Doyi, Israel, Essumang, David, Gbeddy, Gustav Kudjoe Seyram, Dampare, Samuel, Kumassah, Elliot, & Saka, David
(2018)
Spatial distribution, accumulation and human health risk assessment of heavy metals in soil and groundwater of the Tano Basin, Ghana.
Ecotoxicology and Environmental Safety, *165*, pp. 540-546.

https://doi.org/10.1016/j.ecoenv.2018.09.015

Spatial distribution, accumulation and human health risk assessment of heavy metals in soil and groundwater of the Tano Basin, Ghana

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Abstract

Soil serves as a vast matrix for heavy metal accumulation and subsequent redistribution to critical aspects of the environment such as groundwater. Soil pollution study is essential for sustainable human health and ecosystem protection. This study provides vital insight into the fate, accumulation, interactions, and health risk posed by heavy metals in soil and groundwater by employing geochemical accumulation index (Igeo), risk assessment models and multivariate data analysis techniques such as principal component analysis (PCA), preference ranking organisation method for enrichment evaluation (PROMETHEE) and geometrical analysis for interactive aid (GAIA). The median I_{geo} estimates show moderate to strong Pb accumulation levels whilst all the other metals indicate uncontaminated to moderate levels. The PCA output point to anthropogenic origin of Pb and Cd in the Tano Basin and surrounding communities. PROMETHEE-GAIA results indicate that Pb, Cd, Zn and Fe accumulated in the soil matrix may potentially leach into the groundwater resources. The carcinogenic lifetime risks posed by Pb, Cd, and Ni metals to adults are within the tolerable acceptable risk and thus do not present an immediate danger in the study area. Due to the significant toxicity, bioaccumulation and biomagnification properties of Pb and Cd in the environment, areas associated with significant anthropogenic activities require regular monitoring and evaluation in order to ensure that these metals are consistently below the regulatory limits. This study has further elucidated the subject of heavy metal pollution and is therefore expected to enhance sustainable protection of the environment and human health.

Keywords: Heavy metal; geochemical accumulation; multivariate analysis; risk assessment; anthropogenic origin

1. Introduction

Soil is a critical repository for numerous deleterious pollutants thereby serving as a good matrix for assessing the status quo of environmental pollution. These accumulated pollutants can then be redistributed to other compartments of the environment via the influence of anthropogenic and natural factors such as wind, vehicular movement and gravity (Gbeddy et al., 2018). Organisms especially human beings are subsequently exposed to the pollutants through dermal contact, inhalation and ingestion processes. In this context, heavy metals laden in soil pose significant health challenges due to their high toxicity, bioaccumulation and biomagnification properties (Lin et al., 2012; Wuana and Okieimen, 2011). Heavy metals may percolate into the underlining groundwater resources (Ghosh and Singh, 2005) thereby creating further challenges to water quality and sustainable water supply. Accordingly, Lin et al. (2012) noted that heavy metal pollution is a crucial global problem and therefore, merits continuous investigation in order to provide an up-to-date information required for successful risk mitigation.

Heavy metals emanate from myriad of sources in the environment including industrial waste, spillage of petrochemicals, fertilizer application, atmospheric deposition and mine tailings (Wuana and Okieimen, 2011). Areas associated with significant oil and gas exploration and drilling activities are therefore, most likely to experience challenges with heavy metals due to the generation and release of produced water and solid wastes contaminated with heavy metals (Christie, 2012; Namdari et al., 2017). The Tano Basin in Ghana is one such area with high level of commercial oil and gas industry since 2007 (Dailly et al., 2012). The main economic activities of the surrounding communities in the Basin are agriculture and fishing with substantial reliance on the groundwater as potable source of clean water. Contamination of the agricultural soil with heavy metals may therefore present significant risks to the food chain and environmental sustainability due to the direct contact of crops with soil. Although

there may exist significant environmental challenges in these communities, there has not been any corresponding comprehensive study to evaluate the extent of heavy metal pollution.

This study is therefore, aimed at ascertaining the status and impact of heavy metal pollution in the study area by determining the potential sources, inter-relationship between soil and groundwater heavy metal content, determine the scope of heavy metal accumulation, assess the potential risk posed by these pollutants to human health, and also prioritize study sites for continuous future monitoring and pollution control. In this regard, an accurate source apportionment of heavy metals is vital to minimizing potential human exposure. These objectives will be achieved by the application of the most relevant multivariate analytical tools and risk assessment models. The study is expected to provide further insight into the fate and behaviour of heavy metals thereby promoting greater protection of the ecosystem.

2. Methods

2.1 Study area

The Tano Basin situated south-east of Ghana serves a major offshore area for commercial oil and gas exploration and drilling activities. The underlying kerogen source rock types II and III; schists, phyllite and greywacks rocks form the primary geology of the Basin and thus the field's capability to produce commercial amount of oil and gas in Africa (Atta-Peters, 2014; Dailly et al., 2012; Tetteh, 2016). Communities located approximately 60km along the coast of the Basin ranging from Axim (4^o 52'6"N; 2^o14'29"W) to Newtown (5^o 6'60"N; 3^o4'60"W) constitute the study sites as shown in Fig. 1 in this research. The area experiences wet and dry annual seasons with intense agricultural activities during the rainy periods (Doyi et al., 2017).

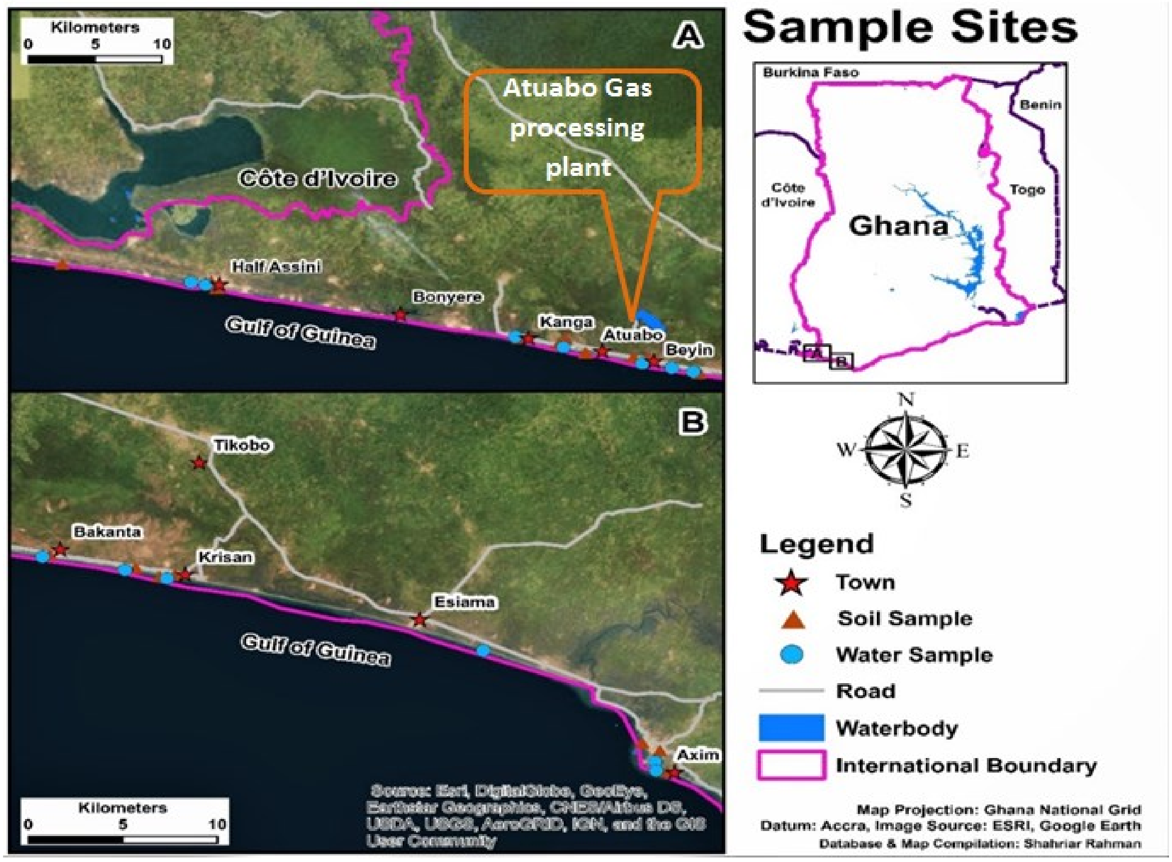


Fig. 1: Study area indicating communities of study sites

2.2 Soil and groundwater sampling

2.2.1 Soil

The sampling area was divided into four sub-areas after the initial survey using a global positioning system (GPS). Twenty seven (27) composite soil samples were collected during the rainy season from a depth of 0 – 2 cm using plastic trowel pre-cleaned with ethanol and deionised water. In order to prevent cross contamination of samples, the trowel was passed through soils adjacent to each sampling site to remove any possible effects associated with the previous site (Doyi et al., 2017; Doyi et al., 2018). Samples were oven dried at a temperature of 105°C for 4 minutes until the samples were well dried. The samples were homogenized by milling and sieving through a 2 mm pore size mesh for subsequent analysis.

2.2.2 Groundwater

Twenty (20) groundwater samples were taken from community mechanized boreholes and dug wells during the rainy season into 500mL polyethylene bottle pre-washed with concentrated HNO_3, methylated spirit and deionized water. The bottles were also rinsed with groundwater samples before filling them to the brim to prevent CO_2 trapping (Diedhiou et al., 2014), labelled, placed on ice chest and then transported to the laboratory for further analysis. Physicochemical parameters such as pH, temperature, electrical conductivity, salinity and total dissolved solids (TDS) were measured in situ using calibrated pH meter and conductivity meter (model number WTW pH 3110).

2.3 Heavy metals analysis

2.3.1 Water and soil samples

Water samples were digested by adding 0.25ml of H_2O_2 (30% v/v), 3 mL of HCl (37% v/v) and 6 mL HNO_3 (65% v/v) to 5 mL of the sample in Teflon digestion tubes. The tubes were firmly closed and placed in an ETHOS 900 microwave digester (Ackah et al., 2014). The digested samples were cooled, poured into clean 25 mL volumetric flask and then diluted to 20 ml with deionized water. On the other hand, 0.25g of powdered homogenized soil sample in Teflon beaker was digested using 6 mL (65 % v/v HNO_3) and 3 mL (37% v/v HCl) based on digestion code 308 (Ackah et al., 2014). The concentrations of cadmium (Cd), manganese (Mn), nickel (Ni), lead (Pb), and arsenic (As) in the digested samples were determined using Varian 240FS atomic absorption spectrometer.

2.3.2 Quality Assurance/Quality Control

High purity analytical grades of chemicals and reagents were used during sample preparation and analysis. Blank solution was analysed after every 10 samples measured. Samples were analysed in triplicates. The recovery of three concentration levels of each analyte spiked samples was determined, and ranged from 90 to 103% with minimal percentage error. The

results from the analysis of IAEA-SOIL-7 certified reference material were within 95% confidence level. The regression coefficients for the calibration curves were approximately 1.0.

2.4 Data analysis

The data generated were analysed using various multivariate data analysis techniques such as principal component analysis (PCA), Geometrical Analysis for Interactive Aid (GAIA) and Preference Ranking Organisation Method for Enrichment Evaluation (PROMETHEE) using StatistiXL Version 1.8 and Visual PROMETHEE Academic Edition Version 1.4.0.0 in line with research objectives. The spatial distribution of heavy metals was determined using Minitab version 17.2.1

2.4.1 Multivariate data analysis

The application of multivariate data analysis techniques in environmental pollution investigation has been necessitated by the increasing complexities in environmental data. The application of these techniques in environmental research is still evolving and the potentials are enormous. Multivariate analysis techniques essentially maximise relevant pollutant information whilst reducing inherent complexities in observed data (Miller and Miller, 2010). In this regard, multivariate analysis is indispensable in characterizing pollutant behaviour patterns, source apportionment and ranking (Ayoko et al., 2007).

2.5 Pollution status and health risk assessment

As a result of the direct contact of food crops with soil and the high probability of heavy metals transfer into the food chain, the degree of heavy metal pollution in soil were assessed using the geoaccumulation indexes (I_{geo}) (Gao et al., 2014).

$$I_{geo} = log_2 \frac{C_n}{1.5B_n} \quad (1)$$

Where C_n is the measured concentration of heavy metal (n) in the soil, B_n is the geochemical background value of element n in the soil and 1.5 is the background matrix correction factor due to lithogenic effects. The B_n of elements in this study were assumed to be the natural worldwide distribution of elements in shales (Turekian and Wedepohl, 1961) due to the lack of site specific values. The seven classes of I_{geo} are: < 0 (uncontaminated), 0 – 1 (uncontaminated to moderately contaminated,) 1 – 2 (moderately contaminated), 2 – 3 (moderately to strongly contaminated), 3 – 4 (strongly contaminated), 4 – 5 (strongly to extremely contaminated), and > 5 (extremely contaminated) (Gao et al., 2014; Jia et al., 2018)

The non-carcinogenic chronic daily intake by ingestion ($CDI_{ingestion}$), inhalation ($CDI_{inhalation}$), dermal (CDI_{dermal}) and carcinogenic chronic daily intake of heavy metals by ingestion (CDI_{ing-ca}), inhalation (CDI_{inh-ca}) and dermal ($CDI_{derm-ca}$) contact pathways were estimated using Equations 2 to 8. The hazard quotient (HQ) for each element for each exposure pathway was obtained by dividing the CDI by the corresponding reference dose (R_fD) whilst for carcinogens the CDI was multiplied by the appropriate slope factor (CSF) in order to assess the risk of cancer. The extent of adverse effects from each heavy metal is assumed to be the sum of the simultaneous exposure via all three routes and thus the hazard index (HI) is estimated as the sum of HQ or non-cancer risk. If HI is greater than 1 (HI > 1), then there exists a significant probability for non-carcinogenic effect to occur (USEPA, 2002; Zheng et al., 2010).

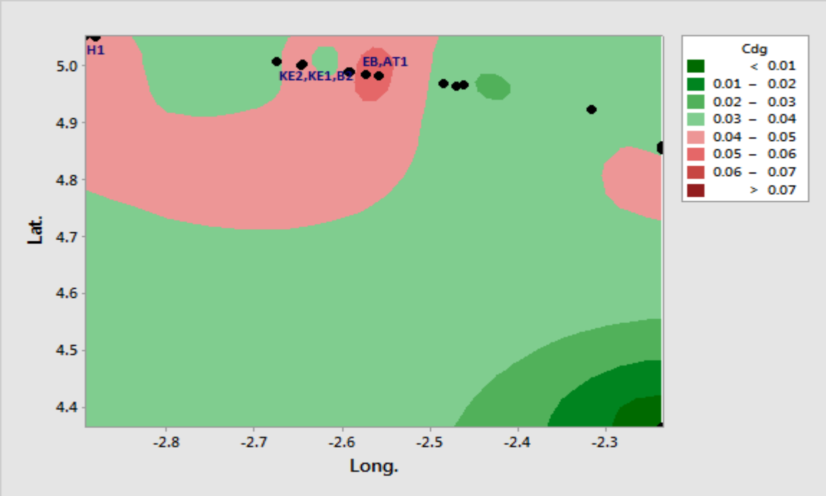
3. Results and discussions

3.1 Spatial distribution of heavy metals

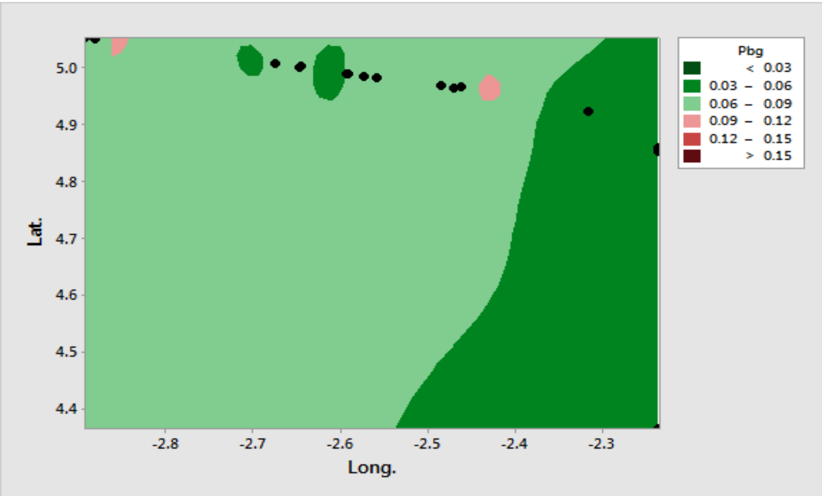
The mean concentrations of heavy metals in soil were in decreasing order of Fe > Zn > Cd > Mn > Pb >Ni. The results however indicate that in comparison to the USEPA regulatory standards for agricultural soil, all the analytes in exception of Cd were relatively low. The

spatial distributions of these analytes are represented in Fig. S1. Cadmium exceeded the regulatory value of 0.48 mg/kg by 0.07 mg/kg. In comparison to studies conducted elsewhere, the levels of most metals quantified were lower. The concentration of Ni is however, comparable to the levels estimated by Grzetic and Ghariani (2008) in soil from the central zone of Belgrade. The spatial distribution of these analytes in soil calls for a critical evaluation of groundwater samples due to the high possibility of migration via leaching.

The average concentration of heavy metals in groundwater samples are generally lower than WHO 2008 maximum contaminant levels (MCLs) in exception of Cd and Pb. The concentrations vary as follows Fe > Pb > Cd > Ni > Zn > Mn. Cadmium and lead however, show approximately 100.0% and 59.0% increases above the WHO MCLs of 0.003mg/L and 0.05mg/L respectively. In this regard, the spatial distributions of Pb and Cd in the study are shown in Fig. 2, which indicates significant levels of Cd in the two communities, Atuabo (AT1) and Ekebaku (EB). The siting of a natural gas processing factory at Atuabo as shown in Fig. 1 may be a key contributory factor since according to Al-Turki and Helal (2004) and Ren et al. (2006), Pb and Cd are anthropogenic origin metals. These observations are highly crucial due to the deleterious impact of these metals such as carcinogenic effects, long biological half-life and tendency to replace Zn biochemically. Pb mimics Ca and has the natural tendency to accumulate with age in bones, the aorta, kidneys, the liver and the spleen. The long term exposure to Cd causes kidney and liver toxicity effects and high blood pressure (Momodu and Anyakora, 2010; Rajappa et al., 2010). In order to maximize the chemical information from these preliminary observations, the research data was subjected to multivariate data analysis.



(a)



(b)

Fig. 2: Spatial distribution of Cadmium (Cd_g); (a), and Lead (Pb_g); (b) metals in groundwater (Note: The concentrations of lead and cadmium in groundwater samples and the corresponding GPS locations (Table S1) were used to generate these diagrams using Minitab software version 17.2.1)

3.2 Multivariate data analysis

3.2.1 PCA of soil samples

The results of a 26x6 soil data matrix PCA analysis indicates that three principal components (PC) are significant with an Eigen value ≥ 1 (Kaiser and Hunka, 1973), contributing 71% of the variance in the data as indicated in Fig. 3 and Fig. S3. The biplot of PC1 and PC2 as

shown in Fig. 3 indicates that Ni and Zn are highly correlated signifying a potential common source most probably lithosphere or parent rocks. Moreover, Cd and Fe are also correlated whilst Fe and Ni are orthogonal implying they emanate from independent sources. The cluster of large number of objects or communities with similar heavy metal distribution patterns further illustrates that anthropogenic source Cd is significantly associated with communities in close proximity to the natural gas processing factory as well as offshore oil and gas rigs in the Tano Basin. Furthermore, few communities with worse case heavy metal accumulation in this cluster can be selected for subsequent assessment of the potential impact of the oil and gas industry on the soil ecosystem.

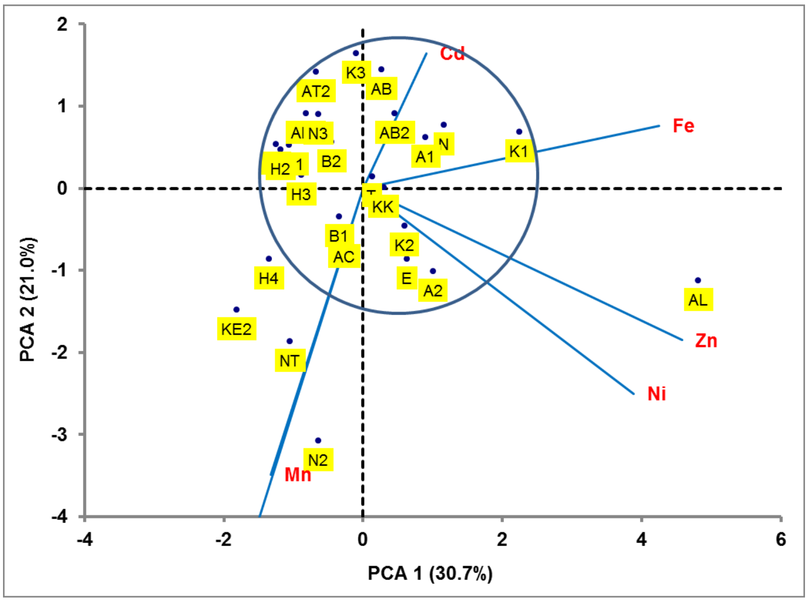


Fig 3**:** Biplot of PC1 and PC2 for soil samples

3.3.1 PCA of groundwater samples

The 17x11 groundwater data matrix was subjected to standardization prior to PCA analysis due to the differences in the units of variables. Using an Eigenvalue ≥ 1, five principal components as shown in Fig. 4 and Fig. S4 have been determined as significant with 82% of the total variance in the data. From Fig. 4, groundwater physicochemical properties such as TDS, Ko, Sal and pH correlates positively with Zn, Cd and Pb (denoted as Zng, Cdg and Pbg

respectively) in Beyin (B1), Kengen (KE), Axim (AX), Half-Assini (H1) and Atuabo (AT1) study areas. This underscores the critical role and influence of these physicochemical parameters on the distribution of these heavy metals which may be related to the overlaying soil matrix. The role of accumulated heavy metals in soil on this observed analyte patterns needs further evaluation. It moreover, indicates the commonality in the anthropogenic source of these three elements.

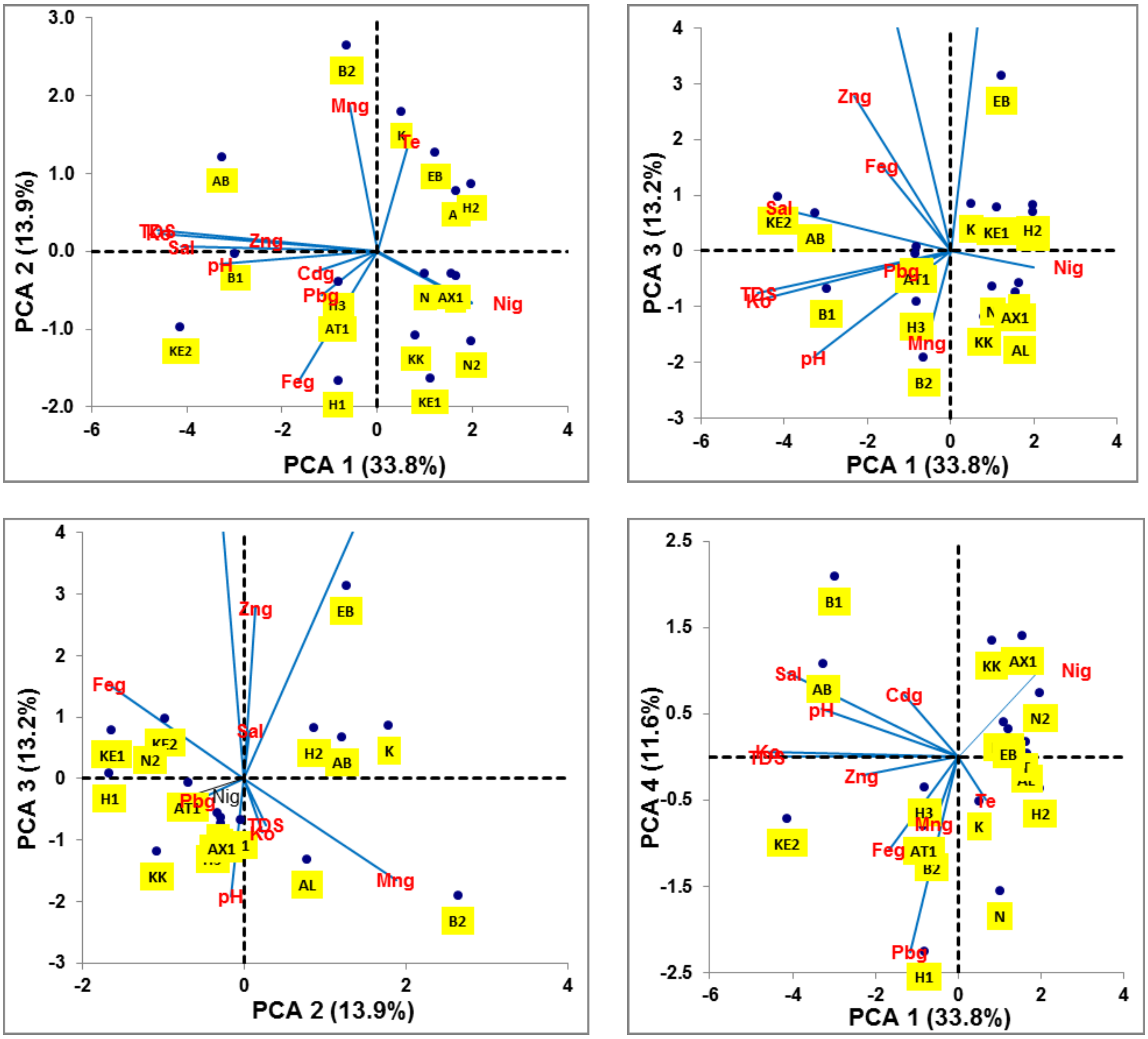
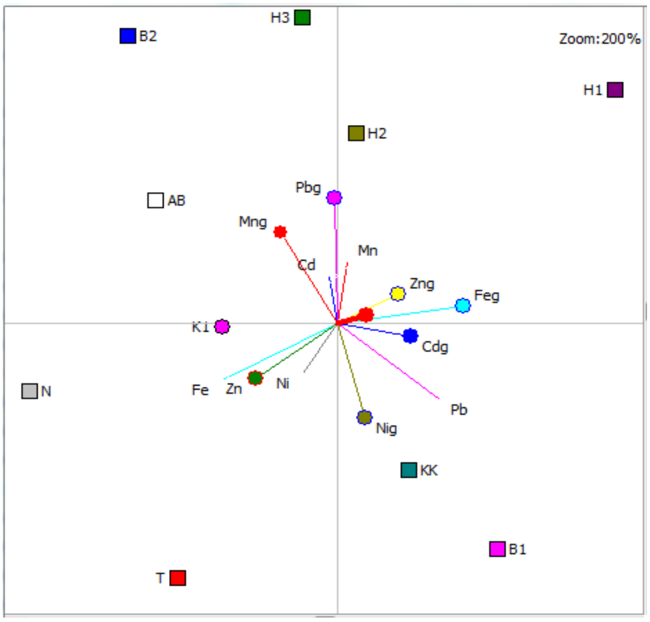


Fig. 4: Biplot of first four principal components for groundwater samples

3.4.1 PROMETHEE-GAIA of soil and groundwater samples

The special relationship between soil and groundwater heavy metal contents was examined using the PROMETHEE-GAIA analysis by considering a data matrix consisting of 14 actions and 12 criteria. Two modelling scenarios were used during the analysis. In the first instance, all the criteria were maximized, assigned equal weighting and subjected to V-shaped preference function under complete ranking PROMETHEE-I and the outputs are shown in Fig. 5. Secondly, all the criteria were maximized, subjected to V-shaped preference function with Cd and Pb given a weighting of 5 due to their higher deleterious impact on human health and the ecosystem and the results are depicted in Fig. 6. The GAIA plots were interpreted in accordance with the guidelines published in Espinasse et al. (1997).



(a)

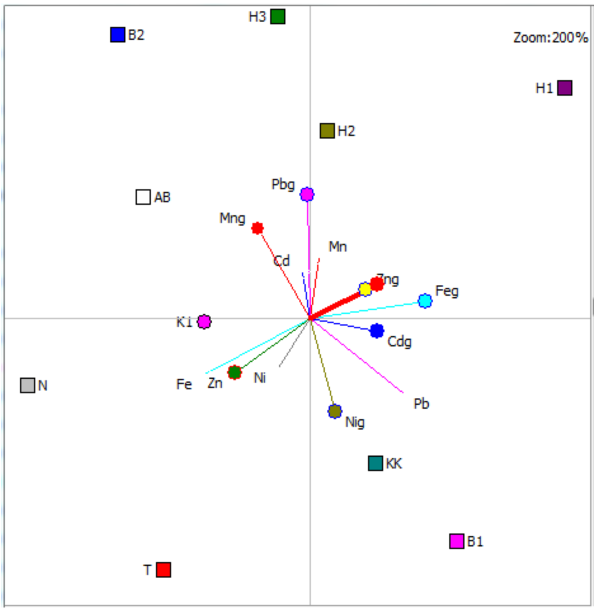
Rank	action	Phi	Phi+	Phi-
1	K1	0,3429	0,6378	0,2949
2	N2	0,1795	0,5481	0,3686
3	H3	0,1058	0,4968	0,3910
4	KK	0,0897	0,5000	0,4103
5	AB	0,0577	0,4776	0,4199
6	KE2	0,0160	0,4615	0,4455
7	B1	-0,0256	0,4455	0,4712
8	B2	-0,0353	0,4423	0,4776
9	H2	-0,0577	0,4167	0,4744
10	KE1	-0,0865	0,3974	0,4840
11	H1	-0,0897	0,4038	0,4936
12	N	-0,0994	0,4006	0,5000
13	T	-0,1699	0,3654	0,5353
14	AL	-0,2276	0,3526	0,5801

(b)

Fig. 5: The GAIA biplots (a) and PROMETHEE-II (b) for scenario 1

From Fig. 5(a), the GAIA biplot represents 46.8% of the information in the data. Zinc and iron play significant role in the metals distribution pattern due to their close proximity to the decision axis, pi (π). In exception of Ni and Mn, the plot shows that Pb, Cd, Zn and Fe accumulated in the soil matrix may be critical leaching points for these metals into

groundwater due to their opposite positions in the diagram, thus indicating an inverse relationship in heavy metal concentrations between the two media. The preference for future monitoring of heavy metals in the study area is as follows Krisan (K1) > Nyale Kplole (N2) > Half-Assini (H3) from Fig. 5(b). Groundwater resources play essential roles in the daily lives of people; however, toxic pollutants such as Cd and Pb hinder the sustainable use of this natural resource and thus the need for regular monitoring in order to guarantee public health and safety.



(a)

Rank	action		Phi	Phi+	Phi-
1	K1		0,3381	0,6522	0,3141
2	KE2		0,2260	0,5897	0,3638
3	H3		0,1683	0,5561	0,3878
4	KK		0,1090	0,5321	0,4231
5	H2		0,0673	0,4968	0,4295
6	T		0,0304	0,4904	0,4599
7	B2		0,0016	0,4712	0,4696
8	KE1		-0,0112	0,4615	0,4728
9	H1		-0,0321	0,4583	0,4904
10	N2		-0,0513	0,4535	0,5048
11	N		-0,0689	0,4375	0,5064
12	AB		-0,0994	0,4183	0,5176
13	B1		-0,1731	0,3894	0,5625
14	AL		-0,5048	0,2276	0,7324

(b)

Fig. 6: The GAIA biplots (a) and PROMETHEE-II (b) for scenario 2

From Fig. 6, the relationship and distribution patterns of heavy metals in both soil and groundwater are analogous to that of Fig. 5. The preference ranking of heavy metals in the study area, however, differs slightly in the order: Krisan (K1) > Kengen (KE2) > Half-Assini (H3). The outcomes of the two scenarios clearly indicate that Kristan is prone to heavy metal pollution and therefore requires regular monitoring and evaluation. The relative location of these sites to various oil and gas rigs off-shore in combination with natural and anthropogenic redistribution factors may be crucial factors underlying the distribution of heavy metals.

3.3 Heavy metal accumulation and human health risk assessment

The geo-accumulation index (I_{geo}) for heavy metals in soil of the study area ranged from 2.5 × 10^{-6} to 6.23 with median I_{geo} levels ranging from Pb (2.03) > Cd (0.31) > Zn (7.0 × 10^{-4}) > Mn (9.3 × 10^{-5}) > Ni (3.0 × 10^{-5}) > Fe (8.6 × 10^{-6}). The results show that in exception of Pb, which exhibits moderate to strong contamination levels the remaining metals manifest uncontaminated to moderate levels. This result attests to earlier observations which show that Pb and Cd are the dominant heavy metals in the study area corresponding to significant anthropogenic sources of these pollutants.

The results for the adult human health risk assessment posed by heavy metals exposure in soil as shown in Table S2 indicates that the non-carcinogenic hazard indices (HI) the heavy metals ranged from 1 × 10^{-6} to 1 × 10^{-3} with Cd as the highest and Ni as the least. In comparison to other studies, the HI estimated in this study is generally low. However, the HI of Ni is comparable to that reported by Grzetic and Ghariani (2008). Carcinogenic lifetime risks posed by the deleterious metals Pb, Cd, and Ni are 3 × 10^{-9}, 1 × 10^{-6} and 4 × 10^{-9} (Table S2) respectively for adults and 2 × 10^{-12}, 2 × × 10^{-6} and 5 × 10^{-9} (Table S3) respectively for children. These estimates are within the tolerable acceptable risk of 1 × 10^{-6} to1 × 10^{-4} for these metals (Jia et al., 2018). The risk assessment further shows that Cd presents the major overall risk in this study in agreement with the I_{geo} results. It must however be stated that due to the lack of available estimates for some parameters in the risk assessment equations, the threat posed by all critical metals could not be assessed. Cadmium is a key deleterious heavy metal known for causing higher incidences of lung cancer and damage to internal organs in the minutest quantities. In this context, constant monitoring of soils and groundwater in the study area because of increased industrial activities such as oil and gas exploration is warranted in order to ensure that the levels are always below the maximum exposure thresholds.

4. Conclusion

Heavy metals in soil and groundwater pose critical challenges to the survival of microorganism, human beings and sustainable ecosystem. In this regard, information on the source apportionment, spatial distribution, geochemical accumulation and human health risk are essential for safeguarding the safety and well-being of the environment. The results indicate that the presence of Pb and Cd in the study area can be attributed to anthropogenic activities. This study has shown that the application of multivariate analysis is highly relevant in the environmental studies of the distribution patterns of heavy metals in soil and groundwater media. This study has further elucidated the subject of heavy metal pollution and is therefore expected to contribute to the greater protection of the environment and human health.

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Conflict of interest statement

The authors can testify that there is no real or perceived conflict of interest such as personal, financial and connection to the person(s) or institution(s) that may have impacted negatively on the outcome of this research.

Acknowledgement

The authors will like to acknowledge the significant role of the Ghana Atomic Energy Commission (GAEC) is highly recognized for instrumentation.