

ABSTRACT

Title of Thesis: Influence of Historic and Current Land Use Practices on PCB Contamination of Soils and Stormwater Sediments in the Chesapeake watershed.

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Polychlorinated biphenyls (PCBs) are persistent organic pollutants (POPs) originating from biphenyl compounds, which were banned in the United States under the Toxic Substances Control Act of 1976. PCBs continue to be detected in various water bodies despite this ban and Total Maximum Daily Loads (TMDLs) have been established in many regions to manage their levels. This study investigates the presence and concentration of PCBs in local soils and roadway sediments, which may become mobilized during stormwater events. Soil and sediment samples from different sites across Anne Arundel County, Maryland, representing various land uses and periods of development were analyzed for 209 PCB congeners, as well as profiles of the ten homologues of chlorinated PCBs. The study correlates PCB concentrations in sediments with land use types (residential, commercial, industrial, greenspace, institutional, energy site) and development eras (before 1970s and after 1970s). The total PCB concentrations ranged from 1.27 to 381 ng/g dry mass. Land use significantly impacted PCB concentration; residential areas (median: 35.6 ng/g) and industry areas (median: 69.8 ng/g) were higher than commercial areas (median: 16.9 ng/g), institutional areas (median: 11.7 ng/g) and greenspace (median: 11.3 ng/g). Within each land use type, no statistically significant differences in total PCB concentrations were observed between pre-1970s and post-1970s development. However, consistent reductions in both median and mean concentrations in post-ban sites indicated a potential influence of development era, with newer infrastructure likely contributing fewer legacy PCB sources. The number of chlorines per biphenyl ranged from 2.48 to 6.11. The toxic equivalency (TEQs) of the PCBs ranged from below 0.0004 to 41.2 pg/g considering all sites. A non-Aroclor congener,

PCB 11, was detected in all samples except in greenspace. These findings provide critical insight into PCB distributions across land use and development histories and support efforts to estimate PCB load reduction efficiencies in stormwater control measures, enabling more targeted remediation strategies in high-risk areas.

INFLUENCE OF HISTORIC AND CURRENT LAND USE PRACTICES ON PCB
CONTAMINATION OF SOILS AND STORMWATER SEDIMENTS IN THE CHESAPEAKE
BAY WATERSHED

by

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Chapter 1: Introduction

1.1. Background of PCBs

Polychlorinated Biphenyls (PCBs) are a group of persistent organic pollutants (POPs) derived from biphenyl with 1-10 hydrogen atoms replaced by chlorine. The formula is $C_{12}H_{10-m}Cl_n$ where $m+n=10$. Totally, 209 congeners exist. The structure of PCBs is shown in Figure 1.1.

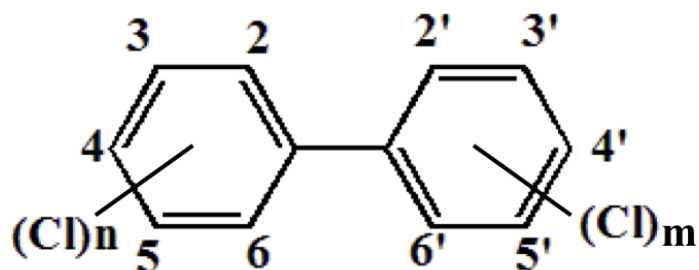


Figure 1.1. Structure of PCBs

Due to the chemical and physical stability of PCBs, and their electrical insulating characteristics, PCBs were commercially produced since 1929 and it was estimated there were totally around 591 million kilograms of PCB produced until 1976 (Erikson, 2018). They are hydrophobic, which means they are relatively insoluble in water. The solubility of PCBs range from 0.0012 to 4830 $\mu\text{g/L}$ (Humans, 2016) and the solubility decreases with increased chlorination.

In the United States, PCBs were primarily produced in the form of Aroclors mixture in the United States. Table 1.1 shows the physical and chemical properties of some Aroclor mixtures. Aroclors are identified by a four-digit code, where the last two digits show the mass

percent of chlorine in the mixture. For example, Aroclor 1260 means that the mixture contains 60 percent chlorine by weight. In the U.S., the most widely manufactured and used commercial mixtures are Aroclor 1016, 1242, 1254 and 1260, which account for approximately 13%, 52%, 16%, and 11%, respectively, of all PCB production from 1958 to 1977 (US EPA, 2004).

Table 1.1. Physical and chemical properties of some aroclors.

Property (Monsanto Company, 1985) (Hutzinger, 2017)	Aroclor 1242	Aroclor 1248	Aroclor 1254	Aroclor 1260
Molecular weight ^b	261	288	327	372
Physical state	Oil	Oil	Viscous liquid	Sticky resin
Boiling point, °C	325–366	340-375	365–390	385–420
Acidity, mg KOH /g maximum	0.015	0.010	0.010	0.014
Solubility:				
Water	Insoluble	Insoluble	Insoluble	Insoluble
Organic solvent(s)	Very soluble	Very soluble	Very soluble	Very soluble

PCBs were extensively utilized in the United States as cooling and lubricating agents in electrical devices such as transformers, building sealants and caulking, and paints. Although they were banned in the U.S. in 1979 by the Toxic Substances Control Act (TSCA), PCBs remain persistent in the environment due to their resistance to degradation and the continued allowance of PCB by-products under current United States Environmental Protection Agency (EPA) regulations (US EPA, 2021).

PCBs are toxic but have a toxicity range. Among the 209 PCB congeners, there are 12 dioxin-like PCBs (DL-PCBs): PCBs 77, 81, 126, 169, 105, 114, 118, 123, 156, 157, 167, 189 due to their structural and toxicological similarities to dioxins. These congeners are considered the most toxic among PCBs (Van den Berg et al., 2006). Toxic equivalency factors (TEFs) were developed for each DL-PCBs by the World Health Organization (WHO) to facilitate risk assessment based on toxicities relative to 2,3,7,8-tetrachlorodibenzodioxin (2,3,7,8-TCDD) (Van den Berg et al., 2006) which are shown in Table 1.2.

Table 1.2. Dioxin-like PCBs and toxic equivalency factors.

DL-PCB	TEF (Van den Berg et al., 2006)
77	0.0001
81	0.0003
126	0.1
169	0.03
105	0.00003
114	0.00003
118	0.00003
123	0.00003
156	0.00003
157	0.00003
167	0.00003
189	0.00003

The U.S. EPA uses total equivalents (TEQs) to account for how dioxin and dioxin-like compounds vary in toxicity, which involves multiplying individual TEFs with the concentrations of DL-PCBs, and summing them up (US EPA, 2015a).

1.2. Motivation

Stormwater runoff serves as a key transport pathway for PCBs in urban environments. The state of Maryland reported in 2018 that around 2500 million m² of Maryland's estuarine waters, approximately 423 km of rivers and streams, and approximately 13 million m² of lakes, reservoirs, and ponds, were contaminated with PCBs (US EPA, 2019). Urban runoff and storm sewers were estimated to account for around 17% of impaired riverine waters and 9% of impaired impoundment (US EPA, 2019), respectively, which were identified as major contributors to PCB pollution.

In response to these widespread impairments, Maryland has established Total Maximum Daily Loads (TMDLs) for PCBs (MDE, 2017) in multiple waterbodies due to their persistence, bioaccumulation potential, and exceedances of fish tissue advisory thresholds (Table 1.3) and Figure 1.2 shows the map of PCB TMDL watersheds in Maryland (MDE, 2017). These TMDLs aim to reduce PCB loads from both legacy sediment sources and ongoing inputs via stormwater and atmospheric deposition.

Table 1.3. PCBs TMDLs of watersheds in Anne Arundel County.

Watershed	TMDL (g/year)
Severn River	725.3
West River	517.7
South River	1237
Magothy River	356.1
Baltimore Harbor	1566.29
Curtis Creek	96.68
Patuxent River oligohaline	98.1

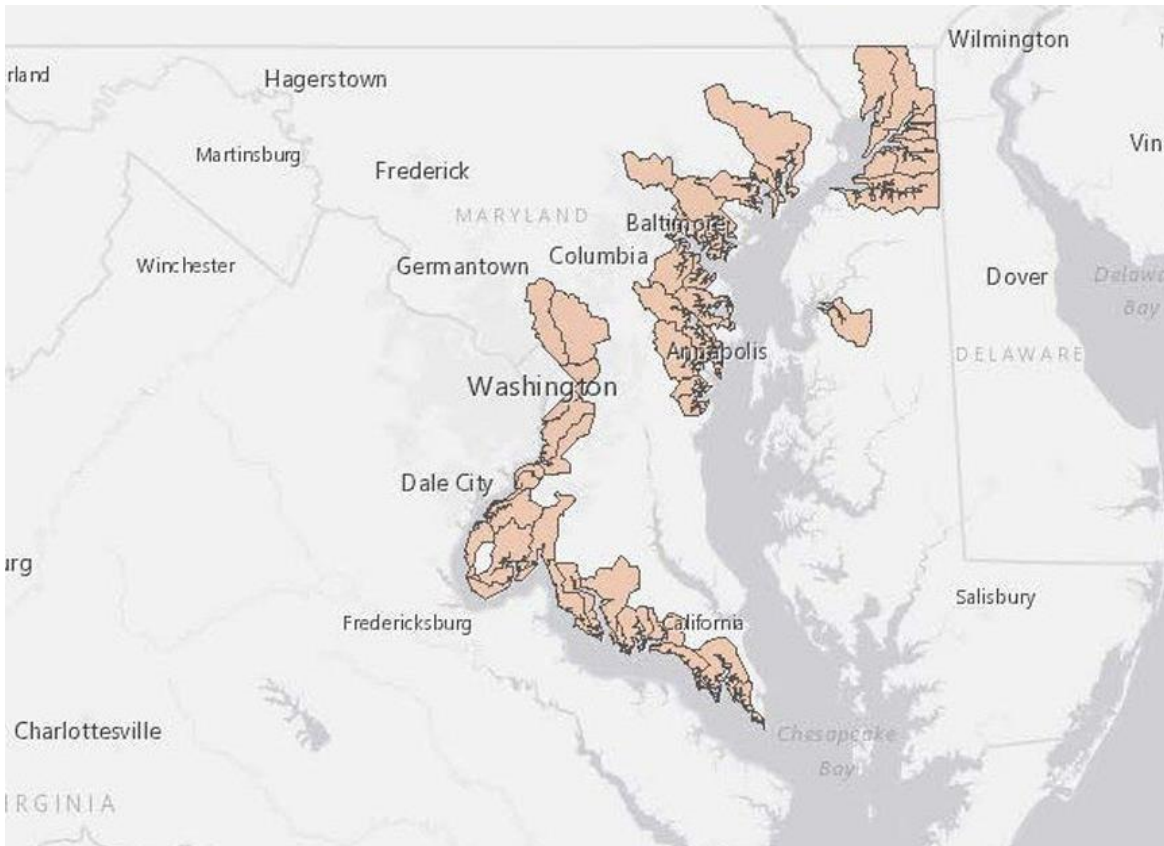


Figure 1.2. Map of PCB TMDL watersheds in Maryland.

Shaded areas represent river and estuarine segments for which the Maryland Department of the Environment has established PCB TMDLs.

Land use can influence PCB distribution through varying levels of impervious surfaces, pollutant generating activities, and runoff dynamics. Moreover, development era (pre- vs. post-1970s) may play a role in legacy contamination patterns due to historical use and disposal practices of PCB-containing materials. Understanding spatial patterns of PCB concentrations across land uses and development eras can inform local implementation strategies and source control efforts under the TMDL framework.

Stormwater mobilizes PCBs when precipitation runs off surfaces like roads and rooftops, picking up contaminants along the way. Studies have shown that a large proportion of PCBs in stormwater are associated with particulate matter, which have been found to account for more than 90% of the total PCBs (Hwang & Foster, 2008), indicating that particulate-bound PCBs should be a primary focus when evaluating PCB congener profiles in stormwater systems.

Previous research by Cao et al., (2019) showed that land use had an impact on PCB concentrations in stormwater sediment, where higher PCB concentrations were found in dense urban areas (average: 39.8 ng/g) and residential areas (average: 35.3 ng/g) compared to lower concentrations in greenspace samples (average: 18.0 ng/g). However, Cao et al., (2019) only focused on five types of land use (Dense Urban, Commercial, Institutional, Greenspace, Residential) and did not address the era of land development. The era of land development may be important because manufacturing of PCBs was banned in the U.S. in 1979. Areas developed before the ban are more likely to be directly impacted by historical PCB use due to the presence of materials such as PCB-containing sealants, hydraulic fluids, and electrical equipment that were widely used prior to regulatory restrictions (ATSDR, 2002). In contrast, areas developed after the ban may reflect lower direct inputs. However, those areas could still be affected by long-term environmental transport processes such as atmospheric deposition and stormwater

runoff. These pathways allow PCBs to persist and migrate through the urban landscape, even in sites without a known history of industrial and commercial PCB use. By including land development era as a factor, we could better understand how historical contamination patterns influence current PCB levels in soils and stormwater sediment. However, PCBs as by-products are still allowed under current regulations, and legacy PCBs continue to persist in the environment due to their strong resistance to thermal and chemical degradation.

Some PCB congeners generate specific concerns, such as non-legacy PCBs, including PCB-11 (3,3'-dichlorobiphenyl), which have emerged as indicators of ongoing, unintentional releases. Unlike legacy PCBs, which were manufactured and used in industrial and commercial applications before PCBs were banned in 1979, PCB-11 is unintentionally produced as a by-product of the manufacturing process for diarylide yellow pigments (Grossman, 2013). Although PCB-11 was not part of any Aroclor mixtures (ATSDR, 2002), it is still been widely detected in various environmental matrices including air (Hites, 2018) and stormwater (Cao et al., 2019). Therefore, understanding the sources and abundance of non-legacy PCBs in stormwater sediment is essential for assessing current contamination and for distinguishing between historical and ongoing inputs of PCBs in the environment.

To estimate potential PCB load reductions and address both legacy and ongoing sources of contamination within watersheds, it is essential to analyze the PCB mass concentrations and congener profiles. The congener profiles analysis can help distinguish between sources of contamination. Legacy Aroclor mixtures have specific homologue congener patterns (Table 1.4) (Frame et al., 1996), whereas non-Aroclor sources tend to exhibit different distributions and lack the patterns of Aroclor formulations. Identifying such patterns could help for source apportionment, distinguishing whether the contamination was due to aging infrastructure,

remobilized sediment, or ongoing diffuse inputs, which could help Stormwater Best Management Practices (BMPs) design improvements.

Table 1.4. Weight Percent of PCB Homologs in Some Aroclors.

	A1221	A1242	A1248	A1254	A1260
Mono	65.5				
Di	29.7	14.7			
Tri	4.8	46	20.9	1.8	
Tetra		30.6	60.3	17.1	
Penta		8.7	18.1	49.3	9.2
Hexa			0.8	27.8	46.9
Hepta				3.9	36.9
Octa					6.3
Nona					0.7
Deca					

1.3. Hypothesis

This study hypothesizes that the distribution and composition of PCBs in stormwater sediments and soils are influenced by both land use type and the land development. The intensity of land use is hypothesized to be a key factor shaping PCB accumulation patterns, with industrial and residential areas contributing higher contaminant loads compared to greenspaces, institutional lands, and commercial areas, which are likely to experience lower direct inputs and. Furthermore, it is expected areas developed before 1970s PCB ban, especially residential and industry areas will show elevated total PCB concentrations and congener profiles more closely aligned with historical Aroclor mixtures, reflecting legacy contamination from past use of PCB-containing materials in construction, electrical infrastructure, and industrial applications. In contrast, post-1970s developments are tended to demonstrate lower concentrations of legacy PCBs. In addition, non-Aroclor congeners, such as PCB-11 may be frequently detectable due to inputs from by-product of pigment manufacturing.

1.4. Objectives and tasks

The overall goal of this study was to better understand the occurrence, distribution, and potential sources of PCBs in stormwater systems to inform effective mitigation strategies. The objectives of this study were (1) to assess the land use and development age impact on the concentrations and congener distributions of PCBs in soils and stormwater sediments; (2) to assess the importance of legacy PCB sources and current sources on PCBs in the urban environment; (3) to use the above information to provide guidance on implementing stormwater control measures (SCMs) and designing recommendations to effectively remove PCBs from stormwater runoff.

These objectives are addressed through sampling of soils and stormwater sediments across diverse land use types and development eras, followed by PCB concentration and congener analysis, evaluation of source contributions, and estimation of PCB removal potential via stormwater control measures.

Chapter 2: Materials and Methods

Figure 2.1 showed the study design and analytical workflow of this research project. Following the flowchart, each component of the methodology would be described in detail in the subsequent sections.

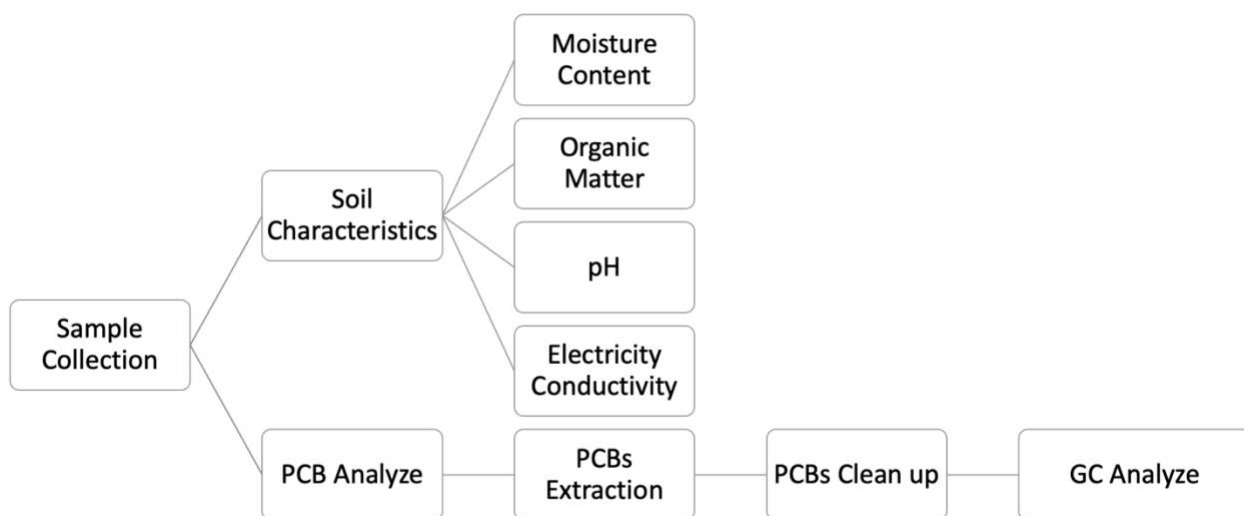


Figure 2.1. Methods flowchart.

2.1 Sites selection and sampling

The study was conducted in Anne Arundel County, Maryland, a region characterized by a mix of urban, suburban, and less developed land uses with different eras of development. From July 2023 to December 2024, a total of 34 samples were collected in Anne Arundel County, from stormwater gutters along the roadway, stormwater drainage points, and stormwater channels to

represent different land use and land era of development. A summary of the sampling sites is presented in Table 2.1.

Table 2.1. Summary of sampling sites and the number of samples collected from each category of land use and land development.

Land Use	Development	Samples	Samples
Residential	pre-70s	5	10
	post-70s	5	
Commercial	pre-70s	1	6
	post-70s	5	
Institutional	pre-70s	2	4
	post-70s	2	
Industry	pre-70s	4	9
	post-70s	5	
Energy Site	N/A	2	2
Green Space	N/A	5	5

Figure 2.2 shows a map of all sampling sites. Explanation of the abbreviation for sample names are given in Appendix A Table A.1. All sites and samples detailed information are in Table A.2. Figure A.1 shows examples of sampling points.

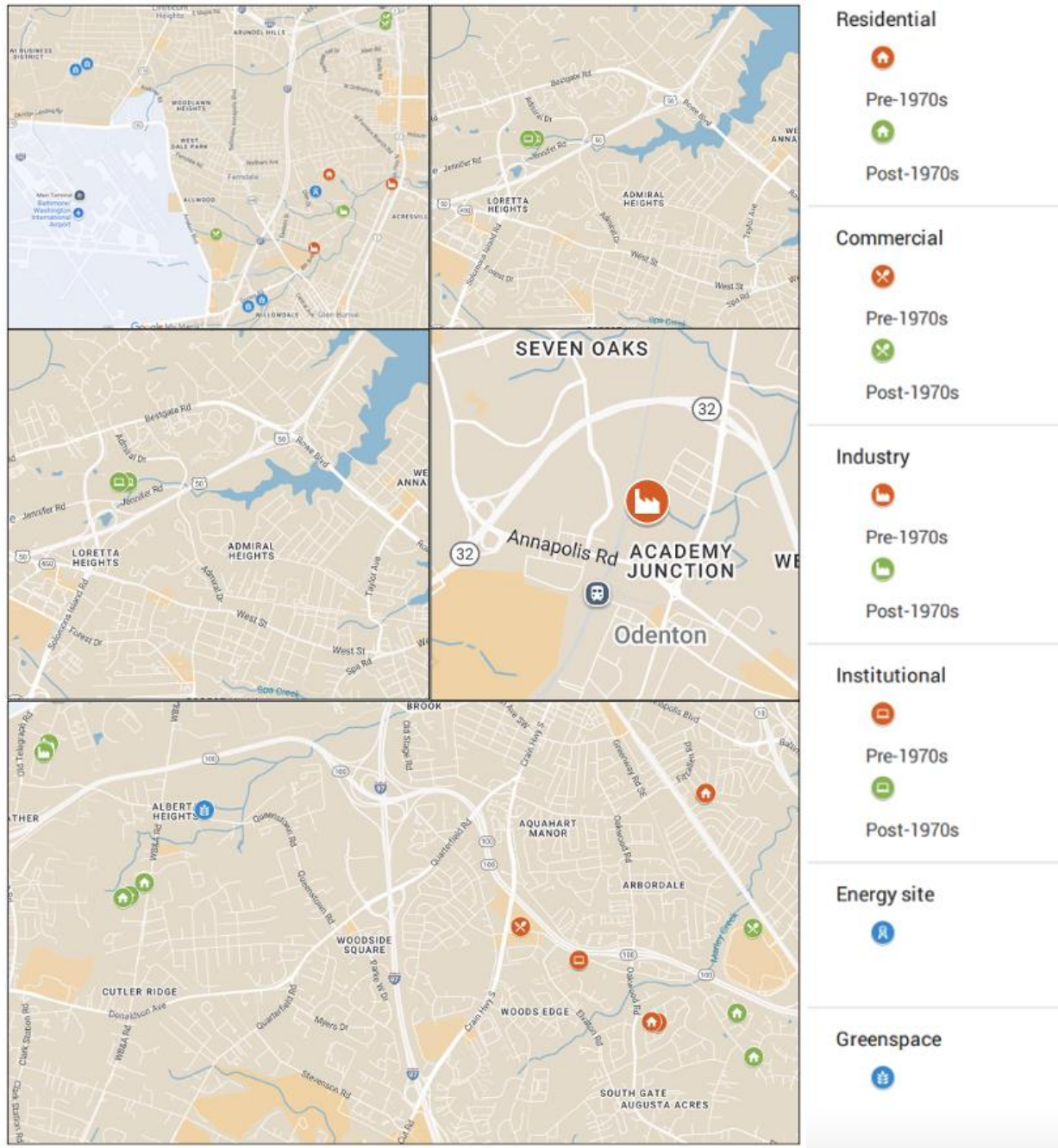


Figure 2.2. Map of the sampling sites.

Sampling locations were selected to represent a range of land use categories, including residential, industrial, commercial, institutional, greenspace, and energy areas.

Samples were collected once at each site. Surface sediment samples (0-7 cm were collected by using clean stainless-steel soil scoop and the sediment was placed in 32 oz glass mason jars with Teflon lids (Ball, USA). All sampling events took place from July 2023 to December 2024, with the air temperature ranging from 10 to 33 °C. To minimize biological and chemical transformations, all samples were stored at 4°C in a cooler in the field and in the University of Maryland, College Park Environmental Engineering laboratory until analysis.

2.2 Soil and stormwater sediment characterization

Moisture content (MC %), organic matter (OM %), pH, and electricity conductivity (EC) were measured for all samples in triplicate. MC % and OM % methods were based on method ASTM D2974 (ASTM, 2020). To determine the MC %, the sample around 2.5g was placed in an oven at 105°C for 24 hours to evaporate the moisture. Moisture content is calculated by weighing the sample before (a) and after drying (b). The difference in mass of the sample is used to calculate the moisture content. Dividing the mass change by the original sample mass determines the % moisture content on a mass basis. Equation is shown in equation 1. The measure limit of the balance was 0.01g which the MC% limit was 0.40%. All MC% below 0.40% would be reported in < MC% limit.

$$MC\% = \frac{a - b}{a} * 100\% \quad (1)$$

OM % is determined by Loss on Ignition. The fraction of organic matter in the soil is calculated by heating the dry soil sample to 550°C to allow the organic matter to burn off. The mineral fraction of the soil is resistant to combustion and remains behind. Dividing the weight change before (b) and after the combustion (c) by the dry sample weight (b) determines the % organic matter on a mass basis. Equation is shown in equation 2.

$$OM \% = \frac{b - c}{b} * 100\% \quad (2)$$

EPA method 9045D (US EPA, 2004) were used for preparing samples to test pH and electricity conductivity. A 10-gram air-dried sample was weighed and transferred to a 50 mL plastic centrifuge tube. Then 20 mL level 2 deionized (DI) water was added into the tube in a 2:1 DI water to soil ratio. An end-over-end mixer was used to mix the soil solution evenly for 30 min, and the samples were left to sit for 1 hour to settle.

Sample pH was measured using a pH meter (B40PCID, VWR, USA). The pH meter was calibrated daily using buffer solutions of pH 4, pH 7, and pH 10 (Sper Scientific, USA). The pH probe was inserted into the soil solution and stirred gently, not touching the sides and bottom of the container. The pH value was recorded when the instrument's indicated a stable value on the digital display.

EC was measured using a conductivity meter (VSTAR, Thermo Scientific, USA). The conductivity meter was calibrated daily using 0.01M potassium chloride (KCl) at 1411 $\mu\text{S}/\text{cm}$ at room temperature. The conductivity probe was inserted into the soil solution and stirred gently, not touching the sides and bottom of the container until the EC value was showed stable on conductivity meter.

2.3 PCB extraction

Microwave-assisted extraction (MAE) was used to extract PCBs from sediment samples using a MARS 6 system (CEM, USA). The method of Cao et al., (2019) was used for extraction with minor modification. A sample consisting of approximately 5 g of sediment that was air-dried for 24 h was weighed into an aluminum dish and transferred into a 100 mL Teflon-lined

extraction vessel (CEM, USA). A solvent mixture of 30 mL hexane and acetone (1:1) was added to each vessel. All solvents were HPLC grade (Fisher Scientific, USA and Lab Alley, USA). Prior to extraction, 20 μ L of a mixed surrogate standard solution (0.5 ppb 2,4,6-trichlorobiphenyl (PCB 30) and 2,2',3,4,4',5,6,6'-octachlorobiphenyl (PCB 204)) was added into each sample to enable recovery assessment. All PCB standards were obtained from AccuStandard, USA. Extraction was conducted at 115°C for 10 minutes under 1000 W. Post extraction, the vessels were cooled to room temperature for approximately 1 hour in a fume hood before opening. The supernatant was transferred into 60 mL amber vials (Fisher Scientific, USA), while the solid residues were sequentially rinsed with 5 mL each of hexane, hexane-acetone (1:1), and acetone, and combined into the supernatant. The extracts were then concentrated to less than 100 μ L under a nitrogen flow evaporation. One mL hexane was added to the amber vials to dissolve the extracts and they were stored at -20 °C until further cleanup.

2.4 Cleanup of PCB extracts

EPA method 3611B (US EPA, 1996), 3620C (US EPA, 2015c) and the method of Cao et al., (2019) were used for the cleanup of PCB extracts, with minor modification. Alumina (Fisher Scientific, USA) was heated at 550°C for at least 24 hours, then cooled to room temperature. DI water was added to the alumina to deactivate it (30 μ L DI water per g alumina) and the alumina was equilibrated in the desiccator for 16 hours before use. Sodium sulfate (Fisher Scientific, USA) was heated at 550°C for at least 4 hours, cooled to room temperature, and left in a desiccator before use. Ten mL glass disposable pipets (Pyrex, USA) were used as cleanup columns, prepared in three layers; glass wool was placed at the bottom, then 5 g of the activated alumina, and finally a layer of oven-dried sodium sulfate at the top to remove moisture from the extracts. The column was first equilibrated with 20 mL of hexane. Afterward the extracts were

transferred into the column, and the effluent was collected. The amber vials were rinsed with 20 mL of hexane, and the rinsate was added to the column. The collected effluent was concentrated by ultra-high purity 99.999% nitrogen (Robert Oxygen, USA) to reduce the volume to less than 1 mL, then spiked with 20 μ L of a mixed internal standard solution containing 0.5 ppm 4-bromobiphenyl (4-BB) and 2,2',4,5,5'-pentabromobiphenyl (penta-BB). Hexane was added to the concentrated effluent to reach a final volume of 1.0 mL, and the samples were vortexed for 10 seconds before being transferred to 1.5 mL GC vials for further analysis.

2.5 GC analysis

All samples were quantified by using gas chromatography with the electron capture detector (GC-ECD; 7890B, Agilent Technologies, USA), fitted with an Agilent J&W HP-5ms capillary column (60 m \times 250 μ m \times 0.25 μ m). Helium served as the carrier gas. Samples were injected by automated autosampler (Model 7693, Agilent Technologies, USA) with a 1 μ L injection volume.

The GC oven temperature program (detailed in Appendix B. Table B.1) was adapted from Cao et al., (2019). The initial oven temperature was 70°C and was increased at a rate of 7 °C/min to 180°C, followed by 1 °C/min to 225°C, then 5.8°C/min to 285°C, holding for 20 min and finally 11.5°C/min to a final temperature of 300°C, holding for 10 min.

2.6 Quality assurance and quality control

All 209 PCB congeners were analyzed in 134 peaks. PCBs were detected in all samples. PCB 30 and PCB 204 were used as the surrogate standards for mono-to tetra- CBs, and penta-to deca-CBs, respectively. Average surrogate recoveries were in range 70.7-121% for PCB30 and 60.3-117% for PCB 204 which were all within the acceptable range of 50-125% (Hermanson &

Johnson, 2007). The measured concentrations were not corrected for the recovery. TCMX and penta-BB were used as the internal standards for mono-to tetra- CBs and penta-to deca-CBs, respectively.

Method detection limits (MDL) for PCB congeners ranged from 0.0103-0.5699 ng/g. The MDL were based on U.S. EPA (2016) with minor modification. Briefly, 7 spiked samples were prepared at 10 times the instrument detection limit and 3 method blank samples were processed through all steps of treatments and analyzed by GC. The MDL of each peak was calculated by multiplying the standard deviation of the replicate spiked sample analyzed concentration by single-tailed 99th percentile t statistic value with n-1 (n is number of replicate spiked samples) degrees of freedom, then divided by the sample mass. More detailed determination steps are given in Appendix B S1.

All sampling and analyses were conducted using pre-cleaned glass or Teflon containers to prevent PCB contamination. All glassware was solvent-rinsed (hexane, acetone, methanol) twice, acid washed by using 10% v/v H₂SO₄ for 8–14 hours, rinsing with DI water. All glassware (other than volumetric glassware) were baked at 550°C for 4 hours before use. Volumetric glassware was rinsed with hexane (HPLC grade) before use. Teflon-material containers were ultrasonicated with hexane and acetone and rinsed with DI water.

To confirm the surrogate standards and internal standards were not present in the environmental sediment samples, one treatment replicate was prepared as a standard control for each sample. The control was treated using the same methods as the other treatments, but without surrogate standards and internal standard addition. Washed and dried standard sand

(Supelco, USA) replicate was used as laboratory blank for each run. If the laboratory blank showed PCBs detected above MDL, all samples of that run were redone.

During each GC run, clean hexane (HPLC grade) was added at the beginning and the end of each run, and between every 3-5 samples. These steps were used to avoid carry over between samples in the GC analysis.

2.7 PCB concentration data analysis

The Kaplan–Meier (KM) survival procedure was used to sum total PCBs considering concentrations less than the detection limit (Helsel, 2005). The results calculated based on the KM method are between the estimated results when 0 and 1/2 MDL were substituted for censored data below MDL (Cao et al., 2023) which can avoid over- or underestimating the PCB concentrations. Briefly, the concentrations of all congeners were arranged in descending order. The MDL value was assigned for congeners with values below MDL. Each congener was then given a ranking r from highest to lowest (n to 1). A status d was then assigned to each congener with 1 indicating that the congener was above the MDL, 0 indicating it was below the MDL. Quantiles were determined for each congener and the area of each congener was calculated. Finally, the total concentration of the target congener group was obtained by multiplying the total area of the congeners by n . An example of this calculation is showed in Table B.2.

2.8 Data analysis

All values are presented as the mean of triplicates plus/minus the standard deviation unless noted. Concentrations of the PCBs were calculated based on data from GC analysis. Concentrations were equally divided when there was co-elution of PCBs into a single GC peak.

Toxicity and concentration of the 12 DL-PCBs: PCBs 77, 81, 126, 169, 105, 114, 118, 123, 156, 157, 167, 189 were calculated. The average number of chlorines per biphenyl was determined based on average molar concentrations.

Pearson's correlation analysis was conducted among soil physicochemical properties and PCB concentrations. Correlation coefficients were computed for all pairwise combinations of variables and statistical significance was assessed at the 5% level ($p < 0.05$). (Significance levels are indicated as: * < 0.05 ; ** < 0.01 ; *** < 0.001).

Due to the non-normal distribution of PCB concentrations (Shapiro-Wilk test, $p < 0.05$), non-parametric methods were used. The Kruskal–Wallis test followed by Conover–Iman post hoc comparisons with Holm correction were performed based on median values to analyze the land use and land development impact on PCB concentrations.

Chapter 3: Results and Discussion

3.1 Soil physicochemical properties and PCB concentrations.

A total of 34 stormwater sediment samples was collected from sites representing a range of land uses and development eras, including industrial, residential, institutional areas, recreational greenspace, and an energy site. All soil samples are loam soil. For all samples, the moisture content ranged from < MC% limit (< 0.4%) with the sample from residential area developed before 1970s (Res-Pre70-4) being the highest at 42.3 %. The highest organic matter content was also observed in the same sample (Res-Pre70-4), reaching 32.4%, with values across all samples ranging from 0.24 to 32.4% (Table 3.1). Electrical conductivity (EC), which reflects the ionic strength of soil and sediment, ranged from 62.3 to 806 $\mu\text{S}/\text{cm}$ among all collected samples. The pH values of the samples varied from 6.15 to 8.84. The soil physicochemical properties for all samples are shown in Table C.1.

Table 3.1. Soil moisture content, organic matter, electricity conductivity, and pH for all collected samples.

n=34				
Parameter	Max	Min	Average	S.D.
MC (%)	42.3	< 0.4	11.9	12.1
OM (%)	32.4	0.24	10.8	6.89
EC ($\mu\text{S}/\text{cm}$)	806	62.3	183	142
pH	8.84	6.15	7.55	0.61

These soil physicochemical properties were measured to explore their potential influences on PCB accumulation in soil and stormwater sediments. These parameters are

commonly considered in contaminant fate studies due to their known roles in controlling the adsorption, mobility, and degradation of hydrophobic organic pollutants like PCBs (Adeyinka & Moodley, 2019; Ćwieląg-Piasecka, 2023).

Each collected soil sample was analyzed for 209 PCB congeners, allowing for detailed assessment of congener specific profiles and total PCB concentrations. PCBs were detected in all samples, as presented in Figure 3.1. The description of total PCB concentrations among all samples is presented in Table 3.2. The variability reflected differences in source influence, land use history, and sediment or soil properties across sampling sites. Congener-specific quantification allowed for calculation of total PCB concentrations as well as evaluation of compositional differences among sites.

Table 3.2. Description of total PCB concentrations in all samples.

	Mean	S.D.	Range		Median
			Min.	Max.	
Total PCB concentration (ng/g) n=34	50.3	68.2	1.27	381	31.2

Samples from an industrial area and residential area developed before 1970s (ID-Pre70-1, Res-Pre70-5) had the two highest total PCB concentrations with a total PCB concentration of 381 ± 13.7 ng/g and 157 ± 1.61 ng/g, respectively, suggesting legacy contamination from historical urban development and industrial practices. The lowest total PCB concentrations in this study were collected from a greenspace and an institutional area, which showed concentrations of 1.27 ± 0.31 ng/g and 5.15 ± 0.59 ng/g, respectively, indicating limited exposure to direct PCB sources or more recent development with fewer legacy inputs. These patterns suggest that both land use

and development era were potential factors of PCB accumulation in soil and stormwater sediments.

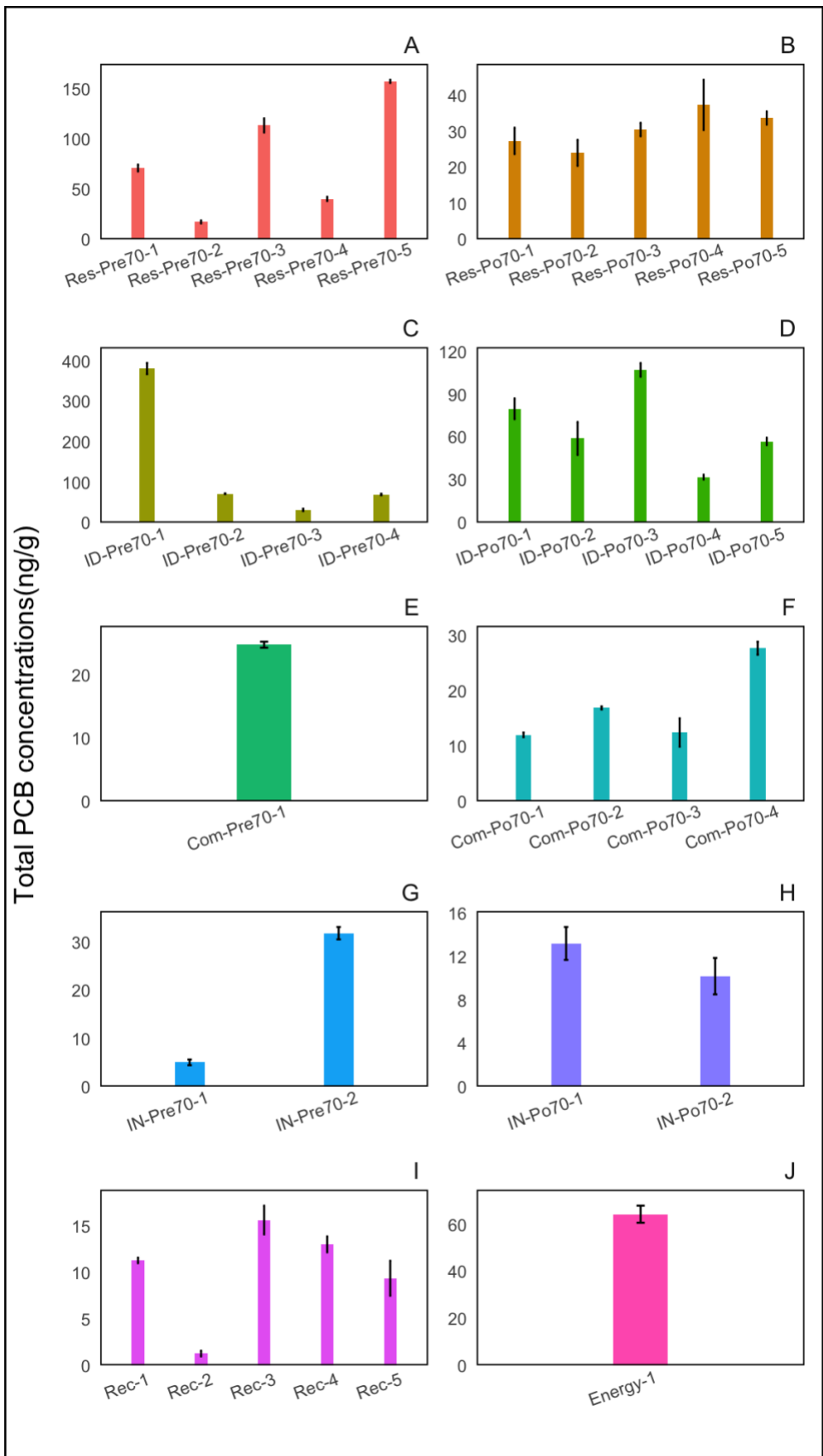


Figure 3.1. Total PCB concentrations for all samples under land use different categories. A.) Residential area developed before 1970s. B.) Residential area developed after 1970s. C.) Industrial area developed before 1970s. D.) Industrial area developed after 1970s. E.) Commercial area developed before 1970s. F.) Commercial area developed after 1970s. G.) Institutional area developed before 1970s. H.) Institutional area developed after 1970s. I.) Greenspace area. J.) Energy site. Note: error bars represent the standard deviation for three replicates; each graph has a different scale.

The correlations between the measured soil characteristics were investigated through Spearman’s rank correlation analysis (Figure 3.2).

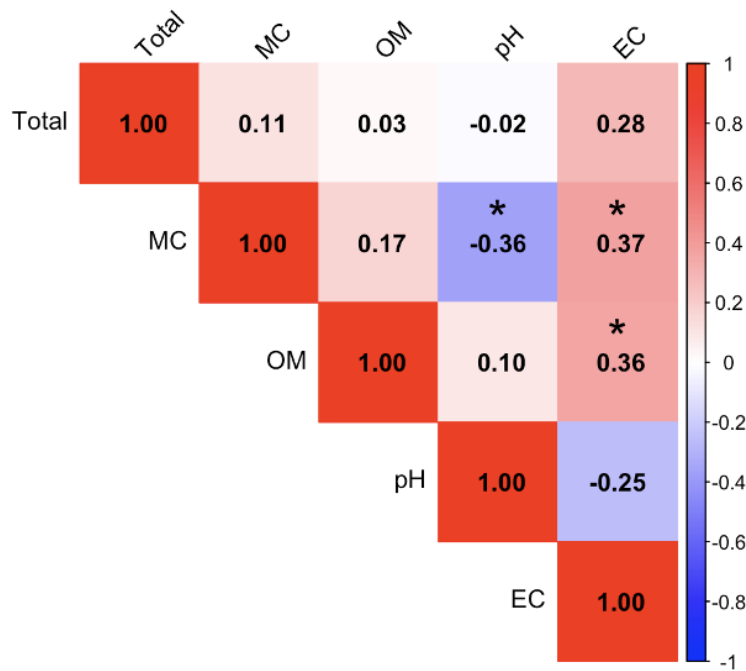


Figure 3.2. Correlation analysis between Total PCBs concentration and soil characteristics.

red = positive, blue = negative; significant level: * < 0.05; ** < 0.01; *** < 0.001.

In this study, no significant correlation between total PCBs and MC and OM have been found ($p > 0.05$). This finding contrasted with research performed by Adeyinka & Moodley (2019) which demonstrated that PCB adsorption onto soil decreased by approximately 48% following the removal of organic matter at 440 °C. The absence of a significant correlation in our study may indicate that other site-specific factors, such as historic PCB inputs, land use, or surface cover characteristics might have more influence in determining PCB levels than current MC and OM content.

EC was measured to provide information into the ionic strength of the soil solution. Similar to a previous study (Niu et al., 2022), we analyzed EC and PCB concentrations directly in soil and sediment samples. However, Niu et al. (2022) showed a significantly positive correlation between EC and total PCB concentrations in soil. In our study, no significant correlation was observed between total PCB concentrations and EC ($p > 0.05$). In contrast, this result aligned more closely with the findings of Adeyinka & Moodley (2019), who conducted a controlled laboratory experiment and showed the ionic strength did not significantly impact the adsorption of PCBs onto soil particles. The contrast between our results and those of Niu et al., (2022) may be due to site-specific differences in source inputs. The land use in their study was typical paddy fields from China. Our results suggest that ionic strength is not a controlling factor in PCB distribution across our sites, and that legacy contamination, land use history, and source proximity are likely more influential.

Similarly, there was a previous research showed that no significant correlation between total PCB concentrations and pH ($p > 0.05$) was found, which agree with the results in this study (Li et al. 2023).

In this study, total PCB concentrations varied widely across sites but showed no significant correlation with any measured soil physicochemical parameters, including organic matter (OM), electrical conductivity (EC), moisture content, and pH ($p > 0.05$). This lack of correlation suggested that inherent soil properties were not the primary factors driving PCB distribution in these urban watersheds and there is no need to normalize PCB concentrations results by these soil parameters in subsequent analyses. Instead, land use type, development era, or historical contamination sources were likely to play a more dominant role in shaping PCB accumulation patterns across the watershed.

3.2 Land use and land development impact on PCB concentration in stormwater sediments.

3.2.1 Effect of land use

Land use type played a critical role in determining the sources, and accumulation of polychlorinated biphenyls (PCBs) in urban environments. Areas with more intense human activity, such as industrial and residential zones, were often associated with higher levels of legacy contaminants due to past usage and infrastructure density, while greenspaces and institutional areas may experience lower contaminant inputs. In this section, we evaluate how different land use categories influence total PCB concentrations in soil and stormwater sediments.

Both median and mean values are reported for descriptive purposes and to compare with other studies (Table 3.3).

Table 3.3. Concentrations of Total PCBs (ng/g) by different land use categories.

Land Use	Mean	S.D.	Range		Median
			Min.	Max.	
Res (n=10)	55.2	45.7	157	17.4	35.6
ID (m=9)	98.8	108	381	31.7	69.8
Com (n=5)	18.8	7.17	27.7	12.2	16.9
IN (n=4)	15.1	11.7	31.9	5.05	11.7
GS (n=5)	10.1	5.47	15.7	1.27	11.3
Energy (n=1)	64.4	n.a	64.4	64.4	64.4

According to the Kruskal–Wallis test ($p < 0.05$) followed by Conover–Iman post hoc comparisons with Holm correction, industrial and residential land uses had significantly higher median concentrations than commercial, institutional, and green space areas, but industrial and residential were not significantly different from each other (Figure 3.3). In addition, industrial areas had a mean concentration of 98.8 ± 108 ng/g, nearly double of the next highest residential area (55.2 ± 45.7 ng/g). In contrast, green spaces had the lowest mean concentration (10.1 ± 5.47 ng/g), with minimum values as low as 1.27 ng/g. These trends are consistent with findings by Cao et al. (2019) who found that dense urban areas had the highest mean PCB concentrations (39.8 ± 10.5 ng/g), followed by residential areas (35.3 ± 6.2 ng/g), while commercial and greenspace zones showed lower levels. Although industrial areas were not specifically analyzed in their study, these results reinforce the importance of considering land use when evaluating pollutant sources and planning stormwater management interventions for PCB reductions.

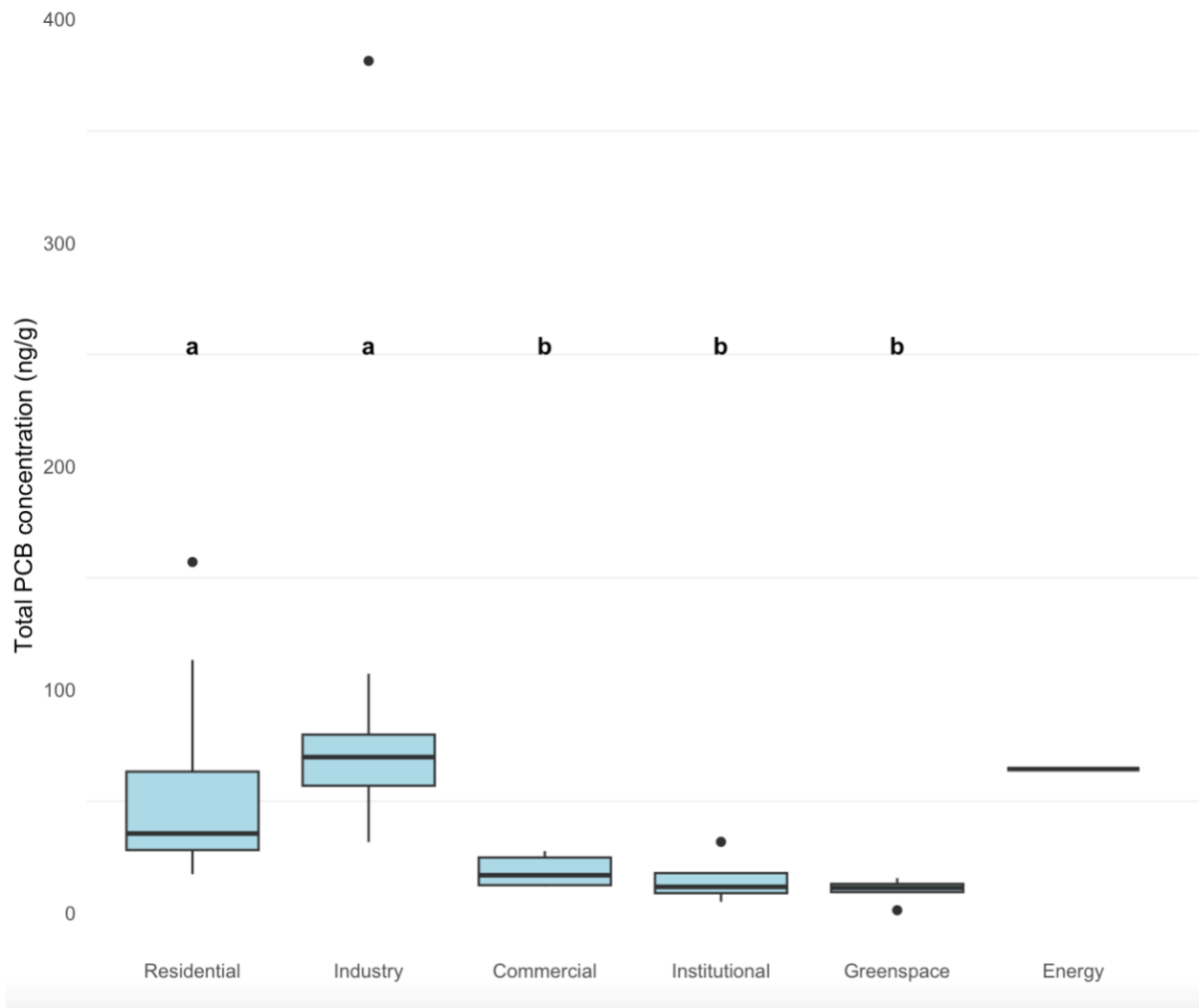


Figure 3.3. Total PCB concentrations across different land use types.

The box represents the interquartile range (IQR), the horizontal line indicates the median, and whiskers extend to $1.5 \times \text{IQR}$. Outliers are shown as individual points. Groups with the same letter indicate no significantly different ($p < 0.05$), according to a post-hoc Conover–Iman test with Holm correction following a Kruskal–Wallis test. Notes: The land use without the letter means there is only one sample.

Higher PCB levels in industrial areas compared with commercial, institutional, and greenspace sites reflected the influence of legacy contamination from historical industrial operations, as well as possible ongoing PCB emissions. Past industrial uses may include storage or disposal of Aroclor products, and contamination from old transformers, capacitors, or hydraulic systems (US EPA, 2004). Additionally, some industrial zones may still act as active sources due to operations such as metal recycling, handling of used electrical equipment, or scrap yards, which have been shown to release PCBs through leaks, volatilization, or contaminated dust and runoff (Seo et al., 2025). In this study, the highest PCB concentration detected in stormwater sediment was 381 ± 13.7 ng/g, collected from an outdoor scrap yard (Figure 3.4). This concentration far exceeds values reported in other industrial samples. For example, Li et al.'s study (2023) investigated PCBs in surface soils around an industrial park in China and found maximum concentrations of 1.24 ng/g (1240 pg/g). The comparison illustrates that the outdoor scrap yard site in this study exhibited PCB concentrations more than 300 times higher, emphasizing the critical localized contamination in areas handling legacy electrical equipment or metal recycling. During rain events, PCBs bound to fine particles can be washed from exposed surfaces into surrounding soils, leading to localized accumulation.

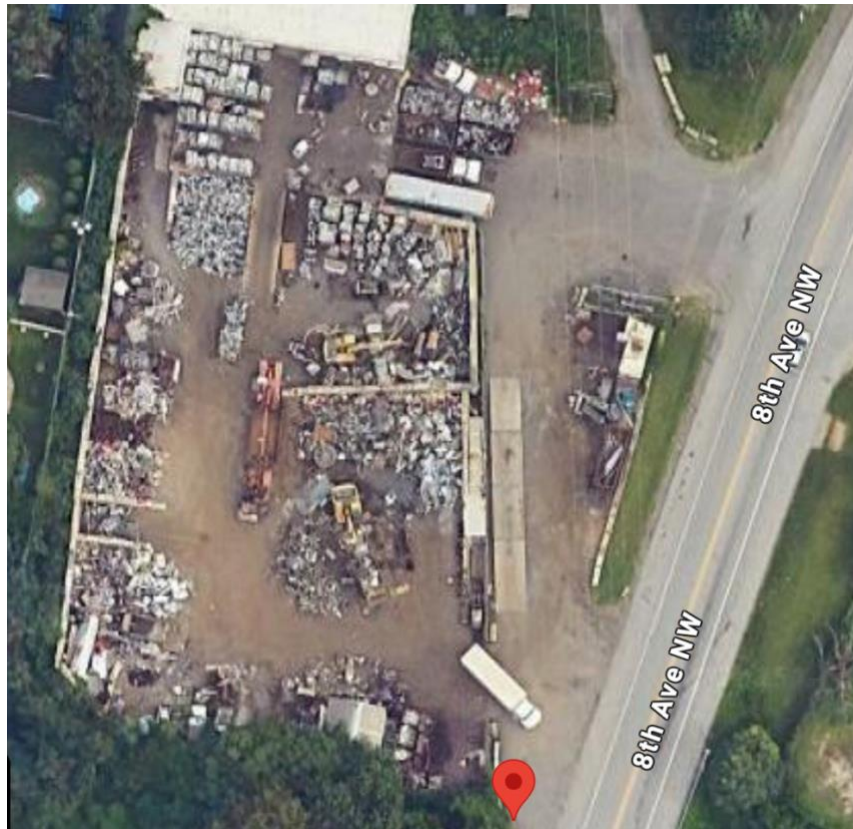


Figure 3.4. Google Map of outdoor scrap yard (39.173698, -76.628208)

where ID-Pre-70-1 collected.

Similarly, residential areas also exhibited higher PCB concentrations than commercial, institutional, and greenspace sites. These levels likely reflect legacy PCB contamination from past uses of sealants, caulks, roofing materials, which were common before the PCB ban (Diamond & Hodge, 2007). These effects may be especially pronounced in densely developed or older neighborhoods; land development history is analyzed separately in the following section.

In contrast, commercial, institutional and greenspace sites had lower PCB levels and showed no significant difference among these land uses. This may be due to newer construction, which phased out PCB containing materials following regulatory bans after 1970s. Most

commercial properties in Anne Arundel County were developed after the PCB ban. Greenspaces included parklands and vegetated open spaces, often located away from industrial zones. Their low impervious surface cover limits contaminated runoff, and natural vegetation may help filter particulate-bound PCBs and possibly promote microbial degradation of PCBs (Jartun et al., 2008).

While statistical comparisons in this study were based on median values to account for the non-normal data distribution, mean concentrations can provide additional insight into the overall burden and variability of PCB contamination across land use types. Particularly, the industrial land use category showed a notable difference between the mean (98.8 ng/g) and median (69.8 ng/g), indicating the presence of one or more samples with especially high PCB concentrations, such as the sample from an outdoor scrap yard (ID-Pre70-1), which reflected a localized source of intense contamination. A similar pattern was observed in residential areas (mean = 55.2 ng/g; median = 35.6 ng/g), likely due to legacy PCB inputs from older infrastructure in select neighborhoods.

In contrast, land use categories with generally lower contamination, such as greenspace (mean = 10.1 ng/g; median = 11.3 ng/g), institutional areas (mean = 15.1 ng/g; median = 11.7 ng/g) and commercial areas (mean= 18.8 ng/g, median=16.9 ng/g), showed much smaller differences between mean and median values. This suggests that PCB concentrations in these categories were more uniform. Comparison between mean and median values helps to illustrate the distribution of contamination within each land use, identifying where PCB hotspots may be skewing the average and where contamination was more evenly distributed.

Overall, the findings showed that land use intensity contributes to PCB accumulation in stormwater sediments and soils. Industrial and residential area had higher levels of total PCB concentration.

3.2.2 Effect of land development era

This study further stratified sampling sites not only by land use but also by the era of development, specifically in pre- and post-1970s matching the PCB ban in the United States in 1979 (US EPA, 2021). This grouping allowed a more targeted examination of whether development timing, relative to the regulatory ban, affected environmental PCB burdens. Both median and mean values are reported for descriptive purposes and to compare with other studies (Table 3.4). Median reduction is calculated by:

$$\text{Median Reduction (\%)} = \frac{(\text{Median Pre} - \text{Median Po})}{\text{Median Pre}} * 100\%$$

Table 3.4. Concentrations of Total PCB (ng/g) by land use and development (Pre-/Post-1970s).

Land Use	Land Development	Mean	S.D.	Range		Median	Median Reduction (%)
				Min.	Max.		
Res	Pre (n= 5)	79.8	56.2	157	17.4	71.0	56.9
	Po (n= 5)	30.6	5.23	37.4	24.1	30.6	
ID	Pre (n=4)	139	163	381	31.7	70.8	16.7
	Po (n= 5)	67.0	28.1	107	32.0	59.0	
Com	Pre (n=1)	24.9	n.a	24.9	24.9	24.9	41.0
	Po (n=4)	17.3	7.30	27.7	12.1	14.7	
IN	Pre (n=2)	18.5	19.0	31.9	5.05	18.5	58.1
	Po (n=2)	11.7	2.14	13.2	10.2	11.7	
GS	n.a (n=5)	10.1	5.47	15.7	1.27	11.3	n.a
Energy	n.a (n=1)	64.4	n.a	64.4	64.4	64.4	n.a

Across residential, industrial, commercial, and institutional land uses, sites developed after the 1970s consistently showed lower mean and median PCB concentrations (Table 3.4). This trend is consistent with the U.S. ban on the production of PCBs in 1979 (US EPA, 2021), suggesting that the post-ban development has reduced inputs of legacy PCBs into the environment.

However, despite these reductions, no statistically significant differences ($p > 0.05$) between pre- and post-1970 groups within each land use type (Figure 3.5) were observed. This result is likely due to a combination of factors, including small sample sizes, high within-group variability, and overlapping concentration ranges. Nevertheless, the observed decreases across all categories provide supporting evidence for the influence of development era on PCB accumulation in stormwater sediments, reinforced the importance of land use history in understanding contaminant distribution.

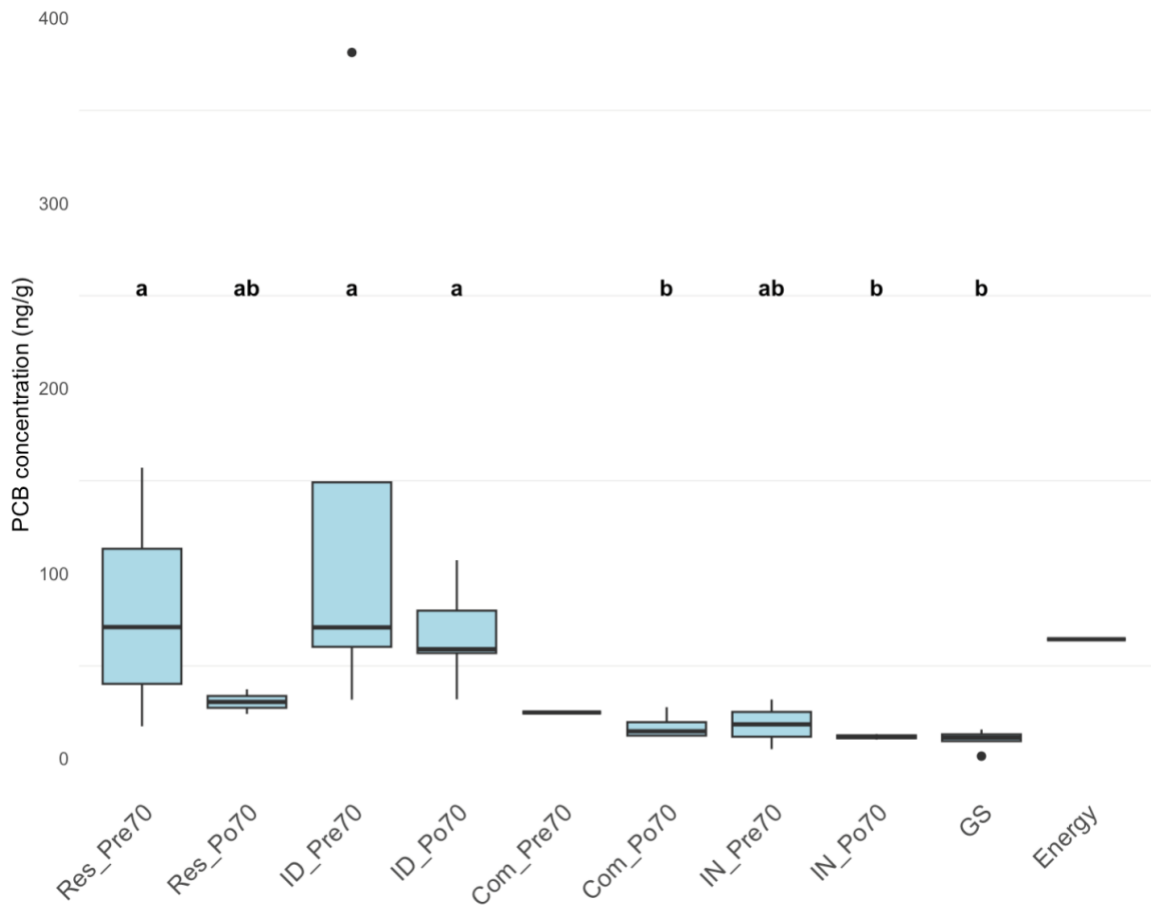


Figure 3.5. Comparison of total PCB concentrations across land use types developed before and after 1970s.

Notes: The category without the letter means there is only one sample.

These patterns are likely explained by differences in historical building materials and infrastructure. Pre-1970s developments were more likely to incorporate materials containing Aroclor mixtures, such as sealants, caulks, transformers, and hydraulic systems, which were commonly used in construction prior to regulatory restrictions. Diamond et al., (2010) reported mean PCB concentrations of 4,630 mg/kg in sealants collected from 80 buildings constructed between 1945 and 1980 in Toronto, Canada. The study further found that residential buildings

constructed between 1950 and 1970 had particularly elevated levels of PCBs in building materials, suggested potential contribution of older buildings to environmental PCB loads via stormwater runoff. In contrast, post 1970s developments may benefit from non-PCB construction materials. Robson et al. (2010) collected 21 building sealant sample from buildings built after 1980 in Toronto, Canada, and none of the samples had detectable PCBs ($< 40 \mu\text{g/g} \Sigma 83\text{PCBs}$).

In many urban areas, sites undergo redevelopment which make it difficult to determine the exact age of the current infrastructure and thus the history of PCB exposure. A site classified as post 1970s may still retain old subsurface soils or sediments, which introduce the possibility of old soils and sediments with contamination inherited from earlier land uses or undocumented material reuse (US EPA, 2011). Weathering and stormwater runoff from such materials can mobilize PCBs into surrounding soils and sediments, particularly in older developments lacking remediation or renovation. Additionally, atmospheric deposition and watershed scale transport can input PCBs from off-site sources (Leister & Baker, 1994). Greenspace areas usually had fewer direct industrial sources but could still accumulate PCBs through urban runoff and atmospheric dry deposition (Holsen et al., 1991). Therefore, even sites without obvious sources such as greenspaces may still retain and can accumulate PCBs due to legacy contamination, atmospheric inputs, and watershed transport.

Overall, despite the lack of statistical significance, the clear and consistent decline in PCB concentrations across all land use types indicates that areas developed after the 1970s were less impacted by legacy PCB sources, highlighting the long-term influence of historical land development practices on current environmental contamination levels.

3.3 Evaluation of homologue profiles

PCBs consist of 209 congeners, which can be grouped into homologues based on the number of chlorine atoms per biphenyl molecule (mono-CBs, di-CBs, tri-CBs, tetra-CBs, penta-CBs, hexa-CBs, octa-CBs, nona-CBs, and deca-CBs). Analyzing homologue profiles can be helpful in revealing patterns of PCB sources since different Aroclor mixtures produce distinct distributions of homologue groups. In this section, we assessed the homologue profiles of environmental samples across land use and development categories to evaluate potential source signatures.

To explore correlation between environmental PCB signatures and known commercial mixtures, the Spearman rank correlation analysis was conducted to evaluate the correlation between the homologue profiles of environmental sample categories and commercial Aroclor mixtures (Figure 3.6). None of the categories showed fully the same homologue patterns originating from Aroclor mixtures, but some showed partial similarity.

Among all site categories, pre-1970 industrial sites (ID_Pre70) exhibited the strongest positive correlations with Aroclor mixtures, particularly A1254 ($\rho = 0.73$, $p < 0.05$), A1248 ($\rho = 0.64$, $p < 0.05$). Similarly, pre-1970 residential areas (Res_Pre70) also demonstrated correlations with A1254 ($\rho = 0.71$, $p < 0.05$). These associations suggest that some samples, particularly from older residential and industrial locations, may still retain partial PCB signatures reflective of historical Aroclor usage, consistent with the widespread use of A1254 in sealants, dielectric fluids, hydraulic systems, and machinery lubricants prior to the ban in the late 1970s (ATSDR, 2002).

In contrast, post-1970 sites across all land use types, including industry, residential commercial and institutional area (ID_Po70, Res_Po70, Com_Po70, and IN_Po70) showed weak

correlations ($\rho < 0.46$) and lacked statistical significance ($p > 0.05$). This trend suggests that more recent developments are either less impacted by legacy PCB mixtures or that their congener patterns have changed from historical Aroclor profiles due to environmental weathering, degradation, or more varied PCB sources (road paint, pigments) (ATSDR, 2002; Hu & Hornbuckle, 2010).

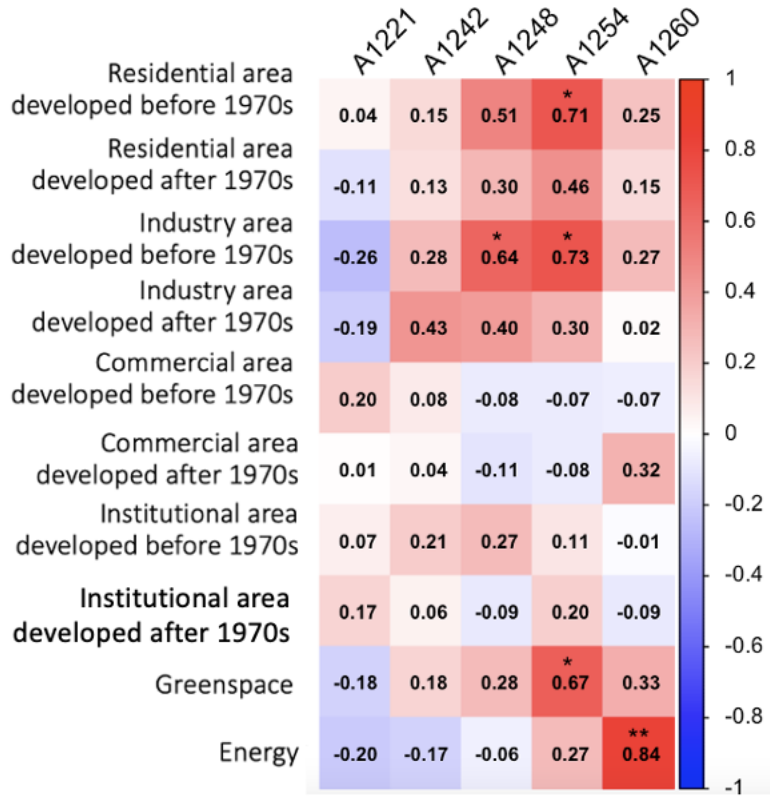


Figure 3.6. Spearman correlation coefficients between homologue distribution of soil and stormwater sediment from different categories and Aroclors.

red = positive, blue = negative; significant level: * < 0.05 ; ** < 0.01 ; *** < 0.001 .

Notes: Commercial area developed before 1970s and Energy only has one sample.

Greenspace (GS) areas exhibited a statistically significant correlation with Aroclor 1254 ($\rho = 0.67, p < 0.05$), suggesting some degree of similarity in homologue distribution. While the

total PCB concentrations in greenspace samples were observed low compared with residential and industrial area, this correlation may reflect trace legacy inputs.

The energy site was associated with highly chlorinated Aroclor mixtures, where significant correlations were observed with A1260 ($\rho = 0.84$, $p < 0.01$). Aroclor 1260 was commonly used in transformers, capacitors, and other closed-system electrical applications (ATSDR, 2000), and the observed correlation may reflect either direct usage or residual contamination associated with energy infrastructure at that location.

Commercial and institutional sites (both pre- and post-1970) displayed low or negative correlations with all Aroclor mixtures ($\rho < 0.1$ in most cases), with no statistically significant relationships detected. This suggests either minimal PCB contamination, highly weathered or degraded profiles, or more heterogeneous sources not well-represented by Aroclor mixtures. The weak correlations may also reflect diverse inputs such as non-Aroclor PCB-11 associated with pigment manufacturing and consumer goods such as newspaper and packaging (Washington State Department of Ecology, 2015). These findings highlight the complexity of PCB sources in urbanized environments where diffuse, modern, and legacy contributions may co-occur.

To evaluate compositional similarities between environmental samples and known Aroclor mixtures, a non-metric multidimensional scaling (NMDS) ordination was used based on Bray–Curtis similarity scores of PCB homologue distributions (Figure 3.7). Bray-Curtis similarity scores are summarized in Table C.2.

In the NMDS plot, the Aroclor mixtures and samples are positioned according to their known homologue compositions. This spatial arrangement serves as a reference framework to evaluate the similarity of environmental samples to commercial PCB formulations.

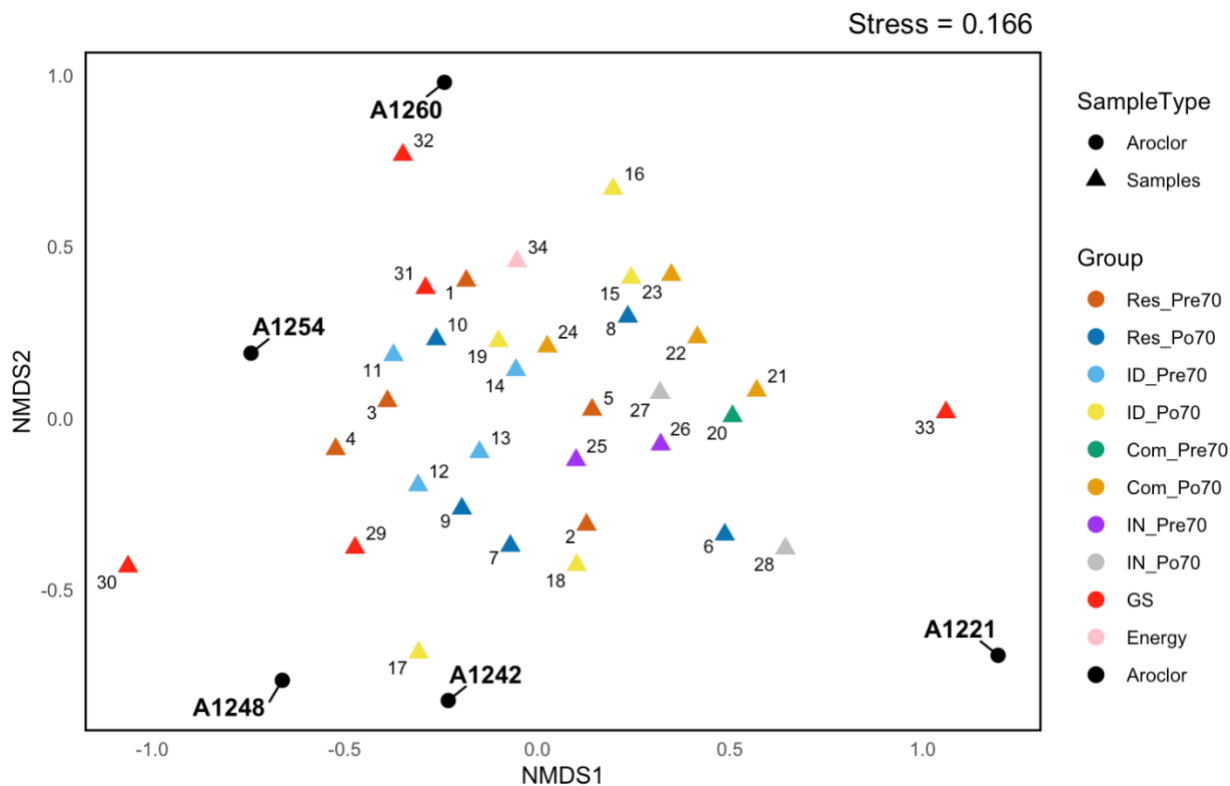


Figure 3.7. NMDS plot of each sample and Aroclor mixtures.

Note: The number of each sample corresponds to the sample name in Table C.2.

None of the environmental samples fully overlapped with the Aroclor mixtures in the similarity analysis. However, several samples showed high similarity scores ($\rho > 0.70$), indicating comparable homologue distribution patterns. Specifically, one sample (Sample 17, ID-Po70-3) from an industry area developed after the 1970s, exhibited strong similarity (0.90) with Aroclor 1242. Two samples from residential areas (Sample 3 (Res-Pre70-3), Sample 4 (Res-Pre70-4)) developed before the 1970s, and one sample from an industry area (Sample 11 (ID-Pre70-1)) developed before the 1970s had high similarity scores (0.77, 0.74, 0.73, respectively) with A1254. Additionally, the sample from the energy site (Sample 34 (Energy-1)) showed high similarity ($\rho = 0.70$) with Aroclor 1260. These high similarity values suggest that the congener patterns in these samples partially resemble those of specific Aroclor formulations historically

used in industrial and commercial applications, and indicate possible historic contamination from capacitor, sealants, cutting oils, transformers, hydraulic fluids, dielectric fluids (ATSDR, 2002).

This analysis was based on pattern similarity rather than the total PCB mass. As a result, even samples with relatively low total PCB concentration, such as one greenspace sample (Sample 29, Rec-1) with total concentration 11.3 ng/g, could display a high similarity score to certain Aroclor patterns ($\rho = 0.70$ with Aroclor 1248). This approach allowed for the detection of legacy source signatures even in areas with minimal contamination and helped identify historical inputs regardless of their current environmental concentration.

In summary, the data indicated that single Aroclor mixtures were not the only source of PCBs in the collected soil and sediment samples. While some samples showed compositional similarity to Aroclor mixtures in the homologue profiles, the overall limited overlap with Aroclor reference points across the NMDS plot (Figure 3.7) suggest the presence of alternative PCB sources.

Some environmental samples might represent mixtures of multiple Aroclor products. In real world applications, especially in older infrastructure and equipment, multiple Aroclors may have been used together. For instance, A1254, and A1260 were all used in transformers (ATSDR, 2002). This resulted blended congener profiles that no longer match any single commercial Aroclor.

Beside the mixture of multiple Aroclor products, the influence of non-Aroclor PCBs could impact the homologue distribution pattern, such as PCB-11 (di-PCB), a known byproduct of pigment and dye manufacturing. PCB-11 has been detected in various urban environments and was not present in any Aroclor mixture, which could significantly alter the homologue distribution when presented (Cao et al., 2019; Hu & Hornbuckle, 2010). Its presence may

indicate modern inputs which unrelated to Aroclor PCB uses. Its presence can alter homologue patterns, especially by increasing the relative abundance of di-chlorinated congeners. Figure 3.8. showed the Spearman rank correlation between environmental PCB signatures and known commercial mixtures after remove mono- and di PCBs. None of the categories still showed fully the same homologue patterns originating from Aroclor mixtures. Compared with the correlation with mono- and di- PCBs, significantly stronger positive correlations were showed after remove mono- and di- PCBs in residential area developed before ($\rho = 0.85, p < 0.01$) and after 1970s ($\rho = 0.81, p < 0.05$), industry area developed before 1970s ($\rho = 0.75, p < 0.05$), institutional area developed after 1970s ($\rho = 0.91, p < 0.01$) and greenspace ($\rho = 0.77, p < 0.01$) with A1254. The energy site also showed significantly stronger positive correlations with A1260 ($\rho = 0.92, p < 0.01$) Aroclor 1260. These data suggested that samples from those categories may better reflect legacy Aroclor signatures in the absence of modern, non-Aroclor influences.

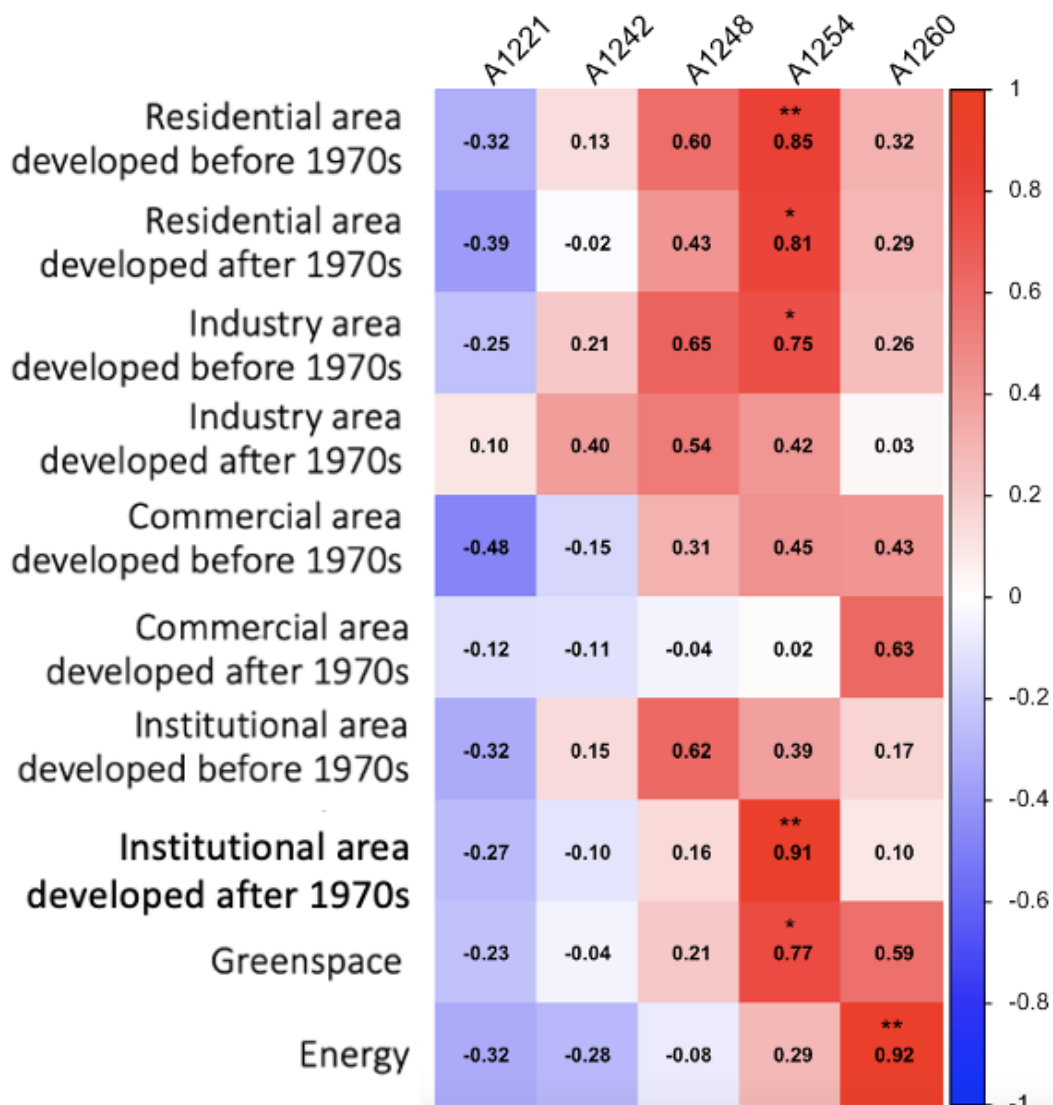


Figure 3.8. Spearman correlation coefficients between homologue distribution of soil and stormwater sediment from different categories and Aroclors after remove mono- and di PCBs.

red = positive, blue = negative; significant level: * < 0.05; ** < 0.01; *** < 0.001.

Furthermore, environmental weathering and degradation processes such as anaerobic and aerobic PCB biodegradation could transform PCB profiles. These processes tend to gradually changed the original profile from Aroclor standard mixtures (Abramowicz, 1995; Passatore et al., 2014). Such transformation is likely in soils and sediments with long term exposure to redox

conditions. For example, anaerobic dichlorination tends to remove chlorine atoms, resulting in the accumulation of lower chlorinated PCB congeners ($Cl < 4$). For example, PCB 2, PCB 4, PCB 19, and PCB 47, PCB 51 are frequently used as indicators of microbial dechlorination activity (Brown et al., 1987; Fagervold et al., 2007).

These findings suggest that the observed congener profiles likely reflect a combination of additional non-Aroclor PCB sources, mixture of multiple Aroclor mixtures and in situ microbial degradation of legacy Aroclors, rather than direct input from a single commercial mixture.

3.4 Evaluation of chlorines per biphenyl among different sites

The average chlorine per biphenyl value serves as an indicator of the chlorination level of PCBs present in a sample. Understanding the chlorine per biphenyl molecule provides insight into the potential sources, and degradation pathways of PCBs at different sites. PCBs with higher degrees of chlorination ($Cl > 4$) are more susceptible to anaerobic reductive dichlorination, while lower chlorinated PCBs ($Cl \leq 4$) tend to undergo aerobic degradation (Abramowicz, 1995). The lower chlorinated PCBs are produced as intermediates or end-products of anaerobic dichlorination and serve as substrates for aerobic biodegradation (Passatore et al., 2014). Some commercial Aroclor mixtures also contain fractions of lower chlorinated PCB congeners (ATSDR, 2002).

The number of chlorines per biphenyl varied among samples and across land use categories (Figure 3.8, Table C.3), ranging from 2.48 to 6.11. Among the samples, one sample from the greenspace category (Rec-4) exhibited the highest number of chlorines per biphenyl (6.11 ± 0.02) despite having the lowest total concentration of PCBs. In total, only 12 out of 34 samples had chlorines per biphenyl values < 4 , indicating the presence of more recalcitrant

PCBs. The lowest chlorines per biphenyl were found at one greenspace sample (Rec-5) due to the dominance of di-chlorinated PCBs (83.8 % of total PCBs by mass) at this site.

Overall, the chlorination patterns observed in this study suggest that highly chlorinated PCB congeners ($Cl > 4$) dominate across most sampling sites (22 out of 34 sites), in agreement with Cao et al (2019) that 12 out of 16 urban sediment samples had mean chlorine per biphenyl values above 4. This indicates that most sites would require anaerobic conditions to initiate reductive dechlorination, followed by aerobic conditions to further break down the resulting lighter congeners. Enhancing such sequential anaerobic–aerobic environments in sediments could promote more effective natural biodegradation of PCBs over time.



Figure 3.9. Chlorines per biphenyl of all samples.

Note: error bars represent the standard deviation between the replicate

3.5 Presence and concentration of non-legacy PCB 11

Non-legacy PCBs represent a group of PCBs that were Aroclor constituents but may be present in other products such as road paint, newspaper, food packaging (Hu & Hornbuckle, 2010; Rodenburg et al., 2010). Among these, PCB 11 is a commonly reported non-Aroclor PCB

congener and was frequently detected (29 out of 34) in this study. The concentration and relative abundance of PCB11 varied among samples (Figure. 3.9). The PCB11 concentration was below the MDL (0.275 ng/g) in all samples from green spaces indicating limited inputs from less undeveloped areas. However, the possibility of nearby developed land contributing via atmospheric deposition or surface runoff could not be excluded. In contrast, PCB-11 was detected in all sites associated with residential, industrial, institutional, and commercial land uses, with variable concentrations and relative abundances (Table 3.7). One sample collected from a residential area developed before 1970s (Res-Pre70-5) had the highest PCB 11 concentration (15.6 ± 2.19 ng/g) among all samples, but the relative mass abundance of PCB 11 was 9.89%. This suggests that the site was contaminated with a mix of PCB sources, including legacy Aroclors, which diluted the proportional contribution of PCB 11 in the total PCB mass. One sample from a residential area developed before the 1970s (Res-Po70-1) and one sample from an institutional area developed after the 1970s (IN-Po70-2) had the highest relative abundance (37.1%) but with concentrations of 10.1 ± 1.81 and 3.83 ± 1.15 ng/g, respectively. These high proportions, but lower concentrations, suggest minimal presence of legacy Aroclor PCBs and point to non-Aroclor sources such as pigment-related inputs being dominant at these sites.

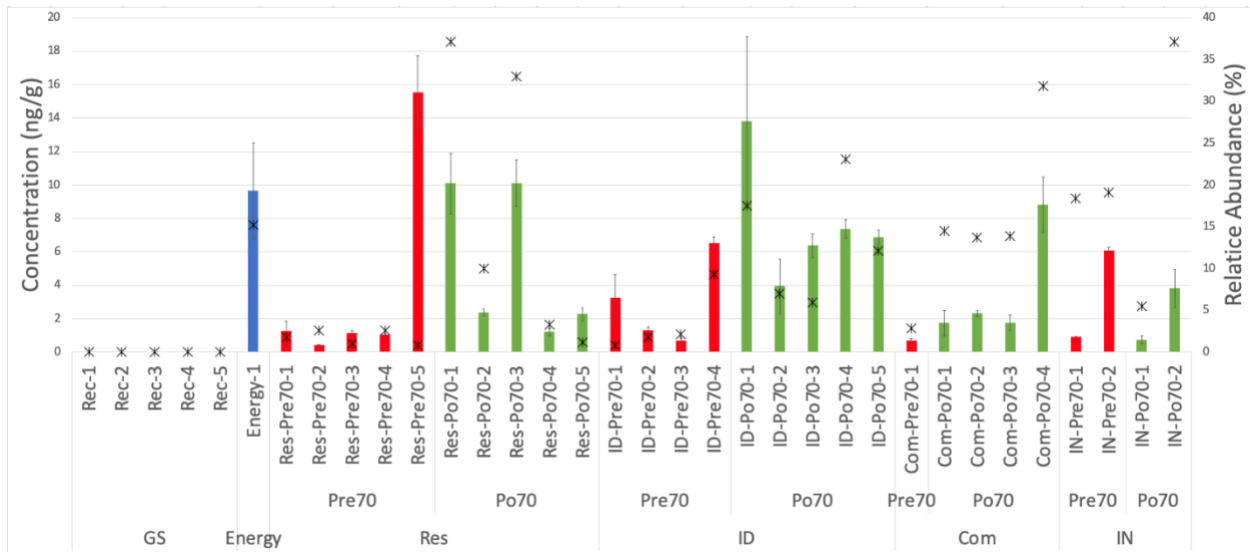


Figure 3.10. PCB 11 concentration (bar (left axis)) and mass relative abundance (x (right axis)).

Table 3.5. Description of PCB 11 concentration and mass relative abundance in residential, industrial, institutional, and commercial land use.

	Mean	S.D.	Range		Median
			Min.	Max.	
PCB 11 concentration (ng/g)	4.56	4.22	0.44	15.6	2.38
PCB 11 mass relative abundance (%)	11.9	11.3	0.85	37.1	9.33

PCB-11 is a non-Aroclor congener and has been frequently detected in urban environments as a by-product of pigment manufacturing, particularly from diarylide yellow pigments (Grossman, 2013) that was used in road paint, traffic markers, bike and bus lanes (City of Spokane Wastewater Management Department, 2015). The influence of pigment-based sources was further supported by visual evidence of yellow paint flakes in some sediment samples. Previous work by Cao et al. (2019) found that the presence of yellow pigment flakes from traffic paint could elevate PCB 11 concentrations by as much as 42%, highlighted the

importance of these physical particulates as a direct input pathway. These findings suggested that targeting sources such as roadway paint and pigment-containing runoff may be effective in reducing PCB-11 loads in stormwater systems.

3.6 Toxicity

PCBs are toxic environmental contaminants known for their persistence, bioaccumulation potential, and adverse health effects (ATSDR, 2002). To guide remediation efforts, the U.S. Environmental Protection Agency (US EPA, 2005) established that action is required if total PCBs in soil exceed 1 mg/kg (1000 ng/g). In the present study, none of the measured soil or sediment samples exceeded this regulatory threshold.

However, due to the higher levels of health risks, dioxin-like PCBs (DL-PCBs) are of specific concern. Among the 209 PCB congeners, twelve are classified as DL-PCBs due to their structural similarity and toxicological behavior akin to 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD) (Van den Berg et al., 1998). The World Health Organization (WHO) established Toxic Equivalency Factors (TEFs) for each DL-PCB (Van den Berg et al., 2006), which are used to calculate Toxic Equivalency (TEQ) values by multiplying the mass concentration of twelve DL-PCBs with the respective TEF. The TEQ metrics enables the US EPA to account for the toxicity of dioxin and dioxin-like compounds (US EPA, 2015a).

In this study, TEQ values for stormwater sediment samples ranged from < 0.0004 to 41.2 ± 2.02 pg TEQ/g (Table 3.5). Toxic equivalency value (TEQ pg/g) for all samples showed in Table C.4. The maximum TEQ_{PCB} concentrations observed in this study value exceeded several reported ranges from comparable soil and sediment environments, such as Eljarrat et al. (2001) in sediments from the Northwest Mediterranean (range: 0.03 and 24.8 pg TEQ/g), and Cao et al. (2019) in urban soils and sediments (range: 1.48 to 18.0 pg TEQ/g) in Maryland, USA (Table

3.6). A recent study by T. Li et al. (2023) in a comprehensive industrial area in China reported lower values, while (Lacomba et al., 2025) documented contamination in Colombian industrial soils, with TEQ_{PCB} values as high as 1612 pg/g. These comparisons highlight the variable nature of dioxin-like PCB contamination across different regions and land use types and emphasize that Anne Arundel County may still have elevated toxicological risks relative to other urban environments worldwide.

Table 3.6. TEQ of DL-PCBs in the soil and stormwater sediment from Anne Arundel County.

TEQ_{PCB} (pg TEQ/g)				
Sample	Mean	S.D.	Range	
			Min.	Max.
Total (n=34)	4.88	11.3	< 0.0004	41.2
	Samples exceeding the quality guideline threshold (20 pg TEQ/g)			
ID-Pre70-1	41.2	2.02	n.a	
ID-Pre70-4	20.9	0.95	n.a	
Res-Pre70-3	26.3	3.69	n.a	

Table 3.7. Comparisons of DL-PCBs (pg TEQ/g) in soil and sediments from different regions.

Location	Range		Reference
	Min.	Max.	
Northwest Mediterranean Sediments, Spain	0.03	24.8	(Eljarrat et al., 2001)
Urban soil and sediments in Maryland State, U.S.	1.48	18.0	(Cao et al., 2019)
Comprehensive industrial area soil, China	0.004	0.27	(T. Li et al., 2023)
Industrial area soil, Colombia	0.03	1612	(Lacomba et al., 2025)

All samples except two collected from an industrial area developed before the 1970s (ID-Pre70-1, ID-Pre70-4), and one sample from a residential area developed before the 1970s (Res-Pre70-3) were below the sediment quality guideline threshold of 20 pg TEQ/g (Eljarrat et al., 2001) (Table 3.7). The highest TEQ value was recorded at ID-Pre70-1, which also had the highest total PCB concentration in the dataset (381.3 ± 13.7 ng/g). Industry area developed before 1970s showed significant higher TEQ than industry area developed after 1970s, commercial area developed before 1970s and greenspace.

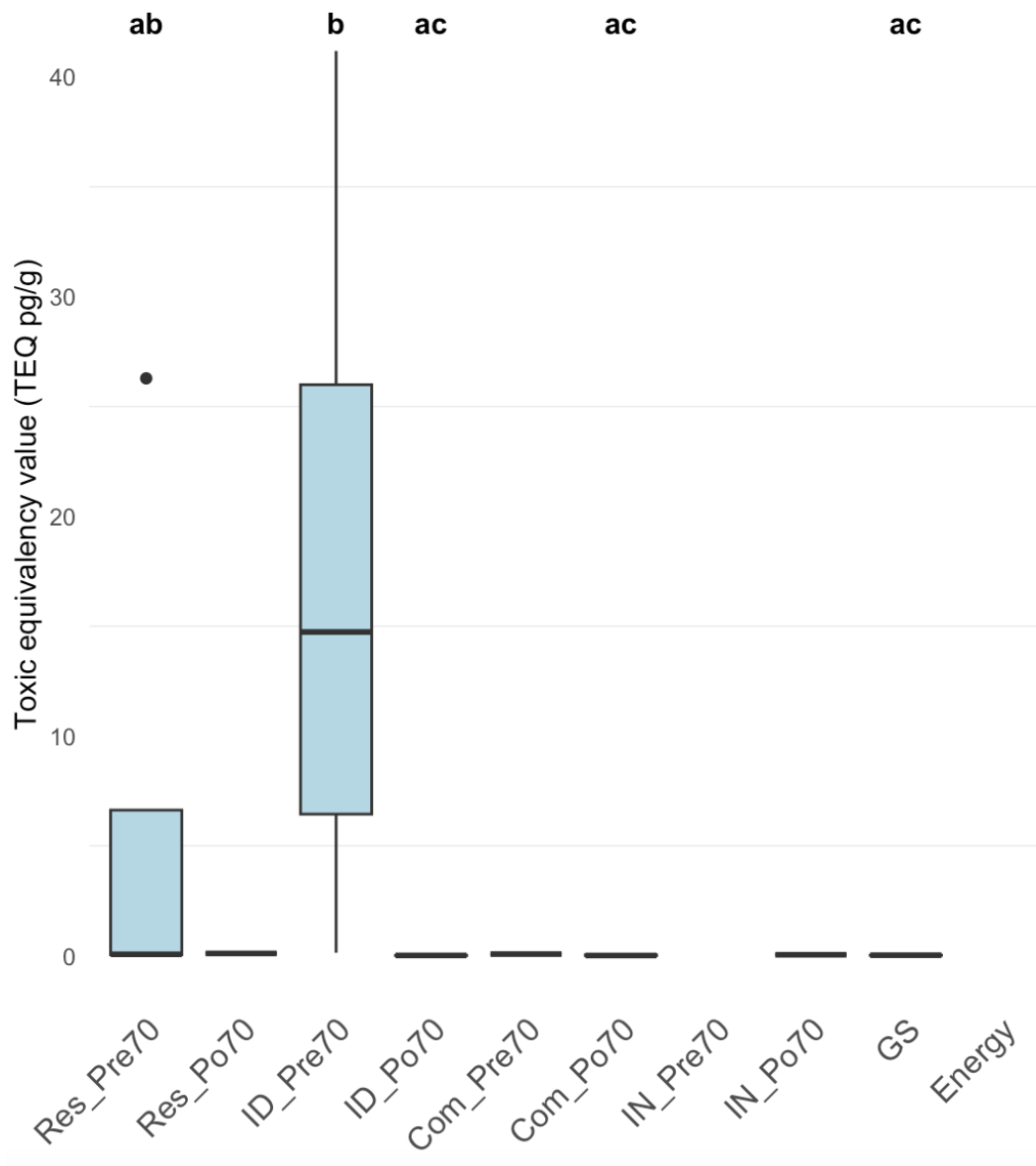


Figure 3.11. Comparison of toxic equivalency value across land use types developed before and after 1970s.

Notes: The category without the letter means there is less than two samples have DL-PCBs detected.

This analysis again highlights the need for targeted remediation and source control at select industrial and older residential locations. While DL-PCBs are a critical indicator of toxicity, they represent only one component of overall sediment toxicity. Eljarrat et al. (2001) noted that PCBs were found to account for 1% to 84% of the total TEQs in marine sediments. Therefore, additional analyses of other dioxin-like compounds, such as polychlorinated dibenzodioxins (PCDDs) and dibenzofurans (PCDFs), would provide a more comprehensive risk profile for the stormwater sediments.

3.7 Stormwater Sediment Removal as a Best Management Practice

Street sweeping is a cost-effective and proactive Best Management Practice (BMP) for controlling the accumulation and mobilization of contaminants in urban stormwater systems (US EPA, 2021). It removes fine sediments and particulate-bound pollutants directly from impervious surfaces before they would be transported into stormwater conveyance systems and discharged into waters.

Anne Arundel County, MD, collected an average of 234 US ton of street sediment in 2024 through regular sweeping operations across 6,685 road miles (AA County, 2024). To estimate the actual mass of dry sediment, a correction based on average moisture content percent in this study of 11.9% would be used. The dry mass was computed as:

$$\begin{aligned} \text{Dry Mass} &= \text{Sediment collection} * (1 - \text{Moisture Content}) \\ &= 234 \text{ US ton} * (1 - 0.119) = 206 \text{ US ton} = 186,880 \text{ kg} \end{aligned}$$

Thus, approximately 186,880 kg of dry sediment were removed from road surfaces via street sweeping in 2024 in Anne Arundel County.

To estimate potential PCB load removal, maximum, minimum, mean and median of total PCB concentrations from soil and sediment samples collected across the study area were used.

The total mass of PCBs removed is calculated as:

$$Total\ PCB\ removed\ (g) = Dry\ Mass\ (kg) * PCB\ concentration\ \left(\frac{ng}{g}\right) * \frac{1g * g}{10^6\ kg * ng}$$

This estimated annual load of PCBs potentially removed is shown in Table 3.8.

Table 3.8. Estimated annual load of PCBs potentially removed in Anne Arundel County in 2024.

	Min	Max	Mean	Median
PCB concentration (ng/g)	1.27	381	50.3	31.2
PCB collected (g/year)	0.237	71.2	9.40	0.583

This estimated value represented the annual load of PCBs potentially intercepted and removed before these contaminants could enter the storm drain and ultimately impair downstream aquatic ecosystems.

Chapter 4: Conclusions

4.1 Conclusions and Recommendations.

This study investigated the occurrence, distribution, and potential sources of PCBs in stormwater sediments and soils across 34 samples representing multiple land use categories and development eras within Anne Arundel County, Maryland. Sampling sites were selected to reflect a range of urban conditions, including industrial, residential, commercial, institutional, greenspace, and energy areas, with both pre-1970s and post-1970s developments (except greenspace and energy area) examined to assess the influence of historical versus modern infrastructure. The goal of this research was to identify how land use characteristics and development history influenced PCB accumulation in urban environments, to determine potential PCB sources based on homologue profiles, and to evaluate the potential for targeted management strategies to reduce PCB loads to local watersheds. The findings from this study include:

- **Influence of Land Use on PCB Accumulation.**

Land use type was found to significantly influence total PCB concentrations in the sediment samples. Industrial (range: 31.7 to 381 ng/g, median: 69.8 ng/g, mean: 98.8 ng/g) and residential areas (range: 17.4 to 157 ng/g, median: 35.6 ng/g, mean: 55.2 ng/g) exhibited the highest levels of total PCBs, indicating that urban intensity and associated sources (building sealants, industrial pollutants, etc.) contributed to the accumulation of PCBs in stormwater sediments and soils. In contrast, commercial (range: 12.2 to 27.2 ng/g, median: 16.9 ng/g, mean: 18.8 ng/g), institutional (range: 5.05 to 31.9 ng/g, median: 11.7 ng/g, mean: 15.1 ng/g), and greenspace (range: 1.27 to 15.7 ng/g, median: 11.3 ng/g, mean: 10.1 ng/g) sites exhibited comparatively lower

concentrations, indicating reduced historical loading, and limited ongoing sources.

Overall, the spatial pattern of PCB occurrence aligned closely with land use intensity, emphasizing the importance of prioritizing source control in heavily developed catchments.

- **Effect of Land Development Era (Pre- vs. Post-1970s)**

While statistical comparisons between pre- and post-1970s sites under the same land use did not show any statistically significant differences, mean and median values indicated higher PCB concentrations in pre-1970s areas. This trend supported the legacy hypothesis that older urban infrastructure and historical industrial practices were an essential contributor to ongoing PCB presence in the environment. The persistence of PCBs in post-1970s sites suggest that contemporary inputs continue to influence stormwater sediments and that Aroclor PCBs are still present in the environment.

- **Homologue Distribution and Source Characterization.**

Homologue profile analyses and comparisons with commercial Aroclor mixtures showed that none of the single Aroclor mixtures was the direct source of PCBs in the sediment samples. Partial similarities between some samples and Aroclor 1254, and 1260 indicated potential, but not exclusive, legacy sources. Notably, PCB 11 which was a non-Aroclor PCBs was frequently detected in all samples except greenspace. The presence of PCB11 is expected to be related to yellow paint used in road paint and other sources such as newspaper and food packaging.

- **Source tracking and Targeted Sediment Removal.**

The observed spatial trends indicate that industrial and residential areas act as persistent PCB hotspots. Targeted removal of stormwater sediment and roadside soil from these areas could reduce PCB mass loadings to the stormwater network. Such management actions would be especially beneficial where sediment accumulation occurs near storm drains and outfalls. Due to the persistence and particle-bound nature of PCBs, selective sediment removal could be a practical and impactful mitigation method.

- **Effect of Street Sweeping on PCB Load Reduction**

Street sweeping provides a quantifiable methodology to intercept PCB contaminated particles before they enter the storm drain system and was evaluated as a practical strategy to reduce PCB transport via stormwater. The potential PCB load removal ranged from 0.237 to 71.2 g/year with 9.40 g per year on average across 6,685 road miles. Strategically targeted and frequent sweeping in residential, industrialized, and high-traffic areas could make a meaningful contribution toward PCB TMDL compliance.

4.2. Limitations and Future Work.

Although this study provides valuable insights into the occurrence, distribution, and potential sources of PCBs in stormwater sediments and soils across different land uses and land development eras, several limitations should be acknowledged, and future studies would be needed in the future:

- **Lack of Sample Size.**

While the study included different land use and development categories, the number of samples per category was limited, especially when comparing pre- and post-1970s development within each land use type. This reduced statistical power and made it difficult to detect significant differences and trends in PCB concentrations.

- **Limited Physicochemical Parameters.**

Only four physicochemical properties were measured. Additional properties such as total organic carbon (TOC) and particle size distribution may provide better insights into the PCB concentrations in soil and stormwater sediments in which TOC could directly provide measure of sorption potential, while particle size could indicate transport behavior and settling characteristics, offering deeper insight into how PCBs are retained or mobilized within sediments.

- **Limited Temporal Dynamics.**

This study collected soil and stormwater sediment sample at a single point in time and could only detect the PCB contamination at that time. However, PCB transport and deposition in urban environments could be influenced by storm frequency and intensity, and maintenance methods, such as street sweeping. A longitudinal sampling approach that crossed multiple seasons and storm events may better characterize temporal variability, accumulation patterns, and pollutant fluxes to enable better source tracking and load estimation over time.

- **Non- Aroclor PCB (PCB 11) Investigations.**

A further and deeper exploration of the origins and pathways of PCB 11 in urban environments would support the suggestion that road paint pigments be regulated as a potential nonpoint PCB source.

- **Street Sweeping Study.**

The estimation of effect of Street Sweeping on PCB Load Reduction in Anne Arundel County each year is just a first approximation. In addition to sweeping frequency and land use type, site-specific environmental conditions such as soil texture, and local climate (rainfall intensity, seasonal patterns) might influence the mobilization and accumulation of PCBs on impervious surfaces and in street sediments. Therefore, future street sweeping study should consider these factors to optimize PCB removal efficiency under varying hydrological and environmental settings. A further study will be needed.

Appendices

APPENDIX A. Sampling Information

Table A.1. Abbreviation of sample name

Abbreviation	Definition
Res	Residential
ID	Industry
Com	Commercial
IN	Institutional
GS	Greenspace
Pre70	Developed before 1970s (before PCB banned)
Post70/Po70	Developed after 1970s (after PCB banned)

Table A.2. Summary of the sampling sites and information

Land Use	Land Development	Sample Name	GPS Coordinate	Sampling Time
Residential	Pre 1970	Res-Pre70-1	39.152957, -76.610989	Aug 2024
		Res-Pre70-2	39.15304, - 76.61084	Aug 2024
		Res-Pre70-3	39.133965, -76.616717	Aug 2024
		Res-Pre70-4	39.133899, -76.616044	Aug 2024
		Res-Pre70-5	39.183506, -76.625590	July 2023
	Post 1970	Res-Po70-1	39.144248, -76.673505	July 2023
		Res-Po70-2	39.144313, -76.673479	July 2023

		Res-Po70-3	39.145563, -76.671205	July 2023
		Res-Po70-4	39.134751, -76.607512	Aug 2024
		Res-Po70-5	39.131056, -76.605688	Aug 2024
Commercial	Pre 1970	Com-Pre70-1	39.141907, -76.630735	Sep 2024
	Post 1970	Com-Po70-1	39.175537, -76.643630	Oct 2023
		Com-Po70-2	39.203438, -76.616005	Sep 2024
		Com-Po70-3	39.141759, -76.605755	Sep 2024
		Com-Po70-4	39.204303, -76.615809	Sep 2024
Institutional	Pre 1970	IN-Pre70-1	39.139096, -76.624462	Sep 2024
		IN-Pre70-2	39.13914, -76.62446	Sep 2024
	Post 1970	IN-Po70-1	38.990589, -76.532814	Dec 2024
		IN-Po70-2	38.990558, -76.531904	Dec 2024
Industry	Pre 1970	ID-Pre70-1	39.173698, -76.628208	July 2023
		ID-Pre70-2	39.094003, -76.701819	Dec 2024
		ID-Pre70-3	39.09406, - 76.70179	Dec 2024
		ID-Pre70-4	39.182266, -76.614906	Dec 2024
	Post 1970	ID-Po70-1	39.178618, -76.623481	July 2023
		ID-Po70-2	39.178602, -76.623094	July 2023
		ID-Po70-3	39.157144, -76.681454	Dec 2024
		ID-Po70-4	39.156561, -76.681977	Dec 2024
		ID-Po70-5	39.15643, - 76.68197	Dec 2024
Energy	N/A	Energy-1	39.181243, -76.627922	Oct 2023
	N/A	Rec-1	39.197440, -76.668949	Oct 2024

Greenspace	Rec-2	39.198175, -76.667010	Oct 2024
	Rec-3	39.166890, -76.637141	Oct 2024
	Rec-4	39.166038, -76.637905	Oct 2024
	Rec-5	39.151604, -76.664778	Oct 2024



Figure A.1. Examples of sampling point. a). Sampling point for energy site. b). Sampling point for commercial area. c). Sampling point from institutional area. d). Sampling point from residential area. e). Sampling point from greenspace.

APPENDIX B. Experimental Methods

Table B.1. Temperature program for GC running.

Run Time (min)	Rate (°C/min)	Hold Time (min)	Temperature (°C)
0		0	70
15.714	7	0	180
60.714	1	0	225
91.059	5.8	20	285
102.36	11.5	10	300

S1. PCB concentration Method detection limit (MDL) determination.

MDLs for the 209 PCB congeners ranged from 0.0103 to 0.570 ng/g. To determine these MDLs, the instrument detection limit (IDL) for each congener was first established based on an instrument signal-to-noise (S/N) ratio in the range of 3 to 5. Seven replicate spiked samples were then prepared at concentrations approximately 10 times the IDL, along with three method blank samples. All samples were processed through the full extraction, cleanup, and analysis procedure and quantified using gas chromatography (GC).

For each congener, the MDL was calculated using the standard deviation (s) of the seven replicate spiked sample measurements and the single-tailed t-statistic at the 99% confidence level ($\alpha = 0.01$) with $n - 1$ degrees of freedom (in this case, $n = 7$). The equation used for MDL determination was:

$$MDL = \frac{t_{(0.99, n-1)} * s}{sample\ mass}$$

When two congeners were fully resolved, peak areas were determined using the valley-to-valley method. In cases where peaks were partially overlapped, the drop method (Figure B.1) was

used to apportion the shared area between adjacent peaks. If multiple congeners co-eluted as a single, unresolved peak, the total peak area was divided equally among the assigned congeners.

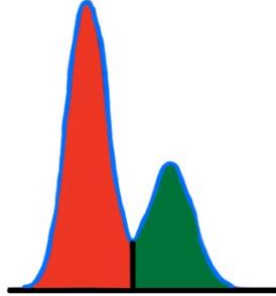


Figure B.1. Drop method for determining the areas under overlapping peaks.

S2. KM method explanation and example calculation.

The concentrations of all congeners, except surrogate standard (PCB30, PCB204) were arranged in descending order. For congeners with values below MDL, the MDL value was assigned. Each congener was then given a ranking (r), where the most concentrated congener received the highest rank (n), and the least concentrated congener received the lowest rank (1). A status (d) was also assigned to each congener with a status of 1 indicated that the congener was above the MDL, while a status of 0 indicated it was below the MDL. Quantiles were determined for each congener using equation (1), and the area of each congener was calculated using equation (2). Finally, the total concentration of the target congener group was obtained by multiplying the total area of the congeners by n using equation (3). Example of calculation is showed in Table B.2.

$$Quantile_n = Quantile_{n+1} * (1 - \frac{d_n}{r_n})$$

$$[\text{For the highest concentration congener, } Quantile_n = 1 - \frac{d_n}{r_n}] \quad (1)$$

$$Area_n = [(1 - Quantile_n) - (1 - Quantile_{n+1})] * C_n$$

$$[\text{For the highest concentration congener, } Area_n = (1 - Quantile_n) * C_n] \quad (2)$$

$$Total\ Concentration = \sum_{n=1}^n Area_n * n \quad (3)$$

Table B.2. Example of KM method using.

PCB congener	PCB concentration (ng/g)	MDL (ng/g)	Concentration (ng/g) <MDLs were substituted by MDL	d	r	d/r	Quantile	1-Quantile	d(1-Quantile)	Area
PCB 79	27.3	0.0143	27.3	1	5	0.200	0.800	0.200	0.200	5.45
PCB 119	17.8	0.0183	17.8	1	4	0.250	0.600	0.400	0.200	3.57
PCB 150	0.000	0.0183	0.0183	0	3	0.000	0.600	0.400	0.000	0.00000
PCB 84	0.000	0.0162	0.0162	0	2	0.000	0.600	0.400	0.000	0.00000
PCB 114	0.000	0.0137	0.0137	0	1	0.000	0.600	0.400	0.000	0.00000

$$Total\ Concentration = \sum_{n=1}^n Area_n * n = 9.02 * 5 = 45.1ng/g$$

APPENDIX C. Additional Results and Supplemental

Table C.1. Soil physicochemical properties for all samples.

Land Use	Land Development	Sample Name	MC (%)	OM (%)	pH	EC ($\mu\text{S}/\text{cm}$)
GS	N/A	Rec-1	3.20 ± 0.40	6.61 ± 0.03	6.48 ± 0.22	84.5 ± 9.79
		Rec-2	15.5 ± 0.23	6.62 ± 0.02	6.74 ± 0.03	454 ± 19.3
		Rec-3	10.9 ± 0.61	8.98 ± 0.51	7.27 ± 0.03	160 ± 9.55
		Rec-4	5.73 ± 0.23	7.78 ± 0.24	7.33 ± 0.03	93.9 ± 5.08
		Rec-5	0.53 ± 0.61	6.57 ± 0.43	7.69 ± 0.06	129 ± 5.18
Energy	N/A	Energy-1	34.1 ± 1.93	6.16 ± 1.00	6.76 ± 0.02	231 ± 18.5
Res	Pre 1970	Res-Pre70-1	< MC% limit	10.4 ± 0.32	7.23 ± 0.12	114 ± 8.24
		Res-Pre70-2	1.46 ± 0.23	15.4 ± 0.16	7.74 ± 0.13	175 ± 8.70
		Res-Pre70-3	< MC% limit	8.15 ± 0.18	7.41 ± 0.07	172 ± 2.52
		Res-Pre70-4	< MC% limit	7.56 ± 0.42	7.55 ± 0.02	95.1 ± 1.73
		Res-Pre70-5	42.3 ± 0.27	32.4 ± 0.83	6.89 ± 0.07	806 ± 20.7
	Post 1970	Res-Po70-1	33.7 ± 0.27	21.2 ± 0.45	6.15 ± 0.07	161 ± 12.6
		Res-Po70-2	11.7 ± 0.30	4.23 ± 0.20	6.43 ± 0.04	62.3 ± 7.45
		Res-Po70-3	12.8 ± 1.23	7.24 ± 0.46	6.55 ± 0.03	102 ± 12.1
		Res-Po70-4	< MC% limit	6.59 ± 2.11	7.63 ± 0.13	138 ± 30.1
		Res-Po70-5	6.58 ± 0.18	14.8 ± 1.76	7.23 ± 0.04	166 ± 4.12
ID	Pre 1970	ID-Pre70-1	5.87 ± 0.62	5.23 ± 0.51	8.84 ± 0.02	111 ± 3.59
		ID-Pre70-2	8.40 ± 0.12	15.2 ± 1.27	8.55 ± 0.04	122 ± 2.72
		ID-Pre70-3	12.2 ± 1.41	12.2 ± 1.27	8.45 ± 0.05	94.9 ± 1.27
		ID-Pre70-4	25.3 ± 1.98	18.1 ± 2.49	7.52 ± 0.06	389 ± 11.3
	Post 1970	ID-Po70-1	17.5 ± 0.80	17.3 ± 0.71	7.37 ± 0.10	373 ± 33.4
		ID-Po70-2	3.56 ± 0.40	5.46 ± 0.16	7.99 ± 0.02	1045 ± 12.3
		ID-Po70-3	32.1 ± 2.33	41.5 ± 0.71	6.98 ± 0.07	268 ± 20.6
		ID-Po70-4	11.1 ± 0.24	12.6 ± 0.95	7.18 ± 0.02	207 ± 13.7
Com	Pre 1970	Com-Pre70-1	2.78 ± 0.81	9.11 ± 2.03	7.26 ± 0.01	158 ± 7.36
	Post 1970	Com-Po70-1	18.5 ± 0.20	14.9 ± 3.02	7.45 ± 0.02	118 ± 1.35
		Com-Po70-2	9.21 ± 0.40	12.4 ± 0.83	7.52 ± 0.03	128 ± 21.1
		Com-Po70-3	3.06 ± 0.46	18.5 ± 0.43	7.76 ± 0.07	141 ± 15.7
		Com-Po70-4	2.00 ± 1.06	13.3 ± 1.16	7.18 ± 0.10	292 ± 19.9
IN	Pre 1970	IN-Pre70-1	6.26 ± 0.76	20.9 ± 0.77	8.24 ± 0.04	152 ± 9.94
		IN-Pre70-2	6.26 ± 0.76	20.9 ± 0.77	7.94 ± 0.01	204 ± 3.61
	Post 1970	IN-Po70-1	15.6 ± 0.79	8.76 ± 0.60	7.73 ± 0.04	182 ± 18.8
		IN-Po70-2	11.7 ± 0.70	3.38 ± 0.30	8.04 ± 0.02	76.7 ± 3.02

Table C.2. Bray–Curtis similarity scores of PCB homologue patterns between the samples and Aroclor mixtures.

#	Sample	A1221	A1242	A1248	A1254	A1260
1	Res-Pre70-1	0.23	0.34	0.27	0.54	0.59
2	Res-Pre70-2	0.32	0.56	0.51	0.46	0.19
3	Res-Pre70-3	0.14	0.45	0.43	0.77	0.41
4	Res-Pre70-4	0.07	0.46	0.56	0.74	0.35
5	Res-Pre70-5	0.50	0.34	0.30	0.47	0.29
6	Res-Po70-1	0.31	0.30	0.25	0.45	0.15
7	Res-Po70-2	0.17	0.52	0.41	0.34	0.24
8	Res-Po70-3	0.30	0.27	0.17	0.47	0.55
9	Res-Po70-4	0.20	0.58	0.57	0.48	0.26
10	Res-Po70-5	0.10	0.39	0.39	0.64	0.57
11	ID-Pre70-1	0.11	0.38	0.42	0.73	0.52
12	ID-Pre70-2	0.16	0.59	0.62	0.54	0.31
13	ID-Pre70-3	0.18	0.56	0.55	0.52	0.36
14	ID-Pre70-4	0.27	0.39	0.34	0.55	0.45
15	ID-Po70-1	0.27	0.30	0.20	0.36	0.40
16	ID-Po70-2	0.12	0.23	0.22	0.33	0.36
17	ID-Po70-3	0.16	0.90	0.63	0.34	0.15
18	ID-Po70-4	0.35	0.67	0.58	0.37	0.17
19	ID-Po70-5	0.21	0.36	0.31	0.56	0.53
20	Com-Pre70-1	0.30	0.33	0.19	0.29	0.31
21	Com-Po70-1	0.35	0.37	0.23	0.21	0.36
22	Com-Po70-2	0.30	0.31	0.17	0.29	0.50
23	Com-Po70-3	0.28	0.25	0.14	0.26	0.60
24	Com-Po70-4	0.25	0.47	0.33	0.40	0.51
25	IN-Pre70-1	0.30	0.51	0.43	0.38	0.35
26	IN-Pre70-2	0.30	0.43	0.32	0.37	0.31

27	IN-Po70-1	0.30	0.31	0.25	0.43	0.38
28	IN-Po70-2	0.30	0.27	0.22	0.35	0.11
29	Rec-1	0.11	0.52	0.70	0.57	0.20
30	Rec-2	0.11	0.32	0.36	0.55	0.09
31	Rec-3	0.15	0.45	0.36	0.54	0.63
32	Rec-4	0.00	0.15	0.22	0.53	0.81
33	Rec-5	0.30	0.15	0.01	0.09	0.16
34	Energy-1	0.18	0.35	0.21	0.52	0.70

Table C.3. Chlorine per biphenyl for all samples.

Land Use	Land Development	Sample Name	Mean	S.D.
GS	N/A	Rec-1	4.30	0.06
		Rec-2	4.45	0.25
		Rec-3	4.87	0.18
		Rec-4	6.11	0.02
		Rec-5	2.48	0.11
Energy	N/A	Energy-1	4.95	0.40
Res	Pre 1970	Res-Pre70-1	4.29	0.04
		Res-Pre70-2	3.29	0.07
		Res-Pre70-3	4.47	0.07
		Res-Pre70-4	4.57	0.02
		Res-Pre70-5	2.90	0.16
	Post 1970	Res-Po70-1	3.11	0.15
		Res-Po70-2	4.90	0.13
		Res-Po70-3	4.05	0.12
		Res-Po70-4	4.37	0.12
		Res-Po70-5	4.98	0.05
ID	Pre 1970	ID-Pre70-1	4.93	0.04
		ID-Pre70-2	4.24	0.03
		ID-Pre70-3	4.36	0.14
		ID-Pre70-4	4.08	0.04
	Post 1970	ID-Po70-1	4.68	0.22
		ID-Po70-2	5.78	0.30
		ID-Po70-3	3.50	0.04

		ID-Po70-4	3.30	0.09
		ID-Po70-5	4.58	0.09
Com	Pre 1970	Com-Pre70-1	3.26	0.06
	Post 1970	Com-Po70-1	3.44	0.30
		Com-Po70-2	3.92	0.05
		Com-Po70-3	4.73	0.03
		Com-Po70-4	4.56	0.16
IN	Pre 1970	IN-Pre70-1	4.30	0.02
		IN-Pre70-2	3.42	0.02
	Post 1970	IN-Po70-1	3.85	0.07
		IN-Po70-2	3.42	0.02

Table C.4. Toxic equivalency value (TEQ pg/g) for all samples.

Land Use	Land Development	Sample Name	Mean	S.D.
GS	N/A	Rec-1	< 0.0004	
		Rec-2	0.0145	0.0022
		Rec-3	0.0278	0.0029
		Rec-4	0.0265	0.0012
		Rec-5	< 0.0004	
Energy	N/A	Energy-1	< 0.0004	
Res	Pre 1970	Res-Pre70-1	0.0317	0.0020
		Res-Pre70-2	< 0.0004	
		Res-Pre70-3	26.3	3.70
		Res-Pre70-4	0.0808	0.0054
		Res-Pre70-5	0.0539	0.0331
	Post 1970	Res-Po70-1	< 0.0004	
		Res-Po70-2	< 0.0004	
		Res-Po70-3	< 0.0004	
		Res-Po70-4	0.106	0.022
		Res-Po70-5	< 0.0004	
ID	Pre 1970	ID-Pre70-1	41.2	2.02
		ID-Pre70-2	8.55	0.22
		ID-Pre70-3	0.138	0.013
		ID-Pre70-4	20.9	0.8
		ID-Po70-1	0.0243	0.0032
		ID-Po70-2	0.00766	0.01047

	Post 1970	ID-Po70-3	< 0.0004	
		ID-Po70-4	< 0.0004	
		ID-Po70-5	< 0.0004	
Com	Pre 1970	Com-Pre70-1	0.0767	0.0079
	Post 1970	Com-Po70-1	0.00513	0.00255
		Com-Po70-2	0.0121	0.0006
		Com-Po70-3	0.0712	0.0076
		Com-Po70-4	< 0.0004	
IN	Pre 1970	IN-Pre70-1	< 0.0004	
		IN-Pre70-2	< 0.0004	
	Post 1970	IN-Po70-1	0.0450	0.0025
		IN-Po70-2	< 0.0004	
		IN-Po70-2	< 0.0004	

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