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Unintentionally produced polychlorinated biphenyls in pigments: An updated review on their formation, emission sources, contamination status, and toxic effects

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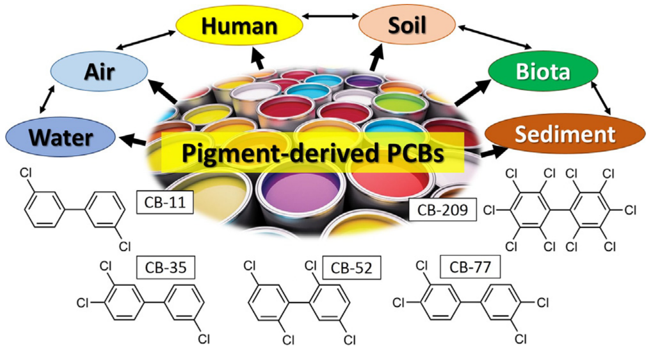
HIGHLIGHTS

- Pigment manufacturing processes serve as unique sources of PCBs.
- The most studied congeners have been CB-11, -35, -52, -77, and -209.
- Pigment-derived PCBs are ubiquitous in the environment.
- Pd-PCBs may exhibit neurotoxicity, endocrine disruption, and carcinogenicity.
- A total solution for management and abatement of pd-PCBs is needed.

GRAPHICAL ABSTRACT



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ABSTRACT

The formation, emission, environmental occurrence, and potential adverse effects of unintentionally produced polychlorinated biphenyls (PCBs) in pigments are reviewed, providing a comprehensive and up-to-date picture on these pollutants. PCBs are typically formed during manufacturing of organic pigments that involve chlorinated intermediates and reaction solvents, rather than those of inorganic pigments. Concentrations and profiles of PCBs vary greatly among pigment types and producers, with total PCB levels ranging from lower than detection limits to several hundred ppm; major components can be low-chlorinated (e.g., CB-11) or high-chlorinated congeners (e.g., CB-209). Pigment-derived PCBs can be released into the environment through different steps including pigment production, application, and disposal. They can contaminate atmospheric, terrestrial, and aquatic ecosystems, and then affect organisms living there. This situation garners scientific and public attention to nonlegacy emissions of PCBs and suggests the need for appropriate monitoring, management, and abatement strategies regarding these pollutants.

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1. Introduction

Polychlorinated biphenyls (PCBs) are a group of organohalogen compounds that comprise 209 possible chemical formulas (also called as congeners) with 1 to 10 chlorine atoms in the molecules (corresponding to 10 homologs) (Stringer and Johnston, 2001). A total of about 1.3 million tons of technical PCB mixtures were produced globally from the late 1920s to the early 1990s in eleven countries now known as the United States (US), Germany, Russia, France, the United Kingdom (UK), Japan, Italy, Spain, Czech Republic, China, and Poland (Breivik et al., 2007). PCBs are generally chemically inert, with outstanding fire retardant, thermal conductivity, and electric insulation properties, leading to their widespread uses as dielectric fluids in transformers and capacitors, heat transfer fluids, hydraulic fluids, and additives in various closed systems and open applications (UNEP, 1999; Erickson and Kaley II, 2011). While technical PCB mixtures were mainly applied in closed electrical and industrial systems, several environmental and human exposure incidents related to these chemicals were reported since the late 1960s (Jensen et al., 1969; Kuratsune et al., 1971). Because PCBs are environmentally persistent and toxic, the production of these compounds was stopped in Japan in 1972, followed by the US in 1977, and in other countries by 1993 (Breivik et al., 2002a, 2007). Besides the intentional production of PCBs described above, these compounds (notably dioxin-like PCBs) can be formed inadvertently during various industrial thermal processes such as waste incineration, cement production, and metallurgy (Gong et al., 2017). In 2001, PCBs were listed under the Annex A (elimination) and Annex C (unintentional production) of the Stockholm Convention on Persistent Organic Pollutants (POPs) (UNEP, 2001).

PCBs have been used as plasticizers in paints and surface coatings but detailed information about their compositions and usage patterns in this sector has not been fully documented (Erickson and Kaley II, 2011). Jartun et al. (2009) measured seven indicator PCBs in old building paint samples from Bergen, Norway at high concentrations (median 0.250; range < 0.001–3390 ppm), indicating the intentional applications of PCBs in paints. On the other hand, the unintentional occurrence of PCBs at concentrations as high as 300 ppm was found in some phthalocyanine pigments prepared by using chlorinated solvents (e.g., trichlorobenzene) as reaction medium, with patterns similar to those of technical mixtures containing 54% chlorine (Uyeta et al., 1976; Buchta et al., 1985). Moreover, initial attention of some novel PCB congeners (e.g., CB-11 and CB-36) with trace levels in technical formulations was described by some analytical environmental chemistry studies, implying new sources from pigment industries (ETAD, 1984; Sistovaris et al., 1990; Rastogi, 1992; Law, 1995). The concern about PCBs as impurities or degradation products in pigments has increased since the 2000s, when CB-11 was found in waste streams (Litten et al., 2002) and in a variety of environmental compartments (King et al., 2002; Choi et al., 2008; Hu et al., 2008; Basu et al., 2009; Du et al., 2009) at unexpectedly higher abundance as compared to other technical PCB congeners. It should be noted that a majority of previous studies concentrated only on a subset of PCB congeners, especially seven indicator congeners (i.e., CB-28, -52, -101, -118, -138, -153, and -180) and twelve dioxin-like PCBs. Percentages of CB-11 and CB-209 in technical PCB mixtures like Aroclors and Kanechlors were generally <0.1% by weight (Frame et al., 1996; Kim et al., 2004), and therefore the presence of these compounds has usually been omitted. The discovery of a novel PCB congener like CB-11 in its emission sources and in the environment was largely due to the application of high resolution gas chromatography/high resolution mass spectrometry (HRGC/HRMS) with isotope dilution/internal standard quantification, which is capable of obtaining full congener-specific profiles of PCBs (US EPA, 2003, 2008, 2010; Litten et al., 2002; Kim et al., 2004).

In the current study, we review literature spanning the last four decades on pigment-derived PCBs (pd-PCBs) to provide comprehensive and updated insights into novel and ongoing emission sources of these legacy, long-ago-banned pollutants. The references included in this review were searched from databases including ScienceDirect, Springer, American Chemical Society (ACS) Publications, Royal Society of Chemistry, Society of Environmental Toxicology and Chemistry (SETAC), Oxford Academic, Taylor & Francis Online, Hindawi, etc., by using keywords such as “PCBs”, “CB-11”, “CB-209”, “pigment”, “paint”, etc. Relevant information prepared as reports of international and national professional authorities was also included. The data presented here discuss the following issues: (1) the formation pathways of PCBs in pigment manufacturing processes, (2) concentrations and profiles of PCBs in commercial pigments and consumer products, (3) the emission routes and fate of pd-PCBs in the environment, (4) environmental levels and profiles of pd-PCBs with new data for developing countries, and (5) human exposure and potential adverse effects of pd-PCBs. General discussion and perspectives regarding academic and management aspects of pd-PCBs are also provided.

2. Unintentional formation of PCBs in pigment manufacturing processes

Pigments are generally classified into inorganic and organic pigments. Among inorganic pigments, the unintentional formation of

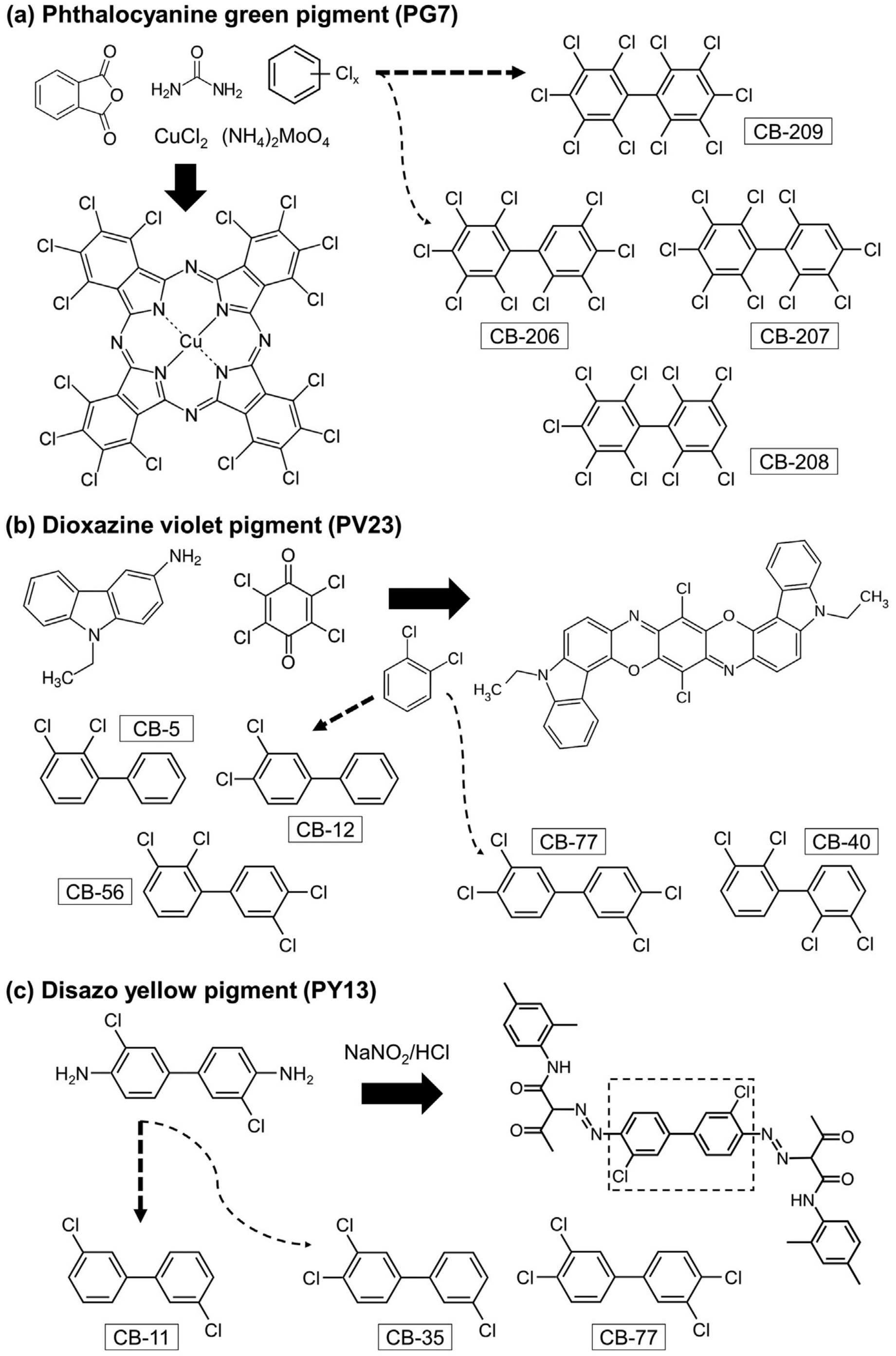
PCBs has been described for titanium dioxide (TiO_2) white pigments, mainly for those produced by the chloride process. This process comprises three steps: (1) production of titanium tetrachloride ($TiCl_4$) by treating ilmenite ore ($FeTiO_3$) at 1000 °C with carbon and chlorine gas, (2) distillation of $TiCl_4$, and (3) thermal oxidation of $TiCl_4$ to produce pure TiO_2 with the release of chlorine gas (Greenwood and Earnshaw, 1997). In the chloride process, PCBs are probably produced by a series of carbochlorination reactions, resulting in the prevalence of highly chlorinated congeners with 9 and 10 chlorine substitutions (Rowe et al., 2007; Du et al., 2008; Praipipat et al., 2013; Vorkamp, 2016). In addition, experiments based on carbochlorination reactions between chlorine and different bed materials indicated that decachlorobiphenyl (CB-209) is one of the most abundant by-products, especially in tests with metallurgical coke and graphite as carbon sources (Landsberg et al., 1985). Trace amounts of non-coplanar PCBs with highly chlorinated PCBs (e.g., CB-101, -138, -153, -180) were also preferably observed in a few TiO_2 nanoparticle samples, as compared to those of coplanar congeners and low-chlorinated compounds (e.g., CB-28 and CB-52) (Ctistis et al., 2016). Information about pd-PCBs in other types of inorganic pigments, for example, aluminum, bismuth, cadmium, chromate, cobalt, copper, gold, manganese, and zinc pigments, is still scarce.

The formation mechanisms of PCBs have been proposed for some typical groups of organic pigments such as azo, polycyclic, and dioxazine pigments (Hu and Hornbuckle, 2010; Anezaki and Nakano, 2014; Shang et al., 2014; Anezaki et al., 2015). It has not been fully characterized whether pd-PCBs are formed simultaneously with pigments in the same reaction medium containing chlorinated intermediates and solvents, or whether they are fragmented from the pigments. The former mechanism likely demonstrates the occurrence of PCBs in phthalocyanine pigments, which have been synthesized by the reaction between phthalic anhydride, urea, and copper or a copper salt with di- or trichlorobenzene reaction media (Uyeta et al., 1976; Buchta et al., 1985; Hu and Hornbuckle, 2010; Anezaki and Nakano, 2014; Anezaki et al., 2015). The formation of PCBs in phthalocyanine pigment manufacturing, especially those for green pigments, is largely attributed to free radical reactions between chlorobenzenes with catalysts copper chloride and ammonium molybdate (Liu et al., 2001; Liu et al., 2004; Hu and Hornbuckle, 2010). The perchlorination processes are responsible for the presence of highly chlorinated congeners (e.g., nona- and deca-CBs) in phthalocyanine green pigments (Hu and Hornbuckle, 2010; Anezaki and Nakano, 2014; Jahnke and Hornbuckle, 2019). Radical reactions with *o*-dichlorobenzene, a solvent used in dioxazine violet pigment synthesis, have resulted in structurally similar congeners as CB-5, CB-12, CB-40, CB-56, and CB-77 detected in the respective pigments (Anezaki et al., 2015). Other chlorinated benzidine, aniline, and benzonitrile intermediates could unintentionally produce PCBs in a variety of azo and polycyclic pigments of yellow, orange, and red shades (Hu and Hornbuckle, 2010; Anezaki and Nakano, 2014; Shang et al., 2014; Anezaki et al., 2015; Jahnke and Hornbuckle, 2019). Among the most predominant pd-PCBs in azo yellow pigments, CB-11 and CB-52 can be produced by the decomposition of diazo compounds originating from starters such as 3,3′-dichlorobenzidine and 2,2′,5,5′-tetrachlorobenzidine, respectively (Hu and Hornbuckle, 2010; Anezaki and Nakano, 2014). However, it should be noted that the above mechanisms were generally based on the structural similarity between major PCBs detected and products or starting materials of pigment manufacturing processes (Fig. 1). Therefore, further confirmation studies in the fields of organic chemistry and chemical technology are needed.

3. Occurrence of PCBs in commercial pigments and consumer products

PCBs were detected in various commercial pigments belonging to organic and inorganic types of yellow, orange, red, violet, green, blue, and white colors (Table 1). Total PCB concentrations ranged from a few ppb to approximately 0.1% by fresh weight of pigments. The highest PCB concentrations were found in some disazo yellow and orange pigments produced in Japan and China with chlorobenzidines as starting materials (Nakano et al., 2013; Anezaki and Nakano, 2014; Shang et al., 2014). Elevated PCB levels were also detected in several dioxazine violet, Naphthol AS red, diketopyrrolopyrrole red, monoazo yellow, and phthalocyanine green pigments, especially for those produced in Japan and China (Anezaki and Nakano, 2014; Shang et al., 2014; Anezaki et al., 2015). Concentrations of PCBs in pigments from the US and some European countries were within the lower range of few ppb to about 300 ppb (Hu and Hornbuckle, 2010; Anezaki et al., 2015; Jahnke and Hornbuckle, 2019). PCBs were not detected or detected at only minor levels in inorganic pigments such as titanium dioxide, iron oxide, and black carbon pigments (Hu and Hornbuckle, 2010; Stone, 2014; Ctistis et al., 2016). These studies revealed that >150 PCB congeners of all 10 homologs were detected at least once in commercial pigment samples. Among them, several "novel" compounds with trace levels in technical PCB mixtures were found as major congeners in some pigments, for example, CB-5, -11, -12/13, -35, -77, and -209. The congener-specific profiles of PCBs that depend on starting materials and pigment synthesis methods were assigned. In particular, disazo yellow pigments derived from 3,3′-dichlorobenzidine were characterized by the abundance of CB-11, -35, and -77, whereas highly chlorinated PCBs (e.g., CB-209, -208, and -206) dominated in phthalocyanine green pigments derived by perchlorination in chlorobenzene (Fig. 1 and Table 1). On the other hand, some legacy congeners such as CB-28, -52, -118, and -153 were also unintentionally produced in pigment manufacturing processes. CB-77 was the most predominant dioxin-like PCBs found in some pigments, followed by CB-126. As a result, toxic equivalents (WHO-TEQs) relative to 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) of dioxin-like PCBs were higher in such pigments with the highest value of 4200 pg WHO-TEQ g^{-1} measured in a Chinese monoazo yellow pigment (Shang et al., 2014).

Due to several applications of PCBs, these substances can be found in different commercial products in which PCB-containing materials were applied, for example, engine oils and lubricants, plastics, inks, paints and surface coatings, carbonless copy papers, papers and paper boards, adhesives, caulks and sealants, and so on (Kuratsune and Masuda, 1972; de Voogt et al., 1984; Jartun et al., 2009; Robson et al., 2010; Erickson and Kaley II, 2011). However, in this review, we mainly focus on the presence of unintentionally produced PCBs, which are associated with the applications of pigments. Rastogi (1992) detected PCBs in seven printing ink samples from Denmark (1.0–184.2 ng g^{-1} for di- to hexa-CBs) with the highest PCB levels found in green and brown inks. Significant concentrations of di-CBs as 78.9 and 15.5 ng g^{-1} were measured in a brown and a yellow ink, respectively, but no specific congeners were reported (Rastogi, 1992). Since then, some studies have focused on the determination of CB-11, a typical indicator of pigment impurities, in consumer goods (Rodenburg et al., 2010a; Guo et al., 2014; Stone, 2014, 2016; Liu and Mullin, 2019). About 350 consumer products of major categories such as paper, fabric, packaging, paint, caulk, and miscellaneous materials, mainly originated from the US, were tested and three fourths of them were contaminated with CB-11 at ppb levels. The highest CB-11 concentrations were found in pigmented samples, usually associated with yellow color, for example, a yellow foam office product, a single serving cereal packaging (2290 ng g^{-1}) and children's yellow sidewalk chalk (1060 ng g^{-1}) (Stone, 2016). Some other tri- to hexachlorinated PCB congeners (e.g., CB-52, -61/70/74/76, and -31) were also detected in consumer products, which were likely treated with azo red, orange, and yellow pigments (Guo et al., 2014; Stone, 2016). A few ppb of CB-209 was measured in some green and yellow spray paint samples (Stone, 2014). Liu and Mullin (2019) detected CB-85, -95, -118, -121, -138, -149, and -153 in a concentration range of about 60 to 140 ng g^{-1} in a fiberboard box sample. In general, the popular use of pigments has resulted in the widespread contamination of



(a) Phthalocyanine green pigment (PG7)

(b) Dioxazine violet pigment (PV23)

(c) Disazo yellow pigment (PY13)

Fig. 1. Potential formation pathways of unintentionally produced PCBs in selected organic pigments (Hu and Hornbuckle, 2010; Anezaki and Nakano, 2014; Anezaki et al., 2015a).

pigment-derived PCBs in consumer products. The most notable congener of this context is CB-11, a trace component of technical PCB mixtures, but it has been recognized as a predominant PCB in many commercial pigments and related products.

4. Emission pathways and environmental fate of pigment-derived PCBs

Environmental emissions of PCBs have been associated with three major processes: (1) production, utilization, and disposal of PCB-treated products; (2) unintentional emission from combustion processes; and (3) re-emission of PCBs from environmental reservoirs (e.g., soil, sediment, and water) (Breivik et al., 2002b). This scheme can also be applied for pd-PCBs with some modifications regarding the life cycle of pigments and pigmented products. PCBs in pigments can be emitted through several pathways such as evaporation, deposition, leaching, and discharging, which are related to domestic and industrial activities (Fig. 2). In addition, environmental behavior and fate of a particular PCB congener also depends on its physicochemical properties. In Table 2, we summarized the principal chemical and physical parameters of some representative PCBs identified as pigment-derived indicators (data mainly retrieved from Mackay et al., 2006 and Paasivirta and Sinkkonen, 2009). In this section, we focus on the emission pathways of some specific compounds (e.g., CB-11, -35, and -209)

Table 1
Concentrations (median and range, ng g^{-1}) and profiles of unintentionally produced PCBs in pigments.

Type	Sub-type	Color (color index name)	Country	Concentrations	Major compounds	References
Azo	Disazo	Yellow (PY81)	Japan	58,000 (8300–740,000)	CB-52, 11, 101, 153	Anezaki and Nakano, 2014
		Yellow (PY12, 13, 16, 17, 81)	China	4400 (980–919,000)	CB-11, 28, 52, 77	Shang et al., 2014
		Yellow (PY12, 13, 14, 17, 55, 83, 152)	Japan	1200 (330–7900)	CB-11, 35, 77	Anezaki and Nakano, 2014
		Yellow (PY97)	US	3.8	CB-52, 90/101/113	Jahnke and Hornbuckle, 2019
		Orange (PO13, 16)	Japan	9400 (340–17,000)	CB-11, 35, 77	Anezaki and Nakano, 2014
	Monoazo	Yellow (PY1, 3, 5, 65, 191)	China	360 (50–19,500)	CB-11, 28, 77, 52	Shang et al., 2014
		Yellow (PY154)	Japan	7.0	CB-11	Anezaki and Nakano, 2014
		Yellow	US	2.8–8.9	CB-4, 8, 11	Hu and Hornbuckle, 2010
		Yellow (PY74)	Japan	ND[a]	–	Anezaki and Nakano, 2014
		Orange (PO36)	Japan	130	CB-77, 35, 11	Anezaki and Nakano, 2014
	Naphthol AS	Red (PR112)	Japan, UK, the Netherlands	220 (4.4–3800)	CB-146, 149, 153, 118, 109, 101	Anezaki et al., 2015
		Red (PR9)	Japan	150 (4.2–430)	CB-52, 101, 31, 18, 26	Anezaki et al., 2015
		Red (PR22, 170, 188)	Japan	ND	–	Anezaki and Nakano, 2014
	Isoindolinone	Red	US	12	CB-15, 12, 8	Hu and Hornbuckle, 2010
Polycyclic	Phthalocyanine	Green (PG7, 36)	Japan	270 (11–2500)	CB-209, 208, 206, 11	Anezaki and Nakano, 2014
		Green	US	180 (94–200)	CB-209, 4, 6, 8, 11, 12	Hu and Hornbuckle, 2010
		Green (PG7)	US	280	CB-209, 206, 208, 6	Jahnke and Hornbuckle, 2019
		Blue	US	2.0–57	CB-5, 12/13, 56	Hu and Hornbuckle, 2010
		Blue (PB15:2)	US	0.69	CB-209, 26, 5	Jahnke and Hornbuckle, 2019
		Blue (PB15, 15:1, 15:3)	Japan	ND	–	Anezaki and Nakano, 2014
	Diketopyrrolo-pyrrole	Red (PR254)	Japan	330–2400	CB-15, 13, 6	Anezaki et al., 2015
		Red (PR254, 255)	UK, Italy, France, the Netherlands	20 (2.0–310)	CB-15, 13, 6	Anezaki et al., 2015
		Orange (PO73)	US	5.0	CB-3, 11, 15, 8	Jahnke and Hornbuckle, 2019
	Quinacridone	Red (PR122)	US	12	CB-12/13, 15, 8, 31	Jahnke and Hornbuckle, 2019
Dioxazine	Dioxazine	Violet (PV37)	Japan	29,000	CB-5, 12, 56, 77, 40	Anezaki et al., 2015
		Violet (PV23)	Japan	1500 (890–6500)	CB-5, 12, 56, 77, 40	Anezaki et al., 2015
		Violet (PV23)	UK, Italy, France	140 (50–1100)	CB-5, 12, 56, 77, 40	Anezaki et al., 2015
		Violet	US	12	CB-56, 5, 12, 77	Hu and Hornbuckle, 2010
Quinophthanone	Quinophthanone	Yellow (PY138)	China	84	CB-11, 28, 52, 77	Shang et al., 2014
Inorganic	Titanium dioxide	White	The Netherlands	2.7	CB-138, 180, 153, 101	Ctistis et al., 2016
		White	US	1.3	CB-209 detected only	Stone, 2014
		White	US	ND	–	Hu and Hornbuckle, 2010
	Iron oxide	Red, yellow	US	ND	–	Hu and Hornbuckle, 2010
	Carbon black	Black	US	ND	–	Hu and Hornbuckle, 2010

[a] Not detected (for more details on the detection limits and blank levels of selected pd-PCBs, see Table S1 of Supplementary data).

with an emphasis on their fate in the atmosphere, hydrosphere, and biosphere.

4.1. Atmosphere

Pigment-derived PCBs can evaporate from products containing them and pollute the air, especially for highly volatile compounds like CB-11 and some other mono- and di-CBs (e.g., CB-1, -2, -3, -4, and -8) (Hu et al., 2008; Hu and Hornbuckle, 2010; Rodenburg et al., 2015a; Shanahan et al., 2015; Zhao et al., 2020). This statement was initially supported by the relationship between air concentrations of CB-11 and ambient temperature (Hu et al., 2008; Anezaki and Nakano, 2014; Zhao et al., 2020). The air emission of CB-11 is thought to link with population density and human activities (Hu et al., 2008; Basu et al., 2009; Hu and Hornbuckle, 2010), implying the intensive application of architectural paints and pigmented products in residential area indoor micro-environments (Baek et al., 2010; Ampleman et al., 2015; Shanahan et al., 2015). The emission behavior of PCBs from paint pigments has been investigated recently (Jahnke and Hornbuckle, 2019). By using polyurethane foam passive emission samplers combined

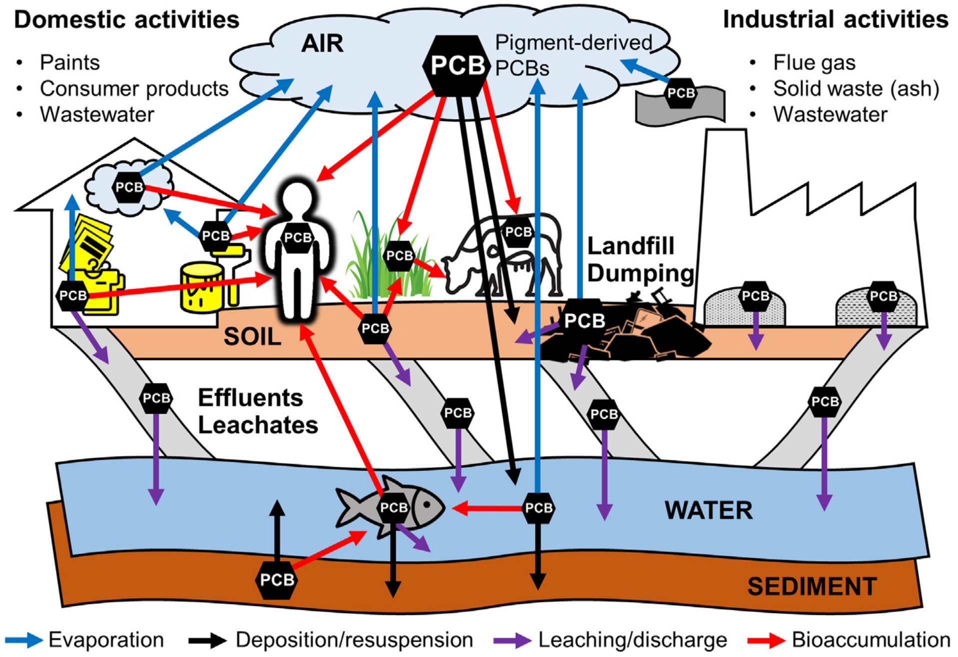


Fig. 2. Emission pathways and environmental fate of pigment-derived PCBs.

with a computational model, Jahnke and Hornbuckle (2019) observed that all PCBs detected in pigments were found in the diffusive materials (i.e., PUF disk) after 6 weeks, even for fully chlorinated CB-209. This study indicated that emission rate of mono- and di-CBs was higher than those of heavier congeners and they can completely effuse from the painted surface; meanwhile, excretion rate of the higher chlorinated congeners was only about 25% or less. Ambient moisture, temperature, wind speed, and air ventilation are probable influencing factors that affect indoor air concentrations of PCBs in painted rooms (Jahnke and Hornbuckle, 2019). Outdoor air likely receives pd-PCBs from indoor sources (Shanahan et al., 2015).

Other sources of atmospheric emission include: (1) re-emission from environmental reservoirs, notably surface water (Bamford et al., 2002; Martinez et al., 2010, 2) evaporation from landfill and open dumping sites; and (3) emission from combustion and waste-processing activities. Persson et al. (2005) reported that landfills containing PCB-treated polysulfide sealants can release PCBs to the air at very slow rates because of the high sorptive capacity of sealants. The emission of pd-PCBs, especially for low-chlorinated congeners, from landfill and open dumping sites, is estimated to be more significant and should be evaluated by future studies. Flue gas and ash from waste incinerators and combustion experiments also contained considerable amounts of several pd-PCBs such as CB-11, -35, -206, -207, -208, and -209 (Ikonomou et al., 2002; Kim et al., 2004; Ishikawa et al., 2007; Jansson et al., 2011). Therefore, the total atmospheric emission of pd-PCBs should account for every stage of pigmented products' life cycle including their production, application, and disposal. In the air, CB-11 is preferably partitioned in the gas phase, while CB-209 is distributed between gas and particulate phases in the same proportions (Wang et al., 2017). The emission of pd-PCBs to the atmosphere, especially for highly volatile congeners, could make them ubiquitous air pollutants, subject to long-range atmospheric transport (Choi et al., 2008; Hu et al., 2008; Baek et al., 2011; Li et al., 2012a, 2012b; Piazza et al., 2013; Wang et al., 2017; Hites, 2018; Bartlett et al., 2019). Also, atmospheric deposition can play an important role in the PCB contamination of aquatic and terrestrial environments. The role of atmospheric deposition (i.e., dry, wet, and gas deposition) as potential source of PCBs in water and soil has been demonstrated by their partitioning behavior between air-water (Totten et al., 2006; Hermanson et al., 2020) and air-soil interface (Tremolada et al., 2015; Cetin et al., 2017a, 2017b; Xu et al., 2019). The deposition tendency of PCBs on surface water and soil is regulated by meteorological conditions (e.g., temperature and precipitation), water and soil properties, and the nature of specific congeners (Totten et al., 2006; Tremolada et al., 2015; Cetin et al., 2017a, 2017b; Xu et al., 2019; Hermanson et al., 2020).

4.2. Hydrosphere

Wastewater discharges have been considered as principal sources of pd-PCBs to the aquatic environment (Litten et al., 2002; Du et al., 2008;

Table 2
Physicochemical properties of some pigment-derived PCBs (data retrieved from Mackay et al., 2006 and Paasivirta and Sinkkonen, 2009).

Parameter	CB-11	CB-35	CB-52	CB-77	CB-209
Chemical formula	$C_{12}H_8Cl_2$	$C_{12}H_7Cl_3$	$C_{12}H_6Cl_4$	$C_{12}H_6Cl_4$	$C_{12}Cl_{10}$
Molecular weight (g mol^{-1})	223.09	257.53	291.98	291.98	498.66
Boiling point (°C)	320	–	–	360	566.2
Melting point (°C)	29	87	87–89	182–184	305.8
Solubility in water (M)	1.59×10^{-6}	4.61×10^{-7}	5.24×10^{-8}	1.95×10^{-9}	1.49×10^{-11}
Vapor pressure (mmHg)	6.49×10^{-4}	–	4.94×10^{-4}	1.64×10^{-5}	1.06×10^{-7}
Henry's law constant (atm m^3 mol^{-1})	2.33×10^{-4}	–	2.00×10^{-4}	9.40×10^{-6}	–
Octanol/water partition coefficient ($\log K_{ow}$)	5.020	5.500	6.025	6.025	8.220
Octanol/air partition coefficient ($\log K_{oa}$)	8.02	8.85	8.47	9.70	13.36

Rodenburg et al., 2010a; Praipipat et al., 2013; Yao et al., 2014). Significant abundance of CB-11 (86%) and CB-35 (9%) as compared with other congeners was found in untreated discharges from a US pigment manufacturing plant (Litten et al., 2002). CB-11 accounted for 36% to 69% of total PCBs in wastewater samples from a Chinese dyeing industrial group (Yao et al., 2014). Wastewater treatment plant (WWTP) effluents, surface runoff, landfill leachates, and combined sewer flows are also responsible for the occurrence of pd-PCBs in the water bodies (Rodenburg et al., 2010a; Praipipat et al., 2013; Yao et al., 2014; Cao et al., 2019). Guo et al. (2014) documented that leaching fractions of CB-11 from printed materials ranged from 6% to 81% by a simple leach test, and they suspected that clothes washing could release CB-11 to wastewater. Landfill leachates can contain some low-chlorinated congeners such as CB-1 and CB-4 with implications of dechlorination (Rodenburg et al., 2010b), but these compounds were also found in pigments (Hu and Hornbuckle, 2010; Anezaki et al., 2015; Jahnke and Hornbuckle, 2019). In addition, the manufacturing of titanium tetrachloride is thought to produce pigments and by-products (e.g., ferric chloride, a potential coagulant used in WWTP) contaminated with CB-209, and then cause near-source pollution (Rowe et al., 2007; Du et al., 2008; Praipipat et al., 2013; Rodenburg and Ralston, 2017). The aquatic emission of pd-PCBs is estimated to be more critical if the wastewater is insufficiently treated or even directly discharged to the receiving water. When a mixture of pd-PCBs is released to the water column, the behavior of its components could vary greatly and be involved in several distribution processes such as air-water, dissolved phase-particulate phase, water-sediment, and water-biota equilibrium. Sediment beds are considered as important sinks for high-chlorinated PCBs like CB-209 and nona-CBs (Hu et al., 2011; Praipipat et al., 2013; Rodenburg and Ralston, 2017). Meanwhile, low-chlorinated congeners including CB-11 are likely dissolved in water and evaporated to the air, rather than absorbed on suspended particles and sediments (Bamford et al., 2002; King et al., 2002; Rowe et al., 2007; Martinez et al., 2010).

4.3. Biosphere

PCBs in general and pd-PCBs in particular can enter the biosphere and undergo bioaccumulation and biotransformation processes. Plant species receive PCBs from surrounding compartments such as air, soil, water, and sediment, and play a key role in transport and elimination of PCBs in the environment (Li et al., 2011; Wang et al., 2012; Yu et al., 2013; Chen et al., 2014; Zhu et al., 2015a). The accumulation of lowly chlorinated PCBs like CB-11 in plants is probably due to direct uptake of dissolved PCBs in soil pore water by roots (Yu et al., 2013) and atmospheric deposition on leaves (Zhu et al., 2015a). PCBs can be metabolized in plant cells, for example, to hydroxylated PCBs (OH-PCBs) via cytochrome P450 (CYP450) enzymes (Van Aken et al., 2010). However, plant PCB uptake, storage, and transformation vary greatly and are species- and congener-specific, and depend on soil characteristics (Li et al., 2011), which should be investigated by further experimental and field studies.

It is well known that the occurrence of PCBs in aquatic organisms results from bioconcentration/accumulation and biomagnification processes. The contributions of PCB intake sources such as water, sediment, and diet depend on congener properties, environmental conditions, and species' living and feeding habits. The top predators accumulate PCBs mainly from prey intake (i.e., biomagnification), whereas low-trophic-level species and benthic organisms receive these compounds from other sources such as water, sediment, and benthic invertebrates (King et al., 2002; Rodenburg et al., 2015b; Jafarabadi et al., 2019). As compared with heavier congeners, CB-11 is estimated to have lower bioaccumulation and biomagnification potential (Khairy et al., 2014; Rodenburg et al., 2015b; Pizzini et al., 2017), but this compound has dominated PCB patterns in biota elsewhere, probably due to its abundance in aquatic environments. The dominance of CB-11 in biota was observed in the Halifax Harbor, Canada (King et al., 2002); an urban river in Beijing, China (Yu et al., 2013); the Terra Nova Bay, Antarctica and the Adriatic Sea, Italy (Pizzini et al., 2017); and the Persian Gulf, Iran (Jafarabadi et al., 2019). Apart from the accumulated fractions, PCBs can be excreted via fecal egestion and gill membrane losses, or metabolized to several oxidation products (Brown, 1994; Paterson et al., 2007). Paterson et al. (2007) reported that the elimination of CB-18 (a dichlorinated congener) from PCB-dosed yellow perch (*Perca flavescens*) was observed, regardless of seasonal variation. Although experimental and field studies on bioaccumulation kinetics of CB-11 are still limited, it has been suggested that this compound and other low-chlorinated congeners are more readily excreted and/or metabolized as compared to heavier congeners (Paterson et al., 2007).

Terrestrial animals and humans are exposed to PCBs, including congeners derived from pigments, through multiple pathways such as inhalation, non-dietary ingestion and dermal contact (e.g., with soil, dust, and painted surface), and food. In general, diet is estimated to play a more important role in human exposure to PCBs as compared to inhalation, except for some low-chlorinated congeners (e.g., CB-8, -11, -18/30) (Ampleman et al., 2015). The occurrence of CB-11 and its hydroxylated and sulfated metabolites in human serum has been reported (Marek et al., 2013, 2014; Koh et al., 2015; Grimm et al., 2017; Sethi et al., 2019). Other pd-PCBs such as CB-35 and CB-209 were also detected in human serum (Koh et al., 2015). Exposure experiments on animals indicated that CB-11 is rapidly absorbed, metabolized, and substantially subject to biliary, fecal, and urinary excretions (Hu et al., 2014). After lung exposure in rats, CB-11 was absorbed (99.8%) and distributed to 41 body tissues within 12 min, then CB-11 and phase I metabolites (e.g., OH-CB-11) decayed to minimal levels within 25 min (Hu et al., 2014).

5. Environmental contamination status of pigment-derived PCBs

In this section, an overview of the available data from monitoring studies that focused on some specific PCB congeners derived from pigments (notably CB-11 and CB-209) is provided, alongside with their spatial and temporal trends. CB-11 has been detected in a variety of environmental compartments including air, soil, settled dust, water, snow, ice, sediment, plant, animal, and human samples from five continents (i.e., Antarctica, Asia, America, Europe, and Oceania) and the Arctic. In many cases, the abundance of CB-11 has surpassed that of legacy congeners that derive from technical PCB mixtures. These findings justify scientific attention on CB-11 as well as other unintentionally produced congeners in pigments. In several monitoring, source apportionment, and inventory studies on PCBs, the pigment-related factors taken into account together with factors of technical mixtures and other unintentional sources (Praipipat et al., 2013; Shanahan et al., 2015; Rodenburg et al., 2019; Zhao et al., 2020).

5.1. Air

The presence of CB-11 in the ambient air was first reported in a study conducted in Jeonju City, South Korea (mean 56, range 15–244 pg m^{-3}, accounting for 8% of total 209 PCBs; Kim et al., 2003). CB-11 was considered as an emerging, ubiquitous air pollutant in several metropolitan and industrial areas in the US since the middle 2000s at mean levels ranging from 3 to 24 pg m^{-3} and maximum values not exceeded 100 pg m^{-3} (Hu et al., 2008; Du et al., 2009; Hites, 2018). In air samples from the US, CB-11 accounted for about 5–10% or even higher percentages (e.g., 15–25%) of total PCBs (Hu et al., 2008; Du et al., 2009; Hites, 2018). Air concentrations of CB-11 in Japan (1.9–37 pg m^{-3}; Anezaki and Nakano, 2014) and Italy (4.9–14 pg m^{-3}; Gregoris et al., 2014) were generally lower than those measured in other countries like China (15–250 pg m^{-3}; Mao et al., 2019; Zhao et al., 2020), Vietnam (68–300 pg m^{-3}; Anh et al., 2020a), South Korea (11–244 pg m^{-3}; Kim et al., 2003; Baek et al., 2010), and Chuuk State of Micronesia

(14–134 pg m^{-3}; Baek et al., 2011). In these air samples, CB-11 contributed about 20–30% with maximum percentages >60% in total PCBs (Baek et al., 2010, 2011; Anh et al., 2020a; Zhao et al., 2020). Interestingly, CB-11 was frequently found in the air of the Polar regions at elevated concentrations (0.43–87 pg m^{-3}) and proportions (up to 85% of total PCBs) as compared with many mainland sites (Choi et al., 2008; Baek et al., 2011; Li et al., 2012a, 2012b; Piazza et al., 2013; Wang et al., 2017). Reasons for the occurrence of CB-11 in the Polar regions' air have not been completely clarified; both long-range atmospheric transport, global fractionation and cold condensation effects, revolatilization from secondary environmental sources, and local anthropogenic sources (e.g., research stations) were suggested (Choi et al., 2008; Piazza et al., 2013; Wang et al., 2017). A comparison of CB-11 and total PCB concentrations in the air is given in Fig. 3, but it is relatively difficult to depict a global spatial trend for CB-11 because of limited data and inconsistent sampling techniques. Nevertheless, CB-11 is estimated to be more abundant in the atmosphere of the tropics and the Southern Hemisphere (Baek et al., 2011).

Data suggest that CB-11 is more concentrated in indoor air than outdoor air (Baek et al., 2010; Ampleman et al., 2015; Marek et al., 2017), implying direct emission of this compound from painted surfaces and pigmented consumer products. However, information about indoor air concentration of CB-11 is very scarce. Baek et al. (2010) mentioned that CB-11 was measured in seven Korean classrooms at a range of 18 to 1819 (mean 521) pg m^{-3}. Average concentrations of CB-11 ranged from 90 to 400 pg m^{-3} in indoor air from homes and schools of East Chicago, Indiana and Columbus Junction, Iowa, in the US (Ampleman et al., 2015). Levels of CB-11 in US indoor school air (median 191; range 94–660 pg m^{-3}) were significantly higher than those recorded outdoors (6.0; 3.0–13 pg m^{-3}) (Marek et al., 2017). This compound was detected in some Vietnamese urban homes and an office (100–280 pg m^{-3}; Anh et al., 2020a). Similar ranges of CB-11 concentrations (median 110; mean ± SD 150 ± 130 pg m^{-3}) that surpassed those of indicator PCBs were found in indoor air from houses and offices in Brisbane and Canberra, Australia (Wang et al., 2019). Because CB-11 was among the most inhaled congeners for US urban mothers (Ampleman et al., 2015) and school children (Marek et al., 2017), additional investigations on its indoor air levels are necessary.

Declining temporal trends of atmospheric levels of technical PCBs have been observed elsewhere (Bruckmann et al., 2013; de la Torre et al., 2016; Wang et al., 2017; Hites, 2018; Hites and Holsen, 2019), but such observations are probably not true for pd-PCBs. Air concentrations of CB-11 did not change significantly during the period 2004–2015 at several sites near the Great Lakes (Hites, 2018). Relatively stable levels of CB-11 without clear time trend were also reported by some long-term monitoring studies conducted in Japan (Anezaki and Nakano, 2014), China (Mao et al., 2019), and the Antarctica (Wang et al., 2017). Previous studies showed that air concentrations of PCBs, especially for low-chlorinated congeners like CB-11, were positively correlated with ambient temperature, and therefore, were higher in hot seasons than cold seasons (Baek et al., 2010; Anezaki and Nakano, 2014; Zhao et al., 2020). In contrast, no distinct seasonal variation and temperature dependence of airborne CB-11 was detected in some remote areas, implying the role of long-range atmospheric transport as an important emission pathway of this congener in such areas (Wang et al., 2017; Mao et al., 2019).

Information about air concentration of CB-209 is relatively scarce. This compound was found in Antarctic air at only fg m^{-3} levels (Li et al., 2012a, 2012b; Piazza et al., 2013). The detection frequency of CB-209 in Chinese air samples was about 46% ($n = 167$) with concentrations ranging from <0.01 to 2.01 pg m^{-3} (Zhao et al., 2020). Kim et al. (2003) detected CB-209 in the air of Jeonju City, South Korea in a concentration range of 0.18–4.24 (mean 1.25) pg m^{-3}. Levels of CB-209 were generally lower than 0.1 pg m^{-3} in the air samples collected

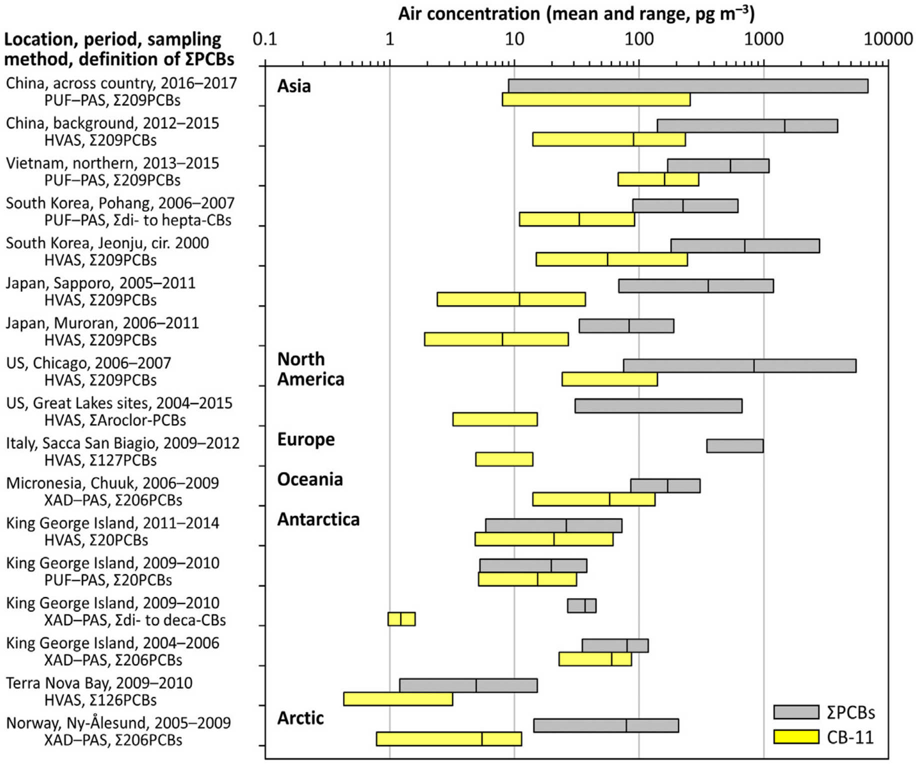


Fig. 3. Comparison of total PCBs and CB-11 concentrations in the air samples from different locations: China (Mao et al., 2019; Zhao et al., 2020), Vietnam (Anh et al., 2020a), South Korea (Kim et al., 2003; Baek et al., 2010), Japan (Anezaki and Nakano, 2014), the US (Hu et al., 2008; Hites, 2018), Italy (Gregoris et al., 2014), Micronesia (Baek et al., 2011), and Polar regions (Choi et al., 2008; Baek et al., 2011; Li et al., 2012a, 2012b; Piazza et al., 2013; Wang et al., 2017). High volume active air sampling (HVAS) and passive air sampling based on polyurethane foam (PUF–PAS) and XAD-2 resin (XAD–PAS) methods were applied.

from Sapporo and Muroran, Japan (Anezaki and Nakano, 2014). Concentrations of CB-209 ranged from 0.11 to 13 pg m^{-3} in the air from heavily urbanized and industrialized areas in Turkey (Cetin et al., 2017a, 2017b). This congener was found in the air around a few Vietnamese end-of-life vehicle dismantling workshops at quite low concentrations (0.12–0.19 pg m^{-3}; Anh et al., 2020a). In almost all the detection cases described above, the proportion of CB-209 in total PCBs was <0.2%. Levels of CB-209 in the air were estimated to be stable and less temperature-dependent (Anezaki and Nakano, 2014). CB-35 and nona-CBs were also frequently detected in the air elsewhere, but their concentrations were relatively low (Kim et al., 2003; Marek et al., 2017; Zhao et al., 2020).

5.2. Soil and dust

CB-11 and CB-209 were found in soil and dust samples, and their concentrations were summarized in Fig. 4 in the comparison with total PCBs. Concentrations of CB-11 in the mainland soils from China (mean 0.60 ng g^{-1} for a composite sample; Yu et al., 2013) and central Vietnam (mean 0.38; range n.d. – 1.3 ng g^{-1}; Romano et al., 2013), and in road dusts from northern Vietnam (0.49; 0.059–1.5 ng g^{-1}; Anh et al., 2019) were higher than those measured in remote areas such as the Antarctica (0.042; 0.019–0.068 ng g^{-1}; Wang et al., 2012) and the Arctic (0.12; 0.070–0.17 ng g^{-1}; Zhu et al., 2015a). In soil samples from the Polar regions, CB-11 accounted for about 10% of total PCBs (Wang et al., 2012; Vecchiato et al., 2015; Zhu et al., 2015a). This compound was also found to be the most important contributor to total PCBs in several Chinese and Vietnamese soil and road dust samples with average percentages ranged from 7% to 19% and maximum values ranging from 15% to 85% (Romano et al., 2013; Yu et al., 2013; Anh et al., 2019). The occurrence of CB-11 in these soil and road dust samples has been likely associated with nearby pigment-related sources and/or long-range transport. A few recent studies have reported the presence of CB-11 in house and workplace dusts from Australia (median 5.6; mean ± SD 6.4 ± 2.5 ng g^{-1}; Wang et al., 2019) and Vietnam (range 0.46–8.7 ng g^{-1}; Anh et al., 2020b).

Levels and proportions of CB-209 were quite minor in almost all the soil and dust samples where it was detected (Wang et al., 2012; Yu et al., 2013; Zhu et al., 2015a; Cetin et al., 2017a; Anh et al., 2019, 2020b), except for two studies (Tremolada et al., 2015; Xu et al., 2020). The abundance of CB-209 in soil was reported for a high-altitude pasture area in the Italian Alps (mean 0.15; range 0.031–0.36 ng g^{-1}), which accounted for 8% to 17% (average 12%) of Σ14PCBs (Tremolada et al., 2015). Several potential sources of CB-209 in this mountain area were suggested, including legacy production of PCB mixtures containing CB-209, incinerators, metal industry plants, and long-range transport of pigment-derived CB-209 (Tremolada et al., 2015). CB-209 was detected in soils from an industrial park in Ningxia, northwestern China at a concentration range of 0.010–0.104 ng g^{-1}, accounting for 28% on average of Σ19PCBs (Xu et al., 2020). Xu et al. (2020) implied that CB-209 originated from a recycled paper mill located in that industrial park, which may use phthalocyanine pigments as paper dye.

5.3. Water, snow, and ice

CB-11 has widely been detected in wastewater and river water samples since the late 1990s (Fig. 5). Concentrations of CB-11 ranged from 6.4 to 44 pg L^{-1} and from 36 to 110 pg L^{-1} in suspended particulate and solution phase, respectively, and this congener accounted for 0.7% to 10% in particulate phase and 5.0% to 45% in solution phase of total 159 PCBs detected in water samples from Bedford Basin of Halifax Harbor, Canada (King et al., 2002). At that time, it was estimated that potential sources of CB-11 may relate to 3,3′-dichlorobenzidine, a precursor of pigment manufacturing and a curing agent for resins, imported to Canada, and intermittent discharges; but no clear evidence was provided (King et al., 2002). Shortly thereafter, Litten et al. (2002) demonstrated that the ubiquitous occurrence of CB-11 and some other congeners like CB-35 and CB-77 in water samples from New York/New Jersey (NY/NJ) Harbor, the US, was associated with pigment manufacturing. Levels of CB-11 drastically decreased in pigment discharges (3600 ng L^{-1}) > WWTP influents (490 ng L^{-1}) > effluents in collecting trunks (0.07–0.6 ng L^{-1}) (Litten et al., 2002). In the early 2000s, concentrations of CB-11 in WWTP effluents in NY/NJ Harbor varied over a wide range of 1.6 to 116,000 pg L^{-1}, and in Hudson River water ranged from 16 to 2100 pg L^{-1} (Rodenburg et al., 2010a). Furthermore, the aquatic emissions of CB-11 and other pd-PCBs have

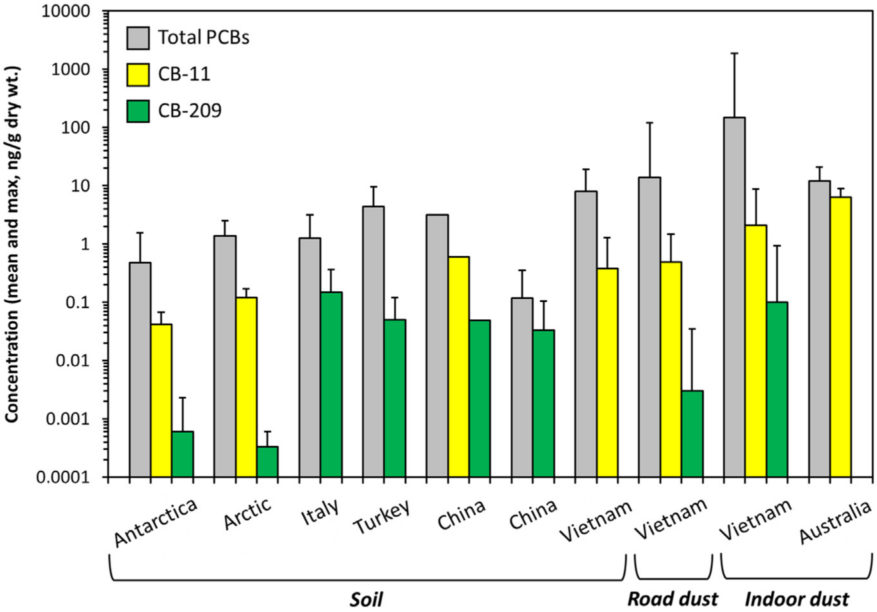


Fig. 4. Comparison of total PCBs, CB-11, and CB-209 concentrations in soil and dust. Data (from left to right) were retrieved for Antarctica (Flides Peninsula, 2009–2010, Σ209PCBs, Wang et al., 2012); Arctic (Svalbard, 2011–2012, Σ25PCBs, Zhu et al., 2015a); Italy (high-altitude area of Italian Alps, 2011, Σ14PCBs, Tremolada et al., 2015); Turkey (metropolitan area of Istanbul, 2014, Σ41PCBs, Cetin et al., 2017a); China (riparian zone in Beijing, 2010, Σ26PCBs, Yu et al., 2013; and an industrial area in Ningxia, 2017, Σ14PCBs, Xu et al., 2020); and Vietnam (soil from Thi Nai Lagoon, central Vietnam, 2010, Σ92PCBs, Romano et al., 2013; road dust from northern Vietnam, 2016, Σ209PCBs, Anh et al., 2019; indoor dust from northern Vietnam, 2015–2016, Σ209PCBs, Anh et al., 2020b; and indoor dust from Brisbane and Canberra, Australia, 2015, Σ8PCBs, Wang et al., 2019).

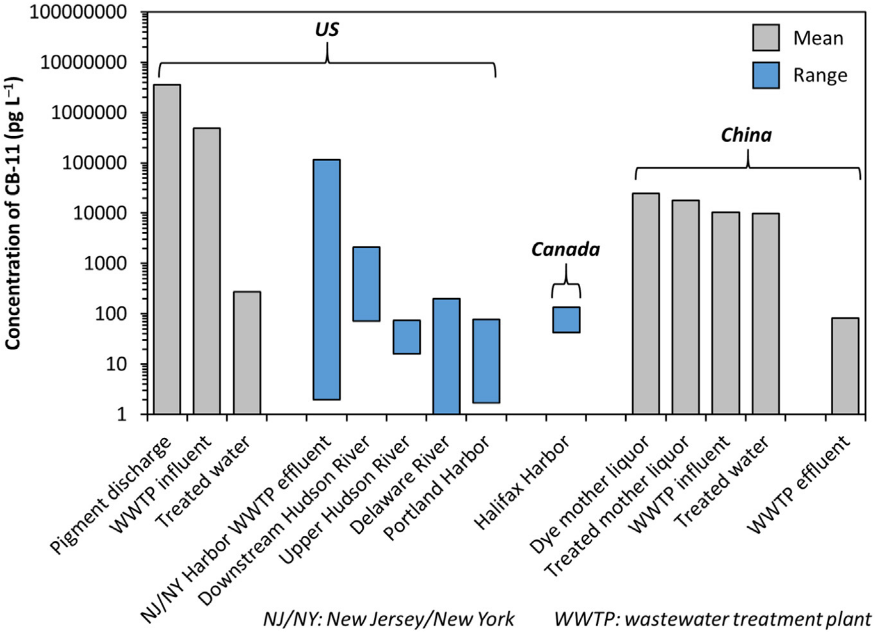


Fig. 5. Comparison of CB-11 concentrations in wastewater and river water. Data were retrieved for the US (Litten et al., 2002; Rodenburg et al., 2010a, 2015c), Canada (King et al., 2002), and China (Yu et al., 2013; Yao et al., 2014).

been characterized for several areas in the US such as the Delaware River (Du et al., 2008), urban channels in Houston, Texas (Howell et al., 2011), the lower Great Lakes (Khairy et al., 2015), and Portland Harbor, Oregon (Rodenburg et al., 2015c). The occurrence of CB-209 in water that are mainly associated with suspended particulates has been reported by a few studies conducted in the Delaware River with implications for a titanium dioxide purification plant (Rowe et al., 2007), and the Houston Ship Channel in Texas with several possible sources such as Aroclor 1268 mixtures, pigments, and atmospheric deposition (Howell et al., 2008).

Information about levels and sources of CB-11 and pd-PCBs in water from other locations is relatively scarce. Freely dissolved CB-11 and CB-209 were detected in a WWTP effluent sample from a river in Beijing, China at concentrations of 81 and 1.4 pg L^{-1}, accounting for about 47% and 1% of Σ26PCBs, respectively (Yu et al., 2013). Yao et al. (2014) measured CB-11 (average 9500–25,000 pg L^{-1}), nona-CBs (32–67 pg L^{-1}), and CB-209 (8.2–27 pg L^{-1}) in water samples from a WWTP of a Chinese dyeing industrial group, and levels of these compounds did not change considerably over different treatment stages. CB-11 was frequently detected in surface water from the Bay of Bengal coast, Bangladesh and contributed to 0.4–2% of Σ209PCBs, but nona- and deca-CBs were not detected (Habibullah-Al-Mamun et al., 2019). Concentrations of Σ127PCBs in lake water samples from Northern Victoria Land, Antarctica ranged from 46 to 143 pg L^{-1} with CB-11 as the most abundant congener (6–19%) (Vecchiato et al., 2015). Full congener-specific profiles of PCBs were also examined in surface snow and ice core samples from the Arctic (Garmash et al., 2013; Bartlett et al., 2019; Hermanson et al., 2020). Such studies have emphasized the occurrence and temporal trends of three congeners as CB-5, CB-11, and CB-52 in surface snow samples at some glacier sites on Svalbard, and indicated increasing fluxes and proportions of CB-5 and CB-52 over the 2010–2014 period (Bartlett et al., 2019; Hermanson et al., 2020). An Arctic ice core from Lomonosovfonna exhibited an increase in CB-11 fluxes during 1974–2009 with its close relationship with coeluting congeners CB-12/13 (Garmash et al., 2013). Besides commercially produced amounts of CB-5 and CB-52 in some German and Russian products, the pigment-related sources of these congeners (including CB-11) were also suggested (Garmash et al., 2013; Bartlett et al., 2019; Hermanson et al., 2020).

5.4. Sediment and sludge

The first notification of pd-PCBs in sediments dates back to 1985 when Law (1995) mentioned contamination of lowly chlorinated PCBs (e.g., CB-11 and CB-36) in downstream sediments of a dye production plant in northern France. This information, however, was cited as the author's personal communication without detailed data (Law, 1995). King et al. (2002) detected CB-11 in two out of twelve sediment samples collected from Bedford Basin, Canada in 1996 at concentrations of 18.7 and 2.6 ng g^{-1}, which accounted for 10.4% and 7.9% of total PCBs, respectively, and suggested that this compound was likely associated with pigments rather than produced by dechlorination of heavier congeners. CB-11 was found as the most predominant congener in sludge samples from a Chinese WWTP with mean concentrations ranging from 1100 to 10,000 ng g^{-1}, accounting for about 70% of total PCBs (Yao et al., 2014). Several studies on PCB contamination in sediments have been conducted over the last decade with special consideration given to pigment-related sources. Furthermore, the input history of these PCB congeners has been reconstructed by analyzing sediment core samples. A comprehensive picture on the spatial distribution of sedimentary PCBs and typical pigment impurities such as CB-11 and CB-209 is shown in Table 3.

Concentrations of CB-11 in sediment samples from the US were generally higher than those detected in other parts of the world. Elevated levels of CB-11 found in sediments from some US specific areas were associated with pigment production and related industries, and relevant input of wastewater and stormwater (Hu et al., 2011; Mohrherr et al., 2012; Praipipat et al., 2013; Rodenburg and Ralston, 2017; Cao et al., 2019). However, proportions of CB-11 in total sediment PCBs at many US sites (e.g., the Great Lakes, Delaware River, NY/NJ Harbor, and Portland Harbor) were relatively low; and this fact can be explained by two reasons: (1) high mobility of CB-11 resulted in its significant freely dissolved and evaporated fractions, and (2) the abundance of Aroclor factors (Hu et al., 2011; Praipipat et al., 2013; Rodenburg et al., 2015c; Rodenburg and Ralston, 2017). Despite that, high percentages of CB-11 in total PCBs were also recorded in surface sediments from Escambia Bay, Florida (4.8% by mole; Mohrherr et al., 2012) and stormwater sediments from multi-functional areas in Laurel, Maryland (1.3–30.9% by weight; Cao et al., 2019). By analyzing sediment core

Table 3
Concentrations (median/mean and range, ng g^{-1}) of PCBs in sediment samples from different locations, with a focus on CB-11 and CB-209.

Location	Year[a]	Sediment type	Total PCBs; N[b]	CB-11 and/or CB-209; % of total PCBs	References
US, New York/New Jersey Harbor	2000–2001	Freshwater, surface and core	All samples 2.3–36,100; surface 28.5–36,100; 132	CB-11: surface 0.22–150; average 0.7%, maximum 6%	Rodenburg and Ralston, 2017
US, Escambia Bay	cir. 2010	Coastal, surface	17,900 (900–125,900); 209	CB-11: 4.8% by mole	Mohrherr et al., 2012
US, Maryland, Laurel	2016–2018	Stormwater, surface	29.2 (8.3–57.4); 209	CB-11: 0.33–6.58; 1.3–30.9%	Cao et al., 2019
Arabian Gulf, Sulaibikhat Bay	cir. 2010	Coastal, core	0.88–15; 26	CB-209: 0.0005–0.006; minor percentages	Gevao et al., 2012
Morocco, Nador Lagoon	2009	Coastal, surface and core	2.5–21; 127	CB-11: not detected (ND) – 1.7; 0–34% CB-209: ND	Giuliani et al., 2015
Italy, western Adriatic Sea	2013	Coastal, surface and core	Surface 0.1–2.2; core ND – 9.0; 28	CB-209: surface ND – 0.2; core ND – 1.8; among the most important congeners	Combi et al., 2017
China, Lake Chaohu	2009	Freshwater, surface	12.3 (2.0–42.7); 28	CB-209: 2.1 (ND – 9.4); 8 (0–77)%	Wang et al., 2014
China, Lake Chaohu	2011	Freshwater, core	0.03–24.11; 41	CB-209: 0.001–18.73; 72.2 (45.5–83.9)% between the late 1950s and 2006	Huo et al., 2017
China, a river in Beijing	2010	Freshwater, surface	2.68–5.40; 26	CB-11: 1.09–1.12; 21–40% CB-209: 0.071–0.074; 1.4–2.6%	Yu et al., 2013
Vietnam, Thi Nai Lagoon	2010	Freshwater, surface	2.0 (0.47–6.4); 92	CB-11: 0.25 (ND – 0.61); 10 (0–17)%	Romano et al., 2013
Japan, off Tohoku coast	2012	Coastal, surface	0.86 (0.18–4.0); 209	CB-209: 0.006 (0.002–0.81); 0.9 (0.3–43)%	Ohkouchi et al., 2016
Japan, Muroran Port	2008	Coastal, surface	29 (0.56–65); 209	CB-11: 0.11 (0.050–0.34); 1.1 (0.2–11)% CB-209: 0.005 (0.002–0.045); 0.05 (0.01–0.3)%	Anezaki et al., 2016
Japan, Beppu Bay	2018	Coastal, core	15 (3.5–150); 209	CB-11: 0.17 (0.029–0.46); 0.1–3.9% CB-209: 0.018 (ND – 0.046); 0–0.6%	Takahashi et al., 2020
South Korea, Nakdong River	1999	Freshwater, surface	3.8 (1.1–141); 43	CB-209: 0.030 (ND–0.17); 0.6 (0–3.1)%	Jeong et al., 2001
Korea Strait	2010	Coastal, core	0.408 (0.045–1.01); 209	CB-209: 0.0008 (0.00003–0.0021); 0.2 (0.1–0.8)%	Guerra et al., 2019
Turkey, Izmit Bay	2005	Coastal, core	16 (5.4–29); 127	CB-11: 0.43 (0.31–1.7); 3.1 (1.6–8.2)%	Giuliani et al., 2017
Antarctica, Great Wall Bay	2009–2010	Coastal, surface	0.182; 209	CB-11: 0.0051; 2.8% CB-209: 0.00017; 0.1%	Wang et al., 2012
Antarctica, Terra Nova Bay	2011–2012	Coastal lake, surface	0.010–0.634; 127	CB-11 was among the most abundant congeners, accounting for up to 12%.	Vecchiato et al., 2015

[a] Sampling years (did not represent sediment deposition years).
[b] Number of analyzed PCB congeners.

samples, Hu et al. (2011) indicated that CB-11 had a temporal trend correlated with the US pigment/dye production history and concentration peaks of about 5 years after total PCB peaks in the early 1970s. Rodenburg and Ralston (2017) measured CB-11 in deep sediment layers corresponding to the 1930s and found its peaks in the early 1970s, which were consistent with those of total PCBs and Aroclor production history. Interestingly, CB-11 was detected even in deeper layers that deposited before the 1930s, implying its non-Aroclor sources and/or other effects (e.g., bioturbation, core extrusion smearing, and selective diffusion) (Hu et al., 2011). However, due to the high mobility of lowly chlorinated congeners, the selective diffusion (if occurred) of CB-11 in sediment cores may lead to its movement and accumulation in shallower depths rather than deeper segments, as similar to those observed by Gevao et al. (1997) for tri-CBs. Concentrations of CB-11 varied over a narrow range (from lower than detection limits to few ppb) in sediment samples from China (Yu et al., 2013), Japan (Anezaki et al., 2016; Takahashi et al., 2020), Morocco (Giuliani et al., 2015), Turkey (Giuliani et al., 2017), and Vietnam (Romano et al., 2013), and were markedly higher than those found in Antarctic samples at ppt levels (Wang et al., 2012; Vecchiato et al., 2015).

Highly chlorinated PCBs such as CB-209 and nona-CBs (i.e., CB-206, -207, and -208) have been monitored in sediments, but their concentrations and proportions in total PCBs were quite minor in the samples collected from the Korea Strait (Guerra et al., 2019), Beppu Bay in Japan (Takahashi et al., 2020), Nador Lagoon in Morocco (Giuliani et al., 2015), Thi Nai Lagoon in Vietnam (Romano et al., 2013), Adriatic Sea (Combi et al., 2017), Arabian Gulf (Gevao et al., 2012), and Antarctica (Wang et al., 2012; Vecchiato et al., 2015). In the US, some studies on source apportionment of PCBs in aquatic environments identified a novel factor comprising elevated proportions of CB-209 and nona-CBs (hereinafter referred to as deca factor), which was likely attributed to local production activities of titanium compounds (Rowe et al., 2007; Du et al., 2008; Rodenburg et al., 2010b). As a result, the deca factor has also been resolved in the sediment beds from some US locations such as Lake Ontario (Hu et al., 2011), NY/NJ Harbor (accounting for only 2% of PCB mass; Rodenburg and Ralston, 2017), and especially in the Delaware River that may be affected by a titanium tetrachloride production plant near Wilmington, Delaware (accounting for 61% of PCB mass; Praipipat et al., 2013). Hu et al. (2011) observed the association between historical deposition of CB-209 and nona-CBs in Lake Ontario sediment and pigment and dye production trends in the US, and the presence of these congeners in sediment layers deposited before the period of Aroclor production in Lake Erie and the Indiana Harbor ship canal. The dominance of CB-209 was recorded in sediment samples collected from Lake Chaohu, a shallow freshwater lake in eastern China, especially in the period between the 1950s and 2000s (Wang et al., 2014; Huo et al., 2017). Potential sources of CB-209 in this lake were assigned to improper discharges from many dye workshops utilizing phthalocyanine pigments located in Hefei City (Huo et al., 2017) or from titanium dioxide purification plants located in its basin (Wang et al., 2014). CB-209 accounted for about 1% (median, $n = 19$) of total PCBs found in 0–5 cm sediment layers from the Pacific coast of Tohoku, Japan with two outliers of 43% and 13%, but no detailed comments were provided (Ohkouchi et al., 2016). CB-209 was identified as an important congener in a sediment core from the western Adriatic Sea, Italy, which contributed up to 40% of Σ28PCBs (Combi et al., 2017). The elaborate congener-specific measurement of PCBs in surface and core sediments can provide relevant information about the spatial and temporal trends of novel compounds such as CB-11 and CB-209, which have usually been omitted from routine analysis. Several local contamination sources of these emerging pollutants were reported, and most of them are thought to associate with pigment manufacturing and disposal processes.

5.5. Plants

The occurrence of pd-PCBs has been observed in several plant species collected from China (Yu et al., 2013; Chen et al., 2014), the Antarctic (Wang et al., 2012), and the Arctic (Zhu et al., 2015a) (Table 4). It is relatively difficult to clarify the difference in total PCB concentrations between plant samples from China and the Polar regions because of inconsistent target compounds and studied species. Yu et al. (2013) detected CB-11 in stem-leaf and root of common reed from a riparian area in Beijing, China at concentrations of 350–660 pg g^{-1} (or 10–28 ng g^{-1} lipid wt.), accounting for about 40% of Σ26PCBs. CB-11 was measured in all plant samples from Fildes Peninsula, the Antarctica and Ny-Ålesund and London Island, the Arctic with concentrations ranged from 11.6 to 258 pg g^{-1} dry wt. (Wang et al., 2012; Zhu et al., 2015a). CB-11 accounted for about 20% of Σ209PCBs in the Antarctic moss and lichen samples (Wang et al., 2012), and 10–30% of Σ25PCBs in six Arctic plant species (Zhu et al., 2015a). Arctic reindeer dung samples were also examined and CB-11 was found at similar concentrations and proportions as plant samples, implying moss and lichen as the main food of reindeers (Zhu et al., 2015a). Di-CBs were also detected in pine and eucalyptus samples from e-waste and rural sites in Qingyuan, southern China (mean values 1500–25,800; maximum values 8250–89,200 pg g^{-1}), however, no congener-specific profile of this minor homolog was described (Chen et al., 2014).

CB-209 was frequently detected in plants at much lower concentrations than those of CB-11 with maximum levels not exceeded 20 pg g^{-1} dry wt. and percentages of about 1% or less (Wang et al., 2012; Yu et al., 2013; Zhu et al., 2015a). Considerable concentrations of CB-209 (mean values 450–3130; maximum values 1110–12,900 pg g^{-1}) were recorded in Masson's pine needle and eucalyptus leaf samples from Qingyuan e-waste and rural sites, but this congener exhibited quite small proportions of 0.5% or less in total PCBs (Chen et al., 2014). The lower accumulation degree of CB-209 as compared to lightly chlorinated homologs is explained by its strong adsorption to soil and low availability for plant uptake (Pier et al., 2002; Zhu et al., 2015a).

Plant–soil bioaccumulation factors (BAFs) greater than unity were observed for CB-11 and CB-209 in the Antarctic lichen and moss (Wang et al., 2012); while BAFs <1 were estimated for PCBs in common reed (Yu et al., 2013), and unclear PCB bioaccumulation from soil was seen in the Arctic plants (Zhu et al., 2015a). A previous experimental study indicated that phytoextraction played insignificant role of PCB decontamination in soil, even for a low-chlorinated congener such as CB-5 (Li et al., 2011). These observations suggest a weak association between PCB contamination levels in plant and soil, as compared to those among plants, air, and water (Yu et al., 2013; Zhu et al., 2015a). Available information about the accumulation, transportation, and transformation of pd-PCBs in plants is still limited, suggesting the need for further experimental and field studies on these special congeners in a wider range of species and locations.

5.6. Animals

The first data available for CB-11 in biota was reported by Bush et al. (1989), showing a PCB chromatogram of a striped bass (*Morone saxatilis*) sample collected from eastern Long Island, New York, the US in 1985 with a very tiny and unquantified peak of CB-11. Concentrations of CB-11 and CB-35 in animal tissues were initially reported for blubber, milk, and plasma samples from six grey seal (*Halichoerus grypus*) mother and pup couples collected on Sable Island, Canada in 1995 (Addison et al., 1999). CB-11 was frequently found in all tissue types, with the highest concentrations measured in plasma samples (3590 ± 1920 pg g^{-1} lipid wt.). However, its levels were much lower than those of other mono-*ortho* PCBs. CB-35 was detected in only one or two samples of each tissue with low concentrations, and was also preferably enriched in blood. Addison et al. (1999) did not provide any discussion about these congeners (i.e., CB-11 and CB-35) because

Table 4
Concentrations (mean and range, pg g^{-1}) of PCBs in plant samples, with a focus on CB-11 and CB-209.

Location (sampling year)	Species	Total PCBs; N[a]	CB-11 and/or CB-209; % of total PCBs	References
China, Qingyuan e-waste site (2007–2008)	Masson pine (*Pinus massoniana*)	670,000 (27,500–1,993,000); 171	CB-209: 3130 (ND[b] – 12,900); average 0.5%	Chen et al., 2014
	Eucalyptus (*Eucalyptus* spp.)	369,000 (88,000–635,000); 171	CB-209: 1210 (ND – 2710); 0.3%	
China, Qingyuan rural site (2007–2008)	Masson pine (*Pinus massoniana*)	105,000 (27,100–191,000); 171	CB-209: 450 (ND – 1110); 0.4%	Chen et al., 2014
	Eucalyptus (*Eucalyptus* spp.)	187,000 (25,400–775,000); 171	CB-209: 470 (ND – 1560); 0.3%	
China, a riparian area in Beijing (2010)	Common reed	1100 (700–1600); 26	CB-11: 490 (350–660); 36–57% CB-209: 7.8 (3.6–13); 0.3–1.2%	Yu et al., 2013
Antarctica, Fildes Peninsula (2009–2010)	Moss (*Sanionia uncinata*)	670 (406–952); 209	CB-11: 131 (27.5–258); 20% CB-209: 2.23 (ND – 5.68); 0.3%	Wang et al., 2012
	Lichen (*Usnea aurantiacoatra*)	544 (404–745); 209	CB-11: 95.4 (45.7–171); 18% CB-209: 3.22 (0.11–16.2); 0.6%	
Arctic, Ny-Ålesund and London Island (2011–2012)	Moss (*Drepanocladus aduncus*)	750 (430–1160); 25	CB-11: 98.3 (69.4–147); 13% CB-209: 8.8 (3.1–19.1); 1%	Zhu et al., 2015a
	Arctic bell-heather (*Cassiope tetragona*)	570 (480–630); 25	CB-11: 47.1 (11.6–102); 8% CB-209: 2.6 (2.3–3.1); 0.5%	
	Mountain avens (*Dryas octopetala*)	490 (420–580); 25	CB-11: 122 (86.7–191); 25% CB-209: 4.6 (3.3–6.6); 1%	
	Sooty sedge (*Carex misandra*)	450 (300–550); 25	CB-11: 25.0 (19.8–31.4); 6% CB-209: 9.2 (4.1–17.2); 2%	
	Alpine hair grass (*Deschampsia alpine*)	410 (370–460); 25	CB-11: 128 (121–135); 31% CB-209: 5.6 (3.3–8.0); 1%	
	Tufted saxifrage (*Saxifraga cespitosa*)	370; 25	CB-11: 79.3; 21% CB-209: 4.0; 1%	

[a] Number of analyzed PCB congeners.
[b] Not detected.

of their minor contributions to total non- and mono-*ortho* PCBs (<1%). The bioaccumulation potential of CB-11 has received scientific attention since King et al. (2002) found elevated concentrations of CB-11 in mussels (*Mytilus edulis*) collected from Halifax Harbor, Canada in 1997–1999. Levels of CB-11 in these mussel samples ranged from 700 to 9100 (median 2700) pg g^{-1} wet wt., comprising 0.6–46% (median 15%) of total PCBs (King et al., 2002). CB-11 was also measured in digestive glands of lobsters (*Homarus americanus*) collected in 1995 from the same harbor at concentrations ranged from <2000 to 20,000 pg g^{-1} wet wt., accounting for up to 1.8% of total PCBs (King et al., 2002). The accumulation of CB-11 in aquatic and terrestrial animals has been investigated by several studies conducted in the last decade; a summary is given in Table 5.

As shown in Table 5, concentrations of PCBs in general and CB-11 in particular in biological samples varied greatly over several orders of magnitude, and were site- and species-specific. Bivalve species can accumulate elevated amounts of CB-11 at ppb levels (accounting for about 15% of total PCBs), directly from the water rather than sediments (King et al., 2002; Pizzini et al., 2017). On the Mediterranean coast, the highest concentrations of CB-11 were found in mussel samples collected from heavily industrialized and high-traffic areas in Venice Lagoon and the Gulf of Trieste; whereas a seasonal variation of CB-11 was observed in clam samples collected from the Po River, with higher levels found in rainy season, implying the roles of runoff water and sediment resuspension as potential sources of CB-11 (Pizzini et al., 2017). CB-11 was also detected in various aquatic and terrestrial invertebrate species; however, no significant bioaccumulation from sediment, soil, or water was noted for this congener (Yu et al., 2013; Jafarabadi et al., 2018). Yu et al. (2013) measured CB-11 in butterfly, chironomid, sludge worm, cicada, dragonfly, grasshopper, scarabaeidae larva, and earth worm samples collected from a riparian area in Beijing, China at concentrations of 1300–10,000 pg g^{-1} lipid wt., which accounted for 6–47% of Σ26PCBs. CB-11 was also identified as dominant congener (contributing to about 11% of Σ22PCBs) in coral samples in Kharg and Lark coral reefs of the Persian Gulf, Iran (Jafarabadi et al., 2018). Previous studies have reported the widespread occurrence of CB-11 in many aquatic and amphibious species such as fish, crab, eel, snail, frog, and toad in the US and China (Khairy et al., 2014; Rodenburg et al., 2015b; Zhu et al., 2015b). However, in these biota samples, CB-11 exhibited quite minor proportions, which may be explained by low bioaccumulation efficiency and/or significant metabolism and excretion of low-chlorinated congeners (Paterson et al., 2007; Rodenburg et al., 2015b; Zhu et al., 2015b). Khairy et al. (2014) reported that CB-11 and other low-chlorinated PCBs were not magnified in the aquatic food web and exhibited trophic dilution. Also, little is known about the accumulation of CB-11 in terrestrial predators. Earlier in vivo exposure studies indicated that ^{14}C-labeled CB-11 was substantially absorbed and distributed to all tissues after begin delivered to the lungs of male rats, and was rapidly metabolized and excreted (Hu et al., 2013, 2014). Therefore, it has been suggested that hydroxylated, sulfated, and glucuronidated metabolites can serve as better biomarkers of exposure to CB-11 than their parent compound (Hu et al., 2014; Grimm et al., 2015).

CB-209 has been frequently detected in a wide variety of animal species from China (Yu et al., 2013; Kong et al., 2014; Zhou et al., 2016), Japan (Ohkouchi et al., 2016), Russia (Imaeda et al., 2009), the Netherlands (van Scheppingen et al., 1996), Canada (Hobbs et al., 2001), the US (Hansen et al., 2004; Fair et al., 2010; Rodenburg et al., 2015b), and the Arctic (Bernhoft et al., 1997; Addison et al., 2005). Concentrations of CB-209 in animal samples varied over several orders of magnitude, from pg g^{-1} to μg g^{-1}. Very high levels of CB-209 up to a few ppm were detected in blubber samples of bottlenose dolphins (*Tursiops truncatus*) from the US Atlantic coast (Hansen et al., 2004; Fair et al., 2010). CB-209 was also preferably accumulated in fatty tissues of high trophic level mammals from other locations at ppb levels (van Scheppingen et al., 1996; Bernhoft et al., 1997; Hobbs et al., 2001; Imaeda et al., 2009). Moderate levels of CB-209 were also found in different aquatic, amphibious, and terrestrial species from a riparian area in Beijing (median 270, range 100–1000 pg g^{-1} lipid wt.; Yu et al., 2013) and from the Yangtze River delta, China (median 4100, range 590–26,000 pg g^{-1} lipid wt.; Zhou et al., 2016). CB-209 was found at lower concentrations in fish samples collected from the Columbia River, the US (median 44 pg g^{-1} wet wt.; Rodenburg et al., 2015b) and in deep-sea organisms from off Tohoku coast, Japan (median 6.7, maximum 96 pg g^{-1} dry wt.; Ohkouchi et al., 2016). In these reports, proportions of CB-209 in total PCBs were not significant as compared to lower chlorinated homologs. These observations are likely attributed to extremely high hydrophobicity and steric hindrance of CB-209, which may lead to its difficulties in migrating across cell membranes and in reaching biota–water equilibrium (Kannan et al., 1998; Fisk et al., 2001; Wang et al., 2007). The abundance of CB-209 and other

Table 5
Concentrations (mean and range, pg g^{-1}) of PCBs in animal samples, with a focus on CB-11.

Location (sampling year)	Species	Total PCBs[a]; N[b]	CB-11; % of total PCBs	References
US, Passaic River estuary (2011)	Blue crab (*Callinectes sapidus*)	76,000,000 ± 1,400,000; 29	Minor percentages	Khairy et al., 2014
US, Hanford Site of Columbia River (2009–2010)	6 fish species[c]	Data unavailable	Detection rate 251/293; mean 41; median 25; minor percentages	Rodenburg et al., 2015b
China, Taizhou e-waste site (2012)	8 aquatic species[d]	25,000–2,270,000; 25	104 (5.5–1178); <1%	Zhu et al., 2015b
China, eastern Shandong Peninsula (2011)	Mackerel, clam, croaker, squid, and shrimp	140–4600; 209	Not detected – 330; 14 (0–35)%	Kong et al., 2014
China, a riparian area in Beijing (2010)	Zooplankton	151,000; 26	32,300; 21%	Yu et al., 2013
	8 invertebrate species[e]	32,000 (7700–72,000); 26	5200 (1300—10,000); 24 (6–47)%	
Antarctica, Terra Nova Bay (1996–2009)	Antarctic scallop (*Adamussium colbecki*)	40,110 (4430–220,000); 127	6950 (250–36,490); 17 (6–31)%	Pizzini et al., 2017
Italy, Adriatic Sea (2013–2014)	Mediterranean mussel (*Mytilus galloprovincialis*)	34,530 (5730–121,780); 127	7400 (610–35,730); 15 (6–29)%	Pizzini et al., 2017
	Manila clam (*Ruditapes philippinarum*)	12,100 (2170–34,920); 127	1980 (260–5120); 16 (10—23)%	
Iran, Persian Gulf (2015)	8 coral species[f]	110–3750; 22	Major compound with about 11%	Jafarabadi et al., 2018

[a] Concentrations based on lipid wt. by Yu et al. (2013) and Khairy et al. (2014); wet wt. by Kong et al. (2014) and Rodenburg et al. (2015b); and dry wt. for other studies.

[b] Number of analyzed PCB congeners.

[c] Studied species included: common carp (*Cyprinus carpio*), mountain whitefish (*Prosopium williamsoni*), walleye (*Stizostedion vitreum*), smallmouth bass (*Micropterus dolomieu*), bridgelip sucker (*Catostomus columbianus*), and white sturgeon (*Acipenser transmontanus*).

[d] Studied species included: crucian carps (*Carassius auratus*), grass carps (*Ctenopharyngodon idellus*), eels (*Monopterus albus*), crabs (*Eriochei rsinensis*), stone snails (*Bellarnya purificata*), apple snails (*Ampullaria gigas spix*), frogs (*Rana plancyi*), and toads (*Bufo raddei*).

[e] Studied species included: butterfly, chironomid, sludge worm (*Tubifex tubifex*), cicada, dragonfly, grasshopper, scarabaeidae larva, and earth worm.

[f] Studied species included: *Acropora robusta*, *Acropora valida*, *Platygyra daedalea*, *Favia favus*, *Favia speciose*, *Porites lutea*, *Sinularia compressa*, and *Sarcophyton trocheliophorum*.

highly chlorinated PCBs in biota was occasionally reported in Brunswick, Georgia, the US, which was resulted from local industrial activities utilizing Aroclor 1268 mixtures (Kannan et al., 1998; Pulster and Maruya, 2008; Balmer et al., 2011; Wirth et al., 2014); and therefore, we did not include these data in our discussion.

5.7. Summary

Air, dust, soil, sediment, water, and biota have been contaminated with unintentionally produced PCBs from pigments. A large number of monitoring studies have revealed the abundance of some specific congeners (notably CB-11 and CB-209), which are associated with pigment manufacturing, utilization, and disposal. Despite their similar origins from pigments, CB-11 and CB-209 are quite different in physicochemical properties and environmental behavior and fate. CB-11 has been considered a global contaminant due to its high mobility in the atmospheric and aquatic environments. Meanwhile, CB-209 has been used as an indicator of localized pollution in soils and sediments. Although the production and use of technical PCB mixtures were globally banned for several decades with a proposed deadline for elimination by 2028 (UNEP, 2016), the emissions and environmental residues of pigment-derived PCBs have not been addressed adequately in the vast majority of the world. Furthermore, many questions about these novel PCB congeners have been raised and need to be answered: Are they toxic to humans and biota? If yes, how may human and animal health be protected from their deleterious effects? What should we do to manage, control, reduce, and eliminate them? These issues will be discussed in the next sections.

6. Human exposure and toxic effects of pigment-derived PCBs

Humans and animals are exposed to PCBs via different pathways and sources such as food consumption, water ingestion, air inhalation, dust and soil ingestion, and dermal contact with PCB-containing materials (ATSDR, 2000; WHO, 2003). The huge body of in vitro, in vivo, in silico, and epidemiological studies conducted over the last five decades has demonstrated toxic effects of PCBs and their metabolites on human and animal health (ATSDR, 2000; WHO, 2003; Ruiz et al., 2008; Grimm et al., 2015; Faroon and Ruiz, 2016; IARC, 2016). Several potentially adverse effects of PCBs have been identified, including systemic, immunological, neurological, reproductive, developmental, genotoxic, and carcinogenic effects (ATSDR, 2000; WHO, 2003; Ulbrich and Stahlmann, 2004; Faroon and Ruiz, 2016). Toxicity and action mechanisms of individual PCBs are structure-related and generally classified into dioxin-like (DL) and non-dioxin-like (NDL) activities, which are mainly attributed to numbers and substitution positions of chlorine atoms in the biphenyl ring (Shain et al., 1991; Giesy and Kannan, 1998; Tilson et al., 1998; White and Birnbaum, 2009). It should be noted that a large number of PCB congeners belonging to all homologs with both DL and NDL structures were found in pigments and consumer products, and many of them were also dominant in technical PCB mixtures and from combustion sources. It is quite difficult to distinguish exposure route and toxicity contributions of PCBs from pigment-related sources and other origins, and thus we only focus on some congeners identified as typical pigment impurities (e.g., CB-11, CB-35, and CB-209).

6.1. Toxicity

Potentially toxic effects of some typical pd-PCBs and their metabolites have been demonstrated by several in vivo and in vitro studies conducted over the last three decades (Table 6). CB-11, 4-OH-CB-11, and 4′-OH-CB-35 can exhibit neurotoxicity in human and rat cell lines via different pathways such as decreasing cell dopamine content (Shain et al., 1991), inducing reactive oxygen species (ROS) production (Brown et al., 1998; Dreiem et al., 2009; Zhu et al., 2013), perturbing

Table 6

A summary of toxicity of pigment-derived PCBs and their metabolites, with a focus on CB-11, CB-35, and CB-209.

Compounds	Testing system	Type of toxicity	Observed toxic effects	References
CB-11	Zebrafish embryos (*Danio rerio*)	Embryotoxicity	Affect liver development, act as aryl hydrocarbon receptor (AhR) agonist/antagonist, inhibit cytochrome P450 1A (CYP1A) activity	Roy et al., 2019
CB-11	Rat PC12 cells	Neurotoxicity	Exhibit intermediate potency in decreasing cell dopamine contents	Shain et al., 1991
CB-11	Rat neutrophils	Neurotoxicity	Stimulate neutrophils to produce reactive oxygen species (ROS)	Brown et al., 1998
CB-11	Rat synaptosomes	Neurotoxicity	Stimulate production of ROS in rat synaptosomes	Voie and Fonnum, 2000
CB-11	Rat cerebellar granule cells	Neurotoxicity	Perturb protein kinase C (PKC) translocation	Kodavanti et al., 1995
CB-11	Transfected rat cortical cells	Neurotoxicity	Promote dendritic growth via cyclic adenosine monophosphate response element binding protein (CREB) mediated mechanism	Sethi et al., 2018
CB-11	Transfected rat GH3 pituitary tumor cells	Endocrine disruption	Inhibit T3 activation of thyroid hormone receptor	Sethi et al., 2019
CB-11	Transfected human prostate carcinoma epithelial cells	Endocrine disruption	Exhibit antagonistic activities against androgen receptor (AR)	Pencikova et al., 2018
CB-11	Rat WB-F344 liver epithelial cells	Carcinogenicity	Inhibit gap junction intercellular communication (GJIC)	Pencikova et al., 2018
CB-11	Human platelets	Cytotoxicity	Exhibit synergistic effect with CB-77 (but not CB-11 itself) in induction of 12-hydroxyeicosatetraenoic acid (12-HETE) generation	Raulf and Konig, 1991
4-OH-CB-11	Human RWPE-1 nonmalignant prostate epithelial cells	Neurotoxicity and cytotoxicity	Induce oxidative stress and cell growth suppression, decrease cell viability and plating efficiency	Zhu et al., 2013
4- and 5-OH-CB-11	Transfected human prostate carcinoma epithelial cells and human breast cancer cells	Endocrine disruption	Act as estrogen receptor (ER) agonist (4-OH-CB-11) and AR antagonist (4- and 5-OH-CB-11)	Pencikova et al., 2018
5-OH-CB-11	Rat WB-F344 liver epithelial cells	Carcinogenicity	Inhibit GJIC	Pencikova et al., 2018
4-CB-11 sulfate	Human MCF-7 breast cancer cells	Endocrine disruption	Exhibit androgenic activity	Flor et al., 2016
4-OH-CB-11 and 4-CB-11 sulfate	Human HepG2 liver cancer cells and rat GH3 pituitary tumor cells	Endocrine disruption	Exhibit agonistic activity towards thyroid hormone receptor alpha (TRα) signaling pathway	Ren et al., 2019
4-OH-CB-11 and 4-CB-11 sulfate	Human SH-SY5Y neuroblastoma cells and rat N27 dopaminergic neural cells	Cytotoxicity	Exhibit selective toxicity (e.g., higher potency in non-hepatic cells) in reduction of cell metabolic activity and lactate dehydrogenase release	Rodriguez et al., 2018
CB-35	Chinese hamster CHO-K1 ovary cells and mouse Hepa1c1c7 hepatoma cells	Endocrine disruption and AhR-mediated toxicity	Exhibit agonistic activities against estrogen receptor alpha (ERα) and AhR receptor, and antagonistic activities against AR	Takeuchi et al., 2017
4′-OH-CB-35	Rat cerebellar granule cells	Neurotoxicity	Stimulate production of ROS	Dreiem et al., 2009
CB-209 and nona-CBs	PCB-exposed mice immunized with sheep red blood cells	Immunotoxicity	Inhibit splenic plaque-forming cell response to sheep red blood cells	Harper et al., 1993

protein kinase C (PKC) translocation (Kodavanti et al., 1995), and promoting dendritic growth (Sethi et al., 2018). CB-11 and its hydroxylated and sulfated metabolites and CB-35 have been found as agonists/antagonists of androgen, estrogen, and thyroid hormone receptors (Flor et al., 2016; Takeuchi et al., 2017; Pencikova et al., 2018; Ren et al., 2019; Sethi et al., 2019). Non-*ortho* substituted CB-35 was estimated to have significant aryl hydrocarbon receptor (AhR) agonistic activity with 20% relative effective concentration $REC_{20} = 1.9 \times 10^{-7}$ M, showing activity just lower than dioxin-like congeners as CB-126 and CB-77 (Takeuchi et al., 2017). Meanwhile CB-11 even without any *ortho*-substitution is an AhR antagonist (Roy et al., 2019). Exposure to CB-11 and 5-OH-CB-11 in vitro may induce arachidonic acid release and/or disrupt gap junction intercellular communication (GJIC), linking them with tumor promotion/carcinogenesis (Pencikova et al., 2018). Although CB-11 induced only a marginal estrogen receptor mediated activity (maximum activity 18% relative to 17β-Estradiol), its metabolite 4-OH-CB-11 had significantly higher activity (maximum 65% relative to 17β-Estradiol) (Pencikova et al., 2018). In addition, computational toxicology was used to predict potential mutagenesis and/or carcinogenesis of CB-11, CB-35, 6′-OH-CB-35, and CB-209 (Ruiz et al., 2008). These findings justify human health concerns associated with pd-PCBs and their metabolites (Zhu et al., 2013; Koh et al., 2015; Rodenburg et al., 2015a; Takeuchi et al., 2017; Sethi et al., 2018, 2019).

6.2. Human levels

CB-11 has been detected in human breast milk (Yang et al., 2002; Nakamura et al., 2008) and blood (Hirai et al., 2005; Nakamura et al., 2008; Marek et al., 2013, 2014; Koh et al., 2015; Sethi et al., 2019). CB-11 was found as one of the most dominant PCB congeners in Korean human milk (1–9% by mole), implying local non-Aroclor sources in urban and industrial areas of Seoul and Incheon (Yang et al., 2002). Meanwhile, CB-11 was only a minor congener detected in breast milk of Japanese women in the Tohoku region (median 0.067, range 0.036–0.164 ng g^{-1} lipid wt., average 0.1% of total PCBs; Nakamura et al., 2008). CB-11 was also listed as one of the most frequently detected congeners in human serum from some US urban and rural communities with 95th percentile value of about 13 ng g^{-1} lipid wt. (Marek et al., 2013, 2014; Koh et al., 2015). The abundance of CB-11 (just behind CB-28) was reported by Sethi et al. (2019) for human maternal serum in northern California, the US. In a Japanese cohort study, levels of CB-11 in maternal blood (median 1.48, range 0.36–15.6 ng g^{-1} lipid wt., average 3.5% of total PCBs) were much higher than those measured in cord blood (0.13; range 0.07–0.29 ng g^{-1} lipid wt., average 0.3%) (Nakamura et al., 2008). In addition, mono-sulfated, mono-hydroxylated, and glucuronidated metabolites of CB-11 were detected in human serum in the US (Zhu et al., 2013; Grimm et al., 2017). Three hydroxylated metabolites of CB-11 (i.e., 2-, 4-, 6-OH-CB-11) were detected in serum samples from three US male donors with concentrations ranging from 1 to 6 pg g^{-1} fresh wt. (Zhu et al., 2013). In 46 serum samples from mothers and their daughters in East Chicago and East Iowa, the US, detectable levels of 4-CB-11 sulfate were found in >20% of the analyzed samples with a maximum quantified concentration of 4.47 pg g^{-1} lipid wt. (Grimm et al., 2017). Beside CB-11, Koh et al. (2015) reported high detection frequencies of other low-molecular-weight and pigment-related PCBs such as CB-14 (73%) and CB-35 (58%) in the US human serum samples.

Due to its high hydrophobicity, CB-209 is ubiquitously distributed in human adipose tissues (Teschke et al., 1993), breast milk (Nakamura et al., 2008; Todaka et al., 2010, 2011), blood (Hirai et al., 2005; Nakamura et al., 2008; Todaka et al., 2008a, 2008b, 2009, 2010, 2011; Koh et al., 2015; Magonia et al., 2016; Zheng et al., 2016), and hair (Zheng et al., 2016; Qiao et al., 2019). Concentrations of CB-209 in omental adipose tissue samples from Canadian residents ranged from 0.54 to 21.3 (mean 7.49) ng g^{-1} lipid wt. and accounted for about 1% of total PCBs (Teschke et al., 1993). In a series of human biomonitoring studies of the Japanese population conducted during 2001–2005, CB-209 was found in all the blood and milk samples with median values of 0.058–0.074 (range 0.005–74.8) and 0.32–1.4 (range 0.063–3.6) ng g^{-1} lipid wt., respectively (Nakamura et al., 2008; Todaka et al., 2008a, 2008b, 2009, 2010, 2011). Elevated levels of CB-209 were detected in serum samples of male (median 34.1 ng g^{-1} lipid wt.) and female (median 38.6 ng g^{-1} lipid wt.) e-waste recycling workers in southern China (Zheng et al., 2016), which were much higher than those measured in normal US residents (median 0.001 ng g^{-1} fresh wt.; Koh et al., 2015). Human hair samples of Chinese e-waste recyclers also contained CB-209 with median values of 0.27–1.55 ng g^{-1} (Zheng et al., 2016; Qiao et al., 2019). Several factors that influenced human levels of CB-209 have been identified, for example, body mass index (negatively correlated with adipose concentrations; Teschke et al., 1993), age (increased with increasing concentrations in blood; Koh et al., 2015; Magonia et al., 2016), and sex (higher concentrations in female hair; Zheng et al., 2016). Nevertheless, almost all the studies investigating CB-209 listed above did not mention its pigment-related sources, except for Koh et al. (2015) who suggested human exposure to CB-209 through contact with products containing phthalocyanine pigments and/or food consumption.

6.3. Exposure pathways and health protection measures

PCBs in general and pigment-derived congeners in particular can enter human bodies through three main pathways: inhalation, ingestion, and dermal contact with materials containing them. The relative contributions of these pathways in total human exposure to PCBs depend on congener-specific patterns in contamination sources and characteristics of exposed persons. The principal exposure route of low-chlorinated PCBs, including CB-11, was estimated to be inhalation of indoor air (Hu et al., 2008; Marek et al., 2013, 2017; Ampleman et al., 2015; Koh et al., 2015; Wang et al., 2019). CB-11 was among the most dominant congeners inhaled by residents in the US (Ampleman et al., 2015), Australia (Wang et al., 2019), and Vietnam (Anh et al., 2020a). Elevated concentrations of CB-11 were found in indoor air of schools in the US (Marek et al., 2017) and South Korea (Baek et al., 2010), implying considerable exposure sources for school-aged persons (e.g., children and adolescents) and teachers (Ampleman et al., 2015; Koh et al., 2015; Marek et al., 2017). In addition, inhalation doses of PCBs may exceed those of dietary exposures in children from some schools where higher indoor concentrations of PCBs were detected, especially for more physically active children as boys than girls (Marek et al., 2017). On the other hand, Koh et al. (2015) found significant correlation between serum levels of pigment-related congeners like CB-11, CB-35, and CB-209 in adolescent-mother pairs from some US communities in East Chicago (Indiana) and Columbus Junction (Iowa), and suspected that the home environments and diet may be exposure sources of pd-PCBs. To our knowledge, studies focusing on dietary exposure to pd-PCBs are still scarce, although CB-11 and CB-209 have been frequently detected in various aquatic species that may be used as food (King et al., 2002; Khairy et al., 2014; Kong et al., 2014; Rodenburg et al., 2015b; Pizzini et al., 2017). Recently, Saktrakulkla et al. (2020) measured 205 PCB congeners in 26 food items purchased in a rural community in Iowa, US, and found CB-11 in catfish, tilapia, and potato. Dermal contact with painted surfaces, printed papers, pigmented fabric materials, and contaminated dust is also a possible pathway of human exposure to pd-PCBs, but its role has not been well-characterized. For the intake of pd-PCB by infants and small children, contributions of breast feeding and unintentional oral/dermal exposure to house dust, paint chips, toys, comics, and textile products (e.g., napkins, handkerchiefs, and towels) should be taken into account together with such general sources as indoor air breathing; because CB-11 and/or CB-209 were detected in breast milk (Yang et al., 2002; Nakamura et al., 2008; Todaka et al., 2010, 2011), house dust (Wang et al., 2019; Anh et al., 2020b), and various pigment-treated products (Guo et al., 2014; Stone, 2014, 2016).

It seems impossible to completely eliminate human exposure to pigment-derived pollutants including PCBs because of their widespread occurrence in the surrounding environments, consumer products, and food. In this section, we listed practices for prevention and reduction of PCB exposure that were previously proposed by professional authorities, with special consideration of their origin from pigments. People should only consume food of guaranteed origin and avoid those that were free-living or farmed in or around contaminated/potentially contaminated areas (FAO, 2006). Appropriate preparation (e.g., removing skin, head, and fatty tissues) and cooking methods (e.g., grilling and deep-frying with disposal of cooking liquids and used oils) can reduce PCB intake by food consumption, especially for fish and meat (Moya et al., 1998; Jin et al., 2017). Other cooking methods such as boiling, simmering, pan broiling, and stir-frying without oils did not exhibit significant efficiency in reducing PCB levels in cooked fish and seafood as compared to raw materials (Moya et al., 1998; Moon et al., 2019). Consumption of food with high dietary fiber contents such as fruits, vegetables, mushrooms, seaweed, rice, nuts, pulses, and tea may reduce exposure to PCBs (Jin et al., 2017). More critically, an overall reduction of animal-based food (notably fish, meat, eggs, and milk) with addition of vegetables and fruits is suggested (Weber et al., 2018). In addition, colored food packaging materials containing PCBs (Guo et al., 2014; Stone et al., 2014, 2016; Liu and Mullin, 2019) and the use of plastic bags and old newspapers or magazines to cover food (e.g., street food in some developing countries) should be avoided.

Measures to prevent PCB exposure from indoor environments and consumer products are also very important. Several remediation methods of indoor PCBs have been proposed for legacy PCB-treated building materials such as fluorescent light ballasts, caulks, joint sealants, paints, and so on (US EPA, 2015; Brown et al., 2016); but such management practices for inadvertently produced PCBs in modern items have not been fully developed (Shanahan et al., 2015; Davies and Delistraty, 2016). General practices for minimizing PCB exposure can be applied such as facilitating air ventilation and more frequent cleaning by proper techniques (e.g., removing dust and residue by wet or damp cloths and vacuum cleaners rather than dry brooms and cloths) (US EPA, 2015). Healthy indoor painting practices have also been introduced such as selection of suitable paint types, maintaining ventilation during painting and for a few days afterwards, and avoiding freshly painted rooms for 2–3 days to reduce exposure to harmful vapors (US EPA, 2000). However, the emissions of PCBs from painted surfaces to indoor air can occur over several weeks or even longer periods (Jahnke and Hornbuckle, 2019), suggesting the need of appropriate scheduling and preparation by painters and house managers with notification for neighbors before painting operation. Human health risks via dermal contact with textiles and clothes have been addressed for various inorganic and organic chemicals (Licina et al., 2019; Rovira and Domingo, 2019); but little is known about this exposure route of PCBs. Some azo-pigment-derived PCBs like CB-11 and CB-52 were detected in a variety of dyed and printed fabric products and their potential for dermal absorption was also suspected (Guo et al., 2014). Newly purchased clothing should be washed several times with elevated temperature water and detergents. In addition, more critical protection measures should be paid to small children such as providing them with a safe diet and good hygienic conditions, strict management of their play activities and toys, and isolating them from potential sources of PCBs and dirty areas (ATSDR, 2000; US EPA, 2015).

7. General discussion and perspectives

Inadvertent formation of PCBs in pigment manufacturing processes and PCB release during production, application, and disposal of pigment-treated products have been considered as ongoing and significant sources of these persistent toxic substances. The first notice of the environmental occurrence of CB-11, a typical pigment-derived congener, was documented in France in 1985 (Law, 1995); but some US sediment core archives showed that CB-11 and highly chlorinated congeners (e.g., nona- and deca-CBs) were found in sediment layers deposited before the start of technical PCB production in the late 1920s (Hu et al., 2011). The intensive applications of pigments and related products have resulted in the omnipresence of such PCB congeners with a wider temporal spectrum than those of intentionally produced PCBs. The abundance (and sometimes with unexpectedly high proportions) and potential adverse effects of pd-PCBs on environmental and human health have stimulated scientific and public attention to these pollutants (Grossman, 2013; Rodenburg et al., 2015a; Vorkamp, 2016; Robertson et al., 2018). Although the number of studies focusing on pd-PCBs has increased since the 2000s, there are several knowledge gaps regarding them that include multiple aspects such as academic, technical, management, and educational issues.

Additional monitoring and assessment investigations on pd-PCBs should be conducted to characterize their environmental occurrence, behavior, fate, and emission sources. The inclusion of pd-PCBs (e.g., CB-11, CB-35, CB-209, and other relevant congeners) as target analytes has altered current understanding about PCB levels and patterns in water, air, and other environmental media (King et al., 2002; Litten et al., 2002; Wang et al., 2019; Anh et al., 2020a, 2020b; Takahashi et al., 2020; Zhao et al., 2020). Indoor air, wastewater effluents, leachates, industrial releases, and dumping fugitive emissions should be monitored more frequently everywhere in the world by sufficient analytical techniques to trace the presence of PCBs from novel sources. A variety of in vivo and in vitro studies have indicated neurotoxicity, endocrine disruption activity, and possible carcinogenicity of pd-PCBs and metabolites, but mechanisms of their metabolism and toxicity need further elucidation (Takeuchi et al., 2017; Pencikova et al., 2018; Roy et al., 2019). As most toxicity guideline values were derived for technical PCB mixtures and dioxin-like congeners, derivation of such values for pigment-derived compounds are necessary, especially for the inhalation exposure pathway (Cogliano, 2016). Improvement of people's awareness and understanding about the presence, sources, potential toxic impacts, and prevention measures of PCBs associated with household materials and consumer products by providing them with appropriate guidance documents, is highly recommended.

On the technical and management aspects, a comprehensive and effective solution for abatement of unintentionally produced PCBs in pigments is still lacking, especially in developing countries. The occurrence of PCBs in various types of pigments is largely explained by the structural similarity between detected PCBs and intermediates and/or solvents employed, but the formation pathway has not been verified in situ with technological considerations by manufacturers (Hu and Hornbuckle, 2010). Pigments that contain chlorine atoms in their molecular structures and use chlorinated intermediates and reaction solvents in their synthesis are likely contaminated with PCBs (ETAD, 2011). Therefore, the replacements of such pigments and raw materials by chlorine-free alternatives have been approved but there are still significant challenges (Rodenburg et al., 2015a). For example, some diarylide yellow pigments showed elevated PCB concentrations up to 700–900 ppm (Table 1), however, replacing them is a very difficult task due to their ability to satisfy specific technical requirements and low cost (Rodenburg et al., 2015a), and these pigments have accounted for the largest fraction of organic yellow pigments on the market (Hunger and Schmidt, 2018). General threshold values of 50 ppm or 25 ppm have been applied as a maximum or allowed residue levels in products containing PCBs in the US, Canada, Japan, and some European countries (ETAD, 2011). With special consideration of inadvertent PCB production, manufacturers and importers dealing with products containing PCBs at levels >2 ppm are required to report to the US EPA (Davies and Delistraty, 2016). In Japan, business operators utilizing organic pigments with PCB concentrations >0.5 ppm identified by the Japan Dyestuff and Industrial Chemicals Association, are requested to confirm PCB contents and report the results to relevant competent authorities (METI, 2012). The compliance of business operators and effectiveness of regulatory agencies might play an important role in management and abatement of unintentional production of PCBs in pigment-related industries.

As a concluding remark, scientific researches and regulatory improvement efforts have shown novel emission sources of PCBs from pigment manufacturing with specific indicators such as CB-11, CB-35, and CB-209, together with those already known as dominant congeners in technical PCB formulations (e.g., CB-52) and combustion sources (e.g., CB-77). The formation and emission of pigment-derived PCBs should be controlled and reduced in a proper manner because of their toxicity, high mobility and/or extreme persistence, and because their abundance may surpass those of legacy congeners in various environmental compartments and biological systems. There is an urgent need for effective reduction, management, and remediation of pigment-derived PCBs, which requires contributions of scientists, engineers, business operators, policy makers, governmental managers, and the general public.

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Declaration of competing interest

The authors declare that there is no conflict of interest regarding the publication of this article.

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