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Estimating the long term leaching potential of PCBs in soil

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Abstract

Polychlorinated Biphenyls (PCBs) are a group of man-made organic chemicals known as chlorinated hydrocarbons. PCBs were banned in 1979 due to its toxicity and persistence in the environment. Variations of PCBs were determined in soils and plants. Degradation of PCB Aroclors and congener patterns were investigated from known contaminated site at a former Military Air Base. PCB concentrations ranged between 17 - 1,040 μg L^{-1} at 0 - 0.5 m depth, between 36 - 898 μg L^{-1} at 1.0 - 1.5 m depth, between 20 - 73 μg L^{-1} at 4 m depth and 21 μg L^{-1} at 8 m depth. There is also a negative correlation between the mean of the Toxicity Characteristic Leaching Procedure (TCLP) of PCBs versus the sampling depth. Linear regression analyses were used to determine the correlation of the factors such as depth, Octanol/Water Partition Coefficient (K_{ow}) and solubility affected Soil/Water Distribution Coefficient (K_d). Effects were found only when the soil depth is at 0.3 - 0.5 m (at 95% confidence level). K_d is positively correlated with K_{ow} while K_d has some level of negative correlation with solubility. No effect was found at 1.0 - 1.5 m, 4.0 m, and 8.0 m. Organic Carbon/Water Partition Coefficient (K_{oc}) showed high results exceeding the log K_{oc} criteria of 4.5; as a result, this is now a matter of concern on the potential adverse effects of the substance on terrestrial organisms within its vicinity.

Keywords: Aroclor, leaching, polychlorinated biphenyls, vertical partitioning.

Introduction

Polychlorinated biphenyls (PCB) are a class of 209 congeners with isomers of chlorinated biphenyls exhibiting varying degrees of chlorination. These organic chlorine compounds have been widely used worldwide since the 1930s in capacitors, transformers, flame retardants, inks, and hydraulic fluids due to their thermal and chemical stability, resistance to alkalis and acids, low flammability, and low electrical conductance (Harrad et al., 1994; Hansen, 1999; Bozlaker et al., 2008; Zhang et al., 2011).

PCBs used for industrial and commercial purposes are known by different names in several countries, e.g., Kanechlor (Japan), Sovol (USSR), Fenchlor (France), Chlophen (Germany), and Aroclor (United States). In particular, Aroclor had a PCB composition with chlorine content comprised of penta- to octa- chlorinated congeners (Hutzinger et al., 1974; US EPA, 1996; ATSDR, 2000). Specifically, Aroclor contains congeners comprising of 12% penta-chloro biphenyls (CBs), 42% of hexa-CBs and 38% hepta-CBs, 7% octa-CBs, and 1% nona-CBs (US EPA, 1996; ATSDR, 2000).

Concerns on PCB contamination in the environment and on human health surfaced during the 1960s, which led to countries scaling down their PCB production and its eventual discontinuation worldwide in the late 1970s (Hansen, 1999; Fang et al., 2017). Eventually, PCBs were considered and listed as one of the 12 Persistent Organic Pollutants (POP) under the Stockholm Convention in 2001 due to its persistence and ease

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in being spread across environments and ecosystems, as well its potential for bioaccumulation and biomagnification (IARC, 2016; Nguyen et al., 2016; Adeyinka and Moodley, 2018).

Due to the lipophilicity and hydrophobicity of these compounds, PCBs are notably stable, persistent, and have long half-lives that promote accumulation in the environment, preferentially in organic matter and lipids (Notarianni et al., 1998; Malina and Mazlova, 2017; Neira et al., 2018). Despite being out of production for several decades, PCBs are still being detected in various environmental media in high concentrations such as in soils (Girvin and Scoot, 1997; Armitage et al., 2006; Fu et al., 2009; Zhang et al., 2011; Melnyk et al., 2015), marine sediments (Vane et al., 2007; Wang et al., 2010; Neira et al., 2018), atmosphere (Choi et al., 2008; Fu et al., 2009; Baek et al., 2010), and water (Harrad et al., 1994). PCBs released and stored in the soil and water media may eventually be distributed and transported widely across environments through local and long-range atmospheric processes, ultimately entering into the food chain (Wania and Mackay, 1996; Nguyen et al., 2016; Malina and Mazlova, 2017).

Although PCBs partition in soil, water, and air media, numerous studies (Cetin et al., 2007; Cetin and Odabasi, 2007; Bozlaker et al., 2008; Glüge et al., 2016) suggest that atmospheric processes remain the main driver in the mobility and deposition of these persistent organic pollutants; especially in soils and sediments. Volatile biphenyls, particularly highly-chlorinated congeners with chlorine atoms > 4 released through air-soil exchange, are easily adsorbed onto organic matter (Hansen, 1999; Cetin et al., 2007; Malina and Mazlova, 2017). Hites and Eisenreich (1987); Steuer et al., (1999) and Malina and Mazlova (2017) reported that these highly-chlorinated biphenyls were sorbed to dust particles in the atmosphere, clay and oxide surfaces, and on suspended solids in water (Wang et al., 2008; Badea et al., 2014).

More specifically, PCBs are known to have very low water solubility and high Octanol/Water Partition Coefficient (K_{ow}), causing strong sorption in soils (Badea et al., 2013). Its solubility decreases as chlorine content becomes higher or chlorinated biphenyls increase. The physical and chemical properties of soil, prevailing weather conditions, and age of contamination affect the varying degrees of PCB contamination in soils (Badea et al., 2013; 2014).

The mobile fractions of PCBs in soil can be evaluated by determining various leaching tests. Toxicity Characteristic Leaching Procedure (TCLP), developed by the United States Environmental Protection Agency (US EPA), estimates the inorganic and organic contaminants of solid wastes that may pose potential hazard to health and environment (US EPA, 1992). Toxicity assessments from these hazardous contaminants recommend the use of TCLP. According to US EPA (1992), this method identifies contaminants present in leachate and their concentrations as TCLP simulates the conditions in landfill over time. This correlates to contaminants present in soil in a period of time with slow movement through external influence of rainwater and slow degradation from sunlight. For a solid or hazardous waste that is 100% physically solid, the maximum theoretical leachate concentration is 5% of the total concentration in the waste (US EPA, 1992).

Elutriate test measures and predicts the release of contaminants in water column (Vicinie et al., 2011). The Soil/Water Distribution Coefficient (K_d) is used to calculate the distribution of contaminants between the soil and leachates. (Badea et al., 2013; 2014) and therefore also used to estimate risks of contamination in groundwater and surface water.

This paper presents the leachate concentrations of PCBs from soil collected in August 2013, 2014, and 2017 in the study area. Partitioning and hydrophobicity of leachate properties of PCBs in soil at different depths were analyzed and studied. The purpose was to examine the patterns of leachability of PCBs as to horizontal and vertical migration and trends including the PCB weathering of Aroclor and congener profiles in the natural environment with respect to time. Furthermore, this research seeks to affirm low leaching rates of highly chlorinated Aroclors and congeners in relation to low water solubility and high octanol/water coefficient in the immediate environment.

Material and Methods

Study Site

The study area is located at the Wagner Aviation, Clark Freeport Zone, Pampanga, Philippines (15°10'14.44"N, 120°32'39.18"E) (Figure 1), which was part of the former Clark Air Base ran by the United States military until 1991. There were 20 PCB transformer units stored in Wagner Aviation in 1997 (CDC-EPMD, 1997). Each transformer unit at Wagner Aviation had an oil capacity of 757 L and a rating of 225 kVA (Weston International, 1997). One unit tested had a PCB Aroclor 1260 concentration of 62,540 mg kg^{-1} based on records of the US Air Force on March 18, 1991.
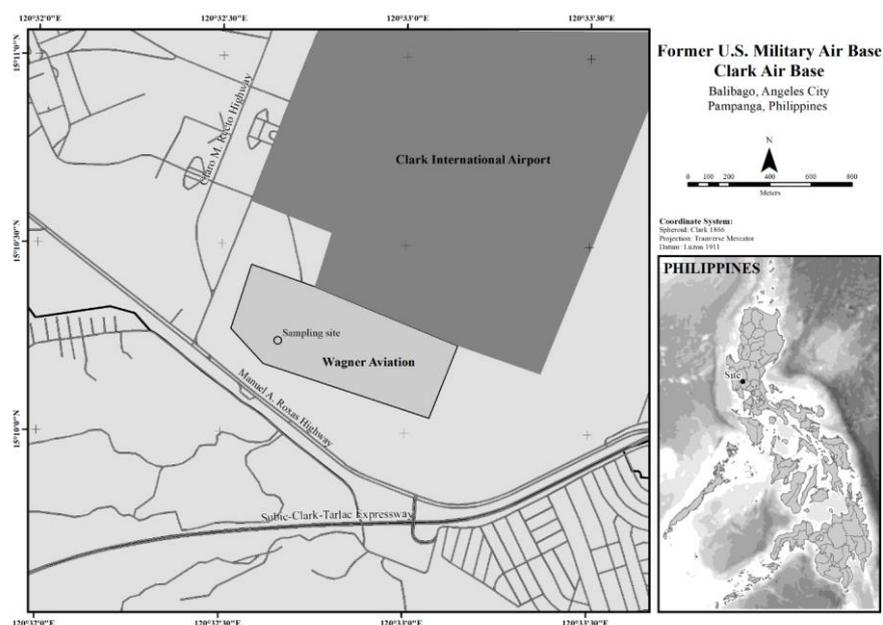
Figure 1. PCB Transformer sampling site at former US Military Air Base, Angeles City, Philippines

In 2002, it was reported that some of the transformer units at the Wagner Aviation site were missing. Moreover, PCB-laced oils have spilled into the underlying soil. Several tests and surveys were done on the soil and surface material of the site. Results of PCB concentrations at Wagner Aviation were 7,800 mg kg^{-1} (Weston International, 1997), 7,200 mg kg^{-1} (Harvard University-School of Public Health cited in CDC-EPMD (1997) and 4,400 mg kg^{-1} (ARRPET-De La Salle University cited in CDC-EPMD (1997) exceeding the US EPA Risk Based Concentration for industrial areas of 10 mg kg^{-1}.

Sample Collection

Samples were collected in the site with an area of approximately 9 sq. m. Surface and sub-surface soil sampling were done in accordance with standard protocols. Soils were collected at sampling depths of 0.3m, 0.5m, 1.0m, 1.5m, 2.0m, 4.0m, and 8.0m. on August 2013, August 2014, and August 2017. Physico-chemical parameters (Table 1), measured were pH, moisture, organic carbon, and particle size for collected surface soils. Grain size distribution within the contaminated area was found to be 25% gravel, 60% sand, and 15% silt with negligible clay fractions (Figure 2, sample 1).

Table 1. Moisture content and pH of soils

Sampling Location	Sampling Depth	Date	pH					% Moisture					Average moisture (%)
			Test 1	Test 2	Test 3	Test 4	Test 5	Test 1	Test 2	Test 3	Test 4	Test 5	
Within the contaminated area	0.5 meters	08.2013	6.85	7.15	-	-	-	14.75	17.99	-	-	-	16.37
		08.2014	6.96	5.02	4.73	4.24	5.12	15.68	21.40	16.67	20.05	16.31	18.02
		08.2017	6.63	6.62	6.33	6.27	-	29.31	22.57	23.19	21.93	-	24.25
	1.0- 1.5 meters	08.2013	7.89	6.92	-	-	-	23.61	24.29	-	-	-	23.95
		08.2014	4.90	5.04	-	-	-	21.11	29.88	-	-	-	25.50
		08.2017	6.69	6.58	6.59	-	-	17.28	18.16	14.69	-	-	16.71
	4.0 meters	08.2013	7.16	7.08	7.16	-	-	12.65	17.49	16.51	-	-	15.55
	8.0 meters	08.2013	6.83	-	-	-	-	12.68	-	-	-	-	12.68

Toxicity Characteristic Leaching Procedure (TCLP) (US EPA Method 1311) tests were done by mixing the soil thoroughly and 50 - 70 g subsamples for each test were collected. A rotary agitation instrument was used and samples were rotated at 30 $\pm$ 2 rpm for 18 $\pm$ 2 hours. Samples were filtered and extracts were collected using US EPA Method 3510C (Separatory Funnel Liquid/liquid extraction method). The average weight of each sample used was 60 g filled to a total 1 L filled with an extraction fluid. The extraction fluid was made by mixing 5.7 mL glacial CH_3CH_2OOH, added to 600 mL distilled water, and finally added with 64.3 mL of 1N NaOH and diluted to a volume of 1 L. Surrogate recovery standards containing Decachlorobiphenyl and Tetrachloro-m-xylene were spiked on each sample prior to extraction. Extracts were concentrated into 5 mL using a Zymark Concentrator. Clean-up using US EPA Method 3630 Silica Gel method was employed after every sample extraction procedure.

Grain Size Distribution
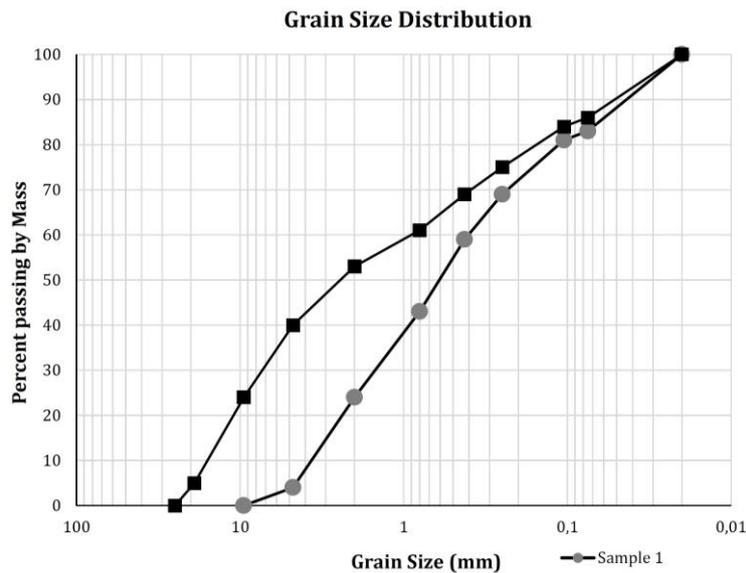
Figure 2. Grain size distribution of soils in Wagner Aviation, Angeles City, Philippines

Gas Chromatography coupled with dual Electron Capture Detector (GC-ECD) - Hewlett Packard 5890 Series II/Agilent 7890B was used in the quantitation of PCB Aroclors and PCB Congeners. Nine Aroclors were selected and analyzed. Around 19 PCB congeners were quantitated based on the US EPA list.

An elutriate test was performed by mixing one part of soil with four parts of water from the area allowing the soil/water mixture to settle, filtering the supernatant and measuring the release of contaminants in the elutriate or filtered supernatant. The filtered supernatant was extracted using US EPA method 351^0C and cleaned up using US EPA method 3630. Detection limit using GC-ECD for PCBs in TCLP and elutriate aqueous extracts were 1.0 µg L^{-1} for Aroclors and 0.25 µg L^{-1} for total congeners.

Computational methods and statistical analyses

Descriptive statistics such as mean, range, and standard deviation were computed for each sample collected at different depths. Calculation of soil-water distribution coefficients used the following equation:

$$K_d = C_s / C_w$$

where, C_s and C_w are the concentrations of a given compound in the soil and aqueous phase, respectively.

Calculation of soil-water distribution coefficients used the following equation:

$$K_{ow} = C_o / C_w$$

where, C_o and C_w are the concentrations of a given compound in octanol and aqueous phase, respectively.

Log10 K_d was then used to normalize its distribution. The relationships between K_d to K_{ow} and solubility of the samples were analyzed through two-factor, simple linear regression between Log K_d vs Log K_{ow} and Log K_d vs Solubility; grouped for each soil depth.

Results and Discussion

Characterization of PCBs in surface soils

Total organic carbon was <0.0001% in all soil samples collected. The low Total Organic Carbon present was a result of the dominantly sand-silt grain composition of the mid to upper horizon soils deposited during the June 1991 Mt. Pinatubo volcanic eruption (Paladio-Melosantos et al., 1996). The eruption of Mt. Pinatubo, located 20 km southwest of the study area, deposited a blanket of silt to fine-grained sand sized ashfall, derived chiefly from andesitic to dacitic magma, over extensive areas surrounding the volcano. Thick beds of volcaniclastic sediments were deposited over the area as heavy rains brought by Typhoon Yunya caused the mobilization of ash and other volcanic products as lahar and mudflows tens of kilometers around the volcano (Paladio-Melosantos et al., 1996).

Figure 3 shows the historical trends of total PCBs from 2013 to 2017. Not all PCB Aroclors were detected except for Aroclor 1260, which was positively detected in all soil depths sampled. Table 3 shows the results of TCLP in soils. The degree of PCB concentrations is expectedly inversely correlated to depth. PCB concentrations range between 17 and 1,040 µg L^{-1} at 0 - 0.5 m depth, between 36 and 898 µg L^{-1} at 1.0 - 1.5 m depth, between 20 and 73 µg L^{-1} at 4.0 m depth, and 21 µg L^{-1} at 8.0 m depth. This suggests that the transport of PCBs to soil is fractionated and mobility is poor (Girvin and Scott, 1997).
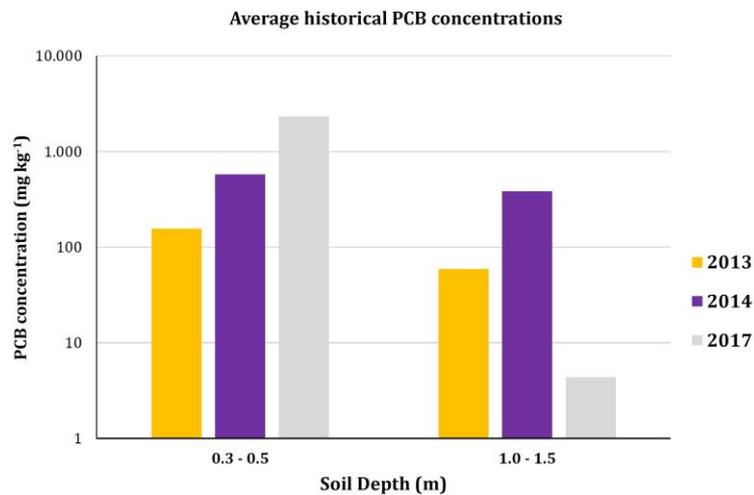
Figure 3. Historical trends of average PCBs from 2013 to 2017 at Wagner Aviation, Angeles City, Philippines

The PCB screening value of the Philippine Department of Environment and Natural Resources (DENR) for soil is 1 mg kg^{-1} and 0.74 mg kg^{-1} for Dutch Intervention Value (DIV) and US EPA Regional Screening Values (Industrial), respectively. PCBs in soil were assessed because they exceeded these screening values. The risk for ingestion from groundwater through leachates was evaluated based on groundwater level. According to groundwater guidelines of DENR Tier 1 Screening Values and Dutch Standard Intervention, total PCBs should not exceed above 0.017 µg L^{-1} and 0.01 µg L^{-1}, respectively. PCBs leachates in all surface and subsurface soils significantly exceed the required screening and intervention values. Based on this screening and intervention standards, the area is heavily polluted with PCBs. Since the site was contaminated with PCBs for almost three decades, this strongly suggests that PCBs are persistent and have poor degradation to sunlight and organisms. It is also noted that Aroclors with larger chlorine composition, particularly Aroclor 1260, are present in the soil and have leached out significantly even if it is naturally liphophilic. As a result, PCB leachates indicate a large risk for groundwater contamination. Unfortunately, there has not been a detailed study on PCB contamination of the groundwater in the area.

Different leaching tests were done to confirm the leaching potentials of PCBs using the congener approach. This test has a short exposure to water from contaminated soil compared to TCLP exposure at 18 hours. Table 3 shows the results of elutriate tests. Hexa-CBs and Hepta-CBs predominantly showed the highest concentrations among all other CBs.

PCB concentrations in the soil profile succession as illustrated in Figure 4, show a distinct distribution pattern. The topsoil is dominated by highly chlorinated biphenyls: 6Cl, 7Cl, and 9Cl, accounting for more than 75% of the total concentration of PCBs. As depth increases however, less chlorinated biphenyls such as 4Cl and 5Cl begin to increase in proportion relative to the total amount of PCB. At 0.3 to 0.5 m depth, K_d is positively correlated with K_{ow} while K_d is inversely correlated with solubility as shown in Figure 5. The same trend was not observed in all other sampling depths.
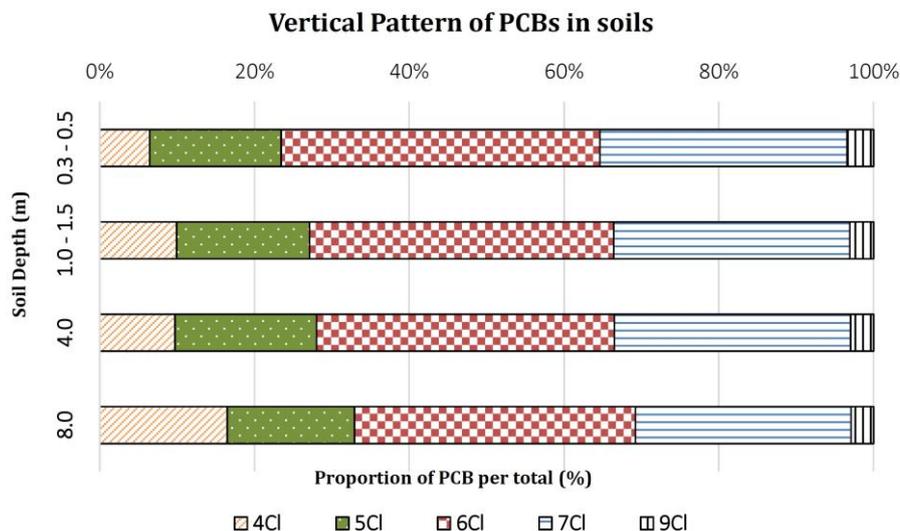
Figure 4. Depth distribution of PCB concentrations in the soils at the Wagner Aviation site

The graphical regression graphs of K_{ow} and Solubility with K_d are shown in Figure 5 and the tabulated statistical results in Table 4. Regression analyses show that the near surface layer (0.3 - 0.5 m layer) is the most susceptible soil profile to contamination as expected. Take note that although there are changes in the pattern, the relationship was not significant, likely because of lack of data points or other factors that changed the concentration of the chemicals being analyzed.
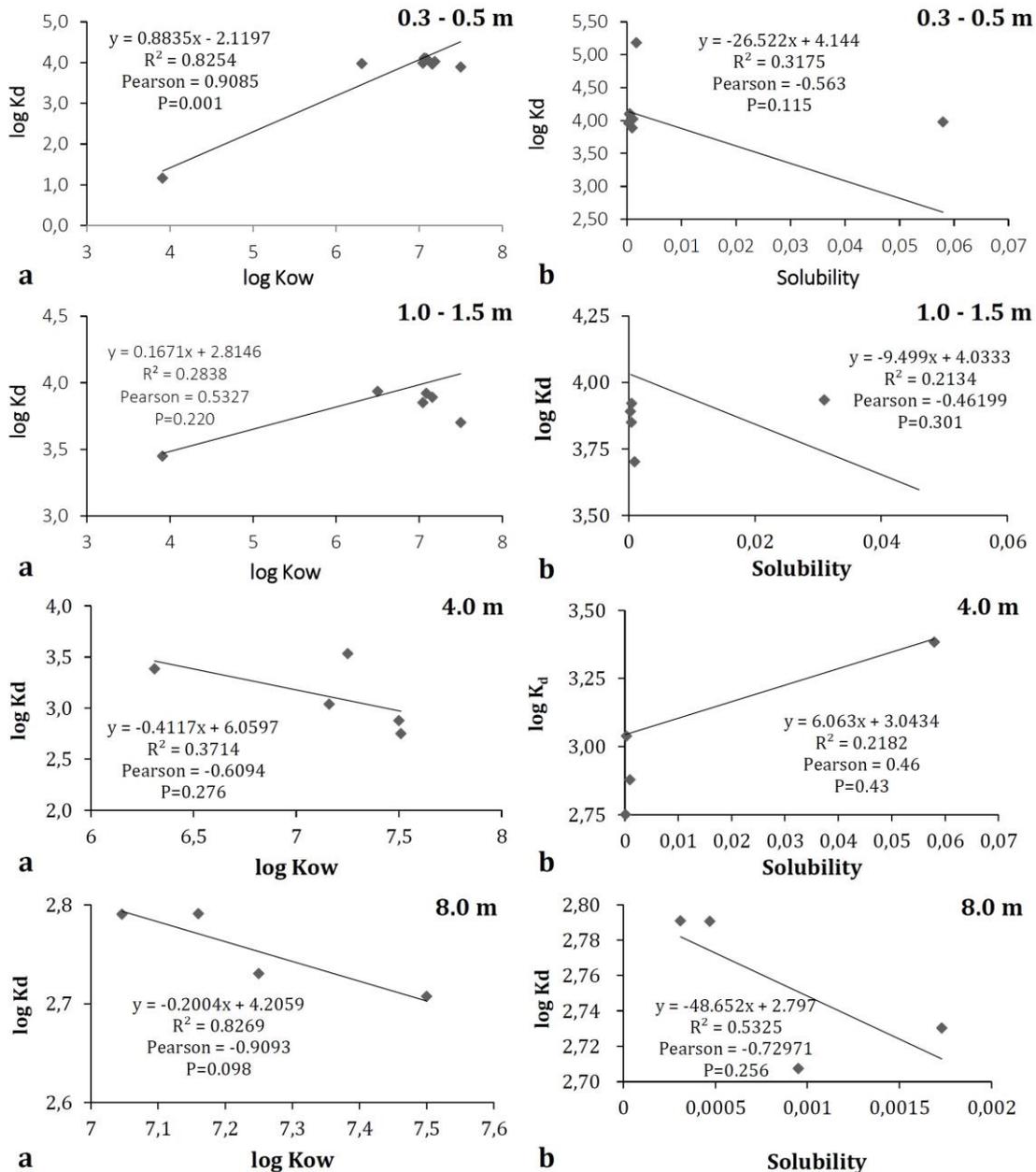
Figure 5. Linear relationship between (a) Kow vs Kd and (b) solubility vs Kd at different soil depths.

This pattern reflects the expected behavior of these PCBs in the soil media. Highly chlorinated biphenyls are lost due to eventual degradation and volatilization in deeper soil profiles and are transported upwards to more surficial soil layers; confirming the same observations found by other researchers (Backe et al., 2004; Wang et al., 2010; Zhang et al., 2011).

Relationships of Organic Carbon, K_d and K_{oc} in soil leachates

The relationship among Organic Carbon/Water Partition Coefficient (K_{oc}), K_d, and Organic Carbon was statistically determined based on the following:

$$K_{oc} = (K_d * 100)/ \% \text{ Organic carbon}$$

where the % Organic carbon is = 0.0001

Figure 6 shows the relationship of the two variables based on the equation. All readings show very high K_{oc} of a substance and exceed the log K_{oc} criteria of 4.5. If log K_{oc} is greater than 4.5, this means that there should be significant concerns on the potential adverse effects of the substance on terrestrial organisms.

Table 2. Elutriate Results of PCB concentration as Congeners, µg L⁻¹

2013 PCB Congeners	0.3-0.5 meters		1.0-1.5 meters		4.0 meters		8.0 meters	
	Average	Range	Average	Range	Average	Range	Average	Range
2,5,2',5'-Tetrachlorobiphenyl (BZ # 52)	0.03	0 ~ 0.08	0.09	0 ~ 0.20	0.04	0 ~ 0.12	0.08	0 ~ 0.14
2,3',4,4'-Tetrachlorobiphenyl (BZ # 66)	1.06	0~ 3.2	0.00	-	0.28	0 ~ 0.85	-	-
2,2',4,5,5'-Pentachlorobiphenyl (BZ # 101)	1.18	0 ~ 3.5	0.43	0 ~ 1.3	-	-	-	-
2,2',3,4,4',5'-Hexachlorobiphenyl (BZ # 138)	0.28	0 ~ 0.83	0.21	0 ~ 0.64	0.50	0 ~ 1.2	0.78	0 ~ 2.3
2,2',3,4,5,5'-Hexachlorobiphenyl (BZ # 141)	1.45	0 ~ 4.3	0.00	-	-	-	-	-
2,2',4,4',5,5'-Hexachlorobiphenyl (BZ # 153)	5.12	0 ~ 14	2.10	0.06 ~ 5.6	2.26	0 ~ 5.5	0.85	0 ~ 2.2
2,2',3,3',4,4',5-Heptachlorobiphenyl (BZ # 170)	1.39	0 ~ 4.2	0.00	-	-	-	-	-
2,2',3,4,4',5,5'-Heptachlorobiphenyl (BZ # 180)	4.97	0 ~ 14	1.62	0 ~ 4.8	1.88	0 ~ 4.5	0.73	0 ~ 2.2
2,2',3,4,4',5',6-Heptachlorobiphenyl (BZ # 183)	0.99	0 ~ 3.0	0.39	0 ~ 1.2	-	-	-	-
2,2',3,4',5,5',6-Heptachlorobiphenyl (BZ # 187)	2.52	0.13 ~ 7.0	0.91	0 ~ 2.7	0.19	0 ~ 0.56	0.44	0 ~ 1.2
2,2',3,3',4,4',5,5',6-Nonachlorobiphenyl (BZ # 206)	-	-	-	-	-	-	-	-
Total PCBs	18.99	0.13~54.11	5.75	0.06~16.44	5.15	0.0-12.73	2.88	0.0-8.04

Table 3. Average log Kd (L kg⁻¹) values obtained for target PCBs from surface to subsurface soils.

PCBs	Elutriate Results of PCBs, µg L⁻¹ (CW)				Concentrations in Soil, µg kg⁻¹ (CS)				K_d, L kg⁻¹				(Mackay et al., 2006)	
	0.3-0.5 m	1.0-1.5 m	4.0 m	8.0 m	0.3-0.5 m	1.0-1.5 m	4.0 m	8.0 m	0.3-0.5 m	1.0-1.5 m	4.0 m	8.0 m	log K_{ow}	Solubility, µg L⁻¹
PCB 52	0.03	0.09	0.04	0.08	0.4	256	-	-	14.52	2,808	-	-	3.910	0.046000
PCB 66	1.06	0.00	0.28	-	10,145	4,076	686	283	9,530	-	2,422	-	6.310	0.058000
PCB 101	-	0.43	-	-	13,328	3,733	643	141	-	8,611	-	-	6.500	0.031000
PCB 138	0.28	0.21	0.50	0.78	41,887	11,061	1,718	419	151,962	51,816	3,419	537.64	7.250	0.001730
PCB 141	1.45	0.00	-	-	15,309	4,004	635	141	10,553	-	-	-	7.190	0.001090
PCB 153	5.12	2.10	2.26	0.85	39,836	10,574	1,711	374	7,778	5036	757	510.02	7.500	0.000950
PCB 170	1.39	0.00	-	-	17,862	4,263	638	141	12,825	-	-	-	7.060	0.000504
PCB 180	4.97	1.62	1.88	0.73	45,441	12,580	2,054	450	9,152	7,784	1,094	618.08	7.160	0.000310
PCB 183	0.99	0.39	-	-	12,343	3,264	545	122	12,423	8,358	-	-	7.087	0.000498
PCB 187	2.52	0.91	-	0.44	24,651	6,418	1,063	243	9,775	7,090	-	617.57	7.046	0.000470
PCB 206	-	-	0.19	-	2,674	667	104	25	-	-	564	-	7.510	0.000112
Total PCBs	17.81	5.75	5.15	2.88	223,476.4	60,896	9,797	2,339	224,012.52	91,503	8256	2283.31	74.523	17.81
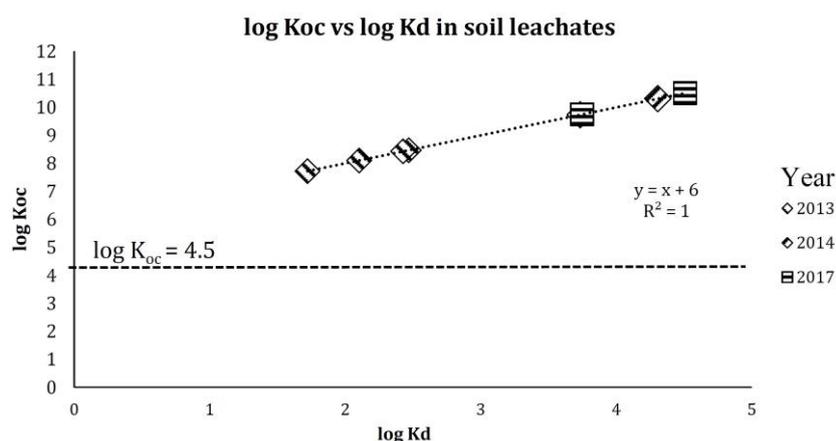
Figure 6. Relationship of log Kd and log Koc in soil leachates

Table 4. Results of K_d, K_{ow}, and Solubility at different depths

Depth	Log K_d vs Log K_{ow}			Log K_d vs Solubility			Log K_{ow} and Solubility Interaction
	R^2	Pearson's R	P	R^2	Pearson	P	
0.3 - 0.5 m	0.8254	0.9085	0.001	0.3175	-0.5630	0.115	not significant (P=0.455)
1.0 - 1.5 m	0.2838	0.5327	0.220	0.2134	-0.4620	0.301	not significant (P=0.827)
4.0 m	0.3714	-0.6094	0.276	0.2182	0.4600	0.430	not significant (P=0.258)
8.0 m	0.8269	-0.9093	0.098	0.5325	-0.7297	0.256	not significant (P not computed)

Conclusion

The data shows the relationship between K_d and predictors K_{ow}, solubility and the interaction between K_{ow} and solubility. Significant effects were found only when the soil depth is near the surface at 0.3 - 0.5 m. K_d is positively correlated with K_{ow} while K_d is weakly inversely correlated with solubility.

Pertaining to the mobility of the substance through the soil profile, the obtained K_d showed high values which indicated a strong adsorption onto soil resulting to lower mobility towards the deeper layers of the soil profile. Still, after 15 years since the PCBs entered the environment, migration of some of its congeners are observed at 8 meters depth and beyond. K_{oc} showed high results which exceeded the log K_{oc} criteria of 4.5. This indicates the potential adverse effects of PCBs on terrestrial organisms. K_{oc} is an important indicator for estimating environmental distribution and environmental exposure level of PCBs. It is recommended to examine the transport of PCBs to terrestrial fauna, groundwater table, and its potential risk to humans.

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