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Polychlorinated biphenyls remediation in soil using moringa seeds and coconut shell based adsorbents

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ABSTRACT

The occurrence and remediation of polychlorinated biphenyls (PCB) in soil from Jos, Plateau State Nigeria was carried out. The polychlorinated biphenyls in the soil was extracted with hexane-acetone mixture (1:1) in ultrasonic bath and cleaned with column chromatography packed with silica gel. The polychlorinated biphenyls were quantified using Agilent 6975 Gas chromatography Mass Spectrophotometer and the concentrations were found as: PCB18 (0.71), PCB20 (0.37), PCB28 (0.38), PCB29 (0.38), PCB44 (0.42), PCB52 (0.66), PCB101 (0.61), PCB105 (0.17), PCB137 (0.64), PCB142 (0.33), PCB153 (0.42), PCB170 (0.13) and PCB180 (0.21) mg/kg. After remediation of PCB with moringa seeds, the concentration were as; PCB20 (0.28), PCB28 (0.28), PCB29 (0.28), PCB52 (0.30), PCB101 (0.18) and PCB143 (0.91), while the others removed completely. The moringa seeds powder removed the PCBs completely except PCB105. The activated carbon (AC) prepared moringa seeds reduced the concentrations of BCBs as; PCB 18 (0.04), PCB 20 (0.011), PCB 28 (0.019), PCB 29 (0.019), PCB 44 (0.021), PCB 52 (0.33), PCB 101 (0.031), PCB137 (0.032), PCB 142 (0.017), PCB 153 (0.021), PCB 170 (0.007) and PCB 180 (0.011) mg/kg. The result suggests that moringa seed powder could possibly be served as an excellent means of remediation for polychlorinated biphenyls in contaminated soil.

Capsule Summary: The polychlorinated biphenyls in soil were monitored and remediated using moringa seeds powder and activated carbon. The moringa seeds powder showed promising efficiency for the remediation of PCBs in contaminated soil.

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INTRODUCTION

Polychlorinated biphenyls (PCBs) are persistent and toxic pollutants that have been widely distributed into the environment (Srishty et al., 2017). They are classified as persistent organic pollutants (POPs) under the Stockholm Convention due to their persistence bioaccumulation, high toxicity and long-range atmospheric transport with potential carcinogenicity (Gouin et al., 2004). Research evidence has

revealed that long range transport is a major source of this contaminant in a remote area (Qibian et al., 2018*)*.

Polychlorinated biphenyls are mainly synthetic chemicals with 2–10 atoms of chlorine attached to the biphenyls molecules and were produced several years ago with annual production estimation of 1.5millions tones (Andersson et al., 2015). Although banned in most of the countries in 1980, yet its emission still continues from old electrical equipment, inadequate management of wastes and electronic equipment leakage or improper disposal of transformer and capacitor oil (Wang et al., 2016).

There are 209 compounds often known as PCBs congeners. Their properties such as heat resistance, low electrical conductivity and thermal degradation performance make them excellent in use as insulators mediums, flame retardant agents, plasticizers and pesticides additives since 1930s (Everaert et al., 2015; Koh et al., 2015; Yuan et al., 2015a*)*. Out of the 1.5million tones production globally, 48% production of the PCBs is used for transformer oils, 21% for small capacitors, 10% for other closed system, these include heat transfer fluids, hydronic fluids, liquid filled cables and circuit breakers and approximately 21% for open systems as paints and pesticides *(*Andersson et al., 2015). They have also been used as organic diluents, plasticizers, adhesives, dust reducing agent cutting oils, flame retardants, sealants and in carbonless copy paper. Some of these PCBs uses have resulted in them directs introduction into the environment.

Contamination of the environmental compartments by polychlorinated biphenyls is a matter of great concern to the environmentalists worldwide. They have been reported to have adverse effects on the environment, human and ecosystem (Yuan et al 2015b*)*. They enter the environment as a mixture containing different individual congeners (Sapozhnikova et al., 2004). Despite their phase out in 1970, PCBs residues still have been reported in soil, water and air all over the world (Ge et al., 2013; Rio et al., 2010; Whitehand et al., 2015; Yang et al., 2014) and their levels are not expected to decrease significantly in the next few decades (Gao et al., 2013).

Table 1: Polychlorinated biphenyls detected in soil samples

PCBs	A	B	C	D	A*	F	G	H
PCB 18	0.71	BDL	BDL	0.03	0.79	BDL	BDL	0.04
PCB 20	0.37	0.28	0.28	0.007	13.27	0.28	ND	0.31
PCB 28	0.38	0.27	0.39	0.015	13.77	0.27	ND	0.52
PCB 29	0.38	0.27	0.39	0.015	13.77	0.27	ND	0.52
PCB 44	0.42	ND	1.11	0.005	7.71	ND	ND	0.1
PCB 52	0.66	0.42	0.4	0.004	27.66	0.3	0.18	0.18
PCB 101	0.61	ND	ND	0.03	3.48	0.18	ND	0.17
PCB 105	0.17	ND	ND	0.109	0.5	ND	ND	0.32
PCB 118	BDL	BDL	BDL	BDL	0.31	BDL	ND	BDL
PCB 137	0.64	ND	ND	0.01	27.81	ND	ND	0.42
PCB 142	0.33	0.28	0.43	0.026	9.47	0.91	ND	0.79
PCB 153	0.42	ND	ND	0.006	37.38	ND	ND	0.53
PCB 170	0.13	ND	BDL	0.005	0.54	ND	ND	0.02
PCB 180	0.21	ND	0.15	0.243	0.38	ND	ND	0.44
PCB 194	BDL	ND	ND	BDL	2.9	ND	ND	BDL
TOTAL	5.43	1.52	3.15	0.505	159.74	2.21	0.18	4.36

Key: A = sample of contaminated soil from Gagere Jos, B = contaminated soil from A + moringa seed powder, C = contaminated soil from A + moringa seed, D = contaminated soil from A + activated carbon. A* = sample of contaminated soil from Unijos old campus, F = sample A* + moringa seed, G = sample A*+ moringa seed powder and H = sample A* + activated carbon. PCB18 = 2,2,5-Trichlorobiphenyl PCB20 = 2,3,3-Trichlorobiphenyl PCB28 = 2,4,4-Trichlorobiphenyl, PCB29 = 2,4,5-Trichlorobiphenyl, PCB44 = 2,2,3,5-Tetrachlorobiphenyl, PCB52 =2,2,5,5-Tetrachlorobiphenyl PCB101 = 2,2,4,5,5-Pentachlorobiphenyl, C105 = 2,3,3,4,4-Pentachlorobiphenyl, PCB118 = 2,3,4,4,5-Pentachlorobiphenyl, PCB137 = 2,2,3,4,4,5-Hexachlorobiphenyl, PCB142 = 2,2,3,4,5,6-Hexachlorobiphenyl,PCB153 = 2,2,4,4,5,5-Hexachlorobiphenyl, PCB170 = 2,2,3,3,4,4,5-Heptachlorobiphenyl, PCB180 = 2,2,3,4,4,5,5-Heptachloribiphenyl, PCB194 = 2,2,3,3,4,4,5,5-Octachlorobiphenyl.

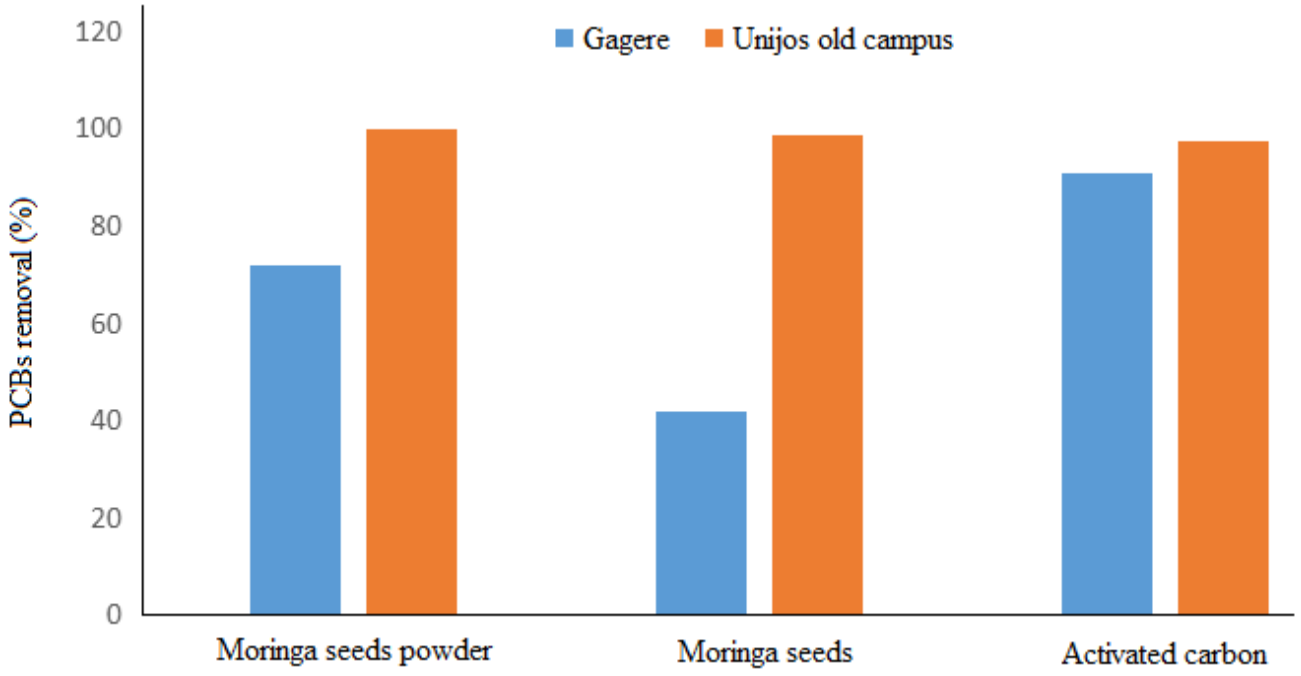

Fig. 1: Percentage removal of PCBs by moringa seed powder, moringa seed and activated compared from contaminated soil

Soil has been an important sink for PCBs and can also acts as a source of pollution (Zhang et al., 2014). Soil contamination has led to the reduction of agricultural areas which is a growing problem in many countries. Meijer et al 2013 reported that nearly 21,000 tons of PCBs have been discharged to the soil in the world. This can be accumulated gradually through the food chain and have a detrimental effect on human (Sirot et al., 2012*)*. The distribution of PCBs in soil can help to assess the level of pollution, infer the source of the emission and evaluate the human health risk (Li et al., 2015*)*. Therefore, the aim of this research is to investigate the occurrence of PCBs in soil and remediate using locally produced activated carbon from Coconut shell and moringa seed on contaminated soil from transformer installation Sites in Jos, Plateau State Nigeria with the objective of evaluating the efficiency of remediation between the activated carbon and the moringa seed.

MATERIAL AND METHODS

Sample collection and treatment

The soil samples were collected in March, 2019 from transformer installation site in Jos, Plateau State, Nigeria within the depth of 15 cm from different sites. The stone, weeds, leaves and roots were removed. All the samples were dried at ambient temperature, crushed and passed through a siever of 10 mm aperture and stored (Guanghui et al., 2017).

Remediation

For the remediation, coconut shell activated carbon was produced as reported elsewhere (Sodeide, 2012). The final product was air-dried after addition of distilled water for two weeks at room temperature instead of oven drying. This was packed in polyethylene bottles for further use. The moringa seeds was purchased from local market, cleaned from debris and crushed with pestle and mortal to obtained the powder seed, which was stored. The contaminated soil (10 g) was mixed with 5 g each of activated carbon, moringa seeds and moringa seed powder and allowed to stand for two weeks, and analyzed for PCBs.

Analysis of PCBs

The soil samples (20 g, moisture free) were mixed thoroughly with 10 g of anhydrous sodium sulphate and ultrasonicated with 30 mL of n-hexane/acetone (1:1 v/v) for 30 min. The extract was cleaned with column chromatography packed with silica gel and concentrated to 2 mL using rotary evaporator. The final extract of 2.0 ml was analyzed for PCBs using a gas chromatograph equipped with 63Ni electron capture detector GC - ECD model CP 3800. The capillary column used was VF 5ms 30 m × 0.25 mm id × 0.25 µm film thickness. The GC conditions were as follows: injection point temperature: 270 ^{0}C; oven temperature programme: 70 ^{0}C (hold 2 min) to 180 ^{0}C at a rate of 25

^{0}C/min (hold 1 min) to 300 ^{0}C at a rate of 5 ^{0}C/min. Temperature of detector was 300 ^{0}C; carrier gas-nitrogen at flow rate: 1.0 ml/min; make-up gas flow rate 29.0 ml/min was used. The total run time was 31.368 min. The PCBs were identified by the comparison of retention time of the PCBs in the samples with standard and PCBs were quantified (mg/kg) on the basis of dry weights.

RESULTS AND DISCUSSION

The polychlorinated biphenyls detected in the soil samples (A & A*) collected from Jos, Plateau State, Nigeria and remediated (B, C, D, F, G and H) are shown in Table 1. Sample A is soil sample collected 2 m away from the installed transformer and the PCB concentration ranges from 0.13 to 0.710 mg/kg. PCB 18 has the highest concentration with the value of 0.710 mg/kg and PCB 170 has the least with the value of 0.13 mg/kg, while others have various values but all less than 1.00 mg/kg. It can be seen that the concentration is in the order of PCB 18 > PCB 52 > PCB 137 > PCB 101 > PCB 44/153 > PCB 28/29 > PCB 20 > PCB 142 > PCB 180 > PCB 105 > PCB 170. Meanwhile PCBs 118 and 194 were not detected probably their values were below instrument detection limit. The total concentration of the PCBs in the soil in mg/kg of both samples (A and A*) were also indicated in Table 1 (4.30 mg/kg and 159.74 mg/kg) respectively, these values were above 2.0mg/kg set by United states under the toxic substance control act (TSCA) which regulates the PCBs in the environment, stipulating that any soil or sediments containing PCBs equal or greater than 2mg/kg that are spilled after 1978 from a source unauthorised for use are regulated as PCB remediation waste (USEPA, 2012).

For sample A* appears to be heavily polluted with the PCBs, the least concentration is PCB118 congener with the concentration of 0.310 mg/kg but highest with PCB 153 with the value of 37.380mg/kg. The various concentration in this site varies from PCB 153 > PCB 137 > PCB 52 > PCB 28/29 > PCB 20 > PCB 142 > PCB 44 > PCB 101 > PCB 194 > PCB 18 > PCB 170 > PCB 105 > PCB 180 > PCB 118. This study agreed with other finding that discovered soil within transformer installed sites in Porthartourt were above the 2.0 mg/kg set by the United States toxic act (Ayomide, et al, 2020). The remediation effiency of the activated carbon, moringa seed and the moringa seed powder were compared, and the use of activated carbon was more effective at lower concentration of PCBs, whereas at higher concentration of PCBs the moringa seed powder appeared more effective (figure 8). This was obtained by calculating the percentage removal of PCBs by the activated carbon, moringa seed and the moringa seed powder using Eq. 1.

$$X\ (\%) = a/b \times 100 \quad (1)$$

Where, X is the percentage of PCBs removed in a particular site, 'a' is the total concentration of PCBs removed in a particular site and 'b' is the total concentration of PCBs in a particular site. From the results obtained there pose a great danger, studies had shown various toxicological effects of PCBs to includes carcinogenicity, tumor promoter-type effects, endocrine disruption, tetratogenicity and possible impairment of psycho-development in human and animals as well (ATSDR, 2000; Walkowiak et al., 2001). The best way to estimate accurate toxicity and assessing the carcinogenic risk for PCBs is achieved with the use of congeners form rather than the total PCBs using their toxicity equivalent fator (TEF) (Davila et al., 1993). The PCBs dioxin like play important role in the evaluation of risk associated to PCBs release and in the development of PCBs clean-up level, this is because evaluation of total PCBs would not give accurate information on the environmental matrix.

Table 2: The toxicity equivalent fator (TEF) values for human risk assessment of PCBs

PCBs	Toxicity equivalent fator [a]	Toxicity equivalent fator [b]
PCB 77	0.0001	0.0001
PCB 81	0.0001	0.0003
PCB 105	0.0001	0.0003
PCB 114	0.0005	0.0003
PCB 118	0.0001	0.0003
PCB 123	0.0001	0.0003
PCB 156	0.0005	0.0003
PCB 157	0.0005	0.0003
PCB 167	0.0001	0.0003
PCB 170	0.0001	0.0003
PCB 180	0.00001	0.0003
PCB 189	0.0001	0.0003

PCB = polychlorinated biphenyl, a = Van der Berg et al. (1998) and b = Van der Berg et al. (2006).

To achieve the compliance with cleanup and remediation levels for environmental matrix like soil for PCBs under the Model Toxic Control Act (MTCA) cleanup regulation, the soil matrix is onsidered a single hazardous substance and the target cancer risk level of one in one million (10^{-6}) is used in estimating cleanup level under method B. The 1998 and 2006 toxicity equivalent factor was used to evaluate the carcinogenicity of four dioxin like PCBs (PCB 105, PCB 118, PCB 170 and PCB 180). The cancer potency factor of the dioxin like PCBs with a mthod B cleanup level 1.3×10^{-6} mg/kg is used as shown in Eq. 2.

$$TTEC\ /TEQ = \sum Cn \times TEFn \quad (2)$$

Where, Cn is the concentration of individual congener in the soil matix, TEFn is the toxic equivalent factor of individual congener, TTEC is the total toxicity equivalent concentration and TEQ is the toxicity equivalence. The 2005 and 2006 Van Der Berg TEF was used in this work (Table 2). The PCB congeners that are affected in the whole fifteen detected are PCB 105, PCB 118, PCB 170 and PCB 180 out of these PCB 170 and PCB 180 toxicity equivalent factors has not been established in the 2006 but have values for 1998. Applying their respective TEF values for 1998 and 2006 and computing using equation (2) the cancer factor (TTEC/TEQ) for site A is $3.\ 21 \times 10^{-5}$ mg/kg, A* is 1.39×10^{-4} mg/kg for 1998 and site A is 5.10×10^{-6} mg/kg, A* is 2.43×10^{-5} mg/kg in 2006, respectively.

Again the values for both sites exceeded the method B clean up levels for dioxin- like PCBs, i.e., 1.3×10^{-6} mg/kg and therefore the sites needed massive clean-up for carcinogenic dioxin-like PCBs. It is pertinent for the relevant authorities to remediate and clean up the transformer locations to avoid further direct human contact and toxicity. This lead the researchers to carry out remediation study having seen the danger posed by this class of pollutants to human and animals. The finding now revealed both moringa seed and activated carbon can be used for PCBs remediation. However, moringa seed powder in this study serves as an excellent means of PCBs remediation in PCBs contaminated soil, which is one of major issue of soil contamination.

CONCLUSIONS

The total PCBs in the soil from two sites of the transformer installations exceeded the maximum permissible limit of 2 mg/kg, which needs cleanup. Both activated carbon and moringa seed can be used for remediation, preferably moringa seed powder showed higher efficiency probably due to increase in the surface area. Due to the economic importance of moringa seed, it can be a potential source for the remediation of soil contaminated with PCBs.

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