Annals of Geographical Studies
Volume 2, Issue 4, 2019, PP 22-35
ISSN 2642-9136

Physicochemical Properties and Heavy Metal Content of Mpraeso and Vume Clays of Ghana

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ABSTRACT

Earthenware clay deposits used in producing ceramics may be a potential source of heavy metals that could leach into food prepared in them. However, little attention has been focused on the heavy metal concentrations in these clays. The study was carried out to determine the physicochemical properties and heavy metal concentration of clays in two popular traditional earthen ware production centres in Ghana; Mpraeso and Vume which produce lot of traditional pottery for food preparation in most homes in Ghana. Munsell Colour Chart was used to determine the colour of the clays. While the pH and EC of samples were measured using pH and EC probe. Heavy metals (Cd, Pb, As, Fe, Zn, Co, Ni, Mn, Cr and Cu) were analysed by atomic absorption spectrometry. Both clays were of hue properties with Mpreaso having a range of 2.5-5YR described as pale to light yellow whiles Vume hue range was between 7.5-10YR described as orange to dull yellowish colour by the Munsell colour chart. The grains of both localities were very fine of less than 4µm. The pH results of three types of pottery materials (raw clay, additive and mixed clay) showed most samples been slightly acidic with a few alkaline ones ranging from 5.2-8.9. The EC of these pottery materials also ranged from 2.2-653. Results of AAS on metals analysed were found to be within the FAO/WHO and EU limits except Cd (4.4- 11.2) in Vume clays and Ni in Mpraeso (52-115) and Vume (61.4-93.3) pottery materials. With the exception of Cd and Ni which need some remediation, the quality of pottery materials used in Mpraeso and Vume were found to be generally good for earthenware production.

Keywords: *Atomic Absorption Spectrometry Analysis; physicochemical properties; heavy metal concentration; clays; ceramics; pottery; Mpreaso; Vume.*

INTRODUCTION

Clay is used as a mineral, particle size and a rock term (Bergaya and Lagaly 2013). According to the Joint Nomenclature Committees (JNC) of the Association International pour l' Etude des Argiles (AIPEA) definition,clay is a naturally occurring material composed primarily of fine-grained minerals, which is generally plastic at appropriate water contents and will harden when dried or fired (Guggenheim and Martin1995). The particle size limit in geology, sedimentology and geoengineering is <4 µm, <2 µm in pedology (Moore and Reynolds 1997) and in colloid science it is <2 µm (Bergaya and Lagaly 2013). The JNC definition and the geology, sedimentology and geoengineering particle size classification were used for this study.

Clay as an industrial raw material may not be high on the radar of valuable minerals but affects life on earth in far reaching ways because of its use in pottery making and other ceramic products like bricks and tiles. From primeval times people all over the world have used clay to mould pots, bowls, vases, floor tiles and a wide range of other products used on regular basis. According to De Guire (2014), as early as about 400 B. C. earthenware pottery was produced on a mass scale in many parts of the world. This wide, purposeful use of pottery supported the local agrarian community, and as such was heartily encouraged (Walsh and Friztlan, 2014).Ghanaian cultural heritage has seen increasing use of grinding earthenware pots for cooking, eating of meals, decorations in many homes and eateries for centuries and for exportation. Ghanaian users of earthenware believe that foods prepared in the traditional pots although takes a longer period, taste better and is much healthier than those cooked in the metal pots. A growing number of Ghanaians also prefer to be served certain traditional meals in the local earthenware pots.

The focus on clay science is usually on its mineralogy, while its colloidal, physico-chemical and heavy mineral aspects are glanced over, if not ignored (Bergaya and Lagaly 2013). And until 1960, clay deposits in Ghana were routinely studied only during occasional geological mapping (Kesse 1985). Though physical and chemical changes occur in the mixed and fired clays in the production process of pottery, glazed clay food wares are known to leach heavy metals into food stored in them sometimes at toxic levels because glazes are said to contain heavy metals like Pb and Cd (e.g. Aderemi et al. 2017; Belgaied, 2003; Dessuyet al. 2011; Halefoğlu et al. 2006; Henden et al. 2011; Nsengimana et al. 2012; Omolaoye et al. 2010).Omolaoye et al. (2010) stated that all ceramic wares contain heavy metals in varying concentrations with nearly 60% of ceramic wares having Pb concentration higher than 500 µgg-1 and that of cadmium levels generally been low.Aderemi et al. 2017 also found that glazes of ceramic food wares and ceramic products leached varying concentrations of Pb, Cd, Zn, As, Cu, Cr, Mn, and Fe into food. The risk of arsenic poisoning by glazed and non glazed potteries was said to be high enough to be of concern by Henden et al. 2011.Nsengimana et al. 2012 found Pb, Cd and Fe in considerable amounts exceeding the safe limits in food established by WHO when foods were cooked in unglazed traditional clay pots. Çiftçi and Henden (2016) also discovered that glazed potteries released arsenic at lower concentrations compared tononglazed potteries.

Though several investigations have been carried out on heavy metal leach ability of pottery products (glazed and unglazed) only a few works have been done on the heavy metal content of pottery raw materials (clays) of unglazed pottery and their propensity to retain and leach metals into foods. Ogah and Ikelle (2015) analyzed heavy metals in edible clays sold in Enyigba village in Abakaliki Ebonyi State, Nigeria and found Ni level to be higher than WHO standards. Aderemi et al. 2017 detected heavy metals in foods of ceramic products to be domiciled from the clay materials used for the ceramics. Samlafoet al. (2016) screened for potentially harmful heavy metals such as Pb, Cd, As and Hg in clay deposits at Vume and found that with the exception of As level which needs some remediation, the quality of clay at Vume is generally good for the production of earthenware products.Once heavy metals are ingested through contaminated food, they pose all sorts of dangers to human health including hepatic diseases, aneamia, nausea etc (Arora et al. 2008; Calaceet al. 2002; Olayinka et al. 2017; Yusufet al. 2009; Zhao et al. 2012);if the recommended daily allowances are exceeded (Dawaki et al. 2013).It is evident that there is less monitoring and information reported on heavy mineral content of clays used in pottery production as well as heavy metals leachability in foods of these unglazed traditional pottery products in Ghana. With the growing usage of traditional pottery in the preparation and serving of foods in homes and local restaurants called 'chop bars', the study sort to investigate the physicochemical properties and heavy metal content of the clays used for pottery production and the leachability of heavy metals into foods prepared in the pots in two popular traditional pottery production centres; Mpraeso and Vume in Ghana.

Clay mineralogical/elemental composition are analysed by destructive (Neutron Activation Analysis, Inductively Coupled Plasma-Optical Emission Spectrometry and Atomic Absorption Spectrometry) (Ogah and Ikelle2015; Samlafo et al. 2016; Viscosi-Shirley et al. 2003) and non-destructive methods (Light Microscope, X-ray fluorescence, X-ray diffraction, Scanning Electron Microscopy etc) (e.g. He et al. 2017; Nakao et al. 2014; van Dongen et al. 2011). The destructive methods are useful in detecting tracer elements whiles the non-destructive methods are very suitable for detecting major elements in clay materials. **A comprehensive study was carried to determine the heavy metal/clay minerals of the clay materials, the leachability of the unglazed pots using** Atomic Absorption Spectrometry **(AAS) and** Scanning Electron Microscopy (SEM) methods. In view of the volume of data, it is only ASS results of clay materials that are presented in this manuscript. Other components of the results; SEM of clay minerals and AAS of leachability of the pots are submitted elsewhere for publication.

Materials and Methods

Geographical Location, Geology and Soils of Study Areas

The two study areas Mpraeso and Vume are well known localities of traditional pottery industries in Ghana. Mpraeso is the capital district of Kwahu South in the Eastern Region of Ghana and it lies between latitude 6° 30ǀ N and 7°N and longitude 0° 30ǀ W and 1° W(Fig.1). It has a population size of 11,190 as at 2013. The altitude of Mpraeso is 308m with a total area of 1462km^2(Fig.1).

Mpraeso has geology of Upper Voltaian sandstones consisting of coarse and fine-grained massive sandstones that are thin bedded, flaggy, impure, ferruginous or feldspathic and locally inter-bedded with shales and mudstone (Fig.1). The sandstones are found along the boundary margins whiles shales and mudstones outcrop within the central part of the District from below the sandstone bed. Clays used in Mpraaso pottery industry are however mined from Nkawkaw and Aman from whose soils are derived from weathered phyllites, schists, tuffs and grey wackes of the Birimian system (Fig.1). The Birimian formation is rich in iron and most of the economically mined mineral resources of the country (Dickson and Benneh, 1995; Kesse, 1985).

Vume is in the South Tongu District in the Volta Region near Sogakope the district capital (Fig.1). Vume is sited at the western bank of the Volta estuary and across to the eastern bank is Sogakope. South Tongu District lies between latitudes 6° 10ˈ and 5°45ˈ N and longitudes 30°30ˈand 0°45ˈ E.

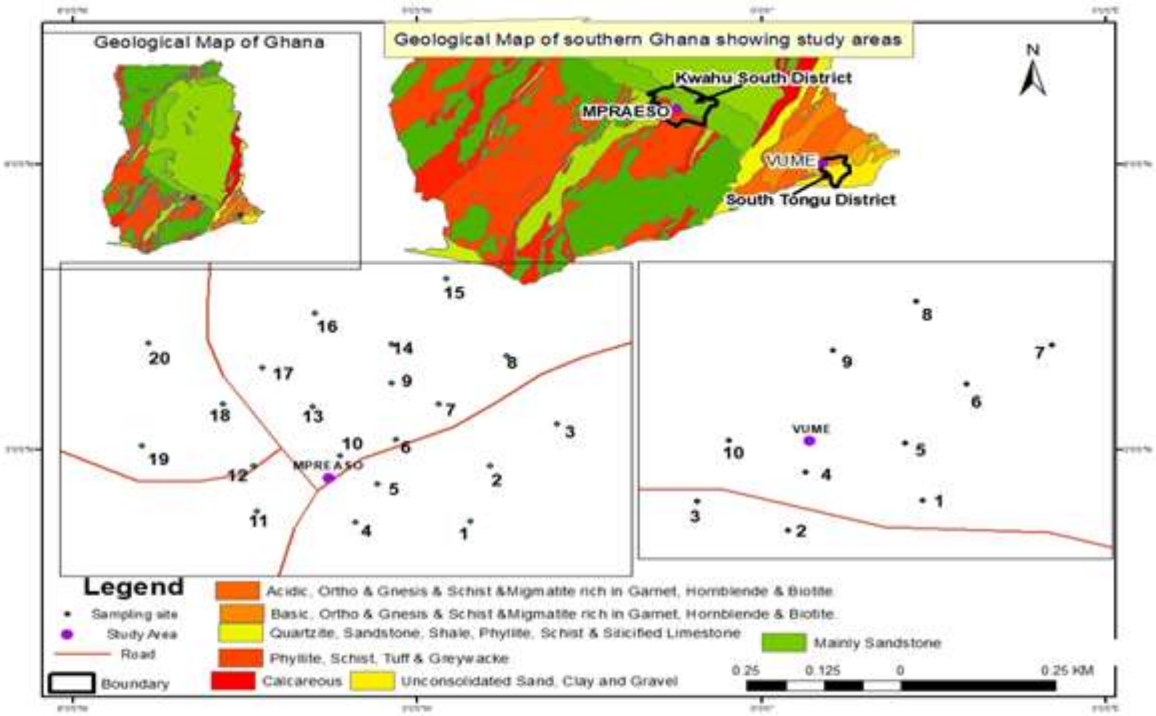

Figure1. *Map of study areas showing sampling sites*

The South Tongu District was carved from Tongu District which was established prior to1988 (Ghana Statistical Service 2014). According to the 2010 Population and Housing Census Report, South Tongu District had a population of 87, 950. The population density is high in communities along the major roads and in few other communities where the road network is good. However, the population is sparse in the north-eastern and south-eastern parts of the District. The district is largely rural with majority of its population (87.1%) living in the rural localities.

South Tongu District is located in the southern part of the Lower Volta Basin and bounded to the north by the Central and North Tongu Districts, to the east by the Akatsi South District, to the west by the Ada East District of the Greater Accra Region and to the south by the Keta Municipality. South Tongu District occupies a total land area of 643.6 km^2. The District is generally low-lying by virtue of its location within the coastal savannah plains but rises gradually to a height of 75m above sea level. The underlying rocks of Vume are metamorphic in origin of the Dahomeyan system. Rocks of the Dahomeyan composed of quartz, feldspar, epidote, hornblende, garnet and mica minerals (Kesse, 1985). The major soils formed over these geological formations are the Ziwai-Zebe complex soils, Tondo-Motawme complex and Agawtaw-Pejeglo complex soils which are formed over the Dahomeyan acidic gneiss rocks (Fig.1).

The district is endowed with large clay deposits at Lolito, Vume and Sokpoe communities, which are predicted by geologist to last for over 100 years if mined commercially in a sustainable way. Though, the manufacture of brick and tiles from clays used in the construction industry has not been fully exploited, pottery products like earthenware, flowerpots and ornaments are produce on a medium to large scale in these communities. The high mineral volume of clay deposits can also be developed into paints and other chemical products.

Clay Deposits and its Applications in Ghana

Clay mineral deposits spread across the entire land area of Ghana with significant amount of clay reserves in commercial quantities (Fig.2). The clays are alluvial materials derived from geomorphological and geological processes of rock weathering, erosion and sedimentation. The expected life span of most deposits in Ghana if mined sustainability ranges from 100 to over 7,00years whiles the regional deposits of the study towns are between 1 – 3,000years (Table 1 and 2) (Kesse 1985). Though research has not indicated the clay reserves in the study areas (Mpraeso and Vume), Tables 1 and 2 show the regional clay reserves for neighbouring towns where the study areas are located. The clay materials of Ghana differ in colour and other properties such as texture, cohesion, viscosity and fired strength (Asamoah 2018).

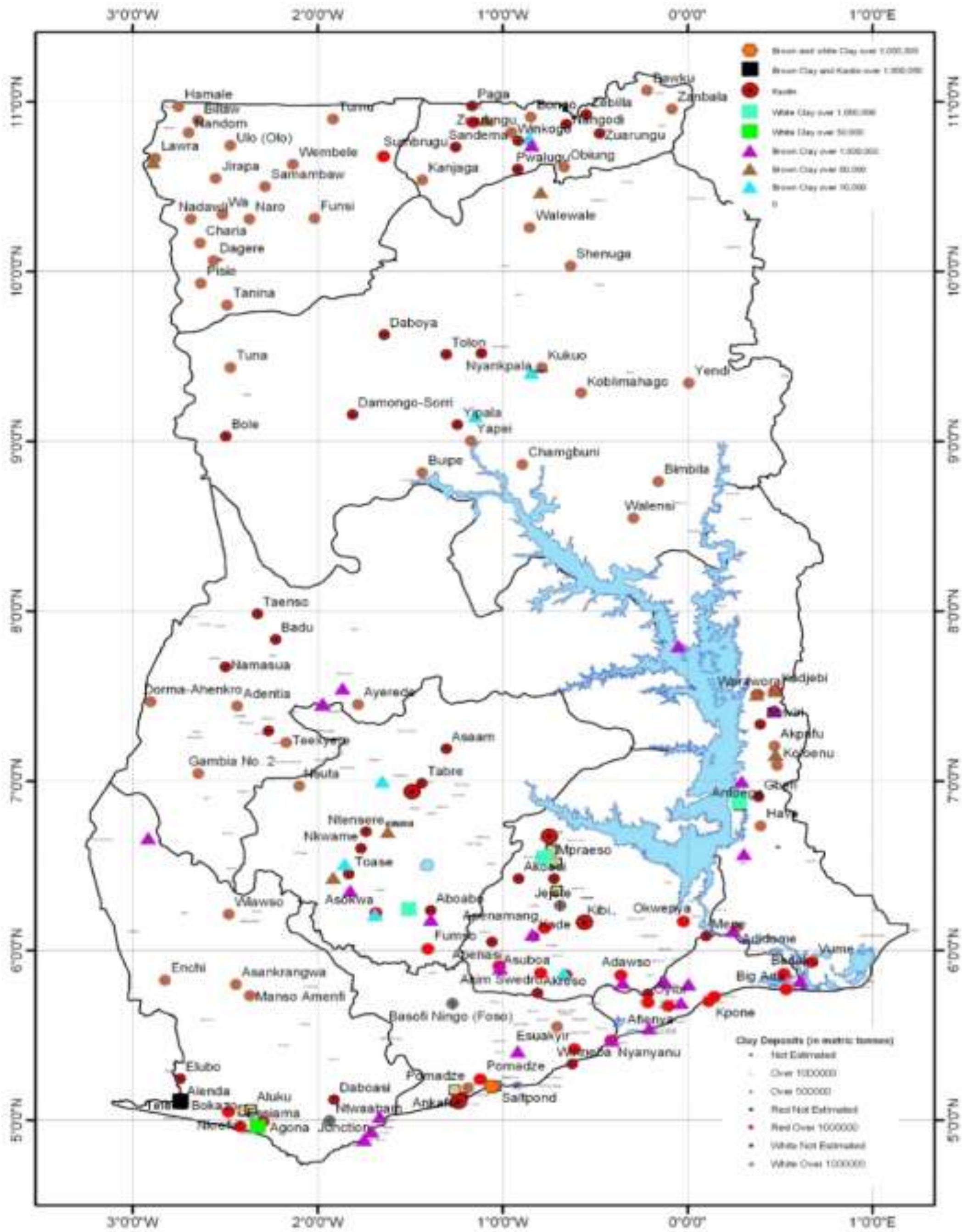

Figure2. A map of clay mineral deposits in Ghana

Source: *Geological Survey Department, Ghana*

The ubiquitous state of this earth material accounts for the widespread pottery activities across the country among different ethnic groups whose different cultures explain the diversity in pottery types in the country. The production of pottery and red bricks, geophagy, sculpture, rituals, plastering and murals are just some of the ways in which the clay materials are used in Ghana (Asante et al 2015; Yaya et al 2017). Though there are huge clay reserves in Ghana, the clay industry's production is limited mostly to the local markets in the country with marginal exports. Ghana as a country has not explored opportunities of the ceramic industry compared to countries like India, China, Brazil, Italy and others where the industry contributes immensely to their GDP. Also, the physicochemical-mineralogical properties and suitability of the clay materials for the various applications have not been fully explored and these account for low applications of the clay materials in the country.

Sample Collection

In all there were 30 sampling sites of potters, 20 sites in Mpraeso and 10 sites in Vume (Fig.1). The sites were chosen based on potter's availability and willingness to assist in the collection of the various data needed for the study. Clay samples were collected using a wooden ladle, composited and a sample taken for laboratory analysis. Sampling equipment were washed with distilled water after each sample collection to prevent contamination of other samples (Inobeme et al. 2014). Samples were put in zip locks and placed in iced-chest with a unique identity. A total of fifty-seven (57) samples consisting of raw clays, mixed clays and clay additives/temper were collected from potters of both study areas for physico-chemical and heavy metal content analyses. Mpreaso samples were 33; consisting 11 samples each of raw clays, mixed clays and clay additives (Tables 1, 2& 3) whereas Vume samples were made up of 9 raw clays, 7 mixed clays and 8 clay additives making a total of 24 samples (Tables 4, 5&6). Sample numbers of mixed and clay additives were lesser than raw clays in Vume because potters did not have those clay samples at the time of sample collection.

Table1. *Clay deposits in Eastern Region of Ghana*

Area	Location	Reserve (tonnes)	Approx Life Span (in yrs)	Area	Location	Reserve	Approx Life Span (in yrs)
Nkwakwa	Adihima/Asuoya	2,240,099	69	Asamankese	Asamankese	840,000	26
	Abepotia	7,614,793	234		Apinmang	2,801,250	86
	Framase	41,687	2	Akim Oda	Akim Swedru	33,173,335	1,021
Kibi	Tamfoi	1,285,084	40		Akim Awisa	1,285,553	140
Anyinam	Moseaso	444,000	14		Akim Abonase	4,561,000	131
	Abomosu	4,081,434	126	Akwapim	Adawso	1,027,000	32

Source: *(Kesse, 1985)*

Table2. *Clay deposits in Volta Region of Ghana*

Area	Location	Reserve	Approx Life Span (in yrs)	Area	Location	Reserve	Approx Life Span (in yrs)
Ho	Adidome No 1	7,755,319	39	Kudzra	Kalakpa	501,440	15
	Adidome No 1	469,800	75		Tuwotsive	1,944	1
Anfoega	Tangidome	7,614	1	Bowiri	Amanfro/Anyinase	2,000,000	62
	Nuzeme	10,083	1	Dayi	Dayi River Basin	997,900	31
Gbefi-Hoeme	Toga	42,163	2	Ketekrachi	Woroto	7,027,707	216
	Kpetoe	29,160	1		Adankpe	2,273,361	70
	Aveyiboe	27,540	1	Hohoe	Adutor	35,854,085	1103
	Valexo	16,300	1	Kadjebi	Kpoglo	9,413,582	290
	Aklamapata	6,318	1		Kadjebi	97,742,979	3008
	Have	6,430	1				
	Agbeditive	12,961	1				

Source: *(Kesse, 1985)*

Sample Preparation and Analysis

Clay samples were disaggregated and air dried for 7 days, pounded in a porcelain mortar and sieved with <4µm mesh. Sieved samples were homogenized, labeled in polythene bags and stored in the laboratory for the various analyses (e.g. Abdulhamid et al. 2015; Acosta et al. 2011; Inobeme et al. 2014).

Two replicate samples were prepared for each sample and labeled for atomic absorption spectrometry analysis (AAS). The Munsell Colour Chart was used to determine the colour of the sieved clay materials. The pH of the clay samples was determined using pH meter in1:1 soil water suspension and the electrical conductivity was determined in the filtrate of the water extract using conductivity meterfollowing procedures of Abdulhamid et al. 2015; Anapuwa and Okolie, 2015 and Olayinka et al. 2017.The atomic absorption spectrometry analysis was used to determine heavy metal concentrations of samples. The AAS analysis involved digestion and reading of heavy metals concentration. The labeled samples were all digested by measuring and transferring 10g of samples (two replicates) into a boiling test tube and treating with aqua regia thus 15ml HCl and 5ml HNO_3 in a ratio 3:1 (Association of Official Analytical Chemist, 1980; Acosta et al. 2011; Zhao et al. 2012). Samples were mixed and heated on a hot plate in a fume chamber with gradual increment in temperature of 90^oC and 150^oC until decomposition was complete (red NO_2 fumes ceasing).

The leachate became colorless and the volume reduced through evaporation to about 5ml. The sample was cooled, filtered through an acid-washed filter paper. The sample was then washed with de-ionized and double distilled water, transferred quantitatively to a 100ml volumetric flask and topped up to the mark with distilled water. The digested samples were covered tightly to prevent contamination with pollutants or other gases in the atmosphere since these could affect the results the filtrate solution of samples was analyzed for Cd, Pb, As, Na, Mg, Fe, Zn, Co, Ni, Mn, Cr and Cu with the aid of Atomic Absorption Spectrophotometer (AAS). The AAS instrument uses a chosen lamp that produces a wavelength of light which is absorbed by these elements of interest. Sample solutions were aspirated into the flame. If ions of the given element are present in the flame, they absorb the light produced by the lamp before it reaches the detector. The amount of light absorbed depends on the amount of the element present in the sample. Absorbance values for unknown samples are then compared to the calibrated curves prepared by running known samples (e.g. Amos -Tautua et al. 2014; Dua et al. 2013; Kusimi and Kusimi, 2012).

For quality assurance and quality control, blanks were measured to evaluate their contributions to error in the measurements. For each extractant, a blank solution of that extractant and water free metals reagent was obtained in the same way as the leachate solution. Each experiment was repeated two times, and parallel sampling was conducted for three times. Precautions taken during digestion included digestion taking place in a fume chamber since Nitrogen (IV) Oxide fumes could cause choking(Du et al. 2013; Inobeme et al. 2014).

Statistical Analysis

Analytical results were processed into tables whiles Principal Component Analysis (PCA) statistical analyses was applied to explain the variability in the heavy metal content of the clays.

RESULTS

Physicochemical Properties and Heavy Metal Concentrations of Mpreaso Clays

Clays are fine materials produced by the weathering of feldspathic and granitic rocks. The clays in Mpraeso were hue between 2.5-5YR which is described as pale to light yellow colour by the Munsell colour chart. The grains were very fine of less than 4µm as indicated by (Guggenheim et al. 1995; Kostorz 2016). The poorly compacted clays could be due to clastic sedimentation in which there is poor sorting. This might also be as a result of less weight overlying the rock which is accompanied by reduced pore space. The residual deposition of the clay through weathering of the parent material undergoes sorting processes during erosion, transportation and finally deposition. Other results of the physicochemical properties of Mpreaso clays are shown in Tables 3 - 5. The pH of a few results of the three typesof clay materials (raw, additives and mixed) were slightly acidic with a range of 5.2-6.3 for Mpraeso clays. The mean pH of the raw clays are higher than the clay additives locally called amodine and that of mixed clays are also slightly lower than the raw clays. The electrical conductivity (EC) of Mpraeso samples ranges between 2-598 for raw clays, 70-653 for clay additives, and 26-316 for mixed clays.

Elements detected in Mpreaso samples are Co, Cu, Fe, Pb, Mn, Ni, Cr, and Zn. Co and Pb were detected in only a few samples with a mean of 5.8mg/kg (Co) and 0.95 mg/kg (Pb) for raw clays, 3.9 mg/kg (Co and Pb) for the additives and 6 mg/kg (Co) and 10mg/kg (Pb) for mixed clays. The mean levels of other metals in the raw clays are 26mg/kg (Zn), 32mg/kg (Mn), 66mg/kg (Ni), 37mg/kg (Cr), 16mg/kg (Cu) with Fe extremely high (1637) (Table3).

Table3. *Concentration of elements in raw clays of Mpreaso*

Sample ID	Elements											
	pH	EC	Cd	Co	Zn	Mn	Ni	Pb	Fe	Cr	Cu	As
Site 2	5.4	2.2	BD	BD	16.5	15.6	**98.8**	10.3	2585	36.9	18.6	BD
Site 3	7.1	2.81	BD	BD	17.3	8.0	**64.7**	0.2	952	18.7	19.6	BD
Site 4	6.7	306	BD	BD	40.8	31.0	**54.44**	BD	1444	41	10.9	BD
Site 5	8.1	184.3	BD	BD	12.7	30.3	**79.4**	BD	1358	38.1	2.4	BD
Site 6	6.5	2.71	BD	0.5	14.3	30.9	**70.4**	BD	2371	34.2	22.8	BD
Site 7	5.3	407	BD	BD	27.0	68.2	**101.3**	BD	2885	26.4	BD	BD
Site 8	6.6	598	BD	9.2	12.4	13.2	**68.8**	BD	1617	33.4	1.6	BD
Site 9	6.9	187.2	BD	19.5	72.0	85.4	**61.9**	BD	2283	58.6	71.3	BD
Site 10	8.4	120.9	BD	10.8	25.8	17.7	5.7	BD	583	16.8	7.8	BD
Site 11	7.3	61.7	BD	23.9	23.8	34.2	43	BD	941	64.9	8.9	BD
Site 12	8.8	152.1	BD	BD	23.5	22.1	34.8	BD	986	32	10.4	BD
Mean	7.0	184.1	BD	5.8	26.0	32.4	**65.7**	0.95	1636.8	36.5	15.9	BD
Limits (FAO/WHO)			3	50	300	2000	50	100	50,000	100	100	20
Limits (EU)			3	n.a	300	n.a	75	300	n.a	150	140	n.a

BD – below detection; n.a – not available; Bold values are those that exceed limits

Except Mn whose mean concentration was slightly higher (436.6mg) in clay additives (Table 4) as compared to 32mg/kg in the raw clays (Table 3) the mean concentration levels of all other metals in clay additives (Table 4) was lower than that of the raw clays (Table 3). Fe was still very high compared with other metals with a mean value of 565.6mg/kg (Table 4). However, mean concentration levels of metals in mixed clays (Table 5) were relatively higher than that of clay additives (Table 4) but lower than that of raw clays (Table 3). Fe levels are still high with a mean of 1137mg/kg (Table 3) compared to 567mg/kg in clay additives (Table 4) and 1637 in raw clays (Table 3).

Nickle concentration levels of most samples of raw, additives and mixed clays were higher than prescribed standards of WHO, FAO and EU. The concentration levels of the other metals are all within permissible limits in soils prescribed by WHO, FAO and EU (Table 3-5).

Table4. *Concentration of elements in clay additives (amodine) of Mpreaso*

Sample	Elements											
	pH	EC	Cd	Co	Zn	Mn	Ni	Pb	Fe	Cr	Cu	As
Site 2	5.3	70.8	BD	BD	1.2	2.6	**80.4**	43.9	1201	BD	BD	BD
Site 3	7.1	70.8	BD	BD	BD	BD	**72.7**	BD	557	BD	BD	BD
Site 4	8.2	280	BD	BD	13.0	91.1	**57.1**	BD	140	23.2	2.1	BD
Site 5	6.3	190.8	BD	BD	7.8	27.5	**75.3**	BD	610	2.3	BD	BD
Site 6	7.1	510	BD	BD	2.8	BD	**50.4**	BD	134	19.9	5.0	BD
Site 7	8.1	158	BD	BD	34.5	72.1	46.3	BD	371	8.5	31.0	BD
Site 8	7.3	153.2	BD	BD	17.0	36.9	47.8	BD	160	13.5	1.7	BD
Site 9	7.2	158.8	BD	29.7	17.8	61.3	**52.7**	BD	96.2	11.2	BD	BD
Site 10	8.3	653	BD	BD	14.1	28.3	45.3	BD	670	11.5	BD	BD
Site 11	5.2	201	BD	12.8	16.0	19.4	**89**	BD	1465	12.7	BD	BD
Site 12	6.2	122	BD	BD	13.8	63.5	**74.6**	BD	817	9.9	BD	BD
Mean	6.9	233.5	BD	3.9	12.5	36.6	**62.9**	3.9	565.6	10.3	3.6	BD
Limits ((FAO/WHO),			3	50	300	2000	50	100	50,000	100	100	20
Limits (EU)			3	n.a	300	n.a	75	300	n.a	150	140	n.a

BD – below detection; n.a – not available; Bold values are those that exceed limits

Table5. *Concentration of elements in mixed clays of Mpreaso*

Sample	Elements											
	pH	EC	Cd	Co	Zn	Mn	Ni	Pb	Fe	Cr	Cu	As
Site 2	5.5	278	BD	BD	17.2	45.1	**115**	20.9	1707	18.2	21.7	BD
Site 3	6.2	26.4	BD	BD	12.4	26.1	44.9	32.0	757	20.4	12.1	BD
Site 4	5.7	219.2	BD	BD	45.0	141.5	**73.1**	16.3	1793	56.8	48.9	BD
Site 5	6.1	77.6	BD	BD	11.8	29.7	**62.0**	13.1	1493	30.0	BD	BD
Site 6	5.9	316	BD	10.5	25.0	86.6	**59.5**	BD	1313	21.7	10.3	BD

Site 7	6.2	78.2	BD	BD	1.4	5.1	**54.9**	BD	1050	35.1	BD	BD
Site 8	5.8	97.0	BD	13.6	31.6	27.6	**55.2**	BD	1316	27.7	2.4	BD
Site 9	7.4	101.4	BD	12.6	17.5	44.9	**57.0**	28.4	633	25.2	9.0	BD
Site 10	6.2	211	BD	5.4	27.7	22.8	**32.5**	BD	1073	31.3	BD	BD
Site 11	5.7	119	BD	6.7	24.7	32.0	**97.0**	BD	1085	19.7	11.0	BD
Site 12	7.3	94.2	BD	17.3	26.4	63.9	**55.1**	BD	271	42.0	17.6	BD
Mean	6.2	147.1	BD	6.0	21.9	47.8	**64.2**	10.1	1135.6	29.8	12.1	BD
Limits ((FAO/WHO			3	50	300	2000	50	100	50,000	100	100	20
Limits (EU)			3	n.a	300	n.a	75	300	n.a	150	140	n.a

BD – below detection; n.a – not available; Bold values are those that exceed limits

Physicochemical Properties and Heavy Metal Concentrations of Vume Clays

Vume clays are produced by the weathering of the granitic acidic gneisses of the Dahomeyan rocks. The clays were hue between 7.5-10YR which is described as orange to dull yellowish colour by the Munsell colour chart. They were well compacted with very fine grains of about a size less than 4µm (Guggenheim et al. 1995; Kostorz 2016) similar to Mpraeso clays.Results of other physical parameters and chemical analysis of elements of Vume clays are illustrated in Tables 6-8. Similar to results of Mpreaso clays, the mean pH of the raw clays are higher than the clay additives(grog) and that of mixed clays are also slightly lower than the raw clays in both communities. Most samples are slightly acidic with mean pH values of 6.5 or less. Only 3 samples of raw clays had pH above 6.5 (Table 6), that of grog clays were two (2) samples (Table 7) and for mixed clays is only one (1) sample (Table 6). Electrical conductivity of raw clays varies between 7 and 399, 78 and 514 for additives (grog) and 16 and 416 for mixed clays of Vume.

Table6. *Concentration of elements in raw clays of Vume*

Samples	**Elements**											
	pH	**EC**	**Zn**	**Mn**	**Ni**	**Cu**	**Fe**	**Cd**	**Pb**	**As**	**Cr**	**Co**
Site 2	5.2	7.0	142.6	184.3	**93.3**	5.7	19350	**6.5**	64.8	BD	BD	BD
Site 3	6.3	195.9	BD	37.9	**66.7**	BD	15370	**8.9**	31	BD	BD	1.4
Site 4	5.5	86.3	9.4	37.4	**87.2**	4.1	7359	**7.3**	86.3	BD	BD	6.3
Site 5	5.8	399	BD	45.7	**81.8**	6.8	9109	**7.5**	59.9	BD	BD	3.2
Site 6	6.7	43	BD	101.6	**69.1**	12	3103	**8.0**	53.8	BD	BD	6.7
Site 7	7.1	323	BD	24.8	**69.3**	3.9	1634	**7.5**	24.8	BD	BD	BD
Site 8	6.4	44.1	BD	37.8	**81.4**	5.3	2635	**9.5**	BD	BD	BD	BD
Site 9	6.4	66.9	BD	36.5	**76.5**	7.2	3994	**11.2**	21	BD	BD	12.8
Site 2	8.9	114.5	BD	32.7	**81.4**	2.8	12210	**8.6**	74.2	BD	BD	8.2
Mean	6.5	142.2	16.9	59.9	**78.5**	5.3	8307.1	**8.3**	46.2	BD	BD	4.3
Limits ((FAO/WHO)			300	2000	50	100	50,000	3	100	20	100	50
Limits (EU)			300	n.a	75	140	n.a	3	300	n.a	150	n.a

BD – below detection; n.a – not available; Bold values are those that exceed limits

Though in varying quantities eight (8) out of the ten (10) elements that were examined were detected in all Vume samples (Tables 6-8). Zinc (Zn) was found in only two (2) raw clay samples with a mean of 16.9mg/kg but was detected in more samples of additives and mixed clays with mean concentrations of 12 and 16mg/kg respectively (Tables 7 & 8). Manganese (Mn), Ni, Fe, Cd, Pb, Cu and Co were detected in almost all raw, additive and mixed clay samples of Vume except in a few for Co. The mean values of Mn, Ni, Cu, Fe, Cd, Pb and Co for raw calys are 60, 79, 5, 8307, 8, 46 and 4.3 respectively (Table 6), that of additive clays are 44, 72, 3, 3818, 7, 56 and 3.4 respectively (Table 7), whiles that of mixed clays are 16, 57, 74, 5, 8049, 8, 58 and 4 respectively (Table 8). Cd and Ni levels are higher than acceptable thresholds in soils. Also Site 8 mixed clay sample Pb concentration is slightly above permissible limts(Table 8). However, the concentration levels of the other elements are all below standard limits in soils of international organizations such as WHO, FAO and European Union.

Table7. *Concentration of elements in clay additives(grog) of Vume*

Samples	**Elements**											
	pH	**EC**	**Zn**	**Mn**	**Ni**	**Cu**	**Fe**	**Cd**	**Pb**	**As**	**Cr**	**Co**
Site 2	7.3	301	28.5	74.4	**73.2**	9.1	3703	**4.4**	90.9	BD	BD	5.7
Site 3	5.3	510	41.5	70	**74.9**	BD	9901	**8.6**	BD	BD	BD	BD

Site 4	6.0	268	9.3	29.7	**63.4**	BD	1386	**5.7**	66.6	BD	BD	6.3
Site 5	7.1	514	BD	34.9	**65.5**	0.7	2327	**8.7**	87.6	BD	BD	BD
Site 6	6.4	79.4	2.9	36.2	**65.8**	4.9	2917	**4.7**	BD	BD	BD	BD
Site 7	5.9	201	BD	30.1	**75.2**	3.5	3679	**9.6**	74.9	BD	BD	4.8
Site 8	5.7	78.3	BD	35.6	**83.6**	0.3	2816	**7.3**	71.3	BD	BD	7.1
Mean	6.2	278.8	11.7	44.4	**71.7**	2.6	3818.4	**7.0**	55.9	BD	BD	3.4
Limits ((FAO/WHO)			300	2000	50	100	50,000	3	100	20	100	50
Limits (EU)			300	n.a	75	140	n.a	3	300	n.a	150	n.a

BD – below detection; n.a – not available; Bold values are those that exceed limits

Table8. *Concentration of elements in mixed clays of Vume*

Samples	**Elements**											
	pH	**EC**	**Zn**	**Mn**	**Ni**	**Cu**	**Fe**	**Cd**	**Pb**	**As**	**Cr**	**Co**
Site 2	5.2	392	103	111.3	**82.8**	7.3	28700	**8.2**	87.7	BD	BD	9.3
Site 3	6	16	BD	44.9	**78.2**	BD	8098	**7.6**	41.3	BD	BD	3.9
Site 4	5.4	119	6.9	92.9	**70.7**	0.7	1617	**7.4**	95	BD	BD	BD
Site 5	5.4	70.4	3.1	45.8	**61.4**	13.5	3536	**10.8**	40.2	BD	BD	BD
Site 6	5.5	416	BD	26.1	**72.9**	2.2	2547	**7.6**	BD	BD	BD	0.8
Site 7	6.4	78.2	BD	33.3	**67.3**	5.2	5069	**8.4**	29.6	BD	BD	7.4
Site 8	5.8	87	13.1	45.6	**79.5**	6.5	6900	**7.6**	67.5	BD	BD	1.5
Site 8	7.4	100.1	5.1	54.3	**77**	2.8	7932	**8.8**	100.3	BD	BD	7.4
Mean	5.9	159.8	16.4	56.8	**73.7**	4.8	8049.9	**8.3**	57.7	BD	BD	3.8
Limits ((FAO/WHO)			300	2000	50	100	50,000	3	100	20	100	50
Limits (EU)			300	n.a	75	140	n.a	3	300	n.a	150	n.a

BD – below detection; n.a – not available; Bold values are those that exceed limits

Descriptive Statistics of Some Heavy Metal Concentrations in the Clays

Principal Component Analysis (PCA)

Principal Component Analysis (PCA) was applied to explain the variability of heavy metals in the clay samples through the application of varimax rotation with Kaiser Normalization. Tables9-11show the results of the factor loadings with a varimax rotation, as well as the eigenvalues and communalities for Mpraeso clays. The results indicate that two eigenvalues were higher than one (1) and these two factors explained 76.97 % of the total variance. The first factor explains 46.7% of the total variance and with major contributions from Zn, Mn, Cr, and Cu and moderate contribution from Co. Factor 2 accounts for 30.3% of the total variance with major positive contribution from Ni and Fe and a negative contribution from Co. Meanwhile the additive samples indicate three eigenvalues that were higher than one (1) (Table10). These three factors explained 75.6 % of the total variance with the first factor explaining 43.3% of the total variance with a positive loading on Zn, Mn (heavily), Cr and Cu (moderately) and a negative loading from Ni, Pb and Fe. Factor 2 positively dominated by Zn, Fe and Cu with a negative contribution from Cr accounted for 17.2% of the total variance. Factor 3 is positively heavily loaded by Co accounted for 15.1% of the total variance.

Table9. *Rotated component matrix for raw clays of Mpraeso*

Elements	**Components**		**Communalities**
	1	**2**	
Co	**0.623**	**-0.597**	0.745
Zn	**0.882**	-0.141	0.798
Mn	**0.856**	0.269	0.804
Ni	0.073	**0.932**	0.873
Fe	0.384	**0.880**	0.923
Cr	**0.736**	-0.163	0.568
Cu	**0.822**	-0.023	0.676
Eigen value	3.269	2.119	
% of Variance explain	46.703	30.272	
% of Cumulative	46.703	76.974	

Loadings >0.4 are shown in bold

Table10. *Varimax rotated PC loading for additives of Mpraeso*

Elements	Components			Communalities
	1	2	3	
Co	0.146	-0.201	**0.839**	0.765
Zn	**0.752**	**0.548**	0.287	0.949
Mn	**0.750**	0.176	0.304	0.685
Ni	**-0.799**	0.157	0.381	0.808
Pb	**-0.640**	0.307	-0.140	0.523
Fe	**-0.736**	**0.409**	0.316	0.809
Cr	**0.639**	**-0.461**	0.031	0.622
Cu	**0.566**	**0.713**	-0.245	0.888
Eigenvalue	3.467	1.379	1.204	
% of Variance explained	43.334	17.234	15.046	

Loadings>0.4 are shown in bold

Table11. *Varimax rotated PC loading for mixed clays ofMpraeso*

Elements	Components			Communalities
	1	2	3	
Co	-0.150	**0.810**	-.158	0.704
Zn	**0.774**	**0.437**	-.195	0.828
Mn	**0.924**	0.175	.091	0.892
Ni	0.349	**-0.595**	**-0.443**	0.673
Pb	0.106	**-0.531**	**0.739**	0.839
Fe	**0.526**	**0.574**	**-0.428**	0.790
Cr	**0.649**	**0.413**	.236	0.647
Cu	**0.934**	0.086	.206	0.923
Eigenvalue	3.180	2.021	1.095	
% of Variance explained	39.745	25.264	13.683	

Loadings>0.4 are shown in bold

When the clay was mixed with the temper the results indicated three eigenvalues higher than one (1) (Table 11). These three factors explained 78.7% of the total variance with the first factor explaining 39.8% of the total variance with a positive heavy load on Zn, Mn and Cu and moderately on Fe and Cr. Factor 2, dominated positively by Co (high), Fe (moderate), Zn and Cr (minor) and negatively by Ni and Pb accounted for 25.3% of the total variance. Factor 3 accounted for 13.7% of the total variance was positively loaded by Pb with a negative contribution from Ni and Fe.

Tables12-14 show PCA analysis of Vume heavy metal content. The results indicated three eigenvalues higher than one (1) (Table 12) for raw clays and these three factors explained 80.3% of the total variance. The first factor explains 45.5% of the total variance and loads positively on Zn, Mn, Ni, Fe (high), Pb (moderate) with a negative influence from Cd. Factor 2 dominated positively by Cu and Co accounts for 19.4% of the total variance. Factor 3 accounting for 15.4% of the total variance has a positive moderate loading by Pb and Co and a negative influence from Cu.

The results of additives of Vume indicate that three eigenvalues were higher than one (1) (Table 13) and these three factors explained 83.8% of the total variance. The first factor explains 38.2% of the total variance and loads heavily on Zn, Mn and Fe positively with a moderate negative contribution from Pb and Co. Factor 2, dominated by Mn, Cu and Co with a negative influence from Cd accounts for 24.9% of the total variance. Factor 3 is positively loaded by Ni, Cd, Pb and Co and accounting for 20.6% of the total variance.

Table 14shows the results of the factor loadings, eigenvalues and communalities for mixed clays of Vume. Two eigenvalues were higher than one (1) and that these two factors explained 74.9% of the total variance. The first factor explained 48.4% of the total variance and loads heavily on Zn, Mn, Ni, Fe, Pb and Co positively. Factor 2 dominated by Cu and Cd positively with a negative influence from Ni accounts for 26.4% of the total variance.

Discussion

Although mineral ions are essential elements in living organisms, there has growing concern about the increasing toxicity of some elements in the environment. Naturally, soils contain mineral ions dissolved from rocks as the water travels along mineral surfaces in the soil (Kusimi and Kusimi, 2012). Thus, depending on the extent of exposure of these minerals, they could be in higher concentrations or vice versa. Human activities also explain the toxicity levels of certain elements in the environment.

Table12. *Rotated component matrix for raw clay of Vume clays*

Elements	Components			Communalities
	1	2	3	
Zn	**0.903**	0.041	-.222	0.867
Mn	**0.821**	0.302	-0.380	0.911
Ni	**0.705**	0.204	0.227	0.590
Cu	-0.036	**0.868**	**-0.451**	0.958
Fe	**0.758**	-0.307	0.325	0.774
Cd	**-0.756**	0.109	0.083	0.590
Pb	**0.594**	0.307	**0.613**	0.823
Co	-0.388	**0.683**	**0.543**	0.912
Eigen Value	3.638	1.554	1.232	
% of Variance explained	45.471	19.421	15.405	

Loadings>0.4 are shown in bold

Table13. *Varimax rotated PC loading for additives of Vume*

Elements	Components			Communalities
	1	2	3	
Zn	**0.904**	0.249	0.082	0.886
Mn	**0.840**	**0.472**	0.141	0.948
Ni	0.185	0.041	**0.874**	0.800
Cu	0.107	**0.834**	-0.168	0.736
Fe	**0.937**	-0.257	0.217	0.991
Cd	0.028	**-0.764**	**0.488**	0.822
Pb	**-0.617**	0.359	**0.437**	0.700
Co	**-0.481**	**0.488**	**0.591**	0.819
Eigen Value	3.060	1.998	1.644	
% of Variance explained	38.247	24.978	20.550	

Loadings>0.4 are shown in bold

Table14. *Varimax rotated PC loading for mix clays ofVume*

Elements	Components		Communalities
	1	2	
Zn	**0.897**	0.299	0.894
Mn	**0.811**	0.133	0.676
Ni	**0.767**	**-0.469**	0.808
Cu	-0.081	**0.944**	0.897
Fe	**0.916**	0.251	0.903
Cd	-0.280	**0.908**	0.903
Pb	**0.657**	0.030	0.433
Co	**0.682**	0.095	0.474
Eigen Value	3.874	2.115	
% of Variance explained	48.421	26.442	

Loadings>0.4 are shown in bold

Chemical elements that exceeded WHO, FAO, EU guidelines in soils are Cd and Ni. Nickel levels were high in some samples of both Mpraeso and Vume whiles Cd was found to be higher in certain samples in Vume clays. The clarification to PCA loadings according to Rodriguez et al. (2006) is that the association of heavy metals can be indicative of their common origins from anthropogenic or natural sources (geogenic and pedogenic sources). Naturally, cadmium exists in the earth's crust in zinc, lead and copper sulphide ores, thus enters the environment through weathering. However, these ores are not present in the geology of the study areas. Cadmium is also anthropogenically introduced into the environment when it is unrecovered from the industrial extraction of cadmium bearing ores or the disposal of cadmium rechargeable batteries or cadmium bearing electronic gadgets. The possible source of high Cd levels in Vume clays is industrial waste containing cadmium. The clay is mined in a valley in the town where domestic waste is dumped. The valley also drains the southern part of the town. Leachate from industrial wastes leaches into the soils increasing Cd levels in the clay materials accounting for the high PCA loading. Clays of Mpraeso are mined outside the towns which is not within the reach of waste from households hence the absence of Cd in Mpraeso clays.

Nickel is the fifth most common element on earth and in soils it is associated geochemically with iron and cobalt (Harasimand Filipek, 2015; Kesse 1985). In soils it occurs mostly in minerals such as pentlandite, garnierite, millerite, niccolite and ullmannite (Harasim and Filipek, 2015; Kesse 1985). These ores are not detected in the geology of the study areas. Anthropogenic sources of nickel include fertilizers like ammonium nitrate and manure from dairy, poultry and swine (Chauhan et al. 2008; Harasimand Filipek, 2015). Staple crops and vegetables are farmed around the surroundings of the clay mining landscapes in both study areas and farmers apply ammonium rich fertilizers. At Mpraeso there is a kraal for cowsupl and of the clay valley and in dry season the cows graze in the clay lands introducing dairy manure into the landscape. The mobility of nickel in soilis site-specific, depending mainly on the soil type and pH. The mobility of nickel in soil is increased at low pH.As shown in Tables 1 – 6, higher Ni concentrations are associated with lower pH or slightly acidic soil conditions. In acidic, organic -rich soils, where fulvic and humic acids are formed by the decomposition of organic material, Ni may be quite mobile, possibly because of complexation by these ligands (Kabata-Pendias, 2001). Thus, Ni source in the clays of both areas is attributable to the above human activities in the locality.

The high loadings of other heavy metals (Co, Zn, Mn, Cr, Fe, and Cu) suggest that these heavy metals are primarily ascribed to natural processes of rock weathering with little or no attribution from anthropogenic activities hence their low levels in the clay materials. For instance, rocks of the Dahomeyen are impregnated withiron: epodote [$Ca_2(Fe, Al)Al_2(SiO_4)(Si_2O_7)O(OH)$] and hornblende [$(Ca, Na)_2(Mg, Fe, Al)_5(Al, Si)_8O_{22}(OH)_2$] (Kesse, 1985). The Birimian formation is also reach in iron (Kesse, 1985). Deep chemical weathering under tropical conditions mobilizes iron in the soils which justifies the high concentration of iron in clay samples in both localities. Anthropogenic activities such as pesticide application, fertilization, industrialization and atmospheric deposition, may generate a strong impact on the accumulation of these heavy metals in the clays in future as result of urbanization and economic development.

Conclusion

The study showed that most clay samples were slightly acidic with a few being alkaline.Three (3) (As, Cd and Cr) out of the ten (10) metals (Cd, Pb, As, Fe, Zn, Co, Ni, Mn, Cr and Cu) were not detected in the clays of the study areas. Arsenic (As) was below detection in clays of both areas whiles Cd was undetected in Mpraeso clays and Crin Vume. Other metals are in tracer quantities exceptNi levels in both Mpraeso and Vume and Cd levels in Vume clays/pottery materials that were above FAO/WHO and EU acceptable limits in soils. Hence with the exception of Cd and Ni which need some remediation, the quality of clays used in Mpraeso and Vume pottery were found to be generally good for earthenware production. Sources of Cd and Ni in the clays are anthropogenic hence measures should be put in place by the local government authorities to control the introduction of these heavy metals into the environment. Environmental and sanitation measures should also be put by the local governments to control human activities that will introduce the other heavy metals into the environment.

Acknowledgement

This paper is from my PhD thesis which was self-funded. I extend my heartfelt gratitude to my supervisors; Prof Augustine Donkor, Dr Kwaku Kyeremeh and Dr Jesse S. Ayivorfor supervising my work. I am also thankful to Ghana Education Trust Fund (GETFUND) for paying my tuition fees of my PhD Programme.

Conflict of Interest Declaration and Data Availability Statement

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. They do not have the right to make the data of the manuscript available to other parties.

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Citation: *Bertha Ansaah Kusimi, John Manyimadin Kusimi, “Physicochemical Properties and Heavy Metal Content of Mpraeso and Vume Clays of Ghana", Annals of Geographical Studies, 2(4), 2019, pp. 22-35.*