INHALATION EXPOSURE TO POLYCHLORINATED BIPHENYLS IN IZMIR

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İZMİR

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ABSTRACT

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Polychlorinated biphenyls (PCBs) are chemicals that are classified as hazardous air pollutants with carcinogenic and chronic-toxic effects on human health. People may be exposed to these chemicals indoors and outdoors via inhalation of indoor and outdoor air. Their production and use were banned worldwide with the Stockholm Convention. Since the Turkish government became an official signatory to the Stockholm Convention in year of 2010, the determination of persistent organic pollutants (POPs) in environmental compartments in Türkiye has gained importance. PCBs are still present in the environment because they are found in materials such as those used in heat transfer, electrical, and hydraulic systems manufactured before the ban due to their persistence to degradation in environment. Industrial processes have an important effect on their environmental concentrations due to unintentional emissions. Therefore, they are still present in air and investigations show that industrial and urban areas have higher levels. The aim of this study was to measure concentrations of PCB compounds in indoor-outdoor air samples collected at homes and schools, and to estimate exposure and associated carcinogenic health risks. Samples were collected at randomly selected sites in urban, semi-urban, and rural areas with polyurethane foam – passive samplers. Samples were analyzed by using gas chromatography–mass spectrometry after Soxhlet extraction. The estimated carcinogenic risks even at the 95^{th} percentile were calculated below the acceptable risk level (10^{-6}), indicating that the population carcinogenic risk associated with inhalation of gas-phase PCBs is not considerable. However, aggregate risks that could be estimated by addition of other plausible exposure pathways would drive the levels above the acceptable level.

ÖZET

İZMİR'DE POLİKLORLU BİFENİLLERE SOLUNUM YOLUYLA MARUZİYET

Poliklorlu bifeniller (PCB'ler), insan sağlığı üzerinde kanserojen ve kronik toksik etkileri olan tehlikeli hava kirleticileri olarak sınıflandırılan kimyasallardır. İnsanlar bu kimyasallara iç ve dış ortam havasının solunması yoluyla maruz kalabilmektedir. Stokholm Sözleşmesi ile bunların üretimi ve kullanımı dünya çapında yasaklanmıştır. Türkiye'nin 2010 yılında Stockholm Sözleşmesinin resmi olarak taraf haline gelmesinden bu yana, Türkiye'deki çevresel ortamlarda kalıcı organik kirleticilerin (KOK) belirlenmesi önem kazanmıştır. PCB'ler yasaklanmalarından önce üretilen ısı transferi, elektrik ve hidrolik sistemleri gibi malzemelerde kullanılması ve çevrede çok kalıcı olmaları nedeniyle halen tespit edilmektedir. Endüstriyel süreçlerin PCB'lerin çevresel konsantrasyonları üzerinde önemli bir etkisi vardır. Araştırmalar, sanayi ve kentsel alanların daha yüksek seviyelere sahip olduğunu göstermektedir. Bu çalışmanın amacı, evlerden ve okullardan toplanan iç-dış mekân hava örneklerinde PCB bileşiklerinin derişimlerini ölçmek, maruziyet ve ilişkili kanserojen sağlık risklerini tahmin etmektir. Örnekler, poliüretan köpük - pasif örnekleyiciler ile kentsel, yarı kentsel ve kırsal alanlarda rastgele seçilen noktalarda toplanmıştır. Numuneler, Soxhlet ekstraksiyonundan sonra gaz kromatografisi-kütle spektrometrisi kullanılarak analiz edilmiştir. Tahmin edilen kanserojen riskler, 95. yüzdelik düzeyinde bile kabul edilebilir risk seviyesinin (10^{-6}) altında bulunmuştur. Bu da gaz fazı PCB'lerin solunmasıyla bağlantılı kanserojen riskinin İzmir nüfusu için yüksek olmadığını göstermiştir. Bununla birlikte, solunuma diğer maruziyet yollarının eklenmesiyle bulunacak toplam risk seviyelerinin kabul edilebilir değerin üzerine çıkacağı tahmin edilmektedir.

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CHAPTER 1

INTRODUCTION

Persistent organic pollutants (POPs) are chemical compounds resistant to environmental degradation owing to their physicochemical properties. They are capable of being transferred over great distances, are widely encountered on a worldwide scale, and accumulate in the food chain, living organisms, and environment. POPs persist in the environment despite the primary emissions reduction and their prohibition of usage. Because POPs are semi-volatile organic molecules with low vapor pressure, they diffuse into the gaseous and organic phases. Since their distribution is primarily dependent on temperature and their affinity, they behave depending on the dominance of these factors among environmental conditions (Jones and De Voogt 1999). After being released into the environment, they are transported by air masses in the atmosphere until they precipitate once again. They are transported over long distances by repeated release-precipitation cycles (Lohmann et al. 2007). As a result, POPs are observed in remote areas, even at polar regions (Baek, Choi, and Chang 2011). Additionally, POPs are known as hydrophobic and lipophilic substances. POP chemicals are released into the atmosphere and deposited with the cooling of the air in the areas where they are transported and adsorb to suspended particles in the soil, water, and organic materials in sediment (Jones and De Voogt 1999). Their decomposition resistance is due to their extended half-lives in water, soil, and air and they accumulate due to their high octanol-air (K_{OA}) and octanol-water (K_{OW}) values.

Polychlorinated biphenyls (PCBs), a famous member of the POPs family, were first discovered in a fish over fifty years ago (Jiménez et al. 1999), which led to research on their environmental effects. Numerous investigations have been conducted since then to determine the prevalence of PCBs in different environmental media (Breivik et al. 2002). PCBs are a class of aromatic organic compounds made up of 209 congeners. They are classified into homologue groups (mono, di, tri, tetra, penta, hexa, hepta, octa, nona, and deca) based on their chlorination number (ranges from 1 to 10).

These chemicals, which are not found in nature, had been produced by a limited number of companies to be used in the industry for commercial purposes due to their non-

flammability, high boiling point, and high dielectric constant values. They are synthesized as a combination of homogenous chemicals. Depending on their chlorine level, they are sold underproduction of trade names, including Aroclor, Clophen, Kanechlor, Fenclor, Apirolio, Ascarele, Delor, Pyralene, and Pyronol. (Kimbrough and Jensen 2012). The most common closed systems that they had been used were capacitors and transformers, dielectric fluids, hydraulic fluids, and heat transfer materials. They had been used extensively in elastic sealants, polymers, and flame-retardant coatings in open systems, more limitedly in inks, adhesives, carbonless copy paper, conveyor belts, rubber products, fluorescent lamp ballasts, incision, lubricants, and metal coatings. From the beginning of the 1950s to the end of the 1970s, PCB-containing building materials were used in school buildings, official institutions, and homes. Although PCBs have been banned, they are still produced as a byproduct of industrial processes such as uncontrolled waste incineration, metal smelting and refining, thermal power generation, and industrial processes such as iron and steel production, and chlorine bleaching of pulp and paper (Thomas et al. 2012).

Six of the 209 congeners (PCB-28, -52, -101, -138, -153, -180) are encountered in relatively high concentrations in environmental media, probably due to their use in technical mixtures, therefore they are characterized as indicator congeners, and predominantly measured in monitoring studies ("IARC Monographs. On the Evaluation of Carcinogenic Risk of Chemicals to Man" 1977). When these compounds are ingested by live organisms, they accumulate in the fat tissues. Due to their widespread presence in the environment, everyone is chronically exposed, and depending on the degree of exposure, persistent toxic and carcinogenic consequences may occur (Jones and De Voogt 1999). Certain PCB congeners (PCB-77, -81, -105, -114, -118, -123, -126, -156, -157, -167, -169, -189) have dioxin-like toxicity, thus have higher cancer-causing potential. Their toxicity is represented as the equivalent of 2,3,7,8-tetrachlorodibenzo-para (TCDD or dioxin for short) (Alcock et al., 1998). Considering past studies, it has been reported that diet is the most important pathway of PCB exposure, especially after the presence of PCBs in high staple food products. However, recent studies have shown that inhalation route has become a comparable to dietary exposure (Chiu et al. 2004).

In Türkiye, PCB concentrations in air have been measured at several locations such as, Bursa, Izmir, Kütahya, Kocaeli, and Antalya (Can-Güven, Gedik, and Kurt-Karakuş 2019; Sari and Esen 2022; Kaya et al. 2012; Aydin et al. 2014). However, as will be presented in Chapter-2 Literature Review, these studies are either limited to one

environmental medium or a specific type of environment. These studies have also focused on either metropoles and industrial areas, mainly by sampling of food, e.g. fish, and ambient air, respectively. Studies in which exposure parameters are identified, and pollutant concentrations are measured, on the other hand, are very rare. There have only been a few studies in which concentrations in major exposure sources such as indoor air has been assessed. Consequently, in Türkiye, there is a lack of concurrently measured concentrations in multiple media and microenvironments pertinent to inhalation exposure, comprehensively estimated level of aggregate exposures, and associated health risks, especially those of children.

The purpose of this research is to determine the extent of environmental inhalation exposure to PCBs, and assess the associated health risks. This is the first study of its type in our country to estimate home and school exposures together for a primary/secondary school age group sub-sample by analyzing PCBs in both indoor and outdoor air, and at homes, schools, and cafe-bar-restaurants.

Detailed information on PCBs in the literature (Chapter 2), the materials and methods used in this research (Chapter 3), the results and discussion (Chapter 4) and the conclusion (Chapter 5) are presented in the following chapters.

CHAPTER 2

LITERATURE REVIEW

POPs have been widely examined internationally due to their detrimental effects. Considerable research has been completed on the PCB concentrations we examined in this study. The relevant data obtained from the research evaluated are discussed in-depth in the following sections. This chapter contains information on PCBs' chemical structures and properties, their potential sources, their transport and fate in the environment, their health effects, and the air concentrations of PCBs.

2.1. Persistent Organic Pollutants

POPs are gained significant attention on a global scale considering their persistency in the environment, transportation ability through the long-range, bio-accumulation tendency in soil media, and their detrimental impacts on the environment and human health (Jones and De Voogt 1999). Another reason it has become an essential issue in the environment, along with industrialization and urbanization, may result from intentional and unintentional sources. Contamination arises from traffic emissions, industrial processes, combustion products, improper waste management techniques (EPA, 2005). Therefore, PCB contamination is critical in industrial, urban, rural, and suburban areas for human exposure. Depending on the level of exposure, chronic toxic and carcinogenic effects may occur. Long-term exposures to PCBs cause health effects on humans, including an increased risk of cancer, reproductive diseases, immune system disruption, neurobehavioral damage, neurological effects, mutagenicity, and birth abnormalities (Vallack et al. 1998).

The United Nations Environment Programme (UNEP) called for a worldwide action plan on POPs in 1996, the first action taken. The International Forum for Chemical Safety (IFCF) recognized and listed 12 POPs in 1997. There was a new agreement made in 2001 called the Stockholm Convention. The countries that signed it, including Türkiye,

agreed to the decisions made with the agreement. These decisions came into effect in 2004. Then, the pollutant list was updated with the chemical groups added first in 2009 and then in 2010, and 2019 (Fiedler et al. 2019). POPs are a broad category of compounds that raise serious concerns. The PBCs are well-known POPs in this category. As a result of analyzing the PCBs' effects, numerous nations have enacted legislation to prohibit their use. PCBs first produced for commercial purposes in 1929 are among the first 12 compounds to be restricted from the environment. They are subject to Annex A (eliminates production and usage) and Annex C (chemicals unintentionally produced and released).

Türkiye and other parties must fulfill their obligations to the Stockholm Convention until 2025 (USEPA).

2.2. Chemical Structures and Properties of PCBs

PCBs are composed of two benzene rings bonded together by a single carbon-carbon bond in the general formula $C_{12}H_{10}$-n Cln, where n ranges from 1 to 10 (Anyasi and Atagana 2011). There are 209 different PCBs identified based on the number of chlorine atoms and where they are in the structure (see Figure 2.1.). As the chlorination level rises, so does the lipophilicity of PCB congeners. Their solubility in water is low, and their boiling points are high. Low electrical conductivity, high thermal conductivity, and resistance to thermal deterioration are some of the characteristics of these materials. PCBs are highly resistant to degradation, only they dissolve well in fats, oils, and organic solvents. A PCB compound becomes more stable and resistant to biodegradation as the number of chlorine atoms increases (WHO 2000).

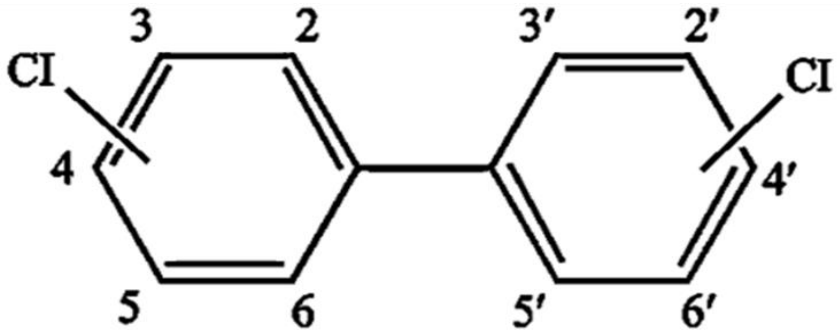

Figure 2.1. The general molecular structure of a PCB molecule in two dimensions

Because of their flame resistance, high chemical and thermal stability, high solubility in organic compounds such as hydrocarbons and oil, low electrical conductivity (dielectric constant), and high boiling temperatures, PCBs were used in various industries across the globe. More than one million tons of PCBs had been manufactured in different countries throughout the globe. PCBs are known by various trade names depending on the country where they are manufactured and the intended use for which they are designed (see Table 2.1.). Aroclor, one of the most widely used commercial compounds produced by Monsanto (Wiegel and Wu 2000).

Table 2.1. Some of the trade names of PCBs

Trade Names	**Country**
Aroclor, Pyranol	USA
Clophen	Germany
Kanechlor and Santothrem	Japan
Phenoclor and Pyralene	France
Askarel	United Kingdom
Fenclor	Italy

PCBs were first produced in the USA in mixtures containing different PCB homologue groups under the name "Aroclor" (such as Aroclor 1221, 1232, 1242, 1248, 1254, 1260) for commercial purposes. In these nomenclatures, the first two digits are the number of 12 carbon atoms, and the last two numbers (such as 21, 32, 42, 48, 54, and 60) indicate the rate of chlorination (Erickson and Kaley 2011). For example, the name "Aroclor" 1260 indicates 12 carbon atoms in the molecule and that it contains 60% chlorine. Table 2.2. shows the Aroclor mixtures for homologue groups of PCBs (Goel, Upadhyay, and Chakraborty 2016).

Table 2.2. Aroclor mixtures percentages by weight

Homologue Groups of PCBs	**Aroclor Names**						
	1221	**1232**	**1016**	**1242**	**1248**	**1254**	**1260**
Mono-CBs	50	26	2	3	–	–	–
Di-CBs	35	29	19	13	2	–	–
Tri-CBs	4	24	57	28	18	–	–
Tetra-CBs	1	15	22	30	40	11	–
Penta-CBs	–	–	–	22	36	49	12
Hexa-CBs	–	–	–	4	4	34	38
Hepta-CBs	–	–	–	–	–	6	41
Octa-CBs	–	–	–	–	–	–	8
Nona-CBs	–	–	–	–	–	–	1
Deca-CBs	–	–	–	–	–	–	–

PCB chemicals are categorized into homologue groups based on their degree of chlorination. Table 2.2. and 2.3. gives details on the properties of homologue groups, their chemical formula, molecular weight, vapor pressure, number of chlorine atom, boiling points and their IUPAC number.

Table 2.3. Detail information by homologue groups of PCBs

(Source: WHO, 2000)

Compounds	**Numbers**	**Chemical Formula**	**Molecular Weight**	**Number of Chlorine Atom**	**Vapor Pressure (Pa)**	**Boiling Points**
Monochlorobiphenyl	1-3	$C_{12}H_9C$	188.7	1	1.1	285
Dichlorobiphenyl	4-15	$C_{12}H_8Cl_2$	223.1	2	0.24	312
Trichlorobiphenyl	16-39	$C_{12}H_7C_{l3}$	257.6	3	0.054	337
Tetrachlorobiphenyl	40-81	$C_{12}H_6C_{l4}$	292	4	0.012	360
Pentachlorobiphenyl	82-127	$C_{12}H_5C_{l5}$	326.4	5	$2.6x10^{-3}$	381
Hexachlorobiphenyl	128-169	$C_{12}H_4C_{l6}$	360.4	6	$5.8x10^{-4}$	400
Heptachlorobiphenyl	170-193	$C_{12}H_3C_{l7}$	395.3	7	$1.3x10^{-4}$	417
Octachlorobiphenyl	194-205	$C_{12}H_2C_{l8}$	429.8	8	$2.8x10^{-5}$	432
Nonachlorobiphenyl	206-208	$C_{12}HC_{l9}$	464.2	9	$6.3x10^{-6}$	445
Decachlorobiphenyl	209	$C_{12}Cl_{10}$	498.7	10	$1.4x10^{-6}$	456

Table 2.4. Chemical names of dioxin-like PCB congeners

(Source:WHO 2000; EPA 1996)

IUPAC Number	Congeners
	Non-ortho congener
77	3,3',4,4'-Tetrachlorobiphenyl
81	3,4,4',5-Tetrachlorobiphenyl
126	3,3',4,4',5-Pentachlorobiphenyl
169	3,3',4,4',5,5'-Hexachlorobiphenyl
	Mono-ortho congener
105	2,3,3',4,4'-Pentachlorobiphenyl
114	2,3,4,4',5-Pentachlorobiphenyl
118	2,3',4,4',5-Pentachlorobiphenyl
123	2',3,4,4',5-Pentachlorobiphenyl
156	2,3,3',4,4',5-Hexachlorobiphenyl
157	2,3,3',4,4',5'-Hexachlorobiphenyl
167	2,3',4,4',5,5'-Hexachlorobiphenyl
189	2,3,3',4,4',5,5'-Heptachlorobiphenyl

2.3. Potential Environmental Sources

PCBs have no natural source, their production had been stopped, but they still exist in the environment. They were produced commercially as complex mixtures from 1929 to the 1970s. It is known that approximately 571,000 metric tons were produced in the USA during the production period. PCBs were used as insulating liquids in transformers and condensers (or capacitors) in heat transfer and hydraulic systems. They were also utilized as a solvent for ink in carbonless copy paper and as a heat transfer fluid. Besides lubricating and cutting oils, PCBs had been used in paints, adhesives, and insulating materials, heat transmission, and electrical and hydraulic systems, among others (Thomas et al. 2012). Despite being banned long time ago, it is believed that old appliances and electrical equipment are the primary sources of their existence. Because this equipment is mainly used indoors, PCB levels in indoor air are often much higher than in outdoor air. Some PCBs are released into the environment through uncontrolled

landfills and hazardous waste sites. Incineration of waste containing PCBs, improper disposal of leaks and spills from old electrical equipment, demolition of structures constructed using PCB-containing materials and processing of recycling scrap metals containing PCBs are sources of PCBs in the environment (W. Wang et al. 2013; Hazrati and Harrad 2006).

2.4. Fate and Transport

POPs released into soil, air, and water in various ways are involved in the transport process because they are resistant to degradation after entering the environment. The physical-chemical properties of each chemical are effective in the transport process. When examining the processes, octanol-air partition coefficient (log K_{OA}), vapor pressure, and solubility are determinants of transport processes. In addition, factors such as ambient temperature and prevailing wind direction are effective (Lohmann et al. 2007). The increasing number of chlorine atoms in PCBs gives information about the transport of the chemical. As the number of chlorine atoms increases, the log K_{OA} increases, while the vapor pressure and solubility decrease. Accordingly, as the number of chlorine atoms increases, their mobility decreases and they accumulate in the close environment. For low-chlorine PCBs, volatility differences due to physicochemical properties result in different atmospheric transport distances. PCBs accumulate in soil or water with dry or wet storage, bioaccumulation in sediment or aquatic organisms due to their low water solubility, or evaporate from the soil again with the heated air and mix with the atmosphere. Therefore, temperature change plays an essential role in the convection process. They tend to evaporate in warm climates and accumulate in cold climates (EPA, 2005).

2.5. Health Effects

Due to the chemical properties mentioned above, PCBs cause adverse health effects with their persistence in the environment and accumulation in the living cells

(ATSDR 200). Human beings are exposed to these chemicals by inhalation, ingestion, and dermal contact routes. So that the absorption of PCBs by humans and animals occurs through the skin, lungs, and digestive systems. The concentration and duration of exposure are essential in resulting adverse health effects.

Considering ingestion, which is one of the exposure routes, the phytoplankton comes to mind as the first step when inclusion of PCBs in the food chain. Phytoplankton, which can be seen as the first and vital layer in the food chain, draws attention. Phytoplankton is the primary food source for all marine organisms and an essential source of oxygen in the atmosphere. Considering the transfer of PCBs, their journey from phytoplankton may result in exposure to invertebrates, fish, and mammals and human exposure through consumption of PCB-containing food sources (Borja et al. 2005). Carcinogenic effects have been observed in animals in studies on PCBs. In addition, other adverse health effects; neurological effects, endocrine effects, diabetes, hypertension, cardiovascular disease, infertility, reproductive system disorders, hypothyroidism, recurrent infections, liver disease, asthma, arthritis, low birth weight have been proven to cause (EPA, 2005). The aryl hydrocarbon receptor (AHR) could call the dioxin receptor. AHR is a protein that in humans is encoded by the AHR gene. The aryl hydrocarbon receptor is a transcription element that controls gene expression. PCBs that are not ortho dioxins (PCB-77, -81, -126, and -169) bind to the AHR and induce dioxin-like toxicity in fish, birds, and mammals. The mono-ortho chlorinated dioxin-like PCBs may also bind the AHR and induce dioxin-like toxicity in birds and mammals. The toxicological consequences of exposure to dioxin-like PCBs in fish, birds, and mammals have been identified. This carcinogenic route is mostly by ingestion (Ludewig et al. 2008). Non-dioxin-like PCBs were appeared toxicological effects. More research on these congeners will likely be classified more specifically into additional subsets based on toxicological effects endpoints. Some toxicological effect examples of non-dioxin-like PCB congeners are endocrine disruption, neurotoxicity, immunotoxicity (Vallack et al. 1998). In other words, mechanisms of action estrogen receptor agonists/antagonists; serotonin biosynthesis inhibitors (Tala R. Henry and Michael J. DeVito 2003).

2.6. Air Concentrations of PCBs

This section provides information on the PCB concentrations in the air in various global locations. While searching the literature, current papers based on the degree of urbanization and air concentration samples collected from indoor and outdoor environments, respectively, using passive sampling as the sampling technique, from research conducted in the world and Türkiye, are included.

Bohlin et al. (2008), conducted simultaneous sampling with passive samplers in Mexico, Sweden, and England. Sampling sites were selected from suitable residential areas representing different household types. In general, the concentration range of PCBs was found to be 59-660 ng/m^3 in the outdoor air. In the sampling conducted in Mexico City, it was found that The urban area had a higher PCB concentration than those in semi-urban area in Mexico City. Pozo et al. (2016), collected samples from the island of Sicily in two periods from urban, rural, and background areas. PCB-28 was the most dominant congener detected during the first period (July-October), while PCB-52 was the second congener. In the second period (October-December), PCB-56 and PCB-44 were dominant congeners. Higher molecular weight PCB distribution has been observed in urban and industrial areas. Melymuk et al. (2016) reported PCB concentrations of the outdoor environments of the homes in Czech Republic in summer and winter seasons. Outdoor summer concentrations of PCB-28, -52, -101, -118, -138, -153, and -180 were higher than those of winter. It is also supported by this study that there is a typical increase for PCBs as observed in the literature due to increased evaporation from PCB sources at higher temperatures. In a study that was carried out in schools in USA, air samples were collected from both indoor and outdoor environments with a passive sampler by Marek et al. (2017). Outdoor PCB concentrations ranged from 0.03 to 3 ng/m^3 in samples collected between January 2012 and November 2015. Indoor air concentrations are mentioned in Table 2.5. Indoor concentrations were almost two times higher when compared to the outdoor environment. According to the samples taken from the urban and rural areas, no significant difference was observed in the outdoor concentrations. Pozo et al. (2017) collected samples from an urban area in India by passive sampling. Among the seven congeners (PCB-28, -52, -101, -118, -153, -138, -180) that were examined, PCB-28 and -52 were the most dominant congeners, while PCB-180 was the least common. Hogarh et al. (2018) conducted a study to assess atmospheric PCB concentrations by the

deployment of passive air samplers for 56 days. The measured PCB concentrations were the highest (4.64 ng/m^3) in the city where e-waste was known to be burned openly without control. Herkert, Jahnke, and Hornbuckle (2018) reported outdoor air PCB concentrations around homes measured by passive sampling. PCB-11 was the dominant congener in the mean outdoor air profile. It was compared with the outdoor concentrations taken from urban, high industrial activity and coastal areas (UK, Toronto, New Jersey) and they indicate that outdoor concentrations lower than in the literature for larger metropolitan areas. It was also found that the air profile was most similar to Aroclor 1254 and also showed contributions from 1248, 1242, and 1016.

Table 2.5. Ambient air PCBs concentrations measured around World

References	**Period**	**Location**	**Sampling Site**	**Mean ∑PCB Concentration ng/m^3**
(Bohlin et al. 2008)	March-April, 2006	Mexico City Gothenburg Lancaster (n=11)	Mexico City Urban	$\sum_{tot}$PCB= 430[a] pg/m^3
			Mexico City Semi-rural	$\sum_{tot}$PCB= 150[a] pg/m^3
			Gothenburg	$\sum_{tot}$PCB= 120[a] pg/m^3
			Lancaster	$\sum_{tot}$PCB= 120[a] pg/m^3

(cont. on next page)

Table 2.5 (cont.)

References	Period	Location	Sampling Site			Mean ∑PCB Concentration ng/m^3
(Pozo et al. 2016)	July-October, October-December, 2007	Island of Sicily, Italy	Period 1	East	RU	$\sum_{28}$PCB= 10 pg/m^3
					UR	$\sum_{28}$PCB= 220 pg/m^3
					UR	$\sum_{28}$PCB= ns
				Northwest	BA	$\sum_{28}$PCB= 70 pg/m^3
					BA	$\sum_{28}$PCB= 300 pg/m^3
					RU	$\sum_{28}$PCB= 10 pg/m^3
				West	RU	$\sum_{28}$PCB= 70 pg/m^3
					UR	$\sum_{28}$PCB= 40 pg/m^3
					RU	$\sum_{28}$PCB= 20 pg/m^3
			Period 2	East	RU	$\sum_{28}$PCB=ns
					UR	$\sum_{28}$PCB= 70 pg/m^3
					UR	$\sum_{28}$PCB= 120 pg/m^3
				Northwest	BA	$\sum_{28}$PCB= 40 pg/m^3
					BA	$\sum_{28}$PCB= 180 pg/m^3
					RU	$\sum_{28}$PCB=BDL
				West	RU	$\sum_{28}$PCB=BDL
					UR	$\sum_{28}$PCB= 70 pg/m^3
					RU	$\sum_{28}$PCB= 30 pg/m^3
(Melymuk et al. 2016)	July-August 2010, February-March 2011	Czech Republic	Urban (Homes)		Summer	$\sum_{7}$PCB= 38 ± 13[b] pg/m^3 $\sum_{7}$PCB= 34[a] pg/m^3
					Winter	$\sum_{7}$PCB= 17 ± 5.9[b] pg/m^3 $\sum_{7}$PCB= 19[a] pg/m^3

(cont. on next page)

Table 2.5. (cont.)

References	Period	Location	Sampling Site		Mean ∑PCB Concentration ng/m^3
(Marek et al. 2017)	January 2012 to November 2015	USA (n=108)	Urban and Rural	Urban School 1	$\sum_{209}$PCB= 0.210[a]
				Urban School 2	$\sum_{209}$PCB= 0.584[a]
				Urban School 3	$\sum_{209}$PCB= 0.183[a]
				Urban School 4	$\sum_{209}$PCB= 0.360[a]
				Rural School 1-2	$\sum_{209}$PCB= 0.159[a]
(Pozo et al. 2017)	December 28-March 27, 2014	India (n=5)	Urban		$\sum_{7}$PCBs=140.6 ± 64.5[b] pg/m^3 $\sum_{7}$PCBs= 53-213[d] pg/m^3
(Hogarh et al. 2018)	May– July 2010	Ghana (n=15)	Urban and Rural		$\sum_{190}$PCB= 0.28-4.64[d]
(Herkert, Jahnke, and Hornbuckle 2018)	August 22 to October 2, 2017	USA, Iowa City (n=16)	Urban	Residential Homes	∑PCB= 142[c] pg/m^3 ∑PCB=70-250[d] pg/m^3

[a]Median [b]Mean±STD [c]Average [d]Range

BDL below detection limit, ns not sampled/not available

Hazrati and Harrad (2006), performed indoor air sampling from various microenvironments with a passive sampler. Concentrations of PCBs in buildings constructed between 1950 and 1979 were substantially higher than those constructed after 1979, which led to the interpretation that the use of PCBs before the ban increased their concentrations in indoor air significantly. The median indoor air concentrations in urban

areas of Mexico City were approximately three times higher than those in semi-urban areas (Bohlin et al., 2008). Melymuk et al. (2016) measured indoor air PCB levels in two seasons. Concentrations in summer were higher than those in winter in residential buildings in the urban and the adjacent suburban areas. High indoor concentrations were associated with areas where buildings were constructed before 1984. The reason for this was stipulated to be Delor 103 PCB mixture was produced in Czechoslovakia before it was banned and being used as a paint additive in insulation, capacitors, and transformer fluid. Audy et al. (2018) also found higher PCB concentrations in old buildings than in new buildings in Canada and the Czech Republic. The levels were similar in the two countries with median values of 455 and 467 pg/m^3, respectively. In a study conducted in Australia, indoor air samples were collected from homes and offices by Herkert, Jahnke, and Hornbuckle (2018) using passive samplers. When compared with indoor concentrations of the schools reported in their previous study (Marek et al. 2017), it was determined that the homes had much lower concentrations than the schools. PCB-11 is the dominant congener, measured in indoor air, with 370 pg/m^3. Demirtepe et al. (2019) examined PCB concentrations in Slovakian homes near a PCB manufacturing plant and found no association between distance to the industry and PCB levels. Additionally, PCB-11 has been detected as a predominant congener in the indoor environment. Wang et al. (2019) was used a modified passive sampler to collect gaseous PCB and airborne particles for air samples. Eight PCB congeners (PCB-11, -28, -52, -101, -118, -138, -153, -180) were compared in offices and homes. The study revealed that PCB congeners showed very similar results in homes and offices. The dominant congeners were PCB-11, -28, and -153 in offices and homes. The presence of PCB-11 in the indoor environment has attracted attention. Because PCB-11 contribution was the highest compared other eight congeners, with a median value of 57 percent. A correlation was made between building age and PCB concentrations, and overall, an increase in PCB concentrations with age was observed. Adesina et al. (2021) measured PCB indoor air concentrations in six bars in Nigeria. A total of 26 PCBs congeners were analyzed and PCB-2 had the highest concentration in all sampling locations, with an average concentration of 0.086 $\mu g/m^3$, representing 21% of the total concentration. Dioxin-like PCBs in indoor air constituted 19% of total PCBs. Dominant homologue groups were determined as mono and tri-chlorinated congeners. Table 2.6. shows the detailed information about indoor air concentrations reported from around the world.

Table 2.6. Indoor air PCBs concentrations reported from around the World

References	Period	Location	Sampling Site		∑PCB Concentration ng/m^3
(Hazrati and Harrad 2006)	September 2003 and November 2005	UK (n=92)	Urban (West Midlands)	Home 1	∑PCB=2530[c] pg/m^3
				Home 2	∑PCB=589[c] pg/m^3
				Office 1	∑PCB=1319[c] pg/m^3
(Bohlin et al. 2008)	March to April, 2006	Mexico City Gothenburg Lancaster (n=35)	Mexico City Urban		$\sum_{tot}$PCB= 460[a] pg/m^3
			Mexico City Semi-rural		$\sum_{tot}$PCB= 160[a] pg/m^3
			Gothenburg		$\sum_{tot}$PCB= 500[a] pg/m^3
			Lancaster		$\sum_{tot}$PCB= 620[a] pg/m^3
(Melymuk et al. 2016)	July-August 2010, February-March 2011	Czech Republic	Urban (Homes)	Indoor Summer	$\sum_7$PCB= 89 ± 33[b] pg/m^3 $\sum_7$PCB= 82 pg/m^3 [a]
				Indoor Winter	$\sum_7$PCB= 61 ± 23[b] pg/m^3 $\sum_7$PCB= 61[a] pg/m^3
(Marek et al. 2017)	January 2012 to November 2015	USA (n=108)	From schools	Urban 1	$\sum_{209}$PCB= 7.873[a]
				Urban 2	$\sum_{209}$PCB= 111.829[a]
				Urban 3	$\sum_{209}$PCB= 2.045[a]
				Urban 4	$\sum_{209}$PCB= 37.010[a]
				Rural 1	$\sum_{209}$PCB= 22.172[a]
				Rural 2	$\sum_{209}$PCB= 7.781[a]

(cont. on next page)

Table 2.6. (cont.)

References	Period	Location	Sampling Site		∑PCB Concentration ng/m^3
(Audy et al. 2018)	June-August 2013	Canada(n=34) and Czech Republic (n=28)	Urban (Homes)	Canada	Σ_7PCB= 734 ± 162 [b] pg/m^3
				Czech Republic	Σ_7PCB= 661 ± 146 [b] pg/m^3
(Herkert, Jahnke, and Hornbuckle 2018)	August 22 to October 2, 2017	USA, Iowa City (n=160)	Urban	Residential Homes	ΣPCB=2830c pg/m^3 ΣPCB=450 to 6970[d] pg/m^3
(Demirtepe et al. 2019)	March–April 2015	Slovak Republic	Homes	Industrial area	$\sum_9$PCBs= 1090[a] pg/m^3
(X. Wang et al. 2019)	2015	Australia (n=28)	Urban (Homes and Offices)	Air (pg/m^3) (n=28)	Σ_7PCB=200 [a] Σ_7PCB= 270 ± 170 [b]
				Airborne particles (ng/g) (n=18)	Σ_7PCB= 32 Σ_7PCB= 60 ± 110 [b]
(Adesina et al. 2021)	-	Nigeria (n=6)	Urban (Public Bar)		Σ_{26}PCB=0.412[c] µg/m^3

[a]Median [b]Mean±STD [c]Average

Like the rest of the globe, research on PCBs have been conducted in Türkiye dominantly in outdoor air. Studies that measured the concentrations of PCBs in ambient air with a passive sampler are given in detail in Table 2.7.

Kaya et al. (2012) collected passive samples between July 2009 and May 2010, and analyzed Σ_{41}PCB congeners from the 40 different location of Aliağa industrial zone of Izmir. PCB levels were varied from 134 to 230,958 pg/m^3. PCBs of a low to medium molecular weight (tri–, tetra–, and penta–CBs) were the most prevalent chemicals in the air throughout the sampling period with increasing concentrations in summer season. In a following study in the same area (Aydin et al. 2014), the PCB congener with the greatest average concentration was PCB-28. In contrast, the congener with the lowest concentration was PCB 206 when considering the whole sample by 1300 and 20 pg/m^3, respectively. Concentrations of low molecular weight congeners (PCB-18, -28, -31, -33, -52, and -49) were higher in industrial areas than those in non-industrial sites throughout the year, regardless of the seasonal variation. This circumstance demonstrated that the Aliağa industrial zone had a role with the high PCB concentrations. The maximum observed total PCB concentration in the atmosphere was 29,051 pg/m^3 in the summer. Birgül et al. (2017) carried out a study in Bursa in 2014, concentration of Σ_{45}PCBs (PCB-18, -22, -28, -31, -41/64, -44, 49, -52, -54, -56, -60, -70 -74, -87, -90/101, -95, -99, -104, -105, -110, -114, -118, -123, -132, -138, -141, -149, -151, -153, -156, -157, -158, -167, -170, -174, -180, -183, -187, -188, -189, -194, -199 and -203) were measured. Concentrations in samples taken from urban, semi-urban, industrial, and agricultural areas ranged from 9.6 to 1,240 pg/m^3. The highest concentration was found in the industrial area with 280 ± 540 pg/m^3. Homologue-group concentrations were in the order from high to low as 3-Cl, 7-Cl, 6-Cl and 5-Cl. Median values of urban1, urban2, suburban1, suburban2, rural, agricultural and industrials by 36.7, 31.6, 45.4, 95.5, 23.4, 22.7 and 120 pg/m^3 respectively. When compared the median values industrial and suburban places concentrations were very close to each other. At 22 industrial/urban and 19 rural locations in Kutahya, Türkiye, Dumanoglu et al. (2017) collected ambient air samples (n = 82) in summer and winter. PCBs ranged from 19.6 to 675 pg/m^3 in winter and 31.6 to 230.2 pg/m^3 in summer. On average, PCB concentrations in the air were 1.5 times greater in the winter than in the summer. Low molecular weight congeners (PCB-17, -18, -28, -31, -33, -44, -49, and -52) dominated the atmospheric Σ_{41}PCB concentrations. Cetin et al. (2017) investigated atmospheric PCB concentrations in Dilovası, an important industrialized region of Türkiye. Concentrations were measured

with a passive sampler at 23 points for one year, with an average sampling period of 30 days. The highest concentration contribution belonged to the congeners with low molecular weight (PCB-18, -28, -31, and -33). When seasonal distributions were examined, it was observed that PCB concentrations decreased in winter. PCB levels were exceptionally high during the summer in urban and industrial areas. The PCB concentration clearly showed a decreasing trend with distance from industrial area. In addition to the three corner locations of Türkiye, sixteen provinces with urban and rural areas in Türkiye were chosen along the centerline from East to West and North to South by Kurt-Karakus et al. (2018). Four three-month sample periods were used: May–July 2014 (first period), August–October 2014 (second period), November 2014–January 2015 (third period), and February–April 2015 (fourth period) (4th period). Total 43 PCBs had an annual average concentration of 108 ± 132 pg/m^3. Kayseri (14.5 ± 14.3 pg/m^3) and Izmir (403 ± 428 pg/m^3) had the lowest and highest mean concentrations of total 43 PCBs in urban areas, respectively. At rural locations, the mean concentration was lowest in Aksaray (19.0 ± 22.7 pg/m^3) and highest in Kastamonu (217 ± 353 pg/m^3). Among the selected PCBs, the congener with the highest annual average was PCB-118, with a value of 26.3 ± 44.6 pg/m3. Rural locations had higher mean concentrations of PCB-104, PCB-114, PCB-118, PCB-123, PCB-151, PCB-167, and PCB-203 than urban sites. Penta-CBs, tetra-CBs, tri-CBs, hepta-CBs, octa-CBs, and hexa-CBs were the greatest to lowest contributors. Can-Güven, Gedik, and Kurt-Karakuş (2019), investigated PCB concentrations in agricultural areas in Antalya. The concentrations of 15 PCBs ranged from nd-213 pg/m^3 with a mean value of 28.0 to 52.1 pg/m^3. PCB-18, -20, -101, -105, and -170 were the most often detected PCB congeners, whereas PCB-52 and -118 were found in just one sample. PCB-28, -31, -44, -138, -149, -153, -180, and -194 were not detected. The following congener groups made a significant contribution to the total contribution of PCBs: Tri-CBs are followed by penta-CBs, hepta-CBs, tetra-CBs, and hexa-CBs. Sari et al. (2020), measured ambient air PCB concentrations in Bursa province in the summer and autumn. Summer and autumn average outdoor $\sum_{40}$PCBs (PCB-4/10, -16/32, -26, -28, -21/53, -22, -45, -52, -49/48, -41/64/71, -74, -66/95, -56/60, -92, -89/101, -119, -81/87, -85, -86, -77/110, -118, -114/149, -123, -153, -138/163, -126 and -180) concentrations were 303±183 pg/m^3 and 41± 23 pg/m^3, respectively. Samples were collected in Gemlik, Bursa, between January and December 2016 by Cindoruk, Sakin, and Tasdemir (2020). PCB levels ranged from 118 to 74 pg/m^3 on average. When the 12-month distribution of PCB concentrations in ambient air is examined, the months with

the highest concentrations were July, August, and September, respectively. The lowest concentrations were observed in December and January. Sari and Esen (2022) collected ambient air samples for 12 months in urban and suburban areas of Bursa. According to the homologue group distributions, tri-, tetra-, and penta-chlorinated PCBs were dominant in both sampling areas. Monthly average PCB concentrations in the urban area of Bursa were found to be between 271.4 and 826.6 pg/m^3 and between 243.2 and 727.2 in the suburban area. In both areas, the highest concentrations of PCBs were found in the months with higher temperatures (June and July). Additionally, the minimum PCB concentrations were identified lowest concentration in the urban and suburban regions in March and November, respectively.

Table 2.7. PCBs concentration reported in Türkiye ambient air

References	**Period**	**Location**	**Sampling Site**	**Mean ∑PCB Concentration pg/m^3**
(Kaya et al. 2012)	2009-2010	Izmir (n=159)	Industrial	$\sum_{41}$PCB= 2085[c] 8727[a]
(Aydin et al. 2014)	July-August, October-November 2009 January-February, April-May 2010	Izmir (n=160)	Industrial and suburban	$\sum_{35}$PCB= 349-94363[d]
(Birgül et al. 2017)	February 2014-December 2014	Bursa (n=5)	Rural	$\sum_{43}$PCB= 24.1± 8.20[b]
			Suburban	$\sum_{43}$PCB= 43.8± 24.4[b] $\sum_{43}$PCB= 180± 210[b]
			Urban	$\sum_{43}$PCB= 42.9±24.60[b] $\sum_{43}$PCB= 160± 280[b]
			Industrial	$\sum_{43}$PCB= 70± 150[b]
			Agricultural	$\sum_{43}$PCB= 84.2± 105[b]

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Table 2.7. (cont.)

References	Period	Location	Sampling Site		Mean ∑PCB Concentration pg/m^3	References
(Dumanoglu et al. 2017)	January 2014-March 2014 and June 02–August 05, 2014	Kütahya (n=82)	Indusrtial (n=22), Urban/Rural (n=19)		Summer	∑41PCB= 187.9± 132.9[b]
					Winter	∑41PCB= 125.3± 33.5[b]
(Cetin et al. 2017)	February 2015 to February 2016	Kocaeli (n=276)	Industrial region (Dilovası)			$\sum_{41}$PCB= 4152 ± 6072[b]
(Kurt-Karakus et al. 2018)	May 2014 to April 2015	Countywide (16 cities of Türkiye) (n=128)	1st Period		Urban	Σ_{43}PCBs= 117.54[a]
					Rural	Σ_{43}PCBs= 68.3[a]
			2nd Period		Urban	Σ_{43}PCBs= 106[a]
					Rural	Σ_{43}PCBs= 85.5[a]
			3rd Period		Urban	Σ_{43}PCBs= 98.7[a]
					Rural	Σ_{43}PCBs= 107[a]
			4th Period		Urban	Σ_{43}PCBs= 93.8[a]
					Rural	Σ_{43}PCBs= 71.4[a]
(Can-Güven, Gedik, and Kurt-Karakuş 2019)	May 22 to November 23, 2013	Antalya (n=17)	Agricultural region			$\sum_{15}$ PCB=28.0 ± 52.1[b]

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Table 2.7. (cont.)

References	Period	Location	Sampling Site	Mean ∑PCB Concentration pg/m^3	References
(Sari et al. 2020)	18th October to December 1, 2014 (44 days)	Bursa (n=8)	Urban (Residential area)	Summer	∑40PCB = 303 ±183[b]
				Autumn	∑40PCB= 41± 23[b]
(Cindoruk, Sakin, and Tasdemir 2020)	January 2016 - December 2016	Bursa (n=12)	Urban		$\sum_{81}$PCBs=118 ± 74[b]
(Sari and Esen 2022)	May 2017 and April 2018	Bursa	Urban		$\sum_{50}$PCBs=522.5 ± 196.9 [b]
			Sub-urban		$\sum_{50}$PCBs=439.5 ± 166.6[b]

[a]Median [b]Mean±STD [c]Average [d]Range

There is only one published study on indoor air concentration related to PCBs in our country, in Bursa carried out with passive sampling in the summer and autumn of 2014 (Sari et al. 2020). Samples were taken from living rooms and kitchens of eight different selected homes. The dominant PCB homologue groups throughout the entire sampling were penta, tetra, and tri by 40%, 23%, and 17%, respectively. In addition, average $\sum_{40}$PCB concentrations in living rooms and kitchens were almost twice as high in summer when compared to autumn concentrations. PCBs in living rooms and kitchens was 604 ± 210 pg/m^3 and 639 ± 2514 pg/m^3, respectively, fall was 362 ±167 pg/m^3 and 309 ± 93 pg/m^3, respectively (Sari et al. 2020).

2.7. Exposure and Health Risk Levels

PCB exposure and health risk assessments have been published in a variety of research conducted all over the world. Several studies performed in Türkiye are presented here to offer insight into our research (Ugranli et al. 2016; Dumanoglu et al. 2017; Cetin et al. 2018; Basaran 2018).

Ugranli et al. (2016) took ambient air samples from a suburban area of Izmir by active sampling method, and estimated exposure to PCBs. The 95th percentile exposure to $\sum_{32}$PCB was determined to be 1.83×10^{-3} and 121 pg/kg/day through inhalation and dermal routes, respectively, resulting in 95th percentile health risk levels of 2.49×10^{-7} and 3.81×10^{-12}. Dumanoglu et al. (2017), Ambient air samples were obtained from 22 industrial/urban and 19 rural sites in Kütahya. After analyzing the concentrations from 82 samples, the related inhalation exposure and risk were calculated by seasonal. In winter, the median cancer risk was 4.29×10^{-8} μg/kg/day as rural+urban+industrial. In summer, the median risk values are rural+urban+industrial 3.36×10^{-8} μg/kg/day.

Cetin et al. (2018) determined the mean risk for $\sum_{41}$PCB in winter, spring, summer and autumn seasons was determined 6.74×10^{-6}, 4.90×10^{-6}, 8.24×10^{-6}, and 7.07×10^{-6} pg/kg/day, respectively, in Dilovası. Mean summer exposure levels of $\sum_{41}$PCB were determined to be 509 pg/kg/day, whereas winter exposure levels were calculated to be 165 pg/kg/day.

2.8. Sources

Research in Türkiye have been published on the identification of PCB sources, findings of some of which are summarized below (Dumanoglu et al. 2017; Basaran 2018; Gungormus et al. 2021; Sari and Esen 2022).

Cetin et al. (2018) apportioned the sources of PCB concentrations measured in Dilovasi, which is characterized as a region with an important permanent source of organic pollutants due to heavy industrialization and heavy traffic in Türkiye. Four main sources were identified. Primary source was iron and steel manufacturing that accounted for 60% of the variation. The second, third, and fourth sources were coal and biomass combustion (29%), technical PCB mixes (6%), and industrial emissions (4%), respectively. In the study conducted by Gungormus et al. (2021), the active sampling method took samples from Istanbul and Izmir, Türkiye. The study shows that the primary sources of PCB emitters as iron-steel plants, ship-breaking plants, oil, and petrochemical industrial plants. In addition, there is a secondary source situation that comes from the soil and sea through re-volatilization. In the study carried out by Sari and Esen (2022) in Bursa, where urban annual average PCB concentrations were higher than those of

suburban area since the urban area was said to be close to the industry. The industry is cited as an important source for PCBs in this case. When the seasonal analysis is observed, it is seen that the increase in temperature increases the dominance of Di-CBs. In addition, the predominance of dichlorinated PCBs was thought to be influenced by atmospheric transport of PCB concentrations during these months. PCBs with low chlorination levels were prevalent in urban and suburban areas. That was explained because the gas phase compounds were collected by diffusion using the PUF-disk sampler.

2.9. Motivation and Objectives

As a result of reviewing studies conducted in İzmir and throughout Türkiye, it has been concluded that PCB concentrations in İzmir are at considerable levels in comparison to the those reported in the literature, even though they were not produced in our country. However, levels of aggregated exposure from air at home and school, and associated health risks are not known at any place in the country. Therefore, the main goal of this study was to estimate the above mentioned exposure/risk for the Izmir population. The objectives of this study were:

i.) To investigate PCB concentrations based on homologue groups

ii.) Comparison of PCB concentrations of indoor and outdoor environments

iii.) Source apportionment based on principal component analysis

iv.) Estimation of lifetime average daily dose as the inhalation exposure measure, and estimation of associated carcinogenic risks for the Izmir population

CHAPTER 3

MATERIALS AND METHODS

The sampling sites, sample collecting techniques, experimental procedures, data analysis methods, and exposure and health risk assessments are presented in this chapter.

3.1. Sampling Sites

Sampling for this study was conducted in the Province of İzmir, located in the Aegean Region of Türkiye with a population of 4,367,251 (TSI 2019), the third most in the country. Summers in İzmir are hot and dry, while winters are warm and rainy. Air samples were taken at 21 (7 urban, 7 semi-urban, 7 rural) points in İzmir. Sampling included indoor-outdoor air (n = 84) samples from schools, and homes. Selection of the 21 points considered the prevailing wind direction that blows from the North of the province where Aliağa Industrial Area (AIA) is located to the metropole and southern districts. AIA is considered a significant source of pollutants due to potential pollution sources such as a large oil refinery, petrochemical complex, steel rolling mills, sorting sites, chemical fertilizer plants, and heavy road and rail traffic. Figure 3.1. shows the sampling points on a map along with AIA and Izmir metropole.

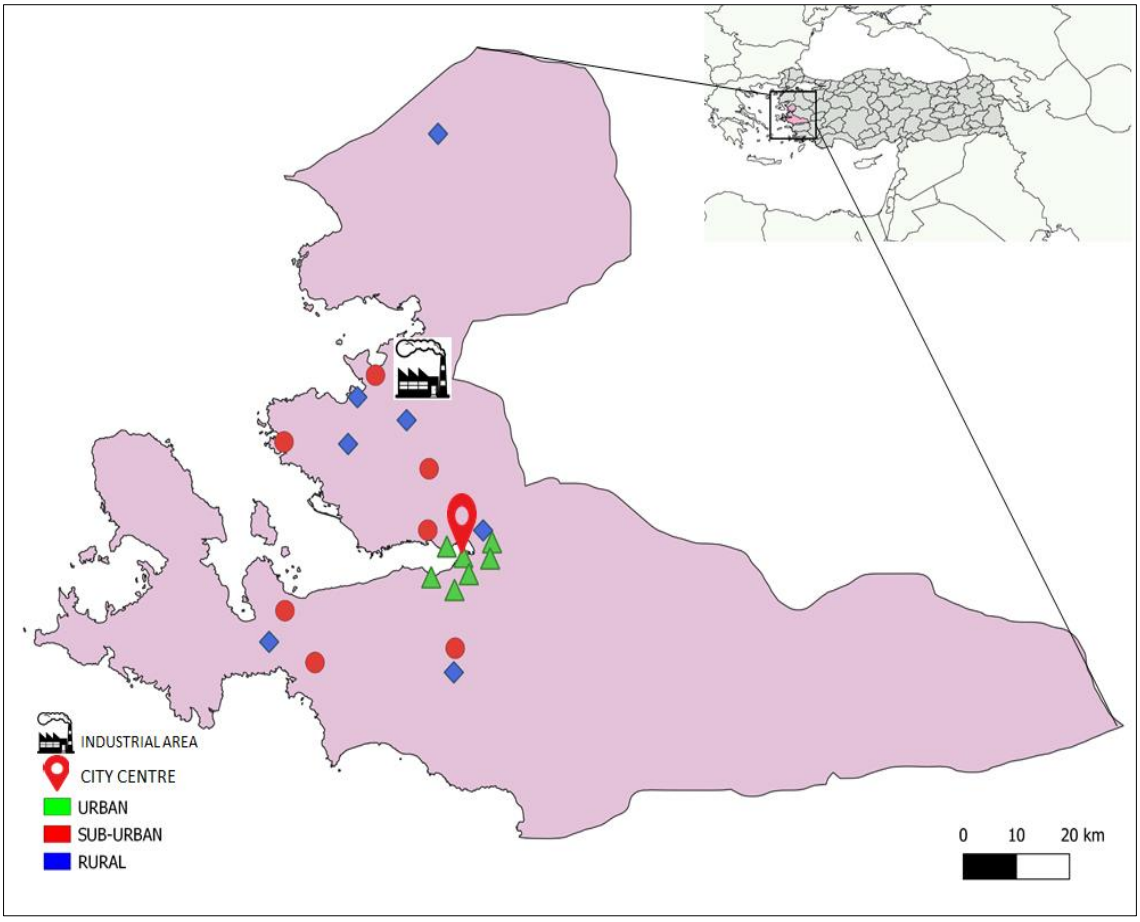

Figure 3.1. Izmir location and sampling points

Selection of the sampling points were random other than being located on a North-South transect through the province, placing them on variable proximity to the AIA. Schools were selected as the main points (Table 3.1.). Then, a residence was sought in close proximity to the schools to participate in the study. Detailed data regarding sampling are given in Table 3.1. Sampling was done in a way that would not disrupt education and training in schools. Indoor samples were taken in classrooms in schools and in the living room in homes while outdoor samples were taken from the playground in the school campus and in the balcony or outside a window at homes. The classrooms close to toilets and types of microenvironments were avoided. The deployment/collection period of each air sample was about one month.

Table 3.1. Sampling location information

Sample Code	Sample Location	Latitude	Longitude	Duration	Days	Average Temperature (°C)
UR1	Karşıyaka	38.45546	27.11238	19.11.2019-20.12.2019	31	12.35
UR2	Bornova	38.46388	27.20907	24.09.2019-22.10.2019	28	21.3
UR3	Bornova	38.43236	27.20491	24.09.2019-20.10.2019	26	21.3
UR4	Alsancak	38.43548	27.14771	24.09.2019-22.10.2019	28	21.3
UR5	Buca	38.40200	27.15947	23.09.2019-23.10.2019	30	21.3
UR6	Karabağlar	38.37151	27.12873	23.09.2019-23.10.2019	30	21.3
UR7	Güzelyalı	38.39594	27.07895	23.09.2019-23.10.2019	30	21.3
SU1	Aliağa	38.78691	26.95967	16.01.2020-24.02.2020	39	9.2
SU2	Foça	38.65786	26.76404	14.01.2020-20.02.2020	37	9.2
SU3	Menemen	38.60586	27.07436	14.01.2020-24.02.2020	41	9.2
SU4	Çiğli	38.48700	27.07159	19.11.2019-20.12.2019	31	12.35
SU5	Menderes	38.25868	27.13023	14.11.2019-16.12.2019	32	12.35
SU6	Seferihisar	38.23098	26.83036	14.11.2019-16.12.2019	32	12.35
SU7	Urla	38.33134	26.76605	17.09.2019-23.10.2019	36	21.3
RU1	Aliağa	38.74440	26.92111	16.01.2020-24.02.2020	39	9.2
RU2	Aliağa	38.69986	27.02637	16.01.2020-24.02.2020	39	9.2
RU3	Gerenköy. Foça	38.65382	26.90110	14.01.2020-20.02.2020	37	9.2

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Table 3.1. (cont.)

Sample Code	Sample Location	Latitude	Longitude	Duration	Days	Average Temperature (°C)
RU4	Kozakyaylası. Bergama	39.25393	27.09344	21.01.2019-28.02.2020	38	9.2
RU5	Laka Köyü. Bornova	38.48660	27.18998	28.11.2019-31.12.2019	33	10.5
RU6	Küner. Menderes	38.21198	27.12728	14.11.2019-16.12.2019	32	12.4
RU7	Kuşçular, Urla	38.27089	26.73222	28.11.2019-31.12.2019	33	10.5

3.2. Sample Collection

For indoor and outdoor air sampling, passive samplers with polyurethane foam (PUF) discs (Tisch TE-1014; 14 cm diameter; 1.35 cm wide; 365 cm surface area; 207 cm^3 in volume; 0.0213 g/cm^3 density; 4.40 g mass) were used. A detailed visual of the sampler is given in Figure 3.2. Indoor and outdoor air samples were collected at schools and homes. A cleaning procedure was applied to all PUF discs to remove possible organic contaminants before sampling. First, PUF discs were soaked in water with Alcanox overnight and then thoroughly rinsed with tap and distilled water. Then, 24-hour cleaning was performed in a Soxhlet extractor with 1:1 acetone:hexane (ACE:HEX). After extraction, the PUF discs were wrapped in aluminum foils pre-burned at 450 °C and placed in a vacuum oven to dry completely. After drying, PUF discs were placed in zip-lock bags and stored in a deep freezer at -20 ˚C until they were taken to the sampling points. At the sampling points, PUF discs were placed in passive samplers with the help of clean tweezers. The passive air sampler was placed in a standard classroom for indoor sampling in schools (away from potential indoor sources such as toilets and canteens). The trees in the school gardens were mainly used for outdoor sampling. The selected points were the living rooms in the residences, which are the rooms where the occupants spend the most time. The balconies of the homes were preferred for outdoor sampling. Indoor and outdoor air samples were collected from schools and homes at the end of

approximately one month. While collecting the samples, the PUF discs were wrapped in pre-burned aluminum foils and stored in locked bags at -20˚C at the laboratory until extraction.

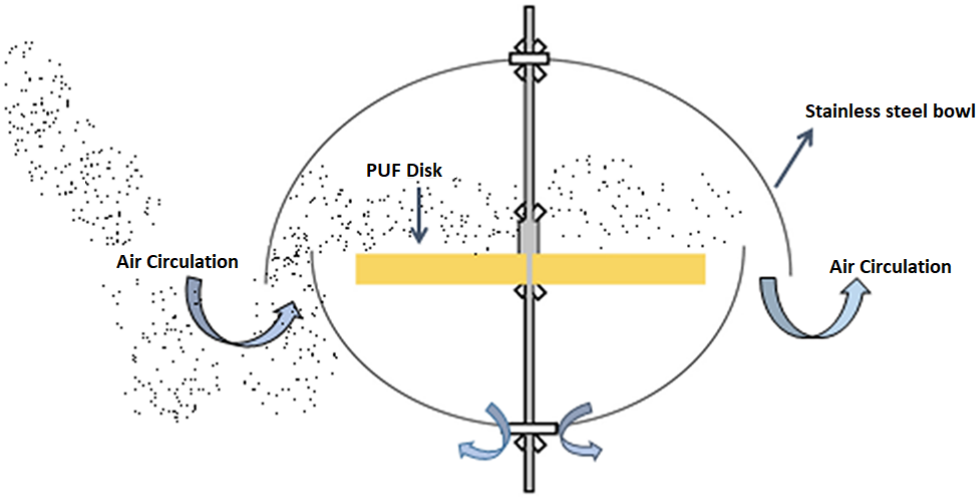

Figure 3.2. Passive air sampler in detail

Table 3.2. Passive air sampler detailed information

PUF disk characteristics		
Volume (cm^3)	207	
Diameter (cm)	14	
Thickness (cm)	1.4	
Density ($g\ cm^{-3}$)	0.021	
Mass (g)	4.4	
Sampler housing dimensions (cm)		
Upper dome	Diameter	28
Upper dome	Depth	8.0
Lower dome	Diameter	22
Lower dome	Depth	8.0

3.3. Sample Preparation and Handling

The sample preparation and management procedure were applied to every material used in the experiment process. It aims to reduce the errors related to laboratory equipment and to obtain high-reliability results by eliminating contaminants before each experiment. For the extraction of the pollutant groups, the materials and chemicals to be used were carefully selected, and appropriate precautions were taken in order not to cause contamination in the samples. The chemicals to be used for the cleaning of the samples were of high quality and baked before use, preventing any contaminant interference from the laboratory environment after production or during waiting. Glass and metal materials to be used were kept in an Alconox bath, rinsed with tap water and distilled water, passed through acetone and hexane, and kept in an oven at 105°C, with their mouths closed with aluminum foil. Glass and metal materials were rinsed again with an organic solvent just before use.

3.4. Sample Extraction and Clean-up

Recovery surrogate standards (PCB-14, PCB-65 and PCB-166) were spiked prior to extraction of indoor and outdoor PUF discs. PUF discs were extracted with 300 mL of a 1:1 mixture of acetone: hexane for approximately 24 hours using the Soxhlet extraction method. Extracts were concentrated to approximately about 2 mL using a rotary evaporator. Following concentration, a single-step clean-up procedure was applied using Si-based solid-phase extraction (SPE) cartridges (Agilent Bond Elute Mega BE, Si 5g) and a vacuum manifold. Applied elution solvents were 30 ml hexane and 30 ml acetone in sequence. Samples were transferred into GC vials after another concentration and solvent exchange (to isooctane) detailed illustration of SPE cartridges given in Figure 3.3.

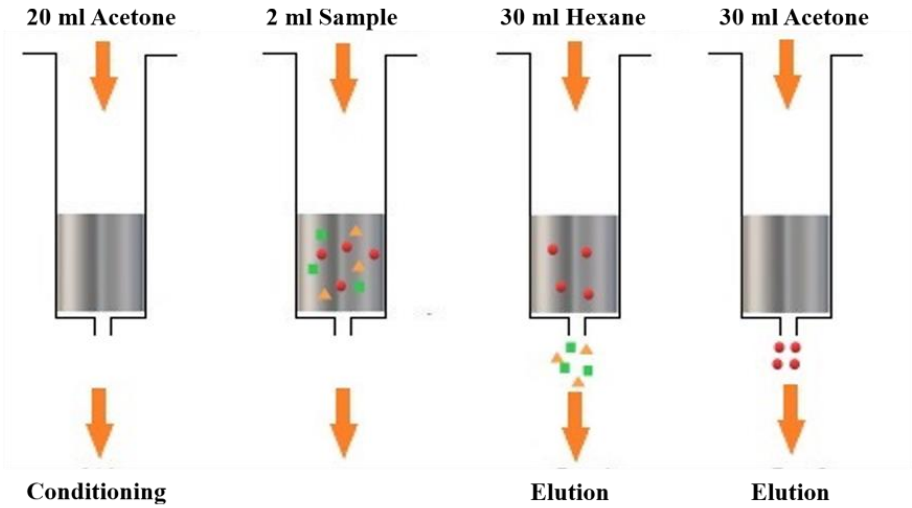

Figure 3.3. Detailed Illustration of Clean-up Procedure

3.5. Instrumental Analysis

The operational parameters utilized to analyze PCBs are described in Table 3.3. and 3.4., Thermo Scientific brand gas chromatography-mass spectroscopy (GC-MS) device (Trace Ultra-ISQ) was used in electron effect ionization mode for PCB analysis with the created SIM windows.

Table 3.3. GC/MS operating conditions

Operating Conditions	**PCB Analysis**
Column	60 m, 0.25 mm, 0.25 μm, Rxi-5Sil MS
Ionization	Electron Ionization (EI)
Carrier Gas	Helium 1.1 mL/min
Injection Mode	Splitless (NCI)
Injection	250 ° C
Injection Volume	2 μL
Ion Source	230 ° C

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Table 3.3 (cont.)

Operating Conditions	PCB Analysis
Quadrupole Temperature	160 ° C
Auxilary temperature	300 ° C
Tempereture program	Initial Oven Temperature 90° C for 1 min. Raised to: 160°C at 15°C/min, 210°C at 3°C/min, 310°C at 10°C/min (holding for 7 min).

Table 3.4. SIM parameters of PCB analysis

PCB Congeners	Ion (*m/z*)	Time (min)
-4, -10, -6, -7, -8, -5, -14, -19, -12, -18, -17, -15, -27, -24, -26, -34, -29, -31, -25	222, 224, 152, 256, 258, 260	14.00-24.00
-32, -28, -20, -33, -22, -45, -46, -69, -52, -49, -47, -48, -65, -104, -44, -59, -42, -37, -71	256, 258, 260, 290, 292, 324, 326, 328	24.00-27.50
-41, -64, -103, -40, -67, -63, -74, -70, -93, -95, -66, -91, -56, -92, -60, -84, -101, -99, -119	290, 292, 220, 324, 326, 328, 254, 256	27.50-30.00
-83, -97, -87, -115, -85, -128, -110, -77, -82, -134, -135, -136, -147	290, 292, 326, 328, 324, 360, 362	30.00-31.00
-107, -149, -123, -118, -144, -114, -131, -146, -153, -132, -105, -179, -141, -137, -176, -164, -138, -151, -178, -129, -166, -187, -183, -157, -167, -185	324, 326, 360, 362, 254, 328, 290, 394, 396	31.00-33.50
-174, -177, -171, -156, -173, -158, -172, -197, -180, -193, -190, -170, -199, -196, -203, -189, -208, -195, -207, -194, -205, -206	394, 396, 360, 362, 426, 428, 462, 392, 237, 272	33.50-44.33

3.6. Quality Assurance/Quality Control

3.6.1. Calibration Linearity

For all congeners, the linear fit was determined to be acceptable (R^2 >0.988; Sofuoglu et al. 2011). Calibration experiments using 7-9 points were conducted. PCB-8 had the lowest calibration linearity (Figure 3.4.).

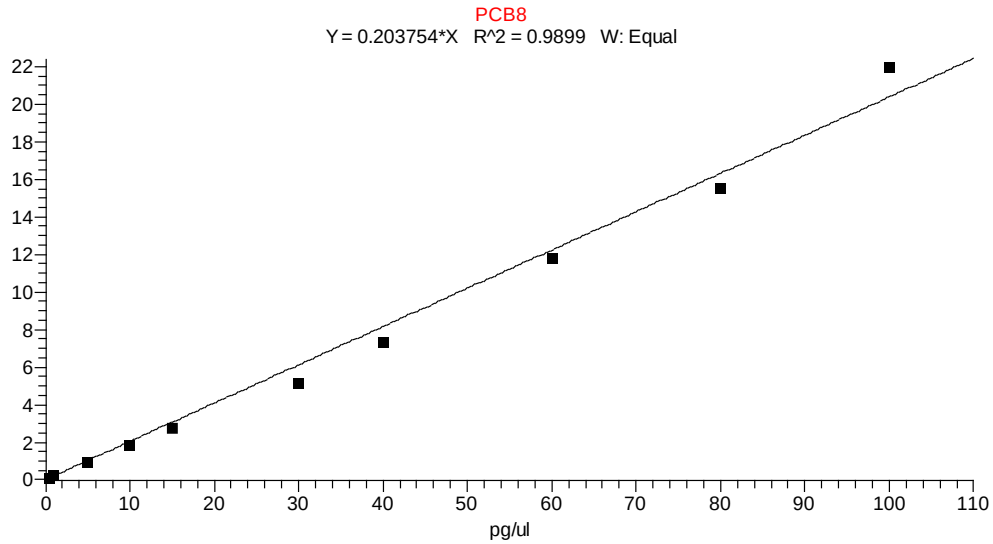

Figure 3.4. The calibration graph obtained from the PCB-8

3.6.2. Procedural Recovery

Prior to extraction, each sample was spiked with 50 ng surrogate standards (PCB-14, -65, -166) to determine the recovery efficiencies. The recovery efficiency of the procedure was computed as follows:

$$\text{Recovery Efficiency (\%)} = 100 \times (Cf / Ci) \qquad (3.1)$$

Cf denotes the concentration of the spiked sample processed as an actual sample, and Ci represents the initial concentration in the spike solution. Table 3.5. lists the statistical recovery (mean±SD) results.

Table 3.5. Recovery of Surrogate Compounds

Surrogate	**Passive (n=51)**
PCB-14	78.12±11.2
PCB-65	75.6±8.2
PCB-166	101±8.7

3.6.3. Detection Limits

This study prepared separate laboratory blank samples for passive air samples. Blank PUF disk were pre-cleaned before use with Soxhlet extraction. Blank PUF disks were used without any sampling process and surrogate standard spiked prior to extraction. MDL (method detection limit) was calculated using the mean blank concentration The MDL value was determined by adding the t-test (the corresponding number of degrees of freedom of the detected sample number) × standard deviation to the mean concentration obtained. In cases such as the MDL cannot be determined in blank samples, the IDL (instrument detection limit) was defined as the concentration at which the congener was not detected. Five separate blanks of clean PUF were processed for passive air samples. IDL was used as the mean blank concentration obtained from the extraction of clean PUFs in circumstances where MDL was not calculated.

3.6.4. Blank Correction

Concentrations reported in this study are blank corrected. Throughout the investigation, five PUF blank samples were analyzed using the procedures described in the previous sections. The mean values for each PCB congener was calculated in the blank samples and subtracted from the sample masses, referred to as "blank correction." PCB-6, -7, -17, -20, -29, -33, -34, -37, -40, -42, -46, -47, -67, -71, -74, -87, -97, -149, -151, -153, -176 were not identified in the blank samples.

3.7. Sampling Rate Calculation

The sampling rate was calculated for the passive air sampler. For samples taken from the outdoor environment, the sampling rate (Rs) was calculated using the Global Atmospheric Passive Sampling (GAPS) model (Herkert et al. 2018). The model calculates the sampling rate based on hourly meteorological data and values such as the chemical's molecular weight, $LogK_{OA}$, and Du_{OA}. The sampling rate is commonly achieved using depuration compounds (Birgül et al. 2017; Wilford et al. 2004). However, the depuration method was not used in the current study because the emission from depuration compounds are of health concern for voluntary study participants. The model has been validated against multiple studies comparing previously calibrated results with depuration compound results (Marek et al. 2017). Outdoor mean sampling rate values in this study ranged from 3.6 to 4.7 $days^{-1}$.

It was not considered appropriate to use the GAP model for samplers deployed indoors. For this reason, literature data were used for indoor air sampling. Among the studies examined in the literature (Hazrati and Harrad 2007; Persoon and Hornbuckle 2009), a sampling rate of 2.6 m^3/day was used (Persoon and Hornbuckle 2009). It was taken into account that Persoon and Hornbuckle (2009) determıned an average samplıng rate in the laboratory environment. The average ventilation rate in laboratory environments (ASHRAE 2016) has been reported to be 0.18 cfm/ft^2 . In our study, natural ventilation is used in all schools and homes. In the literature, the air exchange rate (ACH),

which creates a healthy environment for natural ventilation, is 0.5 ACH (Sundell et al. 2011). The equation 3.2 gives us how Veff related with ACH (Luongo et al. 2016). The average class and home volume in Türkiye was taken as 144 m^3 based on the sampled schools in this study, and the "average" sampling rate value for indoor environments with natural ventilation was found to be close to the average value reported by Persoon and Hornbuckle (2009). Accordingly, the R values we obtained for the indoor environment varied between 2.2 and 3.2 m^3/day.

$$Veff = R \times ACH \times V \tag{3.2}$$

where ACH is the average air exchange rate and V is the volume of the indoor environment.

PCB concentrations were calculated using the following formula (Equation 3.3) with the GAPs model and sampling rate data from the literature.

$$C_{air} = \frac{M_{PCB}}{V_{eff}} \tag{3.3}$$

M_{PUF} is the mass (ng or pg) of the PCB congener on the sample, C_{air} is the PCB concentration in the air (ng or pg/m^3), V_{eff} is the effective sampling volume (m^3).

3.8. Exposure and Health Risk Assessment

Exposure assessment is a process that measures or estimates the magnitude, frequency, and duration values of people's exposure to an agent in the environment (USEPA, 1992). Exposure routes are inhalation, ingestion, and dermal absorption. Scope of this study includes exposure to pollutants only by inhalation. Lifetime average daily dose (LADD) was calculated as the estimate of inhalation exposure in this study (Equation 3.4) recommended by the USEPA (2011). The equation parameters must be expressed in consistent units. In some cases, unit conversion factors may be necessary.

$$LADD_{inh} = \frac{\mathrm{C}\times\mathrm{IR}\times\mathrm{ED}\times\mathrm{EF}}{\mathrm{BW}\times\mathrm{AT}} \quad (3.4)$$

where C is the contaminant concentration (pg/m^3), IR is inhalation rate (m^3/day), ED is exposure duration (yr), EF is exposure frequency (days/yr), BW is body weight (kg), AT is averaging time which was assumed as lifetime, $LADD_{inh}$ is lifetime average daily dose (pg/kg-day) for inhalation exposure. Lifetime exposure was assumed for the assessment (ED = AT = 78.6 years) (TSI, 2019). EF was taken as 350 days/yr. The distribution of BW and IR was taken from Cetin et al. (2018). Parameter values of the assumed probability distributions for variables of exposure and risk are given Table 3.6.

The equation (3.5) was used to estimate the cancer risk associated with inhalation exposure to contaminants in the air (USEPA 1996, 2005).

$$R = LADD \times SF \quad (3.5)$$

Where R is the cancer risk and SF (1/mg kg^{-1} day^{-1}) is the slope factor of the contaminant, and LADD in the units of mg/kg-day (with 10^{-9} conversion factor to pg/m^3 to mg/m^3). Since two different SF values are published for PCBs by the USEPA in the IRIS database, risks for dioxin-like and non-dioxin-like congeners were calculated separately, which then were summed to obtain a cumulative carcinogenic risk. The SF values employed in this study are given in the Table 3.6. The acceptable carcinogenic risk level is 10^{-6} (EPA, 2003). The exposure is therefore considered considerable if the risk factor reaches 10^{-6}.

Table 3.6. Probability distributions assumed for the variables

Input Variables	**Fitted Distribution**	**Distribution Parameters**	**References**
Inhalation Rate (m^3/day)	Extreme Value	Most Likely: 17.68 Scale: 4.71	(Cetin et al. 2018)
BW (kg)	Beta Distribution	Alpha: 12.76 Beta: 8.15	(Cetin et al. 2018)

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Table 3.6. (cont.)

Input Variables	Fitted Distribution	Distribution Parameters	References
SfDL (1/mg kg^{-1} day^{-1})	Uniform Distribution	Minimum: 1 Maximum: 2	This study*
Sf-non-DL (1/mg kg^{-1} day^{-1})	Uniform Distribution	Minimum: 0.3 Maximum: 0.4	This study*

*: values (PCBs; CASRN 1336-36-3" 1989) were used to form and assume uniform distribution

3.9. Statistical Analysis

Statistical analysis was performed using Origin Pro 2021b and Microsoft Excel (Microsoft Office, 2010). Each homologue group is summarized using a box plot with comparisons at the level of urbanization and homologue groups. Percent contribution of data was also represented. The goodness-of-fit of the variables to normality was tested with the Shapiro-Wilk at the significance level of 0.05. Urban, semi-urban, and rural area comparisons were performed with the Kruskal-Wallis and One-Way ANOVA, depending on the normality assumption. When the concentrations were analyzed statistically, groups with less than 50% of the data were determined as below the detection limit and were not included in the calculation between homologue groups. The level of significance was set at $p < 0.05$. Principal component analysis (PCA) was performed to investigate the possible sources of PCBs in air. Before performing PCA all data were normalized. Components with Eigenvalues of >1 were considered significant.

CHAPTER 4

RESULTS AND DISCUSSION

4.1. Air Concentrations of PCBs

This section examines PCB congeners of air concentrations based on homologue groups. Box plots are given on a logarithmic scale since the ranges are rather large. Mono-CBs are not shown because they were not determined in majority of the samples.

The air concentrations taken from the homes were examined based on homologue groups at all sampling points. 91 PCB congeners were quantified after blank correction indoors at home. Figure 4.1. shows the concentration box-plots for homologue groups. Tri-CBs is the highest concentration group with a median value of 261 pg/m^3, followed by Hexa-CBs with a median value of 116 pg/m^3. Octa, Nona and Deca-CBs could not be shown in the graph and included in the comparison because they are not detected frequently. The groups found in the data set above 50% are shown in the graph.

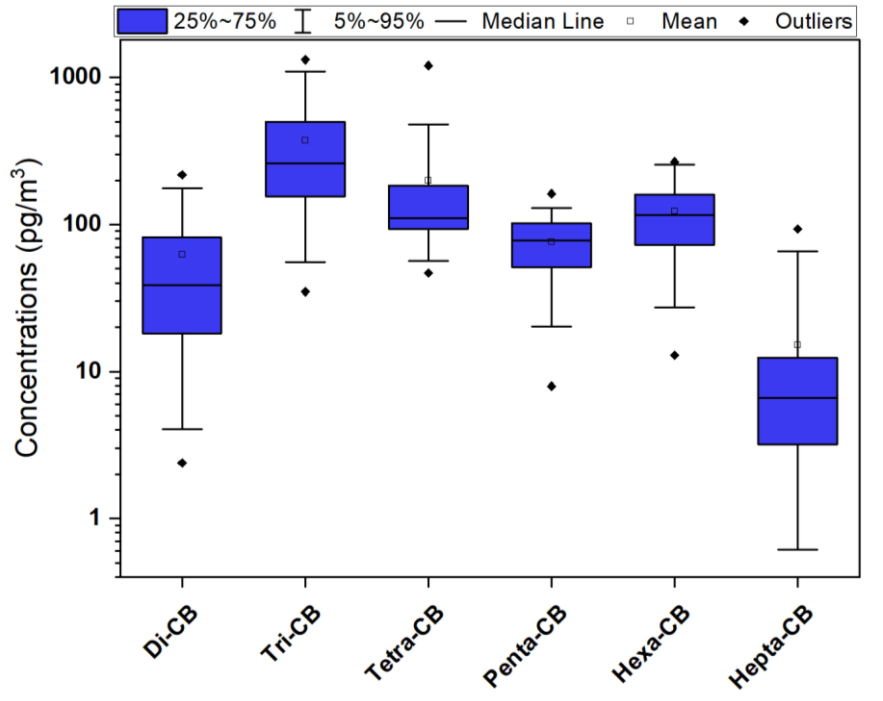


Figure 4.1. Homologue group indoor air concentrations in selected homes

Penta and Hexa-CBs were the dominant homologue group that contributed to Σ_{102}PCB total concentration outdoors (Figure 4.2.). The median values for both of the two groups were approximately 80 pg/m^3. The group with the lowest median value (12.4 pg/m^3) was Di-CBs while Octa, Nona, Deca-CB could not be presented because ratio of BDL values were <50%.

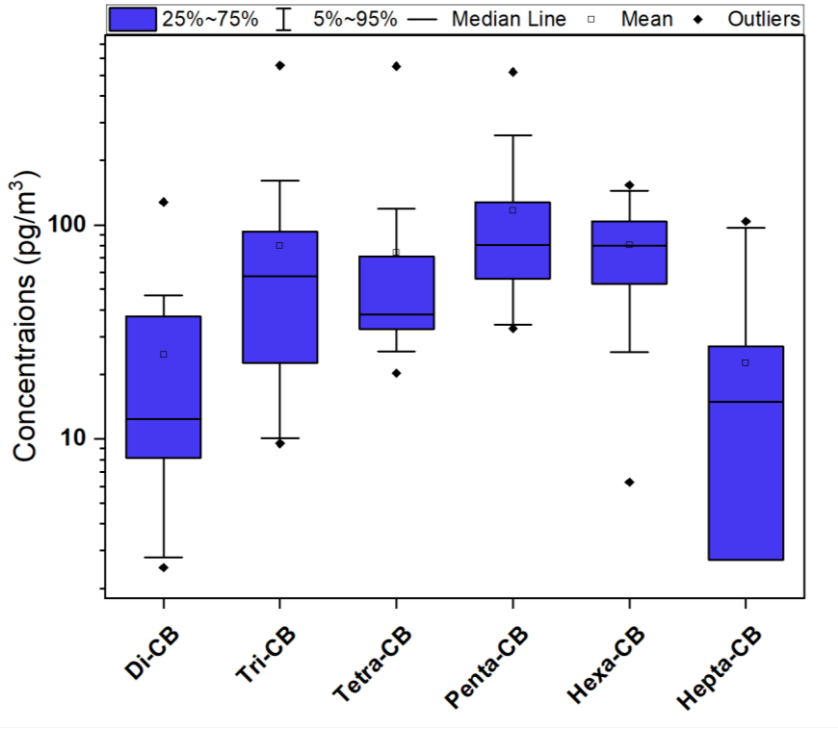


Figure 4.2. Homologue group outdoor air concentrations around selected homes

Similarly, indoor air concentrations in the selected schools are shown in Figure 4.3. There were 97 PCB congeners quantified after blank correction indoors at schools. The highest concentration groups were Tri-CBs and Tetra-CBs with median values of 243 pg/m^3 and 130 pg/m^3, respectively, while the lowest (excluding Octa, Nona, and Deca-CB) was Hepta-CBs with a median value of 5.2 pg/m^3.

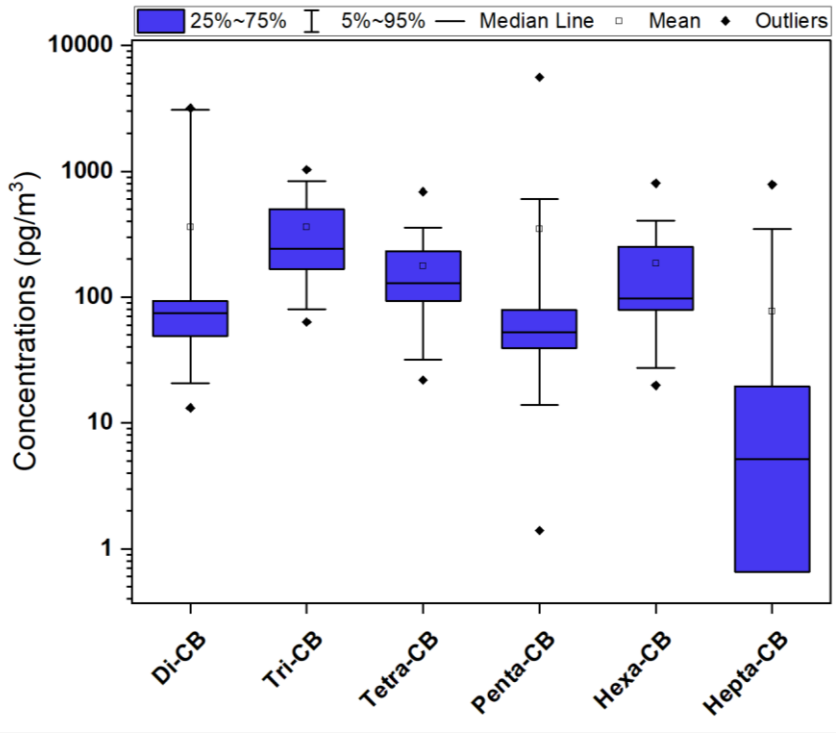


Figure 4.3. Homologue group indoor air concentrations in selected schools

Outdoors around school, Penta-CBs were dominant homologue group with a median value of 156 pg/m^3, while Hexa-CBs followed with 90 pg/m^3 (Figure 4.4.). Differently from the previous, Octa-CBs were detectable with the lowest median value of 3 pg/m^3 while Deca-CBs were still BDL in more than 50% of the samples. There were 94 PCB congeners quantified after blank correction outdoors at schools.

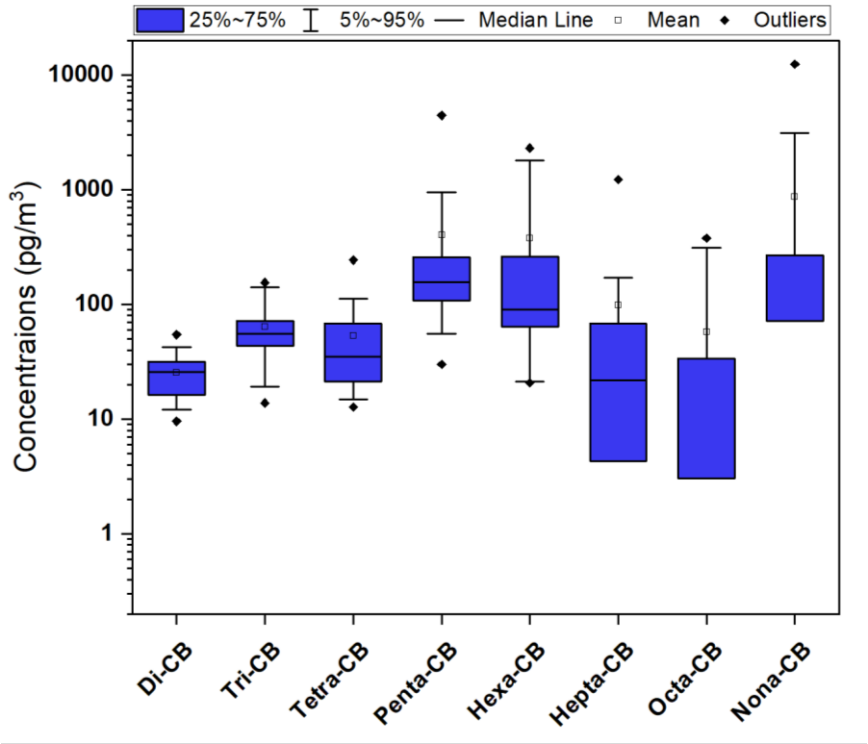


Figure 4.4. Homologue group outdoor air concentrations around selected schools

Low molecular weight groups dominated the indoor air concentrations (Tri, Tetra-CBs). It has been suggested that Di-, Tri-, and Tetra-CBs are related to unintentionally produced PCBs (Xing et al. 2009). Tetra and Hexa-CBs are present indoors at home at similar levels. Because Hexa-CBs are reported to be intentionally produced PCBs, their presence in homes was linked to older transformers and electrical equipment (Cui et al. 2017). Also, potentially polluted soil in the proximity of sampling locations where PCB usage is widely reported could be another source of Hexa-CBs (Salihoglu et al. 2011).

Tri-CBs and Tetra-CBs are the most prevalent homologue groups in the atmosphere in urban, semi-urban, and rural areas in this study. Tri and Tetra-CBs are the dominant groups compared to the other homologue groups in the atmospheric environment; they have low molecular weights and constituted 79 % of Aroclor 1016 and between 41 and 58 % of 1232, 1242, and 1248. The predominance of PCB-28 and PCB-52, two indicator congeners, in these groups is assessed to be evidence of the Aroclor presence. The number of reported congeners in investigations can range from a few indicator (i) to all indicator (i7) PCBs (28, 52, 101, 118, 138, 153, and 180). Indicators were chosen because they are among the most frequently detected congeners at relatively higher concentrations throughout the compositional range of the most common technical

combinations and in the environment. Because of this, they are often used as markers to investigate PCB contamination.

The high concentrations of Penta-CBs found in our study of the outdoor environment can be attributed to factors such as the effect of indoor pollutants on the outdoor concentration and the use of insulation materials on the outdoor buildings containing Aroclor 1260, 1254, and 1248; and Kanechlor 600 (Audy et al. 2018; Zennegg et al. 2005a; Robson et al. 2010; Dumanoglu et al. 2017; Takasuga et al. 2006).

PCBs, mostly known to be of indoor origin, can also be found at high concentrations in industrial activities and outdoor concentrations. Since the city of Izmir, where we sampled, has AIA, it can be seen as the primary source of outdoor PCB concentrations. Penta-CBs iron-steel and shipbreaking plants were cited as PCB emitter sources (Kaya et al. 2012). The high presence of homologue groups in the outdoor environment may be due to the AIA effect.

Also to explain this phenomenon, in addition to the indoor sources, natural events that affect PCB levels outdoors may be considered. Precipitation, that is typical for the sampling campaign period of this study, is one of them. Since low molecular weight components (tri-, tetra-CBs) have approximately 5 to 777 times higher solubility than the high molecular PCBs (>penta-CBs), they are more easily scavenged (removed from the atmosphere) with rain. It was reported that the seasonal contribution of Penta-CBs and Hexa-CBs was at its highest point in the fall (Yeo et al. 2003). Considering the sampling periods (September-January), the fact that the presence of low-molecular pollutants in the outdoor environment is less than as opposed to indoors may be reasonably explained with this argument.

The income levels in the various countries were considered while analyzing the indoor PCB concentrations. The World Bank (2021) was the source of information used to gather the levels of country income. First, research was conducted to determine PCBs found within houses in low-income countries. On the other hand, no studies describe the concentrations of PCBs in the indoor air, particularly in Africa, which is home to many countries with low-income economies (Emmanuel et al. 2020). It is clear that Mexico and Türkiye, both of which were classified as upper-middle-income countries, have concentration ranges that were similar. On the other hand, 111 pg/m^3 was found to be the average concentration of the indicator congeners (CB-28, -52, -101, -118, -138, -153, -180) in Türkiye, which is a lower concentration than most high-income countries. Table

4.1 shows the comparison between countries. Generally, countries with a moderate income tend to have lower concentrations of PCBs than those with a high income.

Table 4.1. Comparison of PCBs with Income Levels

Country	**Income Levels of Country Economies**	**PCB Concentrations (pg/m^3)**	**References**
Türkiye	Upper-Middle Income	Σ_7PCB= 111	This Study
USA, Iowa	High-Income	Σ_{209}PCB= 2830	(Herkert, Jahnke, and Hornbuckle 2018)
Mexico	Upper-Middle Income	Σ_7PCB= 130	(Bohlin et al. 2008)
Slovak Republic	High-Income	Σ_9PCB= 1533	(Demirtepe et al. 2019)
UK	High-Income	Σ_7PCB= 330	(Bohlin et al. 2008)
Sweeden	High-Income	Σ_7PCB= 260	(Bohlin et al. 2008)
Canada	High-Income	Σ_8PCB= 734	(Audy et al. 2018)
Czech Republic	High-Income	Σ_7PCB= 661	(Audy et al. 2018)
Australia	High-Income	Σ_7PCB= 270	(X. Wang et al. 2019)

4.2. Variation due to Location

The level of urbanization was used to classify the contribution of each congener percentage.

Firstly, from high to low MW PCBs-209, -206, -205, -193, -190, -189, -180, -177, -174, -173, -105, -85, -49, -25, -22, -12, -8, and -4/10 were the congeners that made up the profile in rural area homes. Tri-CBs, the group that makes the biggest contribution to

the indoor home concentrations, was seen as the most dominant group in the rural area, followed by sub-urban and urban areas. The congener group of Hexa-CBs is predominantly observed in the sub-urban area. The most dominant group in the urban area was Penta-CBs.

In the PCB samples taken from outdoor the homes, showed almost a similar trend with the indoor air profile. In general, samples taken from the rural area have higher concentrations than sub-urban and urban. Tri, Tetra, Hepta, Octa, Nona, and Deca-CB concentrations were higher in the samples taken from the rural area compared to the other two groups. For urban, Hexa-CBs were dominant compared to rural and sub-urban, while Penta-CBs had a similar value with rural area. Figure 4.5. represents the congener profile for indoor and outdoor homes concentrations.

Contributions of each congener to the total school indoor-outdoor air PCB concentrations are shown in Figure 4.6. The urban and sub-urban areas have higher PCB concentrations in the schools, unlike those in the homes. Tri, Tetra, and Hexa-CBs there are the dominant groups in urban areas. The second highest area was sub-urban with dominance of Penta, Hepta, and Nona-CBs. Di-CBs were at higher concentrations in rural area.

Penta, Hexa, and Hepta-CBs were at higher levels outdoors at schools in urban area. Rural Tri, Tetra, Nona, and Deca-CB concentrations were higher than those in the other areas. The contribution of Di, Tetra, and Octa-CBs in urban and rural regions were similar.

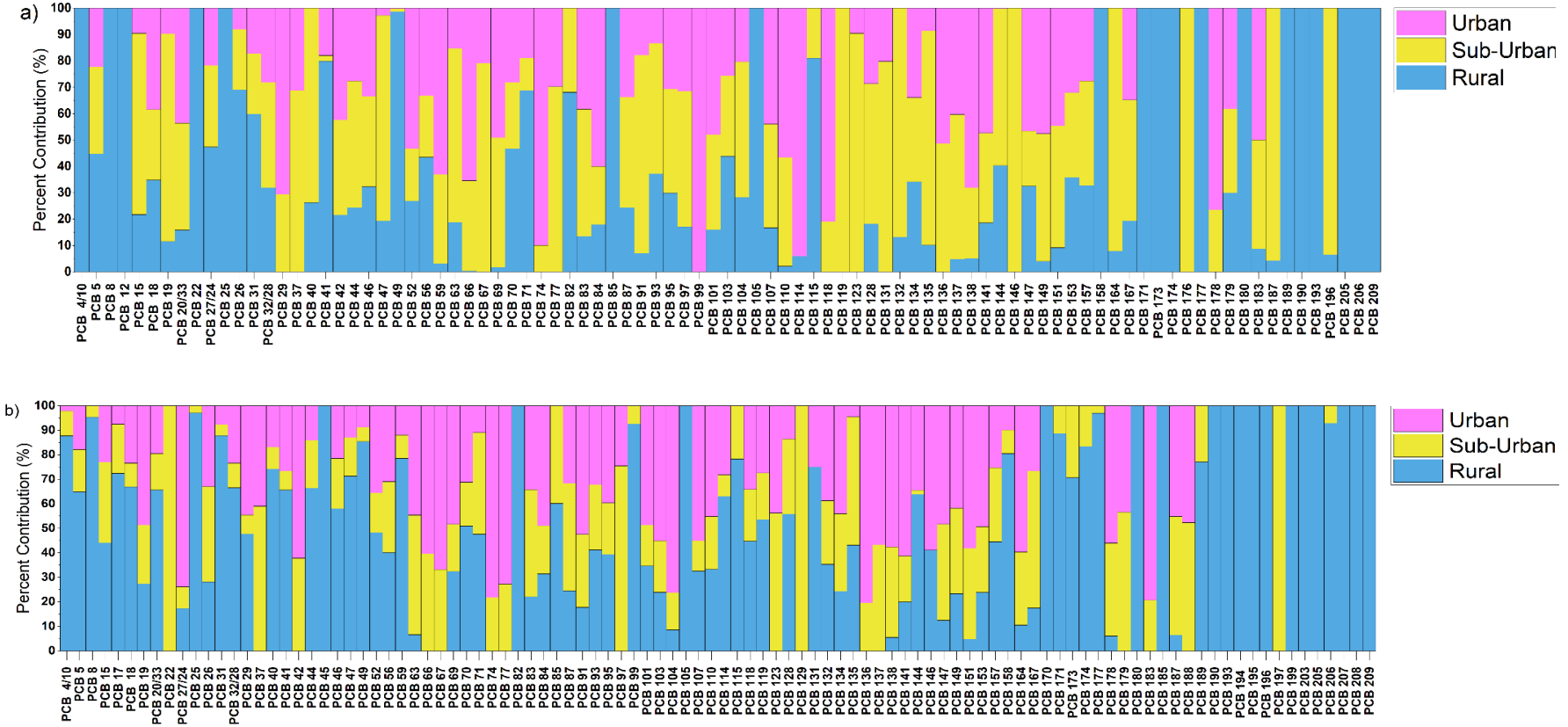


Figure 4.5. Percent Contribution of PCB congeners a) Houses indoor air b) Houses outdoor air

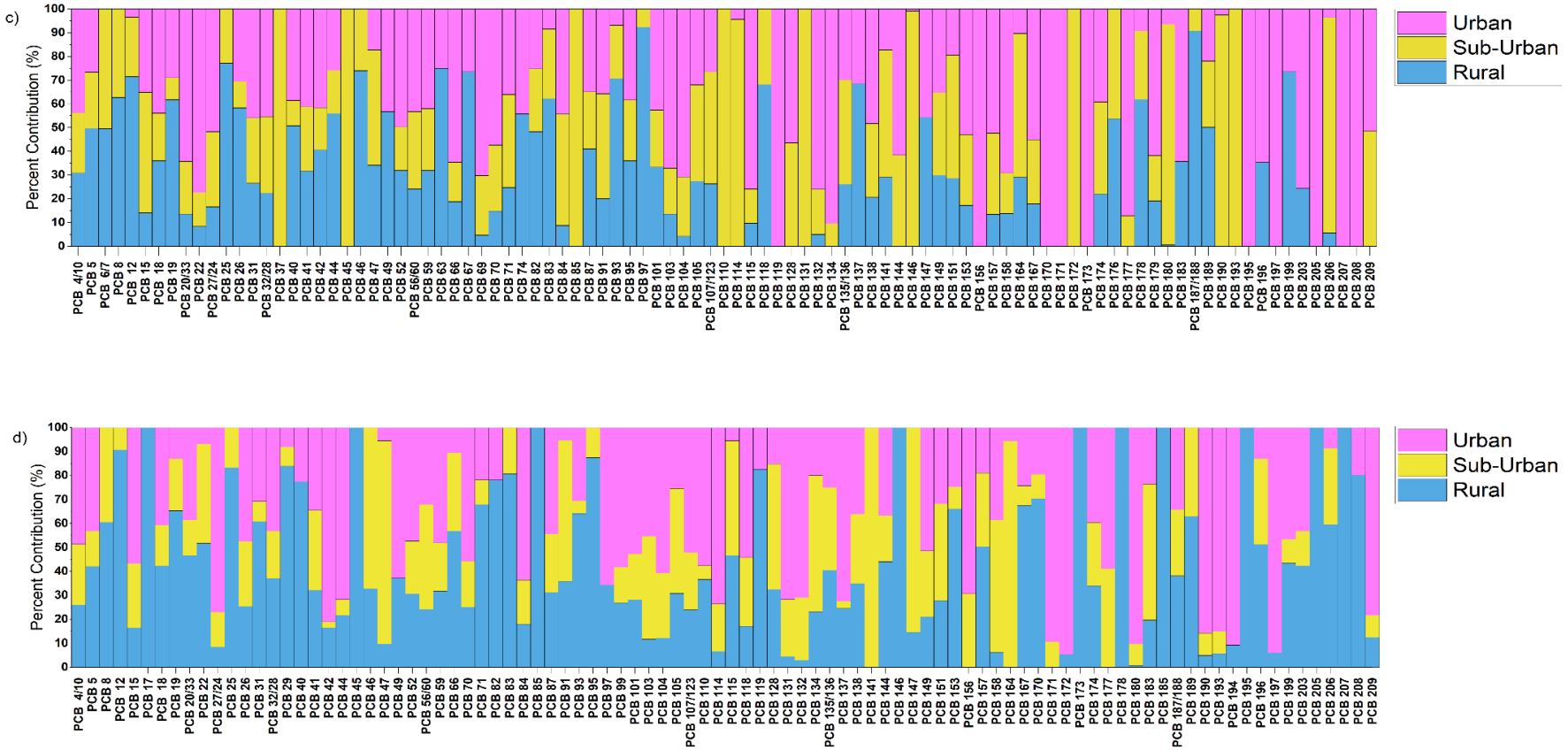


Figure 4.6. Percent Contribution of PCB congeners c) Schools indoor air d) Schools outdoor air

4.3. PCB-AIA Distance Relationship

Study design included sampling points in rural, semi-urban, and urban areas. While urban areas have the highest population density, rural areas are the regions with the lowest population. However, it should be underlined that there is also an industrial area (AIA) to the north of study area. Considering proximity of our sampling sites to AIA, some of rural and suburban sites to the north of İzmir metropolitan area and to the south of AIA are located at closer distances compared to those suburban and rural sites to the south of the metropolitan area. It is known that rural areas generally have lower concentrations compared to urban locations if not locate in close proximity to an industrial area (Harner et al. 2004; Odabasi et al. 2016). Melymuk et al. (2012) reported that PCB concentrations were 39 times higher in urban areas than those in rural areas. However, they argued that this ratio was insufficient, and an found an exponential decrease in ΣPCB concentrations with distance from the urban business center (Melymuk et al. 2012). Odabasi et al. (2017) observed higher PCB concentrations in rural area close to AIA. Our study found high PCB concentrations at points in close proximity to the AIA (ru1, ru2, ru3, su1, su3, and su4, in which *ru* and *su* stand for rural and suburban, respectively). Subsequently, proximity relationship was investigated using simple linear regression analysis (SLR). Figure 4.7. shows the change in total outdoor concentrations of six homologue groups at both homes and schools with distance to AIA. Sampling points were grouped as close, middle, and far. The mean concentrations (with error bars showing one standard deviation) were plotted against distance to AIA. SLR showed that there is a decreasing trend in the concentrations with distance from the AIA for Di, Tri, and Tetra-CBs (slope, ANOVA F-test p-value, and R^2 values are shown in Figure 4.7). The slope flattens for Penta and Hexa-CBs, and reverses for Hepta-CBs, none of which was statistically significant. In consequence, overall total PCB concentrations also showed a decrease with distance. Therefore, it may be argued that the effect of AIA is apparent on the study area exceeding the effect of emissions in the urban area (Izmir metropole). Odabası et al. (2009) and Kuzu et al. (2013) studies have demonstrated the presence of PCB emission in AIA. The AIA has been named the 'hot spot'. In addition, in study Odabası et al. (2009) was determined that the PCB concentration profile from the AIA is generally by low molecular weight PCBs. That may explain why the lower molecular

weight groups are high in the close region and have a decreasing profile with distance from the AIA (Odabasi et al. 2009a; Levent Kuzu et al. 2013).

In addition, we scaled the pollution level by normalizing the data (Table 4.2). For this reason, the sample in the 'Kozakyaylası' region was used as the background concentration while examining the concentrations. The concentration of each sampled location was divided by the background concentration. A value greater than 1 indicates highly polluted, a value less than 1 indicates that the degree of pollution is gradually decreasing. It can be observed that the pollution level decreases as one moves away from the AIA, as in the SLR.

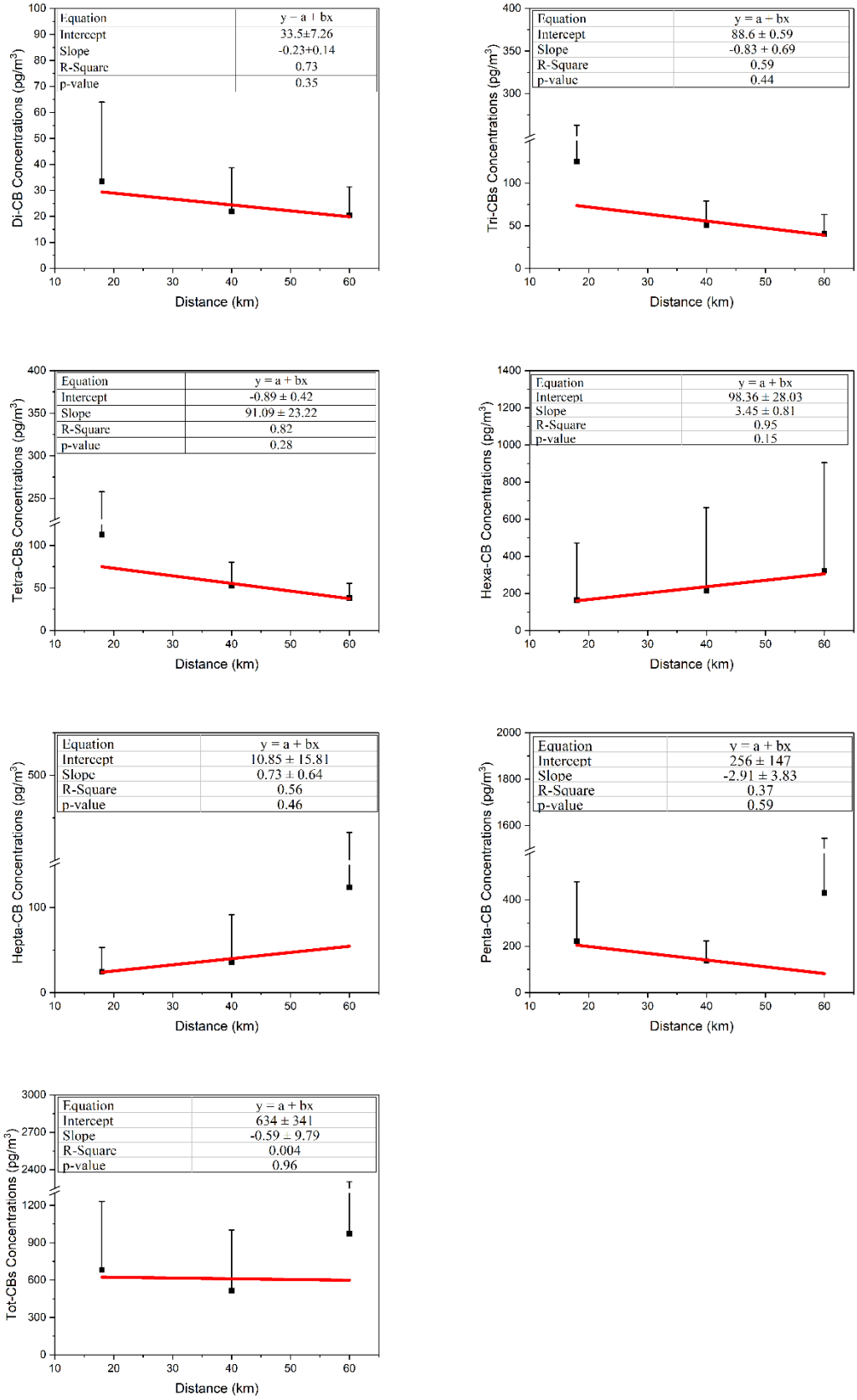


Figure 4.7.The comparison between concentrations and distance from the linear regression parameters

Table 4.2. Pollution levels of PCB concentrations groups of close, middle and far from the AIA

Distance	Tot-CBs	Di-CBs	Tri-CBs	Tetra-CBs	Penta-CBs	Hexa-CBs	Hepta-CBs
Close	1.11	1.54	4.35	1.72	0.59	0.64	0.89
Middle	1.01	0.98	2.68	1.11	0.56	0.73	0.63
Far	0.97	1.35	2.01	0.78	0.67	0.77	1.30

A correlation matrix was constructed to infer on the relationship between concentrations measured indoors and outdoors, along with correlation of homologue groups among themselves (Figure 4.8). Only Di-CBs-Outdoor concentrations were correlated with Di-CBs-Indoor ($p < 0.05$). Apart from this, it was determined that indoor and outdoor PCB concentrations did not correlate even at a significance level of 0.10.

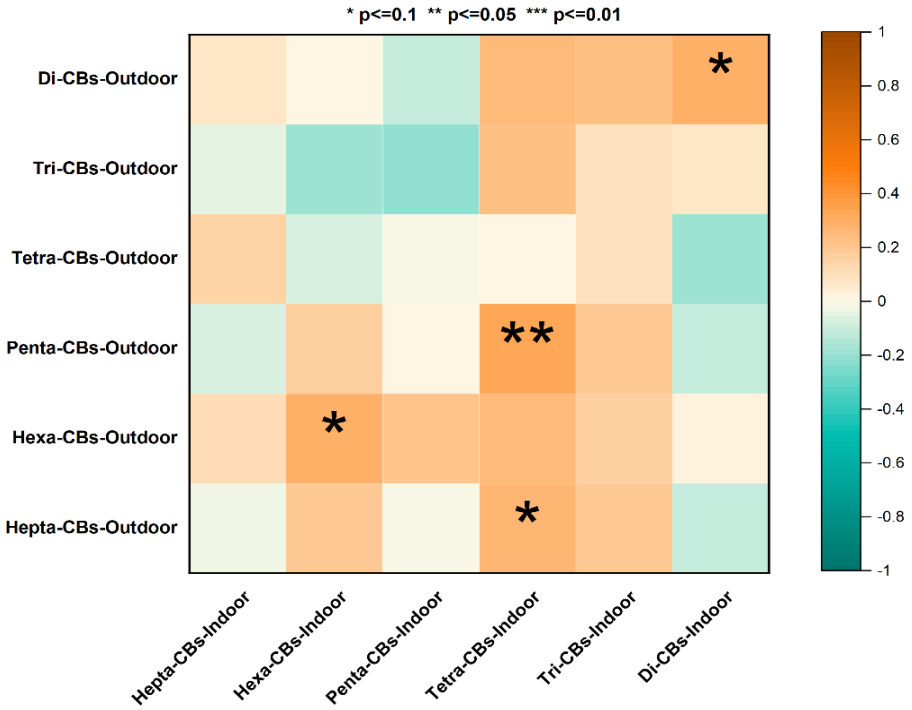


Figure 4.8. Indoor versus outdoor concentrations correlation data

The above analysis was based on the lumped datasets of schools and homes. Although the locations of homes and schools were close at each sampling site (average 2.5 km), different indoor air congener profiles were observed between them, indicating variability difference in PCB sources indoors. However, a difference in outdoor congener profiles was not apparent. Therefore, another correlation matrix was constructed to infer on the relationship between concentrations measured outside the homes and schools (Figure 4.9). Correlations were not significant ($p<0.1$) except for Tri and Tetra-CBs. PCBs has many sources for ambient air including such urban sources as particle emissions from vehicles, residential heating with petroleum or coal (Cetin et al. 2018a). Di-CB and Penta-CBs were not shown a significant correlation. It has been thought that Di-CBs may show different concentrations due to their low molecular weight, being deposited in soil or dust, and re-evaporation due to changing temperature differences. It is thought that these differences will be understood more clearly when the dust profile in the regions where homes or schools were located. For example, homes may be located in an area with more Di-CB deposits. Penta-CBs has no significant correlation between other homologue group. It has been stated in the literature that Penta-CBs was mainly used as a paint additive. (China SEPA, 2003). Penta-CBs, especially its indicator congener, PCB-118, were the most abundant congeners in vehicles, heating, waste incineration, and fly ash emissions (Biterna and Voutsa 2005). For these reasons, Penta-CBs have been evaluated and observed in different profiles due to the effects that may arise from the proximity to above mentioned sources including to traffic or vehicle parking areas. Also, the effects may arise from the differences in the sources used for heating. On the contrary, some groups were correlated. Tri-CBs and Tetra-CBs showed a significant correlation in homes and schools at a significance level of 0.01, which may be attributed to proximity relationship of the locations to AIA (Figure 4.7).

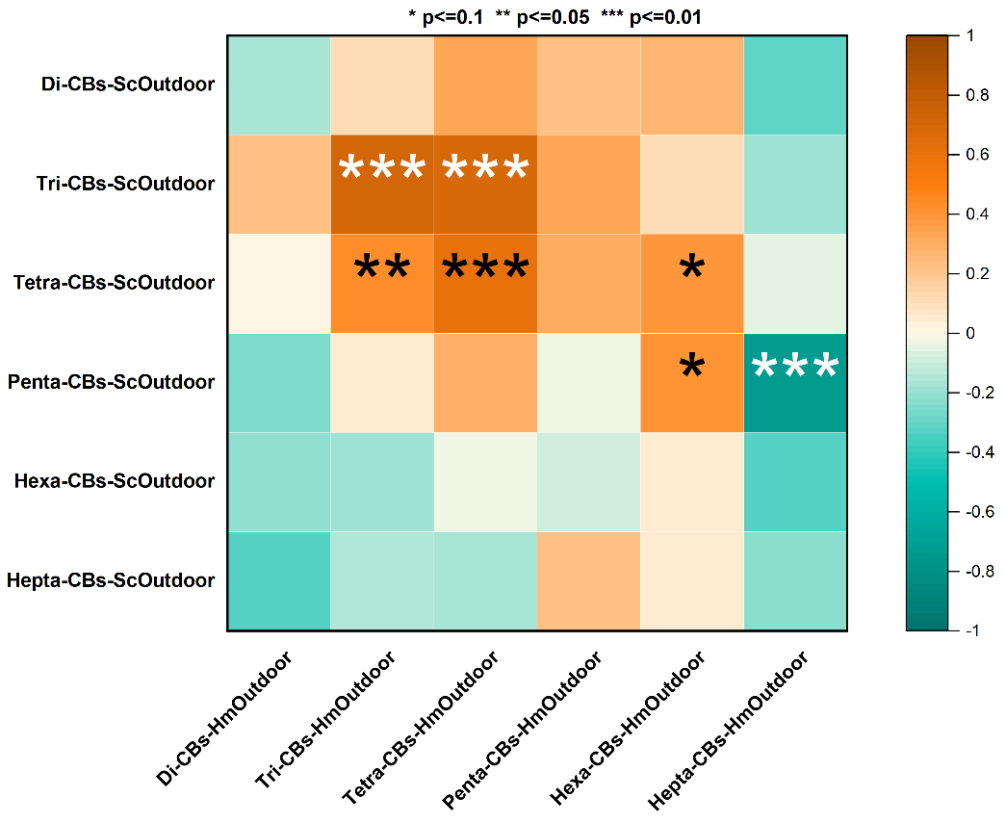


Figure 4.9. Home versus school's outdoor concentrations correlation data

Evidence based on proximity analysis indicated the effect of AIA on the study area. However, the number of data points (n=3) used for SLR and the variation in concentrations in each group were considered as limitations.

Legacy usage of PCBs in industrial and consumer items, negligent manufacture and releases from industries, and unintentionally created no legacy emissions contribute to global PCB pollution. Long-range atmospheric transport (LRAT) affects the PCB pollutants globally. Various countries' laws support the use of PCB pollutants, which the Stockholm Convention prohibits, to eliminate its use. However, Melymuk et al. 2022 investigated that the global management of PCBs claimed the precautions of eliminating it was insufficient. That has been revealed that over the years, the actions enforced by the countries were poor. For instance, the USA is not a party to the Stockholm Convention and, despite being the largest producer, user, and possibly the largest PCB stockholder, lacks effective federal policies for the disposal and safe disposal of PCBs. The Stockholm convention had set a deadline to phase out PCBs after about 50 years. According to the result of the study, after 50 years, the resources (financial and technical) and political will required to solve the problem seem unattainable (Melymuk et al. 2022).

4.4. Indoor/Outdoor Ratios

The contribution of indoor and outdoor sources has been analyzed in various ways in the literature. The most common is the indoor-to-outdoor concentration (I/O) ratio. An I/O value >1 indicates higher contribution from the sources indoors, while values of the ratio <1 indicate the dominance of outdoor sources (Bohlin et al. 2008). However, there is not an agreement in the literature on how large or small a value should be to indicate a dominance (Sofuoglu et al. 2011; Sahin. et al. 2022).

I/O ratios have been interpreted according to the results we obtained from the samples collected from indoors and outdoors at homes and schools based on homologue groups. The calculated I/O values are in Table 4.3 The average values are also illustrated as bar graphs for ease of interpretation (Figure 4.10 for homes and Figure 4.11 for schools). As a result of the comparison made based on homologue groups, we determined that indoor air generally had higher concentrations than the outdoor air in samples taken at homes. In samples taken from homes, groups with values > 1 were Di, Tri, Tetra, and Hexa-CBs, while those with values less than 1 were Hepta-CBs. Penta-CBs were found to be quite close in indoor and outdoor air. Among the homologue groups, Di, Tri, Tetra, and Hexa-CBs were high in the indoor environments of the schools. At the same time, Penta, and Hepta-CBs were the dominant homologue group in the outdoor environment. So we observed a shifted profile at schools compared to homes indicating that there is difference in indoor sources between the types of buildings. Studies in the literature support our results. Studies measuring the indoor PCB concentrations and comparing them with the outdoor environment (Rudel and Perovich 2009; Melymuk et al. 2016; Sari et al. 2020; Bohlin et al. 2008) have also proven higher indoor PCB concentration values. PCBs have been identified in indoor air at amounts one order of magnitude greater than those found in outdoor air. This situation may be linked to the presence of PCB-containing equipment and construction materials in the indoor environment. It has been hypothesized that some electrical appliances and gadgets, such as fluorescent lighting ballasts having PCB-containing components, may release PCBs into the indoor air (Rudel, Seryak, and Brody 2008a). PCBs have also been used in many equipment. The plasticizers in joint sealants may contain up to 30% PCBs which may be abundantly found indoors (Zennegg et al. 2005b). It was discovered that indoor PCB concentrations are approximately 2–50 times greater than outdoor levels (Menichini, Iacovella, and Monfredini 2007). It

demonstrates that the PCB concentration in indoor air is significantly higher, therefore may impact human health, especially of children. Studies conducted in schools showed the presence of PCBs indoors at higher levels than outdoors (Marek et al. 2017; Thomas et al. 2012). The comparison between schools and homes is considered because they may have different indoor sources, e.g. devices, equipment, materials. The difference in sources separating homes and schools may be caused by electrical equipment. Concentrations can be high in homes and schools with the presence of old sources such as material and equipment that were produced/manufactured before the ban of PCBs. In this case, the factor that makes the difference would be the presence of PCB-containing electrical/electronic devices, wires and cables, window frames, wallpapers, paints, adhesives may be sources of PCBs indoors. In addition, the most significant difference in PCB concentrations obtained from schools and homes is stated to be the different floor materials (Rudel, Seryak, and Brody 2008b). Also, some studies identified sealants and caulks as the common primary sources both in homes and schools. In such cases, similar (close) I/O ratios may be observed in home-school environments due to their common sources such sealants and caulks (Gabrio et al. 2000; Herrick et al. 2004; Lund et al. 2016).

Table 4.3. Values of Indoor/Outdoor Ratio for each sampling location

Places	Code	Di-CB	Tri-CB	Tetra-CB	Penta-CB	Hexa-CB	Hepta-CB
Homes	RU1	0.5	0.3	2.2	0.02	0.1	1.8
	RU2	3.7	1.8	4.2	0.6	1.1	
	RU3	2.9	8.2	2.9	0.7	2.7	0.5
	RU4	3.8	9.9	4.9	1.4	0.6	322.6
	RU5	3.5	24.6	3.8	0.2	6.2	0.8
	RU6	1.6	2.2	3.0	0.2	0.9	0.0
	RU7	0.1	0.4	1.5	0.2	1.5	0.9
	SU1	3.9	7.4	1.9	0.9	1.1	0.4
	SU2	12.4	19.5	1.8	1.0	0.4	0.1
	SU3	17.8	52.7	3.9	1.1	1.5	
	SU4	45.4	10.5	4.3	2.2	2.4	0.7
	SU5	1.8	15.4	2.6	3.1	2.6	0.1
	SU6	0.8	11.8	10.7	0.8	40.7	1.7
	SU7	1.0	9.0	3.0	2.1	2.6	0.2

(cont. on next page)

Table 4.3 (cont.)

Places	Code	Di-CB	Tri-CB	Tetra-CB	Penta-CB	Hexa-CB	Hepta-CB
Homes	UR1	0.9	10.9	1.7	2.3	1.0	0.7
	UR2	3.9	1.8	1.2	1.3	1.4	
	UR3	3.1	1.5	0.9	1.1	1.1	4.7
	UR4	1.5	11.7	7.4	1.4	1.0	0.4
	UR5	0.2	0.9	0.6	0.1	0.6	2.7
	UR6	7.7	10.0	7.5	1.1	2.3	0.9
	UR7	3.2	11.1	1.9	1.1	2.2	0.3
Schools	RU1	72.1	0.6	0.3	0.2	0.9	26
	RU2	5.7	3.7	10.5	0.5	1.0	
	RU3	2.6	1.5	1.6	0.2	1.7	0.01
	RU4	2.5	1.4	4.2	0.1	0.2	0.02
	RU5	8.2	12.2	17.6	1.8	2.0	0.1
	RU6	1.8	3.2	3.2	0.8	4.5	58
	RU7	5.2	5.3	7.1	0.9	0.1	0.3
	SU1	2.1	3.2	0.3	0.5	0.3	0.3
	SU2	5.1	2.0	3.2	0.1	4.7	
	SU3	3.4	12.2	9.0	0.6	0.4	
	SU4	2.1	10.5	4.8	0.3	5.1	0.004
	SU5	3.1	21.7	6.6	29.9	0.1	3.1
	SU6	1.2	5.8	7.3	0.4	1.0	62.8
	SU7	235.0	4.4	1.1	0.05	1.0	5.0
	UR1	1.5	18.6	6.3	0.5	0.1	
	UR2	4.9	10.8	25.2	0.2	0.2	
	UR3	2.2	6.8	3.8	0.2	1.5	0.2
	UR4	0.4	3.8	1.2	0.7	3.9	0.4
	UR5	3.0	5.8	2.9	0.01	1.6	
	UR6	1.6	5.4	2.1	0.5	0.7	0.01
	UR7	2.2	6.1	1.9	0.2	3.2	0.3

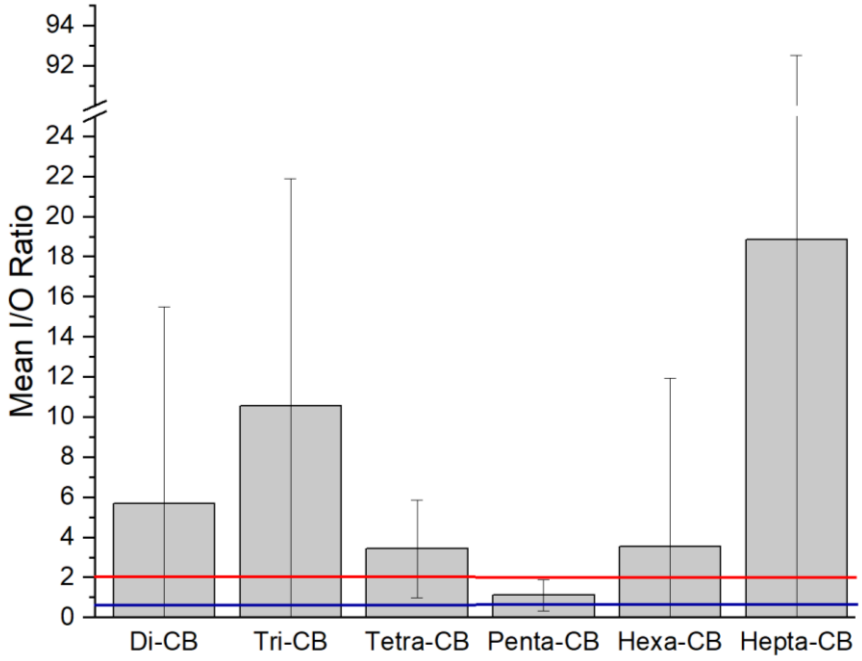


Figure 4.10. Values of the Mean I/O Ratio for homes (Error bars show one standard deviation; horizontal reference lines show demarcation for 0.5<I/O<2.0)

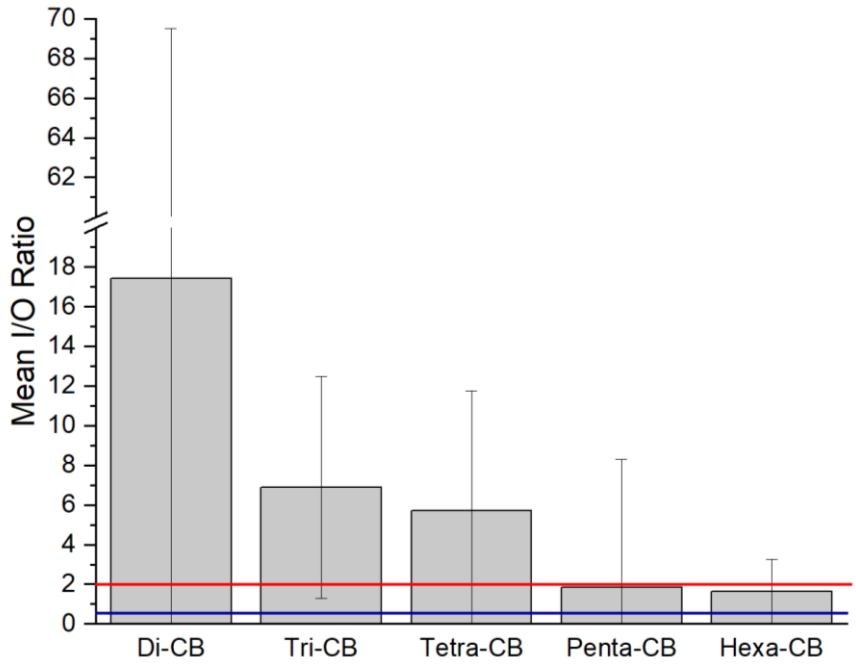


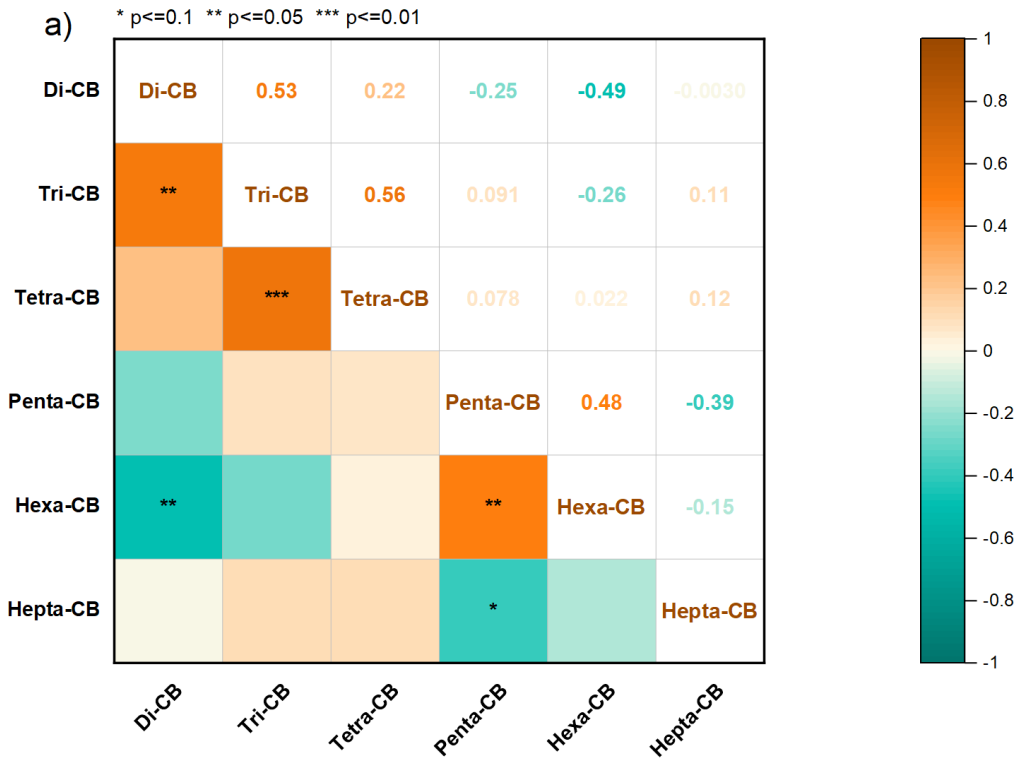
Figure 4.11. Values of the Mean I/O Ratio for schools (Error bars show one standard deviation; horizontal reference lines show demarcation for 0.5<I/O<2.0)

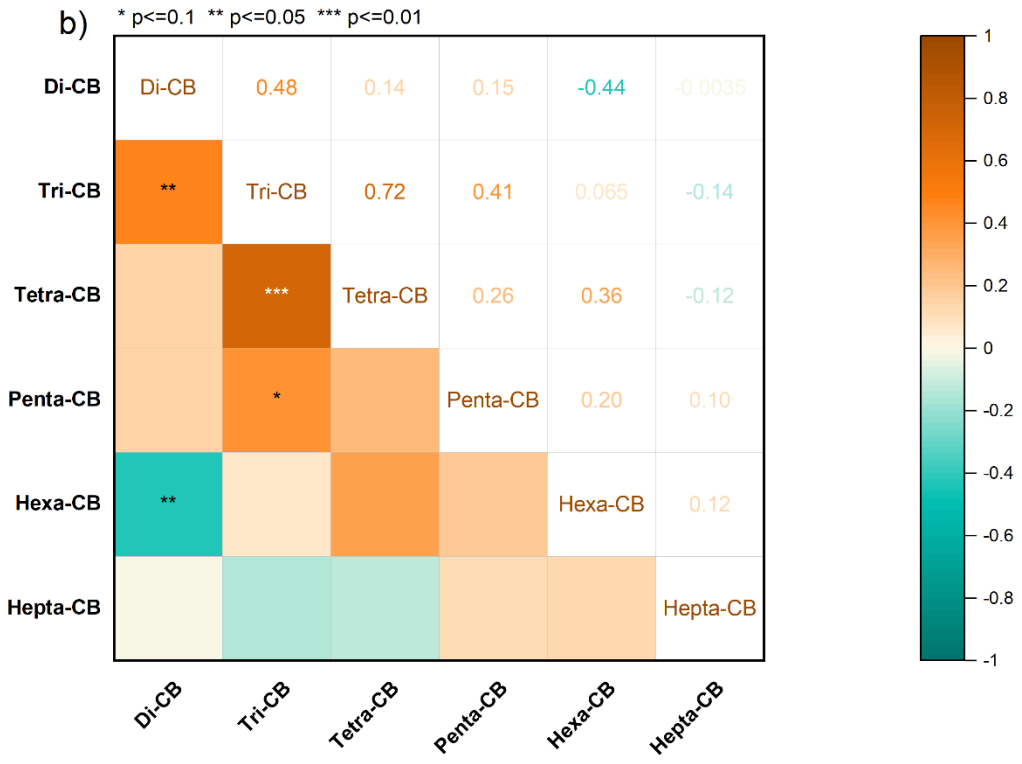
4.5. Correlation for Homes and Schools

In this section, correlation matrices were constructed for homes and schools, indoors and outdoors.

Tetra and Tri-CB homologue groups significantly correlated for all samples. Homes air samples exhibit substantial correlations between Penta and Hexa homologue groups and between Di, Tri, and Tetra groups with lower molecular weights. The heavy molecular weight groups of the outdoor air samples do not show a significant correlation. This condition can be explained by the transport of PCB congeners with low molecular weight released from "hotspots" in the province of Izmir, where Aliağa Industrial activities. It can be noted that the indoor air concentrations of heavy molecular weight congeners are significantly correlated due to the use of commercial mixtures such as Aroclor 1254 in domestic environments (Anh et al. 2020). The lower molecular weight congeners from the AIA may be thought to originate from steel production with electric-arc furnaces from scrap (Odabasi et. al, 2009) and ship-breaking facilities that may be associated with commercial mixtures such as Aroclor 1221, 1242, or Delor (D103).

When the air samples taken from the school are examined, it is seen that there is a significant correlation between the Tri, Tetra, Penta, and Hexa groups. It can be said that commercial mixtures such as Aroclor 1254 and 1242, which support these correlations, are used together in building materials such as insulation materials used in schools (Yagci, 2018). The Di-CB group, which contains congeners with low molecular weight, exhibits the same tendency as the other homologue groups in outdoor air. An effect from AIA is also plausible as discussed for homes. Figure 4.12 shows the all correlation between school and home environment and indoor outdoor concentrations.


a)
* p<=0.1 ** p<=0.05 *** p<=0.01
Di-CB
Tri-CB
Tetra-CB
Penta-CB
Hexa-CB
Hepta-CB
0.53
0.22
-0.25
-0.49
-0.0030
0.56
0.091
-0.26
0.11
0.078
0.022
0.12
0.48
-0.39
-0.15
1
0.8
0.6
0.4
0.2
0
-0.2
-0.4
-0.6
-0.8
-1


b)
* p<=0.1 ** p<=0.05 *** p<=0.01
Di-CB
Tri-CB
Tetra-CB
Penta-CB
Hexa-CB
Hepta-CB
0.48
0.14
0.15
-0.44
-0.0035
0.72
0.41
0.065
-0.14
0.26
0.36
-0.12
0.20
0.10
0.12
1
0.8
0.6
0.4
0.2
0
-0.2
-0.4
-0.6
-0.8
-1

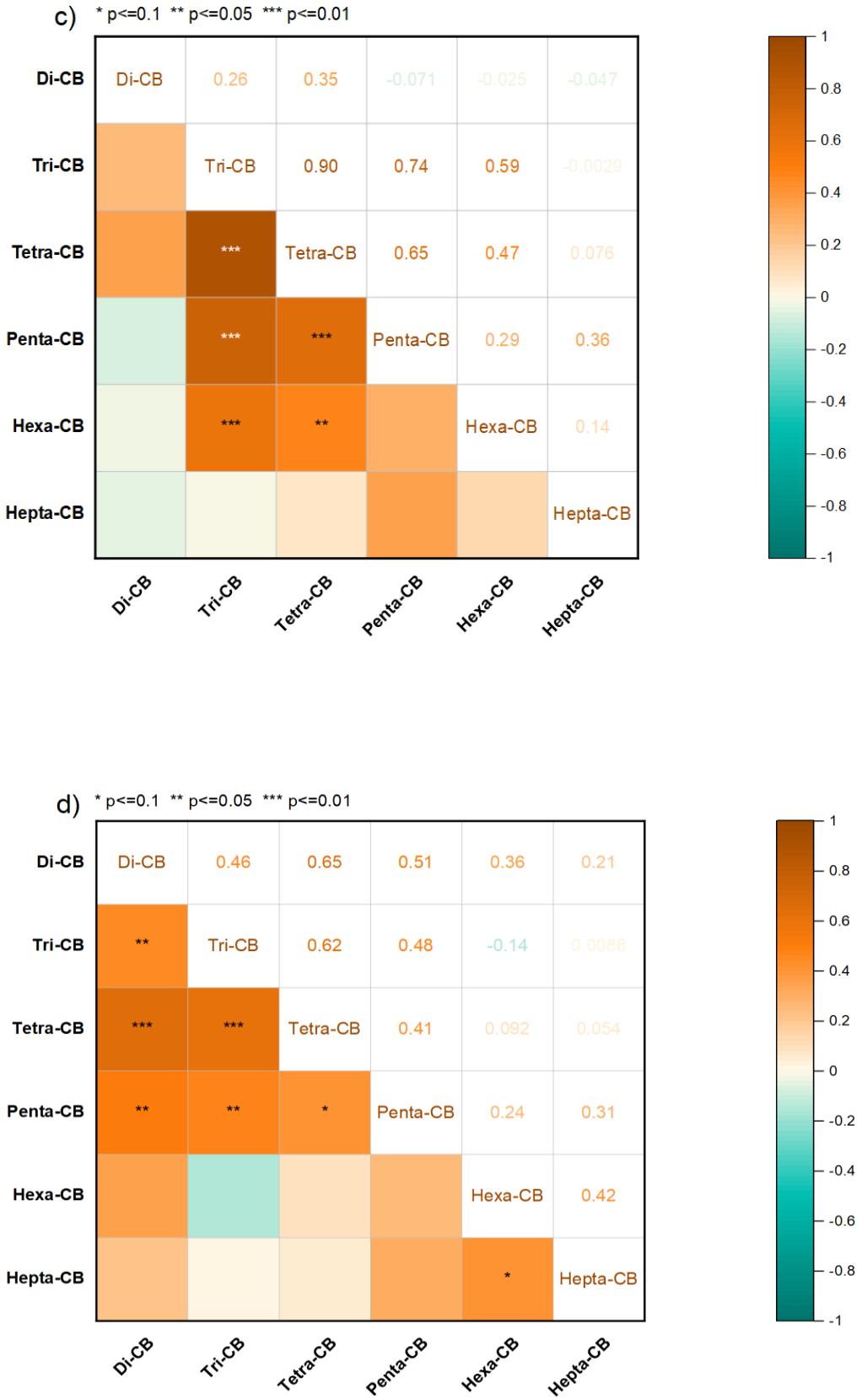


Figure 4.12. Spearman's rho correlation matrix for a) indoor home b) indoor school c) outdoor home d) outdoor school

4.6. Principal Component Analysis

Principal Component Analysis (PCA) was utilized to further assess the variation by homologue groups. PCA was also used in association with diagnostic ratios to apportion PCB sources for indoors and outdoors. If a pollutant was not detected at a rate of >50%, it was eliminated from the analysis. Therefore, PCA excluded Octa, Nona, and Deca-CBs. Before PCA analysis, data were normalized to [0,1] as an attempt to reduce the impact of multicollinearity on regression coefficient estimates and ensure consistency.

Components having an Eigenvalue >1 were considered to be significant. It has been claimed that it is necessary and sufficient for a scale to have positive reliability, which prevents the values with small loads from being perceived as principle components (Braeken and Van Assen 2017). Table 4.4 and Figure 4.13 represent details of the contributions of each component.

For the indoor environment, three components explained 61% of the variation in the concentrations. Contributions to the total were 26% for component 1, 18% for component 2, and 17% for component 3. The dominant homologue groups observed in component 1 were Tri, Tetra, and Penta-CBs. Hexa-CBs are dominant in component 2 while component 3 was loaded by Hepta-CBs. The literature report that possible indoor sources of low molecular weight compounds may be flame retardant coatings of floor and ceiling tiles while medium-molecular-weight congeners may originate from permanent elastic sealants (Heinzow et al. 2007). Therefore, PC 1 may be considered to represent the primary indoor sources. Imamoğlu et al. (2010) reported that Aroclor 1242 was probably used in Türkiye before it was banned. Aroclor 1242 was thought to be another resource enhancer due to its high percentage of low molecular weight congeners. As a result, we can name PC1 as indoor insulation materials. On the other hand, the congeners loaded PC1, especially those lower MW ones, are also found in ambient air emitted from various sources, which include industrial sources in AIA, and transported downwind. Therefore, PC1 may be representing the outdoor air. Further analysis is required to make the elucidation. Hexa and Hepta-CBs loaded PC2 and 3, respectively. It has been reported that high molecular weight PCB congeners were used in dye applications and contribute to indoor PCB concentrations (Rudel and Perovich 2009). As a result, PC2 and 3 might be indicating indoor sources as such.

Table 4.4. Indoor air PCA analysis

	Eigenvalue	Percentage of Variance	Cumulative
1	**1.54353**	**25.73%**	**25.73%**
2	**1.06828**	**17.80%**	**43.53%**
3	**1.04134**	**17.36%**	**60.89%**
4	0.96827	16.14%	77.02%
5	0.71565	11.93%	88.95%
6	0.66292	11.05%	100.00%

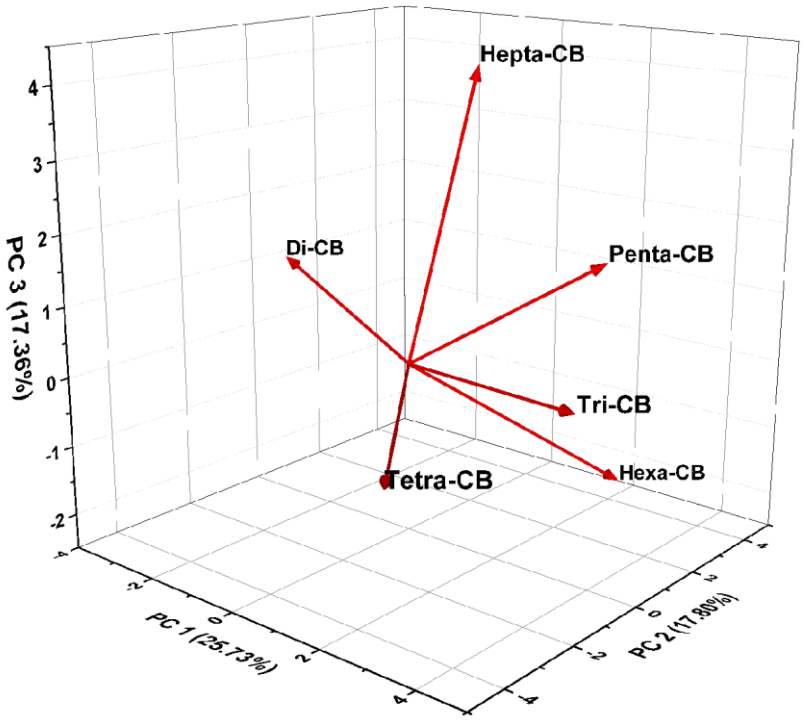


Figure 4.13. Indoor air PCA analysis

When the outdoor air samples were examined, two components were determined. These components explained (PC1 44% and PC2 20%) 64% of the variance. PC1 was loaded by Di, Tri, and Tetra-CBs, while Hexa and Hepta-CBs were dominant in PC2. The Penta-CB also contributed to PC2. The information obtained in the correlation analysis is supported by PCA. The heavy molecular weight groups of outdoor air samples show no significant correlation. Although low molecular weight PCBs are associated with gasoline exhaust emissions, PCB-28, 44, 52, 101, and 118 are shown as emissions from coal and

wood combustion (Salihoglu et al. 2011; Lee et al., 2005). At the same time, it is known that they are of industrial origin and that they are caused by AIA activity (Odabasi et al. 2009b; Aydin et al. 2014). It has been stated that especially ship breaking and iron-steel plants are the sources that make the most important contribution to PCB concentrations. It was found high in total concentrations of low molecular weight compounds such as PCB-17, 18, 22, 28, 31, 33, 44, 49, and 52 among PCB congeners (Aydin et al. 2014). For this reason, PC1 can be defined as industrial vehicle exhaust and combustion emissions for heating purposes. The dominant homologous groups in PC2 were Hexa-CBs and Hepta-CBs. Hepta and Hexa-CBs contain congeners PCB-156, PCB-169, and PCB-187. It was stated in a study that high PCB-156 concentration in Chennai city may be due to diesel engine emissions on highways and the city center (Laroo et al., 2013). PCB-187 is also generally said to show the mixed profile of Aroclor 1254, 1260, and Kanechlor 600 (Aydin et al., 2014). In this case, the high concentrations of PC2 on highways could have resulted from complex sources such as vehicular and industrial emissions and informal incineration.

Table 4.5. Outdoor air PCA analysis

	Eigenvalue	Percentage of Variance	Cumulative
1	**2.64933**	**44.16%**	**44.16%**
2	**1.19894**	**19.98%**	**64.14%**
3	0.96417	16.07%	80.21%
4	0.84675	14.11%	94.32%
5	0.25205	4.20%	98.52%
6	0.08876	1.48%	100.00%

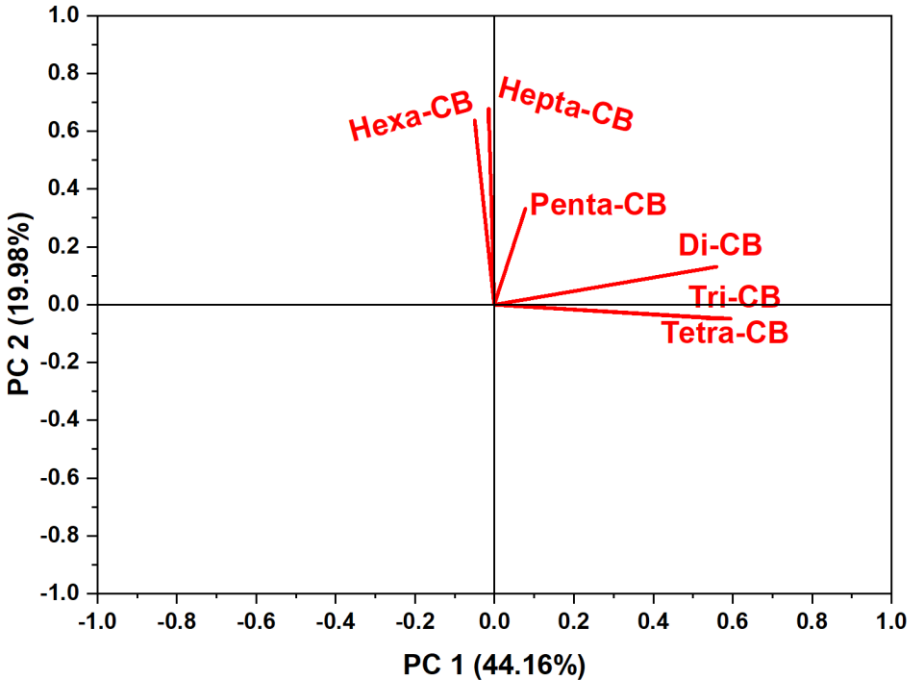


Figure 4.14. Outdoor air PCA analysis

Indoor environments constitute a large portion of exposure areas. Identifying the sources of indoor pollutants plays an important role in reducing them and taking the necessary precautions. For this reason, a comparison was made between home and school environments in samples taken from indoor environments. This comparison would help distinguish between different sources of pollutants in homes and schools. For these reasons, PCA was also performed for the two indoor environments separately, despite the apparent reduction in sample size.

When the indoor concentrations of the homes were examined, it was determined as a result of PCA that it could be explained by two components. PC1 and PC2 contribute with 36 % and 23 %, respectively, for a total explained variance of 59 %. The dominant homologue groups in PC1 was Di and Tri-CBs, while in PC2 was loaded again by Di and Tri CBs but also Penta-CBs. This indicates the existence of two different sources for Di and Tri-CBs, one of which is also a source of Penta-CBs. Although the data were not sufficient to support, Penta-CBs were associated with Hexa-CBs while the heavier groups (Hepta and Tetra-CBs) were originating from another source.

Table 4.6. Home indoor air PCA analysis

	Eigenvalue	Percentage of Variance	Cumulative
1	**2.17684**	**36.28%**	**36.28%**
2	**1.40369**	**23.39%**	**59.68%**
3	0.92853	15.48%	75.15%
4	0.67729	11.29%	86.44%
5	0.467	7.78%	94.22%
6	0.34666	5.78%	100.00%

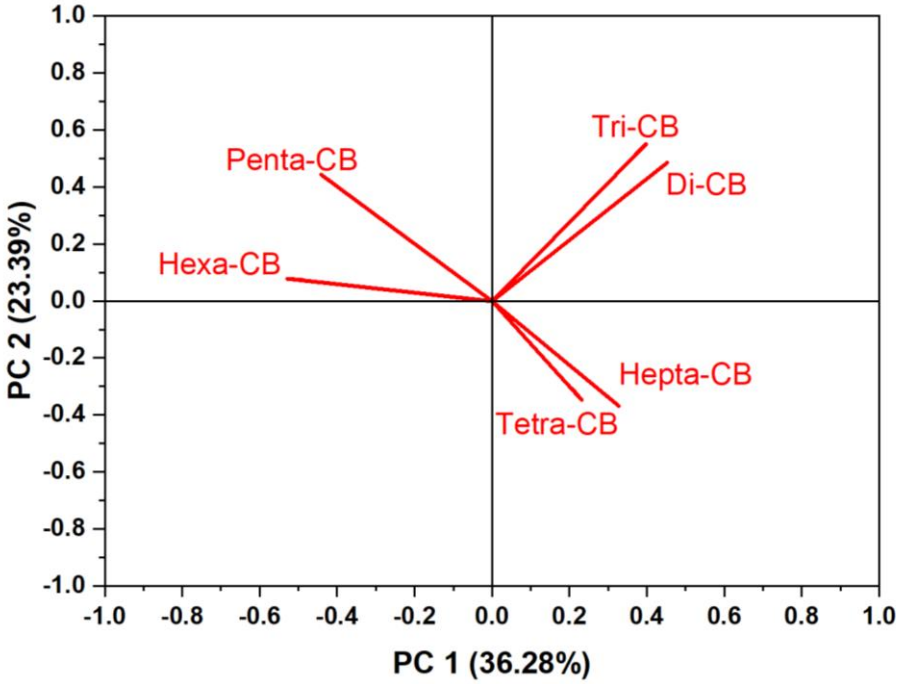


Figure 4.15. Home indoor air PCA analysis

While concentration variation in schools could be explained again with two components with a similar total explained variation of 60 %, PC1 contributed with 42 %, which was loaded by Tri, Tetra and Penta-CBs and PC2 by only Hepta-CBs contributing with 18 %.

So the difference between homes and schools are that there was a source that emits Tri-CBs in both types of buildings but that source emitted Di-CBs at homes while it emitted Tetra and Penta-CBs at schools. The second major source at homes emitted Penta-CBs along with Tri-CBs therefore may be the source that is represented by PC-1 at

schools. The second major source at schools, which was loaded by Hepta-CBs was strong enough to appear as a significant component, the source that emitted Hepta and Tetra-CBs was not as strong at homes.

Table 4.7. School indoor air PCA analysis

	Eigenvalue	Percentage of Variance	Cumulative
1	**2.53088**	**42.18%**	**42.18%**
2	**1.10129**	**18.35%**	**60.54%**
3	0.97816	16.30%	76.84%
4	0.73282	12.21%	89.05%
5	0.54655	9.11%	98.16%
6	0.11031	1.84%	100.00%

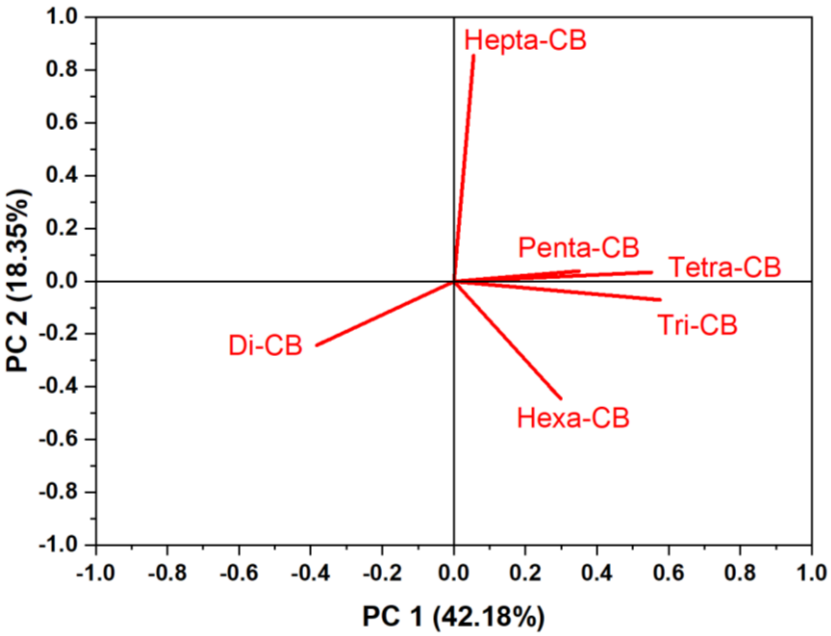


Figure 4.16. School indoor air PCA analysis

Low molecular weight groups predominantly represent all samples' first components throughout the PCA. In this case, the reference for the concentrations in the outdoor environment for low molecular weight PCB congeners can be iron-steel factory activities. Previously, Odabaşı et al. (2009) and Aydın et al. (2014) reported that low molecular weight PCBs dominate emissions of iron and steel plants that utilize scrap, and listed PCB-18, -28, -31, and -33 (Tri-CBs) as indicators for iron and steel production

emissions. Therefore, the source of Di, Tri, and Tetra-CB components in the outdoor environment could be iron-steel production. There are two such plants in AIA that is located in the northern part of the Province of Izmir, also to the north of our sampling sites with various proximity.

Commercial mixtures of PCBs have been produced in different countries and used in indoor materials. Although it is not produced in our country, the materials in our home and school environments imported from other countries can become a situation that increases the PCB content. Components in which low molecular weight groups that were found to be dominant in the indoor environment may be interpreted as technical PCB mixtures used before its ban. In low molecular weight PCB groups, Di, Tri, Tetra-CB, and medium weight Penta-CB groups are Aroclor 1248 and 1242. Aroclor 1242 and 1248 are used as plasticizers and epoxy resins to increase flame retardance and chemical resistance. These materials used indoors can release by emission. Hexa and Hepta mixed PCBs were expressed to be the dominant homologue groups in varnishing, adhesives, and paints (Audy et al. 2018; Taniyasu et al. 2003). Delor 106 is a mixture used in varnishing, adhesives, and paints. Delor commercial PCB mix produced at Czech Republic is expressed to have similar chlorination content as Aroclor mixes. Also, Aroclor 1254 has been used in indoor sealants and caulk compounds, flame retardants, and plasticizers to increase chemical resistance, chemical resistance (Styrene-butadiene copolymers), and pressure-sensitive adhesives and enhanced resistance, flame retardance, electrically insulating materials (Bois et al. 2018). The literature showing PCB profile emitted from paints/cabinets can also be used to compare the indoor profiles (Herkert, Jahnke, and Hornbuckle 2018).

4.7. Exposure and Carcinogenic Risk

In this section, exposure and risk estimates are presented. Monte Carlo Simulation Software Crystal Ball (Version 4.0) was used for simulating the exposure – risk models to obtain probability distribution that could be considered as descriptions of population exposure and risk. The estimations were based on homologue groups for consistence with the previous sections. Fifty percent detection frequency was the criteria to be included in

the assessment. The survey data of the study participants were used to calculate the time spent indoors and outdoors. According to the results of the survey, the time spent indoors during the 24-hour period in İzmir was 20.88 hours, while it was determined as 3.12 for the outdoor environment. Monte Carlo simulation was run for 10,000 trials, generating 10,000 estimates that were used to derive distributions describing population exposure and risk. Probability distributions assumed for the homologue group concentrations are provided in Table 4.8 The best-fitting distribution was selected based on Kolmogorov-Smirnov and Anderson-Darling tests.

Table 4.8. Probability distributions of exposure variables

Input Variables	**Fitted Distribution**	**Distribution Parameters**	**Test**	
[a]Di-non-DlCBs	Lognormal	Mean: 138.49 SD: 327.58	[d]KS: 0.14	[c]AD: 1.04
[b]Di-non-DlCBs	Lognormal	Mean: 25.28 SD: 21.20	[d]KS: 0.7	[c]AD: 0.3
[a]Tri-non-DlCBs	Lognormal	Mean: 382.34 SD: 403.40	[d]KS:0.09	[c]AD: 0.32
[b]Tri-non-DlCBs	Lognormal	Mean: 70.78 SD: 70.80	[d]KS:0.01	[c]AD: 0.40
[a]Tetra-non-DlCBs	Lognormal	Mean: 181.38 SD: 178.83	[d]KS: 0.09	[c]AD: 0.34
[b]Tetra-non-DlCBs	Lognormal	Mean: 61.18 SD: 66.51	[d]KS: 0.09	[c]AD: 0.35
[a]Penta-DlCBs	Lognormal	Mean: 64.42 SD: 290.34	[d]KS: 0.13	[c]AD: 0.86
[b]Penta-DlCBs	Lognormal	Mean: 166.96 SD: 386.53	[d]KS: 0.10	[c]AD: 0.47
[a]Penta-non-DlCBs	Beta	Alpha: 2.53 Beta: 4.27	[d]KS: 0.06	[c]AD: 0.15

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Table 4.8 (cont.)

Input Variables	Fitted Distribution	Distribution Parameters	Test	Input Variables
[b]**Penta-non-DlCBs**	Lognormal	Mean: 45.34 SD: 30.84	[d]KS: 0.01	[c]AD: 0.31
[a]**Hexa-DlCBs**	Lognormal	Mean: 46.67 SD: 111.42	[d]KS: 0.1	[c]AD: 0.32
[b]**Hexa-DlCBs**	Lognormal	Mean: 121.27 SD: 1,173.32	[d]KS: 0.11	[c]AD: 0.58
[a]**Hexa-non-DlCBs**	Lognormal	Mean: 118.46 SD: 108.48	[d]KS: 0.07	[c]AD: 0.17
[b]**Hexa-non-DlCBs**	Lognormal	Mean: 100.80 SD: 123.87	[d]KS: 0.10	[c]AD: 0.60
[b]**Hepta-DlCBs**	Lognormal	Mean: 23.62 SD: 124.64	[d]KS: 0.12	[c]AD: 0.48
[a]**Hepta-non-DlCBs**	Lognormal	Mean: 16.22 SD: 43.27	[d]KS: 0.01	[c]AD: 0.56
[b]**Hepta-non-DlCBs**	Lognormal	Mean: 43.45 SD: 176.70	[d]KS: 0.13	[c]AD: 0.71

[a]Indoor: pg/m3 [b]Outdoor: pg/m3 [c]Anderson Darling [d]Kolmogorov Smirnov

When we consider the activities that make up a typical day, we can see that most of our time is spent indoors. The literature studies and the surveys that we carried out as part of the project's scope allowed us to arrive at a value that we have used as the basis for our evaluation. This statistic implies that we spend around 20.88 hours of each 24 hours indoors. In this context, our goal in measuring the amount of time spent inside is to assist us in gaining a better understanding of the amount of exposure a person has on average during their lifetime to pollutants present in both indoor and outdoor contexts. The equation that we use to determine the degree of exposure indicates that the length of exposure from both indoor and outdoor sources is a significant factor. Calculations of

PCB exposure were carried out, which are shown in Table 4.9 The 95th percentile of the calculation of exposure performed based on homologue groups yields results from that range between 10^{-7} and 10^{-8}.

Table 4.9. The estimated values for PCBs inhalation exposure (mg/kg-day)

Statistics	Mean	Median	Standard Deviation	Minimum	Maximum	95th
∑Di-non-DLCB	3.80×10^{-8}	1.40×10^{-8}	9.49×10^{-8}	2.96×10^{-10}	2.77×10^{-6}	1.36×10^{-7}
∑Di-CB	3.80×10^{-8}	1.40×10^{-8}	9.49×10^{-8}	2.96×10^{-10}	2.77×10^{-6}	1.36×10^{-7}
∑Tri-non-DLCB	1.04×10^{-7}	6.81×10^{-8}	1.21×10^{-7}	2.58×10^{-9}	3.06×10^{-6}	3.05×10^{-7}
∑Tri-CB	1.04×10^{-7}	6.81×10^{-8}	1.21×10^{-7}	2.58×10^{-9}	3.06×10^{-6}	3.05×10^{-7}
∑Tetra-non-DLCB	4.95×10^{-8}	3.42×10^{-8}	5.27×10^{-8}	2.29×10^{-9}	9.87×10^{-7}	1.40×10^{-7}
∑Tetra-CB	4.95×10^{-8}	3.42×10^{-8}	5.27×10^{-8}	2.29×10^{-9}	9.87×10^{-7}	1.40×10^{-7}
∑Penta-DLCB	2.26×10^{-8}	8.97×10^{-9}	6.72×10^{-8}	4.98×10^{-10}	2.96×10^{-6}	7.46×10^{-8}
∑Penta-non-DLCB	1.41×10^{-8}	1.26×10^{-8}	8.28×10^{-9}	3.19×10^{-11}	8.45×10^{-8}	2.98×10^{-8}
∑Penta-CB	3.67×10^{-8}	2.40×10^{-8}	6.82×10^{-8}	2.16×10^{-9}	2.97×10^{-6}	9.10×10^{-8}
∑Hexa-DLCB	1.65×10^{-8}	6.88×10^{-9}	3.88×10^{-8}	4.90×10^{-10}	1.31×10^{-6}	5.75×10^{-8}
∑Hexa-non-DLCB	3.51×10^{-8}	2.53×10^{-8}	3.37×10^{-8}	1.80×10^{-11}	8.55×10^{-7}	9.54×10^{-8}

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Table 4.9 (cont.)

Statistics	Mean	Median	Standard Deviation	Minimum	Maximum	95th
∑Hexa-CB	5.15×10^{-8}	3.77×10^{-8}	5.30×10^{-8}	1.91×10^{-9}	1.37×10^{-6}	1.35×10^{-7}
∑Hepta-DLCB	9.16×10^{-10}	1.63×10^{-10}	4.07×10^{-9}	1.74×-13	2.28×10^{-7}	3.65×10^{-9}
∑Hepta-non-DLCB	5.66×10^{-9}	2.45×10^{-9}	1.09×10^{-8}	2.69×10^{-11}	2.30×10^{-7}	2.01×10^{-8}
∑Hepta-CB	6.57×10^{-9}	3.18×10^{-9}	1.17×10^{-8}	3.31×10^{-11}	2.39×10^{-7}	2.25×10^{-8}

The technique of determining a mathematical link between the quantity of a chemical to which a person is exposed and the chance that there will be an unhealthy reaction to that dosage is referred to as the dose-response method (EPA 1996). The purpose of risk assessment is to try to calculate the likelihood of developing a disease as a result of being exposed to a certain dosage of a chemical. For the purpose of determining the carcinogenic risk distribution for DLCBs and non-DLCBs, the exposure values were multiplied by SF that represents the link between the doses and the response. Homologue groups and total cancer risk are given in Figure 4.17, 4.18, and 4.19 Estimated 95th percentile inhalation exposures for İzmir province were found as 1.87×10^{-7}, 1.95×10^{-7}, and 3.35×10^{-7} respectively ∑DLCBs, ∑non-DLCBs and ∑PCBs.

USEPA (2000b) in general consider 10^{-6} as *de Minimis* and risks above this level to be unacceptable / undesirable. The conducted risk assessment showed that carcinogenic risk of the population associated with inhalation exposure to the concentrations measured by passive sampling were not considerable even at the 95th percentile. However, one should note that passive sampling measures gaseous-phase concentrations although there are reports that particles are also collected by the passive samplers (PUF-PAS) (Genisoglu et al. 2019).

The carcinogenic risk levels of PCBs that were assessed for ambient air in Urla, Izmir are comparable to those of our research. The computed inhalation risk for $\sum_{43}$PCB congeners was determined to be $1.27x10^{-7}$ (Gungormus, 2015). The measurements were

made with samples acquired with active (high-volume) sampling. The risk levels (95th percentile of 2.49x10^{-7}) estimated for a suburban site in Izmir was also similar (Ugranli et al. 2016). The PCB cancer risk was determined to be below the acceptable range (10^{-6}) in a research conducted in the Kütahya province of Türkiye, according to the data obtained from the outdoor air PCB concentrations, which was conducted in a study in which seasonal changes were noted (Dumanoglu et al. 2017). However, another study conducted in Türkiye was carried out in Dilovası. Based on the data obtained, the mean risk levels for PCBs were at the order of 10^{-5} for all sampling points. In addition, a value of 10^{-4} was found in locations considered likely to be contaminated with PCBs. Since Dilovası hosts the most extensive industrial network in Türkiye, a situation that exceeds acceptable risk levels may have been encountered (Cetin et al. 2018a).

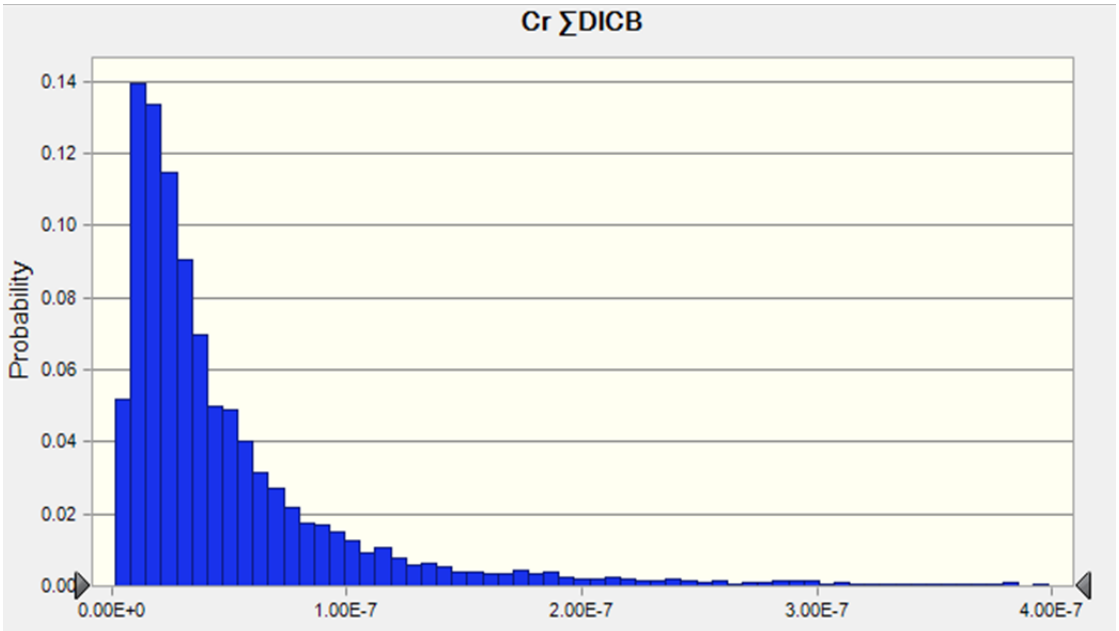

Figure 4.17. Estimated cancer risk (R) frequency histograms due to inhalation exposure of ∑DLCBs

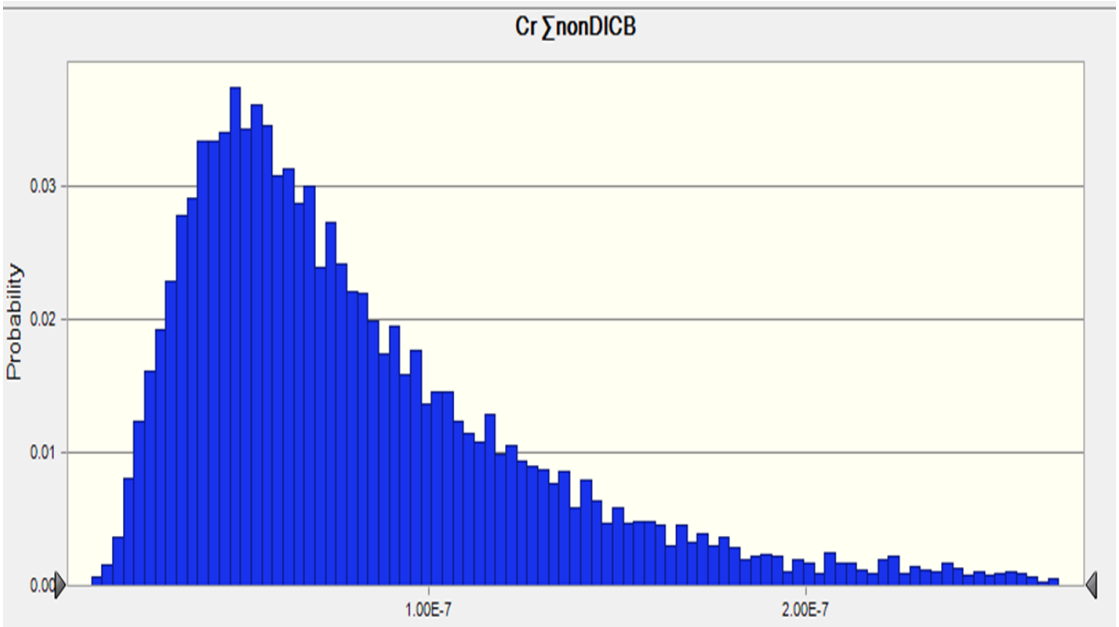

Figure 4.18. Estimated cancer risk (R) frequency histograms due to inhalation exposure of ∑non-DLCBs

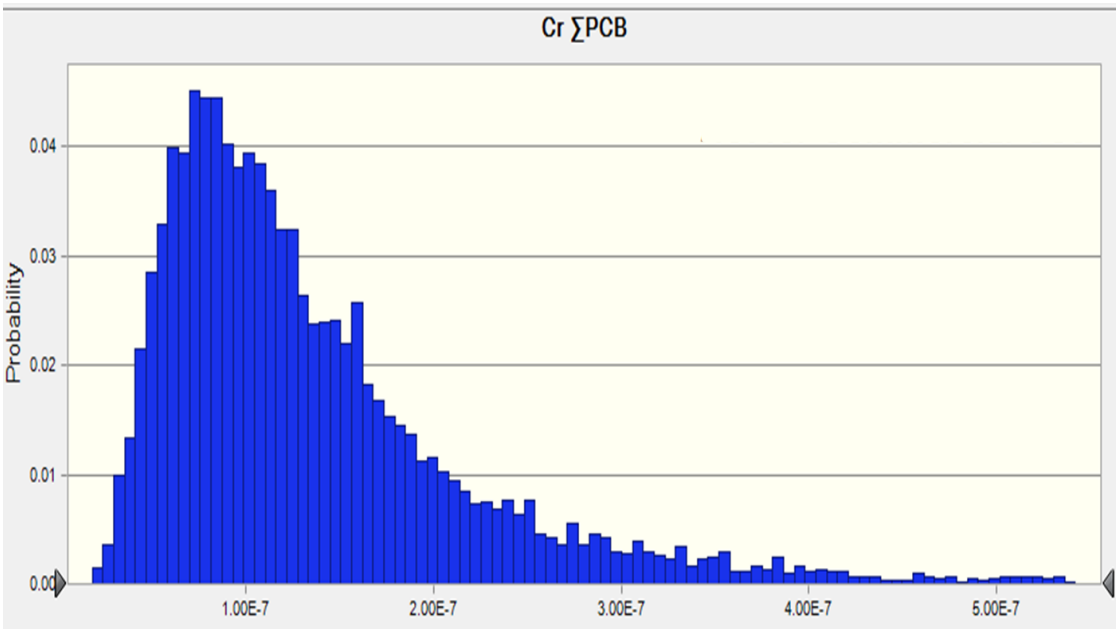

Figure 4.19. Estimated cancer risk (R) frequency histograms due to inhalation exposure of ∑PCBs

4.7.1. Sensitivity Analysis

Sensitivity analysis shows the correlation coefficients between each input and output used in risk assessment, as well as the impacts of input parameters on all other

variables. Depending on variance, variables of exposure-related risk were examined independently. The sensitivity analysis was carried out using Crystal Ball software.

The percentage contributions of the cancer risk data obtained for each homologue group and the overall PCB were examined. Indoor concentrations had the most significant effect on the risk. The overall sensitivity analysis revealed that the risk was most sensitive to the indoor ∑DLCB concentrations and breathing rate with 56.2% and 12.6% on the variance. The outdoor concentrations contributed 11.5% to the risks of ∑DLCBs. The Indoor concentrations contributed with 56.8 % to the risks of ∑non-DLCBs, followed by the inhalation rate with 28.4%. Contribution of the inhalation rate to the variation in the total risk was slightly higher at 32.9%.

In general, indoor concentrations have contributed the most considerable proportion to cancer risk across all homologue groups, assuming a generalization. Indoor congeners with dioxin-like and non-dioxin-like congeners contribute the most to cancer risk via their contribution to total concentration.

4.7.2. Uncertainty Analysis

The uncertainty in the probabilistic risk estimates due to the random selection process from the assumed input variable probability distributions, i.e. due to simulation, were analyzed. The bootstrapping was used for this purpose with 200 simulations of 1000 trials each. Using bootstrapping was feasible to compute the degree of confidence for each estimate of probabilistically assessed risks, as required by the USEPA (1999b) in risk assessment studies. Table 4.10 shows that the uncertainty in ∑DLCB, ∑nonDLCB, and ∑PCB cancer risk levels. These results show that the uncertainties arising from the Monte Carlo simulation remained at low levels.

Table 4.10. Uncertainty in the estimated statistics of carcinogenic risk

PCBs Risk	Statistics	5th	50th	Mean	95th
R ∑DLCB	**Minimum**	7.1×10^{-9}	2.78×10^{-8}	5.24×10^{-8}	1.26×10^{-7}
	Median	7.9×10^{-9}	3.06×10^{-8}	5.98×10^{-8}	1.64×10^{-7}
	Mean	8.02×10^{-9}	3.06×10^{-8}	6.04×10^{-8}	1.67×10^{-7}
	Maximum	8.8×10^{-9}	3.39×10^{-8}	7.45×10^{-8}	2.03×10^{-7}
R ∑nonDLCB	**Minimum**	2.66×10^{-8}	6.34×10^{-8}	7.84×10^{-8}	1.61×10^{-7}
	Median	2.93×10^{-8}	6.70×10^{-8}	8.31×10^{-8}	1.88×10^{-7}
	Mean	2.93×10^{-8}	6.73×10^{-8}	8.33×10^{-8}	1.88×10^{-7}
	Maximum	3.22×10^{-8}	7.22×10^{-8}	8.89×10^{-8}	2.14×10^{-7}
R ∑PCB	**Minimum**	3.89×10^{-8}	1.04×10^{-7}	1.31×10^{-7}	2.86×10^{-7}
	Median	4.45×10^{-8}	1.10×10^{-7}	1.40×10^{-7}	3.59×10^{-7}
	Mean	4.49×10^{-8}	1.09×10^{-7}	1.41×10^{-7}	3.58×10^{-7}
	Maximum	4.93×10^{-8}	1.18×10^{-7}	1.49×10^{-7}	4.25×10^{-7}

CHAPTER 5

CONCLUSION

Passive sampling was used to determine PCB concentrations of air samples taken from 21 homes and 21 schools indoors and outdoors in the province of Izmir. The sites of the homes and schools (7 rural, 7 semi-urban, and 7 urban) were randomly selected. PCB concentrations were investigated based on homologue groups. I/O ratio, correlation analysis, and principle components analysis were conducted to investigate variation in the data set and source apportionment. A Monte Carlo simulation was used to estimate lifetime average daily dose for inhalation exposure and associated carcinogenic risk.

The median values of PCB homologue groups were determined as 39, 261, 110, 78, 116, and 7 pg/m^3 for Di, Tri, Tetra, Penta, Hexa, and Hepta-CBs, respectively, in home indoor air. The median outdoor air concentrations outdoor the homes were determined as 12, 57, 38, 80, 80, and 15 pg/m^3 for Di, Tri, Tetra, Penta, Hexa-CBs, and Hepta-CBs respectively. The median values of the homologue groups were found to be 74, 243, 130, 53, 97, and 5 pg/m^3 for school indoor air, and 26, 56, 35, 156, 90, and 22 pg/m^3 for school outdoor air.

SLR analysis were employed to investigate AIA-PCB distance relationship. The analyses produced evidence for AIA to be a source of PCBs for İzmir because concentrations tend to decrease with distance from AIA. This decreasing trend is observed primarily in Di, Tri, and Tetra-CBs that may be originating from point sources in AIA.

As a consequence of the comparison based on homologue groups, we determined that indoor air concentrations in samples were typically greater than outdoor air concentrations indicating stronger sources indoors. Groups with I/O>1 at home indoors were Di, Tri, Tetra, and Hexa-CBs, while it was <1 for Hepta-CBs. Penta-CBs were found to be about 1 both indoors and outdoors. Di, Tri, Tetra, and Hexa-CB concentrations were higher in the indoor air of schools. Penta and Hepta-CBs was the homologue group with higher outdoor concentrations.

Correlation and principle components analyses indicated that homes and schools probably have different sources of PCBs that include commercial mixtures, materials and equipment, and outdoor air.

The total cancer risk associated with inhalation exposure to PCBs for İzmir population estimated by Monte Carlo simulation was found to be below the acceptable level of one-in-a-million even at the 95^{th} percentile as 1.87×10^{-7}, 1.95×10^{-7}, and 3.35×10^{-7}, respectively for ∑DLCBs, ∑non-DLCBs and ∑PCBs. The sensitivity analysis indicated that the most influential variable on exposure – risk is the concentrations followed by inhalation rate. Bootstrap analysis showed that uncertainty associated with the simulation process are low.

This study may contribute to efforts in developing effective prevention plans based on the dominant homologue groups, the concentration levels and health risk assessment, the current status of indoor air quality, and source apportionment. However, it would be statistically beneficial to increase the number of samples to be able to better represent a metropolitan city within the scope of the study. PCA analysis indicated Aroclor, Delor and Kanechlor mixtures as possible sources. The finding that indoor air PCB concentrations being higher than that of outdoors call for plans to eliminate PCB sources in the indoor environments and to provide a better ventilation to mitigate the indoor air pollution. For this reason, identification of indoor PCB sources is of importance.

Estimated carcinogenic risk levels were just under the acceptable level (10^{-6}). Although, the findings of this study led to the conclusion that carcinogenic risk associated with inhalation exposure to PCBs is not considerable for Izmir population, aggregate exposure through non-dietary dust ingestion, inhalation of particulate matter and dermal absorption may drive the aggregate risk to above the acceptable risk level. Furthermore, an assessment for children may result with a different conclusion because of their disadvantageous situation regarding environmental exposures, and schools having a different congener profile than that of homes. Therefore, recommendations for future study include assessments regarding the mentioned aspects. Additionally, inclusion of dietary ingestion is very important because food is known to be the main exposure media for PCBs. At the same time, a substantial increase in the number of samples and the extend of sampling campaign would help overcome some limitations of this study to recommend mitigation measures.

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