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A review of PCB-11 and other unintentionally produced PCB congeners in outdoor air

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ABSTRACT

This review summarizes up-to-date scientific literature concerning unintentionally produced polychlorinated biphenyls (UP-PCBs), including information on their known or suspected formation pathways, occurrence in air globally, and properties relating to atmospheric persistence and transport. Prior to the listing of PCBs as an original "dirty dozen" persistent organic pollutant (POP) under the Stockholm Convention on Persistent Organic Pollutants, they were already widely regulated, and some monitoring in air was occurring due to environmental and health concerns. So far, the focus of monitoring has been for dominant congeners found in technical PCB formulations, such as Aroclors. However, recent research has shown that processes such as dye/pigment manufacturing, and industrial thermal processes have resulted in UP-PCB emission and detection globally. It is especially concerning that UP-PCBs make up a significant proportion of ∑PCBs (typically from a few percent to as high as 85%), and this contribution continues to increase. Among identified UP-PCBs, PCB-11 is the dominant congener detected in air. Three key recommendations from this review include i.) to include UP-PCBs, such as PCB-11 and PCB-209, as indicator congeners in air monitoring and research programs; ii.) to apply PUF disk passive air samplers as simple and cost-effective tools for generating new information on global air; and iii.) to identify and quantify the ongoing emission sources of UP-PCBs to air. The new information will raise awareness to the growing problem of UP-PCBs and could inform science and policy strategies for assessing and managing this emerging class of chemicals.

1. Introduction

Polychlorinated biphenyls (PCBs) are a family of chemicals composed of 209 anthropogenic persistent organic pollutants (POPs). They consist of chlorine atoms (n = 1–10) located around a biphenyl backbone (Fig. 1). These organic compounds were first manufactured in the 1920s, with their use and production both peaking in the 1960s and 1970s (IARC, 2015). Under trade names such as Aroclor, Clophen, Fenchlor, and Kanechlor (IARC, 2015), it is estimated that at least 1.3 million tons of PCBs were manufactured globally (Breivik et al., 2002). These PCB-containing mixtures were applied to various uses such as dielectric fluids in capacitors and transformers, hydraulic fluids, lubricants, heat-transfer fluids and were incorporated into products and materials such as rubber, carbonless copy paper, and adhesives (IARC, 2015). Of the 209 possible PCB congeners, approximately 130–150 congeners have been found in Aroclor and Clophen mixtures (Frame et al., 1996; Schultz et al., 1989). These congeners are commonly referred to as Aroclor PCBs. The remaining congeners (60–80 species) are either absent or only present in trace quantities (<0.05%, w/w; Schultz et al., 1989), and these are generally referred to as non-Aroclor PCBs.

Issues regarding their environmental persistence, bioaccumulation, and toxicological effects including neurotoxicity, immunosuppression, reproductive effects, thyroid and retinol effects, carcinogenicity, and porphyria led to the gradual elimination of PCBs (AMAP, 2004). PCB production in Japan ceased in 1972, with the USA halting production in 1977 and remaining global production ending by 1993 (Breivik et al., 2002, 2007). Internationally, the production and use of PCBs is currently regulated under Annexes A (Elimination) and C (Unintentional production) of the Stockholm Convention on Persistent Organic Pollutants, which came into effect in 2004 (UNEP, 2009). The Global Monitoring Plan (GMP) is used to evaluate the effectiveness of the Stockholm Convention. Through the GMP, key environmental media, such as ambient air, and biological matrices such as human milk and human

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blood, are monitored to identify changes in POP concentrations over time, including the monitoring of PCB species. The Stockholm Convention provides exemptions for the production and use of select compounds for specific purposes, like the use of DDT for disease vector control (UNEP, 2009). However, no specific exemptions exist regarding the intentional production of PCBs. While historically PCB emissions have been dominated by Aroclor PCB congeners because of their widespread use, recent research indicates that atmospheric PCB profiles are shifting towards current use sources (Khairy et al., 2015). As a result, this profile shift has led to the detection of non-Aroclor or unintentionally produced PCBs (UP-PCBs) in outdoor air (Hu et al., 2008).

As PCBs are anthropogenic chemicals, they have no known natural emission sources. The anthropogenic nature of PCBs, combined with restrictions on their production and use, makes the presence of UP-PCBs in the environment concerning, as it indicates the existence of novel sources responsible for their production and emission. Dye and pigment manufacturing has been linked to the generation and release of UP-PCBs. Other potential UP-PCB sources include waste incineration, pulp production, and other thermal processes such as secondary copper, aluminum, and zinc production, and sintering plants (UNEP, 2009). Given that signatory parties to the Stockholm Convention must take measures to reduce the unintentional release of Annex C compounds (UNEP, 2009), work must be undertaken to better understand the release of UP-PCBs into the atmosphere and to monitor their global emissions.

Previous reviews regarding UP-PCBs have mainly addressed their formation through dye and pigment manufacturing (Anh et al., 2021), and their broad occurrence within various environmental compartments and consumer products (Vorkamp, 2016). In addition, investigation into the presence of UP-PCBs and their sources in China has also been conducted (Liu et al., 2018). However, thus far, no peer-reviewed literature has focused on the global presence of UP-PCBs in the atmosphere.

The objective of this study is to develop a broader understanding of UP-PCBs in outdoor air through a full review of the relevant literature. This includes examining: 1) Information regarding known pathways for the inadvertent formation of PCBs, 2) synthesizing physical properties of UP-PCBs that are relevant to their environmental fate, 3) looking at the current state of sampling and analytical procedures, and 4) a comprehensive look at UP-PCB levels in outdoor air, with a focus on PCB-11. A brief overview of known toxicological impacts related to UP-PCBs is also included to provide further context on these emerging compounds in the environment.

Based on this review, recommendations are provided for future work to improve understanding and to address concerns related to the growing burden of unintentional PCBs in air and other environmental compartments.

2. Review approach

Web of Science (core collection) and Google Scholar were searched during August 2021 for the keywords “non-Aroclor polychlorinated biphenyls” and “unintentional PCB” in conjunction with “air”, “atmosphere”, “ambient”, “airborne”, or “inhalation”. Further background information was sought with the keywords “toxicity”, “toxicology”, or “properties”. The keywords “unintentional PCBs” and “PCB-11″ were searched separately. The search was limited to English language papers published between 1990 and 2021. From this, 225 papers were identified to have titles or abstracts that appeared relevant based on fulfilling one or more of the following criteria:

(1) Title or abstract contains mention of known UP-PCB species,
(2) Production pathways and emission sources of UP-PCBs are addressed,
(3) Provides details of PCB air monitoring, measurements, or analytical methods, or
(4) Outlines toxicological, biological, or other health-related impacts, including mention of PCB metabolites.

Papers were grouped by their primary area of research focus. If articles were deemed satisfactory by fulfilling one of the above criteria, they were subsequently examined individually for results pertaining to non-Aroclor PCBs. Where relevant, studies that were omitted by the original search criteria (such as older publications) have been included where they supplement reviewed materials.

3. Unintentionally produced PCBs

The definition of what constitutes an unintentional PCB has varied throughout the literature. For example, congeners detected in various Aroclor mixtures by Frame et al. (1996) helped outline the definition of what constitutes an Aroclor PCB. Conversely, non-Aroclor PCBs are those congeners not in Aroclor mixtures, a category which encompasses the 60–80 congeners that are either absent from Aroclor mixtures, or those that are only present in trace quantities (Table 1). Other definitions have more broadly included PCBs that have not been intentionally produced for commercial or industrial purposes. The difference in these definitions is that the latter can include PCBs that are detectable in Aroclor mixtures, but are also acknowledged to have alternative and ultimately unintentional formation pathways. The bulk of analysis in this paper will address congeners that fall under the former definition, as there are no intentional pathways through which these congeners are produced.

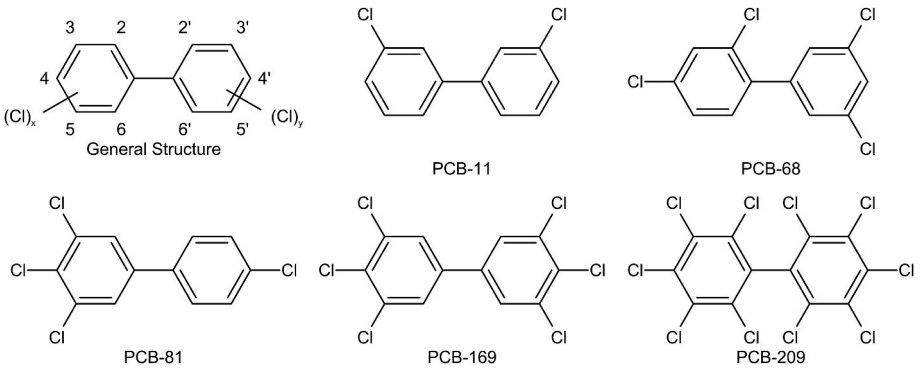


Fig. 1. General chemical structure of polychlorinated biphenyls (modified from Ge et al., 2013), and the structures of unintentionally produced PCB congeners 11, 68, 81, 169, and 209.

Table 1
Summary of PCB congeners either found in trace quantities or non-detectable in Aroclor and Clophen mixtures, or possess known pathways for unintentional formation, and as such are classified as non-Aroclor PCBs in this review. Regular font indicates congeners reported by Schultz et al. (1989) at concentration levels ≤ 0.05%; bolded text indicates congeners reported by Frame et al. (1996) at concentration levels <0.01% (wt), or not detected; italicized font indicates congeners identified by both Schultz et al. (1989) and Frame et al. (1996) to not be major congeners within Aroclor mixtures. Species enclosed in square brackets are congeners that have been detected in outdoor air, and detailed in this review.

Di-CB	[11]	12	13	*14*															
Tri-CB	*21*	23	*30*	*36*	*38*	*39*													
Tetra-CB	43	*50*	54	55	57	*58*	*61*	*62*	*65*	*[68]*	**69**	71	*72*	*73*	76	*78*	*79*	*80*	[81]
Penta-CB	86	89	**90**	93	94	*98*	**100**	102	103	*104*	*106*	107	*108*	109	*111*	*112*	*113*	*116*	117
	120	*121*	124	125	*127*														
Hexa-CB	133	139	*140*	*142*	*143*	144	*145*	147	*148*	*150*	*152*	154	*155*	*159*	**160**	*161*	*162*	163	164
	165	166	*168*	**[169]**															
Hepta-CB	*181*	*182*	*184*	*186*	*188*	*192*													
Octa-CB	200	*204*																	
Deca-CB	**[209]**																		

4. Origin and emission sources

4.1. Organic pigments

Hu and Hornbuckle hypothesized the presence of unintentional PCB congeners in pigments in 2010. This hypothesis stemmed from research that indicated the presence of PCB-11 at high concentrations in pigment manufacturing effluents (Litten et al., 2002). Diarylide yellow in particular has been of interest due to the nature of its chemical structure, which possesses a fragment identical to PCB-11 (Fig. 2). The unintentional production of PCBs in the pigment manufacturing process is suspected to occur through side reactions involving chlorinated solvents and reaction intermediates. Reports have detected PCB-11 in various consumer goods such as newspapers, magazines, cardboard, and other paper products that contained, or were suspected of containing, diarylide yellow (Rodenburg et al., 2010). As indicated by its name, diarylide yellow is part of the diarylide pigment group, a sub-group of azo pigments. Regarding their commercial use, azo pigments are the most important synthetic organic colorants in use, making up over 50% of the organic pigments available (Hu and Hornbuckle, 2010). Various red, orange, and yellow hues are particularly dominant azo pigments found in commercial products (Hu and Hornbuckle, 2010). Other organic pigments of significance include phthalocyanine pigments, which are responsible for many blue and green hues in commercial items. A summary of UP-PCB species by origin or emission source is presented in Fig. 3.

After examining 24 yellow pigment samples from 3 different manufacturing plants in China, PCB-11 was detected in all yellow pigment samples and accounted for a median of 85.5% $\sum_{20}$PCB (Shang et al., 2014). Among the pigments analyzed by Shang et al. (2014), diarylide yellow pigments contained the highest levels of PCB-11. PCBs 28, 52, and 77 were also found at very high levels. The detected levels of PCB-52 in particular contradict previous findings by Hu and Hornbuckle (2010), as it accounted for 92% of $\sum_{20}$PCB in one pigment (P.Y.12). The cause of high PCB-52 levels was unknown. In contrast, Hu & Hornbuckle did not note any significant contributions by PCB-52. Hu and Hornbuckle (2010) examined 33 paint pigments, discovering that PCBs were present primarily in organic pigments at concentrations ranging from 2 to 200 ng/g fresh weight in 15 of 33 samples. Covering a range of pigment colors, the detection frequency of PCB congeners in those 15 samples is presented in Figure S1. Hu & Hornbuckle also reported that of the pigments sampled, PCBs are only found in those of the azo and phthalocyanine types. Among the congeners detected, dioxin-like congeners PCB-77, 114, and 123 were found. PCBs 1, 2, 3, 4, 6, 8, 12/13, and 209 were also detected, and were found at a detection frequency greater than 40% (Hu and Hornbuckle, 2010). PCB-11 was the most detected congener in 13 of 15 pigments where PCBs were identified (Hu and Hornbuckle, 2010).

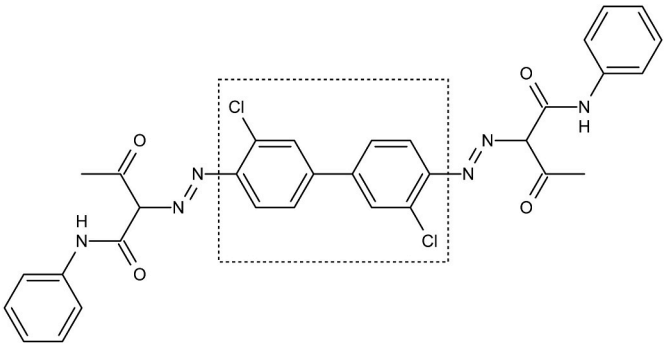


Fig. 2. Illustration of PCB-11 fragment found within the chemical structure of Pigment Yellow 12, a diarylide pigment (modified from Vorkamp, 2016).

Analysis of colorants by Jahnke and Hornbuckle (2019) found some PCBs in samples that matched those previously examined (Hu and Hornbuckle, 2010; Anezaki and Nakano, 2014). However, unlike previous results, PCB-11 was not a dominant congener in sampled yellow colorant samples. PCB-11 comparatively only appeared in the top 10 PCBs by mass for three colorants: phthalo green, medium yellow, and organic orange (Jahnke and Hornbuckle, 2019).

Numerous commercially available organic pigments were examined by Anezaki and Nakano (2014), who reported prominent unintentionally produced PCB congeners and their suspected formation pathways in the pigment manufacturing process. PCBs were detected in 24 of 29 azo-type pigments tested and in 9 of 13 phthalocyanine pigments. PCBs were present at concentrations ranging from 0.0070 to 740 mg/kg within azo-type pigments (Anezaki and Nakano, 2014). PCB-11 was identified as the dominant congener in all but two PCB containing azo pigments, accounting for more than 70% of the total PCB concentration (Anezaki and Nakano, 2014). Other highlighted congeners found in azo-type pigments include PCB-35, PCB-77, and PCB-52. Azo pigments are generally produced using 3,3′-dichlorobenzidine or 2,2′,5,5′-tetrachlorobenzidine, which is converted to a tetrazo form using $NaNO_2$ or HCl, followed by coupling with acetoacetanilide or 3-methyl-1-phenyl-5-pyrazoline (Anezaki and Nakano, 2014). Other azo pigments, such as CI Pigment Red 2 and CI Pigment Red 112, have also been recognized by industry for their potential to inadvertently form PCBs in their production (Rodenburg et al., 2015).

In phthalocyanine pigments, PCBs were found at concentrations ranging from 0.011 to 2.5 mg/kg (Anezaki and Nakano, 2014). PCB-209 accounted for more than 92% of total PCB concentrations in pigment green 7 (PG7), while high concentrations of nonachlorinated congeners 206, 207, and 208 were also detected. Copper phthalocyanine crude blue is a precursor to some phthalocyanine-type pigments. Few PCBs were detected in copper phthalocyanine crude blue, which suggests that

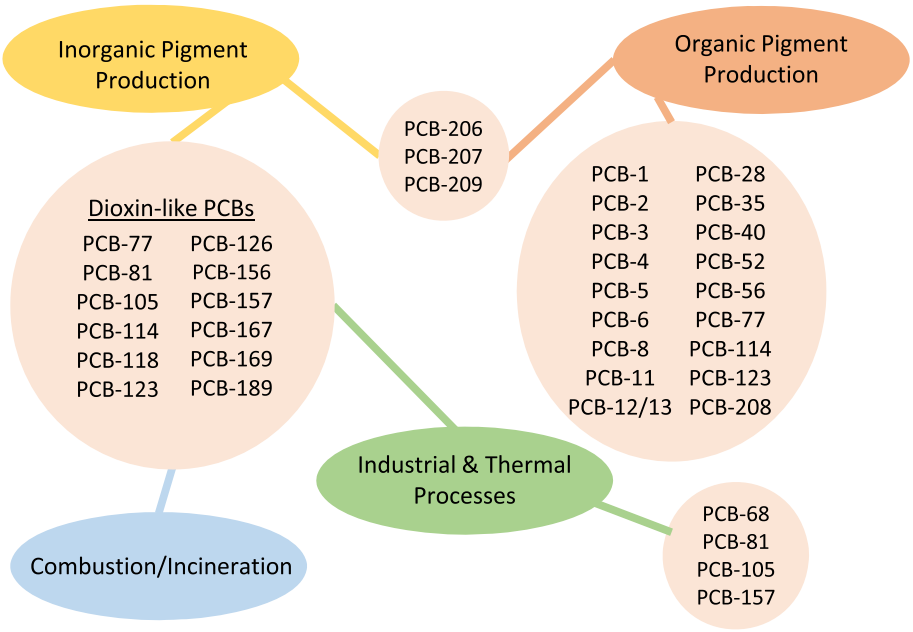


Fig. 3. Summary of PCBs identified in literature with known unintentional production pathways. Their associated origin and emission sources are illustrated.

the chlorination step used to convert copper phthalocyanine crude blue into other pigments is responsible for UP-PCB formation. Chlorination is first suspected to occur with dichlorobenzene and trichlorobenzene, the organic solvents used in phthalocyanine crude blue synthesis, followed by hyperchlorination of the chlorobenzenes and formation of highly chlorinated PCBs (Fig. 4; Anezaki and Nakano, 2014). As such, it is suggested that PCBs may be formed in the production of green pigments such as CI Pigment Green 7, a copper phthalocyanine derivative (Rodenburg et al., 2015).

In polycyclic-type paint pigments, PCB congeners have been detected in diketopyrrolopyrrole (PR254) and dioxazine violet (PV23) pigments (Anezaki and Nakano, 2014). PR254 possessed dichlorinated PCB congeners with only a single chlorine substituent on either aryl group of the biphenyl backbone. PV23 was dominated by dichlorinated or tetrachlorinated congeners including PCB-5, PCB-12, PCB-56, PCB-77, and PCB-40.

In the manufacturing of organic dyes, heterolytic and homolytic processes are suspected to be the primary mechanisms by which PCBs are inadvertently formed (Rodenburg et al., 2015). The heterolytic process forms an aryl cation that reacts with nucleophiles present, such as chloride ions, to form chloroaromatics. However, it is suspected that the homolytic process is the more likely formation pathway for unintended PCBs in manufacturing azo pigments. This process is initiated with an electron transfer from a reducing agent, forming an aryl radical after nitrogen elimination occurs (Rodenburg et al., 2015).

4.2. *Inorganic pigments*

Inorganic pigment production is also suspected of causing the unintentional formation and release of PCBs. Production of titanium tetrachloride ($TiCl_4$), a key precursor to titanium dioxide (TiO_2) pigments, has been associated with the detection of inadvertent PCBs in the environment (Praipipat et al., 2013; Rodenburg and Ralston, 2017), including PCBs 206, 207, and 209 (Rodenburg, 2012). These congeners are likely produced through the purification process of TiO_2, which is described as follows (Gázquez et al., 2014):

$$2TiO_2 + 3C + 4Cl_2 \rightarrow 2TiCl_4 + 2CO + 2CO_2 \quad (1)$$

$$TiCl_4 + O_2 \rightarrow TiO_2 + 2Cl_2 \quad (2)$$

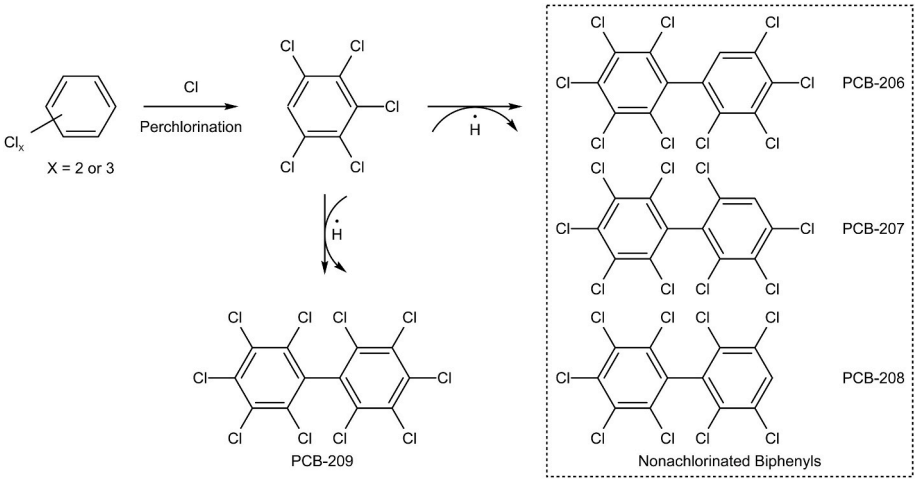


Fig. 4. Formation of unintentional PCBs as by-products of phthalocyanine-type pigment manufacturing (modified from Anezaki and Nakano, 2014).

Through the process illustrated above, unreacted feedstock material could react with chlorine and carbon (usually coke) to generate more highly chlorinated congeners like PCB-206, 208, and 209 as previously mentioned. Rodenburg (2012) illustrated this as follows:

$$2FeTiO_3 + 7Cl_2 + 6C \rightarrow 2TiCl_4 + 2FeCl_3 + 6CO \quad (3)$$

The formation of PCBs through the TiO_2 manufacturing process is supported by the findings of Ctistis et al. (2016), where 18 PCB congeners, including dioxin-like congeners, were found to range from <5 ng/kg up to 802 ng/kg in titanium dioxide pigment. Among the detected congeners however, highly chlorinated PCBs 206, 207, and 209 were not reported. Rather, the detected congeners ranged from tri-chlorinated PCB-28 to hepta-chlorinated PCB-189. Dioxin-like congeners exhibited overall lower concentrations than other PCBs. These results contradict those previously published by Hu and Hornbuckle (2010). There, no PCBs were reported to be present in inorganic pigments that primarily contained titanium dioxide, iron oxide, raw umber, or carbon black. Despite various PCB species being present in titanium dioxide pigments, little direct evidence has been published regarding where UP-PCBs originate in the manufacturing process.

4.3. Thermal, combustion, and decomposition processes

Many studies have shown the potential for PCB formation and release through various thermal and combustion processes. Ishikawa et al. (2007) demonstrated both the formation and decomposition of PCB congeners through the combustion of both refuse-derived fuel (RDF) and automobile shredder residue (ASR), which have been used in electrical power generation, heat recovery systems, and other applications. It was found that PCBs formed in primary combustion chambers largely decomposed by the time they reached secondary combustion. The incineration process increased the concentration of PCBs relative to PCB concentrations in the source material, and also increased the number of congeners present. ASR combustion produced a greater quantity of higher chlorinated PCB congeners than RDF combustion, possibly associated with chlorine abundance, copper, and iron metal present in the input samples (Ishikawa et al., 2007). These findings suggest that the unintentional formation of PCB congeners through combustion is linked to the composition of the source material. Documentation of PCBs being emitted from incineration processes elsewhere has occurred in recent years (Hogarh et al., 2018; Liu et al., 2018; Nguyen et al., 2016; Pham et al., 2019; Zhang et al., 2011), with dioxin-like PCBs commonly cited among the emitted species. However, it is unclear if these are contaminants simply released from incinerated products, or if they have been formed because of the thermal processes.

Sintering, steelmaking, and other smelting processes have also been recognized for their unintentional PCB production potential. In iron ore sintering, a thermal process that converts loose, fine particles into a solid mass through heat and/or pressure (Lu and Ishiyama, 2015), dioxin-like PCBs have been reported in fly ash (Li et al., 2017; Wang et al., 2016). Tian et al. (2012) similarly reported dioxin-like PCBs in sintering processes. However, PCBs were found to account for a relatively small amount of both total mass and toxic equivalency (TEQ) contributions; PCB congeners 81, 105, and 157 were most abundant. PCB-105 has also been detected in air close to iron-steel plants in Turkey, with emission estimates suggesting facilities with electric-arc furnaces might significantly contribute to PCB levels (Kaya et al., 2012). Dioxin-like PCBs have been reported as emissions from iron smelting facilities (Li et al., 2014; Liu et al., 2018), as well as from other non-ferrous smelting locations (Hu et al., 2014; Liu et al., 2018; Nie et al., 2012; Yang et al., 2020). Estimations of future PCB emissions suggest that unintentional production will account for increasingly higher percentages of total PCB emissions (Koshiba et al., 2019).

Dechlorination of higher chlorinated congeners is another possible pathway for unintentional PCB generation. Capozzi et al. (2019) found advanced microbial dechlorination of PCBs in wastewater samples. Decomposition of PCBs through dechlorination has also been detected in sediments (Takahashi et al., 2020), with some research suggesting that UP-PCB congeners can volatilize from bodies of water to the atmosphere (Apell and Gshwend, 2017).

5. Physical-chemical properties

PCBs are environmentally persistent, a result of their physicochemical properties. These properties have been widely studied, and experimentally derived values are easily accessible (e.g., Mackay et al., 2006). Some congeners, however, have garnered less interest and fewer studies have assessed their environmentally relevant properties. This includes many congeners that are considered to be unintentionally produced. Estimation methods, such as EPI Suite (USEPA), are therefore valuable in providing guidance and bridging these gaps. EPI Suite estimates for the notable congeners featured in this review are provided in Table 2. PCB-11, a major UP-PCB, shares in the environmentally persistent properties of other PCBs. It is poorly soluble in water (0.355 mg/L), and its $\log K_{OW}$ and $\log K_{AW}$ (5.27 and −2.02, respectively) would suggest that it is likely to accumulate preferentially in the organic phase (e.g. soil, lipids, vegetation). PCB-11 is a semi-volatile chemical ($V_p = 0.086$ Pa, $\log K_{OA} = 7.29$) and has been detected as a gaseous compound in the atmosphere with a modelled half-life of approximately 2.6 days. K_{OA}--model predictions of its partitioning to particles in air suggest that PCB-11 exists entirely in the gas-phase. Because of its properties, PCB-11 is expected to experience temperature driven surface-air exchange with preferential volatilization to air during warmer periods, and deposition to condensed states during colder periods. Additional information is provided in Table 2 and the Supporting Information.

6. Atmospheric transport and seasonal variation

6.1. Atmospheric transport

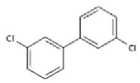
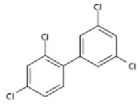
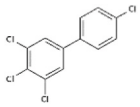
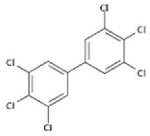
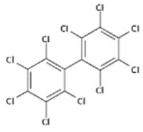
The transportation of contaminants from distant sources is a concern when it comes to UP-PCBs. This concern is amplified when considering remote regions, where there are no primary sources of pollutants. Possible pathways for POPs like PCBs to reach polar regions include cold trapping and fractionation (Wania et al., 1999; Wania and Su, 2004), and long-range atmospheric transport (LRAT; Larsson et al., 1992; Montone et al., 2003; Tanabe et al., 1983; Wania and Mackay, 1993; Wania, 2003). In addition, global distillation, or the grasshopper effect, has also been described as a potential transport mechanism. This form of fractionation is the repeated volatilization and deposition cycle that some POPs undergo (Gouin et al., 2004). For semi-volatile and persistent compounds such as PCBs, this means that even after global emission ceases, there will be a longer delay in their removal from the environment as they are subject to long-range transport through a series of air-surface exchange events.

Evidence for LRAT as a significant transportation process is seen through PCB congener profiles in polar regions. Lighter congeners have been found at increased relative fractions of $\sum$PCBs compared to higher chlorinated and heavier congeners (Hao et al., 2019, 2021; Khairy et al., 2016; Li et al., 2012a). This indicates that LRAT is a dominant cause of PCB presence in polar regions (Gambaro et al., 2005).

6.2. Seasonal variation

Airborne PCB concentrations have previously been shown to exhibit temperature dependence (Wania et al., 1998). Gas-phase concentrations of PCBs are generally impacted by variations in temperature, as there exists a positive correlation between their partial pressure and temperature (Hillery et al., 1997). PCB behavior due to temperature can be modelled through the Clausius-Clapeyron equation (Carlson and Hites, 2005). This equation can assist in determining whether emission sources

Table 2
Estimated physical properties of selected unintentional PCB congeners that are prevalent in outdoor air and toxicological studies.

Acronym	CAS N°	MW (g/mol)	Structure[a]	S_w[b] (mg/L)	V_p[b] (Pa)	logK_{OW}[b]	logK_{OA}[b]	logK_{AW}[b]	Henry's Law[b] (Pa-m^3/mol)	Φ[c] (%)	Half-life in Air[b] (h)
PCB-11	2050-67-1	223.1		0.355	8.65E-02	5.27	7.29	−2.02	17.5	0.020	62.6
PCB-68	73575-52-7	291.99		0.053	6.59E-02	6.34	8.09	−1.75	17.9	0.27	200
PCB-81	70362-50-4	291.99		0.053	1.13E-03	6.34	8.63	−2.29	6.19	1.3	338
PCB-169	32774-16-6	360.88		5.1E-04	7.75E-05	7.41	9.96	−2.55	11.2	26	845
PCB-209	2051-24-3	498.66		7.43E-06	1.41E-05	8.27	8.69	−0.415	0.107	80	1.41E+04

Abbreviations: MW = molecular weight, S_w = water solubility, V_p = subcooled liquid vapour pressure, K_{OW} = octanol-water partition coefficient, K_{OA} = octanol-air partition coefficient, K_{AW} = air-water partition coefficient, Φ = fraction on particles.
[a] Structure images retrieved from SciFinder.
[b] ChemSpider – Predicted Properties (25 °C), EPISuite KOWWIN v1.67 estimate. Where available, EPI Experimental Database values used.
[c] Calculated using GAPS Template (Harner, 2021) at 25 °C with assumed total suspended particle concentration of 25 μg/m.^{3}.

are local in nature, or a result of long-range transport. However, its functionality is limited to locations where concentrations of airborne pollutants are constant (Carlson and Hites, 2005). It is likely that the temperature dependence of PCBs is congener specific due to their varying chemical properties, and it is therefore more prudent to evaluate the seasonal variation and temperature dependence of individual congeners, rather than ∑PCB (Wania et al., 1998). Even within the homologue groups of PCBs, there is considerable variation in the vapour pressure of individual congeners. This variation is due to chlorine atoms and their respective positions, which has differing impacts on a congener's vapour pressure (Carlson and Hites, 2005). Atmospheric concentrations of medium molecular weight PCBs, such as tetra- and penta-chlorinated PCBs, exhibit greater temperature dependence than lower molecular weight PCBs, as their logK_{OA} values are more sensitive to temperature changes (Baek et al., 2010). Multiple studies confirm the seasonality of PCB-11 (Anezaki and Nakano, 2014; Hao et al., 2018; Hu et al., 2008), and total PCBs (Anezaki and Nakano, 2014; Baek et al., 2010). There is no evidence of seasonal variation to atmospheric PCB-209 concentrations (Anezaki and Nakano, 2014).

7. Analytical considerations

7.1. Sampling

Active sampling using high-volume air samplers has been applied to develop long-term data series in regards to atmospheric PCB concentrations. Such sampling programs include the Great Lakes Integrated Atmospheric Deposition Network (IADN, USA; Guo et al., 2018), the European Monitoring and Evaluation Programme (EMEP; Tørseth et al., 2012), the Northern Contaminants Program (NCP, Canada; Hung et al., 2010), and the Great Lakes Basin Monitoring and Surveillance program (Canada; Guo et al., 2018).

Sampling of unintentionally produced PCBs in the atmosphere through active collection has also been thoroughly demonstrated within literature (Anezaki and Nakano, 2014; Hu et al., 2008, 2010a; Khairy et al., 2016). Growing in prominence however are passive sampling methods to assess atmospheric PCBs. Sampling matrices such as polyurethane foam (PUF) disks have been used increasingly, especially under the GMP, to address regional data gaps in the measurement of PCBs (Melymuk et al., 2021; Pozo et al., 2006; Schuster et al., 2021; White et al., 2021). In comparison to active sampling means, passive sampling is advantageous to fill in these data gaps as there are lower costs associated with sample deployment and operation, as well as no need for established infrastructure or an electrical source. This can be an appealing option for sampling, as it allows for the collection of combined gas and particle phase samples (Markovic et al., 2015) in locations that may have otherwise had prohibitive or challenging requirements for sample deployment. Studies using passive sampling techniques for the detection of UP-PCBs are summarized in Table S1.

7.2. Application of PUF disks to PCB-11 sampling

Using the latest revision of the Global Atmospheric Passive Sampling (GAPS) template (Harner, 2021), uptake profiles of PCB-11 were modelled at various average temperatures. This template (see Supplementary Material) can provide the predicted uptake of PCBs (including PCB-11) by PUF disks as a function of deployment days and average site temperature over the deployment period. PCB-11 is used here to demonstrate the PUF uptake modelling by the GAPS template, given the body of literature supporting PCB-11 as a ubiquitous environmental contaminant. As seen in Fig. 5, PCB-11 remains primarily within the linear uptake region through a range of temperatures over 90 days (approximately three months, the standard PUF deployment time used by GAPS). Even for a high temperature of 25 °C, where sorption capacity of PUF disk is reduced, the PCB-11 sample air volume shows a minor reduction as it enters the curvi-linear uptake region towards the end of the sampling period, but does not equilibrate. This ability to maintain a nearly linear uptake of PCB-11, even at warmer temperatures, demonstrates the widespread spatial and temporal applicability of PUF disks as a sampling media for UP-PCBs. The effective air sample volumes

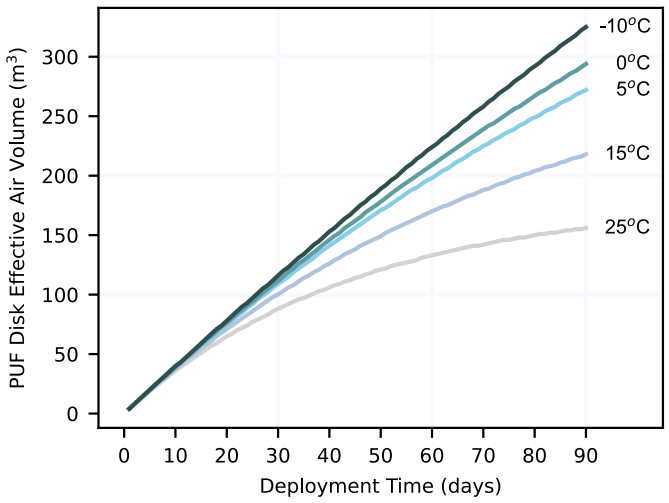


Fig. 5. Predicted uptake profile for PCB-11 using the PUF disk passive air sampler (Pozo et al., 2006) as a function of average temperature and deployment time. Calculations were performed using the GAPS Template and assuming a default sampling rate of 4 m^3/day (see Supporting Information for worksheet).

generated by the template can be used to convert the amount of PCBs collected on the PUF disk samplers to units of concentration in air.

7.3. Sample extraction and analysis

Details pertaining to UP-PCB sample extraction and analysis documented in literature can be found in Section S2. The methods for UP-PCBs are the same as those used for the analysis of PCBs from technical formulations.

8. PCB-11 in global air

PCB-11 has been documented in outdoor air globally (Fig. 6, Table S2). In North America, outdoor air measurements of PCB-11 were first reported in Chicago (Hu et al., 2008). Through multiple studies (Basu et al., 2009; Boesen et al., 2020; Hites, 2018; Hu et al., 2008), urban Chicago PCB-11 concentrations have ranged from undetected to 144 pg/m^3. Industrial sampling in Chicago found similar concentrations, with outdoor air levels ranging from 2.70 to 46.0 pg/m^3 (Martinez et al., 2015). The variation in reported concentrations can be attributed to temperature changes and the semi-volatile nature of PCBs. Hu et al. (2008) found PCB-11 to exhibit higher levels during summer months (48 ± 28 pg/m^3) compared to winter months (5.4 ± 4.9 pg/m^3). When the temperature was normalized, PCB-11 concentrations in Chicago ranged from <MDL (~1.0) to 72 pg/m^3, representing 5% of total PCB congeners in samples (Hu et al., 2008).

Further, PCB-11 has been reported at urban and industrial sites in Cleveland, Philadelphia, Seattle, East Chicago (Indiana), and Iowa City. Cleveland concentrations in urban air have been found to average 15.2 pg/m^3 (Hites, 2018), with a range of undetected to 307 pg/m^3 reported elsewhere (Basu et al., 2009). Levels in Philadelphia were similar to those in both Chicago and Cleveland, with concentrations ranging from 4 to 44 pg/m^3 (Du et al., 2009). In Seattle, the air-water exchange of PCBs was discovered to be a minor sink of PCBs from the Lower Duwamish Waterway Superfund site, resulting in PCB-11 concentrations of 9.1 pg/m^3 emitted into the local atmosphere (Apell & Gshwend, 2017). In East Chicago, located approximately 30 km southeast of Chicago, a range of concentrations from 5 to 8 pg/m^3 have been reported (Marek et al., 2017). Slightly higher levels were also found in Iowa City at 11 ± 9 pg/m^3 (Herkert et al., 2018).

Rural and remote locations, in contrast, exhibit lower PCB-11 levels than urban and industrial regions. Eagle Harbour, Sleeping Bear Dunes, and Sturgeon Point (all USA) show comparable air concentrations of PCB-11 with levels ranging from undetected to 53.8 pg/m^3, undetected to 38.1 pg/m^3, and undetected to 51.8 pg/m^3 respectively (Basu et al., 2009). It was then concluded by Basu et al. (2009) that PCB-11 concentrations are associated with human population density and presumably industrial activities. Average PCB-11 levels for Eagle Harbour, Sleeping Bear Dunes, and Sturgeon Point further exhibit comparable concentrations, confirming the results of Basu et al. (Hites, 2018). As a result of its low Aroclor PCB presence, Eagle Harbour was found to have the highest relative load of PCB-11 (Hites, 2018). The only Canadian location found in this review was Point Petre, where sampling occurred during 2004–2005. PCB-11 concentrations at this site were comparable to other Great Lakes studies with average concentrations of 6.27 pg/m^3 (Hites, 2018).

Studies examining PCB-11 in Asia have been extensive, focusing on atmospheric concentrations in China. Zhao et al. (2020) undertook a comprehensive study looking at 62 urban, rural, remote, and industrial sites. Among the 29 urban locations, the PCB-11 median value was 30.79 pg/m^3, ranging from 6.23 to 165 pg/m^3 (Zhao et al., 2020).

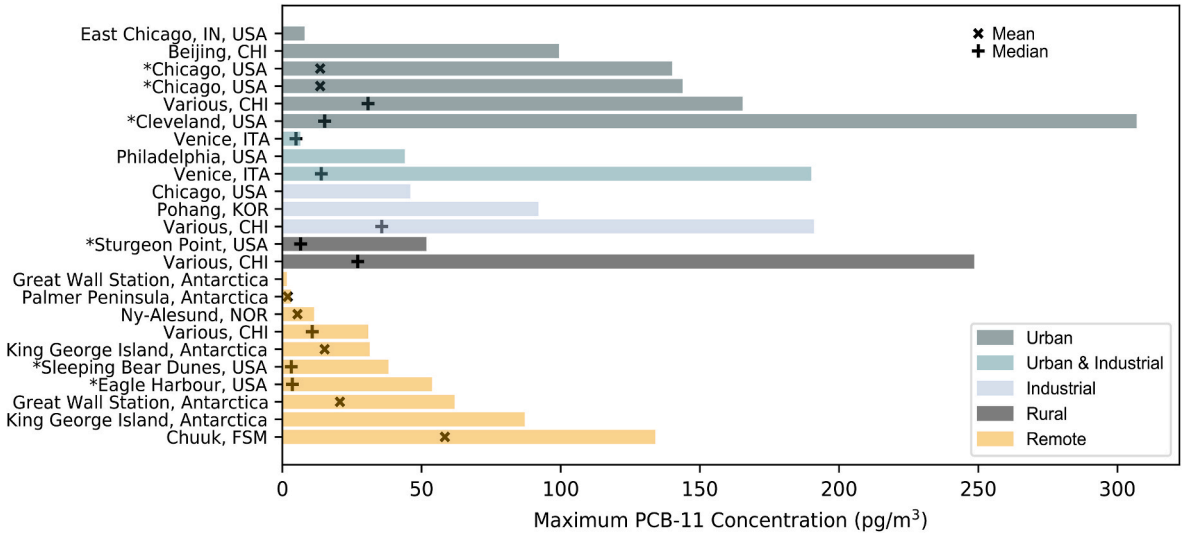


Fig. 6. Maximum observed atmospheric PCB-11 concentrations in air from literature sources, classified by sampling location type. Mean or median values indicated where available. Asterisks indicate locations where maximums and mean/median values are identified in separate studies. Additional details provided in Table S2.

Similar results were reported for the 14 industrial sites, with a median and range of concentrations reported of 35.7 and 10.4–191 pg/m^3, respectively (Zhao et al., 2020). In the more developed and populated regions along the east coast of China, electronic waste (e-waste) sites are more frequently found. These coastal e-waste sites exhibited slightly higher median concentrations than urban and industrial locations (Zhao et al., 2020). Rural sites exhibited comparable median PCB-11 levels to urban and industrial locations (27.1 pg m^3). However, these rural sites exhibited a greater range of results (15.1–249 pg/m^3; Zhao et al., 2020). Zhao et al. (2020) further detected PCB-11 in remote Chinese air ranging from <MDL (15.86) to 30.9 pg/m^3 (median 10.7 pg/m^3). Beijing-specific sampling by Hao et al. (2018) has found PCB-11 concentrations to range from 12 to 99.4 pg/m^3. A separate 2019 study reported background concentrations in Ningbo, China, averaging 89.9 pg/m^3 (Mao et al., 2019). While this is elevated in comparison to the aforementioned rural and remote air concentrations, it should be noted that the Ningbo site exhibited total PCB concentrations much greater than those reported elsewhere (Zhao et al., 2020). PCB-11 in Ningbo accounted for 8 ± 6% of $\sum_{209}$PCB, which is in line with previously mentioned relative PCB-11 loads in Eagle Harbour. Industrial sampling in Pohang, South Korea has shown PCB-11 levels similar to those in Beijing, as reported by Hao et al. (2018), ranging from 11 to 92 pg/m^3 (Baek et al., 2010). Assessments of PCB-11 in Japan have found that the congener is a major constituent (accounting for 3–14% of ∑PCB) in the air at both urban and industrial locations. Over a six-year sampling period, PCB-11 was found at average levels of 11 and 8.0 pg/m^3 in Sapporo (urban) and Muroran (industrial) by Anezaki and Nakano (2014).

The Arctic, Antarctic, and other remote regions have also received considerable interest in atmospheric PCB-11 detection. The majority of Arctic studies have been undertaken in Ny-Ålesund, Norway. Sampling conducted from 2004 to 2007 found average PCB-11 levels of 5.44 pg/m^3 (range of 0.78–11.4 pg/m^3; Baek et al., 2011). Sampling conducted by Choi et al. (2008) overlapped this, with sample collection occurring between 2005 and 2006. Their results showed average concentrations of 9 pg/m^3 at Ny-Ålesund. Later results (Hao et al., 2021) indicate PCB-11 levels at Ny-Ålesund range from 2.2 to 52.0 pg/m^3.

PCB-11 has also been detected in Antarctic air near the Great Wall Station, located in the Fildes Peninsula of King George Island, West Antarctica. Choi et al. (2008), who found average PCB-11 concentrations to be 60 pg/m^3, reported the earliest detection of PCB-11 in Antarctica. These average concentrations exceed some of the levels previously detailed in urban and industrial regions. Sampling conducted at a similar time by Baek et al. (2011) confirm the general range of these results. However, lower average concentrations were reported by Li et al. (2012a), giving mean levels of 15.2 pg/m^3 (range 3.60–31.4 pg/m^3). In both the results presented by Choi et al. (2008) and Li et al. (2012a), PCB-11 accounted for approximately 80% of ∑PCBs. It is suggested that there may be unique sources of PCB-11 present in the Southern Hemisphere, influencing these results (Choi et al., 2008). Other sampling conducted demonstrates overall lower levels than those previous studies (Li et al., 2012b; Wang et al., 2017) but confirms the findings of PCB-11 making up approximately 80% of ∑PCBs (Wang et al., 2017). Remote monitoring has also been conducted also in Chuuk, Micronesia, indicating an average PCB-11 concentration of 58.4 pg/m^3 and resulting in it being the dominant congener at this location (Baek et al., 2011).

Monitoring of PCB-11 is limited in Europe. However, the first study reporting the congener in the outdoor environment found levels in 2009 to range from <DL (1200 pg/sample) to 190 pg/m^3 (median 14 pg/m^3; Gregoris et al., 2014) and 2012 levels to range from 2.7 to 6.5 pg/m^3 (median 4.9 pg/m^3; Gregoris et al., 2014).

Long-term monitoring of PCB-11, beyond individual studies mentioned here, is lacking. Presently, monitoring programs that report findings to the GMP under the Stockholm Convention screen for a number of PCB species, however, PCB-11 is currently not included (UNEP, 2017).

In summary, PCB-11 has been detected at significant and relatively consistent concentrations globally, indicating the ubiquitous nature of the contaminant in outdoor and ambient air. Total PCB concentrations, by comparison, appear to be more varied on a global scale, suggesting that PCB-11 concentrations are not closely linked to Aroclor-PCB sources. Seasonal levels of PCB-11 vary, with numerous studies recognizing higher concentrations during the warmer summer months. Long-range atmospheric transport is suspected, given the presence of PCB-11 in remote Arctic and Antarctic environments.

Overall, determination of PCB-11 concentrations in outdoor air have found the congener to account from as low as 0%, and up to 84.6%, of ∑PCBs (Table S2). With a decline in emission sources of technical PCBs through restrictions of the Stockholm Convention, Aroclor PCB contributions to air profiles are expected to decline. As such, PCB-11 contributions to ∑PCBs are expected to increase, along with continued emission from other UP-PCB sources.

9. Other UP-PCBs in global air

While most non-Aroclor PCB testing has focused on PCB-11, other non-Aroclor PCBs have been reported at detectable levels in outdoor air. There are, however, inconsistencies in which congeners are screened for and reported. In some instances, studies examine the full suite of all 209 PCB congeners (Anezaki and Nakano, 2014; Boesen et al., 2020; Choi et al., 2008; Herkert et al., 2018; Hu et al., 2010a; Mao et al., 2019; Marek et al., 2017; Martinez et al., 2015; Zhao et al., 2020), and (perhaps inadvertently) monitor airborne concentrations of UP-PCBs. Other research has focused on monitoring dioxin-like PCBs, which includes congeners 81 and 169 as possible UP-PCB congeners, when considering the congener analysis of Aroclor mixtures by Shultz et al. (1989) and Frame et al. (1996) seen in Table 1. It should be noted that these co-planar PCBs can be detected within Aroclor mixtures, however they are also produced preferentially within industrial processes, such as combustion reactions. Through monitoring of these dioxin-like compounds, PCBs 81 and 169 have been detected in China (Die et al., 2015; Hao et al., 2018), South Korea (Heo et al., 2014; Shin et al., 2006), Venice (Gregoris et al., 2014), and remote polar regions (Hao et al., 2021; Khairy et al., 2016; Li et al., 2012a, 2012b; Wang et al., 2017).

Beyond these congeners, PCB-68 (Hombrecher et al., 2021), PCB-166 (Mao et al., 2020), PCB-209 (Kaya et al., 2012; Khairy et al., 2016; Li et al., 2012a; Wang et al., 2017), and various congeners that co-elute with known Aroclor congeners (Baek et al., 2010; Du et al., 2009; Gregoris et al., 2014; Hombrecher et al., 2021; Khairy et al., 2016; Mao et al., 2020) have been found in outdoor air samples. Beyond detection of PCB-11, Herkert et al. (2018) also found the presence of PCB-68 in outdoor air at concentrations ranging from <LOQ (50.7 pg/sample) to 0.80 pg/m^3. Their further analysis of sampled outdoor air profiles in Iowa City however indicated that the profile appears to be a mixture of Aroclors and PCB-11, suggesting that PCB-52 is not a prominent outdoor air contaminant. Rather, the emission of PCB-68 was suspected to be of greater importance in regards to indoor air.

Although PCB-11 remains the dominant focus in airborne UP-PCB monitoring, it is not the sole congener of interest. PCBs 81 and 169 have been detected in several instances, with other less frequently discussed congeners found in outdoor air as well.

10. Exposure and toxicity

Research concerning the exposure and toxicity of unintentionally produced PCBs is relatively limited. Most studies focus not on the direct impacts of UP-PCBs, but rather the hydroxylated and sulfonated metabolites of PCB congeners.

With the ubiquitous presence of PCBs in air, their deposition leads to uptake and accumulation within plant and animal species. This ultimately results in their presence in food sources. Dietary exposure

assessed using Toronto and Winnipeg food samples indicated that dietary exposure dominates for ∑PCBs in children and their mothers (Ampleman et al., 2015). Higher chlorinated congeners account for roughly half of all dietary PCB exposure. However, when it comes to individual lower chlorinated congeners, exposure through inhalation accounted for up to a third of total exposure, with mean inhalation exposure greater in children than in their mothers (Ampleman et al., 2015). Non-Aroclor PCBs have been detected in human serum as well, in a similar study also examining urban and rural adolescents and their mothers. It was discovered that non-Aroclor PCBs make up on average 10% of PCBs measured in human serum, with PCBs 11, 14, 35, and 209 being the primary and most prevalent congeners (Koh et al., 2015). Further examination has revealed the detection of PCB-11 (Roy et al., 2020; Sethi et al., 2017), PCB-11 sulfate (Grimm et al., 2017; Roy et al., 2020), and 4-OH-PCB 11 (Roy et al., 2020) in human serum. Unintentional PCBs and their metabolites have also been detected within human and animal organs following exposure including the liver, kidney, lung, and brain (Grimm et al., 2015; Zhang et al., 2021). Characterization of pulmonary exposure in rats showed that inhalation of vaporized PCB mixtures significantly contributes to the body burden of lower chlorinated congeners (Hu et al., 2010b).

Within the existing body of literature, unintentional PCBs and their respective hydroxylated and sulfated metabolites have been linked to several toxicological and health effects. PCB-11 and its metabolites, 4-OH-PCB 11 and 4-OSO_3-PCB 11 (PCB-11 sulfate), have been found to promote dendritic and axonal growth in neuronal cells, which poses the risk of neurodevelopmental effects in humans (Sethi et al., 2017). Further investigations of 4-OH-PCB 11 found that the metabolite induced a significantly higher estrogen-mediated activity than was detected through PCB-11, with PCB-11 itself found to induce the release of arachidonic acid, a precursor to inflammatory mediators. PCB-11 has also been found to inhibit gap junctional intercellular communication, which can lead to the promotion of tumor growth (Pěnčíková et al., 2018). In addition, 4-OH-PCB 11 has been reported to result in higher toxicity to some cell types (N27, SH-SY5Y, and HepG2) in comparison to PCB-11 and the corresponding sulfate (Rodriguez et al., 2018). It also significantly decreases the cellular growth and mitochondrial membrane potential of SIRT3-knockout mouse embryonic fibroblasts and significantly increases the expression of ten genes responsible for controlling fatty acid synthesis, metabolism, and transport (Alam et al., 2018).

Recent research has also found that the airborne deposition of unintentional PCBs is a pathway for biological uptake by marine mammals. In 19 mammal samples from the United Kingdom, at least 145 different PCBs were detected in each sample, with a majority exhibiting concentrations that exceeded toxicity thresholds (Megson et al., in press). In some species, such as the sei whale, it was further discovered that inadvertent PCBs contributed greater than 5% of the total PCB burden (Megson et al., in press).

11. Current research gaps and recommendations

While there exist a number of studies regarding UP-PCBs in air, the monitoring of these compounds is not widespread. Currently, monitoring programs reporting to the GMP are not required to assess levels of PCB-11. In cases where PCB-11 has been included in studies of outdoor air, levels were reported as trace to 20% of ∑PCBs, with values as high as 85% (Table S2). The levels and relative contribution of PCB-11 and other UP-PCBs are likely to increase over time. Airborne deposition of UP-PCBs has been linked to their detection in other environmental media such as marine mammals, where UP-PCBs were found to contribute more than 5% of the total PCB burden (Megson et al., in press). Non-Aroclor PCBs also contribute a substantial amount to PCBs in human serum, where they make up on average 10% of PCBs (Koh et al., 2015).

With the findings presented here, the following recommendations are made:

1.) That PCB-11 and PCB-209 be included as new indictor PCBs to monitor for non-Aroclor emissions, as well as including these congeners as analytes of interest to monitoring programs. This includes those monitoring programs reporting to the GMP, as it would develop a more comprehensive understanding of UP-PCB presence and trends in ambient air and other environmental compartments for assessing risk. Further consideration of congeners such as PCB-68, PCB-90, and PCB-189 as indictor PCBs could provide greater insight for UP-PCB source attribution (Megson et al., 2019).
2.) That PUF disks be considered as a cost-effective and simple tool for measuring PCBs/UP-PCBs in air. The uptake of PCB-11 by PUF has been characterized in this review.
3.) Finally, there is a need to better understand the production and emission sources of UP-PCBs. As presented here, pigment manufacturing and various industrial processes are acknowledged to be notable sources of these congeners. However, it is possible that sources exist beyond those currently recognized. It is therefore recommended that emission inventories be assembled, focusing on UP-PCBs to build on existing knowledge of these emerging compounds.

Author credit statement

Jacob Mastin: Conceptualization, Writing – original draft, Visualization; **Tom Harner:** Supervision, Conceptualization, Writing – review & editing; **Jasmin K. Schuster:** Conceptualization, Writing – review & editing; **Lauren South:** Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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