




International POPs Elimination Project

Fostering Active and Efficient Civil Society Participation in Preparation for Implementation of the Stockholm Convention

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Dioxins, PCBs, HCB in Slovakia - Country Situation Report

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Slovakia
April 2006

About the International POPs Elimination Project

On May 1, 2004, the International POPs Elimination Network (IPEN http://www.ipen.org) began a global NGO project called the International POPs Elimination Project (IPEP) in partnership with the United Nations Industrial Development Organization (UNIDO) and the United Nations Environment Program (UNEP). The Global Environment Facility (GEF) provided core funding for the project.

IPEP has three principal objectives:

- Encourage and enable NGOs in 40 developing and transitional countries to engage in activities that provide concrete and immediate contributions to country efforts in preparing for the implementation of the Stockholm Convention;

- Enhance the skills and knowledge of NGOs to help build their capacity as effective stakeholders in the Convention implementation process;

- Help establish regional and national NGO coordination and capacity in all regions of the world in support of longer term efforts to achieve chemical safety.

IPEP will support preparation of reports on country situation, hotspots, policy briefs, and regional activities. Three principal types of activities will be supported by IPEP: participation in the National Implementation Plan, training and awareness workshops, and public information and awareness campaigns.

For more information, please see http://www.ipen.org

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Table of contents

Introduction
Persistent organic pollutants in the Slovak Republic

Persistent organic pollutants (POPs) are a category of compounds representing one of the most serious environmental problems nowadays. The term ‚persistent organic pollutants' is used for contaminating substances (pollutants) of organic origin, with chemical structure that has changed so much that they degrade very slowly and remain in the environment and organisms for a long time.

Persistent organic pollutants are characterised by their high solubility in fats and ability to accumulate in the fatty tissues of living organisms. Their long-time abundance in the environment, bioaccumulative capabilities and long range transport ability have caused contamination and impairment of living organisms in remote areas where their occurrence had not been assumed.

More accurate scientific knowledge describing the dangerous effects of these substances to human health and the environment has been acknowledged by international institutions such as the United Nations Organisation (UNO) and the World Health Organisation (WHO). These institutions initiated the development and adoption of Stockholm Convention on POPs elimination which was signed on May 22, 2001 in Stockholm.

The Slovak republic ratified the Stockholm Convention on August 5, 2002. It entered into force on May 17, 2004.

Friends of the Earth – SPZ believe it is necessary to pay more attention to POPs. Comparing the risk arising from POPs and from some of the "known" pollutants, it is necessary to consider the effects of POPs are made worse by their persistence, bioaccumulation and toxicity causing effects at very low amounts.

Results of research and reviews made by scientific institutions indicate that there are many sources of the production of polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzo furans (PCDFs) and polychlorinated biphenyls (PCBs) in Slovakia. This has already been negatively displayed in the Michalovce region in Slovakia where people have been exposed to PCB contamination from the former production facility - Chemko Strážske). [1]

Unfortunately this locality is not the only place where contamination with PCBs has occurred. Over the limit amounts of dioxins has been found in food produced in the surrounding area of the Košice waste incineration plant and many other over-the-limit amounts of POPs found in water, food and air indicate that the problem of contamination of the environment with these substances still remains and it is necessary to implement measures to eliminate them.

Unwanted by-products of combustion processes and industrial production

1. Dioxins
Polychlorinated dibenzo-p-dioxins and Polychlorinated dibenzofurans

Physical and chemical properties of dioxins

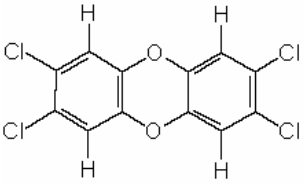
Chemical base:

210 chemical substances belong to the 2 categories scholarly termed as polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) called dioxins. There are 75 possible congeners of PCDDs and 135 congeners of PCDFs. Furans are mostly produced together with dioxins and both types of substances accompany each other in the natural environment [2]. Chemically they belong to the group of chlorinated aromatic compounds. We distinguish mono – octa derivates, as they differ by the number of chlorine atom constituents. Four types of compounds have from 4 to 8 chlorine constituents that can be combined to make numerous isomers.

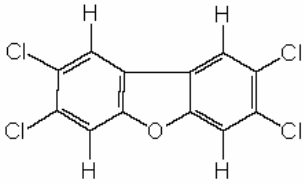
Number of chlorine atoms	Abbrev. PCDD/F	Number of isomers	
		PCDD	PCDF
4	TCDD/F	22	38
5	PeCDD/F	14	28
6	HxCDD/F	10	16
7	HpCDD/F	2	4
8	OCDD/F	1	
Total number of congeners incl. mono – tri PCDDs/Fs		75	135

Carbon skeletons of dioxin and furan (very similar properties) are compounds of aromatic or benzene cores. They contain 8 carbon atoms having hydrogen atoms bonded to them that may be substituted by chlorine or bromine. According to the number of substituted hydrogen atoms we distinguish mono- octa derivates. Polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are originated when hydrogen atoms are substituted by chlorine atoms. Isomers have the same number of chlorine atoms in the molecule, but they differ in the positions of these atoms. All related substances are then called congeners, with no regard to number of hydrogen atoms substituted by chlorine.[3]

Formula:



2,3,7,8-TCDD



2,3,7,8-TCDF

Chemical Abstracts Service Registry Number (CASRN):
Dioxin (Tetrachlorodibenzo-p-dioxin) - 41903-57-5
Furan (Dibenzofuran) - 11141-16-5

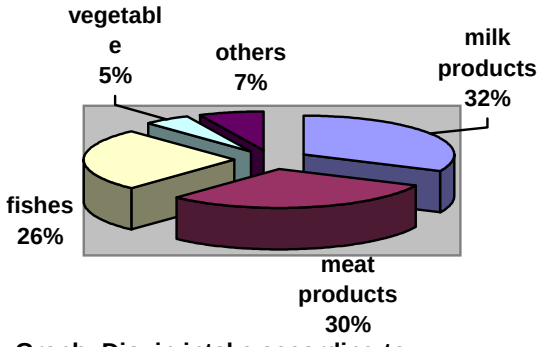
1.1. Properties of PCDDs and PCDFs

PCDDs and PCDFs are **persistent substances** (highly stable). They remain in soil, water, sediments, air and living organisms. They are characterized as resistant to phytological, biological and chemical decay. [2] The half-life of 2,3,7,8 tetrachlordibenzo-p-dioxin in soil can be 12 years [5] and the half-life of elimination from the human body is about 5, 6 [6] – 7, 5 years [9]. It is necessary to stress that **the half-life of elimination is not the same at all times, it depends on dose, body structure, age and sex.** [9] That is why the data in the following table can be taken as a reference only to a certain extent.

Data on half-life of PCDD/F congeners' elimination from the human body[16] a [9]

Congener	Half-life of elimination (years)
2378 – TCDD	5 – 7,5
12378 – PeCDD	5
23478 – PeCDF	6,3
123478 – HxCDD	11
1234678 – HpCDD	9
OCDD	12

It is necessary to realise that the half-life co-efficients presented above are relative, depending on PCDDs/Fs amount/concentration in respective components of the environment and human body. It does not mean that PCDDs will be automatically eliminated after this time; their concentration will be decreased by half.



Graph: Dioxin intake according to sort of food

In living organisms it is mainly congeners substituted in positions 2, 3, 7 and 8 that have been primarily accumulated. Dioxin elimination from the human body is considerably slower than from other researched mammals. As a result there are higher levels of PCDDs/Fs in human tissues than in rodents for example, even when both humans and rodents are exposed to the same doses. The half-life of elimination (the time after which half of the absorbed dioxin has been eliminated) of 2,3,7,8 TCDD from a rat‘s body is 20 days, while the elimination from the human body ranges from 4 to 7,5 years, more than 100 times longer. The same thing can be noted for the most significant form of furans (2,3,7,8-TCDF), its half-time of elimination from a rats body is 2 days, but from the human body it is 16 months, 200 times longer.[2]

These compounds also have extremely **high bioaccumulative properties,** [4] the ability to accumulate and be retained in the fatty tissues of living organisms. They can remain in soil for 3 - 18 years, in living organisms' fats it can be as long as 17 years [4]. They accumulate mostly in fats, both animal and vegetable, in tissues rich in fats, e.g. liver or milk glands. Subsequent accumulation in the food chain causes their higher concentration at the “top” of the food chain. Through eating dairy products, meat and fish, a human absorbs more than 80% of the total amount of dioxins in his/her body.

The main source of dioxin intake is apparently food (but generally they come from emissions originating from the process of combustion before entering the food chain). We intake more than 90% through food and 1 – 5% by breathing. Drinking water is a minor source of dioxins.[11]

Toxicity

PCDDs and PCDFs are highly toxic, they belong to the most toxic of man-made substances and are not produced by nature [8]. According to some sources they are 500 times more toxic than sodium cyanide [4]. There are large differences in congeners' toxicity. Seventeen congeners substituted in positions 2,3,7,8 have strong health-impairing effects [2] – these are regarded as being highly toxic. Compounds with toxicity similar to that of 2,3,7,8 – substituted PCDDs and PCDFs belong to the so-called planar polychlorinated biphenyls [2].

In May 1998 a conference – consultation took place in Switzerland under the patronage of the WHO, and attended by the International Agency for Research Cancer (IARC). The scientists participating at the conference came to a conclusion that dioxins are 2 – 10 times more toxic than it had been anticipated in 1990.

There has been introduced the so-called **toxicity equivalence factors** (TEFs) of respective congeners (Table 1) for dioxins and dioxin-like PCBs and TEFs are used for assessing the toxicity of these. It has been introduced to simplify risk assessment and toxicological dose. Calculation should be made of the total toxicological dose by adding the contributions of individual compounds. Antagonistic or synergistic relations are not considered):

$$TEQ = sum\ (PCDDi \times TEFi) + sum\ (PCDFi \times TEFi) + sum\ (PCBi \times TEFi).$$ [9]

In a simple way – TEQ indicates the concentration has the same toxic effect as if there was only 2, 3, 7, 8-TCDD present in a sample at the same concentration, because the toxicity and other properties of 2, 3, 7, 8- TCDD have been researched most[2]

2,3,7,8 – TCDD is considered to be the most toxic of this category of compounds. [9] Sometimes it is called "Seveso" dioxin, because of industrial incident in the Italian town of Seveso in 1976, where this type of dioxin contaminated the town and the surrounding area.

Mechanism of dioxin effects

A wide range of data shows that the Ah receptor plays a key role in the biological effects of dioxins. Even though the accurate mechanism of action and molecular effect on the activated receptor is still not known exactly, it is assumed that the toxicity of dioxins is caused by changes in the biochemical and cell processes initiated by the activated Ah receptor. Studies acknowledge that the Ah receptor plays a key role in the effects of TCDDs. The activated receptor causes two main changes: intensifies the transcription of a group of genes sensitive to dioxins and immediately activates the tyrosine kinases. The Ah receptor can also directly or indirectly regulate the activity of other groups of genes. The activation of receptors can cause disruption of the endocrine and other systems and changes in cell processes, including growth and division. Some of these effects have been already observed, both in humans and animals indicating that the mechanism of action is the same.[9]

Table 1: Toxicity equivalent factors set forth by WHO to assess the risk for humans based on a meeting that took place in Stockholm, Sweden in 1997 (Van den Berg et al., 1998).

Congener	*TEF Degree*	***Congener***	*TEF Degree*
Dibenzo-p-dioxins		**Non-ortho PCB**	
2,3,7,8-TCDD	1	PCB 77	0,0001
1,2,3,7,8-PnCDD	1	PCB 81	0,0001
1,2,3,4,7,8-HxCDD	0,1	PCB 126	0,1
1,2,3,6,7,8-HxCDD	0,1	PCB 169	0,01
1,2,3,7,8,9-HxCDD	0,1		
1,2,3,4,6,7,8-HpCDD	0,01	**Mono-ortho PCB**	
OCDD	0,0001	PCB 105	0,0001
		PCB 114	0.0005
Dibenzofurans		PCB 118	0,0001
2,3,7,8-TCDF	0,1	PCB 123	0,0001
1,2,3,7,8-PnCDF	0,05	PCB 156	0,0005
2,3,4,7,8-PnCDF	0,5	PCB 157	0,0005
1,2,3,4,7,8-HCDF	0,1	PCB 167	0,00001
1,2,3,6,7,8-HxCDF	0,1	PCB 189	0,0001
1,2,3,7,8,9-HxCDF	0,1		
2,3,4,6,7,8-HxCDF	0,1		
1,2,3,4,6,7,8-HpCDF	0,01		
1,2,3,4,7,8,9-HpCDF	0,01		
OCDF	0,0001		

Acute toxicity of 2, 3, 7, 8-TCDD is really extreme, as can be seen in the following table.

Table 2: Average lethal oral doses LD 50 – **it shows amounts of substances, when toxic effects start to appear** on researched animals (mg/kg of body weight) - rats [3]

Substance	**LD50**	**Substance**	**LD50**
Botulin toxin	3.10^{-8}	Curare	20
2,3,7,8-TCDD (tetrachlordibenzo-p-dioxin)	**0,22**	Mercuric chloride	37
Ditantalum sulfide	15	Arsenic trioxide	45
Strychnine	16,2		

1.2. Negative effects on human health

Dioxins and related compounds belong to highly toxic substances that may cause health malfunctions even when exposure is to minimal doses.

Assessing the effects to health, the European Economic Commission of UNO came to a conclusion that dioxin was the first substance ever, to be considered a global danger for all living organisms.[10]

Dioxins are connected with a wide range of adverse effects to animal and human health [Reinjders 1992, Fox 1992, US EPA 1994]. Many times the effect of dioxins does not necessarily appear on the exposed adults, but appears later in their offspring.[18]

A short summary of proven negative effects directly connected with many congeners of dioxins and furans:

- **dermal diseases** (example chloracne)
- **impairment and subsequent malfunctions of the immune** (immunotoxicity) **system**
- **ability to damage the developing foetus** (teratogenicity)
- **carcinogenicity** (support cancer development, cancer has been reported in every animal species tested); International Agency for Research on Cancer / part of WHO classified the most toxic form of dioxin 2,3,7,8-TCDD as a known human carcinogens and many other dioxins among possible human carcinogens)
- **interferences/disruption of the endocrine system** (can cause malfunctions of male sex organs – threaten male fertility, cause malfunctions of female sex organs – lower fertility, miscarriages, malfunctions of ovaries); [9, 18]
- **negatively affect cardiovascular system,**
- **damage of nervous system** (e.g. certain delay and changes in neurological development of newborn babies whose mothers have been exposed to high doses of PCDDs/Fs, observed interference in behaviour)
- **increase the occurrence of diabetes**
- **cause teeth malfunctions**

[9, 18, 19]

More information on respective negative effects:

The effects of dioxins are different when it is a single high-dose exposure (accidents, etc.) than long-term exposure to small doses. The effects following a single high dose exposure include: chloracne, problems with the nervous system, malfunctions of fats and saccharides metabolism, damage to urinary and breathing organs, pancreas and in many cases the liver. There have also been observed neurological and psychological interference. After a long-term exposure to small doses the endocrine system can be negatively affected, immunity decreased and a contribution made to the development of cancer[4, 17].

Health-damaging (non-carcinogenic) effects on children:

Great attention is focused on the effects on babies because they are exposed to high doses in a short period of time. It could affect their nervous, immune, reproductive or intelligence development [30]. During the first months of their life babies depend on their mother's milk which contains higher levels of dioxins than cow's milk.[11] But the first entry for dioxins to the baby's body is via transplacental passage from mother to baby.

Based on studies conducted on dioxins concentration in breast milk, WHO stated that babies' intake is 20 – 130 pg I-TEQ/kg/day. If we consider the additional contribution of similar congeners of PCBs, a baby's daily intake can be twice as much as the WHO estimate i.e. 40-260 pg I-TEQ/kg/day. A study conducted in Germany, found that a baby's intake is between 27 and 418 pg I-TEQ/kg per day from breast milk; 10% of this exposure was from 2, 3, 7, 8-TCDD. It was found that the percentage of all the remaining congeners in breast milk is very high and exceeds 90%. These numbers exceed the "acceptable" daily intake (ADI) recommended for an adult population. But WHO suggests the ADI should not be used for babies, because its estimate comes from a lifetime intake. WHO says it is balanced for babies because of their smaller intakes during the later years of their life.

WHO says a baby should not intake more than 5% of whole life intake during first 6 months of life when it is breast feeding. US EPA estimates that American babies ingest 4 - 16% of their life-time intake

during this period. Specialists do not recommend solving this problem by reducing breast feeding, but by introducing a radical solution of preventing dioxins entering the food chain [11]. Despite these facts breast fed babies are healthier than those fed with formula/ commercial milk[14]. Babies in a Dutch control group have been characterized as having better neurological development compared to babies fed on formula/commercial milk. But within the group of breast-fed babies, those exposed to the highest levels TEQ of dioxins (more than 50 pg/g of fat in mother's milk) had the worst results. [9]

Delay and changes in neurological development of newborns were observed in two groups of American (exposed to PCBs) and 1 group of Dutch babies (exposed to PCDDs/Fs and PCBs). Neurological problems appeared in both American groups only where there was indicated the highest level of transplacental transport with a hint of nonlinear effect. There were observed many outcomes of transplacental exposition in a control group of Japanese and Taiwanese children (exposed through the placenta to contaminant from rice oil including ectodermal defects, delay of development, low weight at birth, smaller permanent behavioural interference, smaller dimension of penis at puberty, lower height of girls at puberty, and loss of hearing.

With regards to diversity of the substances people are exposed to, it is necessary to say it is not certain to what extent dioxins and similar substances cause these effects. In all studies of babies and children the outcomes were considered to be a consequence of transplacental transport rather than the supply of mother's milk.

The children of Seveso who were exposed to high amounts of TCDD showed the following consequences: small temporary increase of liver enzymes, an increase of number of lymphocytes and their subgroups, temporary chloracne. Children of parents exposed to high levels of TCDD showed an increase in the number of girls born.[9]

WHO states that non-carcinogenic negative effects of dioxins and dioxin-like substances were found as a result of different exposures under different conditions – small delay's in development, small changes in thyroid activity for children with a high extent of transplacental exposure.[9]

There is a study showing that a baby is 100 times more sensitive to dioxin exposure than an adult. A small measured intake of dioxin given to a pregnant rat female at a critical moment of development permanently destroys the reproductive system of the foetuses. Subsequently males exhibit considerably weaker male behaviour and sperm decrease to 40%. The level of dioxin used in this experiment was similar to its level and the level of other dioxin-like substances found in organisms of people living in industrialised areas of Europe, Japan and USA. The Swedish study found a correlation between an amount of PCBs, dioxins and furans in food and a significant reduction of the body's so-called natural "killers," lymphocytes that have an essential role in a organism's defence against infection and cancer. Canadian scientists found that children exposed to high levels of persistent contaminants, had 10 – 15 times more infections than a comparable group of children without these exposures. [WWF: Persistent organic pollutants: hand-me-down poisons that threaten wildlife and people, 1/1999, Washington D.C.]

Learning and behaviour problems Scientists from Detroit found that children exposed to PCBs and other contaminants from Michigan lake fish consumed by their mothers during the 6 years preceding the pregnancy had significant problems with learning and attention span. At the age of 11 the most exposed children had problems with paying attention, problems with both short-and long term memory two times higher probability of a two-year delay in reading skills and three times higher probability of a low IQ. This study is important not only because it showed long-term damage from chemicals to children's health, but also because it found that mothers that ate fish were not themselves considered heavily contaminated with the levels of dioxins in their bodies within the upper limits of "normal" basal level.

In a similar study from New York scientists found a measured interference in the behaviour of new born babies of women that ate 60 pounds of salmon from the contaminated Lake Ontario during their life. These children had abnormal reflexes, could not concentrate for long periods and could not tolerate stress. It was the first study that approved a wide range of affecting temperament when prenatal exposure by a contaminant occurs.

The role that PCBs and dioxins play in learning and behaviour problems: After studying the available literature the US Department of Health and Human Services concluded that PCBs and dioxins are responsible, at least to a certain extent, for neurological problems and behavioural interferences occurring when children are exposed to a contaminant during their mothers' pregnancies. The Agency for Toxic Substances states that there is a significant correlation between epidemiologic studies on people, results found in laboratories, and studies conducted on wild animals: "Quite a lot of information shows that some PCBs or substances similar to them found in fish can lead to interference in behaviour. The same interference was found in people eating fish from the Great Lakes area . [WWF: Persistent organic pollutants: hand-me-down poisons that threaten wildlife and people, 1/1999, Washington D.C.]

Health-damaging (non-carcinogenic) effects on adults:

At the conference of WHO which took place in May 1998 scientists came to a conclusion that there can be expected following negative effects (TCDD) to humans:
- lower level of sperms appearing in the generation of offspring when an exposure is 14 pg / kg of weight per a day.
- lower ability to learn when there is a daily exposure of 21 pg / kg of weight
- interference of immune system appearing in the offspring of people exposed to intakes of about 25 pg / kg of weight per a day
- higher extent of genital deformations in offspring of people exposed to daily intake about 37 pg/kg of weight

[9, 20]

In many groups exposed to higher levels of PCDDs/Fs there was an increase in diabetes. Increased mortality connected with diabetes appeared in Seveso where women were affected. A few industrial studies conducted in Seveso showed an increased mortality caused by cardiovascular diseases. The connection was also found for ischemic heart disease. Many of the observed effects lasted for the period of exposure and then disappeared. Individuals exposed to PCDDs/Fs and PCBs can differ from control groups in changes of lipid concentration, plasma glucose and higher mortality caused by cardiovascular diseases.[9]

Recently proof has come to light on the considerable ability of dioxins to disrupt the endocrine system, even when the dose is very low (trace dose Dioxins can have following effects:
- interference of male sex organs such as: weak sperm and reduced sperm count, testicular atrophy, small dimension of sex organs , decreased level of testosterone, changes in sexual behaviour;
- interference of female sex organs: decreased fertility, miscarriages, malfunctions of ovaries, endocrine changes

[18]

Males exposed to dioxins during their adolescence, father girls more often. This statement was published by Italian doctors in May 2000. It is based on results of study researching men exposed to chemicals after explosion of factory producing herbicides in Seveso in 1976. The study published in the medical magazine *Lancet* showed that dioxins affect the proportion of sex of born children. It seems that very low concentrations of dioxins have long term effects on the male reproductive organs. The concentrations found were 20 times higher normal and were equivalent to the amount causing reproductive interference in laboratory mice.[13] Even low concentrations of dioxins cause an increased production of some enzymes and are potentially genotoxic. [16]

A new study conducted by Finnish dentists who have dealt with the issue for about 10 years, showed some people have their teeth disrupted as a result of exposure to dioxins. They noticed at the beginning of the 1980s that many children have badly developed molars. They were soft and colourless and some areas had no covering of enamel enabling cavities to attack them [21, 19]. Chinese children born to mothers who were exposed to high levels of dioxins had similarly disrupted teeth [22]. The Finish scientists began conducting experiments on animals and monitored those children and their mothers on whom analyses of mother's milk had been done. The Finish study found that children with the worst teeth were born to mothers who had the highest concentrations of dioxins in their milk. A direct relation between the state of the teeth and concentrations of dioxins contained in mother's milk was shown.[19]

Carcinogenic effects:

WHO shifted dioxins to a group of known human carcinogens and subsequently the US EPA (US Environmental Protection Agency) came to this significant decision.[1]

Long lasting studies on the effects of an organisms'exposure to 2, 3, 7, 8-TCDD showed it is carcinogenic to many animal species regardless of sex. Some other PCDDs/Fs and planar and mono-ortho-substituted PCBs are also promoters of carcinogenic diseases.[9]

In the majority of studies an effect of carcinogenic promotion was proven, and to a smaller extent an initiator. It could explain why dioxins cause a general increase in different types of cancer developing after exposure. [14]

Some essential information comes from several studies that for a long time observed the effects on people exposed to higher levels of dioxins (e.g. 4 industrial studies on herbicide producers from USA, Netherlands and 2 from Germany, a study on residents from contaminated area of Seveso, Italy, and studies IARC from different states, etc.)

Epidemiological studies on individuals and groups exposed to the highest levels of 2, 3, 7, 8-TCDD provide strong evidence about an increased risk to cancer (generally) and weaker evidence about risk to a particular type of cancer. Studies from Germany assessed mainly to TCDD and PCDD/F effects (according to I-TEQ) and observed a strong relation between disease and exposure to this dioxin. The majority of these studies found an increased risk of developing soft tissue sarcomas, lung cancer, non-Hodgkin's lymphomas and cancer of the digestive system. Statistically significant increased risk to other types of cancer (mouth and kidney cancer, leukaemia and women's breast cancer) was also observed. [9] WHO states that the public is on average exposed to doses of TCDD of the lower order and to 1-2 times lower doses of PCDDs/Fs than people from these studies or the residents of Seveso have been exposed to.

In 1998 a group of German scientists came to a conclusion that dioxins can be responsible for almost 12% of cancer occurring in industrial world. [15]

Representatives of some industrial sectors tried to convince the public of the existence of a "threshold intake level" of dioxins – supposedly only after crossing this threshold level does dioxins start to cause damage. Unfortunately even the most complex study (US EPA 1994-todate) failed to find the existence of such a threshold level.

Participants of the WHO consultation which took part in the Swiss meeting in 1998 stated that certain small effects can appear in some parts of the population of industrial states as a result of exposure to contemporary average concentrations ingested daily – 2-6 pg/kg of weight. [9, 20]

1.3. Sources of dioxins

When a detailed assessment of dioxin sources is being carried out, it is necessary to distinguish two areas.

Primary sources can be termed those substances whose production and presence at respective facilities is the primary reason for formation of dioxins and similar substances.

The Secondary areas are facilities with technologies that form emissions of dioxins and different residuals containing dioxins (it is being done in the presence of primary sources) – they can be called **secondary sources** of dioxins (or sources of production).

Primary sources of PCDDs/Fs

Dioxins are not produced intentionally, they originate as unwanted by products of incineration processes and the chemical industry if chlorine is present (in any form). Scientific studies have shown that **dioxins are formed when combustion with chlorine** occurs. Several studies -[42, 43, 44] - show a statistical relation between the amount of contained chlorine at the input of waste incinerators (and similar incinerators) and dioxin emissions. **Production of PVC is the biggest consumer of chlorine**. About 35% of chlorine production ends in PVC. [Green paper, EÚ, COM (2000)469, 26/7/2000] Based on this fact and on the conclusions of scientific studies showing a relation between dioxin production in thermal processes (when a chlorine is present), it is highly probable that PVC is one of the most significant primary sources of dioxins. It is expected that without legislative interference its 1/3 proportion will be increased, because chlorine consumption decreases in other industrial sectors (e.g. wood processing and paper industry).

High amounts of dioxins connected with PVC production are described by several studies (Evers 1989, 1993, Adelt 1990, Costner 1994, Johnson 1994). Representatives of the PVC industry have admitted that it is necessary to emit dioxins when producing VCM: "*It is very difficult to find such modification of operational conditions that would prevent PCDDs/Fs production and at the same time not considerably change the reaction that is the purpose of the operating process*." (ICI – 1994).

Another significant **source of dioxin production is the disposal of PVC waste materials**. Studies from 1994-95 conducted by Princeton and Florida universities **show a statistical relationship between chlorine concentration in incinerated waste and emissions of dioxins.** Researchers from laboratories of clean combusting technologies came to the conclusion that:

"Decrease of organically bonded chlorine at inputs leads to a decrease of emissions of chlorinated organic substances. We assume that if maintaining constant levels of other agents there is direct correlation between PVC at input and PCDDs and PCDFs at output. We are convinced that it would be effective to minimalize the amount of chlorinated plastic materials at incinerators' inputs" (Wagner 1993).

This relationship was endorsed by European studies, e.g. a study conducted for Danish Department of the Environment came to the conclusion: *"These new experiments clearly demonstrated relation between PVC concentration in municipal waste and amount of dioxin produced by waste incinerators."* (NME 1994).

In "Concept of dioxin re-evaluation" US EPA (1994) evaluated municipal and hospital waste incinerators as the most significant source of dioxin emissions. PVC is the most significant source of chlorine in hospital waste incinerators (Maraca 1998). The German Environmental Agency estimates that **solid PVC (e.g. bottles and gallipots) is a source of 50% of chlorine present at input to municipal waste incinerators,** even though its proportion in total weight is only 0,5% (Brahms 1989). Also Danish EPA states that PVC is a large chlorine source in wastes entering the municipal waste incinerators [18] One of the last EÚ reports [Green paper, COM (2000)469, 26/7/2000] states that in the European Community incinerated waste flows comprised PVC waste materials – 38% - 66%, decomposing substances (biowaste) – 17% and paper – 10%., which are another source of chlorine.

Anthropogenic versus natural sources of dioxins

Until recently there was a dispute as to whether dioxins come from natural or anthropogenic sources. After several studies it is evident that dioxins in our environment come mainly from industrial processes.

Even if some dioxins arise naturally (forest fires) their amount is minor (according to US EPA <1%). More importantly, these dioxins most likely reflect redistribution of industrial sources of dioxins that settled on leaves and other forest matter. In 1995 a scientific advisory council of US EPA was dealing with issue "Anthropogenic (artificial) versus natural sources of dioxins" and came to the conclusion:

"*Observations carried out at the end of the 1970s showed that dioxins arise when normal materials, including solid municipal waste is being burned which led some of the scientists to a thought that dioxins*

accompany man from use of fire and that they can arise by natural combustion (forest fires). At that time it appeared that observed levels of dioxins are primary a result of burning of coal or perhaps wood in small fire-places".

"These assumptions were for the greater part disproved by studies on sediments conducted in the USA (mainly in Great Lakes) and Europe that showed level of dioxins in the environment has considerably increased by the years 1935-1940 (see file II, p..3-92-94)".

"Because the use of fire had undoubtedly preceded this date we can deduce that dioxins are mainly of anthropogenic origin and their occurrence is connected with events that took place sometime around 1935-1940. What events did happen at that time? We can exclude burning of coal, because coal consumption was in the USA constant from the beginning of the century until approximately 1970. These data do not comply with sediment measures."

So the main source of dioxins in the environment is not burning itself. The source is production and burning of certain artificial (anthropogenic) materials containing chlorine, which were introduced in large amounts for the first time after 1930. U.S. EPA states:

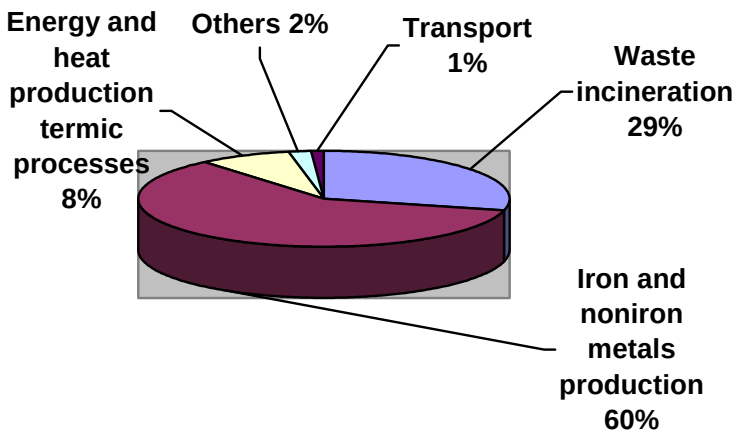
"*Explanation is probably in the fact that there were introduced chlorinated organic compounds (e.g. polyvinylchloride and chlorinated pesticides) around years1935-1940. Significant contribution could be added by other sources, such as lead petrol (which normally contained dichloroethane and dibromomethane), emissions from diesel engines and PCBs".*

" Even if details on arising of dioxins have not been quantitatively known so far, it seems that the occurrence of chlorinated substances in burned waste is the most probable cause of increased measured dioxin depositing in sediments."[41]

Secondary sources of dioxins

Dioxin emissions in the Slovak Republic (SR) were evaluated in „Proposal of the National Implementation Plan under the Stockholm Convention on POPs in the SR" where the following proportion of respective sources is stated:

Contribution of sectors to atmospheric emissions of PCDDs/Fs



(Inventarisation of persistent organic pollutants in the SR, Technical report No. 2, sec. 2)

1. METALLURGICAL INDUSTRY (1990 - 46%, 2001 - 61%) [1]

(20% according to estimation of survey on emissions conducted by Department for Preventive and Clinical Medicine in 1993)

The greatest amount of PCDDs/PCDFs emissions is produced by agglomeration of iron ore and secondary production of iron and steel. It is necessary to stress that this sector is one of the most important industrial sectors in the SR and the amount of processed materials and manufactured products are imposing what corresponds to a great proportion of the sector on the total amount of emissions in the SR There was indicated a decrease of PCDDs/PCDFs emissions by 31% during a monitored period. This tendency was caused by decrease in production compared to 1990 but also by a gradual process of making iron and steel production more environmentally-friendly. From 2001-2003 a reconstruction of agglomeration lines was carried out to comply with the current state of Best Available Techniques (BAT) for this technology. That is why PCDDs/PCDFs emissions coming from ore agglomeration will be decreased by 80% from 2004 at unchanged amounts of production. Amounts of production have been recently rather steady in this sector and there is no assumption that they will decrease in the future. Another decrease in emissions coming from this sector will be possible only as a result of consistent application of BAT/BEP (Best Environmental Practices).

- Measurements carried out in agglomerative finishings generally showed PCDDs/Fs emissions in the scope 0,4 -4 ng TEQ/m^3. One measurement carried out on equipment without introducing a measure for emission decrease showed a concentration of almost 43 ng TEQ/m3.

- Primary and secondary production of copper is characterised by a wide scope of emission values coming from this equipment because of great differences in materials, aggregates and processes. While cleaning waste gas, current equipment can reach levels of PCDDs/Fs emissions varying from a few picograms to 2 ng TE/m^3. One shaft furnace for copper production used to emit 29 ng TEQ/m^3before optimalisation of aggregates.

- PCDDs/Fs emissions coming from melting equipment in secondary production industry of aluminium range from 0,1 to 14 ng TEQ/m^3. Values depend on type of melted aggregate, used materials and forms of cleaning a waste gas. PCDDs/Fs emissions coming from steel production from converter steel plants, cupola furnaces with preheated air, electric and arch furnaces for melting of embedded iron are considerably lower than 0,1 ng TEQ/m^3. Cupola for cold wind and rotating cylinder furnaces have higher PCDDs/Fs emissions. [68]

2. WASTE INCINERATION (37% in 1990, 29% in 2001)

(60% according to an estimation of a survey on emissions conducted by the Department for Preventive and Clinical Medicine in 1993)

As a result of the adoption of new legislation on waste treatment and air pollution prevention, the requirements of EÚ technological equipment in incinerators have been gradually met. This has resulted in a considerable decrease in the number of incinerators in the SR. On the other hand the technical level of the remaining incinerators is being gradually increased. The maximum level of emissions coming from this sector was in 1995 when the already mentioned legislative pressure had not appeared and an economic retrieval meant an increase in industrial waste production. All waste incinerators will have to fulfil strict emission norms based on BAT after 31 December, 2006. [1]

3. FUEL – ENERGETIC SECTOR (16% in 1990, 8% in 2001)

(17% according to an estimation of a survey on emissions conducted by Department for Preventive and Clinical Medicine in 1993)

Heating of commercial sector and households is the most significant contributor to POPs emissions within the energy sector. The main reason being combustion equipment in this sector is not always operating under ideal conditions and not equipped with the necessary anti-pollution devices.

- **Transport(1%)**
- **Chemical production (processes using chlorine)**

Chemical production is also a source of POPs and their emissions. This occurs mainly in processes when chlorine is used. There is not enough information available to find out the level of emissions today, and with regard to the quite small amounts of materials produced by this sector its contribution to the total atmospheric emissions is probably small, but these emissions must not be ignored from a local perspective. A more significant problem for this sector is PCDD concentration in waste. [1]

Known and assumed processes in which dioxins and similar chemicals are formed

Production of gaseous chlorine	**Chlorine electrolyses with carbon electrodes** **Chlorine electrolyses with titan electrodes**
Chemical industry - use of gaseous chlorine	Chlorine aromatic chemicals – production (chlorobenzenes, chlorophenols, PCBs and other) Pesticides Colouring agents Special chemicals Chlorinated solvents- production (trichloroethylene, tetrachloroethylene….) PVC plastic – production… (dichloroethylene, vinyl chloride) Waste production Flows Sludges from flows processing Air emissions PVC plastic articles Other aliphatic organochlorines – production (epichlorohydrin, hexachlorobutadiene) Some inorganic chlorides – production (ferrous and copper chlorides, sodium hypochlorite)
Use of gaseous chlorine - other industrial sectors	* Cellulose and paper – bleaching by chlorine (chlorine compounds**) Plant waste water flows Plant sludges Cellulose and paper articles Emissions from sludge incinerators (side effects of chlorination**) Disinfection of water and waste water Refined metals – production using chlorine (Ni, Mg)
Use of organochlorines	Production of chlorine chemicals by raw chlorine substances (nitrophenols, parathion and other) Decrease/extraction with organochlorine solvents in alkali or reactive conditions Oil refinery with organochlorine catalysts Use of pesticides by heat (wood processing, etc.) Iron/steel clinker with organochlorine cutting oils, solvents or plastic *Burning of petrol or mineral oil fuel with organochlorine additives Use of chlorine whitening substances and detergents in washing machines
Combustion, recycling and fires	* Hospital waste incinerators (PVC) – emissions to air * Municipal waste incinerators (PVC, **paper bleached by chlorine, salt in biowaste, wood treated by coverings containing chlorine compounds/pentachlorophenol) – emissions to air, residuals in ashes (**the same applies to landfills of solid municipal waste in a case of accidental fire, etc.) * Dangerous waste incinerators (used chlorine solvents, chemical industry waste, waste containing PCBs, PCTs) – air emissions, residuals in ashes

	Cement furnaces combusting dangerous waste (solvents, chemical production waste, PCBs) – air emissions, Accidental fires in houses and offices (PVC, pentachlorophenol, chlorinated solvents) Fires in industrial plants (PVC, PCBs, other chlorine-containing chemicals) Recycling of aluminium/melting (PVC) Recycling/melting of iron and steel, cars (PVC, chlorinated remains of mineral oil, chlorinated solvents) * Copper cables – recycling /melting (PVC) *Burning of wood (pentachlorophenol – coverings containing chlorine compounds) ** Car fires (parts made of PVC, chlorinated remains of mineral oil)
Transformations in the environment	Change of chlorophenols to dioxins in the environment ** Forest fires – deposition of organochlorines spread by air, chlorinated pesticides

Notes:

* Addressed by EPA in documents for its new dioxin reassessment (Cleverly 1993, Schaum 1993). List contains sectors in which dioxins and similar compounds are formed (PCBs, chlorodibenzofurans and/or hexachlorobenzene), which were confirmed in chemical analyses, also as sectors in which dioxin formation is" known or suspicious" according to EPA(EPA 1985, PCTN 1985) or NATO (Hutzinger 1988). Source- Pat Costner: Dioxin elimination - A Global Imperative, Greenpeace International, 8.3. 2000, Amsterdam.

** addition of Friends of the Earth from other sources

- **Uncontrolled burning** (of waste...).

Combustion is usually held under improper conditions and combusted substrate can be composed of a mixture of materials. Emission factors published for uncontrolled combusting processes are usually 3 orders of magnitude higher than for optimal combustion.

The contribution of home combustion to the total amount of emissions is lower when an accurate fuel is used – e.g. wood. Adding of packages (or other waste) to solid fuel increases PCDDs/Fs emissions. Using of packages when burning fuel wood can lead to an increase of PCDDs/Fs emissions from 0,06 ng TEQ/m^3 (only wood) to 8 ng TEQ/m^3 (packages added, calculated to 11% of volume O_2). [68]

Emissions from **waste landfills** - especially unofficial, uncontrolled landfills where incomplete combustion or decomposing occurs. Even in controlled landfills when a technically administered combustion of landfill gas operates through on-field burners, this process is a source of dioxins with concentrations 0,1 – 1 ngTEQ.Nm^{-3}. [49]

Many scientific studies have found that **waste incinerators** are significant producers of dioxin emissions. Even if the contribution of incinerators to dioxin emissions in EÚ States has decreased because of legislative limits and is gradually decreasing even more , they are constantly a significant source of PCDDs/Fs. As a result of installing equipment for the absorption of emissions from air, concentrations of PCDDs/Fs emissions in solid waste residues increased, and the treatment of these is unsatisfactory or insufficiently controlled. For example; toxic ashes coming from dangerous waste incinerators in Duslo Šaľa have been (and will be until 2008) placed on a poorly secured waste landfill with only temporary operating permission. There was a disclosure of unsatisfactory and illegal treatment of ashes with concentration of PCDDs/Fs coming from municipal waste incinerators in the Czech Republic. This is why it will be necessary to pay great attention to the issue of PCDDs/Fs elimination from all outputs and sources and that means not only emissions to air, but also to waste residues and waste waters. Unfortunately only minor attention has been paid to this issue by investors and state authorities so far.

1.4. Occurrence in respective components of the environment and human population

Total emissions of dioxins and furans to the environment(g I-TEQ) **in the SR in 2001.**
Surveying of persistent organic substances in the SR, Technical report No. 2, section 2)

Category		Air	Water	Soil	Products	Waste
1	Waste incineration	20,274	0	0	0	58,205
2	Production of ferrous and non ferrous metals	42,575	3E-07	0	0	19,177
3	Production of energy and heat, Thermal processes	5,244	0	0	0	0
4	Production of mineral products	0,61	0	0	0	0,01
5	Transport	0,524	0	0	0	0
6	Uncontrolled combustion processes	0,466	0	0,373	0	0
7	Production and use of chemicals and chemical Articles	1,9E-05	0,002	0	0,781	344,17
8	Miscellaneous	0,062	0	0	0	0,02
9	Landfilling of waste	0	0	0	0	0
1-9	Total	69,8	0,002	0,373	0,781	421,577

Emissions to the air

Measurements of dioxin emissions have not been required by legislators. This is why only rarely have measurements from incinerators been taken.. According to the Department for Preventive and Clinical Medicine, waste incinerators were essential sources of dioxins emitted to air (60%). Adapting our legislation to EÚ norms has started to require emission limits, including those for dioxins that will be valid for all facilities incinerating waste from Jan 1, 2007. Some incinerators, mainly sanitary facilities, have subsequently been phased out for economic reasons. Some of them closed down or are going through reconstruction. Nowadays (2006), 36 waste incinerators are in operation in the SR - 2 municipal waste incinerators, 15 incinerators for industrial waste, 15 hospital waste incinerators (12 of them have temporary permits for 1 year) and 4 facilities for mixed combustion (cement plants). About half of these incinerators have equipment that does not absorb dioxin emissions to the degree required by SR and EÚ legislative and are constantly emitting dioxins in increasing amounts.

According to an inventory of POPs in the SR (Technical report 2, sec. 2 from 2003) in 2001 incinerators acted as a small, but still significant source of dioxins – 29%.

Emission limits set forth by the EÚ have to be met by all facilities incinerating waste in the SR beginning in 2007 at 0,1 ng m3 TEQ. This value has been set using data from systematic monitoring of PCDDs concentrations at exposed places in the surroundings of incinerators. A study on dispersion showed that when an incinerator emitted 2 ng TEQ/m^3 PCDD (and more) dioxins over the wider incinerator's surroundings there was contamination of cow and mothers milk, fish and sheep fats, field vestures, water plankton, in some places this contamination was significant. These effects have not been observed when emissions were under 0,1 ng TEQ/m^3. [4]

It is necessary to realise that only meeting the emission limit of 0,1 ng TEQ/m^3 does not necessarily mean a significant reduction of PCDDs/Fs. What is also important is the total amount of emitted smoke/gases and further treatment of ashes and waste water where dioxin is found. Another problem which

has appeared recently is the method employed in monitoring dioxin emissions. If monitoring was undertaken for longer periods it seems that measured dioxin emissions would probably be higher. (R. De Fré, M. Wevers: Undervaluation of dioxin emission degree, ORGANOHALOGEN COMPOUNDS, volume 36, 1998). Standard emission measurements monitored over a 6 hour period according to norm EN 1948, found emission concentrations of 0,25 ng TEQ/Nm^3, whereas the average value coming from two-week measurements done in the same season varied from 8,2 to 12,9 ng TEQ/Nm^3. This data document shows that standard measurements undervalued average emissions by 30 – 50-fold.

The current greatest source of dioxins emitted to air – the metallurgical industry, does not measure PCDDs/Fs emissions to air, these sources have been assessed using theoretical calculations.. That is why it will be necessary to focus just on this sector and processes of chlorine chemistry in relation to PCDDs/Fs.

Dioxin emissions from a municipal waste incinerator in Košice in 1994 [24]

Report from measurement and evaluation of mass concentration of PCDD, PCDF and PCB emissions coming from waste incinerator CZO s.r.o. in Košice (1994)	
2 samples of stack emissions coming from Košice waste incinerator were measured Dioxin concentrations as I-TEQ :	sample 1 **6,95 ng TEQ/m3,**
	sample 2 **8,07 ng TEQ/m3.**

Dioxin emissions from a municipal waste incinerator in Košice 12 / 2004 (after reconstruction)

Widely published outcomes of PCDDs/Fs measurements coming from incinerator TKO in surroundings of Košice from Dec 12, 2004 (an operator of incinerator is Kosit a.s.)	**0,014 ng TEQ/m3**

Dioxin emissions from Slovalco and Cementáreň Ladce
(National reference centre for dioxins and similar compounds, SZÚ, 2002)

Slovalco Žiar nad Hronom	under 0,1 ng TEQ/m3
Cementáreň Ladce	

"Inventory on POPs in the SR", report 2 sec. 2,0 from 2003 states that the total amount of dioxins emitted to the atmosphere was 69,8 g (I-TEQ). This value has been estimated using theoretical calculations.

Dioxins in outdoor air in the Slovak Republic

Total amount of emissions emitted to the atmosphere in the SR between 2000 and 2004 (SHMÚ)

	2000	**2001**	**2002**	**2003**	**2004**
PCDD/F (g I-TEQ)	89.9	86.9	91.3	70.5	66.7

These data were achieved from an inventory that is made yearly under requirements of "Convention on Long-range Transboundary POP Air Pollution protocol". In 2005 they were recalculated according to new methodology which is mainly based on the Standardized Toolkit for Identification and Quantification of Dioxin and Furan Releases" (UNEP Chemicals, 2005).

The samples were taken by the Department for Preventive and Clinical Medicine, Bratislava, during a period of 1996/1997 as a part of the PHARE project. The results showed that amounts of PCDDs/Fs have increased in the air during the winter heating season. If the source was situated in the area surrounding one of the possible industrial dioxin sources which produces dioxins indifferently to yearly seasons, the effect of winter season did not appear. The Department for Preventive and Clinical Medicine states that observed seasonal concentration of PCDDs/PCDFs (also I-TEQ) could mean that increase of PCDFs emissions is connected to heat production, and PCDDs emissions to seasonally stable emissions arising in industrial processes and transport. [25]

Measured values were mainly around the **indicative value 0,1 pg.m^{-3} WHO for urban air**, which means that they do not indicate more serious problems than in the rest of Europe. [25] WHO states dioxin concentrations in the **outdoor air at the level of O,3 pg I-TE/m^3 or higher** indicate local sources of emissions which is necessary to identify and limit. [3] Considering the acceptable daily limit recommended by WHO and USA-EPA, **the emission limit should be set between 20 – 50 fg/m^3** according to several scientific sources. [e.g.. 11]

In the Czech Republic **a recommended value for outdoor air is 20 fg I-TEQ.m^{-3}** (DPCM Bratislava, MoE ČR, Petrlík 2001).

All measured concentrations of PCDDs/Fs in the outdoor air in the SR were higher than the recommended value in the Czech Republic, **20 fg I-TEQ.m^{-3}**. Some of them were also higher than an indicative value of WHO which is **0,1 pg.m^{-3}** (those measurements were distinguished by grey colour) and in a few cases they were higher than **O,3 pg I-TEQ/m^3** (distinguished by grey and underlined). Indicative values of WHO can be understood only as informative data suitable for assessment of contamination degree or for finding of contamination source.

Dioxins and furans in outdoor air in the SR [25]

Concentrations I-TEQ v pg.m^{-3} (calculated from 2,3,7,8-substituated PCDDs and PCDFs) and concentration (pg.m^{-3}) of total amount of **PCDDs and PCDFs** (printed in cursive) measured **in samples of outdoor air** taken from 15 different places over a period lasting from October 1996 till July 1997.

Take off place	1 BA I	2 BA II	3 BA III	4 BA IV	5 BA V	6 Topoľníky	12 Žilina	13 Ružomberok	14 Krompachy	15 Starina	16 KE I	17 KE II	18 KE III	19 KE IV	20 KE V
Oct 96	0,11 *6,30*	0,06 *2,60*	0,09 *4,80*	0,10 *4,53*	0,11 *6,49*	0,08 *3,92*	0,06 *1,67*	0,18 *7,06*	0,06 *1,47*	0,05 *0,89*	0,09 *3,71*	0,20 *11,20*	0,09 *3,07*	0,06 *1,48*	**0,31** *10,0*
Nov. 96	0,10 *5,28*	0,09 *3,93*	0,03 *1,90*	**0,44** *51,98*	0,08 *4,46*		0,10 *4,02*	0,10 *5,26*	0,07 *3,18*		0,23 *10,78*	0,17 *7,90*	0,19 *5,97*	0,06 *2,67*	0,17 *6,29*
Jan. 97	0,20 *14,02*	0,15 *9,46*	0,16 *8,56*	0,13 *9,74*	**0,30** *18,68*	0,22 *6,21*	**0,37** *17,95*	**0,34** *18,25*	0,08 *2,70*	0,07 *1,90*	**0,69** *29,46*	0,24 *13,22*	**0,44** *18,32*	0,19 *8,57*	0,11 *4,51*
Feb. 97	0,11 *5,21*	0,23 *8,03*	0,20 *8,28*	0,09 *3,13*	0,13 *6,0*		0,06 *1,69*	0,11 *5,03*	0,05 *1,76*		0,09 *3,44*	0,13 *4,60*	0,08 *3,05*	0,04 *1,39*	0,18 *7,42*
Apr. 97	0,04 *1,30*	0,04 *1,61*	0,15 *7,14*	0,04 *1,46*	0,03 *1,16*	0,03 *1,11*	0,11 *2,90*	0,13 *4,86*	0,06 *2,19*	0,03 *0,92*	0,15 *4,30*	0,13 *2,53*	0,05 *1,31*	0,06 *1,31*	0,09 *2,51*
May97	0,07 *2,12*	0,03 *1,0*	0,03 *0,95*	0,04 *1,08*	0,05 *1,76*		0,06 *2,61*	0,08 *2,84*	0,03 *0,91*		0,03 *1,0*	0,16 *4,78*	0,04 *1,19*	0,03 *1,05*	0,06 *1,85*
June 97	0,05 *2,80*	0,04 *2,41*	0,05 *3,93*	0,04 *1,64*	0,05 *1,71*	0,03 *0,92*	0,03 *1,08*	0,15 *8,82*	0,04 *2,26*	0,03 *1,21*	0,06 *2,57*	0,11 *6,82*	0,05 *2,30*	0,04 *1,19*	0,05 *2,29*
July97	0,03 *1,53*	0,03 *1,02*	0,12 *3,51*	0,04 *1,73*	0,04 *1,19*		0,05 *1,56*	0,08 *3,89*	0,04 *1,09*		0,05 *1,49*	0,05 *1,42*	0,03 *1,03*	0,03 *0,88*	0,15 *4,98*
Geom. Average	0,08 *3,61*	0,06 *2,73*	0,08 *3,96*	0,08 *3,66*	0,08 *3,28*	0,06 *2,23*	0,08 *2,67*	0,13 *5,97*	0,05 *1,80*	0,04 *1,17*	0,11 *4,01*	0,14 *5,31*	0,08 *2,80*	0,05 *1,69*	0,12 *4,67*

Take off place 1 - Bratislava I – Kamenné nám., 8 odb.
Take off place 2 - Bratislava II – Trnavské mýto, 8 odb.
Take off place 3 - Bratislava III – Turbínová ul., 8 odb.
Take off place 4 - Bratislava IV – Starohájska ul., 8 odb.
Take off place 5 - Bratislava V – Hviezdna ul., 8 odb.
Take off place 6 – Topoľníky – 4 odb. (agricultural area)
Take off place 12 – Ružomberok – 8 odb. (cellulose and paper production)
Take off place 13 – Krompachy – 8 odb. (copper productuion)
Take off place 14 - Strážske – 8 odb. (PCB production in past)
Take off place 15 – Starina – 4 odb. (water basin – back area)
Take off place 16 – Košice I – Štúrova ul.– 8 odb.
Take off plcce 17 - Košice II – Galaktická ul. – 8 odb.
Take off place 18 – Košice III – Strojárska ul. – 8 odb.
Take off place 19 – Košice IV – Ďumbierska ul. – 8 odb.
Take off place 20 – Košice V – Veľká Ida – 8 odb.

Emission limit values or reference values in some states.[11]

State	**Emission limit (fg I-TE/m^3)** (average value for a year)
Italy	**40**
USA – EPA	**40**
Pennsylvania	**30**

Massachusetts	**45**

Recommended limit values for polluting substances in ČR [45]

Polluting substance	Short time concentration (μg/m3)	Short time concentration (NPK)	Average year concentration (NPK-P) (μg/m3)	Average year concentration RBC
2378 TCDD	**-**	**20 fg/m3**	**-**	**42 fg/m3**

Dioxins in solid waste in the SR

No authorities in the SR monitor POPs concentration in solid waste. That is why we present only an out-of-date table (Department for Preventive and Clinical Medicine) with theoretical calculations of PCDDs/Fs concentration in solid waste arising in waste incinerators.

Several sources [27,28] suggest that the more effective absorption of dioxins by filtrating equipment results in their increased concentrations in solid waste – ashes. Absorption of PCDDs/Fs in a filtrating equipment does not mean their destruction – they become a part of waste fly ash that has to be disposed by landfilling at dangerous waste landfills.

As the table of total dioxin emissions in the SR shows, emissions to air form a smaller part of the total amounts of emissions that are produced by burning materials containing chlorine (69,8 g of emissions going to the air and 421 g of emissions remaining in solid waste). One of the studies held in 11 big municipal waste incinerators in Germany showed that output smokes/gases contained only 12% of all emitted persistent organic pollutants, the other 88% was transported into the environment via other residues remaining as a result of burning process.[34]

Incinerators change waste into solid remains with a considerably lower volume. But it is not true for the burning of substances containing chlorine, e.g. waste containing organic persistent pollutants. When waste rich in chlorine is burnt any arising hydrogen chlorine has to be neutralised, a great amount of salt is formed then which can contain dioxins and other products of incomplete burning and unburned dangerous chemicals. For example when burning waste coming from the production of organochlorine pesticides 2,4-D and 2,4,5-T one of American hazardous waste incinerators produced dioxin contaminated ashes and salts in a volume 80% bigger than that of the burned material. [33]

Toxic metals, dioxins and PCBs contained in ashes have a tendency to concentrate almost exclusively in fly ash. The small dimension of ash particles increases their potential mobility in the environment and transport by air and water passages. For the most part ashes are particles of dimensions smaller than 1 μm and that is why they can be breathed in very easily. [32]

As well as prevention and preferences towards cleaner alternatives. It is necessary to apply an integrated programme of ashes testing, tests in working environment control and tests of environmental monitoring and monitoring of placing contaminated ashes in specialised landfills.

Even after application of all that, it is not possible to expect that the practice of landfilling practice will always prevent an uncontrolled release of these persistent organic pollutants into the environment as Acharya and others noted(1991): *PCDDs/PCDFs (dioxins and furans) absorbed and removed from smoke are transported into other remains that are subsequently introduced into the environment again.* [35] For example scientists from Japan found that dioxins contained in deposited ashes that came from municipal waste incinerators interfuse into water basins. [36]

The US Environmental Preservation Agency (US EPA) released a report[39] that confirms that dioxin concentrations in dust from cement plants burning hazardous wastes are 100 times higher than concentrations in dust coming from furnaces burning conventional fuel. Dioxins were also found in solid residues produced in furnaces burning hazardous waste and conventional fuel at the same time.[40]

Dioxins (PCDDs/Fs) contained in solid remains coming from burning of solid municipal waste in former ČSFR– facilities situated in Slovakia -1992 [2]
9 – 87 ng I-TEQ.g^{-1}

Dioxins and PCBs in slag and ashes from incinerators
(quoted from different international sources) [26]

	Slag	**Ashes**	**Reference**
Dioxins [1)]	**12 – 72 ng/kg** **19-30 ng/kg**	**810 - 1,800 ng/kg** **191 – 1820 ng/kg**	**Environmental Agency 1997** [2)], **ETSU** [3)]
PCBs	**<1 - 8,9 ug/kg**	**<1 – 23 ug/kg**	**ETSU**

The reasons for the wide span of values is that there different sources of waste, different conditions of burning, different filtrating and cleaning equipment and heterogenic ashes.

1)An incinerator incinerating 100.000 ton a year produces 0,35 - 2,1 g TEQ of dioxins in slag per year, and 3,1 - 7 g TEQ in the ashes.[Reference is lower than the one from Environmental Agency England...].

2) Environment Agency (1997). Report on emitting of dioxins to soil and water in Great Britain. Report from AEA Technology plc. Bristol, Environment Agency.

3) ETSU (1996). Properties and use of remains coming from solid municipal waste burning. B/RR/00368/REP. ETSU/DTI. This report thoroughly defines ashes fractions and types of incinerators to be worthy as a consultation material in a case of requiring further information.

Dioxins in the human population in the SR

The following table and graph come from an exposure study conducted by WHO. The second and third round was held with the presence of the SR. It focused on the monitoring of possible health risks for the human population (mainly new born babies) arising from exposure to PCBs, dioxins and furans via intake of mothers milk for a purpose of prevention and control of exposure from this source.

PCDD,PCDF and WHO PCB content in the compound samples of mother's milk from the chosen parts of Slovakia – comparison of the contents found in the 2nd. and 3rd. round of the exposure study

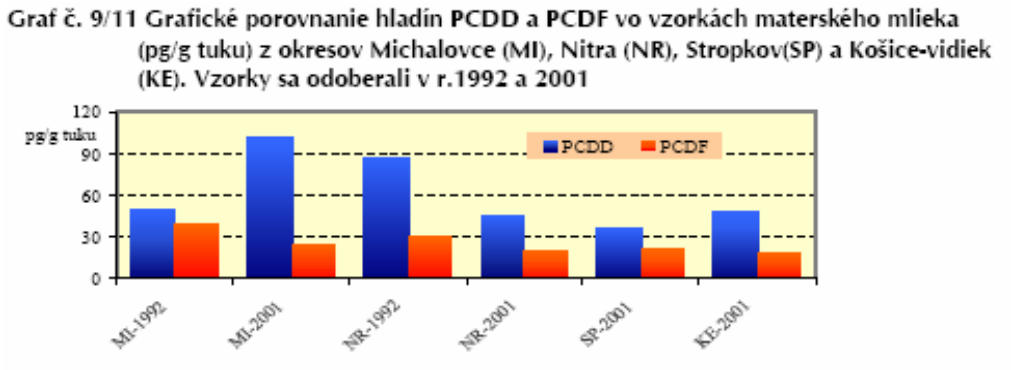
Compound	*Michalovce*		*Nitra*		*Stropkov*	*KE - countryside*
	2.round	*3.round*	*2.round*	*3.round*	*2.round*	*3.round*
*A**	*10*	*10*	*10*	*10*	*10*	*10*
PCDD (pg/g of the lipid)	*49,4*	*102,3*	*87*	*45,1*	*36,0*	*48,6*
PCDF (pg/g of the lipid)	*38,7*	*24,7*	*29,8*	*19,9*	*21.0*	*18,7*
PCDD/F (pg WHO – TEQ/g of the lipid)	*15,1***	*9,9(9,0**)*	*12,6***	*8,6(7,5**)*	*9,5(8,4**)*	*7,8(7,0**)*
PCB (77,126,169,105,118)(pg	*13,3*	*6,3*	*6,1*	*4,5*	*5,1*	*4,2*

WHO – TEQ/g of the lipid						
PCB(12non+monoortho-PCB)(pgTEQ/g of lipid)	-	*19,5*	-	*10,7*	*12,7*	*12,5*

**Pooled sample prepared by the pouring of the aliquot volumes of individual samples of the milk*

In the second round, samples from the districts of Michalovce (MI) and Nitra (NR) were taken (1996). In the third round, samples were taken from the districts of Stropkov (SP) and Košice-surroundings (KE). The district Košice – surroundings was chosen especially to monitor the effects of the Košice municipal solid waste incinerator (2002). Samples were taken in an area assumed to be impacted by the incinerator emissions.. By comparing the measured values in Michalovce and Nitra districts in both rounds of the study, we can observe a tendency of a gentle decrease. Authors of the study state that the combustion of solid municipal waste did not affect the results achieved from samples. [Inventory on persistent organic substances in the SR, Technical report no. 2, sec. 2, 2003] It is possible to dispute this when one considers that the majority of women giving a sample did not consume local food and the highest portion of POPs enters the human body through the food chain, not by breathing. Also, it was later discovered there were over the limit values of PCDDs/Fs in chicken eggs. We can agree with authors of the study that there is an absence of data on PCDDs/Fs in the human population in the SR and it is necessary to intensify monitoring of this area, mainly in the surroundings of potential dioxin sources.

Graph: Graphical comparison of the levels of PCDD and PCDF in the mother's milk samples (pg/g of lipid) from the region of Michalovcen (MI), Nitra(NR), Stropkov (SP) and Kosice-countryside. Samples were from the year 1992 and 2001
pg/g tuku = pg/g of lipid



Concentration of PCDDs/Fs, PCBs and OCCs in human fatty tissue in the Slovak and Czech rep.- **results of PCDDs/Fs** (n=7) (Holoubek a kol. 1995a) [48]			
I-TEQ [pg.g^{-1}]			Total I-TEQ [pg.g^{-1}]
PCDD	PCDF	planar PCB	
6.4 – 10.7	**12.0 – 25.2**	**6.0 – 32.6**	**31.3 – 59.4**

Dioxins in water and sediments

Generally POPs pollution in water comes from waste incinerators and industry sources .The operation of incinerators causes the contamination of waters used for quenching etc. Output flows coming

from filtrating equipment for gas cleaning and water used for grates ashes contain persistent toxic substances. These waste waters can be treated in several ways – they can be used at the place of production again, they can undergo further cleaning, or they can be released through drainage to a sewage tank. State authorities in Great Britain refused to grant planning permission for the building a hazardous waste incinerator because it had been planned near a vulnerable source of water (case Doncaster 1991). [26]

Other facilities that can contribute to the problem are waste waters that come from chlorine bleaching or chlorinated compounds from the paper industry. Scientists have assumed for a long time that dioxins can arise when a cellulose is bleached by chlorine. This theory was later confirmed by several studies – e.g. in US EPA in 1995 when dioxin was found in fish caught near cellulose plants in the States of Maine and Wisconsin. Research of one Swedish environmental organisation on dioxins in the environment from 1998 showed that bleaching of cellulose by chlorine is the most significant source of dioxins after waste combustion and processing of metal scrap. [Dr. Ch. Thies, S. Nemetz, T. Kunz": Wie Papier die Umwelt schädigt und was dagegen getan werden kann, special report Greenpeace Deutschland 2/1991] It is necessary to consider that the paper industry has decreased the use of chlorine and chlorinated substances since 1988 and that it is necessary to re-evaluate the proportion the paper industry contributes to dioxin production.

Dioxins in food in the SR

(concentrations that exceeded maximum limits of
EÚ / SR or reference limits are distinguished with yellow)

Average, minimal and maximum concentrations of PCDDs/Fs in analysed samples of food WHO-TEQ (pg/g of fat)				
Commodity	***Average concentration***	***Minimal concentration***	***Maximum concentration***	***Limit of EÚ / SR***
Pork grease (homemade)	0,47	0,37	0,69	1
Pork (shopping network)	0,30	0,22	0,76	1
Beef (shopping network)	1,15	0,41	2,94	3
Poultry (shopping network)	0,51	0,16	1,18	2
Eggs (chicken, shopping network)	0,74	0,39	1,09	3
Eggs (chicken, homemade)	7,84	1,83	30,5	3
Milk (homemade)	1,43	1,31	1,56	3
Butter (shopping network)	0,83	0,59	1,74	3
Butter (homemade)	1,10	0,74	1,62	3
Fish	0,089	0,0082	0,29	4
Cod liver		75		6

[Anton Kočan, Jana Chovancová, Stanislav Jursa: PCDDs, PCDFs, and PCBs similar to dioxins in food of animal origin (Slovakia), Chemosphere 61 (2005) 1305–1311, Slovak Health University, Bratislava]

POPs values (per gram of fat) **measured in samples of chicken eggs taken from villages Valalíky and Kokšov -Bakša situated in the surroundings of a solid municipal waste incinerator, Košice,** eastern Slovakia
(February 2005, monitoring: Friends of the Earth – SPZ)

	Measured value	**Limits**	Level of activity

PCDDs/Fs in WHO-TEQ pg/g	**11,52**	3,0*	2 (proposal)**
PCB in WHO-TEQ pg/g	**4,60**	2,0 (proposal)**	1,5 (proposal)**
Total WHO-TEQ pg/g	**16,12**	5,0 (proposal)**	-
PCB (7oposites) in ng/g	**189,0**	200***	-
HCB in ng/g	**10,70**	20****	-

*Limit set forth by Regulation of European Council No. 2375/2001 lays down this edge value for eggs and egg products

** EÚ Document – Incidence of dioxins, furans and PCBs similar to dioxins in food is dealing with these new limits.SANCO/0072/2004

*** Limit used in Czech rep. under the Act No. 53/2002.

**** Limit of EÚ

POPs values (per 1 gram of fresh egg weight) **measured in samples of chicken eggs taken from villages Valalíky and Kokšov-Bakše situated in the surroundings of a solid municipal waste incinerator, Košice,** eastern Slovakia
(February 2005, monitoring: Friends of the Earth – SPZ)

	Measured value	**Limits**
PCDD/Fs in WHO-TEQ pg/g	**1.41**	1*
PCBs in WHO-TEQ pg/g	**0.56**	-
Total WHO-TEQ pg/g	**1.97**	-
HCB in ng/g	**1.31**	-

*****US Food Safety and Inspection Service [Circular from July 8, 1997] A guidebook for owners and keepers of poultry, stock and egg producers .Washington, DC: US Department for Agriculture 1997. FSIS informs meat, poultry and egg producers that products containing dioxins at the level of 1,0 ppt in I-TEQ or higher, were devaluated.

POPs values (per 1 gram of weight of a fresh egg) **measured in samples of chicken eggs taken from villages Valalíky a Kokšov-Bakše situated in the surroundings of a solid municipal waste incinerator,** eastern Slovakia

(Slovak State Veterinary and Food Administration , March - May 2005, monitoring realised within confirming of preceding measurements carried out by Friends of the Earth - SPZ)

Locality	Measured value
Košická Polianka, Agrokombinát Torysa a.s.,	2,3
Medzev, Pohm and Gobl	0,9
Nová Polhora, Farma N Pol s.r.o.	1,0

Journal of Slovak MoH No.15-23/2002 and Journal of Slovak MoA No.12/2002

Survey on average finds of dioxins and furans in chosen types of basic food

[Inventorisation of POPs in the SR, Technical report No.2, sec. 2, May 2003; research realised by Department for Preventive and Clinical Medicine– National Reference Centre for Dioxins and similar Substances, Slovak Health University, Bratislava]

Year	Data	Measure	Dioxins			Measure	Furans	
2002		Pg WHO-TEQ/g of fat	Beef	Pork	Eggs	Pg WHO-TEQ/g of fat	Beef	Eggs
	Number of samples		*10*	*10*	*14*		*10*	*14*
	Limit		0,2				0,2	
	Minimal find		0,00334	0,00039	0,00714		0,19929	0,0028
	Average find		0,28303	0,05018	0,28757		0,59302	3,1523

	Maximum find		1,43806	0,44054	1,80104		1,41663	30,0331

Authors of measurements from the National Reference Centre for Dioxins SZU (SZÚ-NRC) observed the following: "Results show that amounts of *PCDDs/Fs contained in food coming from the shopping network are comparable to amounts found in other states of EÚ. Higher concentrations were observed in beef and lower in eggs coming from the shopping network. Higher concentrations of PCDDs/Fs in eggs coming from home rearings of Michalovce district indicate contamination remaining from PCBs production in this locality"*.

It is necessary to add that **concentrations of PCDDs/Fs in eggs coming from home rearings measured by (SZÚ-NRC) considerably exceeded the EÚ and SR limit – on average by 2,5-fold and at a maximum level by 10-fold.** Accordingly we cannot simply state that the only cause of pollution is the remaining contamination from the former production in Chemko Strážske. Certain contributions can also have other sources, e.g. illegal home combustion... Also as measurements of PCDDs/Fs from other localities showed (see monitoring of Friends of the Earth - SPZ) high concentrations in food (eggs) that exceed limits by several-fold are not characteristic only for the surroundings of Chemko Strážske. Higher concentrations of PCDDs/Fs in homemade eggs can indicate a local source of contamination (e.g. local waste incinerator) and the subsequent contamination of the food chain.

In February 2005 a civic organisation Friends of the Earth – SPZ carried out a monitoring of POPs in chicken eggs coming from home rearings in the surroundings of the solid waste incinerator Košice which were subsequently provided for analysis to an accredited laboratory Axys Varilab in Czech Republic Monitoring was part of an international project "Keep the promise – eliminate POPs" held throughout the IPEN network with sampling in 17 countries.

Dioxin values found in eggs from Valaliky and Kokšov-Bakša exceeded EÚ norms by almost 4-fold (see the table**). In addition, limits proposed for PCBs** (in WHO-TEQ) were exceeded in samples by at least two-fold. Dioxin values calculated per weight of fresh eggs exceeded even the limits for eggs sold in the USA. The US Food and Drug Administration estimates a 1:10 000 probability of cancer for eggs contaminated at the level of 1 pg/g I-TEQ. Samples taken from the surroundings of Košice solid waste incinerator exceeded this limit.[1]

If we search for potential sources of dioxins in eggs from Kokšov-Bakša and Valaliky, the following possible sources can be considered: municipal waste incinerator Košice, iron-works US Steel Košice, Heating plant Košice, home heating. A dioxin generator is assessed according to so-called trace - a proportion of respective dioxin congeners.

Research has confirmed that a main source of food contamination characterised by the high dioxin concentrations in villages near Košice solid waste incinerator was probably the incinerator.

"Dioxin traces" were discovered close to the municipal waste incinerator not using a dioxin filter (Košice solid waste incinerator did not operate using a technology for smokes/gases absorption until the end of 2004), even then it was not absolutely compatible. A small contribution to dioxin contamination in the localities of Kokšov-Bakša and Valalíky could also have other sources.

- The direction of prevailing winds (northern, north western 45%, southern 20%, and calm 30%) is the direction from the solid waste incinerator. The iron-works, US Steel, on the other hand is not situated in the direction of prevailing winds. The Heating plant Košice is situated rather further from the locality and regarding its fuel and dioxin trace (those found were significantly different from the one produced by the heating plant) and we can therefore exclude this source as being the main one.

[1] It was estimated (using a factor of a possibility for cancer arising 130 (mg/kg/deň) and by the rounding of results according to valid standards) after consuming of 3-4 eggs a week (30 g of an egg a day) contaminated at a level 1 ppt ITEQ[1, 1]

Regarding high levels of dioxin emissions coming from the Košice incinerator (found in measurements made earlier) and the out-dated technology used until 2004. We can assume that the incinerator is an essential source of dioxins and responsible for the contaminated of the villages of Kokšov-Bakša and Valalíky.

POPs values found in eggs coming from the surroundings of Košice incinerator were comparable to the concentrations found in Rheinfelden, Germany in the second half of 1990s. Rheinfelden was contaminated by pentachlorophenol production and out - dated chlorine production.

Subsequently the Slovak State Veterinary and Food Administration (SVFA) carried out dioxin monitoring in samples of chicken eggs (see table). This monitoring was carried out in a rather strange way as it **was not carried out in the surroundings of Košice solid waste incinerator** which appeared to be an essential source of contamination. Moreover, instead of domestic-laid chicken eggs, eggs from commercial rearings were examined. Despite this the State Veterinary and Food Administration states that its examination was in reaction to our findings and that they did not find a violation of EÚ limits. It is incomprehensible why SVFA did its monitoring of PCDDs/Fs in completely different localities and not in those where contamination was found by FoE. Also disturbing is the fact SVFA did not try to help a local community, and their reaction must be considered improper. Instead of solving the problem it appeared rather as an effort to show that food in the SR is healthy and the SVFA is in control of the situation.

In one sample (taken in v Košická Polianka, from Agrokombinát Tory) concentrations at the level of 2,3 pg/g were found exceeding the SR reference limit. Samples from Medzev showed the lowest contradicts the arguments of the Košice incinerator management that the main proportion of over the limit dioxin concentrations found in chicken eggs in the surroundings of the Košice incinerator comes from illegal waste combustion in households. In reality the situation of waste management in villages Kokšov – Bakša and Valalíky is much better than the one in Medzev where household combustion is also done and probably even to a greater extent though the research data did not show it.

Biota.

Samples of freshwater predaceous fish were taken within the project "Contamination of the environment and human population in the area contaminated by polychlorinated biphenyls" conducted by the Department for Preventive and Clinical Medecine, Bratislava between years 1997 and 1999. A total of two samples were taken. Dioxin and furan concentration was analysed in the samples though this seems to be insufficient for total concentration assessment. [SHMÚ, Slovak MoE , SAŽP, ETV: Inventory on persistent organic pollutants in the SR, Technical report 2, sec2,,2003]

Survey on dioxin and furans concentration in biota (mg/kg of the lipid) **in the year 2002**

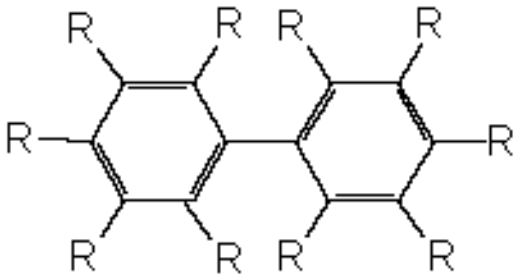
Commodity	Dioxins				Furans			
	Numbers of samples	Min.	Average	Max.	Numbers of samples	Min.	Average	Max.
Freshwater non-carnivorous fishes	2	0,302	0,325	0,348	2	0,259	0,281	0,303

2. Polychlorinated biphenyls (PCBs)

Chemical base :

Polychlorinated biphenyls (PCBs) are a group of 209 isomers. Their formula is $C_{12}H_{10-n}Cl_n$. They have from 1 to 10 chlorine atoms. Accordingly PCBs congeners can be divided into 10 groups, monochlorobiphenyls - decachlorobiphenyls. The variety of substances derived from biphenyls is rather large. Biphenyl is an aromatic hydrocarbon with 2 benzene cores bonded by a single bond. It is a very reactive substance.
[46]

Formula:



Use : [46]

Open systems (leakage of PCBs cannot be prevented, use of these systems leads to environmental contamination)	**Closed systems**
✓ PCB plasticizers ✓ carbonless copy paper ✓ lubricators ✓ components of printer ink ✓ impregnation materials ✓ paint components ✓ glue components ✓ wax components ✓ additives to cements and coatings ✓ materials for lubrication of cast forms ✓ duster materials ✓ sealing liquids ✓ combustion inhibitors ✓ commercial oils ✓	✓ cooling liquids in transformers ✓ dielectric liquids in small and big condensers ✓ fireproof and heat medium anticorrosion hydraulic liquids in mining equipment and vacuum pumps ✓ heat mediums

Commercial names of preparations (commercial preparations PCBs [46])**:**

Commercial name	**Producers**

ALOCLOR, PYROCLOR	Monsanto Industrial Chemicals Co. USA, Great Britain
PYRANOL	Geneva Industries, USA
CLOPHEN, BLACOL	Bayer A.G., FRG
CROPHENE	DSW-VEB, GDR
PHENOCLOR, PYRALENE	Prodelec, France
FENOCLOR, PYRALENE	S.A. Cros, Spain
SOVOL, SOVTOL	USSR
KANECHLOR	Kanegatuchi Chemical Co., Japan
SANTOTERM, AROCLOR	Mitsubishi-Monsanto-Chemical, Japan
FENCLOR APIROLIO, DK	Italy
DELOR, HYDELOR, DELOTHERM	Chemko Strážske, Czechoslovakia

2.1. Properties of PCBs

Physical and chemical properties :

Polychlorinated biphenyls are physically and chemically stable, even at the temperature 300˚C, they are difficult to burn, insoluble in water, are easily absolved in organic solvents and fats. They start to flame at temperatures exceeding 1000˚C. Their nature is strongly lipophilic and this leads to significant bioaccumulation.

PCBs accumulate in fatty tissues where they are persistent.. They are inert to metabolic conversions (only congeners that have less than 4 chlorine atoms undergo a microbial decomposition) . PCBs are chemically stable compounds that undergo slow chemical conversions giving rise to dimers, trimers and oxygen compounds. Substances that arise by their slow conversion are more toxic than PCBs themselves. [46]

Their properties mean that they are found in all biotic and abiotic components of the environment, even in places situated considerably far from places of original production or use. Human population intakes more than 90% of daily PCBs intake through food, mainly animal fats. 47]

2.2. Negative effects to health

PCBs are substances with health damaging effects and 4 congeners out of 209 are highly toxic. Higher PCBs concentrations (10 $mg.m^{-3}$ of air) affect breathing and eyes, and cancer may also be caused. At a concentration of 1 $mg.m^{-3}$ they are absorbed even through the skin and permeate into blood and lymph circulation to enter the spleen and some of the ganglions. These substances change external substances' metabolism in an organism, causing liver disorders and a decrease of immunity. They change the vitamin A and E concentration in the liver, bringing a certain risk of genetic disorder. They can cause blood circulation malfunctions, thyroid structure and function disorders, fatigue, and pregnancy elongation. They also affect reproduction. [46]

2.3. Production and sources of PCBs

PCBs have been formed in a biphenyl catalytic chlorination at a temperature of 150˚C catalysed by iron shavings or ferric chloride carried out in a continual equipment. A mixture of individual congeners is formed (PCBs were never produced as individual congeners) and that is why the product is afterwards refined (thus being cleaned). From the total amount of 209 congener, 120 have been produced in significant amounts.[46]

Commercial production started in 1929, but began to be limited from the end of the 1960s.. Something like 1,2 million of tons of PCBs have been produced world- wide to date.[46]

In the former Czechoslovakia, PCBs were industrially produced from 1959 to 1984 (the production terminated on Dec 31, 1983) in Chemko Strážske under the commercial names Delor, Hydelor and Deloterm. [46] Production and its waste caused a significant contamination of the surrounding environment. From the total amount of 21482 ton produced (+ 1600 ton PCBs waste), 46 % was exported, mainly to eastern Europe, and the rest was used in the former Czechoslovakia.[48] From the beginning of the 1970s OECD states have started to reduce the use of PCBs. Contrary to this (at the same time) the former Czechoslovakia and eastern Europe increased their production. [46]

PCBs exist in open and closed systems. Open systems are applications from which it is not possible to prevent PCBs escaping and their use leads to contamination of the environment. Now there are many releases to the environment, even from close systems, mainly as a result of leakages. Also small condensers are problematic. A considerable risk comes from combustion processes and a major source of release of PCBs is from sediments of big water systems where they come through atmospheric deposition or direct leakage to water. [46]

In PCBs products manufactured in Europe concentrations of tetra-hexa CDFs were around the level 10 ppm. [46] A certain amount of PCDFs (as by-product) can arise when biphenol is being chlorinated. These arise when PCBs products are terminated.

Sources of entry to the environment by PCBs:

- ✓ primary
  - open systems (see table "Use")
  - waste landfills (mainly old, inefficiently secured)
  - waste combustion
  - solid and liquid materials containing PCBs
  - close systems (see table "Use")
- ✓ secondary
  - re-volatilisation from sediments and soil
  - evaporating from applied paints

Main sources of PCB air contamination [46] :

- evaporation of PCBs from older open systems (significant are from applied paints, coverings, but also from mollifiers, heat carrying liquids, wood protection)
- evaporation from operated, disposed or landfilled transformers, condensers, hydraulic systems and equipment containing high amounts of PCBs.
- evaporation from landfills and gases from municipal waste incinerators
- evaporation of PCBs from outer environment (open areas) , as e.g. use of waste oils contaminated with PCBs in transport
- evaporation from waste landfills deposited as dry deposits of industrial waste
- possible evaporation from close systems (the least probable)

A considerable source of environmental contamination is the landfilling of small electronic equipment containing filled condensers where uncontrolled leakage of PCBs is possible.[46]

Global distribution of PCBs [46]	
Total amount of PCBs produced worldwide so far	**1 200 000 ton**
Current pollution in different components of the environment	**374 000 ton**
Amount of degraded and burned PCBs	**43 000 ton**
Reserves (used products and products deposited at the landfills)	**783 000 ton**

Sources of PCBs in the SR

There are many unknown local sources of PCBs in the SR. That is why it is not possible to definitely identify the greatest sources of PCBs in the SR. An inventory of PCB articles and waste was carried out

within the project of Slovak MoE "Contamination of the environment and human population in the areas contaminated with polychlorinated biphenyls" According to results of the project report there is **about 1600 ton of registered waste and about 2700 ton of products based on PCBs** about which no information is available. Nowadays the Slovak MoE is carrying out a minor project on the inventory of PCBs articles still in use. No information is available on the effects of landfills and facilities that have used or are using PCBs (e.g. bitumen mixtures, so called asphalt coaters), incinerators, etc. Despite the possible large releases from these sources, many of them can have only local data (which does not apply to emissions), although they have very negative effects on the environment. For example, releases from the Laborec basin have not been monitored where, according to the estimation of DPCM, great amounts of PCBs (more than 10 ton) have been accumulated in sediments. (DPCM, Bratislava, 2001)

According to one source, old waste landfills are the major relevant source of PCBs environmental pollution in this region. An estimated proportion of PCBs contamination coming from paint application is 5% and from municipal and industrial waste incinerators 9% of total PCBs contamination in the SR.

"Environmental fate" of PCBs

PCBs occur mainly in gaseous state in the atmosphere (87-100%). The rest are PCBs adsorbed on solid particles (floating dust). As a result PCBs can be transported via atmosphere long distances in a relatively short time. (PCBs occurrence has been found in the areas distant from the original source of emissions). [46]

From the atmosphere, PCBs get into soil and water via dry and wet deposition. Dry deposition is important as in the case of PCBs adsorbed on solid particles. Wet deposition is based on washing of solid particles from atmosphere – 99%, the rest is washing from gaseous state. PCBs' reaction with hydroxyl radicals arising in a photochemical reaction with sun rays is a dominant transformation process of PCBs in the gaseous phase. [46]

PCBs' adsorption to sediment or other organic phase is a dominant transport process in a water system. Significant amounts of PCBs have accumulated in water sediments making these sediments a PCBs reservoir from which PCBs will be released for a very long time. [46]

Evaporation from water systems is another significant PCBs transport process occurring in the environment. Transport of PCBs in the system water/atmosphere positively correlates with air temperature. [50]Speed of repeated dissolving of PCBs from sediments into water is higher in summer than in winter when water evaporates faster.. Studies show that transport in a system air/water/sediment leads to increased concentrations of PCBs in surface water layers. [46]

PCBs are usually washed slightly from soil (heavy rains are an exception) [51], because of their low solubility in water and also high extent of adsorption to soil particles. In soils a process of biodegradation is usually very slow, mainly in soils with a high concentration of organic carbon. [46]

Accumulation of PCBs in a terrestrial vegetal ecosystem is realised in several ways:

- through a deposition of atmospheric particles into a leaf system,
- through the adsorption of gaseous phase from the atmosphere by the above ground part of a plant.

The first process has a minor importance. Transport of PCBs from a gaseous phase to vegetation is a dominant source of contamination.[46] however, if we look at water plants we find that bioconcentration factors are even more significant[51]

An adsorption of atmospheric PCBs forms a layer on a water surface with concentrations 500 times higher than concentrations in lower water layers. As a result fish living in this layer contain concentrations higher by a few orders of magnitude. Bioconcentration factors increase with degree of chlorination of PCBs.

A part of PCBs is subject to a photolytic degradation in water. Estimations of its half-time vary from 17 – 310 days for mono- tetra CBs in low water (to 50 cm). Photolytic speed increases with degree of chlorination. There are also to a certain extent anaerobic and aerobic biodegradations taking place in water whose speed is related to degree and type of chlorination. Under aerobic conditions mono-, di- and tri-CB biodegrade more quickly, tetra CB slowly, higher CB do not degrade. [46]

Animals play a significant role in the transport and also the degradation process. PCBs concentrate more in carnivorous animals when we follow the food chain and they act as good accumulators of PCBs in their reserves of fat.

Decrease of bioaccumulation in respective body parts:

tissue>skin>liver>muscles>blood

The half-life of PCBs elimination from living organism is relatively long. [46]

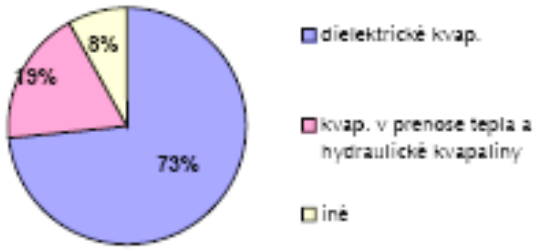
The half-life of elimination of higher chlorinated congeners PCBs is considerably longer, different sources state different values (DPCM, Bratislava, 2001). Some studies on the estimation of PCBs half-time elimination state values are presented in following table:

Half-life of elimination of some congeners and PCB sums (DPCM Bratislava, 2001)

Compound	**Biol. material**	**$t_{1/2}$ [months]**	**Description**	**Literature**
PCB105	blood	6.7	Yu-Cheng	Chen et. al.. 1982
PCB118	blood	9.8	-„-	-„-
PCB138	blood	10.7	Volunteers	Bühler et. al.. 1988
PCB153	blood	11.3	-„-	-„-
PCB180	blood	4.1	-„-	-„-
PCB138	blood	195.6	Prof. exposure	Yakushiji et. al.. 1984
PCB153	blood	330	-„-	-„-
Suma PCB	blood	85.2	-„-	-„-
Aroclor 1248	blood	6-7	?	Steele et. al.. 1986
Aroclor 1260	blood	33-34	?	-„-

Considering physical and chemical properties of PCBs and their similarity to "dioxins" a longer half-life could be expected. For example for 2,3,7,8-TCDD it is 7.1 year – a value very similar to that presented in the table for PCBs sum - 85.2 months (DPCM, Bratislava, 2001)

Distribution of PCBs in the Slovak republic with regard to an area of use
(A. Pilváňová, 2001)



Explanatory notes: 73%: dielectric liquid
19%: liquid in the heat transition and hydraulic liquid
8% : others

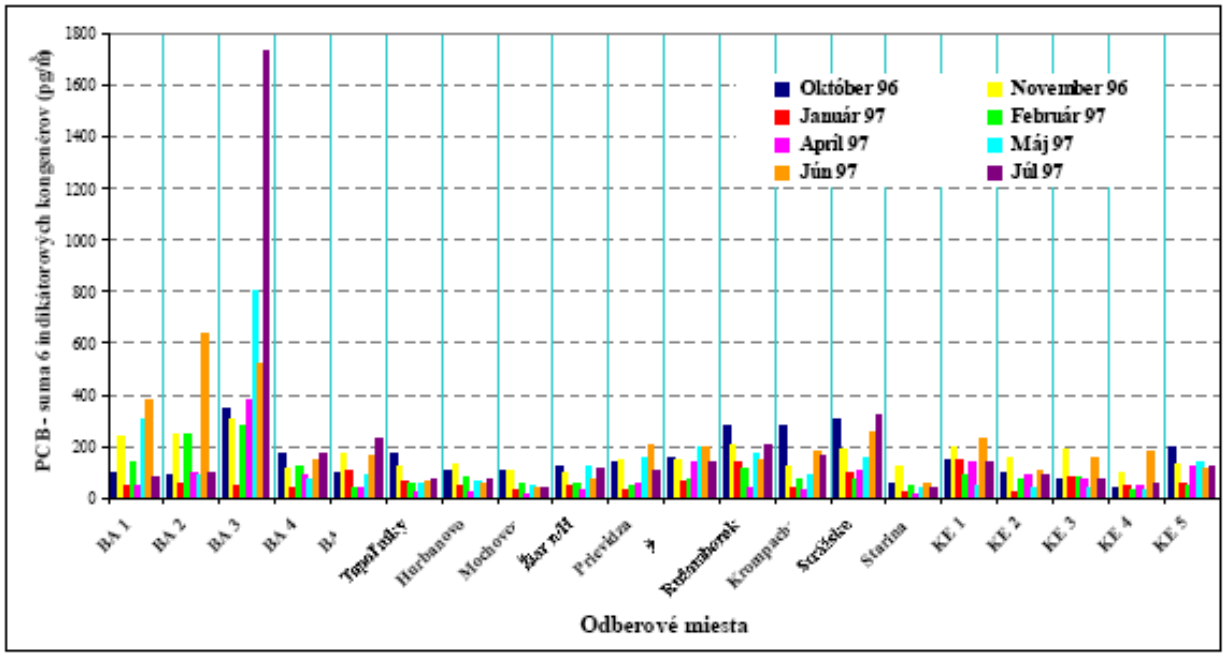
PCB occurrence in respective components of the environment and human population of the SR

PCBs in the outdoor air in the SR

Monitoring of PCBs in the outdoor air carried out within the PHARE project (SZÚ / DPCM, National reference centre for dioxins, Bratislava) showed that concentrations do not considerably differ from other countries. That applies both to urban (industrial) and country areas. Despite the fact that only 8 samples were taken in the course of a year, it was possible to observe that concentrations of lower chlorinated congeners (that dominate in the air) increased with an increase of temperature. The highest concentrations were found in Bratislava at the sample locations on the street Turbínová (BA 3), in the surroundings of the sludge site of a chemical factory. Concentrations of PCBs in the locality of its former production (Strážske) differed from expectations; they were comparable to those found in other towns. But measurements in the third project (Autumn 1997) carried out in Strážske and the neighbouring village Voľa showed **considerably increased concentrations of PCBs** (241 a 217 pg/m3, respectively. The total sum of PCBs congeners 1700 and 1500 pg/m3) **compared to other areas involved in the study.** Concentrations of organochlorine pesticides did not relate to this as a seasonal factor or a sample location. Average concentration in the samples the Stropkov district (11 pg/m3) accounts to minimal values of PCBs concentration found in PHARE project (15 pg/m3 pri VN Starina). DPCM analysed also 4 air samples taken from the **working environment of** a **municipal waste incinerator**. **An average concentration was 43 ng/m3** (minimum 7 ng/m3 and **maximum 86 ng/m3**). An analyses of 1 sample of working air taken at the locality where **power condensers filled with PCBs** were situated (there had been a fire in this compensation locality in the past) **showed that a concentration there was 70 µg/m3 That means about a 700 000 times higher concentration than in the outdoor air.**

Concentrations of dioxins and furans (stated as I-TEQ) **measured in samples of the outdoor air taken from 15 places between 1996 and 1997** within PHARE project

[SZU / DPCM, National reference centre for dioxins and similar substances, Bratislava]
Explanatory notes: PCB – suma 6 indikatorovych kongenerov= PCB – sum of 6 indicatory congeners
Odberove miesta – Places of sampling



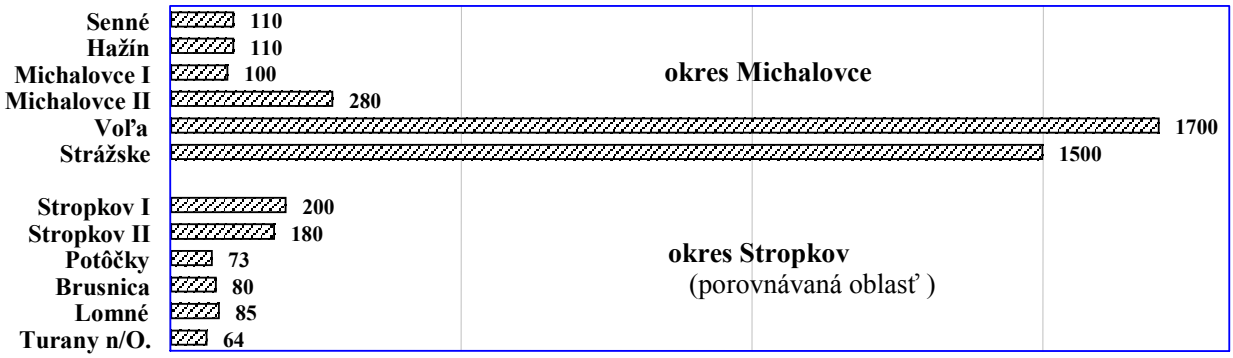
Concentrations of PCBs (sum of congeners 28, 52, 101, 138, 153 and 180)
in samples of the outdoor air taken at chosen localities in Slovakia (pg.m^{-3})
[SZU / DPCM, National reference centre for dioxins and similar substances, Bratislava]

Year of the take off	Number of the samples	Average	Median	Min	Max
1995	15	410	200	40	2420
1996/97	160	136	100	15	1730
1997 district Michalovce	6	82	14	4	241
1997 district Stropkov	6	11	3	2,2	37

Table: Summary concentrations of PCBs in outdoor air [pg.m^{-3}]. Source: Study on Contamination of the environment and human populations in the area contaminated with PCBs [48]

Pollutant	Michalovce district	Stropkov district
Σ PCB	**100 - 1 700**	**64 - 200**

Graph: Concentrations of PCBs in pg.m^{-3} in the outdoor air in samples taken in Michalovce and Stropkov district (Kočan et. al.., 1999b). [48]
Explanatory notes: okres Michalovce = Michalovce district
okres Stropkov (porovnávaná oblasť) – Stropkov district (compared area)



Emissions of PCBs to the air

Calculation of amounts of PCBs emissions arising as a result of waste combustion in the SR in 2000
[SHMI, MoE SR, SEA, ETV Inventory of persistent organic pollutants in the SR, Technical report No. 2, sec 2,2003]

2000	Municipal			Industrial			Hospital			Emissions
Total volume579556,5 ton incinerated waste	Incinerated volume (t)	EF (mg/t)	Emissions (g)	Incinerated volume (t)	EF (mg/t)	Emissions (g)	Incinerated volume (t)	EF (mg/t)	Emissions (g)	Together (kg)
PCB	179 092	5,300	949,2	95904	10,00	959	1967	10,00	19,7	1,93

Total amount of emissions of PCBs in the SR in 1999 (NP of decrease of POPs emissions, SHMI, 2000)

Emissions of PCBs	
Sector Sub sector	PCBs (kg)
Fuel-energetic sector	16,74
System energetic	16,19
Municipal energetic	0,551
Coke production	
Non-industrial combustion	11,27
Heating of non-industrial sector and residential spaces	1,87
Household heating	9,39
Thermal industrial processes	27,94
Industrial energetic	8,86
Production of iron	
Ore agglomeration	8,00
Production of cast-iron	
Other	11,08
Non-thermal industrial processes	0,00
Aluminium production	
Steel production	
Carbon materials	
Wood impregnation	
Road transport	74,85
Other transport	0,941
Waste combustion	2,11
Municipal waste	0,925
Industrial waste	0,778
Sanitary waste	0,410
Total	133,86

Regarding the character of available data e, it is not possible to present an amount of PCB emissions in Slovakia per year. The presented amount of PCBs emissions arising in energy equipment is for 2002 when an inventory was completed. Data on combusted waste for 2001 are just being assessed, so the most recent data available are for 2000. The other data presented are for 1999. If we assume that in 1999 there were operating minimally as much electrical equipment containing PCBs as in 2002, then the total amount of PCB emissions for 1999 was 457,593 kg.

PCBs in water and sediments in the SR

Surface water

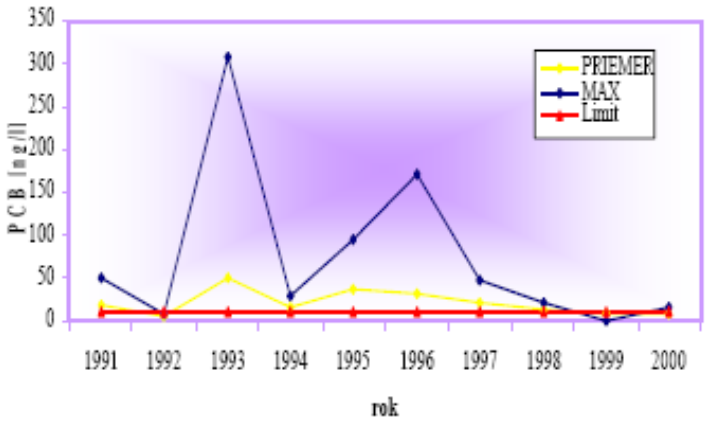
Data on monitoring of PCBs in the water environment were provided by the following organisations - Slovak Hydrometeorological Institute (surface and underground waters), Slovak State Health Institute (surface, drinking, rain, waste and recreation waters), State Veterinary Institute (surface, drinking and feeding waters), Water Research Institute (surface and drinking waters), Department of Hydraulic Engineering Košice (surface, recreative and rain waters), Department of Hydraulic Engineering Bratislava (drinking waters), Water and Sewage works (drinking water), Hydromeliorations Bratislava (surface and feeding waters), Slovak Meteorological Society (surface waters), Department for Preventive and Clinical Medicine, Bratislava (surface waters) and Military Technical Institute Liptovský Mikuláš (feeding waters).

In surface waters PCBs were monitored between 1983-1986 and 1989 – 2001, a total number of analyses carried out was 4398. Approximately **15% - 636 samples exceeded the quality requirement of PCBs for surface waters set forth according to Regulation No. 491/2002 Coll. - 10 ng.l-1. In recreation waters** a total number of 205 analyses for PCB occurrence was realised, and **4 exceeded** the required limit for PCBs **of 50 ng.l-1** (STN 75 7111). In 1987 and between the years 1990-2001 analyses of PCBs were made from samples of **underground waters** in all regions. A total number of analyses carried out was 6474, and 981 samples could not be measured because they exceeded the detection limit. The results of POPs monitoring in waters of the SR are involved in an annex.

There was production of PCBs in the Michalovce district in the past and that is why more attention is paid to these substances in that locality. The highest measured concentrations of PCBs come from the beginning of the 1990s from Laborec basin, municipalities Lastomír and Michalovce, where the highest average concentrations were found in 1993 in the municipality of Lastomír (49,9 ng.l-1) and Michalovce (56,73 ng.l-1) and in 1994 in the municipality Stretava (61,67 ng.l-1). In the following years 1997- 2000 a decrease of measured PCBs concentrations was found in Lastomír. A graph shows the development of average year concentrations and maximum year concentrations between 1991-2000 at the site of Laborec-Lastomír (river km 31) compared to the required limit for surface waters 10 ng.l-1 – a decreasing tendency can be observed. In a water basin Zemplínska Šírava between 1992-1997 maximum concentrations varied from 2 to 44 ng.l-1, but in 2001 concentrations were measured under the detection limit (5 ng.l-1). It is obvious that as a result of PCBs persistence these substances are present in the water environment only minimally, but their increased occurrence can be expected in river sediments in these areas. minimálne, ale [SHMÚ, MoE SR, SAŽP, ETV: Inventorisation on persistent organic pollutants, Technical report 2, sec. 2, 2003]

Development of PCBs occurrence at sampling point Laborec-Lastomír (river km 31) **between 1991 and 2000**

[SHMI, MoE SR, SEA, ETV: Inventorisation of persistent organic pollutants in the SR, Technical report No. 2, sec.2, 2003]



Explanatory notes: priemer=average, rok =year

Recreational waters

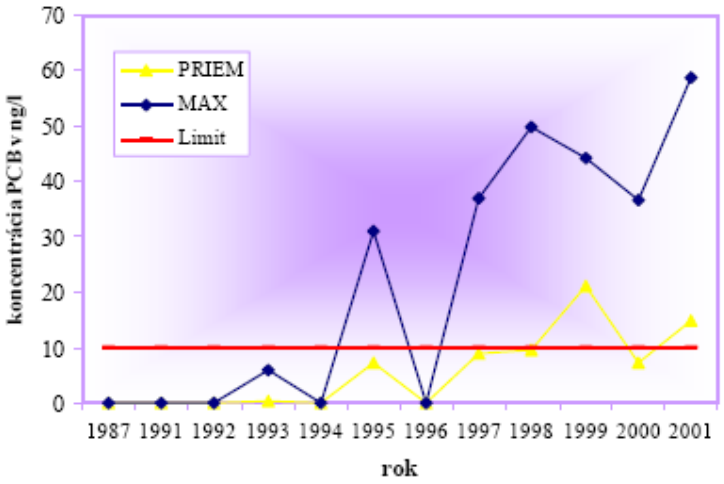
In the recreational waters of the *Bratislava region* (districts of Malacky and Senec, natural grit-sand lakes) PCBs were monitored in 1998 and values found varied from 1,13 to 31,14 ng.l-1. In *Žilina region* recreational waters were monitored in the districts of Źilina (swimming pool), Liptovský Mikuláš (Liptovská Mara dam) and Námestovo (Orava dam) between 1997 – 1999 and in 200. The highest measured value reached 20 ng.l-1. In *Banská Bystrica region* samples were taken from natural lakes in districts of Banská Štiavnica, Lučenec and Žarnovica in 1997 and 1998, and the maximum measured value was 41 ng.l-1. Considering these results we can say that only rare occurrences of PCBs were observed in recreational waters in the chosen localities of these districts during the monitoring period. In the *Košice region,* Zemplínska Šírava basin was the mostly monitored water area between 1993 and 2001 because of the localities' contamination with PCBs, from a total number of 69 samples of surface water, 68 were

undetectable because of concentrations that exceeded relevant limit of detection, **3 measurements exceeded the required PCBs limit for recreational waters (50 ng.l-1) - in 1994 (average year concentration = 187,5 ng.l-1) and in 1998 (average year concentration = 22,3 ng.l-1)**. In following years 1999-2001 no exceeded measurements (50 ng.l-) have been identified in this locality. [SHMI, MoE SR, SEA, ETV: Inventorisation of persistent organic pollutants in the SR, Technical report No. 2, sec. 2, 2003]

Underground waters

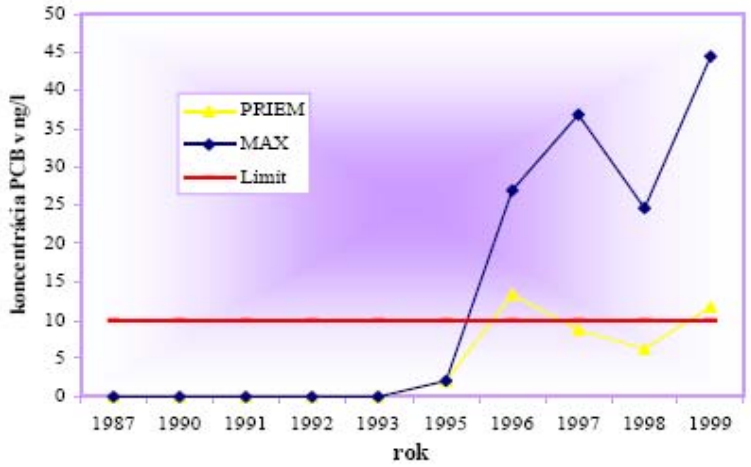
4 % - 230 samples of underground waters from a total number of 6474 samples taken between 1987 and 1990 and 2001 **exceeded the PCBs limit** (10 ng.l-1 according to Regulation No. 491/2002 Coll.). The concentrations reached values up to a level of 58,65 ng.l-1. **The highest number of exceeded measurements** were observed in *Košice and Banská Bystrica regions* where **a curve of PCBs occurrence was increasing** from 1996 and 1997. In Michalovce district increased concentrations (more than10 ng.l-1) were found repeatedly, mainly in villages Lastomír, Michalovce, Krasnovce and Strážske-Pláne. [SHMI, MoE SR, SEA, ETV: Inventorisation of persistent organic pollutants in the SR , Technical report No. 2, sec. 2, 2003]

Trend of PCB occurrence in underground water of the Michalovce district.
[SHMI, MoE SR, SEA, ETV: Inventorisation of persistent organic pollutants in the SR, Technical report No. 2, sec. 2, 2003]



Explanatory notes: priem=average, rok=year

Trend of PCB occurrence in underground water of the Rožňava district
[SHMI, MoE SR, SEA, ETV: Inventorisation of persistent organic pollutants in the SR, Technical report No. 2, sec. 2, 2003]



Explanatory notes: priem=average, rok=year

Drinking waters

There have been 2871 samples of drinking water taken between 1993 and 2001. Twenty-nine samples exceeded the required limit of 10 ng.l-1, most often in the Michalovce district (in municipalities Lastomír and Michalovce). Measured values varied from 0,8 to 86,5 ng.l-1 during a monitoring period (maximum values were identified in 1993). In the following years a decrease of PCBs in measured samples of drinking water from this locality can be observed. Between years 2000-2001 measured values varied from 1,13 to 16,65 ng.l-1. The limit of PCBs detection was exceeded by 702-fold. In the monitored period measured concentrations varied from 0,002 to 270 ng.l-1. [SHMI, MoE SR, SEA, ETV: Inventorisation of persistent organic pollutants in the SR, Technical report No. 2, sec 2, 2003]

Waste waters

In the district of Michalovce a chemical enterprise, Chemko Strážske, is situated. It is the most significant source of industrial pollution. PCBs have been produced in Chemko Strážske in the past. Production of PCBs, but mainly waste arising from this production, has caused considerable contamination of the locality that still exists today.. Nowadays the enterprise has got 2 waste water outlets. One outlet comes from the chemical and biological waste water cleaning plant that carries waste waters to the Ondava stream. The second outlet carries waste waters from two emergency accumulative basins through a waste channel to the river Laborec where mainly waters from once-through water cooling, rain waters, sludge waters from circulating water-towers and a certain amount of sewage waters are concentrated. In a case of emergency state these waste waters are redirected from basins to enterprise waste water cleaning plant.

Between 1993 and 1999 waste waters from enterprise Chemko Strážske were analysed. The waters came from 2 outlets of waste waters from emergency accumulative basins to Laborec and from waste water cleaning plant to Ondava. Concentrations of PCBs measured in waste waters coming from emergency accumulative basins varied from 0,015 to1,22 µg.l-1 and concentrations found in waste waters from waste water cleaning plant from 0,006 to 0,457 µg.l-1.
[SHMI, MoE SR, SEA, ETV: Inventorisation of persistent organic pollutants in the SR , Technical report No. 2, sec. 2, 2003]

PCBs occurrence in waste waters of Chemko Strážske
[SHMI, MoE SR, SEA, ETV: Inventorisation of persistent organic pollutants in the SR , Technical report No. 2, sec. 2, 2003]

terminal of waste water	Water body	year	Year average µg.l^{-1}
Waste water from emergency accumulation basins	Laborec	1993	0,333
		1994	0,380

		1995	0,364
		1996	0,338
		1997	0,146
		1998	0,332
		1999	0,070
Waste water from Clean Centre of waste water of Chemko	Ondava	1994	0,126
		1996	0,144
		1997	0,035
		1998	0,179
		1999	0,019
Waste water from waste canal	Laborec	1993	0,453

Sediments

In 1997-98 samples in a total number of 53 from the surroundings of Chemko Strážske were analysed in DPCM, Bratislava. All results were positive. The maximum value of PCB sum found in waste channel coming from the waste water cleaning plant in Chemko Strážske was 4100 mg/kg. In 2001 SHMI Bratislava analysed PCB contents in river sediments of Slovak rivers (Dunaj, Hron, Ipeľ and Váh) – 21 samples. Positive values were measured in four samples coming from Dunaj and in 1 coming from Váh.

Occurrence of PCBs in soil and sediment samples coming from assumed uncontaminated areas and assumed place of contamination shows they have an ability to evaporate, exist in the atmosphere and return to earth.. Frequency of Delor 106 occurrence comparing to Delor 103 complies with their chemical properties. PCBs with lower chlorine concentration (lower chlorination) are more easily degradable in nature than the more chlorinated PCBs (Delor 106). Self-acting biological degradation of Delor 106 in nature is minimal and very slow. Of course the amount of used substances certainly plays a role. In spite of that, PCBs (as Delor 103 or Delor 106) mean a significant ecological problem whose resolution will take decades because they are still used. e.g. in transformer oils or in different types of oils, paints and liquids deposited at the landfills, even if their production was terminated.

PCBs concentration in sediments in the SR.

Locality	**Analysis by:**	**Year**	**Sum of PCB (mg/kg)**				
			Number of samples	**Min.**	**Max.**	**Average**	**LOD**
Surrounding of Chemko Strazske	ÚPKM Bratislava	1997-98	53	0,002	4100	115	0,002
Dunaj	SHMÚ Bratislava	2001	14	ND	0,0095	0,0018	0,0025
Hron			2	ND			0,0025
Ipeľ			3	ND			0,0025
Váh			2	ND	0,0067	0,0033	0,0025
Vysoké Tatry	ŠGÚDŠ, Spišská Nová Ves	1996	25	ND			0,005
Levice		1996	30	ND	0,953	0,05	0,005
Stredné Považie		1998-2000	85	ND	2,88	0,078	0,005
Galanta		1999-2000	23	ND	0,119	0,039	0,005
Sobrance,		1999-	138	ND	2,76	0,114	0,005

Vranov, Michalovce, Trebišov		2001					
Spiš.-gemer. Rudohorie			73	ND	0,195	0,025	0,005

In 2002 the Slovak Hydrometeorological Institute organised analyses of an extended group of organochlorine pesticides (OCPs) from 81 samples of sediments within a major survey on river contamination in the SR caused by organic pollutants. Within this additional monitoring 10 sediment samples were evaluated to quantify chosen congeners of polychlorinated biphenyls from the area of Zemplínska Šírava. Monitored congeners were PCB 28, 52, 77, 81, 101, 118, 126, 138, 153, 169 and 180. The choice was made according to their effects on human health. Ten PCBs (28, 52, 77, 101, 118, 126, 138, 153, 169, and 180) out of 11 chosen are stated on the list of substances with proven **carcinogenic effects**. This list of carcinogenic substances is part of Proposition 65 which was adopted on May 31, 2002 by the State of California, USA and all substances involved in the list should be labelled as carcinogens. **Maximum acceptable concentration levels in sediments according to the norm of the Netherlands monarchy is 0,004 mg/kg. Congeners stated as the most toxic** (PCB 77, 126 a 169), belonging to planar PCBs **have limits of 0,001 mg/kg**. In a sample of a sediment coming from Strážske stream that acts as an outlet and used for the dilution of waste water coming from Chemko Strážske **the highest concentrations of PCBs congeners were found,** the highest concentration at all was found **for congener PCB180 - 3,7 mg/kg .** In a sample of a sediment from Kyjov stream that carries waste water from the sludge bed Poša, PCBs occurrence was not confirmed**,** although under the outlet of Kyjov stream to Ondava (just under the outlet of waste water channel from the sludge bed Poša), presence of PCBs was again observed in certain concentrations.

An estimated occurrence of polychlorinated biphenyl congeners in rather high concentrations was confirmed from a river sample of Laborec taken under the outlet of Strážske stream (B21S). In a sample of sediment taken from water basin Zemplínska Šírava (B24S) congeners of PCB 28,52,138,163 and 180 were found. PCB congeners were analysed also in river sediments taken from above of waste water outlets exhausting to Laborec (Sediment No. 1) and to Ondava (B23S), in which these substances were not identified..
[SHMI, MoE SR, SEA, ETV: Inventorisation of persistent organic pollutants in the SR, Technical report No. 2, sec.2,2003]

Results of PCBs analyses of the surroundings of Chemko Strážske and Zemplínska Šírava

Number of the congeners/locality	PCB28 mg/kg	PCB52 mg/kg	PCB77 mg/kg	PCB81 mg/kg	PCB101 mg/kg	PCB118 mg/kg	PCB126 mg/kg	PCB138 mg/kg	PCB153 mg/kg	PCB169 mg/kg	PCB180 mg/kg
Sediment n. 1	<0,00096	<0,00097	<0,0018	<0,0009	<0,00122	<0,005	<0,0007	<0,00092	<0,00099	<0,0014	<0,00098
Sediment n. 2	**0,03**	**0,017**	<0,0018	<0,0009	**0,005**	**0,013**	<0,0007	**0,025**	**0,031**	<0,0014	**0,028**
Sediment n. 3	<0,00096	<0,00097	<0,0018	<0,0009	<0,00122	<0,005	<0,0007	<0,00092	<0,00099	<0,0014	<0,00098
B02OVSedPotr	**1,7**	**2,1**	<0,0018	<0,0009	**0,6**	**1,2**	<0,0007	**2,6**	**3,5**	<0,0014	**3.7**
B03OVSedPotr	<0,00096	<0,00097	<0,0018	<0,0009	<0,00122	<0,005	<0,0007	<0,00092	<0,00099	<0,0014	<0,00098
B21S	**0,6**	**0,47**	<0,0018	<0,0009	**0,76**	**0,42**	<0,0007	**0,61**	**1,57**	<0,0014	**1,05**
B22S	**0,04**	**0,006**	<0,001	<0,000	**0,0054**	**0,008**	<0,000	**0,004**	**0,016**	<0,001	**0,007**

			8	9			7			4	
B23S	<0,000 96	<0,000 97	<0,001 8	<0,000 9	<0,001 22	<0,005	<0,000 7	<0,000 92	<0,000 99	<0,001 4	<0,000 98
B24S	**0,004**	**0,003**	<0,001 8	<0,000 9	<0,001 22	<0,005	<0,000 7	**0,003**	**0,003**	<0,001 4	**0,0025**
B25S	**0,006**	**0,0046**	<0,001 8	<0,000 9	<0,001 22	**0,0019**	<0,000 7	**0,008**	**0,006**	<0,001 4	**0,005**

Waste

Considering the data available on the amounts of substances containing PCBs we can assume that the total amounts of such substance that are temporarily stored in various enterprises in the SR reach the level of 4500 – 5000 ton. Approximately 900 ton of the total amount is stored in Chemko, a.s. Strážske. This waste has to be disposed by Dec 31, 2010 according to legal requirements -§ 40a Act No. 223/2001 Coll.. on waste.

Total amount of waste containing PCBs produced in the SR (in t). Source: [SHMI, MoE SR, SEA, ETV: Inventorisation of persistent organic pollutants in the SR, Technical report No. 2, sec.2, 2003]

Region	**Year**				
	1996	**1997**	**1998**	**1999**	**2000**
Bratislavský	2,94	32,378	8,447	8,309	5,867
Trnavský	13,16	6,33	2,5	3,871	3,286
Trenčiansky	26,636	19,594	5,574	3,991	10,611
Nitriansky	19,085	11,016	6,769	4,457	1,197
Žilinský	25,822	12,542	1,482	8,966	6,855
Bansko Bystrický	5,005	23,161	17,6285	7,618	5,24
Prešovský	19,998	10,768	11,54	6,18	6,985
Košický	25,211	25,954	812, 717	852, 668	806,551
Total waste production/year	137,857	141,743	866,6575	896,06	846,592

PCBs in biota of the SR

A survey shows **higher average level of PCBs in musculature of predatory birds** (1,06 mg.kg.-1 of fat) and their accumulation from the natural food chain. PCBs levels in the muscles of a rabbit and a boar vary from 0,16 to 0,32 mg.kg-1 of fat in a relation to a type of consumed vegetal food. **A significant accumulation of PCBs** is observed mainly in **fresh water predatory fish** (2,1 mg.kg-1 of fat) **and fresh water non predatory fish** (3,4 mg.kg-1 of fat). It is necessary to mention the **importance of a locality where game and fish were caught. The majority of finds that exceeded permitted PCBs limits in biota come from eastern Slovakia, mainly from localities surrounding the PCBs production facility,** and may also indicate possible PCBs sources washed from waste landfills situated in the surroundings of the facility to rivers and lakes. Despite the termination of PCBs production in 1984, an increased occurrence of PCBs is observed, mainly in fish coming from some water flows and from Zemplínska Šírava where special veterinary measures were introduced and fish hunt and consumption was banned in 2002. Possible environmental contamination with PCBs can coincidentally occur in other regions, e.g. from contaminated transformer oils or from other sources. Sometimes there are accidental discovery‘s of residuals, e.g. deer or other game caught in a relatively clean areas. But these are rare.

Fish intensively accumulate POPs from the environment they live in (water, sediments). Higher concentrations of polychlorinated biphenyls in samples of non **predatory** fresh water fish were found in *Prešov and Košice region.* In other regions of Slovakia finds reached up to 0.1 mg/kg of fat, except Banská Bystrica and Šaľa district where average founds reached up to 0,4 mg/kg of fat and that are concentrations equivalent to 33% of permitted limit value. **In eastern Slovakia 51 values exceeding regulatory limits have been found.** Samples of fish mainly came from **the** Michalovce district (fish from Zemplínska Šírava),

Poprad and Vranov nad Topľou. Higher levels were also observed in the districts of Stropkov, Trebišov and Sobrance. Samples of fish coming from Zemplínska Šírava showed **extremely high concentrations, reaching up to 23000% of permitted limit** (samples taken by DPCM between 1997 - 1998)).

Higher levels of polychlorinated biphenyls found in samples of hairy game were indicated in Prešov and Košice region. In other regions of Slovakia found values reached up to 0.75 mg/kg of fat, except Martin district **where the average found was 1,4 mg/kg of fat, a value reaching up to 117% of permitted limit**. This high percentage is a result of a find from a locality of Turčianske Jaseno. **There were 9 values exceeding regulatory limits found in a region of eastern Slovakia.** Samples of hairy game, mainly from Michalovce district (mainly samples from surroundings of Chemko Strážske), Humenné – municipality Brekov (3 samples), Poprad (municipality Starý Smokovec) and Kežmarok (municipality Stará Lesná). **Samples of hairy game coming from surroundings of Chemko Strážske showed extremely high concentrations reaching up to 9320% of permitted limit.** (samples taken by Regional Veterinary Service Michalovce in 1999).

Results of hoofed game samples assessment (deer, venison) are similar. Samples exceeding regulatory limits were found mainly in districts of **Michalovce and Vranov nad Topľou where 4 samples exceeding valid limits were found during the monitored period.** Higher average concentrations were also found in **Bytča** district. Maps of PCBs values found in biota (fish, hoofed game) definitely indicate a contamination mostly of eastern Slovakia region. However an incidence of highly persistent substances of POPs type is not excluded where a contamination of the environment and of food sources of game and fish occurred.
[SHMI, MoE SR, SEA, ETV: Inventorisation of persistent organic pollutants in the SR, Technical report No. 2, sec. 2,2003]

Survey on average PCB concentration in biota (mg/kg fat)
(block letter mark level which exceeded limit and/or was at the limit level)

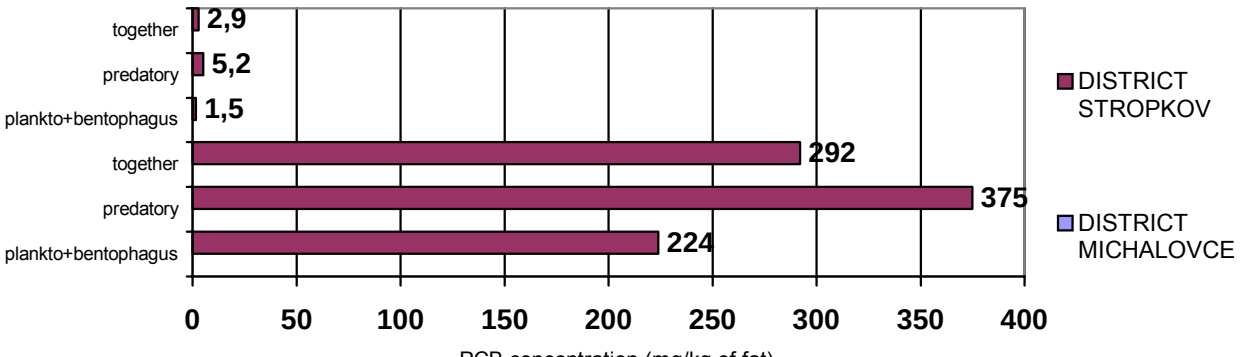
Commodity	**Limit**	**Year**							
		1987	**1988**	**1989**	**1990**	**1991**	**1992**	**1993**	**1994**
wild boar (liver)	1.2	-	-	-	-	-	-	-	-
wild boar (muscle)	1.2-2	ND	0.1400	0.1082	0.0252	0.0702	0.0376	0.0253	-
birds of pray (muscles)	1.2-2	-	-	-	-	-	-	-	**86.500**
birds of pray (eggs)	1.2	-	-	-	-	-	0.0400	-	0.008
birds of pray (entrails)	-	-	-	-	-	-	-	0.8400	-
testaceous animals	1.4	-	-	-	-	-	-	-	-
rabbit, hare (muscles)	1.2-2	0.001	ND	0.0514	0.0868	0.2467	0.0005	-	0.002
hare, rabbit (entrails)	-	-	-	-	0.01	-	-	-	-
game birds (muscles)	1.2-2	-	-	-	0.1	0.02	0.0625	-	0.0018
game birds (entrails)	-	-	-	-	-	-	-	-	-
freshwater carnivorous fishes	0.5-1.4	**7.9240**	-	0.27	-	0.1833	**2.9702**	**33.0145**	0.0058
freshwater noncarnivorous fishes	0.5-1.4	0.2	-	**0.91**	**0.7088**	0.0154	0.2179	0.0278	0.0017
shells	1.4	-	-	-	-	-	-	-	0.0009
deer (muscles)	1.2-2	ND	0.05	0.1445	0.1074	0.0893	0.0653	0.0481	0.0052
deer (lipid)	1.2-2	-	-	-	-	-	-	-	-
deer (entrails)	-	ND	-	-	-	-	-	-	-
furred predatory animals (muscles)	1.2-2	-	0.0017	0.1	0.1774	0.03	0.0037	-	-
furred predatory animals (entrails)	-	-	-	0.005	ND	0.0001	-	0.24	0.0664
Number of samples	-	23	25	92	130	108	202	48	43

Table continued by following years:

Commodity	**Limit**	**Year**	**Togeth**

		1995	1996	1997	1998	1999	2000	2001	er number of sample.
wild boar (liver)	1.2	-	-	-	-	0.06	-	-	2
wild boar (muscle)	1.2-2	-	0.0154	0.1733	0.1603	**4.0503**	0.0117	-	107
birds of pray (muscles)	1.2-2	0.0214	0.1838	0.0261	0.0629	0.0453	-	ND	86
birds of pray (eggs)	1.2	-	-	ND	0.8513	-	-	-	16
birds of pray (entrails)	-	-	ND	ND	-	-	-	-	4
testaceous animals	1.4	0.0015	0.0015	ND	ND	.	ND	-	18
rabbit, hare (muscles)	1.2-2	0.0074	0.0091	0.0333	0.8393	0.4270	0.1819	0.0091	159
hare, rabbit (entrails)	-	ND	0.0015	-	-	-	-	ND	6
game birds (muscles)	1.2-2	0.0043	0.0168	0.0560	0.8177	**5.5676**	**10.8870**	-	131
game birds (entrails)	-	-	0.0082	-	ND	0.0180	-	-	7
freshwater carnivorous fishes	0.5-1.4	0.0698	0.0141	0.1700	0.0121	0.0992	**0.5714**	**11.3541**	351
freshwater noncarnivorous fishes	0.5-1.4	0.0330	0.0126	**23.2337**	0.0452	0.4322	0.4488	**4.3347**	636
shells	1.4	0.0020	-	-	-	-	0.0013	ND	11
deer (muscles)	1.2-2	0.0191	0.0266	0.0431	0.0437	0.0559	0.4712	0.1033	948
deer (lipid)	1.2-2	-	-	-	-	-	-	0.0286	46
deer (entrails)	-	0.0053		0.0690	0.2344	0.1380	0.0380	0.1662	27
furred predatory animals (muscles)	1.2-2	0.7243	0.0323	0.0175	**2.8390**	**26.4563**	**55.1530**	-	125
furred predatory animals (entrails)	-	0.6928	0.1048	0.1707	0.1668	0.1910	-	-	29
Number of samples	-	303	293	306	377	339	250	170	2709

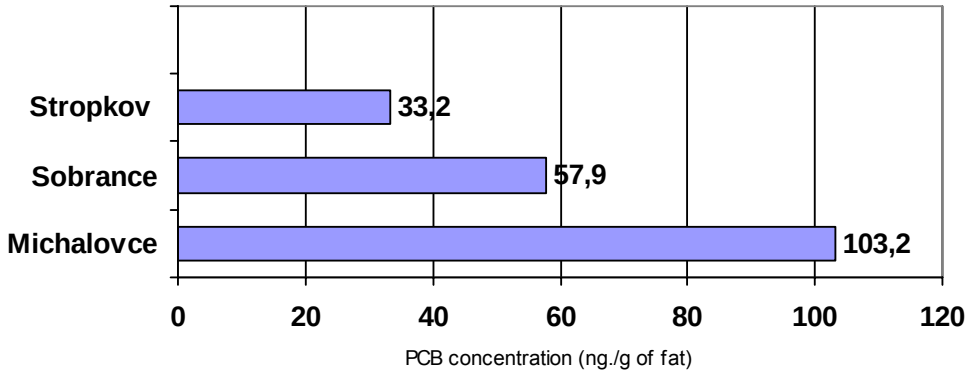
Comparison of average contents of the PCB sum **in** planktophagus, bentophagus and predatory **fish** caught in 1997/98 **in Laborec, Zemplínska Šírava (Michalovce District) and in Ondava and Domaša (Stropkov)**; average contents in all combined samples from the districts Michalovce and Stropkov. [47] [55]



High PCB concentrations were observed within a part of analysed fish. According to the Food Codex, the highest admissible amounts for the fish are 0.2 mg/kg (for congeners 28, 52, 101, 180 – for each individually) and 0.3 mg/kg (for congeners 138 and 153) related per edible portion. The real results of the fish analysis from this measurement are presented in mg/kg of fat. However, we did not evaluate an adiposity of individual samples. After recalculation (ÚPKM) of identified PCB values in the fish fat per edible portion (using average value of fat content in edible portion of fish meat -1 %), for some samples we get high above-limit values. For example, a value of 0.3 mg PCB-153/kg of edible portion means **(considering 1 % content of fat) 30000 ng PCB-153/g of fat. Five combined samples (out of 20) would be above-limit**. **Considering 10 % fat content, this NPM value would mean 3000 ng PCB-153/g of fat and thus, we would get 17 above-limits (out of 20)**.

Within the discovered facts, there have been realized measures in the form of alert (from the side of ÚPKM representatives) to fishermen from the Laborec Basin via local fishermen association to beware of high fish consumption from these rivers, since some of the fish can contain above-limit PCB levels. (ÚPKM Bratislava, 2001)

Graph: **Comparison of average PCB contents** (sum of 9 congeners) **in the samples of wild animals** caught in Michalovce, Sobrance and Stropkov districts. [55] Samples of wild animals came from Michalovce (burdened region), Stropkov (reference region) and Sobrance



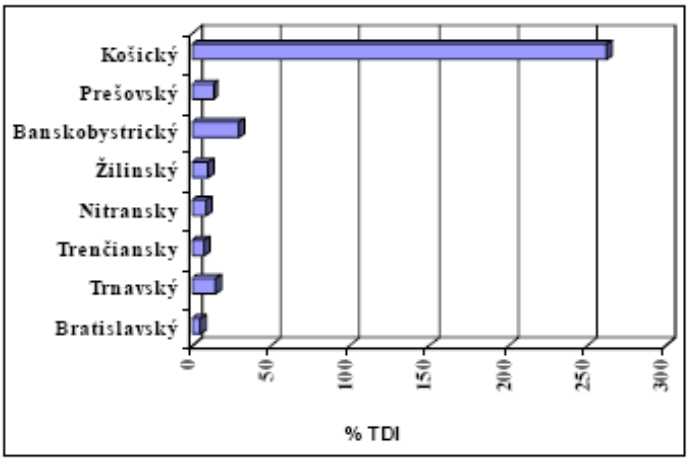
PCB in food in SR

Exposure of the population to PCB congeners (PCB 28, PCB 52, PCB 108, PCB 118, PCB 138, PCB 153, and PCB 180) has been measured since 1984. An intake of the PCB congeners into human organisms was calculated from the data in a database, compared with **TDI** value– tolerable daily intake value, which is unofficially recommended by JECFA FAO/WHO and some of European countries at a level of **0.0004 mg/kg TH/day.**

Considering the unofficial TDI amount, an estimation of population exposure to PCB congeners reaches very high values – 97,9% TDI (1984-1989) in SR), 76,3% TDI (1990-1999) and 26,4% TDI (2000-2002). The highest values of intake were measured in the first years of monitoring. Out of the foods monitored - mainly butter, eggs, milk and meat of freshwater and seawater fish, played a part in the intake. Since 1990, a slight decrease in exposure values was observed, mainly in the last three years at 26.4% TDI, that is, however, still a high value. A high intake calculated in years 1984 to 1989 was affected by a number of investigations on agribusiness companies, poor practices in agricultural through the use of painting materials containing PCB, were found. A similar situation was maintained in the beginning of the 1990`s (1990 to 1993), and have influenced a value of PCB intake during the period of monitoring (1990 to 1999). A high value of PCB intake into the human organism between the years 1994 and 1997 could be explained by a high intake of PCB from domestic eggs, which were taken by ÚPKM within the projects in the Košice

Region (mostly in area of Chemko Strážske). Eliminating these samples from database, the intake of PCBs into the human organism would reach value to 10% of TDI in all regions of the Slovak Republic, except in the Košice Region.

View of PCB intake into the human organism in SR (mg/kg TH/day)



Exposure to PCB congeners represents a health risk to the population, and permanent control, mainly of the commodities of animal origin, support of the elimination of animal fats consumption, and support of improving analytical methods, is needed for a proper health risk evaluation. In the term of regions, the intake of PCB congeners by the population of the Slovak Republic is relatively balanced and planar, except of the Košice Region where the values of PCB intake were several times higher.
[SHMÚ, MŽP SR, SAŽP, ETV: Inventrizácia perzistentných organických látok v SR, Technická správa 2, časť 2, 2003]

Average PCB findings in selected types of food samples (mg/kg of fat, eggs – mg/kg)

Commodity	Year															Total
	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	NS
Beef	0,0529	0,0674	0,2408	0,4825	0,6147	0,1992	0,1313	0,0353	0,0752	0,0318	0,0252	0,0322	0,0242	0,0178	0,0087	6167
Pork	0,0021	0,0385	0,0986	0,0587	0,0633	0,0412	0,0933	0,0159	0,0152	0,0216	0,0147	0,0160		0,0085	0,0025	6057
Eggs	0,0002	0,083	0,0139	0,0152	0,0117	0,0112	0,0140	0,2283	0,0097	-	1,3042	0,0357	0,0129	0,0265	0,0077	1110
Milk	0,0005	0,0462	0,7315	0,9583	0,2133	0,2789	0,2001	0,0542	0,0322	0,0295	0,0140	0,0143	0,0197	0,0178	0,0066	13373
Number of samples	801	834	1581	2441	3638	3697	2603	1652	1058	983	1467	1814	2033	1289	1056	26707

Note: NS = number of samples

Average PCB content in soil (mg/kg)

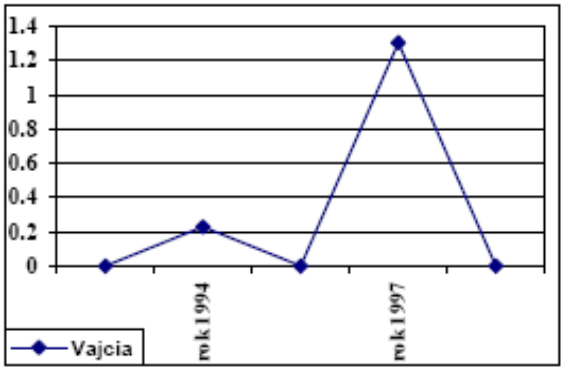
Commodity	Limit	Year										Total number of samples
		1991	1992	1993	1994	1995	1997	1998	1999	2000	2001	
Soil-meadows, grasslands	0,05	0,0053	0,0005	0,0229	-	ND	ND	-	0,00016	0,0001	0,0002	212
Plough land	0,05	0,0116	-	0,0234	-	-	-	ND	0,00015	0,0001	0,0002	1250
Soil-orchards	0,05	0,0015	-	-	-	-	-	-	0,00016	-	-	8
Soil-vineyards	0,05	ND	-	-	-	-	-	-	0,00016	0,0001	-	32
Soil CV	0,05	-	-	**0,66**	0,1	-	**1053**	-	-	-	0,0227	57
Number of samples		372	2	138	8	1	53	10	198	322	455	1559

Note: CV =

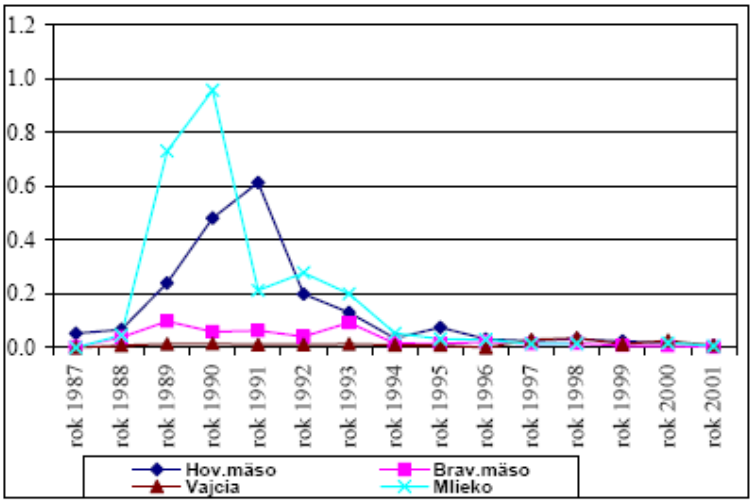
The table "Average PCB findings in selected types of food samples" gives overview about PCBs levels in selected types of food during the period 1987 - 2001.

In the years 1994, 1997 and 1998 within the project, SZU - ÚPKM performed intended sampling of eggs **from domestic poultry-raising in the districts Michalovce and Stropkov. Together, they found 82 above-limit egg samples**.

Time trends of PCB contamination in eggs from domestic poultry-raising (mg/kg)

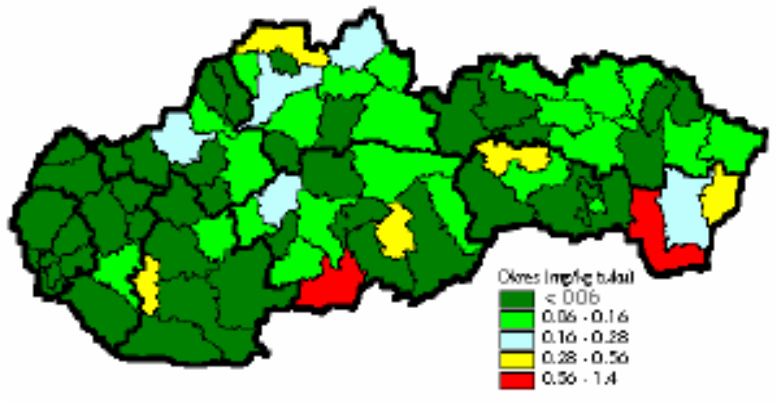


Time trends of PCB contamination in selected types of materials of animal origin in SR (mg/kg of fat)



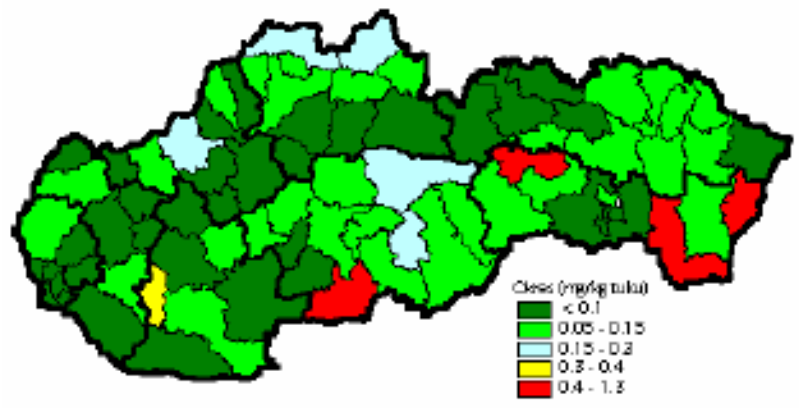
Explanatory notes: Hov.maso= beef Brav.maso=pork vajcia=eggs mlieko=milk

View of PCB content in the beef (mg/kg of fat) **in the years 1987 to 2001**



Regional evaluation of PCB content in beef. PCBs have contaminated the food because of poor practices (contamination from painting materials). Those are mainly the districts of Veľký Krtíš and Trebišov.

View of PCB content in milk (mg/kg of fat) in years 1987 to 2001



In most of the districts, PCB contents in milk ranged at relatively low values. Similarly as in the beef, in the milk there are four districts where feedstuff and materials of animal origin were contaminated from painting materials. These are districts of Veľký Krtíš, Spišská Nová Ves, Trebišov and Sobrance.

Identifying high PCB values, the Regional veterinary and food administration performed measures. Detailed investigation of PCB sources was performed; in some cases, after a commission inspection a decision to liquidate breeding facilities was issued. Further, other measures were taken, for example, the careful removal of wooden skids, adjustment of iron constructions and troughs, reconstruction and hygienic cleaning of objects. This allowed the establishing of new breeding facilities. In the case of finding unsuitable milk samples, a extra regime of milk processing was drafted in the mobile dairy of Mliekoservis Zvolen. If there was contamination of silage pits, an order of feeding by mentioned feedstuff was issued. Often, the demolition of feeding troughs was provided. Measures to quicken PCB eradication were proposed and often, orders for animal transport and slaughter were issued.

Dietary exposure

Observing exposure of the SR population to PCB congeners, it was found that the average exposure amounts from the consumption of daily food in the observed years 1996-2001 were very low (from 0.77% ADI to 12.18% ADI). Higher PCB values in the food were analyzed in year 1997, however, comparing this with the ADI limit this was not exceeded (max. 40.73% ADI). In the last three years, the values have been under 10% of ADI limit (in maximums), what proves only small exposure of the population.

Observing exposure levels of the child population to processed food in 1995-1996, the highest values of observed pollutants were analyzed in the area of Bratislava, where, when calculating burdening, an acceptable daily amount was exceeded by 40%. For observed food types, taken from pre-school facilities, no important differences in PCB levels were measured, in contrast to relatively high maximal values (0.356 mg/kg of overall weight). PCB levels in 30% of non-vegetarian food samples and 15% of vegetarian food samples **reach higher values than the acceptable level , thus, it is necessary to consider a particular risk. Comparing a ratio of acceptable daily intake with the ADI limit, in the both food types these values were exceeded (from 235.8% ADI), pointing to a high exposure by the observed contaminant.**
[SHMÚ, MŽP SR, SAŽP, ETV: Inventarizácia perzistentných organických látok v SR, Technická správa 2, časť 2,2003]

Average PCB contents (sum of 9 congeners) **in observed food commodities of the districts Michalovce** (burdened region) **and Stropkov** (reference region), listed in increasing order. [56]			
Commodity	**Origin**	**District**	**PCB** Content ($ng.g^{-1}_{tuku}$)
Pork	chain store	Michalovce	4.0
Milk	chain store / import	Stropkov	6.6
Pork	chain store	Stropkov	7.5
Sausage	chain store	Michalovce	8.0
Milk	chain store / import	Michalovce	8.3
Frankfurters	chain store	Stropkov	9.0
Ham sausage	chain store	Michalovce	10.3
Beef	chain store	Stropkov	11.3
Frankfurters	chain store	Michalovce	11.5
Eggs	poultry house / producer	Michalovce	15.4
Eggs	chain store	Stropkov	16.2
Lard	private breeding facility	Stropkov	16.5
Butter	private breeding facility	Stropkov	17.0
Eggs	chain store / import	Michalovce	18.3
Milk	private breeding facility	Stropkov	19.3
Milk	dairy facility / producer	Michalovce	20.0
Ham sausage	chain store	Stropkov	21.6
Chickens	chain store	Stropkov	24.5
Sausage	chain store	Stropkov	25.4
Beef	chain store	Michalovce	28.5
Butter	private breeding facility	Michalovce	28.5
Lard	private breeding facility	Michalovce	49.3
Milk	private breeding facility	Michalovce	52.7
Eggs	private breeding facility	Stropkov	128.9
Chickens	chain store + private breeding	Michalovce	374.3
Eggs	private breeding facility	Michalovce	2764.2

PCBs in the soil in SR

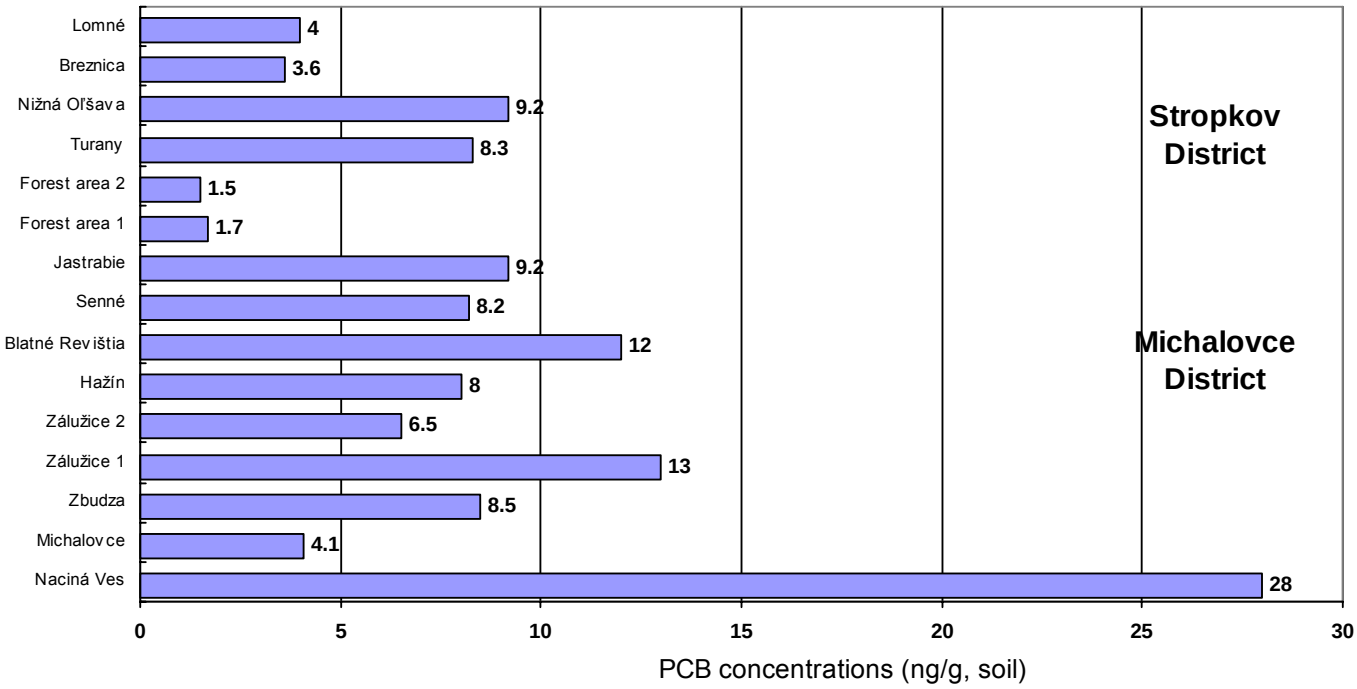
PCBs in agricultural soil were monitored during 1991-2001. Over a period of eleven years, 1021 agricultural soil samples were taken from 73 agribusiness facilities from 20 districts of the Slovak Republic, (representing seven Slovak Republic regions) and analysed. Within investigations of the burdened areas (surroundings of covering plants, Chemko Strážske, etc.), The Preventive And Clinic Medicine Agency has, apart from the agricultural soil, already analysed 538 soil samples. Average PCB values range to 14% of the allowed limit value for all soil types, revealing very low levels of PCB soil contamination in the sampling locations. The average contents of 6 PCB congeners sum for arable part of agricultural soils are around 0.7 to 7.5 µg.kg-1. At the surrounding of plough-lands, average values are from two to three times lower. Since year 1993, average PCB findings in arable soil decreased from 0.023 mg/kg to 0.00016 mg/kg in 2001. In some agricultural soils, where higher PCB levels were found, it is probably spot contamination. During a five-year monitoring cycle, it has been possible to specify a decreasing trend of PCB content in agricultural soils, which are, despite this state, still monitored.

From a regional point of view, **the highest PCB presence was found in contaminated soils in the districts of Stropkov and Michalovce**. **Average values were up to 47341 mg/kg, meaning that with the valid limit value of 0.05 mg/kg for agricultural soils, a remarkable exceedence of that figure.**

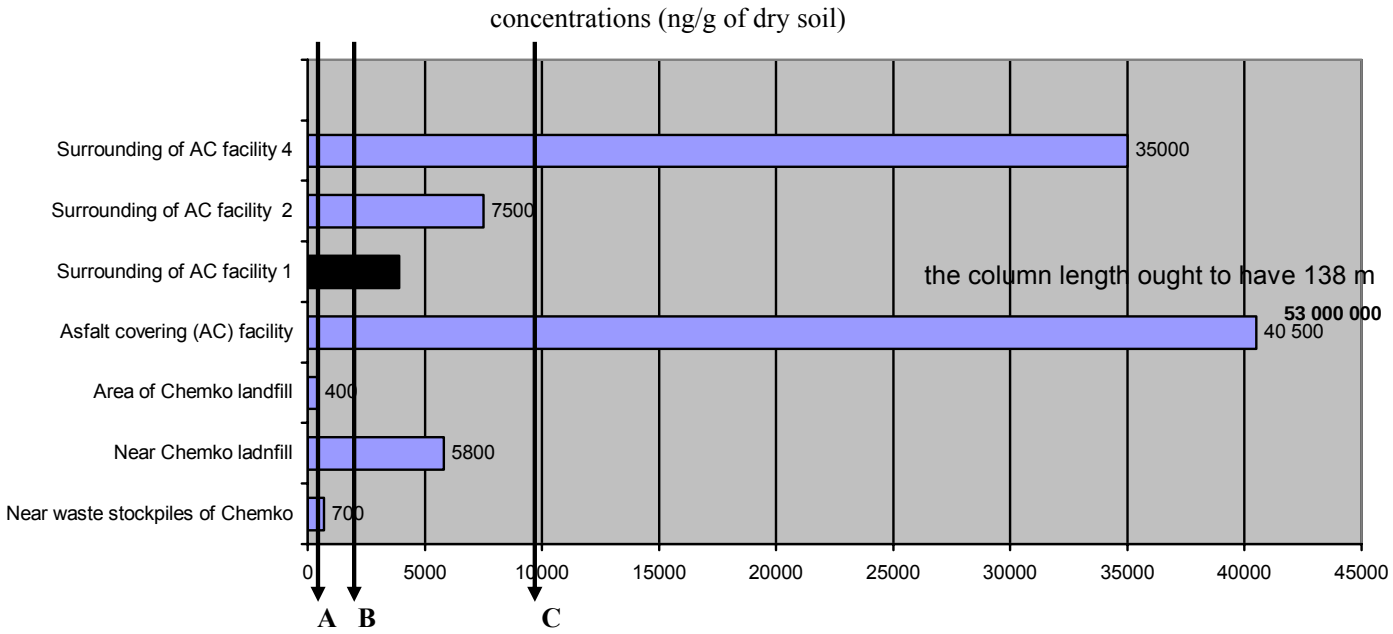
In agricultural soils, the PCB presence was significantly lower; while the highest average values occurred in the districts Poltár and Topoľčany (up to 70% of allowed limit value), in districts Žilina and Žiar

nad Hronom (up to 20% of allowed limit value) and in districts Nitra and Michalovce (up to 10% of allowed limit value).

Average PCB content in soil (mg/kg)" (see page 48§§§§§§§§) gives overview about average levels of PCB measured in soils in Slovakia during the period 1991 - 2001.



PCB levels in the soil samples taken **in some areas of the districts Michalovce and Stropkov.** [48]



PCB levels in the soil samples from surrounding accumulation and landfill of the plant Chemko Strážske and asphalt covering facility. [48]

A - 0.1 mg.kg^{-1} for each indication congener (28,52, 101, 138, 153, 180) – **reference value** means that under this value the soil is not considered contaminated
B - 1 mg.kg^{-1} – indication value – further study and control of the contaminated place are required, when creation, area and concentration could have negative impact on the human health or on the other elements of the environment
C - 10 mg.kg^{-1} – indication value for sanitation – it is inevitable to immediately perform definite, analytical monitoring of the range of destruction of particular place and to decide on the way of revitalisation measures.

PCBs in the human population in SR

In the last years, PCBs in the human population were monitored within various projects. One of them was the project realised in 1996-2000 in cooperation with CDC in Atlanta, USA. Part of the project was a mother's exposure to organochlorine insecticides and PCBs in selected regions of SR (Michalovce, Nové Zámky, Levice and Snina) via setting of concentrations in the placenta.

The highest concentrations of all the 6 analyzed PCB congeners (28, 52, 101, 138, 153 and 180) and 5 out of 6 analyzed organochlorine insecticides (α-HCH, β-HCH, γ-HCH, pp´-DDT, pp´-DDE) **were measured in the region of Michalovce, proving the continued presence of organochlorine compound residues in the environment as a consequence of intensive industrial chemical production in this area.**

Median and maximal concentrations of organochlorine pesticides and PCBs in human placenta taken in industrial and rural areas of Slovakia

Compound	Concentration (ng. g^{-1})			
	Industrial area (N=57)		Countryside (N=63)	
	Median	Max	Median	Max
PCB-28	0,1	4	nd	0,2
PCB52	0,1	0,6	nd	2
PCB-101	0,2	109	nd	8,9
PCB-118	0,1	23,5	nd	0,4
PCB-138	0,2	7,9	nd	6,4
PCB-153	0,2	124,8	0,1	24,4
PCB-180	0,1	1,9	nd	0,1

Notes: N= number of samples, Max. – maximum value, nd - non detect

Another work was the study of WHO, in which the 2nd and 3rd round the Slovak republic participated. It focused on the health risk to the human population (mostly the newborns) from exposure to PCBs, dioxins and furans via maternal milk intake. In the 2nd round the samples from the districts of Michalovce (MI) and Nitra (NR) were taken. In the 3rd round samples from the districts of Stropkov (SP) a Košice-vidiek (KE) were taken. The Košice-vidiek district was chosen especially for investigating the influence of Košice MSW incinerator. Sampling was performed at the expected area of emission grounding from this incinerator. Comparing measured contents in the districts of Michalovce and Nitra, in both rounds of exposure study it was possible to observe a decreasing trend. The authors also say that the samples taken in the area of Košice-vidiek did not show the influence of municipal solid waste (MSW) incineration. [SHMÚ, MŽP SR, SAŽP, ETV: Inventarizácia perzistentných organických látok v SR, Technická správa 2, časť 2,2003]

The last statement is disputable with regards to the fact that the most of the donors did not use local food, and POPs are transferred mainly via the food-chain. and this fact, together with the findings of almost 4-times exceeded PCDD/F content in the eggs from surrounding of Košice MSW incinerator, indicate the opposite. In addition, a recent study from the Slovak Medical University measured 15 PCB congeners in 2047 people from Michalovce, Svidnik, Stropkov (Petrik J, Drobna B, Pavuk M, Jursa S, Wimmerova S, Chovancova J, Serum PCBs and organochlorine pesticides in Slovakia: Age, gender, and residence as determinants of organochlorine concentrations; Chemosphere 2006, 10 March 2006). The results showed high concentrations of PCBs in children (766 ng/g lipid) and adults (3105 ng/g lipid). The authors concluded that „PCBs still remains a source of relatively high environmental exposure for subjects in teh Michalovce district."

Content of 6 indicatory PCB sum in combined samples of maternal milk from selected areas of Slovakia

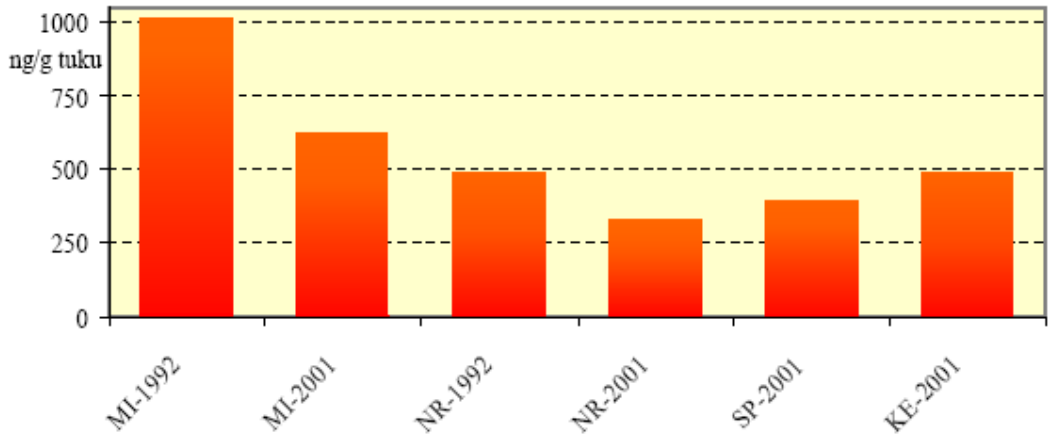
[SHMÚ, MŽP SR, SAŽP, ETV: Inventarizácia perzistentných organických látok v SR, Technická správa 2, časť 2,2003]

Compound	Michalovce		Nitra		Stropkov		Košice - countryside
	2 round	3 round	2 round	3 round	3 round	3 round	3 round
N*	10	10	10	10	10	10	10
Σ indicator PCBs (28,52,101,138,153,180) (ng/g lipid)	1015	621	489	331	397	490	

* pool sample prepared by pouring of aliquot individual samples of mother milk

** calculated with I-TEQ

Graphical comparison of PCB levels (sum of 28, 52, 101, 138, 153 and 180 congeners) **in the samples of maternal milk** (ng/g of fat) **from the districts Michalovce, Nitra, Stropkov and Košice-vidiek** (samples taken in 1992 and 2001).
[SHMÚ, MŽP SR, SAŽP, ETV: Inventarizácia perzistentných organických látok v SR, Technická správa 2, časť 2,2003]



Levels of PCB (9 congeners addition), HCB, γ-HCH, p,p'-DDE and p,p'-DDT in **the samples of the blood serum fat** in different types of the human population in districts Michalovce (MI) and Stropkov (SP) –**PCB results**. [48]

	Content [ng.g^{-1} of blood serum fat]	
	Σ PCB[1]	HCH
Workers exposed to PCB (n=38)	**8 567**	22
Men (n=27)	**10 115**	21
Women (n=11)	**4 767**	22
Fishermen (MI, n=11)	**12 205**	33
Overall population MI (without workers exposed to PCB, n=215)	**4 166**	30
Together with the workers exposed to PCB (n=253)	**4 827**	29
Randomly chosen from the workers exposed to PCB (n=225)	**4 222**	30
Without fishermen (n=204)	**3 753**	30
Overall population SP (n=205)	**1 206**	38
Men MI (n=107); overall population	**4 802**	33
Women MI (n=108); overall popul.	**3 536**	27
Men SP (n=101); overall population	**1 385**	36
Women SP (n=104); overall popul.	**1 033**	37

Addition of congeners: 28, 52, 101, 138, 153, 180, 118, 156 and 170.

PCB levels (sum of congeners 28, 52, 101, 138, 153 and 180) in the samples of human blood serum (calculated per fat ratio) **from the Michalovce District** (contaminated area) **and the districts Stropkov and Svidník** (reference areas). Samples taken in y. 2001.

District	**Number of samples**	**PCB contents (ng.g^{-1} lipid share)**			
		Average	Median	Min.	Max.
Michalovce	378 (162 m, 216w)	2414	1438	308	61425
Svidník + Stropkov	1038 (402m,636w)	692	595	113	13513

Notes: m =men, w=women

PCB monitoring projects in SR, conducted mainly by the Preventive and Clinic Medicine Agency in Bratislava, from which above mentioned values come from (mainly from the project [47]). Problem-solving team summarize the results as follows:

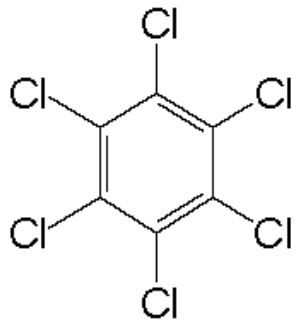
- Yet **20 –times higher PCB concentrations** (around 1.6 µg.m^{-3}) **were measured in outdoor air taken from surrounding** (Strážske, Vôľa) **of former PCB production in Chemko Strážske, or from waste landfills of this plant, in comparison with the reference area** (villages in the Stropkov District).
- Very high PCB content (2-4 mg.g^{-1}) was measured in the samples of Chemko Strážske waste channel sediments coming to Laborec River. **Sediments in Laborec and Zemplínska Šírava contained PCB at the levels 100 – 2000 times higher** (tenths - units of µg.g^{-1}) **than sediment from a controlled area** (Ondava River and water reservoir Domaša). Preliminarily, it can be **estimated that in the contaminated water objects in the Michalovce District several tons of PCB lies**.
- Substantially higher PCB levels were set in the soil samples taken in the surrounding of asphalt covering facilities, from which PCB were released into the soil [probably] by poorly maintained thermo-exchanging systems. In the extreme case, when the sample was taken **directly into the area of plant, already 53 kg of PCB was measured in a ton of the soil. Even in the samples taken out of the area of the asphalt covering plant, 38 g of PCB was found in a ton of the soil (according to the corresponding legislation, indication value for sanitation is 10 g.t^{-1}**). However, in the soils of the Michalovce District a remarkable increase of PCB concentration in comparison with the other soils from the controlled region was not noticed; the **surroundings of one of the Chemko, Inc. landfills have shown several 100 – times increase in PCB content (units g.t^{-1})**.
- Contamination of the elements of the environment was definitely shown by the findings of increased PCB contents in wild animals. In particular, **the fish caught in contaminated waters of Zemplínska Šírava and Laborec contain (in comparison with the fish from Domaša and Ondava) an average 100 – times higher PCB levels** (around300 µg.g^{-1} of fat). PCB exposure reaches above-average levels among frequent consumers of such contaminated fish.
- The analyzed food, bought in the chain store had visibly lower PCB content than the food coming from domestic breeding or those produced for materials, whose origin was in the burdened area. Since the food from the chain store or the materials for its production were imported into the monitored districts, the difference between the compared districts was not noticed. The reverse situation occurred in the case of the food coming from the animals kept for domestic breeding. **On average, approximately 2.8 µg of PCB in a gram of fat was found in the eggs from Michalovce domestic breeding, whereas in those from Stropkov it was "only" 0.13 µg.g$^{-1}_{fat}$. In the milk from domestic breeding from Michalovce was 0.053 µg.g$^{-1}_{fat}$, from Stropkov 0.019 µg.g$^{-1}_{tuk}$. In the milk from Michalovce dairy plant (producer) was 0.020 and in the milk imported to both of the districts around 0.007 µg.g$^{-1}_{fat}$.**
- Higher PCB content in some food types from the burdened district Michalovce has undoubtedly led to increased levels of these pollutants in the population of this district. In the fat isolated from the blood serum samples taken from the general population of the Michalovce District average PCB concentration 4.2 µg.g^{-1} was found, of the Stropkov District 1.2 µg.g^{-1} and the workers professionally exposed to the PCB production in Chemko Strážske 8.6 µg.g^{-1}. **Among the general population, there were 11 fishermen, who consume the fish caught in the contaminated waters of Laborec and Zemplínska Šírava – average PCB level in their fat was 10.1 µg.g^{-1}. <u>It is necessary to highlight that PCB levels and even the levels of HCB and p.p.´-DDE,, not excluding the Stropkov District, are substantially higher than in West-European and North-American countries</u>**.
- There was observed a connection (more remarkably in men) between increased PCB levels and enlarged volumes of thyroid gland and indicators of damaged structure and function of thyroid gland. In the workers of Chemko, affection of thyroid gland was recognised in much larger extend. **Even the**

correlation between PCB content in organism and number of pathological changes related with thyroid gland was noticed. [47]

3. Hexachlorobenzene

Chemical name: Hexachlorobenzene

Formula:



CAS Number: 118-74-1

Some physical properties: summary formula: C_6Cl_6
molecular mass: *284.76* g/mol
boiling point: *326 unit, 322°C sublimes*
state: *solid*
conflagration point: *242°C*
density: *2.044 at 23°C*
steam pressure: *1.45 mPa at 20°C*
solubility in water: *1E-5 g/l at 25°C*
other solubility: *partially soluble in cold alcohol, soluble in benzene, chloroform and ether*
structure: *spike crystals*
colour: *white*
smell: *specific, unpleasant*
impurities: *hexachlorobutadiene if HCB is produced from tar; hepta-and octachlorodibenzofurans and octachlorodibenzo-para-dioxin were recognised in commercial HCB.*

Commercial names of the preparations: Amatin, Anticarie, Bunt-cure, Bunt-no-more, Co-op hexa, Julin's carbon chloride, No bunt, No bunt 40

Use:

It was used as selective fungicide for modification of the seeds of wheat, onion and other plants; used also for disinfection of the soil. Many pesticides contain it as an impurity. It is also an industrial by-product. In industry, it is used for production of fireworks, munitions, synthetic rubber and aluminium. It is created as a by-product during production of industrial chemicals, including carbon tetrachloride, perchloroethylene, trichloroethylene and pentachlorobenzene and as an unwanted by-product of incineration processes (waste incineration), cellulose whitening by chlorine, with similar mechanism of creation as PCDD/F. HCB was in former Czechoslovakia used in agriculture as selective fungicide since 1945 (ÚPKM Bratislava, 2001). A

use of HCB is since 1985 forbidden [54] HCB was produced in form. Czechoslovakia in Spolana Neratovice and its production was finished in year 1968.
[Prof. RNDr. Ivan Holoubek, CSc. et. al..: Persistentní organické polutanty, RECETOX – TOCOEN & Associates Brno, TOCOEN REPORT No. 149; 1999]

3.1. Negative influence to health

Experiments on animals proved mutagenic, teratogenic and carcinogenic properties.
In humans: pathological creation and secretion of porphyrins (blisters and in following pigment scars on the skin), liver enlargement, temperature decrease, loss of appetite, loss of weight, muscle atrophy, change in the hair
IARC and US EPA classified HCB as potential carcinogens for the human (Class 2B) (IARC 1987, US DHHS 1994b). In the reproduction, maternal risk studies, it was found that HCB was hepatotoxic and/or damaged foetus, affected neurological behaviour. Various studies suggested that HCB damages the immune system.

According to the Environmental Health Criteria 195, there is not enough of the toxicokinetic information related to the human. It was proven at experimental animals that HCB is very well absorbed by oral way and just a little by skin. HCB is accumulated in animals and the human in adipose tissues and could be transported to children via placenta and maternal milk. HCB is just partially metabolised, creating pentachlorophenol, tetrachlorohydroquinone and pentachlorothiophenol as the main metabolites in urine. The halftime of elimination for HCB ranges from approximately one month in rats and rabbits to 2-3 years in monkeys. (ÚPKM Bratislava, 2001)

3.2. HCB sources

Except for the application in agricultural preparations, which has been in the SR stopped since 1985, its sources are some of the incineration and industrial processes.
HCB emissions are generated in the same thermo and chemical processes as the emissions of PCDD/F and HCB is created by a similar mechanism. The main sources of HCB emissions can be:
a) waste incineration, including co-incineration,
b) thermal sources of metallurgic processes,
c) use of the fuels with chlorine content in blast furnaces. [68]

Intake of HCB:

Generally, the human population is endangered by HCB mainly by intake from food (around 92%), much less by breathing (7%), drinking (1%) or by skin contact. [48]

3.3. Occurrence in elements of the environment and at the human population of SR

HCB in the air of SR

The highest HCB concentrations (in outdoor air) were noticed in the samples from agricultural regions (Levice – 4.74 ng.m^{-3}, Topoľníky – 4.45 ng.m$^{-3)}$,a busy crossroad (Bratislava – Patrónka - 3.62 ng.m^{-3}) and the surrounding of waste incinerators (Bratislava - 3.6 and 3.57 ng.m^{-3}). This could be caused by HCB use as fungicide (now forbidden), and also by its formation during incineration processes. [48]

HCB concentrations in the samples of free air from selected regions of Slovakia
[SHMÚ, MŽP SR, SAŽP, ETV: Inventarizácia perzistentných organických látok v SR, Technická správa 2, časť 2,2003]

Sampling year	No. of samples	Concentration (pg. m^{-3})			
		Average	Median	Min.	Max.
1995	15	2089	1800	127	4740
1996/97	160	127	95	17	900
1997	12	50	45	21	100

Table: **Summary concentrations <u>in outdoor air</u>** [pg.m^{-3}]. Source: Štúdia zaťaženia životného prostredia a ľudskej populácie v oblasti kontaminovanej PCBv(Ing. A. Kočan CSc. et. al.., 1999) [48]

Pollutant	Michalovce District	Stropkov District
HCB	**21 - 37**	**53 - 100**

Chlorinated pesticides concentrations in outdoor air
– HCB results [pg.m^{-3}] [48]

Locality	Concentrations	References
Slovakia - industrial, urban area (1995/6)	**99 - 3 620**	Kočan et. al..
- agricultural	**4 450 - 4 740**	
- rural, forest area	**147**	

HCB in waters and sediments of SR

From the available data during the period of 1991-2001, 1574 samples of **surface waters** were analysed, out of these 198 were findings (values measured above the detection limit), within which **8 concentration values were measured above required limit in surface waters** 0,05 µg.l-1.

In the **backwaters (lakes and water reservoirs, including thermal waters**) **from the view of recreation use,** during 1998, 2000 and 2001, 410 samples were analysed, out of which 211 were measured above the detection limit, while **13 concentration values, which exceeded the limit set at 0.01µg.l-1** were registered.

In drinking waters, hexachlorobenzene was analysed between 1991-2002 in 3301 samples, while 541 concentration values were measured above detection limit and out of this number **29 exceeded the limit for drinking waters 0.01µg.l-1**. Occurrence of higher hexachlorobenzene concentrations in drinking waters was measured in 2000 and 2001 in the *Košice Region* (Michalovce and Sobrance District), where the values range from 0.5 to 41.74 ng.l-1 and in *the Bratislava Region* (District Bratislava I) with measured values from 0.1 to 15.4 ng.l-1 in 2002.

In underground waters during the period of years, altogether 9841 samples were evaluated, 1051 of them were measured above the detection limit and **3 concentration values exceeded required limit 0.01µg.l-1**. The concentrations measured ranged from 0.001 to 48.6 ng.l-1.
[SHMÚ, MŽP SR, SAŽP, ETV: Inventarizácia perzistentných organických látok v SR, Technická správa 2, časť 2,2003]

Surface waters

Hexachlorobenzene occurrence was noticed in the *Košice Region* (the Michalovce District), in the *Nitra Region* (districts Nitra and Topoľčany) and in the *Trenčín Region* (district Prievidza). In the *Košice Region,* during the period of 1996-2001, 152 surface water samples were analysed, while in 49 samples hexachlorobenzene was measured above corresponding detection limit and 3 concentration values out of this exceeded required limit for hexachlorobenzene (0.05 µg.l-1). The highest concentrations were registered in surface waters in the Michalovce District in 1999 and ranged between the values 0.082-0.118 µg.l-1 (Laborec, WR Zemplínska Šírava). Between 2000 - 2001 in the same locality low concentrations in interval 0.,0003 – 0.04 µg.l-1 were measured. In the *Nitra Region,* the most often occurrence of hexachlorobenzene was registered in surface waters in the districts Nitra and Topoľčany, however, in no sample of surface water the required limit 0.05 µg.l-1 was exceeded. Between 2000-2001 in these districts, 57 samples were analysed, while 48 concentrations measured ranged in interval 0.001-0.04 µg.l-1 (the Topoľčany District, River Nitra, river km 91.1). In the *Trenčín Region* hexachlorobenzene was found in the Prievidza District (village Chalmová, River Nitra, river km 123,8), where it was observed during the period of 1992 – 2000, while increased occurrence was noticed in years 1995 to 1997, when there were measured values in the range from 0.002 to 0.14 µg.l-1. Between 1998 – 2000 in this locality, a decrease in hexachlorobenzene concentration values, which ranged from 0.001 – 0.03 µg.l-1, was noticed. In backwaters (lakes and water reservoirs, including thermal waters), monitored for the aim of recreational use, in the period of 1998, 2000 and 2001, 410 samples were analysed, 211 out of them were measured above the detection limit, while 13 concentration values were noticed as exceeding the set limit 10 ng.l-1. In the *Bratislava Region* between the years 2000-2001 in natural lakes (gravel deposits), concentrations in the range from 0.068-57.97 ng.l-1 were measured. In the *Košice Region* between the period of 2000-2001, hexachlorobenzene occurrence in water reservoir Zemplínska Šírava was noticed with an interval measured from 0.5 to 39.8 ng.l-1 (year 2000, village Vinné).
[SHMÚ, MŽP SR, SAŽP, ETV: Inventarizácia perzistentných organických látok v SR, Technická správa 2, časť 2,2003]

Sediments

Hexachlorobenzene in sediments was evaluated in the surrounding of Chemko Strážske, water reservoir Domaša, Šírava, Laborec and Ondava. The highest concentrations were found in water reservoir Šírava – western part 1100 mg/kg, Zemplínska Šírava – middle part 100 mg/kg, Domaša- Turany nad Ondavou 1100 mg/kg, Domaša –Bžany 2500 mg/kg. In other samples, the contents are under 1 mg/kg.
[SHMÚ, MŽP SR, SAŽP, ETV: Inventarizácia perzistentných organických látok v SR, Technická správa 2, časť 2,2003]

Concentrations of monitored pollutants in the sediments of the Michalovce and Stropkov districts (µg/kg) [47] - samples from the Stropkov District are in cursive

Sample	Sampling place	HCB	Date
S-01	Zemp. Šírava 1 (west. part)	**1.1**	X/97
S-30	Zemp. Šírava 1 (west.part)	**<0.2**	VI/98
S-02	Zemp. Šírava 2 (mid. part)	**0.10**	X/97
S-31	Zemp. Šírava 2 (mid. part)	**<0.7**	VI/98

S-03	Zemp. Šírava 3 (east. part)	**<0.07**	X/97
S-32	Zemp. Šírava 3 (east. part)	**<0.4**	VI/98
S-04	filling channel to Z.Š. (village Zbudza)	**2.0**	X/97
S-16	filling channel to Z.Š. (village Zbudza)	**<0.7**	VI/98
S-15	draining channel from Z.Š.	**<0.3**	X/97
S-22	draining channel from Z.Š.	**<1.1**	VI/98
S-05	Laborec nad Strážskym (village Krivošťany)	**0.31**	X/97
S-17	Laborec nad Strážskym (village Krivošťany)	**<2.7**	VI/98
S-07	Laborec (1) pod Strážskym (village Vôľa)	**<0.5**	X/97
S-24	Laborec (1) pod Strážskym (village Vôľa)	**<0.9**	VI/98
S-25	Laborec (2) pod Strážskym (village Lastomír)	**<0.9**	VI/98
S-08	channel Čierna voda (village Blatné Revištia)	**<0.1**	X/97
S-21	channel Čierna voda (village Blatné Revištia)	**<0.25**	VI/98
S-09	Senné rybníky (village Iňačovce)	**<0.06**	X/97
S-23	Senné rybníky (village Iňačovce)	**<0.4**	VI/98
S-18	waste channel from Chemko sewage tank	**<0.6**	VI/98
S-19	waste channel from Strážske sewage tank	**24**	VI/98
S-06	waste channel from Chemko and Strážske sewage tanks	**120**	X/97
S-20	waste channel from Chemko and Strážske sewage tanks	**72**	VI/98
S-10	*Domaša 1 (village Turany n/Ondavou)*	***1.1***	*X/97*
S-28	*Domaša 1 (village Turany n/Ondavou)*	**<0.3**	*VI/98*
S-11	*Domaša 2 (village Bžany)*	***0.50***	*X/97*
S-29	*Domaša 2 (village Bžany)*	***2.5***	*VI/98*
S-12	*Ondava 1 (near the outfall of Domaša)*	***0.70***	*X/97*
S-27	*Ondava 1 (near the outfall of Domaše)*	**<0.8**	*VI/98*
S-13	*Ondava 2 (village Nižná Olšava)*	**<0.04**	*X/97*
S-26	*Ondava 2 (village Nižná Olšava)*	**<0.2**	*VI/98*

Hexachlorobenzene content in sediments
[SHMÚ, MŽP SR, SAŽP, ETV: Inventarizácia perzistentných organických látok v SR, Technická správa 2, časť 2,2003]

Locality	Analysed by	Year	HCB mg/kg				
			PV	Min.	Max.	Average	LOD
Surrounding of Chemko Strážske	ÚPKM - SZU	1997-98	53	0.001	2500	151	0.001

Soil

Average hexachlorobenzene values ranged up to 2% of permitted limit value for all types of soils; this represents very low level of soil contamination by hexachlorobenzene.

Food

Considering the long-time introduction of hexachlorobenzene (since 1985), the residues of hexachlorobenzene in the food of SR today occur in relatively low concentrations. From 28921 tested foods, analysed in the years 1984-2001, 51% of samples were under the detection boundary. During the whole monitoring period, the fraction of samples above the limit of hexachlorobenzene was 0.3%, representing 73 samples of the overall number of those monitored. Hexachlorobenzene was in milk monitored since 1987. Together were investigated 3062 milk samples from all the Slovak republic regions. The highest values of average findings were registered in 1988 and 1992 (0.055 and 0.059 mg/kg of fat). In the last 8 years all measured values were under the limit boundary 0.01 mg/kg of fat, while in last four years the average values were lower than 0.003 mg/kg of fat. Out of overall number of the milk samples analysed, 39 samples did not meet valid limit values. Out of these above-limit samples 19 were in the years 1986-1989 and 20 in 1991-1996. Since 1997 no invalid samples have been found at the area of the Slovak republic. Per hexachlorobenzene content, the eggs investigated were at a substantially lower rate. Together 1124 egg samples were analyzed. All values were very low. An exception being 1994, when 32 samples were analysed, and 19 did not meet valid hygienic limits, representing 59.4%. Nineteen samples were taken from the households in the districts Michalovce and Stropkov.

Out of the overall number of the samples of beef analysed (2474 samples) only two above-limit samples, coming from the Nitra Region, were found during the whole monitoring period. With pork meat the situation is similar. Altogether 4229 samples were analysed, out of which only three did not meet valid hygienic limits. These samples were analysed in the 80`s and came from the Prešov Region.

Population exposition to hexachlorobenzene in observed stages
[SHMÚ, MŽP SR, SAŽP, ETV: Inventarizácia perzistentných organických látok v SR, Technická správa 2, časť 2,2003]

Kg / person / day	1984 - 1989	1990 – 1999	2000 – 2002
Receive mg / person / day	0.00223	0.00154	0.00096
% TDI (mg/kg/TH)	18.7%	12.9	8.1

View of average hexachlorobenzene findings in selected food staples
(mg/kg of fat, eggs – mg/kg)
[SHMÚ, MŽP SR, SAŽP, ETV: Inventarizácia perzistentných organických látok v SR, Technická správa 2, časť 2,2003]

Commodity	**Year**								
	1984	**1985**	**1986**	**1987**	**1988**	**1989**	**1990**	**1991**	**1992**
Beef	0,065	0,050	0,042	0,069	0,046	0,015	0,022	0,013	-
Pork	0,060	0,027	0,020	0,023	0,025	0,004	0,021	0,009	-
Eggs	ND	-	0,001	0,007	0,010	0,002	0,003	ND	-
Milk	-	-	0,030	0,035	0,055	0,038	0,034	0,013	0,059
Number of samples	9	8	896	787	836	948	686	483	10

Table continued by following years:

Commodity	**Year**									**Total**
	1993	**1994**	**1995**	**1996**	**1997**	**1998**	**1999**	**2000**	**2001**	**N. of S.**
Beef	0,010	0,005	0,002	0,004	0,006	0,002	0,003	0,002	0,003	2474
Pork	0,010	0,001	0,0004	0,002	0,001	0,0004	0,001	0,001	0,001	4229
Eggs	-	0,184	ND	-	0,021	0,001	0,001	0,002	0,002	1124
Milk	0,010	0,009	0,002	0,002	0,0003	0,002	0,002	0,003	0,001	3062
Number of samples	12	638	289	227	602	1301	1491	870	598	10889

Average HCB contents in monitored food commodities in the districts Michalovce (burdened area) and Stropkov (reference area). [56]			
Commodity	**Origin**	**District**	**Content** ($ng.g^{-1}_{of\ fat}$)
			HCB
Pork	chain store	Michalovce	**1.6**
Milk	chain store / import	Stropkov	**9.0**
Pork	chain store	Stropkov	**1.8**
Sausage	chain store	Michalovce	**2.4**
Milk	chain store / import	Michalovce	**12.8**
Frankfurters	chain store	Stropkov	**3.2**
Ham sausage	chain store	Michalovce	**1.3**
Beef	chain store	Stropkov	**1.8**
Frankfurters	chain store	Michalovce	**3.5**
Eggs	poultry house / producer	Michalovce	**3.0**
Eggs	chain store	Stropkov	**3.0**
Lard	private breeding facility	Stropkov	**6.9**
Butter	private breeding facility	Stropkov	**7.4**
Eggs	chain store / import	Michalovce	**2.7**
Milk	private breeding facility	Stropkov	**4.9**
Milk	dairy facility / producer	Michalovce	**11.4**
Ham sausage	chain store	Stropkov	**2.7**
Chickens	chain store	Stropkov	**1.9**
Sausage	chain store	Stropkov	**2.6**
Beef	chain store	Michalovce	**14.7**
Butter	private breeding facility	Michalovce	**13.2**
Lard	private breeding facility	Michalovce	**6.4**
Milk	private breeding facility	Michalovce	**13.1**
Eggs	private breeding facility	Stropkov	**16.6**
Chickens	chain store + private breeding	Michalovce	**10.6**
Eggs	private breeding facility	Michalovce	**40.7**

Dietary exposure

Comparing average and maximal HCB values from intake of daily food with ADI limits - in year 1996, significant exceeding of average and mainly maximal exposition amounts was registered. In following years, the decrease of the values was measured. In year 2001, maximal pesticides values reached 55.41% ADI. [SHMÚ, MŽP SR, SAŽP, ETV: Inventarizácia perzistentných organických látok v SR, Technická správa 2, časť 2,2003]

Exposition to HCB through food - population of districts Michalovce and Sobrance (mg/kg body weight/day). Source: [SHMÚ, MŽP SR, SAŽP, ETV: Inventarizácia perzistentných organických látok v SR, Technická správa 2, časť 2,2003]

Years	**Number of samples**	**Limit**	**Minimum**	**Average**	**Maximum**
1996	21	0,0006	0,000069	0,001557	0,01417
1998	20	0,0006	0,000086	0,000527	0,00143
2001	25	0,0006	0,000063	0,000183	0,00033
Total	66	-	0,000063	0,000756	0,01417

(average human weigh 60 kg)

One of the monitoring projects was aimed at investigating the actual state of environmental pollution (air, surface water, sediment, soil, fish and wild animals) in the surroundings of sources (mainly in the Michalovce District) of mainly PCB, but also HCB, γ-HCH contamination; further food contamination in the Michalovce District; levels in the human population via blood serum analysis, and to assess some parameters characterising the state of human health; all of this was to be compared to the reference area (the Stropkov District). Altogether 322 samples from adult donors from the Michalovce District were analysed (out of these 38 were exposed professionally, 66 were oncology patients, 14 fishermen consuming the fish from the water entities directly joined to the waste channel of Chemko a.s.) and 205 were from donors from the Stropkov District.

It was proved that consumption of the contaminated food from domestic breeding (mainly eggs and the chicken) leads to higher POPs findings, including HCB in the human organism. Not only the PCB levels, but also the **HCB and p,p'-DDE levels were in the both districts substantially higher than in West-European and North-American countries.** The difference in average HCB levels between the both monitored districts was minimal (Kočan et al. 1999, Petrík et al. 1999, Petrík et al. 2000, Petrík et al. 2001).

Within the project "Maternal milk", even the hygienic service deals with POPs monitoring. The project is focused on the monitoring of microbiological contamination and nutrition value and on reflection of quality of the environment to quality of maternal milk given to the infants. Twenty-one samples of maternal milk taken within the project, (2001-2002), were analysed for PCB congeners and selected organochlorine pesticides content. HCB levels in the milk samples ranged from 15.2 to 184 ng.g-1 of fat, with average value 96.6 ng.g-1 of fat (Miklánková a Horecká, 2003, Horecká et al., 2002).

HCB levels in the samples of the human blood serum, adipose tissue and maternal milk (calculated per fat ratio) **from six Slovak districts** (the samples taken in 1992-1994).
[SHMÚ, MŽP SR, SAŽP, ETV: Inventarizácia perzistentných organických látok v SR, Technická správa 2, časť 2,2003]

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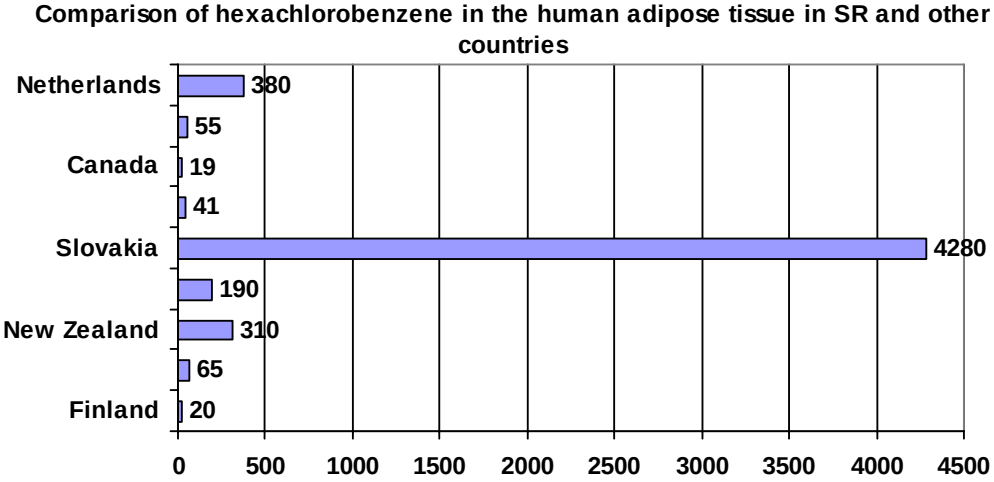
(average human weighs 60 kg)

Table: Measurements of PCBs (9 congeners addition), HCB, γ-HCH, p,p'-DDE a p,p'-DDT levels in the samples of **blood serum fat** in the different groups of the human population in the Michalovce (MI) a Stropkov (SP) districts –**HCB results** [48]

Definition of the selected group of population	**Content [ng.g^{-1} of blood serum fat]**
	HCB
Workers exposed to PCB (n=38)	**1 699**
Men (n=27)	**1 674**
Women (n=11)	**1 761**
Fishermen (MI, n=11)	**1 453**
Overall population MI (without workers exposed to PCB, n=215)	**1 921**
Together with the workers exposed to PCB (n=253)	**1 888**
Randomly selected from the workers exposed to PCB (n=225)	**1 912**



With fishermen (n=204)	**1 945**
Overall population SP (n=205)	**1 622**
Men MI (n=107); overall population	**1 419**
Women MI (n=108); overall population	**2 418**
Men SP (n=101); overall population	**1 217**
Women SP (n=104); overall population	**2 015**

Average levels in lipid. serums from 5 Slovak regions in 1992 [µg.kg^{-1}] [48]

Pollutant	Average	Median	Range
HCB	**5.38**	**4.27**	**0.16 - 23.20**

Source: Kočan, A., Petrík, J., Chovancová, J., Drobná, B., Uhrinová, H.: Chlórované aromatické zlúčeniny v ľudskom organizme z vybratých modelových oblastí Slovenskej republiky [Záverečná správa vedecko-technického projektu č. 93-535-03-32]. Bratislava, Ústav preventívnej a klinickej medicíny, dec. 1995, 170 s. The samples were taken in 1992-95 from 7 selected regions of SR – together 69 adipose tissues. The level in the graph represents average value from all the samples.

Fifty samples of the human blood were taken in 1992 from the general population living in five selected areas of the SR (Michalovce, Veľký Krtíš, and in the city wards of Nitra, from the region of Myjava and Bratislava). These were analysed, (apart from others), for HCB. HCB levels were about 100-times higher than in USA, Japan, Finland or Canada. [48]

HCB levels are note-worthy high in the maternal milk samples; they are one to two times higher than the values registered in other countries (see following table.). In the samples from Slovakia, surprisingly high HCB concentrations were found and they are probably caused by its use in agriculture and their creation during industrial production of some chlorine solvents. [48]

To the question about the causes / reasons of high HCB concentrations in adipose tissue in SR the ÚPKM Bratislava (2001) states:

The average age of donors of the human adipose samples taken and analysed within afore mentioned project is 48.4 y (16-83), that means that all the donors spent during the period to interdiction of HCB use around 8 to 75 years, this could **testify to residues of exposition by high HCB values in the past.**

In the blood serum from this study, the PCB levels are an average 4600 ng.g^{-1} (per fat), presenting a good match with the results in the human depot fats. In contrast to this, in the blood serum of 536 people from the districts Michalovce and Stropkov the average HCB level was 1980 ng.g^{-1}. These samples were taken in 1998-99, thus, 5 years later than those in afore mentioned study.

HCB levels in the food of biological origin taken in 1998-99, are at the level of units, maximally tenth of ng.g^{-1} of fat. Abroad, the HCB concentrations in food are comparable with our results, while the concentrations in the human fat range at a level only approximately 10 to15-times higher compared with the

values in the food. From this , **today the food in the SR should not be the cause of such a high exposure of the people.**

Yet Uhnák and col. in the almanac of lectures from VIII whole-state conference „Allochthonous agents in consumables“ (Starý Smokovec 1977), present **in the human fat 4820 ng HCB.g^{-1}** (thus, the value comparable with the results of ÚPKM), but for food the authors state much higher values: maternal milk 3470, cheese 623, butter 496, poultry fat 373, egg yolk 80 ng.g^{-1} of fat. From this data can be seen that **HCB levels in the food decreased over approximately 20 years by 15-20-fold, but the values in human fat just about a tenth to half**.

Conclusion – for evaluating the average value of HCB contents in the human biological material it would be necessary to provide a study with representative sample of the population of the SR, including the children. Above mentioned values do not meet this condition – in one case are, otherwise, taken from 7 SR districts, but just 10 samples from each district. In the second case, 536 samples were taken just in the Eastern-Slovak districts Michalovce and Stropkov.

What is related to HCB source – to this question considering present results of analysis it is not possible to answer exactly. **Since the HCB content in the food during the last 20 years has decreased significantly, it is possible that the food consumed in the 1970s and 1980s caused increased exposure of the Slovak population.**

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