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Accumulation of Metals and Polychlorinated Biphenyls (PCBs) in Soils around Electric Transformers in the Central Region of Ghana

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ABSTRACT

Twenty six hot spots of PCBs pollution in the central Region of Ghana were investigated. Soil samples were collected from around twenty six transformers in the central region. The soil samples were extracted with hexane-acetone mixture (1:1 v/v) for 16 hours and the extract cleaned up with concentrated sulphuric acid (1+1), 5% (w/v) aqueous potassium permanganate and copper granules. The extract was eluate with hexane: dichloromethane (97:3 v/v), concentrated to 1 ml, transferred into a pre cleaned 2 ml vial with acetate ethyl resins and acetate ethyl added to the vial to make the total volume 2 ml. This was analyzed for PC Bsusing GC – ECD. The soils were digested withHNO₃ and HClO₄ acid, and analyzed for metals by Varian 235 AAS. The mean concentration of the $\sum PCB$ (8.17 \pm 2.96 µg/kg) was much lower than the 25 mg/kg recommended level. The $\sum PCB$ ranged between 1.32 and 12.94 µg/kg. The mean concentration (in mg/kg) of the metals were Zn (94.70 \pm 123.87) >Pb (26.68 \pm 37.27) > Cu (21.63 \pm 20.51) > Al (0.70 \pm 0.38) > Fe (0.06 \pm 0.03)> Ag (0.06 \pm 0.20). The variations in the levels of the metals were in the order Ag >Pb> Zn >Cu >Al > Fe. Very few site were found to be contaminated with metals, but the level of metal contamination was very low. There was no significant correlation between the PCBs and any of the metals. However, significant relations were observer among some metals. Correlations between the metals agreed with the results obtained by PCA. The sources of the PCBs and metal were anthropogenic.

INTRODUCTION

The polychlorinated biphenyls (PCBs) constitute a large class of compounds produced by the partial or complete chlorination of the biphenyl molecule. PCBs were first synthesized in 1864[1], but the commercial production of PCBs began in the United States in 1929 in response to the electrical industry's need. PCBs are not natural components of the environment. They were first identified as environmental contaminants in 1966, during the analysis of environmental extracts for DDT and related metabolites. The manufacture of PCBs was banned in the United States in 1977, due to potential health hazard[2].Despite its ban in Western countries, a large proportion of PCBs remain in transformers and capacitors in most developing countries [3,4]. PCB pollution in the environment is worldwide [5-9].Their unusual persistence coupled with their tendency to accumulate in living organism raises concern because of the potential adverse effects they can have on various organisms, including humans and their endocrine disrupting capacities [10-12].

The source of PCBs in developing countries has been attributed mainly to the importation and use of transformers and capacitors containing PCB transformer oils. These oils enter the environment through poor handling of damaged electrical equipment, leakages, spillage during retro filling and illegal dumping of PCB containing waste in the seas of these countries. Transformer oils have been found to contain some metals such as aluminum, copper, iron, lead,

silver, tin and zinc. For example, copper would be found in the windings and also in any bronze or brass components. Lead is found in soldered joints, connectors and other peripheral components. Iron is located in the transformer core and tank, whereas aluminum can be found in the windings, corona shields and ceramic bushings. Lugs, bolts, connectors and some peripheral components may also contain tin, silver and zinc[13-17].

Even though metals are found in all soils, it is the concentration levels that present the risk, and it is the mechanism of enrichment that defines the enriched environment as "polluted". The Earth crust comprises metals combined with various non-metallic elements, mainly oxygen and silicon, distributed over the surface of the Earth [18]. Soil contamination by metals may occur as a result of various mechanisms. Worldwide, the most important sources of metal pollution are mine tailings, smelter emissions, waste incineration and atmospheric deposition [19, 21]. But the main contribution to the trace-element load in urban top soils and dust is almost unanimously considered to be traffic emissions derived from atmospheric deposition [22].

The release of transformer oils into the environment, particularly soil, could result in PCB and heavy metal contamination. One route *via* which PCBs enter soils is by accidental spills of PCB-contaminated fluids associated with transformers and large capacitors. If the risk arising from such incidents is to be understood and effectively managed, the rate of decline in soil contamination must be accurately predicted, along with the relative significance of the different fate mechanisms involved [23]. The application of multivariable statistical methods offers a better understanding for interpreting complicated environmental data sets. These approaches have been used successfully to support the interpretation of complex field measurements and extract meaningful information from such databases [24-28].

Some PCBs have been found in the Ghanaian environment [15, 29-30]. It has been established that 455 pre-1972 possible PCB-containing transformers could be found countrywide [31]. This study therefore seeks to investigate and assess the level of concentrations of PCBs and some metals in soils around transformer sites in the Central Region of Ghana, and identify the source of these heavy metals in the soils by multivariate analysis.

MATERIALS AND METHODS

Sampling

The Soil samples were collected from the immediate surroundings of Electricity Corporation Ghana (ECG) transformers in five districts in the Central Region of Ghana in September, 2008. A total of seventy eight (78) composite soil samples were collected from twenty six different locations in the five districts. At each site three (3) composite samples were collected with a garden hand towel, at depth of 0 - 10 cm, 10-20 cm and 20-30 cm. Each composite sample weighed between 880 - 950 g, and consisted of ten core samples which were collected randomly from each site, thoroughly mixed and sub sampled to obtain the composite. The samples were placed in labeled polythene bags and sent to the laboratory. They were air dried, and each sample was sieved using 250 μ m mesh sieve. The sampling sites are shown in figure 1.

Analysis of metals

The soil samples were dried at 100 °C for 48 hours in the oven. The dried samples were passed through standard screen to remove large particles. For the digestion of the soil sample, one gram of dried and homogenized soil was weighed and placed in an acid washed Teflon vessel. The digestion was performed with a mixture HNO₃ and HClO₄ acid. The digested samples were analyzed for metals [32]. The analytical precision and accuracy of the method was accomplished by analyzing a blank and duplicate spike samples. The Varian 235 AAS was used for the metal analysis.

Extraction and cleanup of PCBs

Ten grams of air dried soil (<250 μ m) was put into a cellulose timble and covered with a layer of glass wool. This was placed in an automated soxhlet apparatus and extracted with 100 ml of hexane-acetone mixture (1:1 v/v) for 16 hours. When the extraction was completed, the inner surface of the condenser was rinsed three times with acetone-hexane and allowed to cool. The extract was evaporated and concentrated to 10 ml by rotary vacuum evaporator at 40 °C; 50 ml of hexane was added and concentrated to remove the acetone. This was then drained through a funnel containing hexane - rinsed sodium sulphate and glass wool to dry the extract. The transfer was completed by rinsing



the vessel containing the extract followed by sodium sulphate with 5 ml portions of hexane. This was evaporated and concentrated to 5 ml before clean up.

Figure 1: Map showing sampling sites in the Central Region

In order to remove co-extractive, organochlorines and other organic compounds, sulphuric acid cleanup was carried out, as the PCB is not affected by the acid. Ten milliliters of concentrated extract was transferred, with three 5 ml hexane rinses of the flask into a 125 ml separatory funnel. Twenty milliliters of concentrated sulphuric acid (1+1) was added and shaken vigorously for two minutes; gas built up was carefully discharged, and the mixture was allowed to stand for 20-30 minutes for the phase to separate. When the sample had partitioned into the discrete layers the acid layer (lower layer) was drained and discarded. The organic layer was rinsed with two 5 ml of double distilled water which was drained out and discharged.

Five millimeters of 5% (w/v) aqueous potassium permanganate was added to separator containing the acid cleaned extract, this was shaken vigorously and then allowed to stand for phase separation. Permanganate layer was drained and discarded. The organic layer was washed with two 10 ml deionized water and the aqueous layer discarded. The sample was passed through sodium sulphate in a funnel to dry the extract before the copper clean up [33-35]

The copper cleanup was necessary to remove any sulphur present which might significantly interfere with the GC/ECD analysis. Copper granules were rinsed with concentrate sulphuric acid, washed with water and then with the extraction solvent ('hexane -acetone). Approximately 0.5 g of the freshly treated copper was added to 10 ml extract, agitated and allowed to stand for 10 minutes and the extract quantitatively transferred through a funnel plugged with a glass wool to remove the copper. The process was repeated and the extract obtained concentrated by the rotary evaporator to 5ml. The extract was further cleaned with silica gel [36, 37].

The column (0.75 m, 16 mm ID) was plugged with glass wool, filled with silica gel and 2 g Na₂SO₄ placed on top. The column was pre washed with 30 ml of 3% dichloromethane in hexane (v/v). Two hundred and fifty millimeter flask was placed under the column and the solvent was drained to reach the top of sodium sulphate layer. The concentrated extract was transferred using a pipette onto the column followed by three 5 ml rinses of the sample

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flask using the pipette each time. The solvent was drained to the top of the sodium sulphate and the column eluted with 100 ml of hexane: dichloromethane (97:3 v/v). The eluate was concentrated to about 1 ml. The final clean up extract was transferred into a pre cleaned 2 ml vial with ethyl acetate resins andethyl acetate added to the vial to make the total volume 2 ml. This was stored in a refrigerator below 10 °C until GC - ECD analysis.

2.4.2 Gas Chromatography - Electron Capture Detector (ECD) Analysis

The final extract of 2.0 ml was analyzed for PCBs using a gas chromatograph equipped with $_{63}$ Ni 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 °C; oven temperature programme: 70 °C (hold 2 min) to 180 °C at a rate of 25 °C/min (hold 1min) to 300 °C at a rate of 5 °C/min. Temperature of detector was 300 °C; carrier gas-nitrogen at flow rate: 1.0 ml/min; make-up gas flow rate - 29.0 ml/min. The total runtime was 31.368 min.

The retention time for the PCBs standard were PCB 28, 12.54 min; PCB 52, 14.161; PCB 101, 17.341 min; PCB 153, 20.14 min; PCB 138, 21.027 min; and PCB 180 23.272 min. The identification of PCBs congeners in the sample was conducted by comparing the retention times of the PCBs congeners in sample to that of the PCB standards (Fig. 2). The concentrations of the individual PCBs congeners in mg/kg were calculated on dry weight basis, and the total PCBs concentration (Σ PCB) calculated by summing up the concentrations of individual PCB congeners.

Assessment of metal Contamination

Enrichment Index (or pollution index) is very useful in evaluating the degree of multiple enrichment or contamination in the environment was calculated using the relation:

Enrichment Index =
$$\sum [(Metal concentration)/(Permissible level of metal)]$$

Number of metals

The Contamination factor CF is the ratio obtained by dividing the concentration of each metal in the soil by the baseline or background value (concentration in unpolluted soil):

 $CF = [\underline{C}_{heavy metal}]/[\underline{C}_{background}]$ and Contamination degree $(\underline{C}_{deg}) = \sum CF$.

The contamination levels may be classified based on their intensities on a scale ranging from 1 to 6 (0 = none, 1 = none to medium, 2 = moderate, 3 = moderately to strong, 4 = strongly polluted, 5 = strong to very strong, 6 = very strong) [38].

The descriptive contamination classes with values of $C_{deg} < 8$ to >32 are $C_{deg} < 8$ implies low degree of contamination, 8–20 signifies medium level contamination, 20–32 implies high contamination and C_{deg} > 32 indicates very high degree of contamination [39].

RESULTS AND DISCUSSION

Recovery and reproducibility studies

The recovery of 0.2 ppm mix standards from the spiked duplicate soil samples indicated 89% recovery, which was within the 75 -125% (100% \pm 25%) acceptable criteria. The precision of the analysis calculated as the relative percent difference (%RPD) was 8% and was within the acceptable range of \pm 15%. The mean percentage recovery of PCBs from the referenced standard reference material was 79%. The chromatogram of the PCB mix standard is presented in figure 2.



The retention time for the PCB standard were PCB 28, 12.54 min; PCB 52, 14.161; PCB 101, 17.341 min; PCB 153, 20.14 min; PCB 138, 21.027 min; and PCB 180 23.272 min. The identification of PCBs congeners in the sample was conducted by comparing the retention time of the PCB congeners in the sample to that of the PCB standards (Fig. 2). The concentrations of the individual PCBs congeners in mg/kg were calculated on dry weight basis, and the total PCBs concentration (Σ PCB) calculated by summing up the concentrations of individual PCB congeners.

Statistical Analysis

The results of the analysis were evaluated statistically using Microsoft Excel 2010 and SPSS computer software package version 16. The analyses were done at 0.01 and 0.05 levels. All congener data were first tested for normality using the Kolmogorov-Smirnov test [40]. The Kolmogorov-Smirnov test indicated that the log-transformed data came from a normally distributed population.

The results of the analysis of PCBs and metals are presented in table 1.

The mean concentration and ranges of the \sum PC Band metals (Tab.2) indicated that there were variations in the levels of both the PCBs and the metals. The \sum PCB ranged between 1.32 and 12.94µg/kg and with relative standard deviation of 36.23%. The variations in the levels of the metals were in the order Ag (330%) >Pb (139.58%) > Zn (130.80%) >(Cu (94.82%) > Al (54.29%) > Fe (50%). The very wide variations are an indication of the varied sources of these metals. The sources could be from vehicular emissions, from the transformer oil, deposited into the soils together with the PCBs or originally present in the soils. The very large variation in the levels of Ag and the large number of measurement below the detection limit suggest that the soils do not contain and significant amount of Ag. The Ag detected in some of the soil might be from the transformers. The mean concentration of the \sum PCB (8.17 ± 2.96 µg/kg) was much lower than the 25mg/kg level recommended, by the Canadian Council of Ministers of the Environment (CCME), for the protection of environment and human health CCMC [41]. Thus the levels of PCBs in the soils may not pose any significant hazards.

Sample	$\sum PCB (\mu g/kg)$				Metal (mg/kg)					
_	0-10 cm	10-20 cm	20-30 cm	mean	Ag	Al	Cu	Fe	Zn	Pb
UL	7.94	5.5	7.14	6.86	nd	0.99	7.29	0.05	27.97	5.91
UA	10.77	9.71	16.14	12.21	nd	0.48	13.11	0.06	59.31	44.21
UCDS	9.58	2.2	18.18	9.99	nd	0.61	23.69	0.04	18.33	6.01
NF	5.23	6.98	11.06	7.76	nd	0.86	14.95	0.09	33.44	22.93
Р	1.68	3.78	5.29	3.58	0.09	0.23	8.33	0.02	16.5	11.75
MSS	8.99	6.26	11.62	8.96	nd	0.83	14.41	0.05	60.31	10.24
AR	11.73	11.14	11.3	11.39	nd	0.66	13.88	0.06	38.78	12.66
В	3.02	2.94	9.54	5.17	nd	0.76	23.64	0.08	52.02	8.35
EM	8.36	11.38	8.76	9.50	0.09	0.35	10.64	0.08	249.3	47.76
EC	4.24	10.06	6.13	6.81	nd	0.69	32.63	0.08	56.4	11.24
ESF	4.52	11.36	8.98	8.29	nd	0.58	18.25	0.04	102.35	184.26
А	8.48	10.42	8.87	9.26	nd	0.96	76.96	0.05	395.71	25.8
SA	11.84	16.13	10.84	12.94	0.11	0.33	6.23	0.03	31.95	8.28
SEC	10.69	5.41	5.48	7.19	nd	0.7	12.06	0.03	29.91	9.89
М	11.17	7.06	10.72	9.65	nd	1.19	2.36	0.07	23.5	8.71
MEC	15.98	6.68	3.72	8.79	0.26	0.22	13.85	0.04	107.12	17.01
AM	18.01	14.98	2.6	11.86	nd	1.08	39.83	0.13	538.97	40.02
ASS	13.26	7.42	11.51	10.73	nd	1.73	70.48	0.13	140.72	85.3
NVTI	5.5	5.31	2.56	4.46	nd	0.1	4.56	0.01	11.33	3.19
WBL	15.68	5.78	4.13	8.53	nd	0.53	7.17	0.04	38.55	5.81
WPD	16.16	12.74	9.26	12.72	nd	0.1	5.23	0.01	22.61	4.05
BC	9.54	5.05	7.58	7.39	nd	0.72	20.82	0.03	35.46	14.3
ASEC	15.26	2.38	2.2	6.61	0.01	0.72	19.53	0.04	69.03	13.97
ASSP	2.32	12.88	5.16	6.79	1.02	0.63	69.26	0.07	142.69	46.52
AN	1.5	1.24	1.22	1.32	nd	1.36	14.1	0.13	129.82	28.36
KGW	6.5	1.48	3.34	3.77	nd	0.75	19.19	0.03	30.14	17.01

Table 1: Concentration of PCBs and metals in soils around transformers in the Central Region of Ghana

nd= below limit of detection (LOD) LOD=0.001mg/kg

Table 2: Descriptive statistics

Analyte	Ν	Mean	Std. Dev.	Min.	Max.
PCB (µg/kg)	26	8.17	2.96	1.32	12.94
Metal (mg/kg)					
Ag	26	0.06	0.20	0	1.02
Al	26	0.70	0.38	0.1	1.73
Cu	26	21.63	20.51	2.36	76.96
Fe	26	0.06	0.03	0.01	0.13
Zn	26	94.70	123.87	11.33	538.97
Pb	26	26.68	37.24	3.19	184.26

There was no significant correlation between the PCBs and the metals at 0.05 level (Tab. 3). However, significant relations were observer among some metals (Tab 3).Cu showed significant positive correlation with Ag (r = 0.41, p < 0.05, n =26); with Al (r = 0.47, p< 0.05. n = 26): with Fe (r = 0.43, p < 0.05, n=26) and with Zn (r = 0.58, p < 0.01, n - 26). Also there were significant positive correlations between Zn and Al (r = 0.58, p < 0.01, n =26) and between Zn and Fe (r = 0.52, p < 0.01, n=26); Significant positive correlation was also observed between Fe and Al(r = 0.75, p < 0.01, n =26). Pb did not show significant correlation with any of the metals, indicating that significant amounts of Pb in the soilsmighte from sources differentfrom those of the other metals.

The non-significant correlation between the PCBs and the metals suggest that these metals do not have any significant effect on the persistence PCBs or on the amounts of PCBs extracted from the soils; or the non-existence of any significant relation between the levels of PCBs in the transformer oils and the metals introduced into the oils through wear and tear as the transformers function.

Table 3: Pearson's Correlation matrix for PCBs and metals in soils around transformers in	the Central Region of Ghana

Parameter	Ag	Al	Cu	Fe	Zn	Pb	PCB
Ag	1	-0.159	0.408*	0.023	0.082	0.085	-0.069
P value		0.438	0.039	0.91	0.69	0.69	0.739
Al		1	0.472*	0.753**	0.304	0.19	-0.082
P value			0.15	0	0.131	0.352	0.691
Cu			1	0.428*	0.575**	0.292	0.082
p value				0.029	0.002	0.148	0.692
Fe				1	0.523**	0.25	0.014
P value					0.006	0.218	0.945
Zn					1	0.261	0.234
p value						0.197	0.25
Pb						1	0.108
p value							0.598
PCB							1
p value							

*Correlation is significant at the 0.05 level(2-tailed) **Correlation is significant at the 0.01 level(2-tailed)

Multivariate analysis, principal component analysis (PCA) and cluster analysis (CA) and correlation matrix were used in the study to provide a better insight and understanding of the the dynamics of the PCBs and metals in the surface soils.Principal component analysis was used following standard procedure reported in literature[42-44],for evaluation fextent of metal contamination in the study area and source identification, PCA was performed on the logarithmic form of the metal data. Varimax rotation [42], was used to maximize the sum of the variance of the factor coefficients. This technique clusters variables into groups, such that variables belonging to one group are highly correlated with one another (with sum of squared loadings > 0.3) were grouped into one component. The number of components indicate the total number of possible sources of variation in the data. For the cluster analysis hierarchicalagglomerative clustering by the Ward's method wasselected for sample classification because it possesses a small space distorting effect, uses more information on cluster contents than other methods, and has been proved to be an extremely powerfulgrouping mechanism. The method was applied to normalized data using squared Euclidean distances as a measure of similarity. The results of the PCA (Tab 4) and CA (Fig 3) are shown.Only factor loadings greater than 0.3 are significant.

Table 4: Rotated component matrix of three-factor model with strong loading in bold type-face

	Component		
Parameter	PC1	PC2	PC3
Ag	-0.14	0.93	-0.06
Al	0.92	0.09	-0.07
Cu	0.55	0.64	0.26
Fe	0.89	0.06	0.11
Zn	0.54	0.27	0.54
Pb	0.86	0.24	0.40
PCB	-0.15	-0.14	0.90
Eigenvalues	2.36	1.44	1.34
% ofvariance	33.68	20.50	19.10
Cumulative %	33.68	54.17	73.27

The results of the PCA of heavy metal contents are shown in Table 4. Three principal components (PCs) with eigenvalues greater than 1 were extracted. PCA leads to a reduction of the initial dimension of the dataset to three components which explain 73.27% of the data variation. Therefore, these three factors play a significant role in explaining PCB and metal contamination in the study area.

The components one PC1, which had high loadings of Al, Fe and Pb; moderate positive loading ofCu and Zn accounted for 33.68 % of variance, and is the most important component. PC1 could be better explained as anthropogenic and lithogenic sources. Theanthropogenic source may have been derived from from transformers andvehicular emmisions. These metals showed significant correlations amongs each other (Tab 3). The second

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componen PC2, which had high positive loadings of Ag and moderate positive loading of Cu, accountedfor 20.5% of variance. PC2 can be considered as a anthropogenic and derived from the transformers. Only Cu showed significant correlation with Ag (Tab 3).The third component PC3, is highly loaded with PCBs and moderately loaded with Zn and Pb accounted for 20.1% variance; and indicating a mixed source from both lithogenic and anthropogenic inputs. The PCBscan be considered as an anthropogenic component, from transformers while Zn and Pb werefrom both lithogenic and anthropogenic sources.No significant correlations were observed amongst the elements in PC3. In general, correlations between metals agreed with the results obtained by PCA,

Based on information assessed from principal component analysis,hierarchical cluster analysis was performed [45,46]. Two mainclusters can be distinguished in the dendrogram (Fig 3), obtained from theCA performed on the analyzed parameters with Ward's methodand the squared Euclidean distance as a similarity measure (Fig. 3). Cluster 1 includes elements Ag, Fe,Al, Cuand Pb, which in the previoussection were identified as contaminants derived from both lithogenic and anthropogenic sources. Cluster 2, which containsPCBis derived from anthropogenic sources, specifically from transformers.



Figure 3: Dendrogram obtained by hierarchical clustering analysis for parameters

Contamination factor (Cf)and Enrichment Index

The calculation of Contamination factor indicated that none of the sites was contaminated with Al, Fe, Ag and Pb. However, three site AM, ASS and ASSP (11.54% of the sites) had Cf of Cu<1 implying none to medium contamination and one site A (3.85% of the sites) had moderate contamination of Cu and two sites A and ASS (7.7% of the sites) had moderate contamination Zn (Cf>2). Only site AM was moderately to strongly contaminated with Zn (Cf>3). Site A and AM had mean enrichment quotient (MEQ) or enrichment Index greater than lindicatingthat the average concentration of the metals were above permissible levels, and any enrichment may be from anthropogenic input. The degree of contamination (Cd) was low since Cd < 8.

CONCLUSION

The mean concentration of the \sum PCB (8.17 ± 2.96 µg/kg) was much lower than the 25 mg/kg level recommended the CCMCand may not pose any significant hazards. The \sum PCB ranged between 1.32 and 12.94 µg/kg. The mean concentration (in mg/kg) of the metals were Zn (94.70 ±123.87) >Pb (26.68± 37.27) > Cu (21.63±20.51) > Al (0.70± 0.38) > Fe (0.06± 0.03) > Ag (0.06±0.20). There were variations in the levels of the metals. There was no significant correlation between the PCBs the metals. However, significant relations were observer among some metals. Multivariate analysis indicated the correlations between the metals agreed with the results obtained by PCA; and the sources of the PCBs and the metals were anthropogenic. The mean enrichment quotient indicated that the average concentrations of the metals were above permissible levels but the degree of contamination was low.

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