

Heavy Metals Distribution in Soil Profile Exposed to Leachate of Septic Tanks in a School Environment

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Abstract

This study examined the physicochemical properties of soil exposed to leachate from septic tanks within the International Secondary School of the Rivers State University, Rivers State, Nigeria. Soil samples were collected in quadruplicates from depths 0.0 m (surf), 0.5 m (sub), 1.0 m (deeps) and 1.5 m (Msub) for each 0.5 m, 1.0 m, 1.5 m and 2.0 m horizontal distances away from all eight sides of two septic tanks yielding a total of 128 samples. The samples were analyzed using standard physicochemical and AAS techniques, and the obtained experimental outcomes subjected to standard statistical treatment. The results showed that the means of the levels of pH, PO₄, and Zn followed the order, surf>sub>deeps>Msub. Other properties such as EC (Electrical Conductivity), TKN (Total Kjeldahl Nitrogen), Ni and Cu also had highest levels of concentration or counts at the surface depth but did not follow this trend. Mn showed the highest occurrences at the Msub depth. The mean Ni level for the site was above the FME_{env} and WHO maximum permissible levels respectively.

Keywords: Leachate • Heavy metals • Physicochemical • Soil • Environment

Introduction

Septic leachate is the clarified effluent liquid from a septic system underground tank which is normally emptied into the soil absorption area for further treatment in many developing nations. Its composition may include heavy metals, microorganisms, fats and oils, protozoa, and inorganic macro-components such as magnesium, calcium, sodium, potassium, iron, chloride, sulphate depending on the soil properties and how frequently the leachates infiltrate the soil absorption area [1].

Sadly, some soils in Africa which serve as soil absorption areas have soil properties ill-suited to accomplish large scale dispersal of leachates. In addition, effluents are generally treated over a small soil percolation area. Therefore, the treatment capacity of the soil is frequently exceeded resulting in contamination. Worse still, the very soil media that should provide additional leachate treatment area is lost to trench excavation to allow for septic tank component installation.

These drawbacks in addition to limitations in requisite bedrock and ground water level geologies and the density of septic tanks situated in an area (which is often high in most urban areas of Africa), frequently lead to mal-functions and failures of domestic septic tank systems. Released heavy metals and microorganisms in pollutant effluents or leachates profit from subsurface transport due to incomplete degradation or sorption/filtration by the soil thereby altering soil biota and eventually ending up in aquatic ecosystems [2]. In this way, septic systems become significant contributors of heavy metals and fecal pollution to surface waters and to domestic wells especially in high septic tank density areas.

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Materials and Methods

Study area

The study area (Figure 1), International Secondary School, is situated within the premises of Rivers State University, Nkpulu Oroworukwo, Port Harcourt and lies within the following projected coordinates: Universal Transverse Mercator (UTM), Zone 32N, World Geodetic System (WGS) 84; 276010.902 m & 531656.115 m; 276315.407 m & 531687.070 m; 276312.875 m & 531522.450 m; 276011.803 m & 531520.308 m (Eastings & Northings respectively) while the septic tanks of interest had details listed in Table 1.

The city, Port Harcourt, is metropolitan with a population of approximately 3 million sitting on approximately 181km² of land with a sub-equatorial wetland climate, a rainfall of about 2000mm and annual mean temperature of about 29^oC. Its surface cover geography may be summarized as a porous and permeable soil surface during the rainy season which allows waste leachates access to the underlying soil layers as well as bedrock ground water levels [3,4].

Field activities

Samples were collected on 7th September, 2020 from 6.30am to 7pm using a modified hand soil auger from vertical depths 0.0 m (surface or top soil), 0.5 m (sub soil), 1.0 m (deep subsoil) and 1.5 m (Maximum sub surface soils) in quadruplicates, for each 0.5 m, 1.0 m, 1.5 m and 2 m distance away from the soakaway sides. A total of 128 samples were collected, thoroughly homogenized, stored in polyethylene bags and used for physicochemical determinations.

Methods of analysis

The soil samples for the physicochemical analyses were air dried, ground and sieved through meshes (ASTM E-11 specification) to remove coarse particles and obtain particle size < than 2 mm. Soil pH and EC were determined on a 1: 2.5 (w/v) soil/ water slurry and ISO 11265:1994 respectively.

10g of sample was weighed on a Mettler Toledo ME 204 Analytical balance, correct to four decimal places and transferred quantitatively into a 100 ml beaker. 25 ml of deionized water (conductivity< 0-1.0μS/cm) was added and the content stirred on an electromagnetic stirrer for 30 mins. The slurry was allowed to settle for 30 mins and the pH measured with a Thermo Scientific Orion Star A211 temperature compensated pH meter, pre-calibrated with OEM provided 3-point buffer (4,7 and 10) standards. For EC, 10g of

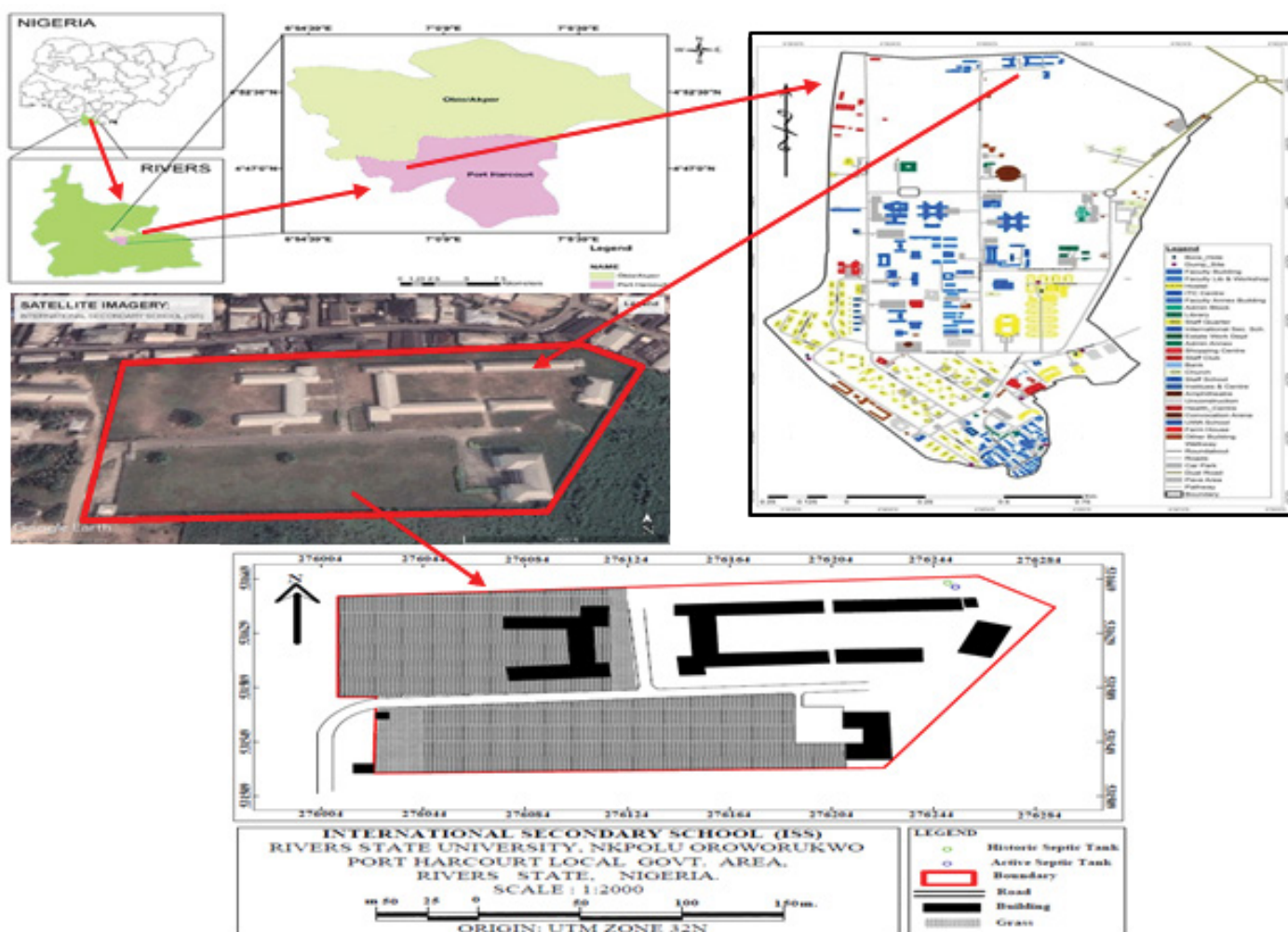


Figure 1. Map showing the study area and sampling points.

Table 1. Relevant information on the septic tanks of the study.

Septic Tank Id	Colour on Map	Status	Dimension			Geo-location	
			L & B (m)	Perimeter	Area	Northing	Easting
1	Green	Historical	5.47 m × 2.75 m	16.44 m	15.04 m ²	531666 m	276250 m
2	Blue	Active	5.45 m × 1.95 m	14.80 m	10.63 m ²	531663 m	276253 m

sample was weighed on a Mettler Toledo ME 204 Analytical balance to four decimal places and transferred quantitatively into a 300ml beaker. 50 mls of deionized water was added to the beaker and the content stirred on an electromagnetic stirrer for 30 mins. The suspension was allowed to settle for 30mins and the conductivities determined using a Fischer 145A multi-function conductivity meter.

Total Kjeldahl Nitrogen using ISO 11261:1995 soil Quality-2016 adapted method by accurately weighing and transferring 10g of soil sample into a 500 ml kjedahl flask. 10g Na₂SO₄, 0.6g of anhydrous CuSO₄ and 30 ml of conc. H₂SO₄ were added gently to the flask. The content was digested on LabConco block digester auto set to 410 oC for 1hr, allowed to cool and then quantitatively transferred into a 500 ml volumetric flask slowly and made up to mark. 100 ml of this made-up solution was transferred into a 500 ml round bottom distillation flask. 30 ml of 30% (30 g NaOH pellets in 100 ml distilled water) NaOH solution was added to the flask and some zinc metal powder as anti-bump and set on an electrothermal heating mantle. To another 250 ml graduated Erlenmeyer flask was added 100 ml of freshly prepared 0.4% (40g of boric acid granules in 1liter of pre heated H₂O) H₃BO₃ solution and 3 drops of methyl orange indicator and into which the receiver of the distillation unit immerses. The distillation was then initiated and 100ml of distillate collected into the Erlenmeyer flask containing the boric acid. The received solution was

then back-titrated against 0.2N H₂SO₄ from a yellow to a pink end point. The titre was value was recorded.

Total Phosphate was determined using a Modified Troug Method. 10g of soil sample was weighed into a 250 ml beaker containing 200 ml deionized water. 2.5 ml of 0.025M H₂SO₄ was added to the content of the beaker and stirred for 30 mins. The slurry was allowed to settle clearly and then filtered through Whatman No.42 filter paper into a 400 ml beaker. Thereafter, 100 ml of filtered solution was collected and used as working volume. 5 ml of Ammonium Molybdate solution (48g/l) prepared by dissolving 48grams of [(NH₄)₆ MO₇O₂₄·4H₂O] in 800 ml preheated distilled water to which 2.5 ml of NH₄ (OH) and distilled H₂O was added to make it up to 1 litre mark was added and 5 ml of Amino solution made by dissolving 18.5g Sodium Sulfite, 0.50g 1-Amino-2-Napthtol-4-sulphonic acid and 31 g of Sodium Metabisulphite (Na₂S₂O₃) were added in that order. The absorbance of the reaction mixture was read at 650 nm after 8mins on a Hach 3900 series UV-Visible Spectrophotometer (Modified Troug Method).

Results and Discussion

Levels of physicochemical parameters

The Descriptive Statistics Summary of the means with their standard

deviations and the ranges of the Physicochemical Parameters were determined using Minitab Statistical Software (Version 20) on the experimental data from soil analysis. The obtained results are presented in Table 2 while the variations of their levels with respect to the vertical depths and horizontal distances are presented in Figures 2-6.

pH

The variation of soil pH (Figures 2; IA and IB) show that the mean pH levels ($M \pm SD$) followed the order 5.55 ± 0.49 (Surf) $> 4.88 \pm 0.39$ (Sub) $> 4.83 \pm 0.59$ (deep sub) $> 4.73 \pm 0.52$ (Msub) respectively indicating a decrease in soil pH. The decrease may be attributed to leaching caused by high amounts of rainfall containing soot or carbon particles from anthropogenic sources. It may also have been due to anaerobic digestion or decomposition of soil organic matter which is richer at the soil surface.

Furthermore, the site mean pH was 5.01 ± 0.587 which indicates that the soil is acidic to slightly acidic. Although there is paucity of data with respect to pH variation with vertical depths at three or more sample depths (beyond the 1.0 m depth) in Nigerian soils, the mean surface pH obtained from this study (5.55) was in agreement with those reported in previous multi-depth studies. According to Rahman MA, et al. [5], acidity in this range (< 5.5 and < 3.8) frequently aids Mn oxides and Fe_2O_3 solubilization, leading to predominance of free metal cations which play a key role in phytotoxicity. The observed acidity, may also have contributed to the observed decrease in phosphate down the soil depth as well as aided fungal survival at the Max sub depths as reported by the authors elsewhere. On the other hand, the pH was highest at the distance closest to the side of the soakaway (0.5 m) and lowest at the farthest point

Table 2. Table showing levels of all the studied soil properties in the sample.

Parameter/Units	Mean \pm SD	Range	WHO/FAO/FMEv Max Limits
Ph	5.02 ± 0.59	4.31 – 6.94	6.0 -7.5 (FAO)
EC ($\mu S/cm$)	32.11 ± 20.78	8.70 – 132.50	-
Total Nitrogen (%)	0.083 ± 0.02	0.06 -0.13	-
Total Phosphate (mg/kg)	35.39 ± 5.19	27.14 – 49.74	-
Zn (mg/kg)	34.70 ± 17.33	8.30 – 73.00	50- 300 mg/kg (WHO)
Mn (mg/kg)	134.10 ± 109	9.50 – 350.30	300 mg/kg (WHO)
Fe (mg/kg)	3409 ± 3537	732 – 15.821	5000-50,000 mg/kg (FMEv/WHO)
Ni (mg/kg)	34 ± 23.42	0.00 – 99	30 mg/kg (FMEv)
Cu (mg/kg)	41.33 ± 22.09	14.4 – 113.70	50 mg/kg (FMEv)

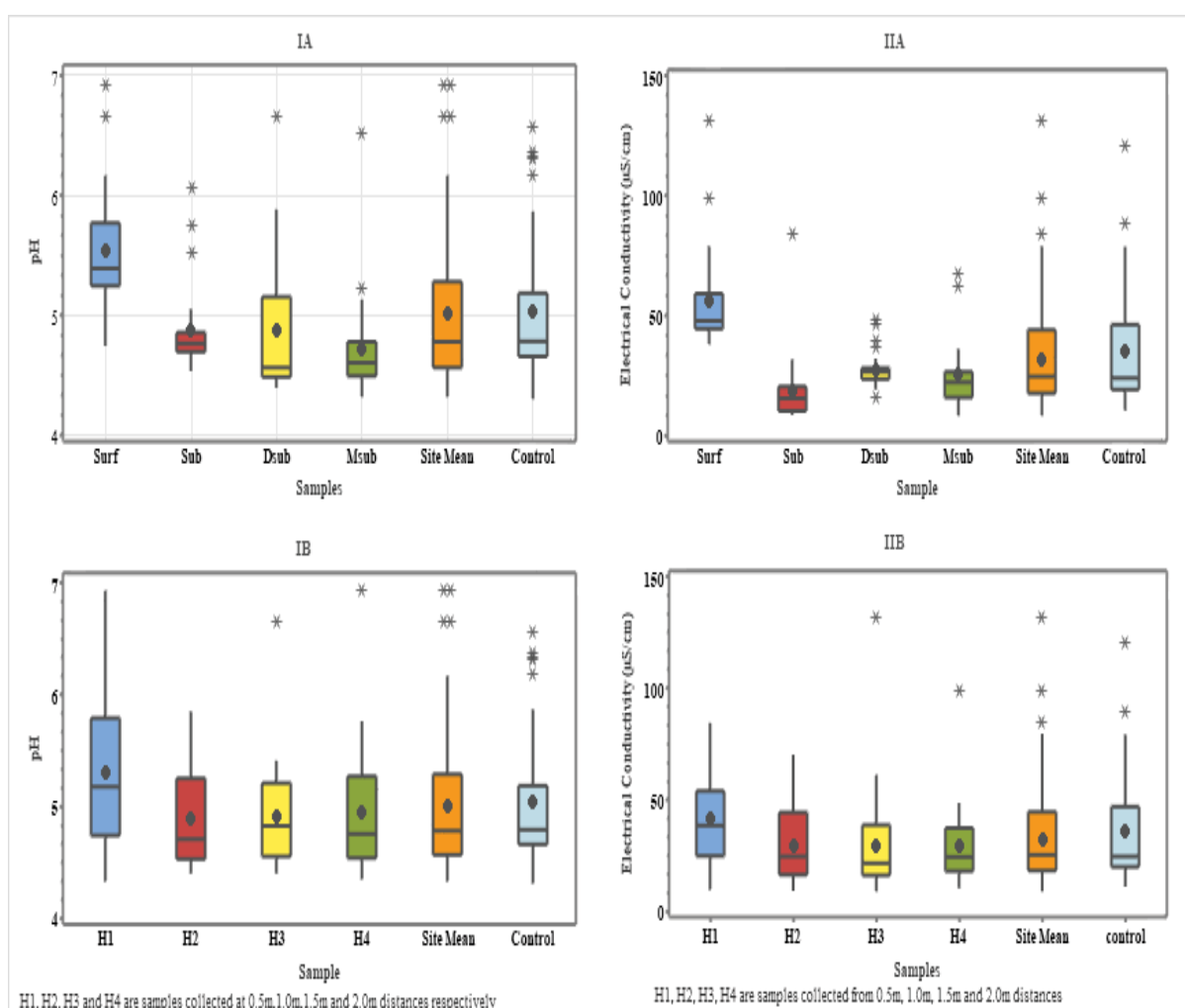
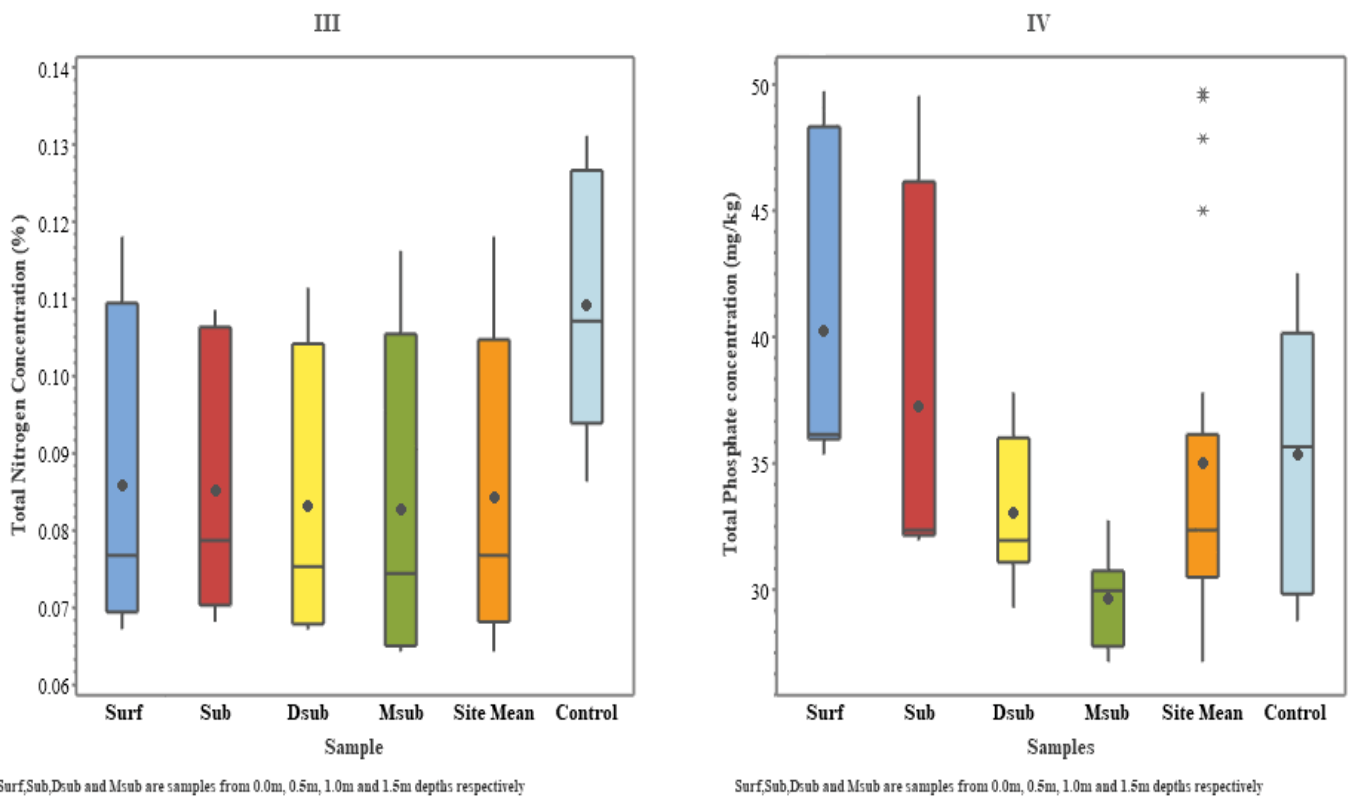


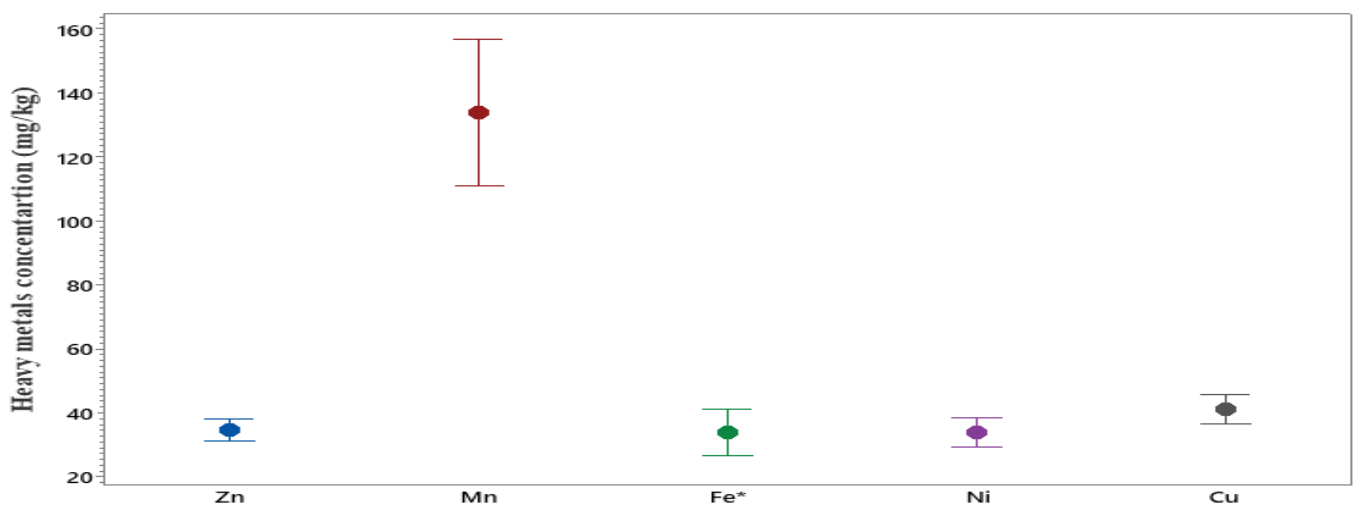
Figure 2. Boxplot of the variation of pH and EC of the soil sample across the vertical depths (1A and 2A) and horizontal distances (1B and 2B), the site mean and the experiment control. The boxes extend from the 25th to the 75th percentiles; the whisker represent the lowest and highest coefficient; the horizontal line and the black circle indicate the median and mean respectively ($n=128$).



Surf,Sub,Dsub and Msub are samples from 0.0m, 0.5m, 1.0m and 1.5m depths respectively

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Figure 3. Boxplot of the variation of total nitrogen (III) and phosphate (IV) of soil samples across the vertical depths, site mean and experiment control. The boxes extend from the 25th to the 75 percentile; the whisker represent the lowest and highest coefficients; the horizontal line and the black circle indicate the median and mean respectively (n=32).



Note : Mean levels are Zn = 34.70mg/kg, Mn= 134.08mg/kg, Fe= 3409mg/kg.7, Ni= 34.00mg/kg and Cu= 41.33 (mg/kg)
Fe* = n x 100 (mg/kg)

Figure 4. Box plot showing variation in mean concentration of heavy metals for the site.

(2.0 m) with respect to the horizontal distances which may indicate that soil organic and mineral matter as well as weathering influences may be uniformly distributed across the study site horizontally.

For EC, the range of values were within the WHO recommended safe values and may be an indication of very low presence of dissolved salts as well as anthropogenic interferences.

The order of variation of the Electrical Conductivity with depth was surf>sub>dsub>Msub, showing a general decrease of EC with vertical soil depth while for distances away the observed mean ECs indicated that ECs were higher at the point closest to the septic leachate plume. That it remained unchanged, thereafter, between 1.0 m and 2.0 m away from the soakaway may suggest absence of horizontal mobility of ions, as well as possible presence of very similar biota and abiotic factors within these points.

Nitrogen was low at the site and did not significantly change with soil vertical depth while mean phosphate concentrations 40.25 ± 6.66 mg/kg, 37.26 ± 7.91 mg/kg, 33.04 ± 3.06 mg/kg and 29.67 ± 1.96 mg/kg respectively for surf, sub, deep sub and Max sub depths were moderately high, and indicate that the phosphate content decreased with depth. However, the increasing soil acidity with soil depth may also have enhanced solubilization of the adsorbed phosphate making the phosphate available and liable to leaching with increasing soil depth as recorded in this study.

Heavy metals

The variation of the mean levels for the heavy metals, presented in Figure 4 showed the decreasing heavy metals content order $Fe \gg Mn > Cu > Zn > Ni$. This is close to the order of relative mobility of the cations of this study through soil as previously reported. Zn is known to be more mobile in acidic conditions

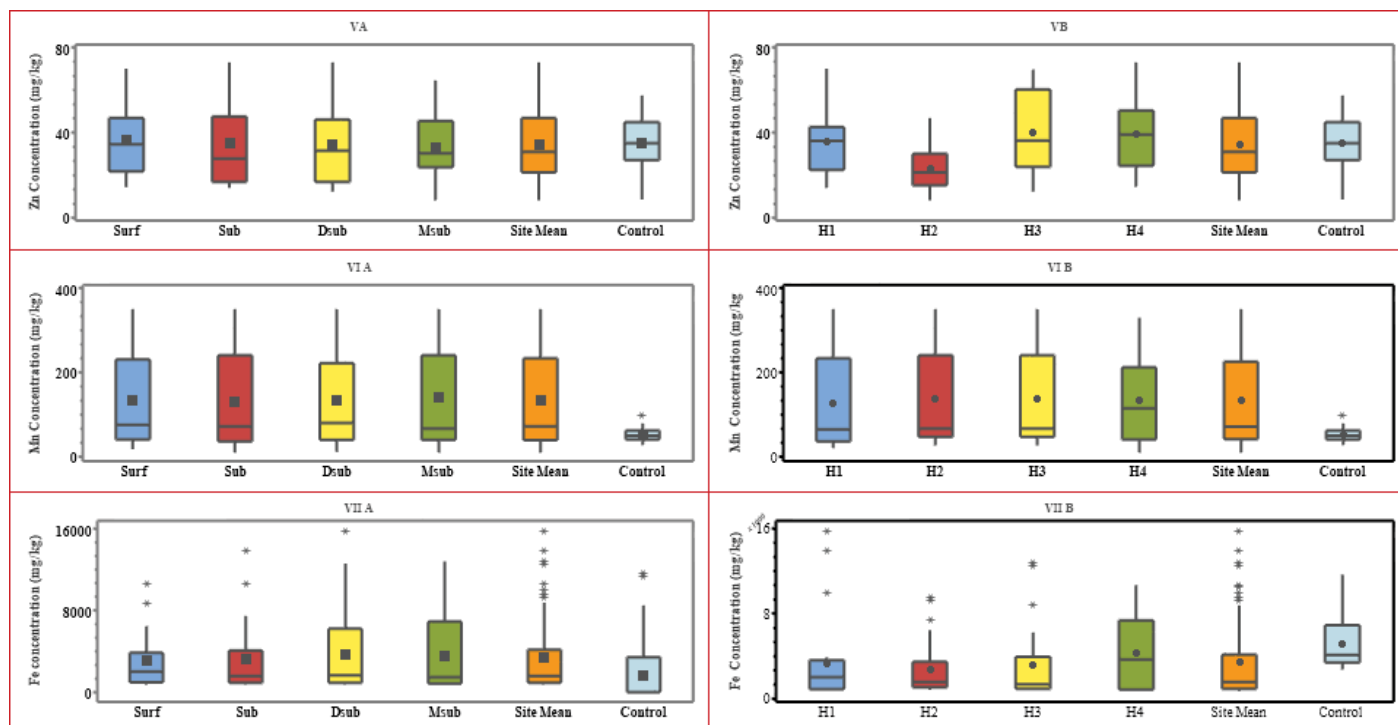


Figure 5. Boxplot of the variation of Zn, Mn and Fe across the vertical depths (VA, VIA, VIIA) and along horizontal distances (VB, VIB and VII B), site mean and experiment control. The boxes extend from the 25th to the 75 percentile; the whisker represent the lowest and highest coefficients; the horizontal line and the black circle indicate the median and mean respectively (n=128).

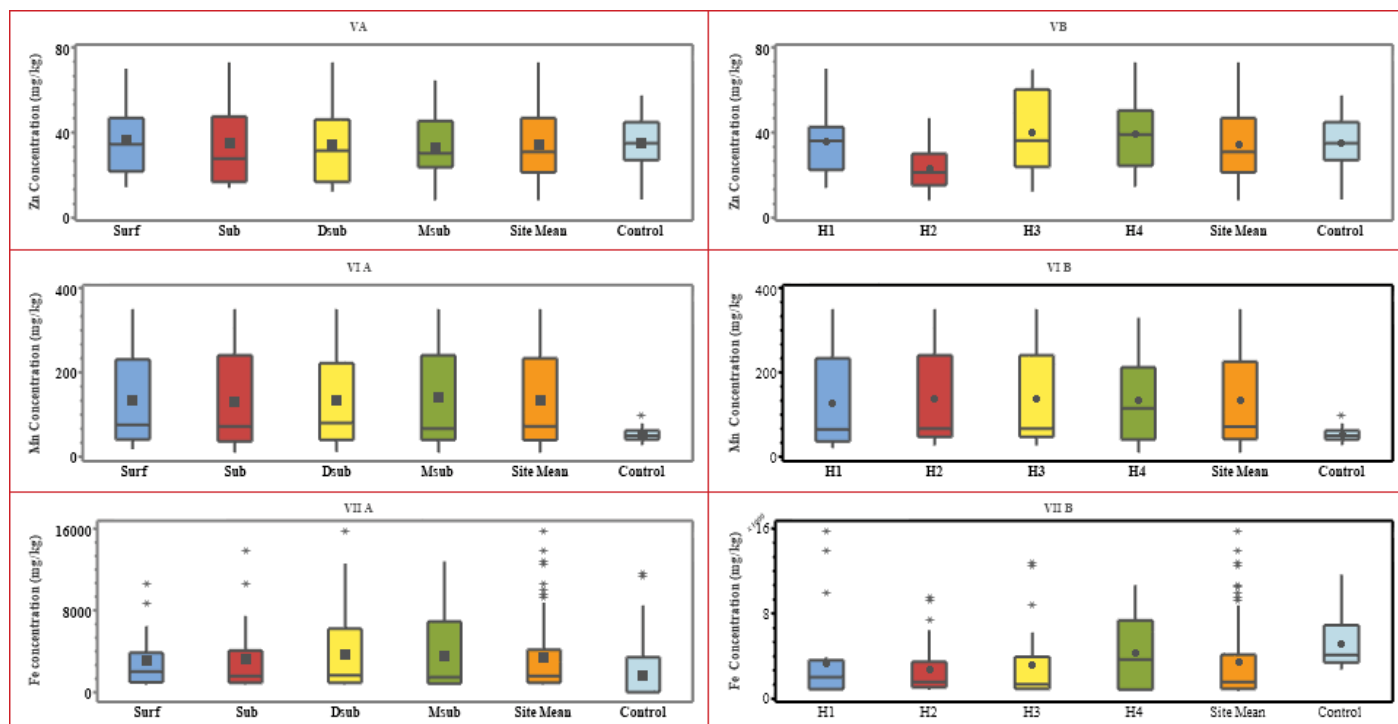


Figure 6. Boxplot of the variation of Ni and Cu across the vertical depths (VIII A and IXA) and along Horizontal distances (VIII B and IX B), site mean and experiment control. The boxes extend from the 25th to the 75 percentile; the whisker represent the lowest and highest coefficient; the horizontal line and the black circle indicate the median and mean respectively (n=128).

supporting its lower concentration than Zn. Ni behavior in soil solution which is copiously influenced by Mn and Fe oxides chemistry may account for its low values and may have been the most mobile due to formation of stable complexes with soluble organic matter. Furthermore, Cu, Zn and Ni showed a decrease of their levels with increasing soil vertical depth probably by being more tightly bound at the surface soils to soil organic matter and clay minerals. It may also have resulted from the preferential adsorption of hydrolysed metal species produced by the increasing acidic soil solution with increasing soil

depth compared to their free metals [6], the presence of strongly oxidizing soil environment and/or Fe rich particulates which made these cations more mobile. The greater retention of Cu content may indicate low presence of soluble Cu – Organic Ligand complexes which are a common route of Cu mobility in soils.

Zinc: The site mean concentration variation of Zinc (Figure 4), showed that the Zn content was highest at the surface depths and lowest at the Msub

depth (Surf>Sub>Dsub>Msub). This decrease suggests an initial reversible adsorption of Zn through the formation of weak complexes with organic matter at the surface soils, followed by later desorption into soil solution as the soil conditions became more acidic.

However, the low desorption (in terms of difference between the surface and Msub depth concentrations, 3.17 mg/kg) suggests the underlying presence of irreversibly bound and probably non-exchangeable lattice entrapped Zn. This is plausible because this study's results show higher desorption of Cu (10 mg/kg) between the surface and Msub depths even though Zn is known to be the more mobile heavy metal of the two in acidic soil conditions, suggesting indeed that the clayey soils of the 1.0m-1.5m depths were effective in holding Zn quite strongly in agreement with previous findings.

The site mean concentrations at distances away from the soakaway were (mg/kg) 37.61, 23.84, 38.44 and 39.15 for 0.5 m, 1.0 m, 1.5 m and 2.0 m respectively indicating the order 2.0 m>1.5 m>0.5 m>0.5 m which shows the slight mobility of Zn in the horizontal direction away from the soakaway sides or accumulation through input from anthropogenic sources of Zinc such as batteries, smelting and electrical equipment.

The ranges of Zn for all the vertical depths investigated are within the WHO/FAO acceptable range of Zinc in uncontaminated soils (20-300 mg/kg).

Manganese: The manganese mean concentrations were 135.2 ± 113 mg/kg (surface), 129.9 ± 110.3 mg/kg (sub surface), 132.4 ± 104.3 mg/kg (deep sub surface) and 139.0 ± 117.6 mg/kg (Msub depth) presenting the concentration order Msub>Surf>Dsub>Sub. The ranges were slightly compatible with previous findings of 28.9-101.2 mg/kg. However, the means were higher than those reported under similar acidic soil conditions (pH range 3.67-6.7), for road side soils of Jos Metropolis, and for motor park soils in Gombe. They are however lower than 203.48 mg/kg reported for the soils and sediments along banks of River Benue, in Taraba State. The similarity in spatial vertical distribution of the mean Mn content across the depths suggests that the metal is native to this soil pedon probably from basalt, trachyte and rhyolite, while the small net Mn increase between the surface and Msub depths may indicate that a significant portion of soil Mn may be concentrated in ferromanganese concretions.

Furthermore, this study's results agree with findings which suggest that Mn is concentrated in residual sediment and soil deposits in tropical and sub-tropical environments while in humid environments Mn is leached by acidic conditions, although its distribution is rarely uniformly distributed in soil substrata (also in agreement with this research findings), it rather exists in concentric nodules, spots or concretions that imply seasonal growth.

The Mn distribution along the horizontal distance was similarly fairly unchanged. The manganese content suggests anthropogenic interference from sources such as fertilizers, fungicides, paints, fireworks and cosmetics. Manganese may therefore serve as an indicator of manganese from human sewage.

Iron: The mean total iron contents for the study area were 3108 ± 2767 mg/kg, 3230 ± 3512 mg/kg, 3695 ± 4203 mg/kg and 3601 ± 3701 mg/kg for surface, subsurface, deep sub and Msub depths respectively revealing the decreasing order, deep sub>Msub>Sub>surf. The mean values were variously different from mean values of 1534 mg/kg and 1411 mg/kg for wet and dry seasons respectively, previously reported [7] for solid waste dumpsites at Uyo in their studies. The lower Fe concentration values between the 0.0 m-0.5 m soil depth compared to the 1.0 m - 1.5 m depth, may indicate more solubilization of iron at these depths in agreement with some authors, who suggested that reducing and acid conditions enhance the mobilization of Fe compounds because the growing soil acidification with vertical soil depth enhanced the ability of the iron oxides and (hydro)oxides to release Fe into the soil solution leading to higher availability and mobility. However, it is not clear if, the increasing amounts of <0.02 mm granulometric fractions of the clay sandy loam soil system native to the study site which also generally results in

increased Fe content, played a role since iron content was higher in the Msub and subsoil depths which were more acidic than at the surface depth. Again, the closeness in the values of the Fe concentration across all the vertical depths may indicate strong iron concretions from same parent material.

For the horizontal distances, the spatial distribution of iron showed the highest concentration obtained at the 2.0 m distance while the lowest was recorded at the 0.5m horizontal distance. The order observed was 1.0 m<1.5 m<0.5 m< 2.0 m which enhances the argument of a similar HM origin for the study site.

Nickel: The mean Ni contents at surf, sub, deepsub and Msub depths were 36.65 ± 22.88 mg/kg, 35.14 ± 22.24 mg/kg, 29.24 ± 21.37 mg and 34.97 ± 27.50 mg/kg respectively, which are very similar to those reported elsewhere [8] who also noted that although Ni concentration is generally uniform across soil profiles, it is generally higher at the soil surface due to accumulation via anthropogenic activities such as industrial waste materials, lime, fertilizer and sewage sludge. According to several authors, soil pH is the major factor controlling nickel solubility, mobility and sorption while clay content, iron- manganese mineral and soil organic matter are more of a secondary importance. Therefore, the decreasing soil pH of this study may have led to increased solubility and mobility of Ni at the lower soil depths leading to the lower Ni contents at those depths compared to the surface depth in agreement with some findings as Senwo ZN and Tazisong IA [9].

The mean of the total concentrations at the horizontal distances, 0.5 m, 1.0 m, 1.5 m and 2.0 m away from the soakaway were 32.65 ± 26.81 mg/kg, 34.08 ± 23.04 mg/kg, 31.52 ± 23.97 mg/kg and 37.76 ± 20.43 mg/kg respectively which did not differ much but followed the order 2.0 m>0.5 m>0.5 m>1.0 m suggesting a similar source of nickel.

The mean total nickel concentration for the study site, 34.00 ± 23.42 mg/kg is slightly higher than the United Kingdom, urban and rural nickel concentration mean of 28.5 mg/kg, the 21.1 mg/kg previously reported as well as the FMEV Standard (30 mg/kg). Nickel accumulates in plants and when bio-accessed by humans, can enhance health challenges. It also may affect cell viability in microorganisms as well as inhibit enzyme activity by denaturing the enzyme thereby inhibiting some useful bacteria growth.

Copper: The Cu levels decreased across the vertical soil depths in the order surf>sub>dsub>Msub depth. Therefore, Cu was accumulated in the surface layers of soils which is consistent with previous work, although according to Lock K, et al. [10], it usually displays a tendency to be adsorbed by organic matter, carbonates, clay minerals, and oxy-hydroxides of Mn and Fe in deeper soil layers. Furthermore, the lower Cu content at Dsub and Msub depths may also have been aided by the more acidic soil pH at these profiles which enhanced Cu mobility as Cu shows greater solubility at pH in the 5.5 range. It is however, not clear if the high surface area of the ferro manganese oxide nodules or crusts and Cu²⁺ ability to replace Fe²⁺ once adsorbed onto these sites also aided the depletion of Cu ions, and solicits further interest.

The site mean copper concentration across all depths (41.33 ± 22.09 mg/kg) is close to the maximum permissible limit by WHO/FAO standard (50mg/kg) requires closer monitoring and early attention to avoid copper pollution. Copper ions affect the transcription mechanism in cell leading to disruptions in cell divisions [11].

Correlation

The correlation amongst physicochemical parameters for the study site (Figure 7) revealed a positive and strong correlation between Ni & Mn (r=0.74) with respect to depth and also with horizontal distance away from the soakaways. There was also a moderate and positive correlation between pH & EC (r=0.65) with respect to vertical depth. The positive relationship between Mn & Ni may be the consequence of the great ability of manganese compounds such as its minerals in soils to strongly bind bivalent cations such as nickel while that between pH & EC may indicate that they are governed by similar regulation and partition of water and solutes characteristics. The very consistently negative correlation between Fe & Mn observed in this study, is in agreement with the ability of manganese to occupy the sites of other divalent cations such as Fe²⁺.

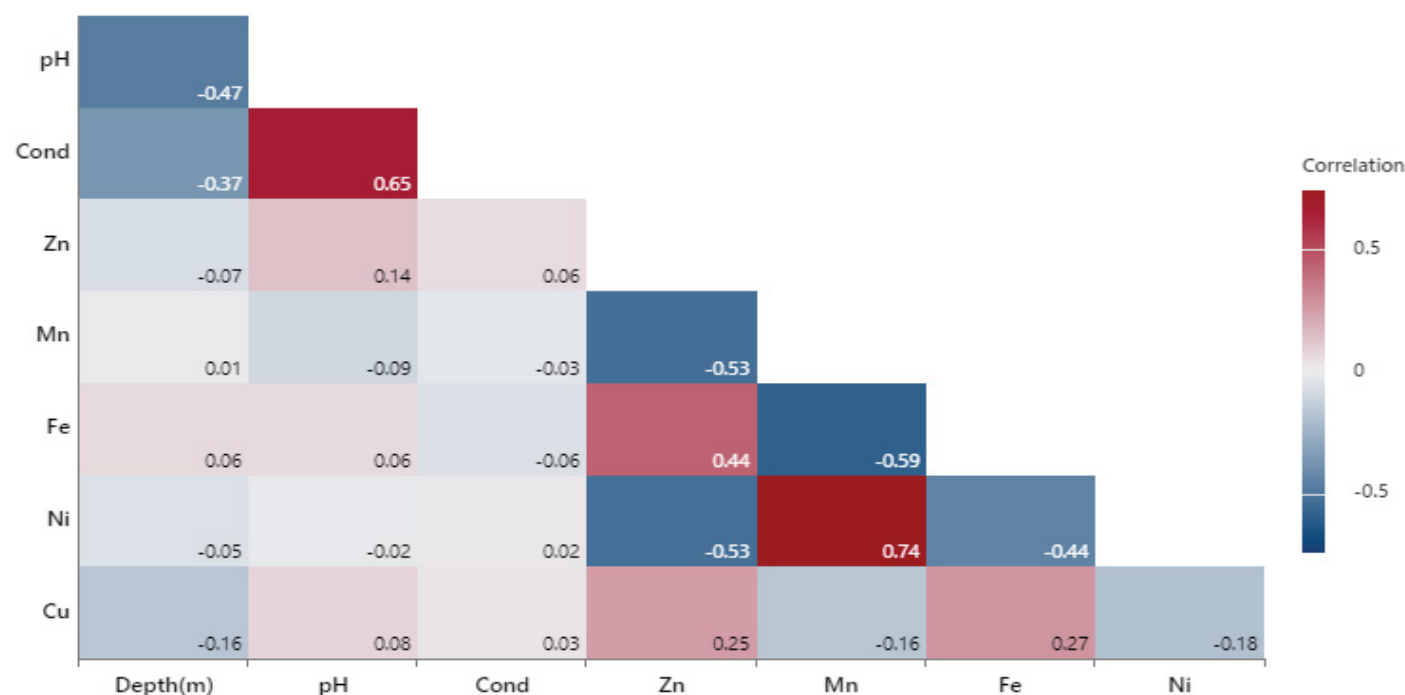


Figure 7. Correlogram showing the general relationships between pH, EC and the heavy Metals for the site. Deeper blue and red colour shades indicate higher Pearson correlation coefficient values.

Conclusion

This study revealed that the soils studied were acidic to slightly acidic and that the Nickel average level is higher than the UK urban and regional standard of 28 mg/kg, as well as the FMEV maximum permissible limit of 30 mg/kg for Ni in soils. It also showed that the Cu level is in very close proximity of the FAO/WHO maximum permissible limit for copper in soils and therefore soil protective action has to commence.

As leachates seep into lower pedons of soil, they will eventually come in contact with the ground water geology and contaminate ground and well waters and their courses. They may also be carried by rain or runoff surface water and get into rivers and other water bodies that serve human and animal needs and thereby cause serious health problems.

In the immediate, the educational or academic community around will be at great risk Nickel pollution. Productive school hours could be lost through possible illnesses. Therefore, human and animal health forms around the vicinity should be protected through additional health and environmental management interventions and accelerated planning towards implementation of a more modern sewage management system vigorously pursued to avoid leaching of septic tank effluent.

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Conflict of Interest

None.

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