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How are polychlorinated biphenyls currently being produced, despite the production ban, and do they pose a risk to the environment?

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Abstract

Polychlorinated biphenyls (PCBs) are a group consisting of 209 congeners, differing in the number and site of substitution of chlorine atoms to the biphenyl molecule. Due to their physicochemical properties, they have found wide industry use. As a result of many years of large-scale use of PCBs, their toxic properties began to be noticed as they manifested in poisoning among humans. After thorough testing of PCBs, they were classified in the group of persistent organic pollutants (POPs), and their production was banned and they were withdrawn from use. Although PCBs are no longer produced in factories, they can still be formed. In this article, will be presented and explained methods of accidental formation of PCBs. Based on data from the literature, we performed an environmental risk assessment for PCB-contaminated soils.

1. INTRODUCTION

Polychlorinated biphenyls (PCBs) are aromatic compounds that occur as mixtures of congeners. There are 209 congeners of PCBs, but only about 100-130 are found in industrial mixtures [ATSDR 2000, Erickson 2001]. The substitution site and the number of chlorine atoms in the molecule determine a particular congener's chemical and physical properties, and consequently, its environmental behaviour and toxicity. The single, central carbon-carbon bond connecting the phenyl rings in the chlorobiphenyl molecule makes the rings relatively free to rotate relative to each other. Therefore, depending on the substitution site and the number of substituents, the two rings can arrange themselves spatially, differently to each other [Falandysz 1999]. A special group of PCBs compounds characterised by high biological activity and toxicity includes planar (coplanar) PCBs, i.e. chlorobiphenyls with a flat spatial structure, resulting from the alignment of the phenyl rings almost in one plane. The coplanar position is characterised by the absence of chlorine atoms near the interring bond, meaning that these are chlorobiphenyls free of chlorine atoms in the ortho position. They are called non-ortho chlorine-substituted PCBs, or non-ortho coplanar PCBs [ATSDR 2000, Erickson 2001]. In 1998, due to the similarity of toxic effects on living organisms, the WHO classified non-ortho and mono-ortho PCBs as 'dioxin-like' PCBs, which should be analysed together with dioxins to determine toxicity levels [Van der Berg 1998, Dudzińska

2004]. The dioxin-like PCBs distinguished by the WHO include 12 congeners with IUPAC numbers 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, and 189. Individual, chemically pure PCBs congeners are colourless and usually occur in crystalline form. Depending on their composition, technical preparations are colourless, or yellow or dark brown thick oils [Falandysz 1999]. PCBs are characterised by poor electric conductivity, low flammability, high chemical stability, resistance to thermal decomposition, and environmental degradation by biological, photolytic, and chemical processes [Willey 2007]. Due to these properties, PCBs were widely applied in the industry sector [Erickson 1997, ATSDR 2000, Erickson 2001, Dudzińska et al. 2004]. The WHO classified PCBs usage into three categories [WHO 1993]:

- completely closed systems: electrical transformers, electrical capacitors, electrical switches, relays and other device, electrical cables, electric motors, and electromagnets;
- incompletely closed systems: plumbing systems and heat transfer systems (heaters, heat exchangers);
- open systems: softener in polyvinyl chloride, neoprene, and other types of artificial rubber; components in paints and coatings; components in carbonless ink and carbon paper; components in adhesives; a volumetric admixture of pesticide; components in lubricants, sealants, and sealants; and a fireproof material in fabrics, carpets, and polyurethane foam.

2. METHODOLOGY

Based on the scientific literature related to the occurrence of PCBs in the environment, this review has two main objectives: (1) to describe the different possibilities surrounding the production of PCBs, and (2) to highlight PCBs' existence in the environment although they have been withdrawn from use. Our keywords – polychlorinated biphenyls, environmental risk assessment, earthworm, synthesis *de novo*, incineration, road transport, and dioxin-like – were selected individually or jointly to search for articles on the Scopus database, Springer database, and Google Scholar. The literature search included publications from 1966–2022.

3. PRODUCTION OF PCBs

PCBs were originally synthesised in 1881 by German chemists [Ododo et al. 2019]. Production of PCB-based oils on a mass scale began in 1929. PCBs were produced under various trade names. Among the most common are: Clophen (Germany), Aroclor (USA), Keneclor (Japan), Pyralen, Flix (France), and Sovol (USSR) [Ericsson 1997, Falandysz 1999]. In Poland, polychlorinated biphenyls were produced in small quantities for a short time, while imports of these substances from other countries were more significant. PCBs were produced at two industrial plants: the Tarnów-Mosice Nitrogen Plant in 1971–1976, under the name Tarnol or Chlorinated Biphenyl, and the ERG plant in Żabkowice Będzińskie, under the name Chlorophene. It is estimated that by 1980, about 2,000,000 tons of PCBs were produced worldwide (according to WHO). The largest producers were the United States, Germany, the USSR, France, and Italy. In the 1970s, PCBs production was banned in most countries. The United States was the first to ban it in 1977; in Europe (France and Spain), production continued until 1985 [WHO 1993]. 48% of the PCBs produced were used in transformer cooling oils and 21% were used in dielectric fluids of capacitors [Whyllie et al. 2010]. Polychlorinated biphenyls are persistent, lipophilic, and hydrophobic, with a high potential for bioaccumulation in living organisms [Kaya et al. 2018]. In addition, they have high stability and low susceptibility to degradation [Erickson 2001]. The half-life of PCBs in the soil varies from 3 months to 47 years [EA 2007; Cachada et al. 2009]. Due to their toxicity and stability, the production of PCBs was banned in the 1970s in most countries [Cachada et al. 2009]. Polychlorinated biphenyls have been managed globally under the Stockholm Convention on POPs since 2004, requiring environmentally sound management of PCBs by 2028 [Melymuk et al. 2022]. In Europe, their production took place until 1985 (France and Spain) [WHO 1993]. About 1.3 million tonnes of pure PCBs, through dilution and poor management, were expanded to 17 million tonnes of PCBs contaminated materials and waste, with an estimated 20–35% of PCBs being released into the environment [UNEP 2016, Melymuk et al. 2022].

4. PCBs DE NOVO SYNTHESIS

PCBs were industrially produced during electrophilic substitution reactions with catalysts (Lewis acids like FeCl_3), as complex mixtures with the direct chlorination of biphenyls using anhydrous chlorine [Robinson et al. 1994]. Small amounts of PCBs can be formed spontaneously, from suitable precursors, such as waste incineration, chlorination of drinking water, or chlorine bleaching of pulp [Falandysz 1999, Erikson 2001, Ludwicki et al. 2002]. PCBs can be formed in synthesis *de novo* as by-products during thermal processes, like incomplete petrol combustion in motor engines or waste incineration [Jin et al. 2011]. During petrol combustion *de novo*, PCBs synthesis is initiated by the compounds in petrol, such as dienes, benzene, chlorobenzenes phenols, and diphenyl esters [Brož et al. 2000]. Because of this, road transport can be a source of PCBs being released into the environment. In Poland, soil samples were collected along exit routes from Warsaw roads leading to Gdańsk, Lublin and Poznań, and tested. The total content of PCBs in this soil was within 0.331–37.077 ng/g [Gabryszewska et al. 2018]. This study proved that PCBs content in the soil depends on the speed of the cars and the braking distance, which is related to the amount of flue gases, and also that the age of the road relates to a longer deposit time and a larger PCBs content in the soil. Studies have shown that PCBs emissions from modern diesel engines equipped with catalysed exhaust after-treatment systems for PM and NOx removal were low [Laroo et al. 2011, Laroo et al. 2012, Liu et al. 2011].

Studies carried out in areas along the Bohai and Yellow Sea regions showed that mean PCBs concentrations were higher in urban soils (20.7 ng/g) compared to rural areas. It was shown that unintentionally produced PCBs emissions have a major impact on PCBs content in soils, and they largely come from the production of (42%), pig iron (37%), crude steel (18%), and rolled steel (3%) [Song et al. 2018]. It is known that PCBs 47, 51, 68, and other congeners exist in polymer products in trace levels, like silicone, but polymers have not been identified as a significant environmental source of PCBs. Herkert et al. took air samples at 16 residences and found PCB 47, PCB 51, and PCB 68, which accounted for up to 50% of measured indoor ΣPCBs (2700 pg/m^{-3}) [Herkert et al. 2018]. Congeners PCB 47, PCB 51, and PCB 68 were also found in samples of bio-indicators (dandelion and kale) and soil in the vicinity of a silicone rubber factory in Germany [Hombrecher et al. 2021]. Hombrecher et al. discovered that silicone rubber production using bis(2,4)-dichloro benzoyl peroxide (2,4-DCBP) as a cross-linking agent emits significant amounts of the non-Aroclor PCBs congeners PCB 47, PCB 51, and PCB 68 into the ambient air. PCBs emissions were reported in the range of 150–300 mg/kg in flue gas condensate flakes [Hombrecher et al. 2021].

In China, copper smelting is an important source of unintentional PCBs production [Jiang et al. 2015]. Studies have been carried out showing that fly ash can contribute to PCBs formation in the cooling zone of a SeCu smelter.

In China, fly ash is recycled to recover residual metal; however, exposing fly ash to air promotes the formation of PCBs. The thermal treatment of fly ash without a controlled treatment atmosphere can increase the risk of formation of dl-PCBs [Jiang et al. 2015]. Tests of agricultural soils in China showed that the sum of 209 PCBs congeners' concentrations in soils ranged from 64.3 to 4358 pg/g. The dominant PCBs congeners were PCB 11 and PCB 209 in eastern China, and PCB 44+47+65 and PCB 68 in southern China. Their spatial distributions depended on local sources for PCBs. The source apportionment results indicated that historically produced and commercially used mixtures of PCBs were the dominant contributors to the concentrations of highly chlorinated PCBs congeners in China's agricultural soils. Sources of unintentional PCB production (pigment/paint, combustion-related sources, and polymer sealant) contributed to 57.4% of the total PCBs, making up the majority of PCB buildup in agricultural soils [Mao et al. 2021].

The effect of waste incinerators on PCBs content in the environment has been tested in Poland. Soil and plant samples were collected around the municipal waste incinerator plant (MWI) and the industrial waste incinerator plant (IWI). The highest accumulation of PCBs in plants near MWI was recorded in *Tanacetum vulgare* leaves (12.45 ng/g), and near IWI in the grass (4.3 ng/g) [Gabryszewska, Gworek 2020]. The accumulation of PCBs in soils, for both kinds of waste incinerators, was similar, at approximately 3 ng/g [Gabryszewska, Gworek 2020]. PCBs may also be formed during the combustion of biomass in the household stove, where concentration in the exhaust gases was 1.68 ng/kg [Moltó et al. 2010]. In Turkey, around a cogeneration heat and power plant, PCBs contents were found to be 1.05 ng/g in soil and 0.38 ng/g in ashes [Gedik et al. 2011].

5. ENVIRONMENTAL RISK ASSESSMENT

The effect of PCBs production during *de novo* synthesis on PCBs content in soils appears to be small. However, is it safe for the environment? To answer this, environmental risk assessment calculations were performed. The environmental risk assessment from PCBs-contaminated soils was based on calculations of the quotient of PEC to PNEC. Due to the impossibility of calculating PEC values for PCBs (with no data needed for calculations), the worst case was assumed for the risk assessment. Actual measured values of PCBs content were assumed as PEC values. If the result of the quotient calculated in this way exceeded the value of 1, these PCBs' contents in the environment posed a risk to the test organism, and if the value of the quotient was equal to or less than 1, the amount of these PCBs in the environment was considered acceptable [Gworek et al. 2002].

Due to the limited availability of ecotoxicological literature data, we tested soils from areas along Poznańska, Gdańska, and Lubelska, and also soils from areas around an industrial waste incinerator and municipal waste incinerator. The results of these calculations are presented in Table 1.

Abbreviations used:

PEC - predicted environmental concentrations

PNEC - predicted no-effect concentration

AF - assessment adjustment factor

$E_b C_{50}$ - concentration of the test substance concentration for which a 50% reduction in biomass growth is observed

$LC_{50\ 96h}$ - concentration of the substance at which half of the test animals die (after 96 hours of the test)

NOEC - concentration of the substance at which no effect is observed

LC_{50} - concentration of the substance at which half of the test population dies

Earthworms are key organisms in the decomposition of organic plant matter; their population increases with the availability of organic matter. Due to their widespread occurrence and importance in soil systems, they are considered to be very useful organisms for assessing soil contamination. Residual chemicals accumulate in earthworms and can be transferred by them to the tissues of animals with higher trophic levels in the food chain [Edwards, Thompson 1973]. Concerns have also been raised about the environmental impact of persistent pollutants such as dioxins and PCBs. Earthworms play a significant role in biological monitoring because they can bioaccumulate or bio-concentrate xeno-biotic chemicals [Tharakan et al. 2004]. They are used to measure heavy metal levels and persistent organic soil contamination.

5.1. Eisenia foetida

$LC_{50} = 30.4\ \mu\text{g}/\text{cm}^2$ [Fitzpatrick et al. 1992].

To convert the LC_{50} unit (from $\mu\text{g}/\text{cm}^2$ to $\mu\text{g}/\text{kg}$), assumptions were made following the guidelines for performing exposure assessments, in terms of ecotoxicology for plant protection products.

1 m² of soil weighs 75 kg, with the following assumptions: soil depth of 5 cm, soil density of 1.5 g/cm³

AF = 1000 [ChemSafetyPro 2016]

$PNEC = LC_{50} / 1000$

$PNEC_{dgs} = 4053.3\ \mu\text{g}/\text{kg}$

If we assume that: flakes emitted in outlet gases of the rubber factory end up directly in the soil, PEC_{soil} is equal to PCB concentration in flakes, and flakes will be eaten by earthworms, then half of the earthworm population exposed to the dose of PCBs near the rubber factory will die. None of the above quotient values (Table 1), except for the value from the rubber factory, exceeded the value of 1. Hence, the conclusion is that PCBs' contents in soils from the study areas in Poland do not pose a threat to earthworms.

Under Polish law, risk-causing substances of particular relevance to the protection of the earth's surface and the permissible contents of these substances in the soil [mg/kg dry weight of the earthen parts of the soil (<2 mm)] were determined for a depth of 0-0.25 m ppt.

Table 1. Soil PCBs exposure assessment results for earthworms

Tested area	Highest total PCBs' concentration in soil (µg/kg)	PEC/PNEC _{earthworms}
Poznańska route*	37,07	0,009146
Lubelska route*	20,87	0,005149
Gdańska route*	4,16	0,001026
Industrial waste incinerator**	2,59	0,000639
Municipal waste incinerator**	3,38	0,000834
Flakes from silicone rubber factory in Germany ***	300000	74,0137

Data source:

* - [Gabryszewska et al. 2018]

** - [Gabryszewska, Gworek 2020]

*** - [Hombrecher et al. 2021]

Permissible contents of risk-causing substances by the land group [Dz. U. 2016]:

I group - built-up and urbanised land - 0.02 mg/kg (20 µg/kg)

II group - agricultural land - 0.02 mg/kg (20 µg/kg)

III group - forest land - 0.02 mg/kg (20 µg/kg)

IV group - industrial land - 2 mg/kg (2000 µg/kg)

The threshold values given apply to each of the compounds in the PCBs group, specifically: PCB 28, PCB 52, PCB 101, PCB 118, PCB 138, PCB 153, PCB 180. No exceedances of PCBs' permissible contents in soil were detected.

6. SUMMARY

Despite the cessation of the production of PCBs and their withdrawal from use, PCBs still exist in the environment, even in places where they were not used previously. PCBs have been identified in Baltic fish [Jensen 1966], and subsequently in human and animal tissues [Jensen 1966, Erickson 2001, Ciereszko et al. 2004]. Traces of PCBs have also been discovered in remote regions of the Arctic [Bartlett et al. 2019] and Antarctica, that is, in regions

where industrial pollution should not occur [Wania et al. 1996]. The reason PCBs occur in areas where PCBs have not been used is that PCBs can be formed spontaneously from suitable precursors. Additionally, PCBs can be transported by the dust in the air for long distances, and PCBs from the air can be transferred to ground surfaces via dry particulate deposition [Güzel et al. 2020]. An understanding of unintentional PCBs formation mechanisms could be used to guide the development of methods for the control and reduction of PCBs content in the environment. However, mechanisms of unintentional PCBs formation during industrial processes are not well known.

Considering data from the literature for PCBs concentrations in soil, a risk assessment for earthworms was performed. None of the PCBs' concentrations in soil posed a risk to earthworms. For PCBs concentrations in the soil to pose a risk to earthworms, the concentration should have exceeded 4053.3 µg/kg, in which case the value of the PEC/PNEC_{earthworms} ratio would have been greater than 1. If flakes from the silicone rubber factory in Germany would have fallen on the soil and been eaten by earthworms, then half of these earthworms would die.

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