

CONTAMINATION OF WATER AND SOIL BY POLYCHLORINATED BIPHENYLS

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Polychlorinated biphenyls (PCBs) are stable organic substances that are formed by chlorinating biphenyls and used as insulating fluids in transformers and condensers, in lubricants, plasticizers, paints, adhesives or seals. After contact with living organisms from sources of pollution of soil and water, they cause weakening of the immune system and damage to vital organs. Such ecological burdens have arisen as a result of the production of PCBs in industrial plants in different places of the world in 30-years of 20th century. This paper includes basic physico-chemical properties of PCBs, environment contamination, health risks and possibilities of PCBs degradation.

KEY WORDS: polychlorinated biphenyls, physico-chemical properties, contamination, degradation

KONTAMINÁCIA VODY A PÔDY POLYCHLOROVANÝMI BIFENYLMI. Polychlorované bifenyly (PCB) sú stabilné organické látky, ktoré vznikajú chloráciou bifenylov a používali sa ako izolačné kvapaliny v transformátoroch a kondenzátoroch, v mazadlách, zmäkčovačoch, farbách, lepidlach alebo tesneniach. Po kontakte so živými organizmami zo zdrojov znečistenia pôdy a vody spôsobujú oslabovanie imunitného systému a poškodzovanie životne dôležitých orgánov. Takáto ekologická záťaž vznikla ako výsledok produkcie PCB v priemyselných závodoch v rôznych miestach sveta v 30. rokoch 20. storočia. Tento príspevok zahŕňa základné fyzikálno-chemické vlastnosti PCB, kontamináciu prostredia, zdravotné riziká a možnosti degradácie PCB.

KLÚČOVÉ SLOVÁ: polychlorované bifenyly, fyzikálno-chemické vlastnosti, kontaminácia, degradácia

Introduction

Polychlorinated biphenyls (PCBs) are organic aromatic compounds with the chemical formula $C_{12}H_{10-n}Cl_n$ and characteristic general structure (Fig. 1).

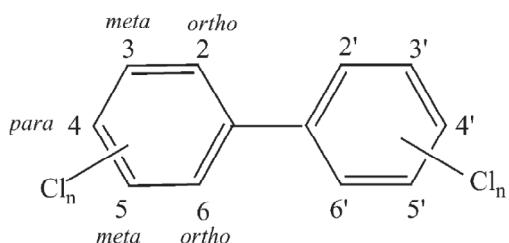


Fig. 1. Structural formula of PCB (Wiegel, Wu, 2000).

Obr. 1. Štruktúrny vzorec PCB (Wiegel, Wu, 2000).

PCBs were produced commercially by synthetic chlorination of biphenyls at higher temperature using catalysts as iron salts (Sahni et al., 2005). Obtained reaction mixture was neutralized, distilled and final product was refined. Such produced complex mixture of PCBs differed in number and position of chlorine atoms yielding up to 209 congeners. The degree of chlorination was affected by the amount of chlorine used in the reaction. PCB congeners with the same number of chlorine atoms are called homologs. Different chlorine positions in homologs form isomers. Unique physico-chemical properties of PCBs mixtures containing up to 90 congeners provided them use in a wide range of industrial applications expanded from 1930s to 1970s (Borja et al., 2005). In various locations of PCB production, mixtures of PCB were marked differently, e. g. Aroclor in USA, Kaneclor in Japan, Fenclor in Italy, Pyralene in France, Delor in ČSSR or Clophen in West Germany, which were used as electrical insulating liquid, heat-carrying medium or hydraulic fluid (Watts,

1998). Due to their use, PCBs have come into certain areas of the ecosystem, where they are very dangerous pollutants. In the 1960s they proved to be global contaminants of the environment. Nevertheless, that these substances are no longer produced and their production is banned, there some pollution impends, because some PCBs sources still exist in the ecosystem today. They have been often found in the tissues of organisms, especially in human tissues, and thus have entered the food chain. The transfer of PCBs into water poses an increased risk during deposition in the sediment, which is considered as PCBs reservoir (Borja et al., 2005). In 1984 PCBs production in Czechoslovakia was banned (Breivik et al., 2002), but there are still risks of leaks from earlier production, accidents or bad waste management (Nisbet, Sarofim, 1972). The main risk of PCBs is associated with their ability to remain for a long time in the soil and water (up to several decades), high resistance to degradation and lipophilic character, which is the reason of PCBs bioaccumulation in cells (Borja et al., 2005). One way how to eliminate PCBs from the environment is to use various physico-chemical methods, which are usually very costly and technically demanding. Therefore more suitable degradation mechanisms search in recent years. One of options is the use of living organisms in the process of biodegradation, when dangerous pollutants can be transformed to less toxic substances or mineralized (Cookson, 1995). This paper reviews literature overview and studies on PCBs, their basic physico-chemical properties, water and soil contamination and possibilities of degradation.

Basic physico-chemical properties of polychlorinated biphenyls

Manufacture of PCB led to production of congener mixture with molecular weight ranging from 188 to 439.7 depending on the number of chlorine atoms attached to the biphenyl rings. Physico-chemical properties and toxicity of PCBs depends on the degree of chlorination and the position of chlorine. The most toxic congeners are with 5 and 10 chlorine atoms in the para-, meta-positions (Sylvestre, 1985) and the 3,4-ortho positions (Albro, McKinney, 1981). The boiling point of the products was directly related to chlorination. It ranged from 34°C for monochlorobiphenyls up to 300°C for decachlorobiphenyls (Danielovič et al., 2009). PCBs are strongly hydrophobic. Lower chlorinated PCBs forms usually colorless oily liquids. Pentachlorobiphenyls are more viscous oils. The highly chlorinated PCBs tends to be greases and waxy substances yellow color. PCBs are soluble in organic solvents, oils and fats and slightly soluble in water, which decreases with increase in the number of chlorine atoms. They have very low ionizability. Polychlorinated biphenyls are very resistant to the effects of bases and acids, usual oxidizing and reducing agents. They are also resistant to hydrolysis and alcoholysis (Borja et al.,

2005). Only 19 congeners are characterized by chirality, spatially geometric property, while the chiral substance is not identifiable with its mirror image and has no center or plane of symmetry, however, it may have an axis of symmetry (Kaiser, 1970). Properties of PCBs suitable for industrial applications include chemical stability, thermal resistance, non-flammability, high density, chemical inertness, high electrical resistivity or high dielectric constant and low acute toxicity (Hutzinger, 1974). It has been shown that these properties are a great threat for the environment and the quality of living organisms life.

Water and soil contamination by polychlorinated biphenyls and health aspects

Increased PCB content was found primarily in highly industrial areas. Therefore, contaminated sites include areas where PCBs were used. PCBs persist in the environment and some amounts of them were confirmed in air, water, soil, and food. Manufacture, use, transport and disposal of PCBs led to the accidental spills. Fires in products containing PCBs entered the air, in which they were able travel long distances and contaminate the environment (Borja et al., 2005). It has been found that an important cyclical mobility mechanism of PCBs is an evaporation from surfaces. Contaminants are transported by the movement of air masses with water vapor up to large distances. PCBs have been identified even in remote parts of the world where they have never been used, i.e. in the polar region, as evidenced by their distant transport in the air (Muir et al., 1988; Vecchiato et al., 2015; Ubl et al., 2012). In Czechoslovakia PCBs were produced in Chemko Strážske in Eastern Slovakia in 1959-1984. The estimated amount of PCBs was around 25000 tons, of which a large volume leak out into the environment. Despite the fact that the production of PCBs was forbidden here in 1984, due to the persistence of the substances, the contamination is still present (Kocan et al., 2001; Kaštánek, Kaštánek, 2005). In Eastern Slovakia an important source of contamination by PCBs is Strážsky Canal, which flows into the Laborec River (Danielovič et al., 2009). PCBs are sorbed on the organic matter in the soil (Zheng et al., 2014) and sediment (Smith et al., 2009). So sediment of Zemplínska šírava is contaminated because it is fed by the Laborec River (Danielovič et al., 2009). Fig. 2 illustrates the schematic map of PCBs contaminations location in Eastern Slovakia, where PCBs migrated in aqueous environment from Stražsky canal, through the Laborec River into the Zemplínska šírava water reservoir. PCBs came into the groundwater from the Laborec River in the riverine zone. From mentioned sources of pollution, they get into the bodies of the organism and into the food chain. It has been shown that PCBs are toxic to aquatic organisms affecting their reproduction and growth. Polychlorinated biphenyls are teratogenic, adversely affecting the enzymatic system and liver. They have a negative effect on the immune



Fig. 2. The schematic map of PCBs contaminations location in Eastern Slovakia (Brázová et al., 2012).

Obr. 2. Schématická mapa miesta kontaminácie PCB na Východnom Slovensku (Brázová et al., 2012).

system and cause acne-like eruption of blackheads (Bherstilee, 1994). The main source of PCBs for human is food. PCBs are rapidly absorbed from the digestive system and, due to their lipophilic character, accumulate in adipose tissue and liver. PCBs migrate from the mother to the offspring via the placenta and breast milk (Farooq et al., 2003). PCBs negatively affect the immune, hormonal, nerve and reproductive system (Passatore et al., 2014). PCBs are included in the list of potentially carcinogenic substances (Lauby-Secretan et al., 2013). Due to their high biological persistence, there is a big problem with liquidation these substances. Commercially used technologies are limited to only landfilling and thermal destruction processes. In the chemical practice, due to the difficulty of the work associated with the determination of congeners, only the PCBs indicators representing representatives of commercial mixtures are analyzed. In environmental samples, as well as samples of plant and animal materials, congeners number 28 (trichlorobiphenyl), 52 (tetrachlorobiphenyl), 101 (pentachlorobiphenyl), 138, 153 (hexachlorobiphenyl) a 180 (heptachlorobiphenyl) are monitored (Danielovič et al., 2009). The main often used method for quantitative determination of PCBs is high resolution gas chromatography usually combined with mass spectrometry (Castelli et al., 1983).

Possibilities of polychlorinated biphenyls degradation

PCBs are subject to spontaneous degradation very slowly and they persist in environment for a long time. Therefore, searching of new mechanisms for their uptake and removal from sources of contamination is the main goal since the canceling of their production. Methods used to decontaminate soil and sediment containing PCBs include incineration, chemical oxidation, solvent extraction, reductive dechlorination, vitrification or thermal desorption (Gomes et al., 2013). During the incineration of contaminated soil, it must be pulled off and mostly transferred to the site of decontamination, in addition, incineration causes irreversible soil degradation. During combusting of PCBs, it is necessary to keep a constant high temperature above 1300 °C in order to avoid the formation of highly toxic polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (Erickson et al., 1989; Totevová et al., 1997). Different sorbents can be used for decontamination, e.g. activated carbon (Beless et al., 2014), bentonite or various polymers (Kaštánek et al., 1995), which must then be dumped or incinerated. Reductive dechlorination of PCBs can be achieved also by application of nanosized Fe/Pd bimetallic particles (Xu, Bhattacha-

ryya, 2007). One technically less demanding possibility is also to use a bioremediation method, i.e. to use live organisms for pollutants removing. Congeners with a higher number of chlorine atoms can be degraded by anaerobic reductive dehalogenation in which halogenated substances serve as an alternative electron acceptor in anaerobic respiration (Mohn, Tiedje, 1992). This process is carried out, for example, by bacteria of the genus Dehalococcoides (Field, Sierra-Alvarez, 2008). Aerobic degradation of PCBs is realized, for example, by some bacterial strains of Pseudomonas (Nováková et al., 2002), Burkholderia (Tillmann et al., 2005), Alcaligenes (Murínová et al., 2014), or Rhodococcus (Mukerjee-Dha et al., 1998). In general, congeners with lower number of chlorine atoms and congeners substituted on only one aromatic ring are better degradable in this way (Nováková et al., 2002, Tillmann et al., 2005, Murínová et al., 2014, Mukerjee-Dha et al., 1998). The ability to transform PCBs has been described, for example, in Solanum nigrum (Mackova et al., 1997), Nicotiana tabacum (Rezek et al., 2008) or in mushroom organisms as Ascomycota a Zygomycota (Tigini et al., 2009; Mouhamadou et al., 2013; Sage et al., 2014). An excellent ability to degrade PCBs with an efficiency of about 99% in the fluid culture medium containing ligninolytic mushroom Pleurotus ostreatus was observed. This strain is among the few species described, which are able to degrade both penta- and hexachloropenic biphenyls and reduce the acute toxicity of the medium (Čvančarová et al., 2012). It was confirmed also photodegradation of PCBs by UV irradiation (Maio et al., 1999). For cleaning of the contaminated water can be used also magnetoferitin (Urban et al., 2010), which has ability to uptake PCBs into its unique nanoscale structure. In addition, PCBs inside magnetoferitin can be easily separated from surrounds thanks to the presence of magnetic nanoparticles using magnetic separator. There is premise, that magnetic nanoparticles of iron oxides in magnetoferitin can also degrade PCBs (Sun et al., 2015).

Conclusion

In this paper, basic knowledges about polychlorinated biphenyls including their production, physico-chemical properties, circulation in the ecosystem and processes of degradation were discussed. Polychlorinated biphenyls are characterized by high chemical and temperature stability, toxicity and hydrophobicity, depending on the position and number of chlorine atoms. Polychlorinated biphenyls persist in the ecosystem and through the food chain settle in the body of living organisms by binding to the adipose tissue followed by the disruption of immunity and organs. Primary sources of PCBs have been almost eliminated, but secondary sources are still dangerous. Therefore, the important part of this review is to introduce with the possibilities of polychlorinated biphenyls degradation. There are several ways how to transform toxic PCBs to less harmful substances, as

physical methods or technically less demanding bioremediation methods with the use of living organisms. The new perspective material for polychlorinated biphenyls separation and degradation could be also magnetoferitin. Before development of suitable rapid and non-invasive methods of PCBs destroying it is necessary to protect the ecosystem by the continuous monitoring of these substances.

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KONTAMINÁCIA VODY A PÔDY POLYCHLOROVANÝMI BIFENYLMI

Polychlórované bifenyls sú syntetické organické látky zložené z dvoch bifenylových kruhov, na ktorých je naviazaný rôzny počet chlórov. Polychlórované bifenyls boli komerčne produkované v mnohých priemyselných závodoch sveta od roku 1930 približne do roku 1980. Ich fyzikálno-chemické vlastnosti, najmä vysoká tepelná stabilita a chemická odolnosť ich predurčili napríklad na použitie ako izolačných kvapalín v kondenzátoroch alebo transformátoroch. Neskôr sa ukázalo, že práve tieto vlastnosti sú veľkou hrozobou pre nás ekosystém. Bola zistená cyklická mobilita polychlórovaných bifenylov po vyparovanie z kontaminovaných vodných povrchov. Kontaminanty môžu byť transportované pohybom vzdušných mäs do veľkých vzdialenosí, ale hlavným prostredím ich migrácie je vodné prostredie. Napriek tomu, že je výroba polychlórovaných bifenylov už pozastavená, stále existujú zdroje znečistenia vody a pôdy. Na Východnom Slovensku ide o Strážsky kanál, do ktorého ústili odpadové vody z chemického závodu Chemko (Strážske) a ktorý sa napája na rieku Laborec. Odtiaľ sa znečistenie rozšíriло do vodnej nádrže Zemplínska šírrava, ktorej sediment tvorí hlavný rezervoár polychlórovaných bifenylov. Tie sa dostávajú do potravinového reťazca, do tel rôznych organizmov, vrátane človeka, a spôsobujú oslabovanie imunitného systému a poškodzovanie životne dôležitých orgánov. Polychlórované bifenyls boli taktiež zahrnuté do zoznamu potenciálne karcinogénnych látok. Hlavné

riziko polychlórovaných bifenylov je spojené s ich schopnosťou pretrvávať veľmi dlhy čas (až niekoľko desiatok rokov) vo vode a v pôde, s vysokou odolnosťou voči degradácii a lipofilným charakterom, ktorý je príčinou ich bioakumulácie v bunkách. Preto je klúčovým cieľom výskumu v tomto smere nájsť vhodný spôsob na separáciu, elimináciu a degradáciu polychlórovaných bifenylov. Jednou z možností môže byť použitie rôznych fyzikálno-chemických metód, ktoré sú zvyčajne drahé a technicky náročné. Preto sa hľadajú vhodnejšie spôsoby degradácie, ako napríklad použitie živých organizmov (baktérií, hub alebo rastlín), ktoré sú schopné rozložiť polychlórované bifenyls na menej toxickej zlúčeniny. Nový perspektívny materiál na separáciu a degradáciu polychlórovaných bifenylov môže byť aj magnetoferitín, ktorý je schopný prijať tieto škodlivé látky do svojej unikátnej nanorozmernej štruktúry. Navyše, polychlórované bifenyls vnútri magnetoferitínu môžu byť ľahko odseparované z okolitého roztoku vďaka prítomným magnetickým nanočasticiam použitím magnetického separátora. Takisto tu existuje predpoklad, že magnetické nanočastice oxidov železa v magnetoferitíne môžu tiež degradovať polychlórované bifenyls. Je dôležité pripomenúť, že pred vývojom vhodnej, rýchlej a neinvazívnej metódy na destrukciu polychlórovaných bifenylov, je nevyhnutné chrániť ekosystém neustálym monitorovaním týchto látok a predchádzať ich styku s organizmami.

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