the environment respectively. The recommended HCB emission factor is 2 mg/t chlorine-containing solvents use.

### Waste treatment and incineration

Waste incineration and, first of all incineration of chlorinated solvents and vinyl chloride wastes, are HCB emission sources.

HCB emission factors for wastes incineration are given in the table 8.3.

**Table 8.3 HCB emission factors for wastes incineration**

Substrate	Emission factor, g/t	Reference
Tetrachloromethane	8	Duiser, Veldt, 1989
Trichloroethylene	3	-“-
Tetrachloroethylene	6	-“-
Polyvinyl chloride	5	-“-
Wastes	0.002	Berdowski et al., 1997
Solid wastes	0.01	Cohen et al., 1995
Sewage sludge	0.5	-“-
Medical wastes	0.019	-“-
	0.857	Supplementary Guide..., 2000

In other cases the following HCB emission factors may be used: for hazardous wastes (including paint production wastes and chlorine-containing wastes) – 10 mg/t, contaminated timber (wood) – 2 mg/t, other solid wastes – 0.1 mg/t.

### **Pesticides application**

The application in agriculture and gardening as a fungicide especially for seed protection made up to 100 – 600 g per 100 kg of cereal and beet seed, for soil treatment and as a pesticide for plant protection.

The estimates for HCB emissions from pesticide application depend on the usage rates of HCB-containing pesticides, the HCB content in those pesticides, and the rate of HCB volatilization from the pesticide formulation when it is applied. In accordance with (Technical Paper..., 1995), the emission factor for HCB during field use is 40 % of HCB application.

Table 8.4 includes some available HCB emission factors.

**Table 8.4 HCB emission factors for HCB use in agriculture**

Source	Emission factor	Reference
Pesticide use (agriculture)	50 – 150 mg/ha (total arable land and permanent crop) per year	Duiser & Veldt, 1989
	100 mg/ha (arable land and permanent crops)	Axenfeld et al., 1992
Field application in agriculture	0.40 t/t (greenhouses 0.20)	Berdowski et al. 1997
	0.50 t/t (< 0.1 % of total fungicide consumption)	Chapter Use of Pesticides (agriculture)

### **Open burning of municipal refuse**

The practice of waste utilization by open burning is used rather widely in many countries (Introduction to Area ..., 1999; The European Dioxin..., 2000). A list of wastes burnt in bonfires is rather large: paper, paperboard, packaging material, polyethylene film, contaminated wood, rags, tyres, plastic bottles, waste food, etc. Often street sweep which contains together with dry vegetation and leaf fall a great amount of domestic wastes are burnt in bonfires. In some of the regions, plastics (particularly PVC) are an increasingly large share of domestic waste (Zero Toxics..., 2001). Because of increased use of drinking water and milk packaged in disposable plastic containers, domestic waste in some countries contains 12–15 percent plastics. This can be compared to the 2–3 percent of plastics in domestic waste in the U.S. and U.K. (Qdais et al., 1997). According to USEPA estimates, about 21 million people in the U.S. are burning about 8 million metric tons of household waste in piles or barrels in their backyards each year (The Inventory of Sources..., 1998).

The emission factor recommended by EPA (1997) is 20 µg/t wastes incinerated.

The table 8.5 includes the set of default emission factors which may be used if no more data available on emission of HCB.

**Table 8.5 Default emissions factors of HCB**

Source category	SNAP	Emission factor	Unit
<b>Stationary fuel combustion</b>	<b>01+02+03</b>		
Hard coal		16	µg/t
Firewood		60	µg/t
<b>Production processes</b>	<b>03&amp;04</b>		
Sinter plants	030301	32	µg/t
Secondary aluminium production	030310	5	g/t
Magnesium production	030323	0.3	g/t
Non-ferrous casting	N/A	0.5	g/t
Cement	030311	11	µg/t
<b>Solvent production</b>	<b>040413</b>		
Tetrachloromethane		10	g/t
Trichloroethylene		4	g/t
Tetrachloroethylene		6	g/t
<b>Solvent and other product use</b>	<b>060406</b>	2	mg/t
<b>Road transport &amp; other mobile sources and machinery</b>	<b>07+08</b>		
Leaded gasoline		0.87	ng/km
Unleaded gasoline		0.024	ng/km
Diesel fuel		21	ng/km
<b>Waste treatment and disposal</b>	<b>09</b>		
Incineration of domestic or municipal wastes	090201	0.001	g/t
Incineration of industrial wastes	090202		
Tetrachloromethane		8	g/t
Trichloroethylene		3	g/t
Tetrachloroethylene		6	g/t
Polyvinyl chloride		5	g/t
Hazardous wastes*		0.01	g/t
Contaminated wood		0.002	g/t
Other solid wastes		0.0001	g/t
Incineration of hospital wastes	090207	0.019	g/t
Incineration of sludge from waste water treatment	090205	0.5	g/t
<b>Agriculture</b>	<b>10</b>		
Use of pesticides (HCB)	100600	0.5	t/t

\* - including paint production wastes and chlorine-containing wastes

## 9 SPECIES PROFILES

## 10 UNCERTAINTY ESTIMATES

An uncertainty of HCB emission calculation consists of uncertainties of emission factors and uncertainties of statistical (production / activity) data. First type of uncertainty can be estimated statistically on the basis of experimental data on HCB concentration variability in emission; second type depends on accuracy of statistical data and varies from country to country.



On the whole, the precision and accuracy of HCB emission calculation depends mainly on the quality of emission factors. But lack of statistical data for some source categories also results in high level of uncertainty.

## **11 WEAKEST ASPECTS IN CURRENT METHODOLOGY**

There are a lot of weak spots in HCB emission estimates due to poor level of investigation of their sources. Obtaining of additional data on application of products polluted with HCB, distribution of HCB-contaminated substrates, combustion and disposal of HCB-contaminated wastes should be considered as priority elements of HCB emission inventory improvement.

## **12 SPATIAL DISAGGREGATION CRITERIA FOR AREA SOURCES**

## **13 TEMPORAL DISAGGREGATION CRITERIA**

## **14 ADDITIONAL COMMENTS**

## **15 SUPPLEMENTARY DOCUMENTS**

## **16 VERIFICATION PROCESSES**

Simplest verification of emissions can be done for by comparing the results of emission inventory of HCB in similar countries.

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## 19. RELEASE VERSION, DATE AND SOURCE

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Authors: Sergey Kakareka  
Tamara Kukharchyk  
Institute for Problems of Natural Resources Use and Ecology  
National Academy of Sciences of Belarus  
10 Staroborysovski trakt, Minsk 220114  
Belarus  
Tel/fax: +375 17 290 34 27  
[kakareka@ns.ecology.ac.by](mailto:kakareka@ns.ecology.ac.by)