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Intrapedal Variability of Mercury in Two Soil as Affected by Clay Type and Sesquioxides

[1]E.U. Onweremadu and [2]A.E. Ibe
[1]Department of Soil Science and Technology
[2]Department of Crop Science and Technology, Federal University of Technology,
P.M.B. 1526, Owerri, Nigeria

Abstract: Influence of clay type and sesquioxides on the distribution of mercury (Hg) in a soilsphere in southeastern Nigeria was studied in 2006. A base map derived from geological map of the study area guided field sampling. Routine and special analyses were conducted on soil samples for the purpose of this study. Soil data were subjected to statistical analysis using SAS computer software. Results showed that Ibeku soils were younger, had more sesquioxides with crystalline forms dominating the amorphous oxides. Similar trend was followed by older Umuahia soils. Hemiexpansible minerals and smectites were dominant clay minerals in Ibeku soils unlike Umuahia soils dominated by kaolinite. Higher values of Hg were recorded in the Ibeku soils (9.2-15.6 mg kg^{-1}) compared with Umuahia soils (5.6-8.1 mg kg^{-1}). Degree of relationship between Hg and soil properties differed. While organic matter (R = 0.92), hemi-expansible minerals (R = 0.91) and oxalate iron (Fe_{ox}) (R = 0.88) influenced Hg distribution in Ibeku soils, kaolinite (R = 0.89) and Fe_{ox} (R = 0.89) had the highest influence over Hg contents in pedogenic horizons of Umuahia soils. More information on Hg forms, dispersability and flocculation properties of these soil colloids and edaphic factors influencing them are needed for practical usages in agriculture and environment.

Key words: Degradation, pedology, mineralogy, translocation parent materials

INTRODUCTION

Mercury (Hg) is a neurotoxic heavy metal (Farella *et al*., 2001) which is found in soil (Brabo *et al*., 2003). Soil Hg can result from long history of accumulation (Fadini and Jardim, 2001), from emission (Hernandez *et al*., 2004), soil erosion (Mainville *et al*., 2006) and land clearing (Fostier *et al*., 2000). Land use changes affect biodiversity (Myers *et al*., 2000) which affect plant soil-plant interactions and consequently human health.

Food is the most basic of human need for survival, health and productivity (Smith *et al*., 2006) but land degradation brought about by a prolonged interface between human-induced and natural factors exacerbated low food productivity (Di Falco *et al*., 2006) as well as its quality (Aikore *et al*., 2003). One of the problems of land degradation is the accumulation of toxic heavy metals, such as mercury (Hg), whose Hg (11) form is strongly retained by soil (Isirimah *et al*., 2003).

Soil Hg content and bioavailability may be dependent on the depth and organic matter content of the soil (Mainville *et al*., 2006), form of Hg and presence of sesquioxides (Isirimah *et al*., 2003), seasonal change (Onweremadu *et al*., 2007) and vegetal form (Roulet *et al*., 1999). The Hg contents are

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Corresponding Author: Dr. E.U. Onweremadu, Department of Soil Science and Technology,
Federal University of Technology, P.M.B. 1526, Owerri, Nigeria
Tel: + 2348034935502

high in upper horizons of the soil profile resulting from anthropogenic contributions to the predosphere. Yet in deeper horizons, Hg also occurs possibly resulting from weathering of rocks and consequent pedogenesis (lithogenic Hg) Agbozu *et al.* (2007) reported that heavy metals enter the aquatic environment through natural and anthropogenic source.

Soil properties such as clay content, organic carbon content, aluminium and iron oxides were highly correlated with P-sorption (Dodor and Oya, 2000; Zhang *et al*., 2005), implying that such properties influence adsorption of heavy metals (Basta *et al*., 1993; Krishnamy and Mathan, 2001).

Studies on soil Hg distribution in relation to soil clay type and sesquioxides are non-existent in the study area. However, to the best of my knowledge, only studies on general distribution have been conducted (Ano, 1994; Odoemena and Ano, 2003; Aiyesanmi, 2005; Onweremadu *et al*., 2007). This study investigated variability of soil Hg as influenced by clay type and sesquioxides.

MATERIALS AND METHODS

Study Area

Two study sites, namely Ajata Ibeku and Amakama Umuahia lying on latitude 5°32′151″N and 7°33′34″E and longitudes 5°26′40″N and 7°28′49″E, respectively are located in Abia State, Southeastern Nigeria. While soils of Ajata Ibeku are underlain by Bende-Ameki formation (Coarse sandstones, intercalations of Shales and thin shelly limestones), Amakama Umuahia soils are formed from Benin formation (Coastal Plain Sands). Both sites are in the humid tropical climate with marked wet and dry seasons of nine and three months duration, respectively. The mean annual rainfall is about 2201.92 mm while annual temperature varies between a minimum of 22°C and a mean maximum of 32°C. The vegetation is the tropical rainforest made up of several woody perennials notably *Elaeis guneensis*, *Dactyldenia barteri*, *Mangifera indica*, *Gmelina arborea*, *Anarcadium occidentale* etce tera. Low input agriculture predominates in the area with the traditional slash and burn type of land clearing after soil fertility restoration by bush fallowing.

Field Sampling

Field studies were conducted in the early months of 2006. Guided by a geological map of the study area, two study sites, namely Ajata Ibeku-(Bende-Ameki formation) and Amakama Umuahia (Benin formation), were chosen for field study. These formations are recorded geological periods. On each soil group, a soil profile pit was dug, described and sampled based on the degree of pedogenic horizonation (FAO, 1990). Five soil samples were collected from each pedogenic horizon, giving a total of 25 soil samples per sampling site. Soil samples were air-dried, crushed gently and sieved through 2 mm sieve.

Laboratory Methods

Particle size distribution of the less than 2 mm fractions was measured by hydrometer method (Gee and Or, 2002). The soil pH was measured in 1:2.5 suspensions of soil in 0.1 M KCl. Total carbon was determined by loss on ignition method using the LECO equipment. Cation exchange capacity (CEC) was estimated using N ammonium acetate (Thomas, 1982). Sodium-citrate-dithionite was used to extract iron (Fe) and aluminium (Al) on the < 2 mm fraction of all genetic horizons (Loeppert and Inskeep, 1996) and results were written in subscripted d. For the determination of minerals and elemental oxides in clay fractions, < 2 mm fractions were mechanically fractionated in deionized water at a 1:8 (W/W) soil/water ratio after shaking for 16 h on a reciprocal shaker at 60 revolutions per minute X-Ray Diffractometry (XRD) of clay was determined with oriented clay specimens using the Siemens 1500 diffractometer with Ni-filter and CuKα-radiation. Prior to mounting the oriented clay, sub-samples were first pre-treated as follows: saturation with Mg, solvation with glycerol, saturation with K and subsequent heating of the sample to 110, 350 and 550°C. This was done to isolate different mineral forms because of their variable properties under different temperature regimes. The semi-quantitative evaluation minerals in the soil clay fractions was determined using a computer package DIFFRAC AT V3.3 (Siemens, 1993). This method assumes that available minerals in the sample sum to 100% and the individual mineral is fraction of the total. Oxides of elements were estimated using Siemens SRS 200 X-ray fluorescence (XRF). In this methods, the < 2 mm soil samples and fractions were ground to powder, out of which 2.666 g was weighed and further ground and mixed with 1.333 g cellulose material with the trade name Spectromelt CIO Merck, which acts as a binding agent to the soil sample meant for analysis. This sample was then transferred to tablet making machine and turned into tablet with a pressure of 30 t for 5 min. The soil/cellulose sample in a ratio of 2:1 was then analyzed using XRF against standards to obtain the concentration of the elements which was further converted to their oxide forms using factors. The elements determined in this manner are referred to a total elements and are expressed as oxides. These standards were of soil and rock materials of various mineralogies purchased commercially from Breitlander Eichproben und Labormaterial GmbH (Hans-Sachs-Strasse 12, D-/59077 Hamm, Germany, Available Online at: http://www.breitlander.com). The standards, of which were 50, were prepared in the same way as the soil samples and analysed with XRF using the Cr-Tube at the intensity of 50 kV and 50 mA. This method has been calibrated using a Siemens computer package Spectra 300, V2.0 (Siemens, 1993). Specific surface area was determined through N_2-adsorption using a single point BET method.

Total soil mercury levels were determined with the Cold Vapour Atomic Fluorescence (CVAF) technique (Pichet *et al*., 1999). In this method 250 mg of soil was digested in a 10:1 nitric and hydrochloric mixture, heated at 120°C and then injected in an Atomic Absorption Spectrophotometer (AAS).

Statistical Analysis

Soil data were subjected to correlation analysis using Statistical Analysis System (SAS) General Linear Model procedures (SAS Institute, 1990).

RESULTS AND DISCUSSION

Soil Properties

Both soils are deep and of strong to moderate acidity. Soils from Ibeku were clayey with high values of Cation Exchange Capacity (CEC) and Organic Matter (OM) unlike Umuahia soils. But Umuahia soils were more highly weathered (SCR = 0.11-0.17) than Ibeku soils (SCR = 0.13-0.68) (Table 1). In both soils, Silt Clay Ratios (SCR) decreased with depth. Variability in soil properties is attributed to

Table 1: Profile distribution of soil characteristics

Horizon	Depth (cm)	Clay (g kg^{-1})	Specific surface areas (m^2 g^{-1})	Silt (g kg^{-1})	Sand (g kg^{-1})	pH KCl	ECE (cmol kg^{-1})	OM (g kg^{-1})	SCR
Ibeku									
Ap$_1$	0-9	392	18	267	341	5.1	24.4	39.6	0.68
Ap$_2$	9-28	455	28	228	317	5.0	21.6	18.9	0.50
Bt	28-60	749	36	148	103	4.9	19.3	9.8	0.19
2 Btg	60-80	819	68	121	60	4.7	24.6	7.0	0.14
2 Bssgl	80-101	805	54	111	84	4.4	23.8	4.3	0.13
Umuahia									
Ap$_1$	0-12	206	12	36	758	4.3	6.5	11.6	0.17
Ap$_2$	12-29	243	13	32	725	4.5	5.2	10.3	0.13
Bo$_1$	29-65	292	15	38	670	4.6	3.2	7.7	0.13
Bo$_2$	65-102	320	17	50	630	4.8	2.8	3.6	0.15
Bo$_3$	102-126	313	16	36	651	4.8	2.5	2.6	0.11

CEC = Cation Exchange Capacity, OM = Organic Matter, SCR = Silt Clay Ratio differences in parent material since the two locations are geographically associated. However, land use history may have a localized influence on soil differences. Higher values of specific surface area were recorded in Ibeku soils, implying greater availability of moisture content in its exchange sites.

Iron and Aluminium Mineralogy

The dithionite extractable Fe (Fe_{di}) of Ibeku soils (younger soils) are higher than those of Umuadhia (older soils) (Table 2). This result is consistent with the findings of Igwe *et al.* (2005) that Fe_{di} content was higher in recent and very recent soils than older ones of the River Niger floodplain in eastern Nigeria. The Fe_{di} is the crystalline plus the amorphous or poorly crystalline forms of Fe, whereas the Fe_{ox} is the amorphous form. In both soils, the crystalline forms of Fe and Al dominated over the amorphous types probably to the age of these soils. Lower Fe_{di}/clay ratios in Ap/horizons can be attributed to long periods of waterlogging of these soils, dissolution of Fe oxides and their eventual removal from the soils.

Clay Mineralogy

Kaolinite, followed by hemi expansible minerals dominated soil clays of Umuahia while hemi-expansible minerals followed by smectites were predominant in Ibeku soils (Table 3). Low pH and local temperature > 25°C favour formation of kaolinite. Micas are altered at acidic ranges of soil pH giving rise to smectite (Aoudjit *et al*., 1996). The distribution of kaolinite is related to the age of these soils, as more highly weathered Umuahia soils (SCR = 0.11-0.17) had more kaolinite than the younger Ibeku soils (SCR = 0.13-0.68). The abundance of kaolinite in Umuahia soils indicates that most primary minerals (feldspars and micas) have disappeared due to weathering and this portends low fertility.

Table 2: Iron and aluminium mineralogy in studied soils

		Fe_{di}	Al_{di}	Fe_{ox}	Al_{ox}	Fe_{ox}/Fe_{di}	Fe_{di}/clay	Fe_{di}/total Fe
Horizon	Depth (cm)	--g kg^{-1}--						
Ibeku								
Ap_1	0-9	18.2	3.1	14.6	2.1	0.80	0.04	0.55
Ap_2	9-28	48.6	2.6	8.2	1.6	0.17	0.01	0.85
Bt	28-60	52.6	3.4	8.0	1.6	0.15	0.07	0.86
2 Btg	60-80	56.4	3.6	5.2	1.5	0.09	0.06	0.91
2 Bssg1	80-101	54.2	2.8	2.8	1.1	0.05	0.06	0.95
Umuahia								
Ap_1	0-12	5.2	1.4	2.2	1.0	0.40	0.02	0.70
Ap_2	12-29	6.8	1.5	2.8	1.1	0.41	0.02	0.70
Bo_1	29-65	8.2	2.8	3.0	2.4	0.36	0.03	0.73
Bo_2	65-102	8.4	2.6	3.1	1.3	0.37	0.02	0.73
Bo_3	102-126	6.2	2.4	2.6	1.2	0.42	0.01	0.70

Fe_{di} and Al_{di} = Dithionite extractable Fe and Al, Fe_{ox} and Alox = Oxalate extractable Fe and Al

Table 3: Distribution of major clay types in the study sites (loss on ignition)

Horizon	Depth (cm)	Illite	Hemiexpansible minerals	Smectites	Kaolinite
Ibeku					
Ap_1	0-9	8	25	22	30
Ap_2	9-28	7	18	31	33
Bt	28-60	9	16	32	31
2 Btg	60-80	10	21	30	30
2 Bssg1	80-101	6	14	39	29
Umuahia					
Ap_1	0-12	4	26	16	53
Ap_2	12-29	4	27	17	55
Bo_1	29-65	3	24	19	58
Bo_2	65-102	3	23	14	59
Bo_3	102-126	2	25	13	61

Mercury Content

Table 4 shows the distribution of Hg in these soil, with higher values observed in Ibeku soils. While in Ibeku soils Hg decreased with depth, the reverse was the case in Umuahia soils. This variability could be attributed to the distribution of OM in Ibeku soils while soils of Umuahia were highly leached. Higher Hg values in the epipedon is in line with the findings of Mainville *et al.* (2006) that there are higher Hg burden in surficial layers due to organic matter abundance. Results of Hg distribution in Umuahia soils show that the influence of OM was less prominent in sub-surface Hg distribution.

Relationships Between Mercury and Soil Properties

There was a more significant ($p<0.05$) correlation between Hg and OM ($R = 0.92$) in Ibeku soils when compared with soils of Umuahia ($R = 0.78$), suggesting that OM has more influence on Hg content in Ibeku soils while in less OM-containing soils of Umuahia, other soil attributes are contributing meaningfully to Hg distribution (Table 5). Higher correlation coefficient values between Fe and Al oxides were recorded, implying that these sesquioxides influence greatly the distribution of soil Hg. These results show that OM and oxides of Fe and Al could be used as good predictors of Hg distribution. Organic matter provided sorption sites for the adsorption of soil Hg but its impact is felt at surficial layers (Mainville *et al.*, 2006) while Fe and Al oxides may form complexes with OM. Gerke and Hermann (1992) showed that P-sorption in Fe-humic substance mixtures was six to seven times larger than amorphous Fe oxides and this may be due to the formation of ternary complexes between organically bound Fe and phosphates. This relationship can also be likened to the formation organo-mercuric substances in soils which makes Hg immobile and less bioavailable to crops. In deeper horizons, oxides of Fe and Al may have greater influence as OM decreased with depth.

Clay minerals had more significant ($p<0.05$) with Hg in Umuahia soils. Kaolinite had a greater influence ($R = 0.89$) on soils of Umuahia while the hemi-expansible minerals ($R = 0.92$) and illites ($R = 0.89$) predominate in Ibeku soils. Higher values of clay content in the subsurface horizons of Ibeku soils, dominated by hemi-expansible mineral (vermiculite, Al-interlayered vermiculite) and

Table 4: Mercury distribution among soils ($n = 25$)

Horizon	Depth (cm)	Hg (mg kg^{-1})
Ibeku		
Ap_1	0-9	15.6
Ap_2	9-28	9.2
Bt	28-60	9.6
2 Btg	60-80	9.8
2 Bssg1	80-101	9.7
Umuahia		
Ap_1	0-12	7.7
Ap_2	12-29	7.8
Bo_1	29-65	7.9
Bo_2	65-102	8.1
Bo_3	102-126	5.6

Table 5: Relationship between Hg and selected soil properties ($p<0.05$; $n = 25$)

Factors correlated			Factors correlated		
Ibeku	R	R^2	Umuahia	R	R^2
Hg Vs Fe_{di}	0.86	0.73	Hg Vs Fe_{di}	0.80	0.64
Hg Vs Fe_{ox}	0.88	0.77	Hg Vs Fe_{ox}	0.89	0.79
Hg Vs Al_{di}	0.79	0.62	Hg Vs Al_{di}	0.82	0.67
Hg Vs Al_{ox}	0.81	0.65	Hg Vs Al_{ox}	0.87	0.75
Hg Vs Kaolinite	0.78	0.61	Hg Vs Kaolinite	0.89	0.79
Hg Vs Smectite	0.82	0.67	Hg Vs Smectite	0.68	0.46
Hg Vs HExp	0.91	0.82	Hg Vs HExp	0.66	0.43
Hg Vs Illite	0.89	0.79	Hg Vs Illite	0.54	0.29
Hg Vs OM	0.92	0.84	Hg Vs OM	0.78	0.60

smectite is attributed to dispersion and translocation of clay colloids forming illuvial pedogenic horizons. This is consistent with the findings of Kjaergaard *et al.* (2004) in Danish clayey till soils. The above implies that kaolinite-dominated soils of Umuahia are less dispersable and may retain less Hg while easily dispersed Ibeku soils will allow rapid movement of Hg in the pedosphere. This could constitute a danger to environmental health as it causes non-point source pollution., especially in aquatic habitats. These positive correlations between Hg and mineralogical properties in both soils point to the fact that Fe and Al oxides in addition clay type influence dispersability and transportability of Hg. The above result is in line with reports of some scholars (Seta and Karathanasis, 1997; de Jonge *et al.*, 2000) that soil contaminants, such as pesticides and heavy metals sorb strongly soil to colloids (sesquioxides and clays). However, there may need for more investigations on Hg form, dispersability and flocculability of colloids as they affect Hg mobility in these humid tropical soils. Again, an evaluation of more soil properties related to pedogenesis of Hg may be necessary. These soil and soil-related factors when studied should be subjected to Principal Component Analysis (PCA) to determine main factors for purposes of modelling for precision agriculture and environmental friendliness.

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