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Full Length Research Paper

Determination of some polycyclic aromatic hydrocarbons (PAHs) associated with airborne particulate matter by high performance liquid chromatography (HPLC) method

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In this study, polycyclic aromatic hydrocarbons (PAHs) associated with airborne particulate pollutants of aerodynamic size 10 µm (PM10) were studied for three months, from October to December 2010 in the Vanderbijlpark area. Some PAHs are highly carcinogenic and could be more harmful when combined with inhalable PM10. A dual E-Sampler which combines the light scatter and the gravimetric filter methods was used. A 10 mg/L standard stock solution that contained naphthalene (Naph), 2-methyl naphthalene (2-MNaph), phenanthrene (Phe), anthracene (Anth), benzo(b)fluoranthene (BbFl), benzo(k)fluoranthene (BkFl), benzo(a)pyrene (BaPy) and dibenzo(a,h)anthracene (DiBahAn) was prepared, compounds were indentified and quantified with an Agilent high performance liquid chromatography (HPLC). A dichloromethane (DCM) and n-hexane (1:1) extraction mixture was used to extract the pollutants from both exposed and unexposed (blank) filters. Detection limits obtained ranged from 0.001 to 0.0305 mg/L and R-values ranged from 0.996 – 0.999. Very good percentage recoveries were obtained with the lowest 97.63% and highest 101.57% associated with DiBahFl and 2-MNaph, respectively. Total concentration of 2-MNaph obtained per month were 325.2 ng/L (October 2010), 162.4 ng/L (November 2010) and 381.2 ng/L (December 2010). Relatively high levels of 2-MNaph were detected consistently when compared with other pollutants in the three months. Concentration ranges of other PAH compounds were Anth (7.2 - 14.76 ng/L), BbFl (6.7 - 13.6 ng/L), BaPy (6.8 - 13.0 ng/L) and BkFl (6.7 - 10.8 ng/L). Daily and monthly mass concentration levels obtained were lower than the strict regional daily limit of 0.075 µg/m^3, as well as national and international daily limits of 0.150 µg/m^3. These results could be used as the basis for undertaking a comprehensive study on the status of these organic compounds from the heavily industrialized Vaal Triangle region.

Key words: Polycyclic aromatic hydrocarbons (PAHs), PM10, distribution, concentration, high performance liquid chromatography (HPLC), Vanderbijlpark.

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are a class of organic pollutants which are known to be carcinogenic to animals (Hailin et al., 1998; Menzie et al., 1992). They are lipophilic and present in the atmosphere due to emissions from vehicles, residents heating systems that utilize coal, wood, gas and industrial processes

(Söderström et al., 2005). PAHs are composed of carbon and hydrogen atoms arranged in the form of fused benzene ring as cluster, linear or angular (Maliszewska-Kordybach, 1999). They exist in the atmosphere in both vapor and particulate phase (Guo et al., 2003; Van Jaarsveld et al., 1997). PAHs with low molecular weight have propensity to be more concentrated in the vapor-phase whereas those with higher molecular weight are often associated with particulates (Maliszewska-Kordybach, 1999; Guo et al., 2003). When in air, PAHs can be transported over a long distance before they are deposited with atmospheric precipitation on soils, vegetation, sea and inland (Wania and Mackay, 1996; Wild and Jones, 1995). PAHs have been associated with adverse health effects in the human population (Halsall et al., 1994; IARC, 1987). They are linked to morphological, physiological and developmental abnormalities in test animals, increased allergic immune responses in human at low levels and may act synergically with other air toxics to cause adverse health effects (Li et al., 2005; ATSDR, 1995; Diaz-Sanchez et al., 1996; Harvey, 1991). Estimated potential doses of carcinogenic PAHs by inhalation to range is between about 0.02 and 3 µg/day with a median value of 0.16 µg/day, which is about 20 times more than the calculated food dose and about 25 times more than the potential dose with drinkable water (Menzie et al., 1992).

Vanderbijlpark city is located close to anthropogenic pollution sources such as mining, a coal power station, steel manufacturing companies, an oil refinery, major roads and fossil fuel burning residential areas. This city has been identified as one of the national air pollution hot spots according to the National Environment Management Air Quality Act 2004 (Act No. 39 of 2004) of South Africa (Vaal Environmental News, 2011). A number of reports exist on the general environmental pollution by PAHs within this city and the surrounding areas, however, not much if any has been reported on airborne pollution by PAHs.

The aim of this study was to evaluate the levels of PAHs associated with airborne PM10 particles from the Vanderbijlpark. Cities of Vanderbijlpark, Sasolburg and Meyerton form the Vaal Triangle region. These cities are also close to townships such as Sebokeng, Evaton and Sharpville.

MATERIALS AND METHODS

Sample collection, preparation and analyses

Sample collection

A particulates E-Sampler from Met One Instruments Inc, Oregon United States of America, was used to collect PM10 samples (Figure 1). The E-sampler is a dual technology instrument that combines the real time measurement of the light scatter method and the gravimetric filter method where particles are pre-concentrated. An internal rotary vane pump draws air at the rate of 2 L per minute (LPM) into the visible laser light sensing chamber used for measuring the number of particles in a particular volume of air. The instrument is capable of making 40 measurements per second and averaging them to get a representative particulate data per hour, day or month. Rugged state of the art electronics measure the intensity of the focused light and output a signal to the central processing unit (CPU).

The E-Sampler was placed in the backyard of a suburban house (26°42′01″S, 27°51′15″E) and sampling occurred in three consecutive months from October to December 2010. Continuous daily samples were averaged every 30 min and then arranged per month. Data measured on the day which experienced the highest PM10 loading was further arranged per hour to study the diurnal distribution. High purity quartz filters (20 x 25 cm Whatmann 41, Whatman Corp, USA) were used and dried in a desiccator overnight. A mass of filter paper was weighed before and after exposure to attain the total particulate mass loading per sampling period on a Mettler Toledo AG245 analytical balance.

Standard and sample preparation

A 1.0 mg/L standard stock solution was prepared by dissolving 1.0 mg of each of Naph, 2-MNaph, Phe, Anth, BbFl, BkFl, BaPy and DiBahFl PAH compounds (Dr. Ehrenstorfer Reference Materials, Atlanta, U.S.A.) in 1000.0 ml acetonitrile solvent. All solvents used were of analytical grade and were obtained from Sigma-Aldrich, Johannesburg, S.A. A series of calibration standard solutions ranging between 0.01 and 1.0 mg/L were prepared from the stock solution.

A benchmark method for the extraction of PAHs from soils and sediments (was adopted during this study (Lau et al., 2010). The soxhlet extraction apparatus were cleaned by evaporating methanol at 180°C in a rotaevaporator three times, followed by three times evaporation of the extraction mixture of dichloromethane (DCM) and n-hexane (1:1) for an hour. The fourth extract of the extraction solvent mixture was collected from the receiving flask for analysis. PAH compounds were extracted from both unexposed and exposed filters with the prepared extraction mixture for an hour at 180°C in the rotaevaporator. Extracts were analyzed on the same day or stored in amber sample bottles in a fridge at below 4°C temperatures. About 20.0 µl of sample were injected five times per sample in a high performance liquid chromatographic (HPLC) instrument.

Instrumentation

An Agilent 1100 model HPLC (Agilent Technology Inc, Santa Clara, California, U. S. A) with a programmable wavelength diode array and ultraviolet (on 254 nm) detectors were used. Operating conditions were: sample volume = 20.0 µL, run time = 25 min, flow rate = 1.0 mL/min, column temperature = 23.0°C (ambient), column = eclipse XDB-C18 column (4.6 mmID x 250.0 mm (5.0 µm) 80.0 Á), mobile phase = 50.0% DCM and 5.00% n-hexane.

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Figure 1. Filter loading area of the E-Sampler (Model 9800 Rev G).

Table 1. Average daily and monthly PM10 mass concentration results levels (mg/m^3).

Day of the month	2010		
	October	November	December
1	0.00067	0.00042	0.00025
2	0.00038	0.00004	0.00025
3	0.00033	0.00033	0.00067
4	0.02283	0.00063	0.00042
5	0.01477	0.00033	0.00054
6	0.03554	0.00021	0.00067
7	0.00121	0.00046	0.00071
8	0.00079	0.00008	0.01413
9	0.00017	0.00021	0.01158
10	0.00021	0.00001	0.00573
11	0.00025	0.00002	0.00688
12	0.01225	0	0.00013
13	0.01396	0.02046	0.00042
14	0.00008	0.01883	0.00071
15	0	0.0001	0.00021
16	0.00183	0	0.00017
17	0.00425	0.00046	0.00042
18	0.00225	0.00108	0.00021
19	0.00975	0.00146	0.00004
20	0.048	0.00188	0.00001
21	0.00204	0.00104	0.00002
22	0.01542	0.00029	0
23	0.00967	0.00046	0.00004
24	0.0001	0.00021	0.01387
25	0.0007	0.00013	0.01667
26	0	0.00008	0
27	0	0.00633	0.00001
28	0.01809	0.02754	0.00017
29	0.02196	0.00046	0.00025
30	0.00038	0.00021	0.00004
Sum	0.23788	0.08376	0.07522

RESULTS AND DISCUSSION

PM10 mass concentration

Average daily and monthly PM10 mass concentrations for three months are shown in Table 1. The highest mass concentration data per month were: 0.048 mg/m^3 (20th), 0.03554 mg/m^3 (6th), 0.02283 mg/m^3 (4th) and 0.02196 mg/m^3 (29^{nth}) in October; 0.02754 mg/m^3 (28th); 0.02046 mg/m^3 (13th) in November and the December data were all below 0.02 mg/m^3. These results are also in agreement with those obtained in summer seasons as reported (Moja et al., 2012). Average daily mass concentrations were below the stringent Vaal Triangle regional standard of 0.075 mg/m^3. Department of Environmental Affairs (DEA's) national daily limit together with international EPA's standard and WHO's guidelines of 0.150 mg/m^3 were also not exceeded (NAAQS, 2009; Annergan and Scorgie, 2002).

The highest daily PM10 loading was obtained on the 20th October. Hourly distribution of this data is shown in Figure 2, with the largest peak starting from 2:00 (0.0001 mg/m^3) and ending at 16:00 (0.0011 mg/m^3) with a peak maxima occurring at 13:00 (0.0043 mg/m^3). A smaller peak was measured from 18:00 (0.0011 mg/m^3) to 22:00 (0.001 mg/m^3) with a peak maxima at 19:00 (0.0021 mg/m^3). Similar PM10 diurnal distribution pattern within this study area was previously reported (Moja et al., 2012). This distribution pattern resemble the morning traffic flow to work, as well as the optimized industrial activities at around midday.

Concentrations of PAHs

Detection limits for the instrument were taken as three times the standard deviation of the lowest detectable concentration of PAHs from the mean of triplicate analyses and ranged from 0.001 to 0.0305 mg/L (Table 2).

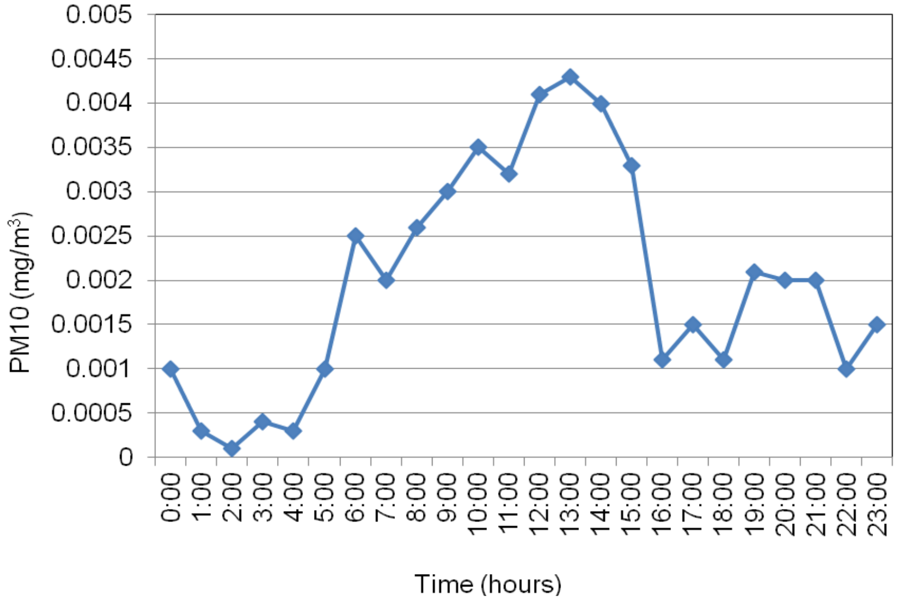

Figure 2. Hourly PM10 distribution on the 20th of October.

Table 2. Calibration data.

PAHs	Linear calibration ranges (mg/L)	Detection Limits (mg/L)	Linear equation	R^2	Recovery (%)
Naph	0.2 - 1.0	0.0305	y = 23.73x + 0.11	0.999	98.34
2-MNaph	0.2 - 1.0	0.0301	y = 27.17x + 0.06	0.999	101.57
Phe	0.02 - 1.0	0.0035	y = 335.4x + 0.04	0.998	100.39
Anth	0.01 - 1.0	0.001	y = 1135x + 0.15	0.999	100.82
BbFl	0.03 - 1.0	0.0062	y = 174.5x + 0.09	0.996	100.7
BkFl	0.04 - 1.0	0.0032	y = 132.9x + 0.12	0.999	101.01
BaPy	0.04 - 1.0	0.0057	y = 132.2x + 0.31	0.996	100.68
DiBahFl	0.2 - 1.0	0.0146	y = 35.53x + 0.14	0.999	97.63

R-values obtained during the standard calibration process ranged from 0.996 - 0.999. Very good percentage recoveries were obtained with the lowest 97.63% and highest 101.57% associated with DiBahFl and 2-MNaph, respectively.

2-MNaph and Anth compounds were detected from a prepared extracting solvent mixture at 8.967 (min) and 10.979 (min) retention times (t_R), respectively (Figure 3). Their peak areas were 249.907 for 2-MNaph and 10.6032 for Anth.

Three PAH compounds were also detected after extracting an unexposed blank filter paper with the prepared solvent mixture. Figure 4 shows a chromatogram with peaks of 2-MNaph (t_R = 8.976 min), Anth (t_R = 10.976 min) and BbFl (t_R = 11.830 min). Peak areas were 222.465 for 2-MNaph, 7.94263 for Anth and 8.17125 for BbFl. Since higher peak areas of 2-MNaph and Anth were obtained from the prepared extracting solvent mixture than from the extract of a blank filter paper, it could be concluded that the blank filter did not contain these two compounds. However, this filter did contain BbFl.

The highest concentration of PAHs standard mixture (1.0 mg/L) was used to optimize the HPLC. Standard PAH compounds had the following retention times: Naph (6.933 min), 2-MNaph (8.712 min), Phe (9.700 min), Anth (10.119 min), BbFl (13.936 min), BkFl (14.108 min), BaPy (14.454 min) and DiBahFl (14.976 min) as shown in Figure 5.

PAH compounds detected from the exposed filters are shown in Figures 6a to c. The five PAH compounds detected from all the samples analyzed in the order of appearance of the peaks on the chromatogram are 2-MNaph, Anth, BbFl, BkFl and BaPy and the retention

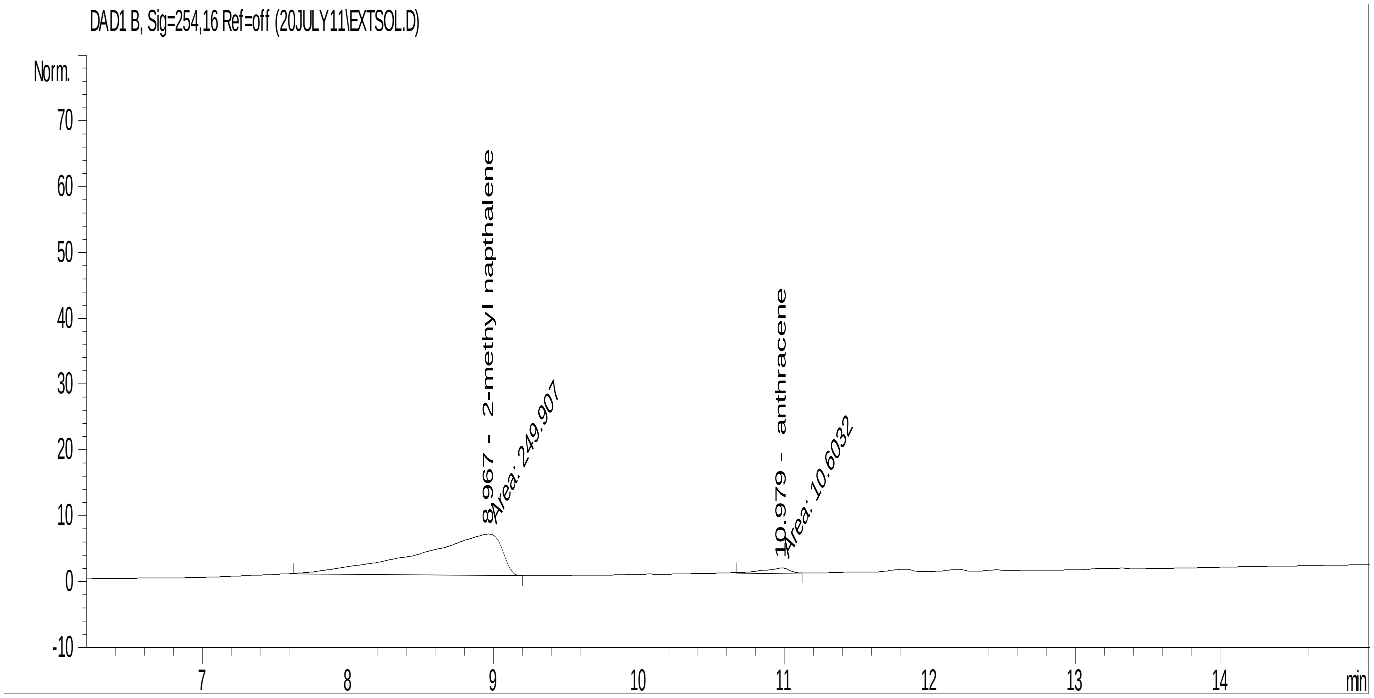

Figure 3. Chromatogram of PAHs from the prepared extracting solvent mixture.

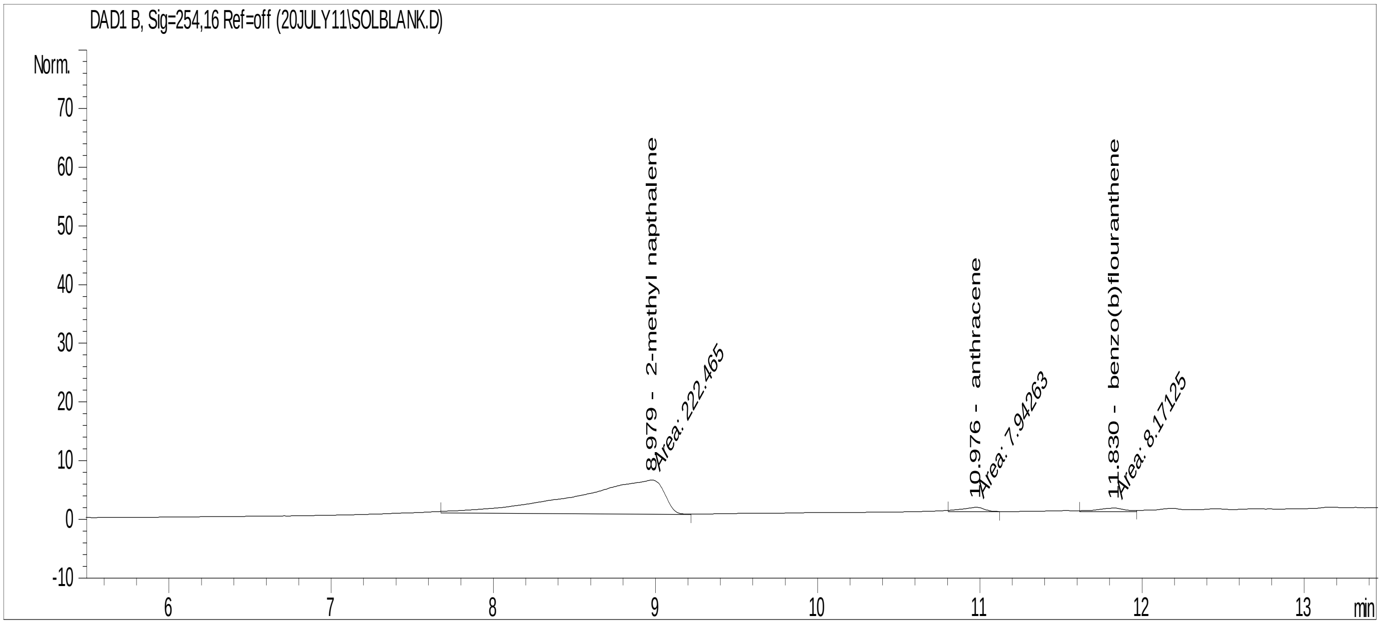

Figure 4. Chromatogram of PAHs extracted from an unexposed filter paper.

times are listed in the same order. Figure 6a had retention times: 9.007, 11.026, 11.871, 12.232 and 13.200 min. Figure 6b had 8.978, 10.998, 11.834, 12.209 and 13.288 min and Figure 6c had 9.008, 11.019, 11.869, 12.225 and 13.172 min.

Total concentration of 2-MNaph obtained per month were 325.2 ng/L (October 2010), 162.4 ng/L (November 2010) and 381.2 ng/L (December 2010). Relatively high

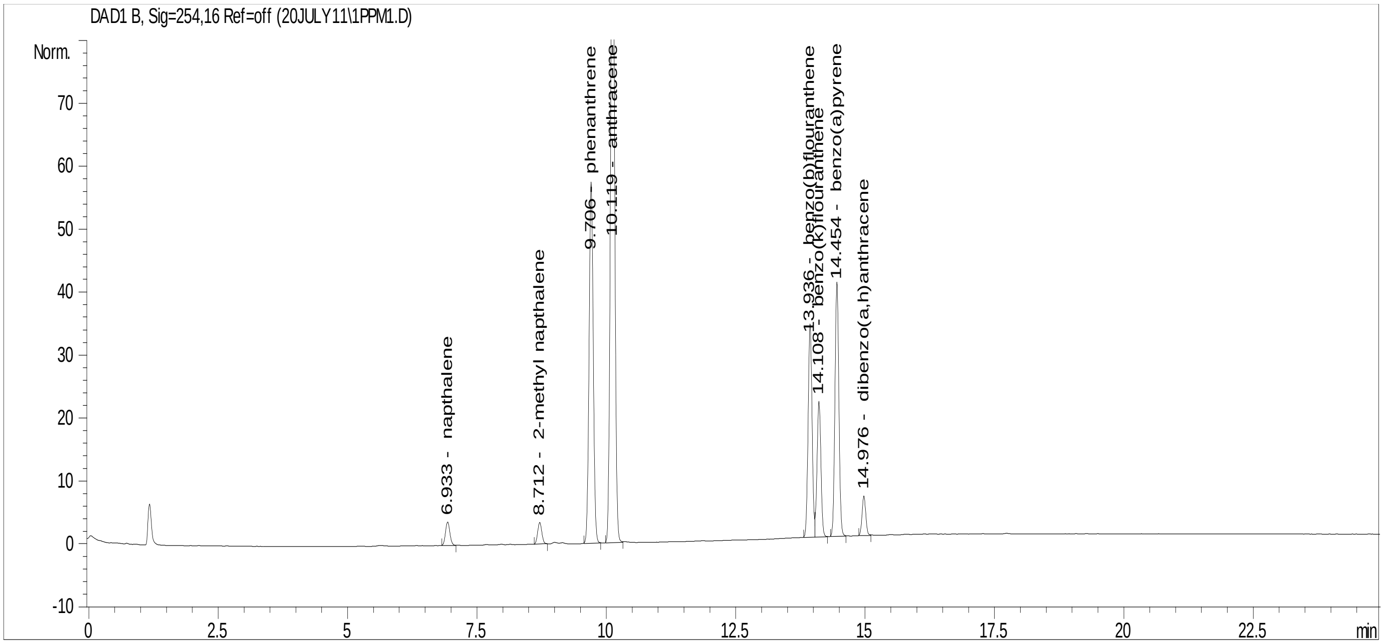

Figure 5. A chromatogram of standard PAHs mixture.

levels of 2-MNaph were detected consistently when compared with other pollutants in the three months of this study (Figure 7). Concentration ranges of other PAH compounds were Anth (7.2 - 14.76 ng/L, BbFl (6.7 - 13.6 ng/L), BaPy (6.8 - 13.0 ng/L and BkFl (6.7 - 10.8 ng/L).

2-MNaph is formed through the distillation of methylnaphthalene, where 1-methylnaphthalene is removed leaving behind 2-MNaph (ATSDR, 1995). Mixtures containing 2-MNaph are used in the formulation of alkyl-naphthalenesulfonates (used for detergents and textile wetting agents), chlorinated naphthalenes, and hydronaphthalenes (used as solvents). Pure 2-methylnaphthalene is a component used in the manufacture of vitamin K and the insecticide carbaryl (1-naphthyl-N-methylcarbamate) (HSDB, 2002). Possible PAH sources within the study area would include combustion activities from a coal power station, coal using steel manufacturing companies, an oil refinery and diesel and petrol powered vehicles or informal burning of solid waste. Another possible source could be insecticides used at a nearby farm area.

Since inhalable PM10 particles are a cause of health concern on their own, the elevated presence of 2-MNaph adsorbed on the surfaces of these particles could exacerbate these health effects (ATSDR, 1995; Diaz-Sanchez et al., 1996; Harvey, 1991; Vaal Environmental News, 2011; Lau et al., 2010; Moja et al., 2012). Collectively, PAHs cause skin irritation and inflammation, while Anth, BbFl, BkFl and BaPy cause adverse health effects to humans (ADH, 2009; WHO-IPCS, 1998). BbFl and BaPy are known as animal carcinogens, but together with BkFl are also classified as possible human carcinogens (HSDB, 2002; WHO-IPCS, 1998; Wang et al., 2011).

This study successfully evaluated the levels of some PAHs association with airborne PM10 particles within the Vanderbijlpark study area. Based on these results and the outcome of other similar studies (Liu et al., 2006), airborne PAHs and PM10 pollutants could be used as indicators of urban air pollution.

Conclusion

Average daily and monthly mass concentration levels obtained were lower than the strict regional daily limit, as well as national and international daily limits.

The presence and wide distribution of some PAHs within the Vanderbijilpark environment is a major challenge Moja et al. (2013) reported the presence of some carcinogenic compounds such as BbFl and InPy in water samples used for domestic and agricultural purposes in the same study area. The current study also reports the existence in the atmosphere of PAH compounds that could cause adverse human health effects, for example, Anth, BbFl, BkFl and BaPy. The association of respirable fraction of airborne particles with toxic PAHs elevates local air to dangerous levels. These results could be used as the basis for undertaking a comprehensive study on the status of these organic

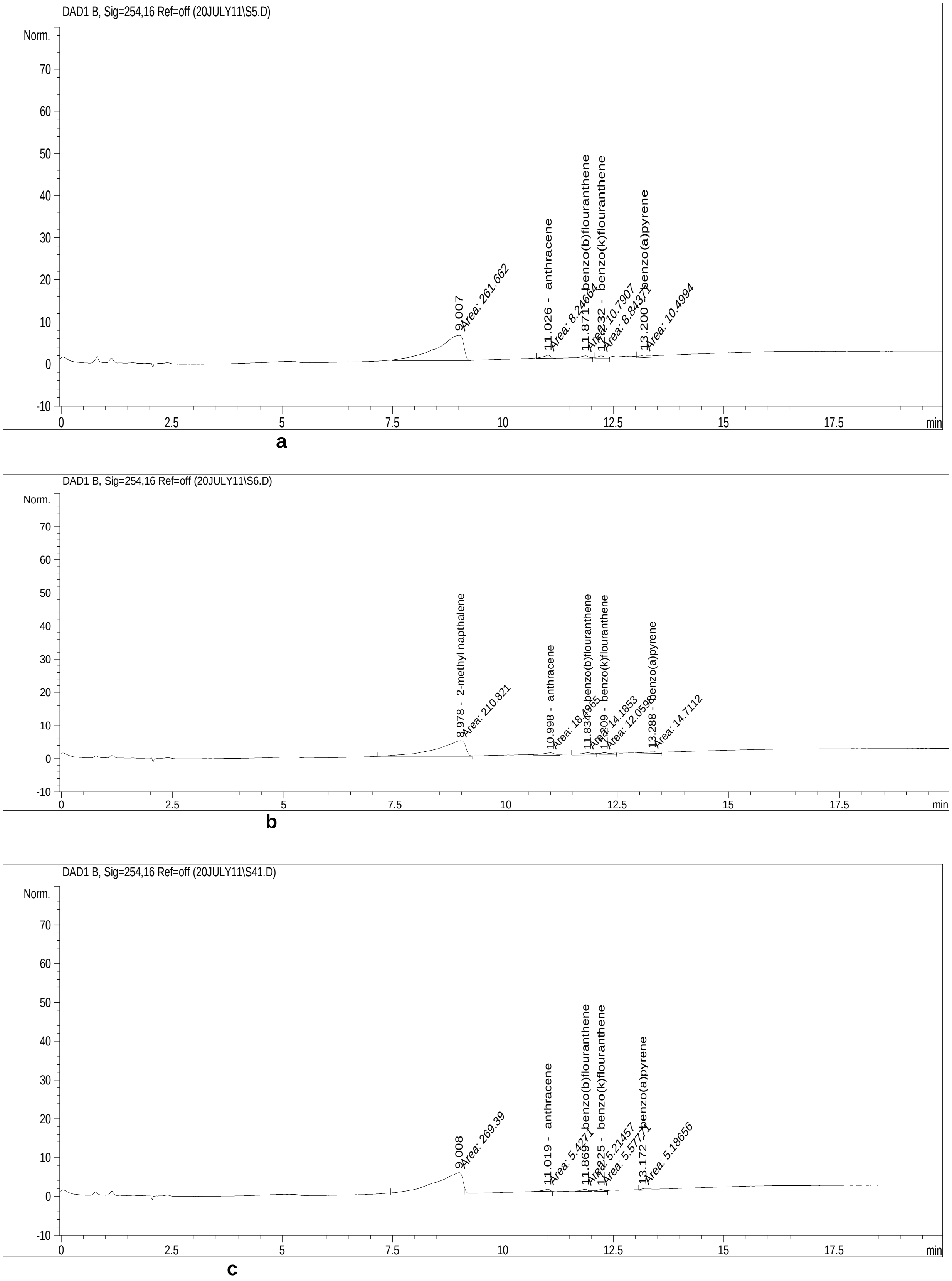

Figure 6. Chromatograms of (a) October sample, (b) November sample and (c) December sample.

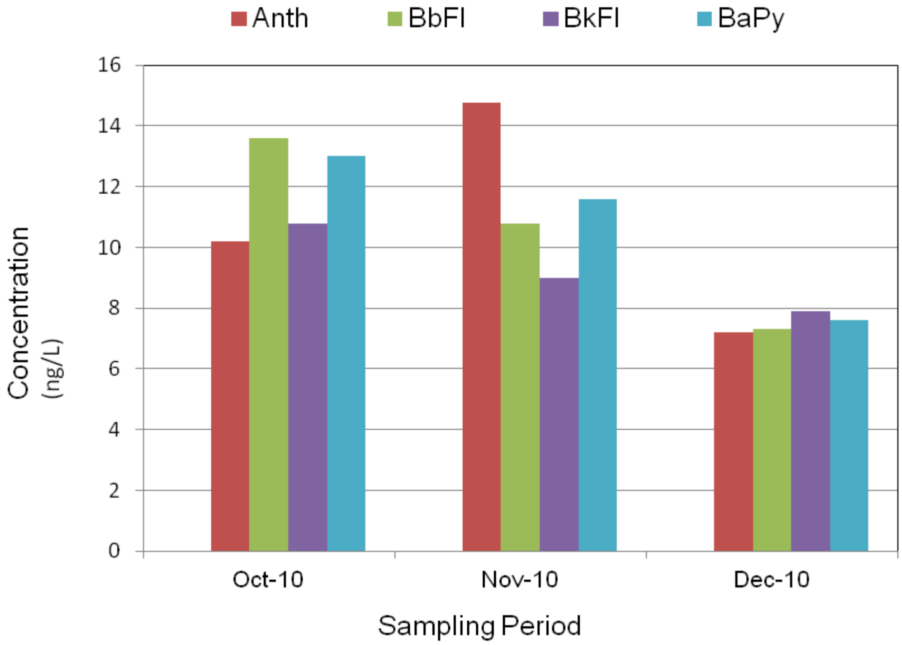

Figure 7. Concentration of different PAH compounds detected from samples.

compounds from the heavily industrialized Vaal Triangle region.

Conflict of Interests

The author(s) have not declared any conflict of interests.

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Household-scale environmental health in the Ezulwini Valley, Swaziland

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An environmental health assessment of 210 households located in four communities in the Ezulwini Valley, Swaziland, is summarized. The assessment focused on household-scale environmental health in the context of four key resource sectors: drinking water, energy, solid waste and human waste, with availability and perceived adequacy considered for each sector. The survey was administered in the field by small teams of students alongside Swazi community members, utilizing a snowball sampling strategy with stratification by economic class. Electronic administration via mobile devices assisted in geolocating records, minimizing entry error and rapidly compiling results for daily review and analysis. Results indicate challenges in household access to basic resources and resource impacts, even in this relatively developed part of Swaziland; these results varied considerably by community and economic class, and were only somewhat comparable to previous national-scale assessments. In a larger context, international efforts toward improving household-scale environmental health conditions (e.g., via related UN Millennium Development Goals) are laudable, yet these results corroborate other research suggesting that progress can be difficult to measure, and is decidedly uneven by household location and socioeconomic status.

Key words: Swaziland, environmental health, water, sanitation, solid waste, fuelwood, Millennium Development Goals (MDGs).

INTRODUCTION

Situating global environmental health

The field of environmental health is as broad as our many needs for well-being and safe surroundings. According to the World Health Organization (WHO), environmental health "…addresses all the physical, chemical and biological factors external to a person, and all the related factors impacting behaviours. It encompasses the assessment and control of those environmental factors that can potentially affect health" (www.who.int/topics/environmental_health). The most recent WHO report (2013a) launches with a foreword citing the eight UN Millennium Development Goals (MDGs) for 2015, in which environment and health are

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central to at least four (www.un.org/millenniumgoals).

As with most global health statistics, however, environmental health is a key axis of differentiation between socioeconomic classes. For instance, WHO/UNICEF Joint Monitoring Programme for Water Supply and Sanitation (JMP) statistics for 2011 indicate that 67% of the population from World Bank low income group countries have access to improved drinking water sources, as compared to 99% in high income group countries, and only 37% from low income countries have access to improved sanitation as compared to 100% from high income countries (for further details, see WHO Global Health Observatory Data Repository, apps.who.int/gho/data; all statistics from 2011). One would expect that inequities in environmental health are present at the sub-country scale as well, given evidence suggesting marked disparities in other health indicators among the most and least wealthy members of a wide range of countries (WHO, 2013b, 143ff). Eradication of these disparities has been a major goal of the UN MDGs, yet the empirical question remains as to whether progress in global environmental health is moving quickly among poor people of the world; at least some published reviews of progress toward MDGs suggest otherwise (Poku and Whitman, 2011; Usua et al., 2012; Fehling et al., 2013).

Environmental health in Africa

Many people living in Africa face particular challenges relative to the rest of the world. The most recent World Health Organization statistical summary (2013b) launches by comparing regions of the world in progress toward MDG goals relative to 1990, and Africa's improvement in under-five mortality rates, as one indicator of health conditions, is higher than many other parts of the world, with an annual rate of decline close to 2% (p. 13). Yet absolute rates in maternal mortality, tuberculosis, HIV/AIDS and access to improved drinking water and sanitation are still considerably worse in Africa than in other parts of the world (pp. 14-17). These statistics suggest good progress over the last several decades, offset by continued struggles in securing adequate standards of general and environmental health (Gabay, 2011; Groenewald, 2011; Mukonka et al., 2014), especially in an equitable manner across socioeconomic class (Kangalawe et al., 2008; Yanda and William, 2010).

In the last five years, a great deal of attention has been paid to environmental health conditions in Africa. The proceedings of one major conference including 52 African country delegations and held in Libreville, Gabon in August 2008 launches by stating that "Africa continues to face the 'traditional' challenges of poor access to safe drinking water, hygiene and sanitation. Yet the continent must now also deal with new and emerging challenges, including the effects of climate change on health, accelerated urbanization and indoor and outdoor air pollution" (WHO, 2009a, 7). A frequently cited statistic in the proceedings is the estimate that nearly one-quarter of all deaths in Africa in 2002 were attributable to "environmental risk factors" (WHO, 2009a, 23; cf. Ogunseitan, 2007). The conference resulted in the Libreville Declaration on Health and Environment in Africa (WHO, 2009b), suggesting widespread commitment to improving environmental health conditions on the continent. The conference was followed by a smaller implementation meeting (WHO, 2009c). Overall, WHO pursued a wide range of public and environmental health initiatives in Africa in the latter part of the first decade of the 21st century (WHO, 2010).

The status of Swaziland

Swaziland is a small landlocked country in southern Africa, with a 2011 population of approximately 1.2 million inhabitants. According to World Bank statistics (data.worldbank.org), its general economic conditions are somewhat better than those of its peers: for instance, its 2011 gross national per capita income was estimated at US$2,830, over twice the average for developing countries in sub-Saharan Africa (US$1,248). Yet its health struggles are considerable: Swaziland has the highest HIV infection rate of any country in the world. According to the World Health Organization, in 2011 190,000 people or 15.8% of the overall population were HIV positive, including 26% of the population aged 15 to 49. This has contributed in large part to a life expectancy at birth of only 49 years, and at least 75,000 orphans due to parent death from AIDS (WHO Global Health Observatory Data Repository, apps.who.int/gho/data, all statistics from 2011).

The general environmental status of Swaziland has been summarized in a recent comprehensive assessment by the Swaziland Environment Authority (2012), focusing on five themes: land, water, atmosphere, biodiversity and human development. In the context of human health, the report concludes that "Swaziland faces many challenges in the human health sector among which limited capacity in terms of human and financial resources is one, and weak information systems particularly in relation to monitoring and evaluation of different priority health programmes is another" (p. 258). To compare this status with other countries in Africa, WHO health statistics estimate that 28% of Swaziland's population did not have access to improved drinking water sources in 2011, and 43% did not have improved sanitation (WHO, 2013b, 33-4), placing Swaziland in the 46th and 79th percentiles, respectively, among African countries (with higher percentiles being better status).

A more detailed health assessment was done via the 2010 Multiple Indicator Cluster Survey (MICS), administered in conjunction with UNICEF, and involving

Table 1. Selected Swaziland Multiple Indicator Cluster Survey (MICS) results, in percentage (n≈4800 households).

Item	Urban	Rural	Overall
Improved water	91	60	67
Water treatment	24	14	15
Improved sanitation	94	73	78

Table 2. Percent household access to improved water in Swaziland by wealth quintile (5 = richest; 1 = poorest).

Wealth index quintile	Improved water access
5	92
4	77
3	66
2	60
1	41

nearly 5000 Swazi households (Central Statistical Office and UNICEF, 2011). The UNICEF MICS instrument focuses on key MDGs, in particular those related to human health. As suggested in Table 1, the MICS found that 67% of the overall population had access to improved water sources and 78% had access to improved sanitation facilities. Though the MICS improved water access result is in rough agreement with the WHO figure provided above, the marked disparity between the MICS improved sanitation result of 78% and the WHO figure of 57% may suggest the need for greater methodological agreement. Table 1 also suggests that only 15% of the population had access to treated water sources, a much smaller proportion than those with access to “improved” sources, suggesting a key disparity to be discussed further below. A comparison between the overall population with access to improved water and those from rural vs. urban areas (Table 1) and poor vs. rich wealth quintiles (Table 2) demonstrates significant geographic and economic class variation masked in the overall percentage, as suggested in the JMP statistics above.

Further evidence on the state of Swaziland’s rural environmental health can be gained from a series of annual assessments produced by the Swaziland Vulnerability Assessment Committee (2011, 2012). The SVAC reports include statistics on basic household resources such as water and energy necessary to secure livelihoods: for instance, in 2011 31% of rural respondents indicated an environmental hazard (solid or liquid waste, or liquid or sewage discharges) near their water source (SVAC, 2011, 48), and in 2012, 82% of rural households relied on fuelwood for cooking, vs. only 18% using electricity (SVAC, 2012, 13).

Overall, however, a more comprehensive picture of household access to safe resources, and impacts on these resources, has not been developed for Swaziland. By way of terminology, a household in Swaziland comprises an extended family unit dwelling on a homestead, which typically includes a number of adjacent dwelling and storage structures. Historically, Swazis generally dwelled in a more dispersed fashion on homesteads rather than in a nucleated fashion in villages, and some vestiges of this cultural past remain even in more densely settled parts of the country.

In light of the key role played by basic resources such as water and energy for household well-being, potential health effects of these resources on households (e.g., waterborne disease or indoor air pollution via fuelwood burning), and potential impacts of households on these resources (e.g., contamination by solid or human waste), this study focuses on household-scale environmental health in the context of four key resource sectors: drinking water, energy, solid waste and human waste. These resources are commonly considered in household-scale assessments in Africa (Joséphine et al., 2008; Oyelola and Babatunde, 2008; Ewodo et al., 2009; Awe et al., 2011; Mughogho and Kosamu, 2012), though they are less commonly brought together to present a more comprehensive view.

MATERIALS AND METHODS

Study site: Ezulwini Valley

The Ezulwini Valley (literally, Valley of Heaven) is classified in SVAC reports as part of a peri-urban corridor stretching between Mbabane and Manzini, the two largest urban areas in Swaziland (Figure 1). The Ezulwini Valley has great significance to Swazis, with the Parliament, National Archives, Somhlolo National Stadium, and major traditional sites located here. Additionally, Ezulwini has been a site of considerable commercial development, with numerous high-end resorts and a major shopping center. Based on this traditional and commercial significance, the Ezulwini Valley would appear to be superior to many other locations in Swaziland in terms of basic household resource needs. As a study site, then, Ezulwini is not “representative” of Swaziland (an impossibility at any rate, given geographic and economic class variability in basic resources as noted above); rather, it suggests how households are faring in a relatively developed part of Swaziland outside of Mbabane and Manzini.

Our connection to the Ezulwini Valley involved students from Lewis & Clark College in Portland, Oregon volunteering May through July 2013 in local community neighborhood care points, or NCPs, as part of an overseas program. Neighborhood care points were created in Swaziland in response to the HIV-AIDS crisis, as a means of providing basic food, health, and educational services to vulnerable populations, in particular orphans and vulnerable children (U.S. Fund for UNICEF, 2011). The original NCPs were created following a severe drought in 2003; there are now over 1500 distributed across Swaziland, providing a variety of services and exhibiting a wide range of structural improvements (NERCHA, 2011).

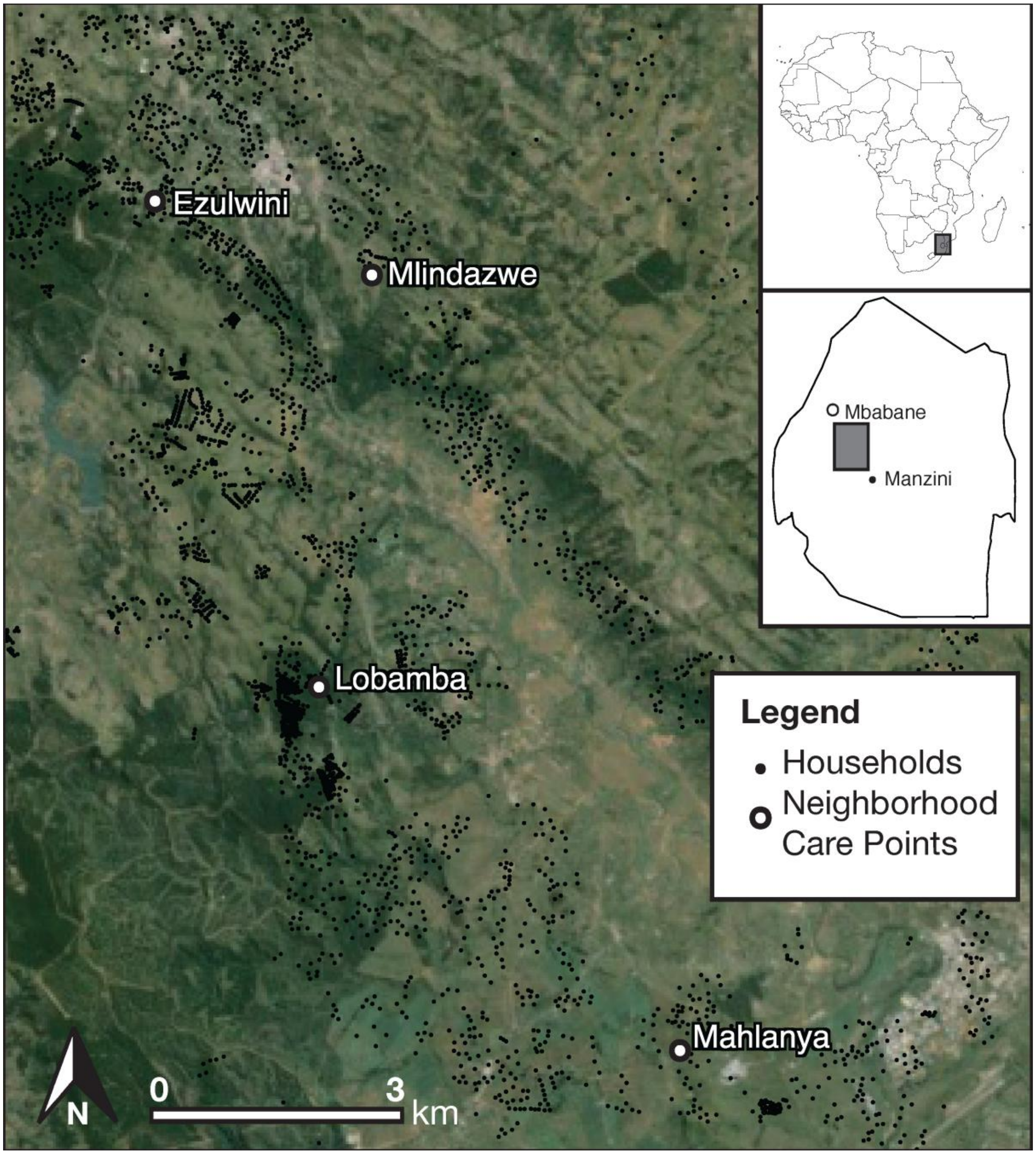

Figure 1. Location map of Ezulwini Valley study site in Swaziland, with four neighborhood care points and surrounding communities included in environmental health assessment.

Students worked in partnership with All Out Africa (www.alloutafrica.com), an organization based in the Ezulwini Valley that places volunteers in a number of local NCPs, which they also support via building assistance, educational materials, and other services. Students volunteered in four NCPs and conducted our environmental health assessment among households in the surrounding communities (Figure 1). Of these four NCPs, Ezulwini and Lobamba NCP are found in relatively densely settled peri-urban areas, whereas Mlindazwe and Mahlanya NCP are found in less densely settled areas with more rural characteristics. All four NCPs we selected are relatively close together, lying within a 12 km span of the Ezulwini Valley.

Research question

Our research question was “What is the condition and perception of water, energy and solid/human waste among households in communities surrounding four NCPs in the Ezulwini Valley?” In addition, we planned to compare results by community and economic level, and with country-scale Swaziland data. As is evident in these questions we were interested not only in the current state of these resources and resource impacts, but in how they were perceived and prioritized among the Swazis who lived in these communities, as any attempt to improve environmental health conditions must be grounded in knowledge of both. We also

Table 3. Environmental health assessment sections.

Section	Content
Pre- Survey Information	Survey location, reference number Economic class surrogates Respondent age/gender
Household General	Settlement duration Number of minor/adults Most important issues
Drinking Water	Sources Treatment Quantitative/qualitative adequacy Notes/recommendations
Household Energy	General energy sources Cooking energy sources Fuel wood cooking method/location Cooking energy adequacy Notes/recommendations
Solid Waste	Disposal locations Community adequacy Notes/recommendations
Human Waste	Sanitation facility locations Community adequacy Notes/recommendations

anticipated that differences would arise across communities and economic class, and in spite of the relatively unrepresentative nature of the Ezulwini Valley as a study location we wanted to compare our results with related data collected for Swaziland.

Survey design

The survey instrument was constructed in consultation with related assessments. Input and translation assistance to SiSwati was provided via key Swazi contacts (including Swazi students attending Lewis & Clark College). The survey was pretested in the field to ensure straightforward and consistent administration. We used actual photos of households in the Ezulwini Valley to train teams in calibrating the low/medium/high economic class categories, thus aiding consistency across community survey teams.

The instrument included six major sections, with primary survey items noted for each in Table 3. The overall flow of the survey was intended to begin with background information collected by survey administrators without prompting respondents, e.g., economic class surrogates and respondent approximate age and gender. It further solicited general household information, followed by each of the four priority resource sectors, starting with drinking water as a straightforward, recognized sector and ending with solid and human waste as more sensitive sectors. A general discussion of respondents' most important community issues was included prior to resource-specific sections to gauge the larger significance of these resources. For all sectors, respondent perception/prioritization was elicited via perceived adequacy items using a three-point scale (inadequate/somewhat adequate/adequate), where adequacy involved the household scale for drinking water (which included both qualitative and quantitative adequacy, that is, sufficient purity and volume) and energy, and the community scale for solid and human waste given the potential for cross-household impacts. The overall survey took between 10 and 30 min to administer per household.

Survey administration and analysis

Following student NCP volunteer work in the mornings, the environmental health assessment was conducted after the NCPs closed at midday, in teams of 1-3 students together with 1-4 Swazis (including Swazi Lewis & Clark students and NCP community members). Our goal was to survey approximately 50 households in each of the four communities, a decision based primarily on afternoon time available in each community, though we did oversample in Lobamba given its larger population, ending up with an overall sample of 210 households (Table 4). Due to technical and practical challenges in surveying a random sample of households, we adopted a snowball sampling strategy, starting with the households of contacts (e.g., cooks or participating youth) in the NCPs, then moving on to neighbors or other contacts. Given our interest in the effects of differing levels of wealth on household-scale environmental health, we informally stratified our community

Table 4. Neighborhood care point (NCP) communities and households.

Household	Ezulwini	Lobamba	Mahlanya	Mlindazwe	Total
Households within 0.5 km radius of NCP	201	401	66	72	740
Households surveyed	50	72	51	37	210

samples by economic class surrogates, primarily structural improvements and amenities (e.g., wall/roof materials, structure size, presence of satellite dish antenna). These sampling and quota decisions mean that our survey is not necessarily representative of these four communities in the Ezulwini Valley, but it does suggest some important characteristics of the households we surveyed, as well as similarities and differences by community and economic class.

The survey was administered using iPad Mini mobile devices running a survey app called Fulcrum (www.fulcrumapp.com). The iPads and Fulcrum app offered a number of advantages, including (a) concurrent administration via survey downloading to multiple devices, followed by uploading of data at the end of each day; (b) automatic geolocation of households, (c) minimization of entry error via response lists, branching logic and required questions; and (d) accumulation of data onto the Fulcrum site for ready monitoring and downloading. We were also able to provide daily updates and reminders directly on the app. The iPads provided community benefits in addition to survey administration: for instance, our students used them with NCP children during their volunteer time, and took (and printed for distribution) photos of NCP and community members upon request.

A small team of students reviewed data at the end of each day to check for entry error, geolocation error, and other potential problems as well as sample coverage. During the first few days of administering the final survey, these data review meetings proved invaluable in monitoring progress and making team-specific recommendations as well. Additional teams did follow-up reconnaissance of community water and fuelwood sources to determine quality. Water sources were tested for total and fecal coliform and inspected visually for probability of surface runoff contamination; fuelwood sources were assayed for extent, major species and reproduction rate.

Data from the household surveys were downloaded from the Fulcrum website in spreadsheet format, then recoded as necessary for descriptive statistical analysis; text responses were summarized to provide better interpretation of numerical results. Data were also uploaded in shapefile format to QGIS, an open-source GIS and mapping program, to discover spatial patterns. Analysis results were shared with Ezulwini Valley community members and Swaziland government officials in a variety of public events.

RESULTS

Drinking water

Selected drinking water results are shown in Figure 2 and Table 5. As noted above, the concept of “improved” drinking water, which has been central to MDG and Swaziland rural development goals, is arguably vague, as most assessments consider as “improved” any piped water irrespective of source protection or treatment. We thus adopted a more restrictive definition of “improved” in our survey. In brief, we distinguished in our survey between piped water (drinking water obtained via a piped distribution system, whether a community or private tap) and improved water (water considered safe to drink as a result of source protection or treatment); whereas virtually all improved water is piped, not all piped water is improved (safe to drink).

Table 5 thus lists the percentage of households in each community and overall with access to piped water, improved water and springs and streams (which are potentially unsafe due to surface runoff contamination), as well as those households who treated their drinking water (e.g., by boiling or adding bleach). Since some piped water was deemed improved (safe to drink), and some unimproved spring/stream sources were piped, the top three rows add up to more than 100%.

Overall, fully 70% of the 210 households we surveyed had access to piped water sources, which is understandable given the relatively developed nature of the Ezulwini Valley. Yet only 37% had access to what we determined as improved (safe to drink) sources, many (32%) regularly fetched drinking water from springs or streams, and only 9% did any form of household treatment. These results alone are highly important: nearly two out of three surveyed households in one of the most developed parts of the country regularly access unsafe drinking water sources, and less than one in ten regularly purifies their drinking water. Additionally, access time to water sources was significant even for this relatively developed area, with a mean roundtrip time of 50 min. Figure 2 and Table 9 suggest important differences between the four surveyed communities in their access to safe drinking water. The two more urban communities, Ezulwini and Lobamba, had relatively high proportions of households supplied by piped water, in contrast to the more rural community of Mahlanya, which relied heavily on springs and streams. Mlindazwe, the other community of a more rural nature, also had a high proportion of piped water, but in all cases these distribution systems relied on unimproved and untreated water sources. Access to improved water sources ranged from nil in Mlindazwe to 79% in Lobamba, the site of a former royal village and an important traditional and administrative center.

Perhaps surprisingly, access to piped water predictably was found to increase beyond the low economic class sector, but access to improved (safe to drink) water

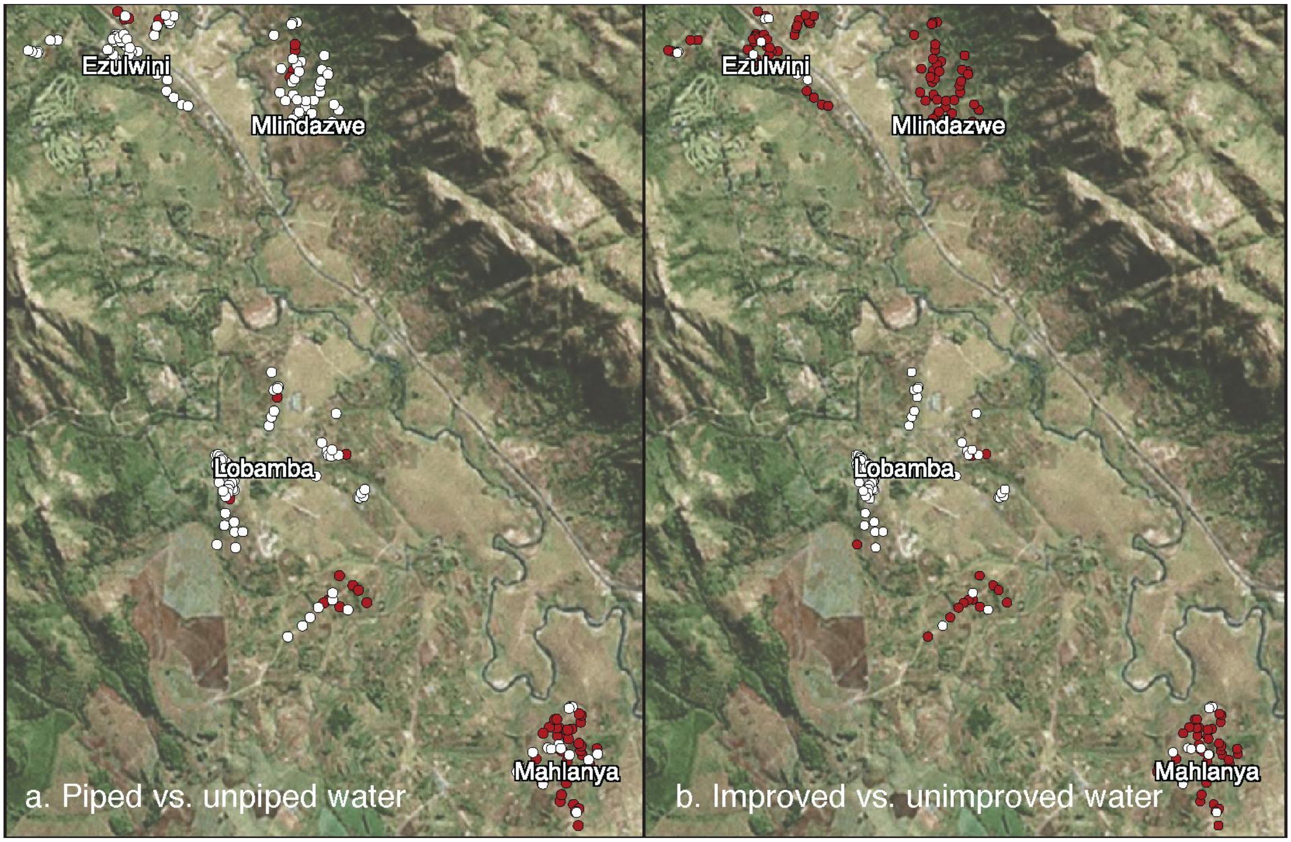

Figure 2. Distribution of households using (a) piped (white) versus unpiped domestic water and (b) improved (white) versus unimproved domestic water sources.

Table 5. Water source type and water treatment practice (percent households surveyed) by community.

Item	Ezulwini	Lobamba	Mahlanya	Mlindazwe	Overall
Piped water	86	76	31	89	70
Improved water	20	79	20	0	37
Springs/streams	18	21	73	16	32
Any household treatment	14	3	10	13	9

actually decreased as economic class increased. How could this be? To be sure, a small number of wealthier households utilized their own private deep wells (boreholes), but most relied on piped systems shared with their communities, which as noted above were not consistently improved. One possible explanation for this anomalous result is that economic class may be a surrogate here for particular communities, given the vastly disparate community-scale results: in Lobamba, where improved water sources are commonly used, only one household in eight was rated upper economic class, whereas in Mlindazwe, for instance, where no surveyed households used improved water, almost seven out of eight were rated medium or high economic class.

Energy

Figure 3 and Table 6 present results of the household energy portion of our environmental health assessment. As could be expected in this relatively developed part of Swaziland, fully 71% of surveyed households utilized electricity as a general energy source (usually for lighting and outlets). In contrast, only 32% of households used electricity for cooking, a function of its perceived high cost relative to other options. The most common source of cooking energy was harvested wood, which roughly one-half of all households used; some respondents also purchased fuelwood from nearby vendors, who generally transport wood from other parts of the country to the

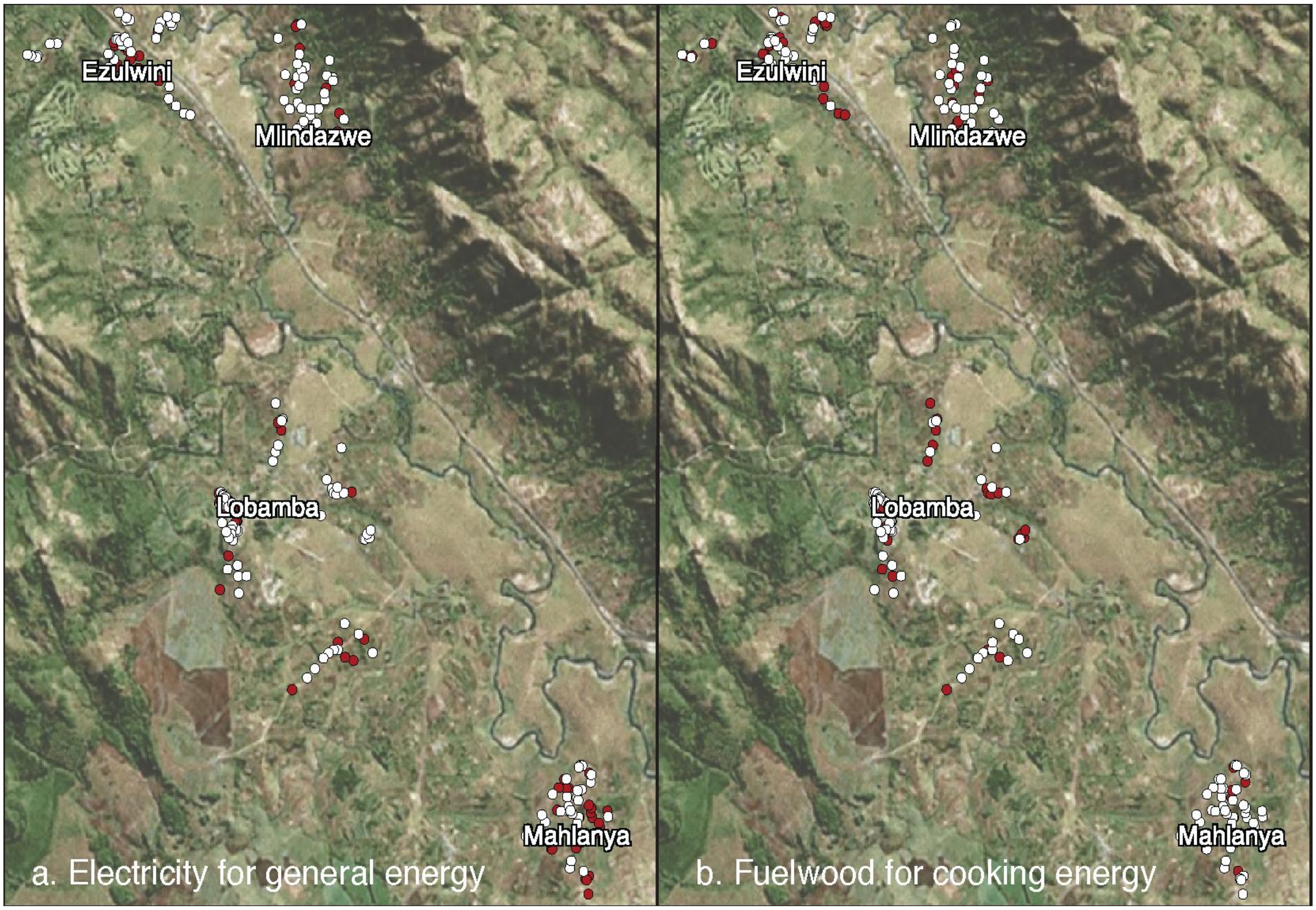

Figure 3. Distribution of households using (a) electricity (white) for general energy needs and (b) fuelwood (white) for cooking energy needs.

Table 6. Energy source type (percent households surveyed) by community.

Energy Source	Ezulwini	Lobamba	Mahlanya	Mlindazwe	Overall
Electricity (general)	76	72	59	81	71
Electricity (cooking)	50	28	14	43	32
Compressed gas	34	17	6	0	15
Fuelwood (harvested)	42	46	53	60	49
Fuelwood (purchased)	24	19	43	22	27

Ezulwini Valley given high demand relative to supply. Though fuelwood is traditionally used for cooking in Swaziland, this high percentage of harvested wood in the Ezulwini Valley is surprising given the relatively high settlement density as indicated in Figure 1. In all, 71% of households used harvested or purchased wood for cooking, exactly equal to the proportion that used electricity for general (non-cooking) energy needs.

A comparison of energy use by community suggests quite large differences in use of electricity for cooking (ranging from 14% in rural Mahlanya to 50% in peri-urban Ezulwini), presumably due in part to readily availability of fuelwood nearby for harvest or purchase. Trends in energy use by economic class (Table 9) are marked: high economic class households used electricity far more frequently for cooking than medium and low economic class households (e.g., roughly seven in ten among high vs. one in ten among low economic class households), and far less harvested wood. Evidently, decisions as to cooking energy are based on a tradeoff of cost verse labor,

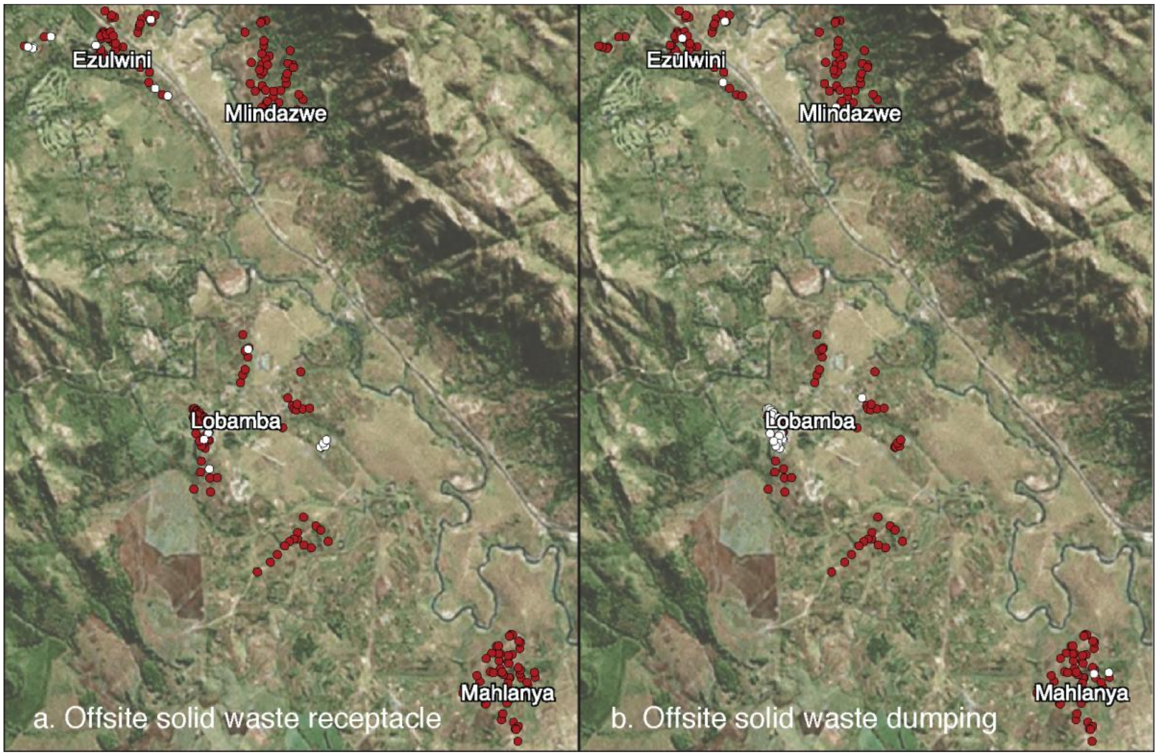

Figure 4. Distribution of households using (a) offsite solid waste receptacle (white) and (b) dumping solid waste offsite (white) in non-designated location.

Table 7. Solid waste disposal method (percent households surveyed) by community.

Disposal method	Ezulwini	Lobamba	Mahlanya	Mlindazwe	Overall
Burn onsite	62	51	94	95	72
Bury onsite	60	13	0	5	20
Offsite receptacle	18	11	0	0	8
Offsite dumping	8	26	4	3	12

as fetching fuelwood can take a great deal of time (2.8 h average roundtrip among reporting households) but communal fuelwood reserves are free.

This heavy reliance on fuelwood for cooking via open-air stoves, which are common in Swaziland, results in the well documented environmental health problem of indoor air pollution, for which sub-Saharan Africa is particularly afflicted (Legros et al., 2009). These results suggest that indoor air pollution is widespread even in relatively developed parts of Swaziland such as the Ezulwini Valley, and especially so among low economic class households, and among women who do almost all the cooking in a typical Swazi household. Similar to the relatively low proportion of households that purify their drinking water, however, the ubiquity of this indoor air pollution problem may render it difficult to successfully address.

Solid waste

The relation between households and their surrounding communities differs in the context of waste: though household drinking water and energy supply options depend on community-scale infrastructure in the Ezulwini Valley, solid and human waste decisions made by households generally have a cumulative impact on their surrounding communities. The household results for waste, then, should be considered at the scale of potential community as well as household impacts.

Figure 4 and Table 7 summarize selected solid waste survey results: overall, by far the most common method of dealing with solid waste was for households to burn or bury their waste onsite (in the immediate vicinity of their dwelling structures), but one in eight dumped their waste offsite (in a non-designated location vs. a landfill). The

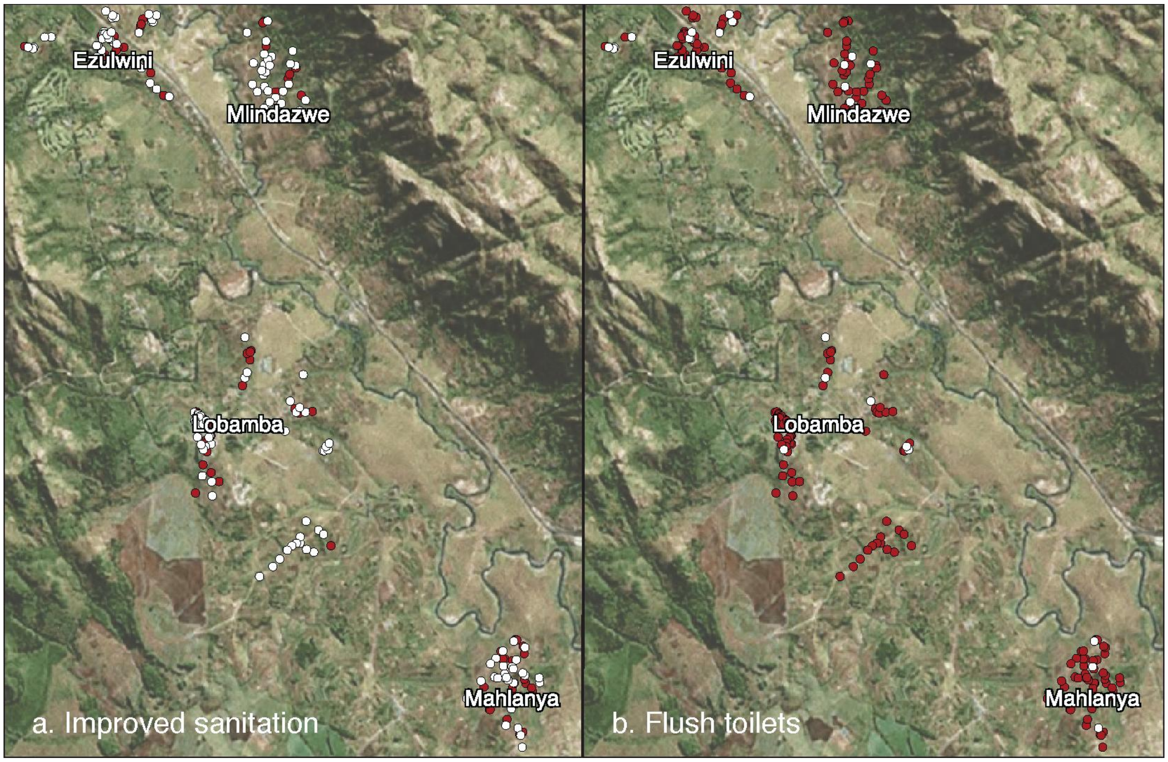

Figure 5. Distribution of households using (a) improved sanitation (white), and (b) flush toilets (white).

Table 8. Human waste disposal method (percent households surveyed) by community.

Disposal method	Ezulwini	Lobamba	Mahlanya	Mlindazwe	Overall
Overall improved sanitation	70	67	61	78	68
Flush toilet	22	8	6	14	12
Flush toilet	54	54	61	76	60
Improved latrine	28	19	35	19	25

Ezulwini Town Council maintains a set of solid waste collection receptacles in Ezulwini, with some receptacles in Lobamba as well; for the most part, however, these do not seem to be frequently used, and they are not available in the more rural communities of Mahlanya and Mlindazwe.

A comparison by community reveals, for instance, the stark conditions of peri-urban Lobamba, where crowded homesteads and a lack of collection receptacles have resulted in over one in four households dumping their waste offsite in non-designated locations. The accumulation of solid waste was clearly evident in these parts of Lobamba as well, increasing potential for disease transmission, especially among children who frequently walked or played near affected areas. Economic class seems to play a role as well (Table 9), with higher economic class households using offsite receptacles far more and dumping offsite far less.

Human waste

Globally, progress in access to sanitation has generally lagged behind provision of water supply, for a variety of political and other reasons (Rosemarin et al., 2008). This larger pattern may not be evident in the Ezulwini Valley: as shown in the human waste survey results in Figure 5 and Table 8. Overall, over two in three surveyed households (68%) used improved sanitation facilities for disposal of human waste, in contrast to only 37% having access to improved (safe to drink) water. These sanitation facilities are generally an improved pit latrine on the homestead (60 percent), though a small proportion (12 percent) of homesteads had flush toilets. Additionally, a full one-quarter of households used latrines that respondents or surveyors deemed unimproved (e.g., without a concrete slab or outdoors).

Comparison by community and economic level reveals

Table 9. Selected resource results (percent households surveyed) by economic level.

Item	Low economy	Medium economy	High economy	Overall
Piped water	52	78	78	70
Improved water	42	38	22	37
Electricity (general)	39	81	97	71
Wood (harvested)	61	49	28	49
Offsite receptacle	3	7	19	8
Offsite dumping	18	12	6	12
Overall improved sanitation	48	74	83	68
Flush toilet	2	7	44	12

some patterns in human waste disposal. Community results were generally consistent, with some community-specific differences: for instance, Ezulwini had a higher proportion of flush toilets (22%), in part connected to a community sewer scheme; and more Mahlanya households (35%) used unimproved latrines, perhaps in part due to the more rural nature of this community. Differences by economic level (Table 9) are more marked,with higher economic class households reporting much higher use of improved sanitation facilities (83% high vs. 48% low economic class). This difference is understandably even greater in the use of flush toilets (44% high vs. 2% low economic class). The generally strong performance of Ezulwini Valley homesteads in their use of improved sanitation, therefore, masks important differences by economic class.

Perceived adequacy

The results above reflect reported reality in the context of four key resources, all with environmental health implications, essential to household well-being in the Ezulwini Valley. To successfully address issues related to these four resources, empirical data on household perceptions of resource adequacy can help us understand the priorities of surveyed communities.

Figure 6 and Table 10 summarize these results, on a Likert-type scale, we devised ranging from 1 (low reported adequacy) to 3 (high reported adequacy). Overall, respondents rated water quality the most adequate (2.6 average), and sources of cooking energy the least (1.9 average). In comparison to the results above, respondents seemed more concerned about adequate drinking water quantity (2.3) than quality (2.6), even though our results indicate relatively few households have access to safe drinking sources. This may suggest challenges in gaining community support for improving drinking water quality if quantitative shortages are not also prioritized. The overall results also suggest that initiatives to address cooking energy challenges may receive strong community support.

Community comparisons summarized in Table 10 suggest considerable variance in perceptions of solid waste disposal adequacy, with the more peri-urban areas of Ezulwini and Lobamba perhaps understandably reporting much poorer adequacy than the more rural areas of Mahlanya and Mlindazwe. A similar difference between peri-urban and more rural communities was found with respect to human waste, and some variance was found in perceived cooking energy adequacy, though likely as a function of fuelwood availability. Water quantity and quality displayed less variance between communities.

When comparing adequacy by economic class (Table 11), the general pattern is one of the lower perceived adequacy among lower economic groups; for instance, perceived cooking energy adequacy ranged from an average of 1.5 among low economic class households to 2.4 among high economic class households, a possible function of the differing energy utilization patterns summarized above. This pattern is particular evident in perceived solid and human waste disposal adequacy, suggesting that lower economic class households may be disproportionately impacted. Less difference is evident in perceived quantitative and qualitative water adequacy.

DISCUSSION

Summary of results

A synthesis of the survey results above suggests clear overall patterns, as well as important differences across the four surveyed communities and three economic classes. Given that the Ezulwini Valley is one of the most developed areas in Swaziland, it is unsurprising that over two out of three surveyed households utilize piped drinking water, yet far more surprising that only slightly more than one out of three utilize water from protected or treated sources- a result that proved highly community specific. Access to safe drinking water would be a major environmental health priority, then, in this location; yet the perceived adequacy results summarized above remind us that drinking water quality alone is not the most important priority in these communities.

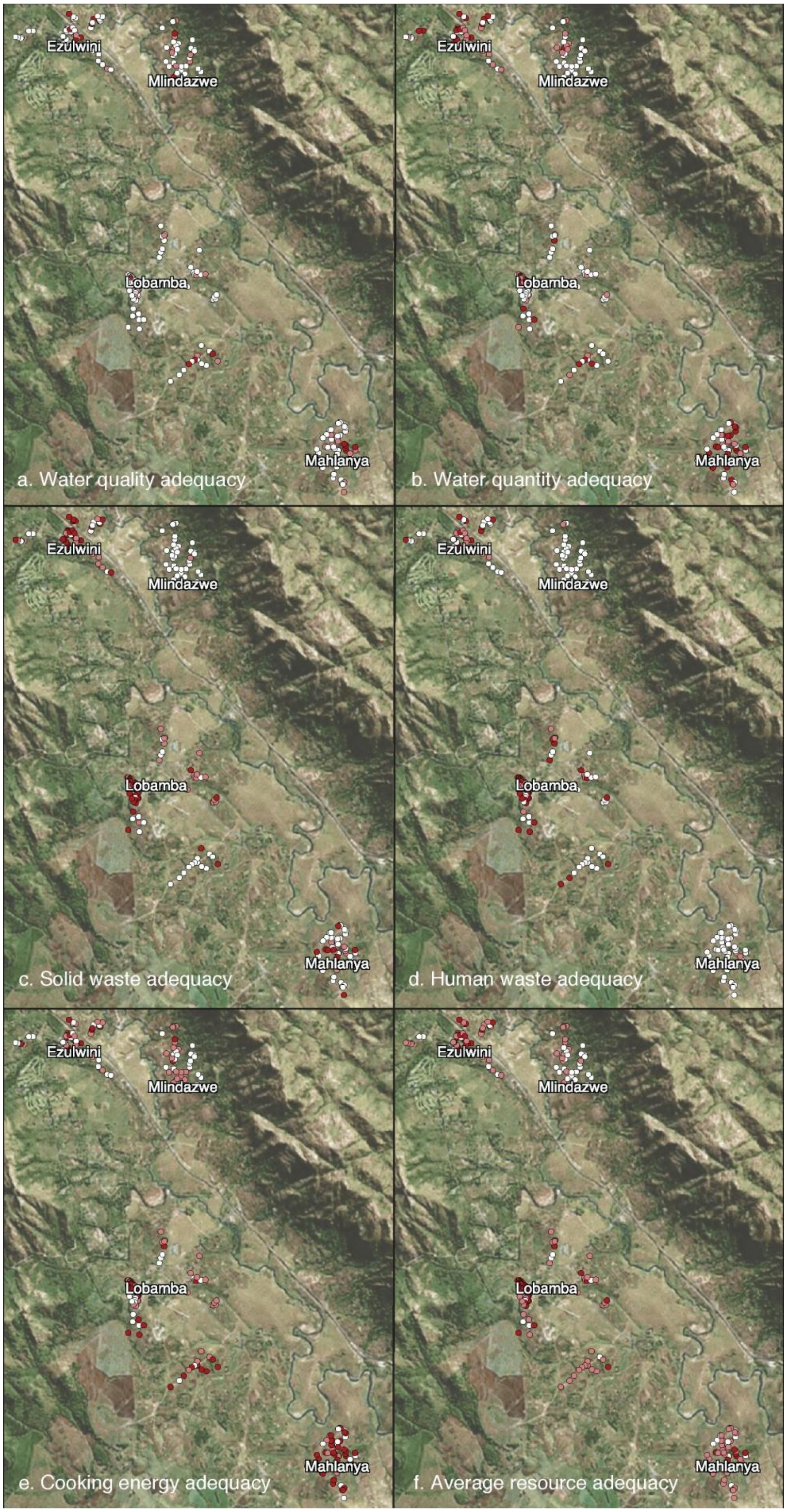

Figure 6. Perceived resource adequacy (white = adequate; red = inadequate) for (a) drinking water quality, (b) drinking water quantity, (c) solid waste, (d) human waste, (e) cooking energy and (f) overall average.

Table 10. Resource adequacy comparison by community (1 = inadequate; 3 = adequate).

Resource sector	Ezulwini	Lobamba	Mahlanya	Mlindazwe	Overall
Water (quantity)	2	2.5	2.1	2.6	2.3
Water (quality)	2.6	2.8	2.5	2.6	2.6
Cooking energy	2.1	1.8	1.5	2.4	1.9
Solid waste	1.7	1.9	2.6	3	2.2
Human waste	2.2	2	3	3	2.4

Table 11. Resource adequacy comparison by economic level (1 = inadequate; 3 = adequate).

Resource sector	Low economy	Medium economy	High economy	Overall
Water (quantity)	2.0	2.4	2.3	2.3
Water (quality)	2.5	2.7	2.8	2.6
Cooking energy	1.5	2.0	2.4	1.9
Solid waste	2.0	2.2	2.6	2.2
Human waste	2.3	2.5	2.7	2.4

A similar disparity is evident in overall household energy results, where over two out of three surveyed households have access to electricity, yet less than one in three use electricity for cooking- in this case, a result that varied significantly according to household economic class. From an indoor air pollution standpoint, and given the increasingly dense settlement (thus fuelwood demand) in the Ezulwini Valley, results from this important resource sector are unacceptable as well. In contrast to drinking water quality, however, there appears to be strong community prioritization of the need to address cooking energy challenges.

Solid and human waste disposal in surveyed communities of the Ezulwini Valley differs in part as a function of community type, where relatively dense peri-urban areas understandably struggle more with waste than do the more rural communities. Overall, relatively little attention has been devoted to solid waste disposal, and the vast majority of households fend for themselves, though perceived adequacy results suggest strong potential support for improved solid waste disposal in more peri-urban communities. Much more attention has gone to human waste, an even greater environmental health concern: here, over two in three surveyed households utilize improved sanitation facilities, though results differ significantly by household economic class.

Comparison of Ezulwini Valley and Swaziland

In comparing results from the Ezulwini Valley with the rest of Swaziland, recall the country-scale WHO and UNICEF-sponsored results above, where 67-72% of the country was reported to have access to improved (piped) water, and 57-78% was reported to have access to improved sanitation facilities. Relative to these estimates, the Ezulwini Valley is not significantly ahead of nor behind the national average- a relatively surprising result, given the Ezulwini Valley is one of the most developed areas in Swaziland. Households in the Ezulwini Valley do, however, utilize relatively more electricity and less fuelwood for cooking than in the country as a whole. Yet geographic and economic class differences in the Ezulwini Valley appear to be reproduced in the national-scale picture as well; the overall statistics at both the scale of this area and Swaziland thus apparently mask important differences by location and wealth.

Larger implications

The prominent role of environmental health in our most important development agendas, such as the UN Millennium Development Goals, is heartening. Yet, the above results from one of the most developed regions in Swaziland are sobering, as the status of drinking water, cooking energy and solid and human waste disposal reveals continued challenges for many households in the area. Indeed, simply assessing progress toward these important MDGs is methodologically challenging: in the key case of drinking water, our study reveals that relatively few "improved" sources may be safe for drinking. More accurately ascertaining progress toward ensuring safe drinking water for all is thus a far more demanding task, and may reveal even poorer overall performance, than efforts to date have suggested. Globally, this disparity between "safe" and "improved" drinking water sources has been recognized by the WHO/UNICEF Joint Monitoring Programme for Water Supply and Sanitation, which is planning a post-2015 (post-MDG) initiative focusing on evidence-based drinking water, sanitation, and hygiene assessment (WHO 2013b, 19). Additional research tends to corroborate the inadequacy of the MDG definition to ensure provision of truly safe drinking water (Lenton et al., 2008, Dar and Khan, 2011; Clasen, 2012; Sambu and Tarhule, 2013).

Our survey also suggests that overall results mean relatively little in the context of specific communities and socioeconomic classes. We selected four communities located quite close to each other, yet overall household-scale patterns were in many ways unique to each (with the exception of differences between peri-urban and more rural communities). Some enjoy good treated water schemes; some have better fuelwood resources; some worry less about waste. Effectively improving environmental health at the scale of household resource access and impacts, therefore, can only be done by gathering information on, and paying close attention to, community-specific needs and desires.

The additional layer of economic class is a critical one, perhaps no more striking than in the Ezulwini Valley, where among the most wealthy and many poorer households in Swaziland dwell in relatively close proximity. As suggested at the outset, however, this reality is not limited to the Ezulwini Valley nor Swaziland: in the context of urbanizing areas, for instance, "…the health impacts of the most serious problems are largely confined to lower-income groups….It is common for the residential areas of middle- and upper-income groups…to receive good quality water supplies, sewers, drains, electricity supplies and regular services to remove solid waste while 30% or more of the city population on the poorer residential areas receive little or nothing" (Hardoy et al., 2001).

Household-scale environmental health thus continues to be a challenge in our world, certainly more so among the poor, and with specific needs and priorities among particular communities; though the MDGs have contributed toward improvement, there is a long way to go, and the post-MDG scenario following 2015 is anything but clear (Poku and Whitman, 2011; Vandemoortele, 2011; Usua et al., 2012; Van Norren, 2012; Nayyar, 2013; Kesavan and Swaminathan, 2014). This study of the Ezulwini Valley, the Valley of Heaven, should remind us that there are households located even in relatively developed parts of the world that still face these challenges on a daily basis. Continued attention to fine-scaled data gathering and sharing, as was our survey's intent, may help these areas better identify their needs and desires as they move closer toward creating environments that are safe and healthy for their inhabitants.

Conflict of Interests

The author(s) have not declared any conflict of interests.

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Full Length Research Paper

Benthic macroinvertebrate community of a fourth order stream in Kashmir Himalaya, India

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The study was conducted in order to determine the abundance of benthic macroinvertebrates in Doodhganga stream passing through Brenwar forest (Yousmarg) Kashmir. Altogether 6 orders under 2 phyla namely: Arthropoda and Annelida were recorded from the stream. Inspite of the torrential flow of the stream, the hard substrata like boulders and cobbles provided a stable habitat for diverse number of macroinvertebrates. The diversity of benthic community helped to assess the health of the stream. On the basis of Hilsenhoff biotic index the study area was found pristine with negligible organic pollution. The seasonal dynamics showed greater diversity and density in summers than in winters.

Key words: Benthic, diversity, pollution, stream.

INTRODUCTION

Benthic macroinvertebrates are by far the most common groups used in the assessment of water quality (Williams and Feltmate, 1992; Rosenberg and Resh, 1993; Lin and Yo, 2008; Martins et al., 2008). These animals of size 200-500 µm (Rosenberg and Resh, 1993) inhabit the bottom substrate of fresh water, estuarine and marine ecosystem (A.P.H.A, 1998). The distribution of benthic macro invertebrate species and communities is controlled by a variety of environmental factors such as habitat characteristics (Peeters and Gardeniers, 1998), water quality (Hellawell, 1986), sediment quality (Chapman, 2001), sediment grain size (Tolkamp, 1980), contaminants (Phipps et al., 1995) and by biological factors such as competition and predation (Macneil et al., 1999).

The benthic community shows a range of taxa-specific responses to environmental stressors, these may be with respect to alteration in the food webs (Goedkoop and Johnson, 1996; Lodge et al., 1998; Stockley et al., 1998) or due to floods or drought (Covich, 1993; Power, 1995; Johnson et al., 1998), that alter the species composition of the benthic macrofauna.

This paper provides useful information pertaining to the type of invertebrates inhabiting the bottom substrate, thereby reflecting the pollution level in the stream.

MATERIALS AND METHODS

Study area

Doodganga stream is the principal left bank tributary of the River

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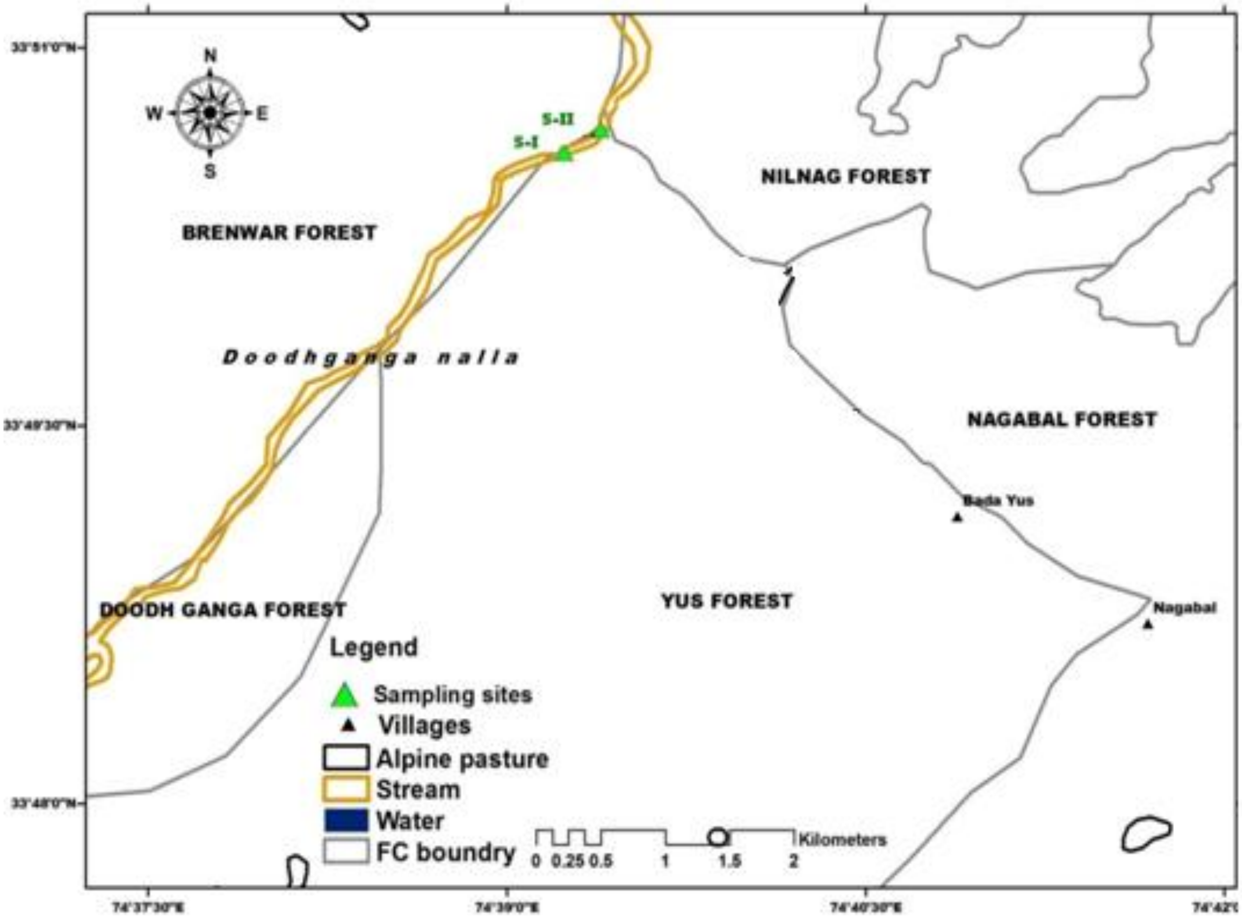

Figure 1. Doodhganga stream through Brenwar forest, Yousmarg and identification of two sampling sites.

Jehlum. The sampling sites (Figure 1) were located in the stream passing through the Brenwar forest of Yousmarg at an altitude of 2,304 m a.m.s.l. The coordinates for the two sites (I & II) were 33° 50′ 34.4″N and 74° 39′ 12.4″E and 33°50'41.2" N and 74°39'24.7" E, respectively. The study area belonged to stream order four (Hussain and Pandit, 2011) according to Rosgen classification. Sampling was carried in two different sites and the results obtained were averaged to provide a holistic information about the stream.

Methodology

Quantitative sampling was done during May, June, November and December (2010) via following the protocol for hard-bottomed streams (Hoffsten and Malmqvist, 2000; Stark et al., 2001; Ilmonen and Paasivirta, 2005). Fifteen replicates were taken from each site per month.

Samples collected were preserved in 4% formalin (for specimen with exoskeleton) and soft bodied organisms were preserved in 70% ethanol (Borror et al., 1976). The identification was done upto genus level with the help of standard works of McCafferty and Provonsha (1998), Wetzel and Likens (2000), and Ward (1992).

The population density was determined by calculating the number of individuals recorded per meter square. Biotic indices like Shannon diversity index (Shannon and Weaver, 1976), Simpson index (Simpson, 1949), evenness index (Pielou, 1966) and Hilsenhoff Biotic Index (Hilsenhoff, 1977, 1982, 1987) were also determined.

RESULTS AND DISCUSSION

The general characteristics of the stream are given in Table 1. During the period of investigation, 22 species of macrozoobenthos were recorded (Table 2). Arthropoda was found to be the most dominant group, comprising of 21 species followed by Annelida with 1 species. The former was represented by class Insecta (5 order) and Crustacea (1 order).

Highest relative density (94.5%) was recorded for phylum Arthropoda. The hard bottomed stream provided a stable habitat (Williams and Feltmate, 1992; Emere and Nasiru, 2007; Arimoro et al., 2007) to the macroinvertebrates against the increased water flow rate. The other phyla Annelida was represented by only *Erpobdella octoculata*. Here their contribution to the total invertebrate

Table 1. General characteristics of the study area.

Parameter	Average values
Depth (m)	1.23
Dissolved oxygen (mg/l)	6.52
Water flow rate (m^3/s)	2.06
Substrate type	Hard-bottomed stream (leaf litter from adjoining forest)

Table 2. Community structure of macrozoobenthos species in Doodhganga stream

Phylum	Order	Family	Taxa/Species
Arthropoda	Diptera	Athericidae	*Atherix* sp.
		Blephariceridae	*Bibiocephala* sp.
		Chironomidae	*Diamessinae* sp. *Glyptotendipes* sp.
		Empididae	*Clinocera* sp.
		Tabanidae	*Tabanus* sp. *Chrysops* sp.
		Tipulidae	*Hexatoma* sp. *Tipula* sp.
		Simuliidae	*Simulium* sp.
	Trichoptera	Glossosomatidae	*Glossosoma* sp.
		Hydropsychidae	*Hydropsyche* sp
		Limnephilidae	*Limnephilus* sp.
		Rhyacophilidae	*Rhyacophila* sp.
		Brachycentridae	*Brachycentrus* sp.
	Plecoptera	Capniidae	*Allocapnia* sp
		Chloroperlidae	*Xanthoperla sp.*
	Ephemeroptera	Baetidae	*Alainites* sp.
		Heptageniidae	*Epeorus* sp.
	Coleoptera	Chrysomelidae	Unidentified
	Amphipoda	Gammaridae	*Gammarus pulex*
Annelida	Pharyngobdellida	Erpobdellidae	*Erpobdella octoculata*

community was little, as the annelids have been observed to thrive better in soft depositing substrates which were not the present case.

The total number of individuals varied 632 ind.m^2 (May) to 185 ind.m^2 (December). During the individual sampling months (Figure 2) more or less constant trend of trichoptera and diptera dominating over other orders was reported. The macroinvertebrates belonging to these two orders are known to have a wide range of feeding behavior ranging from shredders, collectors to predators (Hutchinson, 1993) thereby occupy every possible trophic level (Williams and Feltmate, 1994; Mackie, 2001).

The benthic fauna showed a seasonal variation in density and diversity as is seen in the Figure 2. The

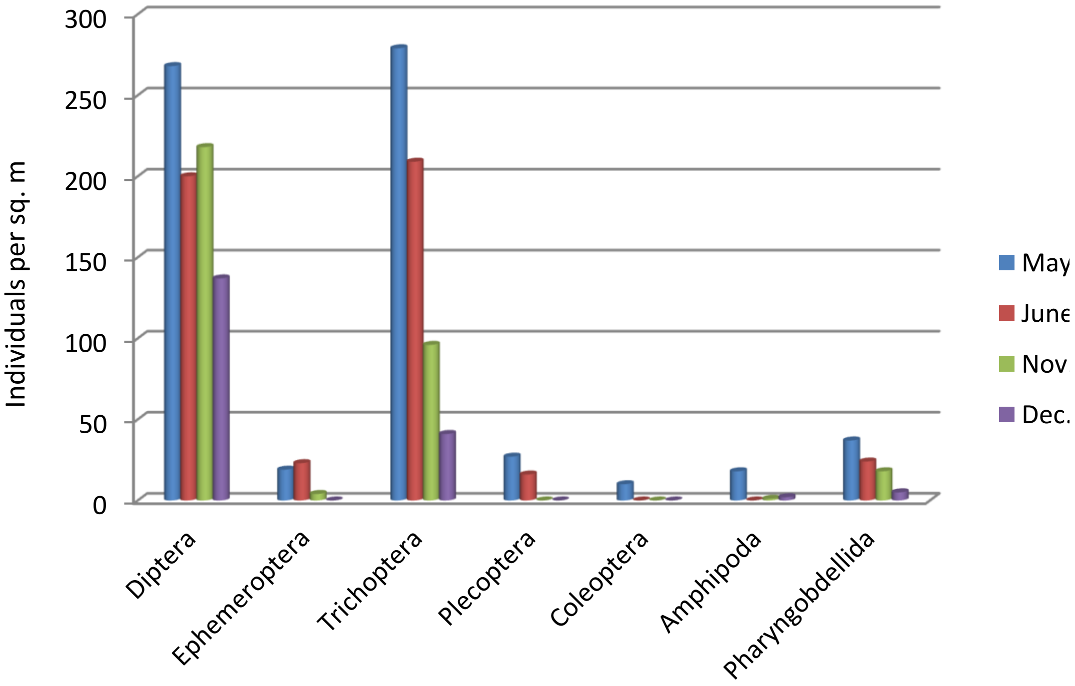

Figure 2. Population densities (number of individuals per sq. m.).

abundance was found to be highest during summer as compared to winters (Bruce et al., 2003; Lamp and Haube, 2004). The freezing temperature in winter in this high altitude temperate stream limits the density and species richness however warmer conditions in summers favors establishment of diverse fauna (Gupta and Michael, 1983; Allan, 1995; Cowell et al., 1997; Stark and Armitage, , 2004). The mean values of Shannon-, Simpson- and equitability indices were found to be 2.18, 0.16 and 0.85 respectively. The Hilsenhoff Biotic Index (3.6) indicated no apparent organic pollution in Doodhganga stream has apparently no organic pollution.

Conclusions

The study shows that the substrate type has an effect on the distribution of benthic organisms. A seasonal variation in the abundance of individuals indicates that temperature and its factors have a pronounced influence on the life cycle of invertebrates. The diversity indices and biotic index shows high abundance, richness and diversity with no organic pollution in the study area hence indicating good stream health.

Conflict of Interests

The author(s) have not declared any conflict of interests.

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Full Length Research Paper

Batch adsorption of heavy metals (Cu, Pb, Fe, Cr and Cd) from aqueous solutions using coconut husk

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This study was carried out to evaluate the efficiency of metals (Cu, Fe, Pb, Cr and Cd) removal from mixed metal ions solution using coconut husk as adsorbent. The effects of varying contact time, initial metal ion concentration, adsorbent dose and pH on adsorption process of these metals were studied using synthetically prepared wastewater. The percentage removal of metals increased with increasing weight (0.4-1.2 g) in 50 ml of adsorbent dose and the observed trend was: Cr>Cu>Pb>Fe>Cd. The adsorption efficiency increased with increasing initial metal ion concentration (0.3-0.9 mg/l) and the observed trend was: Cr>Cu>Cd>Fe>Pb. Similarly, percentage removal of metal ions increased with increasing pH of the mixed metal ions solution (pH values of 2, 6 and 10). The observed trend of percentage adsorption of metals by varying pH was: Cd>Fe>Cr>Cu>Pb. The effect of contact time on the adsorption efficiency at different time intervals of 20, 40 and 60 min in mixed metal ions solution showed that the removal of tested metals was rapidly achieved during a short interval of 20 min. Generally, the study showed that coconut husk (a waste material) is a viable material for removal of metals from waste water as the percentage adsorbed varies from 95.2-98.8, 91.1-99.3 and 75.0-98.5% for Cd, Cr and Cu, respectively while the percentage removal of Fe and Pb from the waste water varies from 84.9-97.0 and 81.1-98.7%, respectively. Isothermal studies showed that the experimental data are best fitted on Langmuir model.

Key words: Batch adsorption, heavy metals, wastewater, coconut husk.

INTRODUCTION

Increased use of metals and chemicals in process industries has resulted in generation of large quantities of effluent that contain high level of toxic heavy metals and their presence pose environmental-disposal problems due to their non-degradable and persistence nature. Unlike organic pollutants, the majority of which are susceptible to biological degradation, heavy metal ions do not degrade into harmless end products (Gupta et al., 2001). The presence of heavy metal ions is a major concern due to their toxicity to many life forms. Heavy metal contamination exists in aqueous wastes stream of many industries, such as metal plating, mining operations, tanneries, chloralkali, radiator manufacturing, smelting, alloy industries and storage batteries industries, etc. (Goyal and Ahluwalia, 2007; Olayinka et al., 2007, 2009; Kadirvelu et al., 2001). Treatment processes for

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heavy metal removal from wastewater include: precipitation, membrane filtration, ion exchange, adsorption and coprecipitation/adsorption. The removal of toxic heavy metal contaminants from aqueous waste streams is currently one of the most important environmental issues being investigated (Igwe et al., 2005; Mondal, 2009; Suthipong and Siranee, 2009). Studies on the treatment of effluent bearing heavy metals have revealed adsorption to be a highly effective technique for the removal of heavy metal from waste stream and activated carbon has been widely used as an adsorbent (Chand et al., 1994). Despite its extensive use in the water and wastewater treatment industries, activated carbon remains an expensive material (Abdel-Ghani and El-Chaghaby, 2009).

Adsorption on low cost-adsorbent for removal of toxic metals from wastewater has been investigated extensively. These materials include thioglycolic acid modified oil-palm (Akaninwor et al., 2007), wild cocoyam biomass (Horsfall and Spiff, 2004), brewery biomass (Kim et al., 2005), sodium hydroxide modified Lalang (*Imperata cylindrica*) and leaf powder (Hanafiah et al., 2006). Recently, efforts have been made to use cheap and available agricultural wastes such as coconut shell, orange peel, rice husk, peanut husk and sawdust as adsorbents (Vaishnav et al., 2012) to remove heavy metals from wastewater (Abia and Igwe, 2005).

The use of the coconut shell as a biosorbent material presents strong potential due to its high content of lignin of about 35-45%, and cellulose of about 23-43% (Carrijo et al., 2002). As a result of its low cost, powder of coconut shell- *Cocos nucifera* is an attractive and inexpensive option for the biosorption removal of dissolved metals. Various metal-binding mechanisms are thought to be involved in the biosorption process including ion exchange, surface adsorption, chemisorption, complexation and adsorption-complexation (Pino, 2005; Matheickal et al., 1999).

Coconut shell is a material composed of several constituents, among them lignin acid and cellulose bear various polar functional groups including carboxylic and phenolic acid groups which can be involved in metal binding (Matheickal et al., 1999; Ting et al., 1991). The cellulose and lignin are biopolymers admittedly to be associated to the removal of heavy metals (Gaballah and Kilbertus, 1994; Gaballah et al., 1997; Hunt, 1986). Adsorption on low cost-adsorbent for removal of toxic metals from wastewater has been investigated extensively. These materials include thioglycolic acid modified oil-palm (Akaninwor et al., 2007), wild cocoyam biomass (Horsfall and Spiff, 2004), brewery biomass (Kim et al., 2005), sodium hydroxide modified Lalang (*Imperatacylindrica*) and leaf powder (Hanafiah et al., 2006).

However, these studies did not involve batch adsorption process but rather single adsorption of metal ions from their aqueous solutions, although study carried out by Abdel-Ghani and El-Chaghaby (2009) involved batch adsorption process.

In this study, the use of unmodified coconut husk in the removal of metal ions from aqueous solutions through batch adsorption studies was investigated. Coconut husk which is generally considered as a waste is abundant in Nigeria and has a high sorption capacity due to its high tannin content. This study involved the examination of four variables such as pH of the solution, metal ion concentration, contact time and adsorbent loading on the removal of Cu(II), Fe(II), Cd(III), Cr(III) and Pb(II) ions from aqueous solutions, simultaneously.

MATERIALS AND METHODS

Adsorbent

Coconut *(C. nucifera)* shell/husk were collected from Abusoro at Okitipupa Local Government Area of Ondo State, Nigeria; sun dried for about 2-5 days before being ground into fine particles using the manual grinding machine and sieved with sifter to obtain 120 mm (micrometer mesh) finer dust particles. The finer dust particles were treated with 0.1 M HCl and was later re-introduced into an oven at a temperature of 30°C for 30 min and then preserved in a sample container for future use.

The concentrations of Cu, Cd, Fe, Cr and Pb ions in the adsorbent were determined by placing 5 g of the adsorbent in 50 ml de-ionized water for 50 min in a 50 cm long and 2 cm diameter glass column. Aliquot portions of the eluate from the pre-treatment of the organic waste were carefully decanted into 50 ml plastic bottles and analyzed for the heavy metals using Atomic Absorption Spectrophotometer (AAS) GBC Scientific with oxy-acetylene flame at temperature of about 2500°C.

Adsorption technique

A glass column was fitted with cotton wool and held firmly in a vertical position with the aid of a clamp fixed at one end to a retort stand as illustrated in Figure 1. Atmospheric pressure helped to push the sample through the organic material.

Adsorption experiment was done by measuring 50 ml of the wastewater sample and poured into a 100 ml conical flask. 5 g of the pre-treated fine particle coconut husk was added to the wastewater.

Adsorbates

The solutions of Cu, Cd, Fe, Cr and Pb metal ions were prepared from analytical grade $CuSO_4.5H_2O$, $3Cd(SO_4).8H_2O$, $FeSO_4.7H_2O$, $Cr(NO_3)_3.9H_2O$ and $Pb(NO_3)_2$ respectively from BDH Chemicals Ltd, Pool England. 30 mg/L aqueous solutions (stock solutions) of these salts were prepared with de-ionized water in 250 ml volumetric flask and these stock solutions were diluted with de-ionized water to obtain the working standard solutions.

In each set of experiment, the effect of one factor was evaluated by varying this factor while keeping all other factors constants.

Adsorption experiments

Batch adsorption process was carried out at laboratory room temperature. The different factors affecting adsorption process of

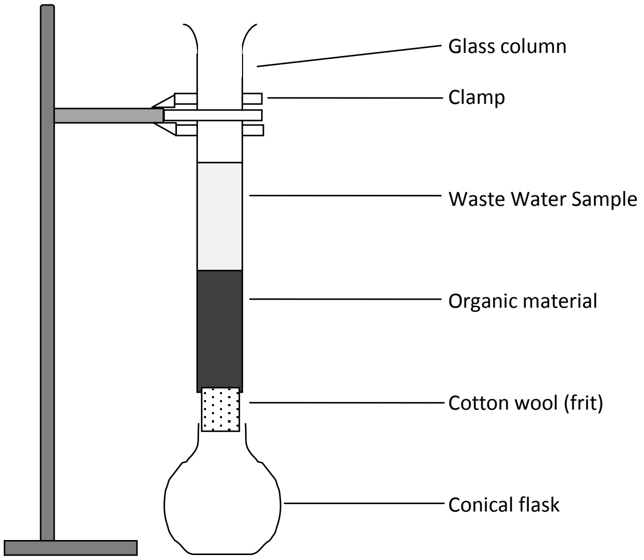

Figure 1. Sketch diagram of experimental set-up.

the metal ions under study (Cu^{2+}, Pb^{2+}, Fe^{2+}, Cr^{3+} and Cd^{2+}) such as contact time, concentration, adsorbent dose and pH have been studied using synthetically prepared wastewater. The sorption capacity qe mg/g and removal efficiency Q were obtained according to the Equations 1 and 2, respectively:

$$qe = \frac{(Co - Ce)V}{W} \quad (1)$$

$$Q = \frac{(Co - Ce) \times 100\%}{Co} \quad (2)$$

Where V is the volume of the solution, W is the amount of adsorbent, C_o and C_e are the initial and concentration of the solution after adsorption in mg/l.

Statistical analysis

The relationship between pairs of metal adsorption at the various variables was tested using Pearson Moment Correlation Coefficient. All statistical analyses were tested using SPSS 21.00 with significance based on 95% confidence level (Ogbeibu, 2005).

RESULTS AND DISCUSSION

Effect of adsorbent dosing

The availability and accessibility of adsorption site is controlled by adsorbent dosage (Rafeah et al., 2009). The effect of mass of adsorbent loading on heavy metal removal using coconut husk was investigated by varying adsorbent loading weight from 0.4 to 1.2 g per 50 ml of mixed metal ions solutions (Appendix 1). The effect of coconut husk weight is graphically presented in Figure 2. It can easily be inferred that the percentage removal of metal ions increased with increasing weight of coconut husk. This is due to the greater availability of the exchangeable sites or surface area at higher dose of the adsorbent. This result is in agreement with previous studies on many other adsorbents (Bin et al., 2001; Ajmal et al., 1998; Abdel-Ghani and El-Chaghaby, 2009). The observed trend of percentage removal of metal ions was: Cr>Cu>Pb>Fe>Cd. Furthermore, the P-values of 0.058 and 0.090 at 95% confidence level ($P>0.05$) shows statistically that there was no significant correlation in adsorption pattern between Cu and Cd and that of Cu and Fe. However, there exist significant correlation in adsorption pattern between Cu and Cr and that of Cu and Pb, since $P<0.05$ at 95% confidence level. Similarly, there exist a significant correlation between the pairs of Cd and Fe ($P<0.05$) and that of Cd and Pb ($P<0.05$) at 95% confidence level. The same trend was observed statistically between the pairs of Fe and Cr ($P<0.05$) and Fe and Pb ($P<0.05$). However, there was no significant correlation statistically between the adsorption pattern of Cr and Pb at 95% confidence level ($P>0.05$).

Effect of concentration on adsorption of the metal ions

The effect of initial metal concentration on the adsorption efficiency of coconut husk is shown in Figure 3. Adsorption experiments were carried out at different initial metal ion concentrations of 0.03, 0.06 and 0.09 mg/l in mixed metal ions solution (Appendix 2). The adsorption efficiency increased with increasing initial metal ion concentration. This result is in accordance with the work of Okieimen and Onyenkpa (2000). It is generally expected that as the concentration of the adsorbate increases the metal ions removed should increase. It is believed that increase in concentration of the adsorbate bring about increase in competition of adsorbate molecule for few available binding sites on the surface of the adsorbent hence increase in the amount of metal ions removed. The observed trend of percentage removal of metal ions was: Cr>Cu>Cd>Fe>Pb. The P-values of 0.041 and 0.018 at 95% confidence level ($P>0.05$) shows statistically that there was significant correlation in adsorption pattern between Cu and Cd and that of Cu and Cr. However, there was no significant correlation in adsorption pattern between Cu and Fe ($P<0.05$). Cd shows statistically that there was significant correlation in the adsorption pattern with the rest metals (Cu, Fe, Cr and Pb) as their $P<0.05$. Fe also shows statistically significant correlation with Pb ($P<0.05$) at 95% confidence level.

Effect of hydrogen ion concentration

The pH adsorption edges of the constant concentration for Cu, Cd, Fe, Cr and Pb for coconut husk are shown in Figure 4. All experiments were carried out in the pH

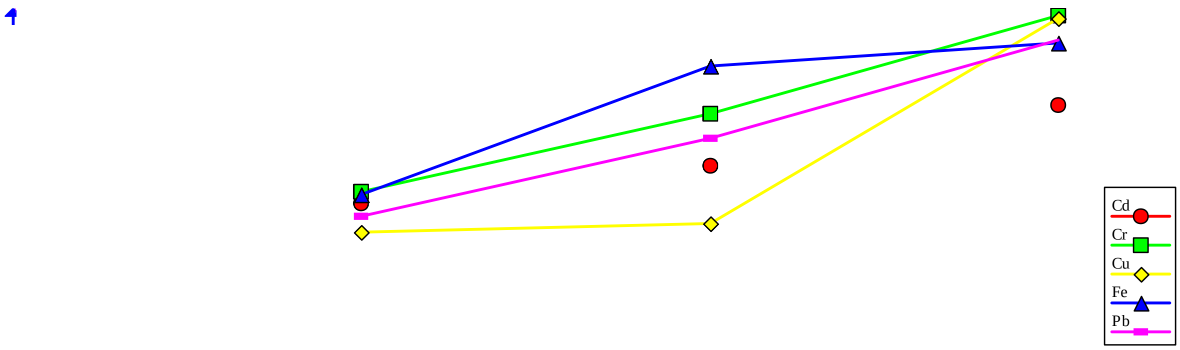

Figure 2. Effect of coconut loading weight on metal ion adsorption in a mixed metal ion solution.

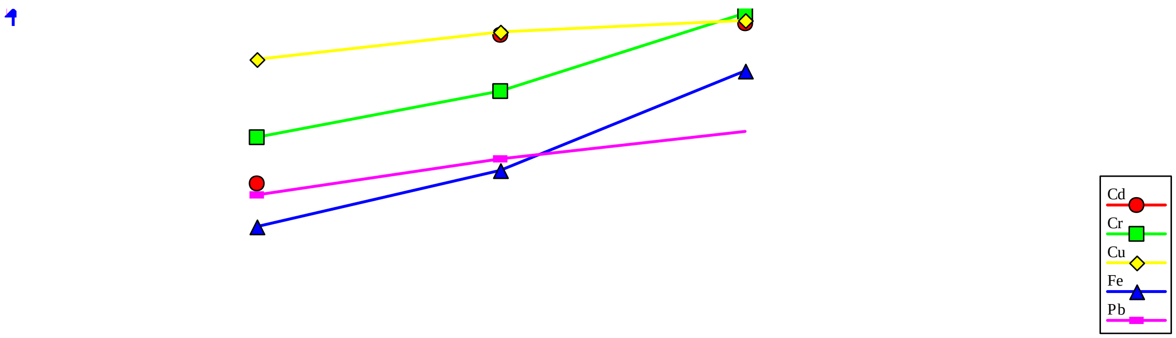

Figure 3. Effect of variation in initial metal ions concentration on adsorption using coconut husk.

values of 2, 6 and 10 (Appendix 3) where chemical precipitation is almost avoided, so that metal removal could be related to the adsorption process (Abdel-Ghani and El-Chaghaby, 2009).

The susceptibility of the system pH changes may be attributed to the nature of the ions in solution and the nature of the adsorbent used. The lower the pH, the more H^+ ions competing with the metal ions for adsorption

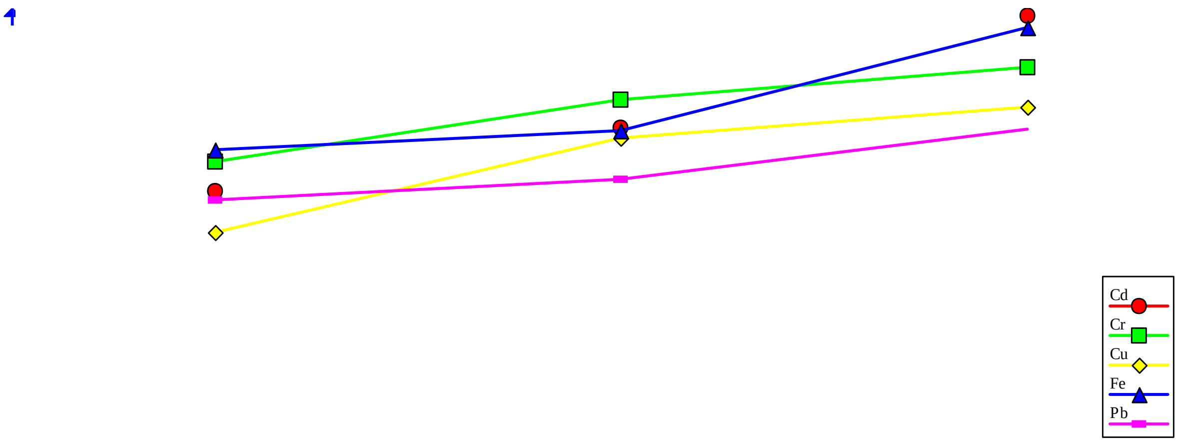

Figure 4. Effect of hydrogen ion concentration (pH) on metal ions adsorption.

sites, thus reducing their adsorption. On the other hand, the higher the pH, the less the H^+ ions competing with metal ions for adsorption sites, thus increasing their adsorption, which explains the obtained results in Figure 4. The observed trend of percentage adsorption of metal ions was: Cd>Fe>Cr>Cu>Pb. Furthermore, the $P<0.05$ at 95% confidence level shows statistically that there was significant correlation in adsorption pattern between Cu and Cd; Cu and Fe; and Cu and Pb, respectively. Similar trend of adsorption pattern statistically was observed between Cd and Cr ($P<0.05$) and Cd and Pb ($P<0.05$).

Effect of contact time

The effect of contact time on the adsorption efficiency is shown in Figure 5. Adsorption experiments were carried out at different time intervals: 20, 40 and 60 min in mixed metal ions (Appendix 4). It was observed that removal of tested metals was rapidly achieved, within a short period of 20 min. Adsorption of Cd and Cr ions attained maximum within 20 min while that of Cu was within 40 min. Adsorption of Fe and Pb increases with increase in contact time. Generally, the observed trend of metal removal was: Pb>Cu>Cr>Fe>Cd. Previous results revealed that removal of all tested metals was rapidly removed within a short period of 30 min (Olayinka et al., 2009). The effect of contact time on adsorption process of metal ions from wastewaters were studied by many authors (Dakiky et al., 2002; Saeed et al., 2005; Abdel-Ghani et al.,2007a; Abdel-Ghani et al., 2007b). The results indicated that the equilibrium time was dependent on the nature of the adsorbent and on metal ions concentration. Furthermore, the $P<0.05$ at 95% confidence level shows statistically that there was significant correlation in adsorption pattern between Cu and the rest metals (Cd, Fe, Cr and Pb, respectively). Similarly, Cd exhibited statistically significant correlation with Fe ($P<0.05$) but there was no significant correlation with Pb ($P>0.05$). However, there exist statistically significant correlation in adsorption pattern between Fe and Cr ($P<0.05$) and Pb ($P<0.05$). The $P>0.05$ shows statistically that there was no significant correlation between the adsorption pattern of Cr and Pb.

Isothermal studies

The analysis of equilibrium data for the adsorption of Cd, Cr, Cu, Fe and Pb on coconut husk was done using the Langmuir and Freundlich isotherm model as shown in Tables 1 and 2, respectively. The extremely high R^2 values provided by the Langmuir isotherm suggest that the data best fitted the Langmuir isotherm given by the equation:

The adsorption capacity of the adsorbent,

$$qe\left(\frac{mg}{g}\right)=\frac{v}{m(Co-Ce)}. \quad (3)$$

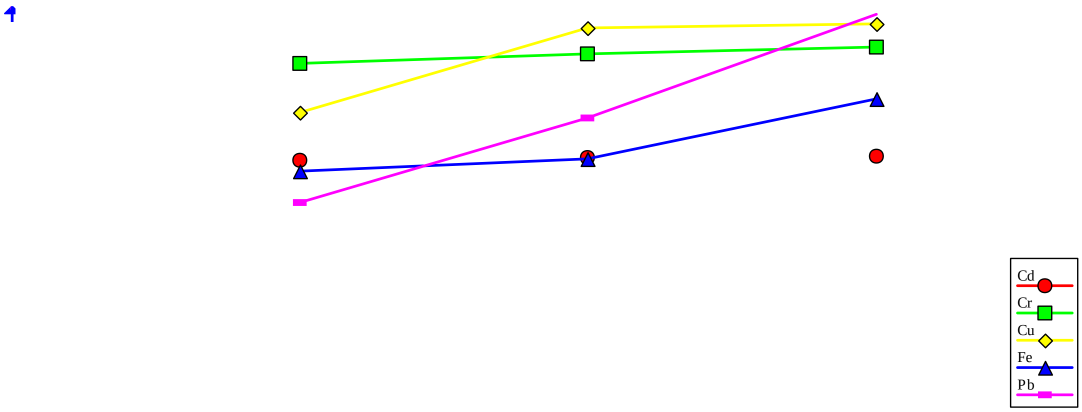

Figure 5. Effect of contact time on metal ions adsorption.

Table 1. Values of Langmuir Isotherm constants for sorption of Cd, Cr, Cu, Fe and Pb metal ions.

Metal ions	q_{max} (mg/g)	K_l (l/mg)	$R_L=1/(1+K_l C_o)$	R^2 values
Cu	0.00010	786.1	0.041	0.965
Cd	0.00010	610.02	0.051	0.992
Fe	0.00009	118.4	0.22	0.919
Cr	0.00031	1355.00	0.024	0.835
Pb	0.00039	38.44	0.46	0.999

Table 2. Values of Freundlich Isotherm constants for sorption of Cd, Cr, Cu, Fe and Pb metal ions.

Metal ions	K_F (mg/l)	$\frac{1}{n}$	N	R^2 values
Cu	4.4E-6	0.810	1.23	0.148
Cd	1.24E-6	1.047	0.96	0.783
Fe	3.7E-7	1.603	0.62	0.346
Cr	2.0E-5	0.585	1.709	0.729
Pb	7.96	2.253	0.44	0.974

Where C_o (mg/l) and C_e (mg/l) are initial and equilibrium concentration of adsorbate solution respectively.

The Langmuir isotherm equation is written as:

$$qe = \frac{qmaxKlCe}{(1 + klCe)} \qquad 4$$

q_{max} (mg/g) is the maximum adsorption capacity upon complete saturation of the adsorbent surface, K_l(dm^3/g) is a constant related to the adsorption/desorption energy.

The equation above can be rearranged to form the Scatchard regression:

$$\frac{qe}{Ce} = qmaxKl - Kl\, qe \qquad (5)$$

A plot of $\frac{q_e}{C_e}$ versus q_e yields a slope $-K_l$ and intercept $q_{max}\, K_l$

The isotherm constants were determined from the respective plots, and are presented in Table 1. Regression values (R^2) presented in Table 1; indicate that the adsorption data for Cd, Cr, Cu, Fe and Pb metal ion removal fitted well the Langmuir isotherm.

Conclusion

This study was carried out to evaluate the efficiency of metal removal from mixed metal ions solution using coconut husk as adsorbent. Contact time, initial metal ion concentration, adsorbent dose and pH as factors that affect adsorption process of metals were studied using synthetically prepared wastewater. The percentage removal of metals increased with increasing weight of coconut husk and the observed trend of percentage removal of metal ions was: Cr>Cu>Pb>Fe>Cd. The adsorption efficiency increased with increasing initial metal ion concentration and the observed trend of percentage removal of metal ions was: Cr>Cu>Cd>Fe>Pb, while percentage removal of metal ions increased with increasing pH and the observed trend of percentage adsorption of metal ions was: Cd>Fe>Cr>Cu>Pb. The effect of contact time on the adsorption efficiency at different time intervals reveals that the removal of tested metals was rapidly achieved during a short period of 20 min. Generally, the study revealed that coconut husk (a waste material) is a viable material for removal of metals from waste water and therefore could be applied in large scale industrial effluents replete with heavy metals. Isothermal studies showed that the experimental data are best fitted on Langmuir model.

Conflict of Interests

The author(s) have not declared any conflict of interests.

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Appendix 1. Percentage removal (%) of metal ion from synthetic wastewater by varying adsorption weight.

Metals	Cu	Cd	Fe	Cr	Pb
0.4 g	65.80	70.03	71.47	71.83	68.10
0.8 g	67.03	75.97	91.13	83.77	80.03
1.2 g	98.53	85.17	94.70	98.93	95.20

Appendix 2. Percentage removal (%) of metal ion from synthetic wastewater by varying molar concentration.

Metals	Cu	Cd	Fe	Cr	Pb
0.03 M	92.27	73.27	66.63	80.37	71.40
0.06 M	96.47	96.07	75.23	87.43	77.03
0.09 M	98.33	97.83	90.47	99.30	81.10

Appendix 3. Percentage removal (%) of metal ion from synthetic wastewater by varying pH.

Metals	Cu	Cd	Fe	Cr	Pb
pH 2	66.40	72.53	78.70	77.03	71.17
pH 6	80.47	82.03	84.93	86.37	74.43
pH 10	85.13	98.83	97.03	91.10	81.90

Appendix 4. Percentage removal (%) of metal ion from synthetic wastewater by varying contact time.

Metals	Cu	Cd	Fe	Cr	Pb
20 Min	82.77	74.90	73.30	90.73	68.17
40 Min	96.57	75.50	75.30	92.23	81.90
60 Min	97.10	75.60	84.93	93.27	98.67

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Full Length Research Paper

Determinants of domestic water consumption in a growing urban centre in Osun State, Nigeria

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Household access to potable water is required for sanitation and general well-being. This challenge is under the influence of variables that play both temporal and spatial roles. This research examines the determinants of domestic water use in Iwo, Osun State, Nigeria. Ten households each were randomly selected from ten of the fifteen political wards in the town for the administration of 100 copies of questionnaire. Female respondents in the investigation were 84.30%. Factor analysis extracted nine out of the thirty one water use components in the analysis. These variables explain 76.0% of the total variance in domestic water use. Multiple regression analysis shows r^2 value of 80.60%. The all-inclusive standardized model generated by stepwise regression analysis showed that five variables are strong predictors of domestic water use in the study area. Water planners need to consider these variables in water supply planning. It is suggested that further investigations be conducted on the quality of water from these sources due to its closeness to the respondents to ensure its fitness for human consumption.

Key words: Domestic water demand, water demand modelling, water accessibility, growing town.

INTRODUCTION

A timely and spatially accessibility to potable water is salient to human well being. It is considered an essential resource for the possibility of life, regardless of amount or proportion (USEPA, 2000) Even so, urban access to potable water in Nigeria is about 42% in 2008 (WHO and UNICEF, 2010). Thus, several published works have been geared towards the development of predictive models which can be applied to estimate the prospective water use at a given period of time and space, for instance, Xinming et al. (1990), Arbues et al. (2003), Okeola and Sule (2010), Al-Amin et al. (2011), Aper (2011), Ayanshola et al. (2010), Ifabiyi and Ahmed, (2011) and Adeoye et al. (2013) among others. The relevance of water use forecasting according to Cook et al. (2001) and Ifabiyi et al. (2012) are as follows: Firstly, it ensures better water management, secondly, it ensures fair sharing and distribution of this resource thereby preventing crisis often associated with water accessibility,

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thirdly, water use forecasting assists in overcoming the challenges of urban and suburban growth; fourthly, it assists system optimization which leads to least cost; fifthly, it allows an understanding of the underlining factors that will affect water demand; and lastly, forecasts provide information about the past and future water use. Boumann et al. (1998) listed four main methodologies in forecasting water demand namely per capita and per unit approaches, end-use models, extrapolation methods and structural or causal models. Galan et al. (2009), however noted that the choice of any of these approaches is based mainly on the intended use and time frame of the prediction and the available data.

Thomas (1998) had noted that domestic water use varies according to the living standards of the consumers in urban and rural areas. In addition to this, Al-Amin et al. (2011) also observed that the quantity of water is variable depending on the cultural habit, settlement pattern, typed of supply, water source among others. However, it is observed that the space is not equally endowed with adequate water resources. Some areas are sufficiently endowed with both surface and subsurface water resources as in the tropical areas making the accessibility easier with all other factors held constant. On the other hand, some areas have to invest a lot in other to have water required to keep life going such as in the arid areas (Arab Water Council, undated; Foster et al., 2011).

The challenge of ensuring timely and spatially accessibility to potable water has become an important issue. The reason is that the quality and quantity required for human health and leisure depends on several factors, especially when we try to determine the quantity of water that may be demanded at any point in time at household level (for instance Onda et al., (2012). According to Ayanshola et al. (2010), accurate estimation of water demand should put into consideration variables such as income, population and sex, while Al-Amin et al. (2011) listed cultural habit, settlement pattern, type of supply and water source as water use determinants in homes. Ifabiyi et al. (2012) found in Sokoto, Nigeria that levels of education, income levels and marital status correlated positively with total household water use while time cost and the distance to water points correlated negatively.

Unfortunately, lack of data has been considered as the principal factor hampering proper and adequate water demand estimation especially in the developing nations (Ayanshola et al., 2010). Metering of water use which could have helped in efficient water use is not in use in Nigeria, thus bases for proper definition of the actual water use, according to Bilthas (2008) is lacked. Zhou et al. (2002) and Ruijs et al. (2008) noted that sufficient data is a required tool for planning water demand management and studies. This work is aimed at evaluating the variables that determine domestic water use in a growing urban centre and their strength in forecasting domestic water use. This will invariably assist in result-oriented water demand planning and designing.

MATERIALS AND METHODS

This study was carried out in Iwo Township (Figure 1). Iwo is one of the 30 Local Government Areas in Osun State, Nigeria. The town has an area of 245 km^2 with a population of 191,348 according to (National Population census, 2006). It is located between the coordinate axis of 7°38'N and 4°11'E. Iwo is subdivided into five quarters which are subdivided into 15 political wards (Table 1). The town consists of Muslims, Christians and traditionalist but the former were observed to form the dominant ones. The central part of the town which consists of the palace and other ancient buildings and compounds now incorporates modern buildings (Enclopedia, Britanica). It has witnessed a tremendous increase in the number of people and spatial coverage as a result of the location of Bowen University, a private University owned by the Nigerian Baptist Convention established in 2002 and also, the location of Reality Radiovision Station owned by the Osun State Government among other developmental projects. The most popular periodical market in the town, Odo-Ori Market attracts people from nooks and crannies of Osun State and other neighbouring States. These establishments have led to the influx of people of diverse professions and areas of life into the town. Some of whom have either settled in their own private buildings while others stay in rented apartments or guest houses located within the town. The major source of potable water in Iwo is Aiba Water Reservoir located within Government Forest Reservation Area in the town. The inadequate supply of water from the Water Works has led to the exploitation of underground and surface sources in the town (Olutona et al., 2012).

Multistage sampling technique was used in arriving at the sample size. Ten wards out of which ten households were randomly selected formed the area of coverage for the purpose of this survey. The quarters and the wards selected are shown in Table 1.

Data was collected through the administration of a pre-tested 100 questionnaire among the randomly selected households (see Appendix 1). This work intentionally focused on age groups from 18 to 65 years because they form active category of the age group and so will be able to give relevant information on domestic water use. The data was subjected to factor analysis for normalisation and to determine the factors that explain water use for domestic purposes in the study area. Multiple regression model was applied to generate predictive model of water use in the town. All statistical analyses were performed using SPSS software, Version 16.0, 2007.

RESULTS AND DISCUSSION

Table 2 shows that about 58.60% are of primary level, 27.30% post primary level, 12.10% are of tertiary level while the remaining 2.00% are other levels, one of which could be Islamic education. Generally, literacy level in Iwo is not different from what is obtainable in Nigeria as a whole, of about 56.9% (UNESCO, 2012). The literacy level is equally high and this has been found to have influence on domestic water use (Ifabiyi, 2011).

Table 3 shows that female gender was 85.9% while the male gender was 14.1%. The investigation focused on heads of households especially women by virtue of their traditional role in water provision for home use. However, the proportion of males in the study was only accommodated where female is not available or indisposed in the course of the investigation. Table 4 shows that respondents that were within the age range of 46 to 65 years were 75.8, 22.2% were from 18 to 45

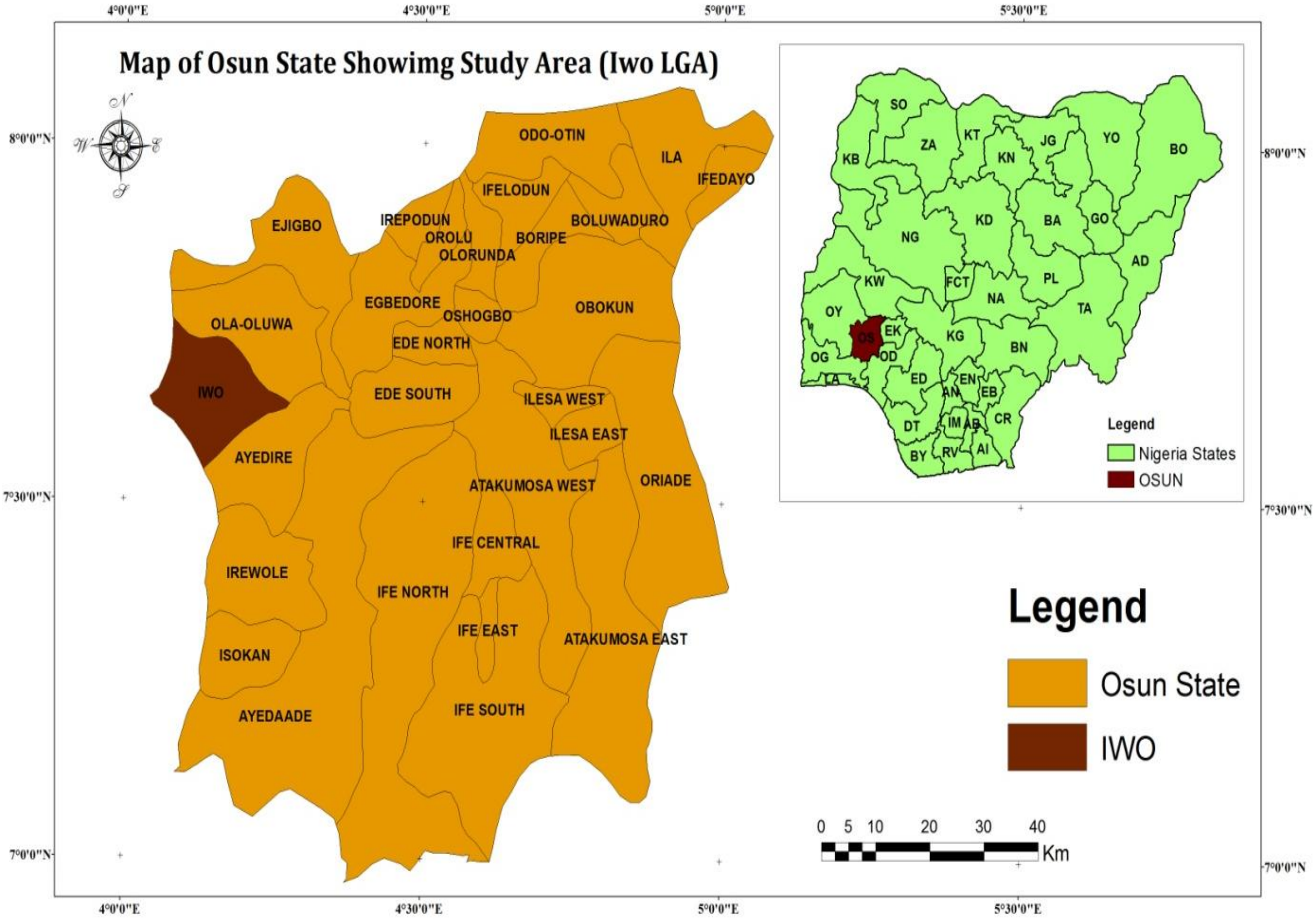

Figure 1. Map of Iwo, Osun State, Nigeria.

Table 1. Iwo LGA quarters and their respective wards with their selection status.

S/N	Name of Quarters	Name of Wards	Status
1.	Gidigbo	Ward One	Selected
		Ward Two	Selected
		Ward Three	Not selected
2.	Isale-Oba	Ward One	Selected
		Ward Two	Selected
		Ward Three	Not selected
		Ward Four	Selected
3.	Molete	Ward One	Not selected
		Ward Two	Selected
		Ward Three	Selected
4.	Oke-Adan	Ward One	Selected
		Ward Two	Not selected
		Ward Three	Selected
5.	Oke-Oba	Ward One	Not selected
		Ward Two	Selected

Source: Author’s field compilation (2013).

Table 2. Respondents' distribution by level of education.

S/N	Level of education	Frequency	Percentage
1.	Primary	59	58.6
2.	Post primary	27	27.3
3.	Tertiary	12	12.1
4.	Others	2	2.0
	Total	100	100%

Source: Author's fieldwork (2013).

Table 3. Respondents' gender distribution.

S/N	Gender	Frequency	%
1.	Male	14	14.1
2.	Female	86	85.9
		100	100.0

Source: Author's fieldwork (2013).

Table 4. Respondents' age distribution.

S/N	Age distribution	Frequency	%
1	<18	0	0.0
1.	18-45	22	22.2
2.	46-65	76	75.8
3.	>65	2	2.0
	Total	100	100.0

Source: Author's fieldwork (2013).

Table 5. Respondents' religious group distribution.

S/N	Religious group	Frequency	%
1.	Christians	51	50.5
2.	Muslims	49	49.5
	Total	100	100.0%

Source: Author's fieldwork (2013).

Table 6. Respondents' distance to water sources.

S/N	Distance to water source (minutes)	Frequency	%
1.	0-10	70	69.7
2.	11-20	28	28.3
3.	21-30	1	1.0
4.	>30	1	1.0
	Total	100	100.0

Source: Author's fieldwork (2013)

years while 2% were 65 years and above. Table 5 shows that Christians form 50.5% of the respondents while Muslims were 49.5%. The proportion of the Christians involved slightly exceeded that of Muslim. Table 6 shows that 69.7% of the respondents were within 10 min trek from water source, 28.3% has maximum of 20 min to water source while the remaining 1.0% have maximum of 30 min to the source. Also, 1.0% have greater than 30 min to water source in the study area. It was observed during the investigation that most homes have their own hand-dug wells or borehole. This reduces the stress of trekking a long distance in search of water. More so, this study was carried out during the rainy season when acquifer yields are appreciable.

Table 7. Respondents' sources of water.

S/N	Source of water	Frequency	%
1.	Hand-dug well	92	92.0
2.	Borehole	8	8.0
3.	Rivers/streams	0	0.0
4.	Rainfall	0	0
5.	Pipe-borne	0	0
	Total	100	100.0

Source: Author's fieldwork (2013).

Table 7 shows the various sources of water for domestic purposes in the study area. The finding shows that 92% of the respondents claimed to rely on hand-dug wells as their source of water while the remaining 8% get their water from boreholes. None of the respondents claimed relying on surface streams, rainfall and pipe-borne water for domestic water uses probably because it is far from them or the unreliable quality of water from such sources.

Determinants of domestic water use

The results of factorability show that Kaisser-Meyer-Olkin (KMO) measure of sampling adequacy is 0.69. Bartlet's test of sphericity is significant at 99.00% level of significance. This implies that the data is factorable. Table 8 shows the variables that influence domestic water use as extracted by factor analysis. Analysis revealed that the first two axes explained 31.22% of data variability. The 9 components extracted from the 31 components analysed (Appendix 2) explain 76.00% of the variation in domestic water use in the study area. The components are household size, water supplied by the suppliers, household preference for water source, age range of the suppliers, water supply for dish washing and

Table 8. Extracted water use components and their respective factor 350 characteristics.

S/N	Water use components[1]	Loading[1]	Eigenvalue[2]	Variance (%)[2]	Cumulative (%)[2]
1.	Household Size	0.964	5.20	16.77	16.77
2.	Quantity Supplied by fetchers	0.795	4.48	14.46	31.22
3.	Preference for a source	0.832	2.46	7.94	39.17
4.	Age Range of suppliers	0.801	2.26	7.30	46.47
5.	Religious Use of water	0.964	2.05	6.62	53.08
6.	Water supplied for Dish washing	0.757	2.00	6.45	59.54
7.	Water supplied for bathing	0.708	1.80	5.89	65.35
8.	Age of Respondents	0.720	1.70	5.49	70.84
9.	Gender Composition	0.642	1.60	5.16	76.00

Source: [1] Extracted from SPSS-generated Table 7; [2]Extracted from SPSS-generated total of variance.

Table **9.** Model summary.

Model	R	R Square	Adjusted R square	Std. error of the estimate
1	0.897	0.805	0.786	72.94145

Source: SPSS-generated table.

bathing, age of the respondents, religion and gender composition.

The relevance of household size in domestic water use cannot be overlooked. This study revealed that this variable has the highest explanation of 16.77% of variance out of the 9 components. It implies that the larger the household is, the likely the higher the domestic water use. This finding is similar to those of Keshavarzi et al. (2006) and Ayanshola et al. (2010) where household size was observed to be one of the determinants of domestic water demand. Also, the quantity of water supplied by the supplier explains 14.46% of the total variance in domestic water use in the study area. This implies that the more the water supplied by the suppliers or fetchers, the likely the higher the household water use and vice versa, assuming all other factors are held constant. This is similar to the observation of Olajuyigbe (2010) in the south western Nigeria.

In the same vein, household preference for a water source also influences domestic water use in the study area with 7.94% of variance. The observation is the central focus of Vásquez (2011). This factor is relevant where family prefers a particular source for a given home use. This may arise in a situation where a given source is preferred for drinking or washing. Hard water may not be preferred for washing because of it does not foam easily.

Another component extracted is the age range of the suppliers that explains 7.30% of the total variance. Households dominated by young adults are more likely to have more supply of water than those homes dominated by aged or children of less than school age. Religious use of water explains 6.62% of the variance in Iwo. This component, also observed by Ruma and Sheikh (2010), becomes important because of the presence of Muslims in Iwo that use water for ablution purposes.

The supply of water for dish washing and bathing purposes with 6.45 and 5.89% of the total variance, respectively, becomes important in Iwo because the closer the water source the more likely is the higher water supplied for these purposes. Similar observation was made by Environment Agency (2008). Water rationing for these domestic activities may not be relevant as largest proportion of the respondents have maximum of 10 min to water sources. The age component has 5.49% of the total variance. This component may be important especially where the respondent is within the age of working class. Such group of people are likely to use more water for various domestic purposes (e.g. toilet cleaning, lawn watering) which may not be relevant in homes with aged and teenager. The last and the least percentage of variance of 5.16 which is for gender composition. The more the females in a given household, the higher the domestic water use it is likely to be. Females have been found to use more water than their male counterparts. This observation of female gender contribution to domestic water use is similar to that of Xinming et al. (1990).

Domestic water use modelling

The results of multiple regression analysis as revealed in Table 9, show a high coefficient of determination (r^2 = 80.60% ± 72.94 SE) at 95% significance level. This shows that the variables extracted are valid for explanation of variation in domestic water use in the study area.

Table 10. Coefficients of the predictors of domestic water use in the study area.

Model	Unstandardized coefficients		Standardized coefficients	T	Significance
	B	Std. Error	Beta		
(Constant)	476.020	7.371		64.584	0.000
Water supplied for bathing	71.976	7.408	0.457	9.716	0.000
Age range of suppliers	65.428	7.408	0.415	8.832	0.000
Quantity of water supplied	61.848	7.408	0.392	8.349	0.000
Household size	-59.188	7.408	-0.376	-7.990	0.000
Water supplied for dish washing	54.299	7.408	0.345	7.330	0.000

Source: SPSS-generated table.

The data was further subjected to stepwise regression analysis. The model which was generated show that five components were strong predictors of domestic water use in the study area. These are water supply for bathing, age range of fetchers, quantity of water supplied, household size and water supply for dish washing. Equation 1 shown is valid at 95% level of significance (R^2= 79.5% and S.E=73.34): The coefficients of the variables are presented in Table 10. The standardised model generated is presented in equation below:

$$Y = 476.02 + 0.457_{BAT} + 0.415_{AGR} + 0.392_{QTS} - 0.376_{HSZ} + 0.345_{DSW}$$

Where: Y is the predicted daily total household water use, BAT is water supplied for bathing, AGR is age range of water suppliers/fetchers, QTS is quantity of water supplied, HSZ is the Household size, and DSW is water supplied for dish washing.

Conclusion

An investigation into the determinants of domestic water use in Iwo, a growing city in Osun State, Nigeria has been examined. The results of descriptive statistics showed that females form the larger proportion of the respondents and 98.00% have maximum of 20 min to their various water sources. Also, the dominant sources of water of 100% of the respondents are hand-dug wells and boreholes. Factor analysis extracted nine water use components out of the 31 components involved in the analysis. The nine components explain 76.00% of the variations in domestic water use in the study area. Multiple regression analysis shows a high r^2 value of 80.60%. The all-inclusive model generated from stepwise regression analysis show that five water use components are strong predictors of domestic water use. Daily household water use model derived is valid at 95% level of significance (r^2= 79.50% ± 73.34SE). The components are water supply for bathing, age range of water suppliers/fetchers, quantity of water supplied, household size and water supplied for dish washing.

In conclusion, it is evident from this study that domestic water use on daily basis in Iwo is not static but rather under the influence of certain variables. The implication of the results here is that policy makers in urban water supply planning should incorporate such variables as discovered here in water supply planning for its sustainability. However, since water sources were observed to be within the reach of the respondents, it is suggested that investigation be conducted on the reliability of water sources in terms of its quality to ensure its conformity with the standard recommended to safeguard human health.

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Appendix 1. Questionnaire on the determinants of domestic water demand in a growing city in Osun State, Nigeria.

Section A

1. Name of Ward___________________________________
2. Level of Education: A No education___. B. Primary___C. Secondary____D. Tertiary___E. Others___
3. Sex: Male_____Female_____
4. Age:A. <18_____B.19-45______C. 46-65____ D>65___
5. Household size A. <5____B. 6-10____C. 11-15____D. 16-20____E. >20______
6. Type of the House: Traditional Compund___ B. Modern Self-Contained____
7. Number of Rooms in your house A. 2__ B. 3___. C. 4___ D. 5____ E.>5___
8. No of Females in the family____________ No of Males in the family_______________
9. No of Children of school age in the family A. <3___ B. 4-6____ C. 7-9___ D. >9____
10. Occupation A. Farming___ B. Trading___ C. Civil Service___ D. Others ___
11. What is your monthly gross income? A.<N10,000____B. N10,000- N25,000____

C. N26,000 – N40,000_____D.N41,000-N55,000_____E. >N55000_______

12. What is the distance of the nearest water source to your house?___

Section B: Kindly complete the table below appropriately.

	Micro-component uses	Quantity of household water use per day (in litres)	Sources of water (Please tick appropriately)						
			River	Pipe-borne	Hand-Dug Well	Powered bore hole	Rain water	Vendor	Bottled/ sachet water
13.	Drinking								
14.	Cooking								
15.	Bathing								
16.	Cloth Washing								
17.	Dish washing								
18.	Toilet flushing								
19.	Car washing								
20.	Others								

21. Is your house connected to a running pipe borne water ? Yes_____ No_____
22. Which of these sources is located within your house? Hand-dug well___Borehole___Tap water.___
23. Do you conserve water? A. No____ B. Yes____
24. If yes to question 19, how do you conserve water?

A. Drums_____B. Overhead tanks_____C.underground tank____D.others______

25. If No, to question 19, why? A. water runs 24hrs/day___B.No container____ C.Distance of the water source_____
26. How often do you fetch water? A. Daily___B. every 2days___C. Every 3days___D. weekly___
27. How long does it take you to collect water for home use?

A. <10minutes ____B. 11-20minutes____C. 21-30minutes____D. >30minutes._____

28. Do you pay for water? Yes_____ No_____
29. If yes to 25, how much do you pay monthly?A.<#250__B. #250-#500__

C. #500-#750___ D. >#750____

30. How often does your tap run? A. Daily____ B. Once a week___ C. Twice a week D. weekly
31. Which of the sources will you prefer? A. Hand-dug well____B. Borehole___C. Stream/river____D.Pipe Borne____
32. State the reason for your choice in No 27 ____

____

33. Is your water source reliable'? A. Yes_______ B. No_______
34. If your answer in 31 is No, can you please give the reason? ______________

____

35 . Who is responsible for fetching water in your family? A. Females___ B. children____ C. Men_____
D. Vendors____ E. All of the above______

36. What is the age range of those family members that are responsible for fetching water in your family?
A. 5-12yrs____B. 13-18yrs____C.18-25yrs___D. >25yrs____
37. How many litres of water would they fetch in a day?
A. <50litres ____ B. 51-100litres____C. 100-150litres____D. >150-200litres______E. >200litres
38. What time of the day do you use more water? A. Morning time___ B. Afternoon time___ C. Evening time_____
39. Give your general view of the water supply situation in your village A. Adequate__B. Inadequate__ C. Poor____

Appendix 2. Water use components analysed.

1. Level of education
2. Sex
3. Age of the Head
4. Household size
5. Religion
6. No of females
7. No of males
8. No of children
9. Monthly Income
10. Distance from source
11. Reliability of the source
12. Water supply for drinking
13. Water supply for cooking
14. Water supply for bathing
15. Water supply for cloth washing
16. Water supply for dish washing
17. Water supply for sanitation
18. Water supply for car washing
19. Other uses
20. Time spent
21. Price of water
22. Tap availability
23. Source preference
24. Regular supply of water
25. Reason for the regularity
26. Reliability during dry period
27. Reason for reliability (dry)
28. Alternative source
29. Age range of fetchers
30. Quantity fetched by the fetchers
31. Respondents’ view on water accessibility.

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Full Length Research Paper

Assessment of heavy metals concentration in soils around oil filling and service stations in the Tamale Metropolis, Ghana

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This study was conducted to assess the level of heavy metals contamination of soil around oil filling and service stations in the Tamale Metropolis. Soil samples were collected from various oil filling and service stations. Elemental analysis of samples was conducted using atomic absorption spectrometer at Atomic Energy Laboratory, Accra. The metals concentrations ranged from 2.37 to 15.00 mg/kg for Cr; 0.01 to 0.03 mg/kg for Hg; 3.2 to 22.68 mg/kg for Cu; 0.12 to 6.63 mg/kg for Cd and 4.93 to 74.20 mg/kg for Pb. The mathematical models: Index of geoaccumulation (Igeo), enrichment factors (EF), contamination factor and degree of contamination were employed to identify possible levels of pollution from anthropogenic sources. The enrichment factor means places the elements in a decreasing order as Cd > Pb > Cr > Cu > Ni> Fe > Zn > As > Hg > Mn that agreed with others models such as contamination factor, pollution load index and degree of contamination. Elements such as chromium (Cr), copper (Cu), lead (Pb) and manganese (Mg) gave enrichment factor values ranging from 2-5 signifying moderate enrichment. The study revealed that soil contamination by the metals originated from a common anthropogenic source such as the oil filling activities, brake wear, tyres wear and corroded vehicles engine materials since these sources are noted to contribute one or two correlated metals to the natural environment. Hence, pose potential threat to humans and critical environmental media such as water bodies. It is therefore recommended that Environmental Protection Agency (EPA) should regularly monitor the oil filling and service stations to check the levels of heavy metals in the metropolis.

Key words: Oil filling, service station soils, enrichment factor, heavy metals, cadmium, Tamale Metropolis.

INTRODUCTION

Heavy metals are among the more serious pollutants in our natural environment due to their toxicity, persistence and bioaccumulation potential (Caeiro et al., 2005). Following the introduction of heavy metal contaminants

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into a river, whether via natural or anthropogenic sources, they partition between aqueous (pore water, overlying water) and solid phases (sediment, suspended particulate matter and biota) (Prudencioa et al., 2007; Zhang et al., 2007). Heavy metals are natural constituents of the earth crust. A number of these elements are biologically essential and are introduced into aquatic enrichments by various anthropogenic activities (Al-Khashman, 2004). The main anthropogenic sources of heavy metals are the industrial point sources, the present and former mining activities, foundries, smelters and diffuse sources such as piping, constituents of products, combustion of by products, traffic and human activities (Nilgun et al., 2004).

Heavy metals at trace levels present in natural water, air, dusts, soils and sediments play an important role in human life (Isaac et al., 2004). Soils are critical environment where rock, air and water interface (Facchinelli et al., 2001; Jonathan et al., 2004). Topsoil and dusts in urban areas are indicators of heavy metal contamination from atmospheric deposition. It has been noted that location close to roads are severally polluted by heavy metals such as Pb, Zn, Cu, Cd among others from traffic (Guvenc et al., 2004; Wilson et al., 2005).

With the rapid industrialisation and economic development, heavy metals are continuously introduced to soils and sediments via several pathways, including fertilisation, irrigation, rivers, runoff, atmospheric deposition and point sources, where metals are produced as a result of metal mining, refining and refinishing by-products. Soils are usually regarded as the ultimate sink for heavy metals discharged into the environment (Banat et al., 2005), and sediments can be sensitive indicators for monitoring contaminants in aquatic environments (Pekey et al., 2004). Therefore, the environmental problem of soil and sediment pollution by heavy metals has received increasing attention in the last few decades in both developing and developed countries throughout the world (Zhang et al., 2007).

Pollution of the natural environment by heavy metals is a universal problem because these metals are indestructible and most of them have toxic effects on living organisms when permissible concentration levels are exceeded. Heavy metals frequently reported in literature with regards to potential hazards and occurrences in contaminated soils are Cd, Cr, Pb, Zn, Fe and Cu (Akoto et al., 2008). Environmental contamination by hydrocarbons and petroleum products constitute nuisance to the environment due to their persistent nature and tendency to spread into ground and surface waters. Environmental pollution with petroleum and petrochemical products has attracted much attention in recent decades. The presence of various kinds of automobiles and machinery vehicles has caused an increase in the use of motor oil. Oil spillages into the environment have become one of the major problems. Used motor oils such as diesel or jet fuel contaminate natural environment with hydrocarbon (Husaini et al., 2008).

The environment can potentially be affected by numerous operations in petroleum exploration, production and transportation, with common sources of contamination being leaking underground storage tanks (Nadim et al., 2000). The chemical composition of soil, particularly its metal content is environmentally important, because toxic metals concentration can reduce soil fertility, can increase input to food chain, which leads to accumulate toxic metals in food stuffs and ultimately can endanger human health. Metals occur naturally in the earth's crust and their contents in the environment can vary between different regions resulting in spatial variations of background concentrations. The distribution of metals in the environment is governed by the properties of the metal and influences of environmental factors (Khlifi and Hamza-Chaffai, 2010).

People living in industrial cities are particularly exposed to this decline in environmental quality leading to human health problems. Urban environments are affected by natural cycles, where air, water and soil are altered by products ultimately returned to the environment in the form of nuisance. The main sources of pollution is fumes from vehicles, sediments from service stations and oils washed into water bodies, organic matter of plant and animal origin, industrial and sewage effluents. In recent times, many investors are into the oil vending (petrol and diesel) business, resulting into uncontrolled sprouting of oil filling and service stations. Some of these oil filling and service stations fails to go through environmental impact assessment process due to cost, political reasons and bureaucratic nature of the process. Hence, due to improper siting of the stations, improper storage tanks or material use for storage and mishandling of fuel during delivery can leach or contribute heavy metals concentrations that can pose threat to environmental media. The objective of this study was to assess the levels of heavy metal concentration around the oil filling and service stations in the Tamale Metropolis.

MATERIALS AND METHODS

Study area

Tamale is the capital of Northern Region of Ghana (Figure 1). Tamale is the third most populous settlement in Ghana with 537,986 inhabitants according to the 2012 census (GhanaWeb.com). The town is located 600 km north of Accra. The metropolis experiences one rainy season from April to September or October with a peak in July and August. The mean annual rainfall is 1100 mm within 95 days of rainfall in the form of tropical showers. Consequently, staple crop farming is highly restricted by the short rain season. The dry season is usually from November to early April. It is influenced by the dry North-Easterly (Harmattan) winds while the rainy season is influenced by the moist South Westerly winds. The mean day temperatures range from 28°C (December and mid-April) to 43°C (March, early April) while mean night temperatures range from 18°C (December) to 25°C

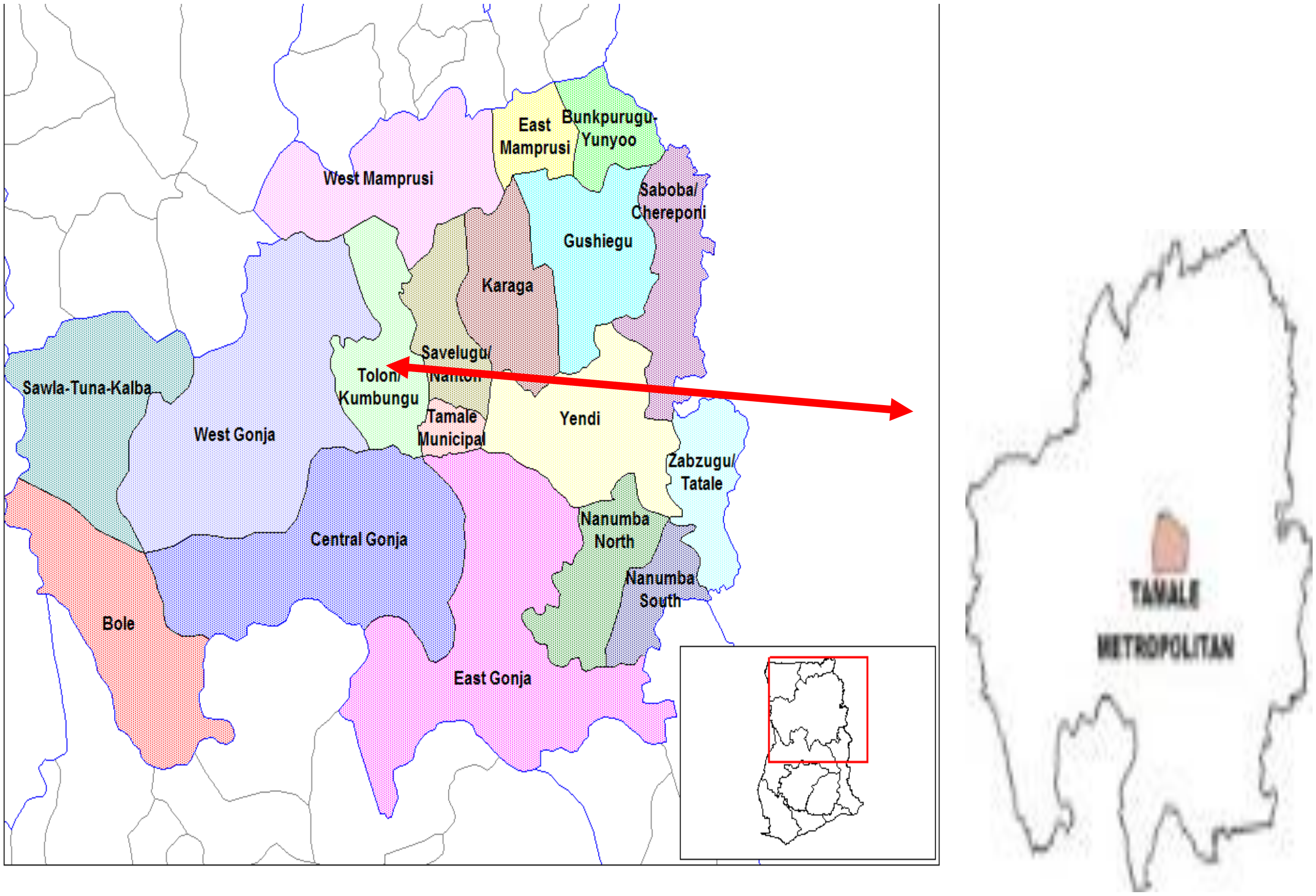

Figure 1. Map of the northern region with Tamale Metropolis singled out.

(February, March). The mean annual day sunshine is approximately 7.5 h.

Sampling techniques

Soil samples were taken from twenty one (21) oil filling and service stations in the Tamale Metropolis. At the sampling sites, about 500 g of soil sample were collected by sweeping using soft touch brush, scoop, trowel and plastic containers. One sample was collected from each filling or service station. The oil filling and service station soil samples were stored in sealed plastic containers, carefully labelled and taken to National Nuclear Research Institute of the Ghana Atomic Energy Commission laboratory for elemental analysis.

Digestion of oil filling and service stations soil samples and analysis

Determination of heavy metals was done in accordance with protocols at National Nuclear Research Institute of the Ghana Atomic Energy Commission. The soil samples collected were air dried to a constant weight in the laboratory under room temperature for 24 hours and then sieved using 200 µm mesh. Sample of 0.5 g was weighed into a tufflin bucket and 6 ml H_2NO_3 acid, 3 ml HCl with 5 drops of H_2O_2 added to the samples on a hot plate (this step was carried out in the hood). The tufflin buckets was tightened and fixed in the rotar. The rotar was placed in the microwave oven and the machine programmed to commence the digestion process (Each sample had its own programme). The digested samples were diluted to 20 ml and transferred into the appropriate test tube. The concentrations of trace metals such as Cr, Hg, Cu, Zn, Mn, Fe, Pb, Ni, As, Co and Cd in the filtrate were determined using atomic absorption spectrometer.

Quality assurance

Strict QA/QC measures were adopted to ensure reliability of the results. All chemicals and reagents used were of high purity. Glassware used was cleaned thoroughly with detergent and rinsed several times using deionized water. Deionized water was used for all dilution purposes. For the purposes of detection and quantification limits of the AAS, a blank solution was read 25 times, and the standard deviations were calculated for the noise levels generated for each of the elements of interest. The detection limit (LOD) for each element was achieved as follows:

$$\mathrm{LOD} = \frac{3 \times S}{m}$$

Where S is the standard deviation of the blank readings and m represents the gradient of the calibration curve for each element. The limit of quantification was calculated using 10 s/m. The accuracy and reproducibility of the analytical procedure was determined by spiking and homogenizing three replicates of each of three samples selected at random. Triplicate of each sample was spiked with three different concentrations of the element of interest as follows: Cd (0.5, 2.0 and 3.0 mg/l), Cr (1.0, 2.0 and 5 mg/l), Ni (2.0, 5.0 and 10.0 mg/l), Co (2.0, 5.0 and 8.0 mg/l), Fe (2.0, 5.0 and 10.0 mg/l), Zn (0.25, 0.5, and 1.0 mg/l) and Pb (2.0, 5.0, and 10.0 mg/l) and treated in a similar manner as the samples. The absorbances measured by the AAS were converted to concentrations using standard calibration curves. One thousand milligrams per liter single element standards of the elements of interest, obtained from Fluka Analytical (Sigma Aldrich Chemie GmbH, Switzerland), were diluted using 10 % HNO and used to generate the calibration curves for the AAS analysis.

The oil filling and service station's soil contamination were assessed using various indices: contamination factor (CF) (degree of contamination), pollution load index (PLI), enrichment factor (EF) and index of geoaccumulation (Igeo).

Contamination factor

To assess the extent of contamination of heavy metals in oil filling and service station soils, contamination factor and degree of contamination was used (Rastmanesh et al., 2010). The C^if is the single element index which is determined by the relation:

$$C^if = \frac{C^i_{o-1}}{C^i_n} \quad (1)$$

Where C^if is the contamination factor of the element of interest, C^i_{o-1} is the concentration of the element sample, C^i_n is the background concentration, in this study the continental crustal average was used (Taylor and Meclenan, 1985). C^if is defined according to four categories: <1 low contamination factor, 1-3 moderate contamination factors, 3-6 considerable contamination factors and > 6 very high contamination factor.

Degree of contamination

The sum of the contamination factors of all the elements in the sample gives the degree of contamination as indicated in the equation below:

$$C_{deg} = \Sigma\, C^if \quad (2)$$

Four categories has been defined for the degree of contamination as follows: <8 low degree of contamination, 8-16 moderate degree of contamination, 16-32 considerable degree of contamination and > 32 very high degree of contamination.

Pollution load index

Each oil filling and service station soil was evaluated for the extent of metal pollution by employing the method based on the pollution load index (PLI) developed by Thomilson et al. (1980) as follows:

$$PLI = {}^n\surd\, (CF1 \times CF2 \times CF3 \times \ldots\ldots CFn) \quad (3)$$

where n is the number of metals studied and CF is the contamination factor calculated as described in an earlier equation. The PLI provides simple but comparative means for assessing a site quality, where a value of PLI < 1 denote perfection; PLI = 1 shows that only baseline levels of pollutants are present and PLI >1 indicate deterioration of site quality (Thomilson et al., 1980).

Enrichment factor

Enrichment factor (EF) of an element in the studied samples is based on the standardization of a measured element against a reference element. A reference element is often the one characterized by low occurrence variability. It is used to differentiate heavy metals originating from human activities and those of natural sources. This is determined by the relation:

$$EF_X = [X_S / E_{S\ (ref)}] / [X_C / E_{C\ (ref)}] \quad (4)$$

where EFx is the enrichment factor for the element X, Xs is the concentration of element of interest in sample, Es $_{(ref)}$ is the concentration of the reference element used for normalization in the sample, Xc is the concentration of the element in the crust and Ec $_{(ref)}$ is the concentration of the reference element used for normalization in the crust (Taylor and Meclenan, 1985). Five contamination categories were recognized on the basis of the enrichment factor: EF < 2 deficiency to minimal enrichment, EF = 2-5 moderate enrichment, EF = 5-20 significant enrichment, EF = 20-40 very high enrichment and EF > 40 extremely high enrichment (Yongming et al., 2006; Kartal et al., 2006).

Index of geoaccumulation

The index of geoaccumulation (Igeo) is widely used in the assessment of contamination by comparing the levels of heavy metal obtained to a background levels originally used with bottom sediments (Muller, 1969). It can also be applied for the assessment of oil filling and service station soil samples contamination. It is calculated using the equation:

$$Igeo = log_2\,(C_n / 1.5B_n) \quad (5)$$

C_n is the measured concentration of the heavy metal in oil filling and service station soil and B_n is the geochemical background concentration of the heavy metal (crustal average) (Taylor and Meclenan, 1985). The constant 1.5 is introduced to minimize the effect of possible variations in the background values which may be attributed to lithologic variations in the sediments (Lu et al., 2010). The following classification is given for geoaccumulation index (Huu et al., 2010; Muller, 1969): < 0 = practically unpolluted, 0-1 = unpolluted to moderately polluted, 1-2 = moderately polluted, 2-3 = moderately to strongly polluted, 3-4 = strongly polluted, 4-5 = strongly to extremely polluted and > 5 = extremely polluted.

Statistical analysis

The heavy metals concentrations in oil filling and service station soils were subjected to Pearson's significant correlation analysis using SPSS version 16 to determine the relationship and characteristic between the metals.

RESULTS AND DISCUSSION

Heavy metal concentration in oil filling and service station

Heavy metals enter the environment by natural and anthropogenic means such as natural weathering of the earth's crust, mining, soil erosion, industrial discharge, urban runoff, sewage effluents, pest and disease control agents applied to plants, air pollution fallout, oil leaking

Table 1. Mean concentration of heavy metals (mg/kg) in soils within oil filling and service stations.

Elements	Minimum	Maximum	Mean	Std. Deviation	Soils worldwide
Cr	2.37	15	7.35	2.6	80
Hg	0.01	0.03	0.02	0.01	-
Cu	3.29	22.68	7.86	4.79	25
Zn	3.15	10.43	5.57	2.02	70
Mn	0.23	3.69	2.54	0.75	9
Fe	1.5	1.68	1.53	0.04	35
Pb	4.93	74.2	19.51	15.13	17
Cd	0.12	6.63	2.29	3.76	0.3
Ni	1.79	5.29	2.82	0.87	20
As	0.13	0.71	0.33	0.16	-

from underground storage tanks, corroded metallic storage tanks among others. The mean concentration of the elements obtained from soil samples from oil filling and service stations in the Tamale Metropolis shows decreasing order of Pb> Cu > Cr > Zn > Ni > Mn > Cd > Fe > As> Hg. The concentration of chromium (Cr) ranged from 2.37 to 15.00 mg/kg with a mean of 7.35±2.60 mg/kg (Table 1). Variety of small large scale industrial activities for example metal plating, anodizing, dyes, pigments, ceramic, glues, tanning, wood preserving and textiles are reported to contribute to Cr levels (Alloway, 1995). The concentration of mercury (Hg) ranged from 0.01 to 0.03 mg/kg with a mean of 0.02±0.01 mg/kg (Table 1). Exposure to high levels of metallic, inorganic or organic mercury can permanently damage the brain, kidneys and developing fetus.

Concentrations of copper (Cu) ranged from 3.2 to 22.68 mg/kg with a mean of 7.86±4.79 mg/kg 9 (Table 1). Cu is an essential element, but may be toxic to both humans and animals when its concentration exceeds the safe limits. Cu is used in numerous applications because of its physical properties. The toxicity for humans is not very high (Poggio et al., 2009). Cu normally accumulates in the surface horizons, a phenomenon explained by the bioaccumulation of the metal and recent anthropogenic sources (Abdul Hameed et al., 2013). The observed values of the Cu content did not exceed the normal threshold value prescribed in soil (20 - 30 mg/kg) (Alloway, 1995). Manganese (Mn) recorded concentration levels ranging from 0.23 to 3.69 mg/kg with a mean of 2.54±0.75 mg/kg (Table 1). Manganese deficiency in the human body can produce severe skeletal and reproductive abnormalities in mammals. High doses of manganese can cause adverse effects primarily on the lungs and on the brain. The concentration of Zinc (Zn) ranged from 3.15 to 10.43 mg/kg with a mean of 5.57±2.02 mg/kg (Table 1).

Environmental contamination of Zn is mainly related to anthropogenic input. The anthropogenic sources of Zn are related to industries and the use of liquid manure, composted materials and agrochemicals such as fertilizers and pesticides in agriculture (Romic and Romic, 2003). Presence of Zn may be due to oil leaking from underground storage tanks. Iron (Fe) concentration ranged from 1.50 to 1.68 mg/kg with mean of 1.53±0.04 mg/kg. Iron is vital for almost all living organisms, participating in a wide variety of metabolic processes, including oxygen transport, DNA synthesis and electron transport. It is known that adequate iron in a diet is very important for decreasing the incidence of anaemia. The concentration of lead (Pb) ranged from 4.93 to 74.20 mg/kg with a mean of 19.51±15.13 mg/kg (Table 1). Some of the lead concentration recorded in some of the oil filling and service stations exceeded the worldwide soils value of 17 mg/kg for lead. This can be attributed to leakage of oil (petrol, diesel among others) from the storage tanks or corroded metallic storage tanks. This therefore poses a potential threat to humans and critical environmental media such as water bodies. Food is one of the major sources of lead exposure; the others are air (mainly lead dust originating from petrol) and drinking water. Lead as a toxicologically relevant element has been brought into the environment by man in extreme amounts, despite its low geochemical mobility and has been distributed worldwide (Oehlenschläger, 2002).

The concentration of cadmium (Cd) ranged from 0.12 to 6.63 mg/kg with a mean of 2.29±3.76 mg/kg as compared to worldwide soil value of 0.3 mg/kg. Widespread distribution of Cd and its high mobility makes it a potential contaminant in a wide range of natural environments. Generally, soil Cd concentrations exceeding 0.5 mg/kg are considered evidence of soil pollution (McBride, 1994). Cadmium is naturally present in the environment: in air, soils, sediments and even in unpolluted seawater. Cadmium is emitted to air by mines, metal smelters and industries using cadmium compounds for alloys, batteries, pigments and in plastics, although many countries have stringent controls in place on such emissions (Harrison, 2001). Human and environmental safety cannot be guaranteed from Cd exposure as it is obvious that the operation of some of these oil filling and service stations in the metropolis are contributing to Cd

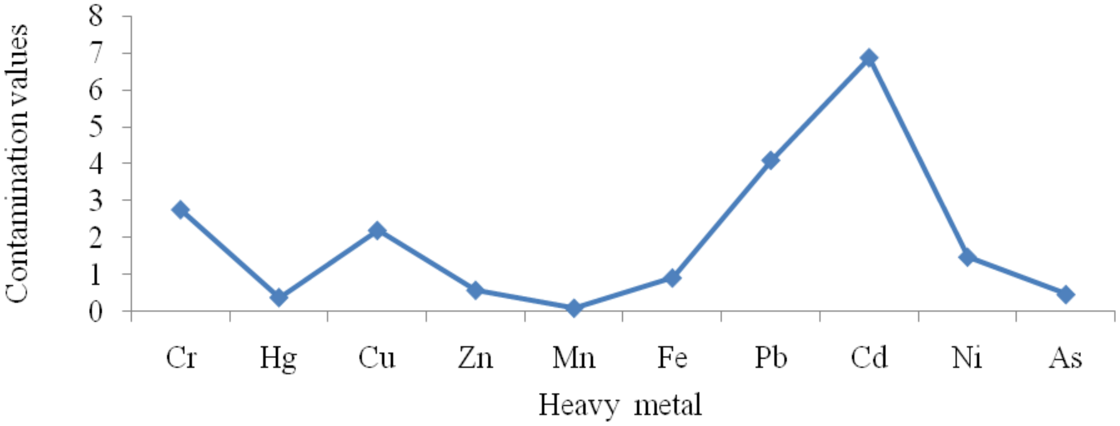

Figure 2. Degree of contamination of soil sampled from oil filling and service stations.

concentration in the soils. Many anthropogenic activities within the metropolis can increases Cd concentrations in soils well above background levels, such as sewage sludge, oil leaking from underground storage tanks and phosphate fertilizer application. Cadmium accumulates in the human body affecting negatively several organs: liver, kidney, lung, bones, placenta, brain and the central nervous system (Castro-González and Méndez-Armenta, 2008).

The concentration of Nickel (Ni) ranged from 1.79 to 5.29 mg/kg with a mean of 2.82±0.8 mg/kg (Table 1). Nickel has many common industrial uses due to its unique chemical properties. Industrially, it is used in electroplating, electroforming, in circuitry, and in nickel-cadmium batteries. Metallic nickel is non-carcinogenic to humans; however, all other nickel compounds, such as nickel sulfides, oxides, and silicates and other soluble salts are carcinogens. Carcinogenic nickel exposure is greatest through the inhalation of nickel containing particulates. The burning of fossil fuels as well as the refining of metals such as copper introduces considerable amounts of nickel into the atmosphere (Lee et al., 2005). Ni is widely used in electroplating and in the manufacture of batteries. Ni toxicity for human beings is not very high, but it can cause respiratory diseases (Poggio et al., 2009).

The concentration of arsenic (As) ranged from 0.13 to 0.71 mg/kg with a mean 0.33±0.16 mg/kg (Table 1). Arsenic is quite widely distributed in natural waters and is often associated with geological sources, but in some locations anthropogenic inputs, such as the use of arsenical insecticides and the combustion of fossil fuels, can be extremely important additional sources. Inorganic arsenic is considered carcinogenic and is related mainly to lung, kidney, bladder and skin disorders (ATSDR, 2003). Arsenic is a contaminant of public concern since it is highly toxic and carcinogenic. It may be accumulated in plants and eventually be transferred to humans through the food chain.

Contamination factor and degree of contamination

Based on the results presented in Table 2, the mean contamination factor of heavy metals shows a decreasing order as follows: Cd > Pb > Cr > Cu > Ni> Fe > Zn > Hg = As **>** Mn. The study observed highest contamination factor of 2.29 (Cd) from only one station and least contamination factor of < 0.002 (Mn) from about sixteen oil filling stations sampled. The contamination factor for elements such as Cr, Hg, Cu, Zn, Mn, Fe, Pb, Ni and As were less than 1 signifying low contamination from anthropogenic sources (Taylor and Meclenan, 1985).

Moderate contamination was obtained in Cd (2.29) in one of the oil filling station. This might be due to oil leaking from underground storage tanks or rusted fuel tanks. The degree of contamination of soil samples from various oil filling and service stations are presented in Figure 2. Generally, the study obtained degree of contamination factors less than 8 mg/kg signifies that the samples from the oil filling and service stations have low degree of contamination (Taylor and Meclenan, 1985). The study reveals that the variability in the range of all the metal distributions as compared to their degree of contamination respectively is an indication of low pollution of the sample. The degree of contamination of heavy metals shows a decreasing order as follows: Cd > Pb > Cr > Cu > Ni> Fe > Zn > As > Hg > Mn.

Pollution load index

The study recorded pollution load index less than 1mg/kg for all the heavy metals parameters considered which denotes perfection (Thomilson et al., 1980). Based on the results presented in Figure 3, the decreasing trend of pollution load index of heavy metal levels was as follows: Cd > Pb > Cr > Cu > Ni > Fe > Zn > As > Hg > Mn. This suggests that oil filling and service stations within the various sites have not contributed to the heavy metal concentrations in the soils. Possible inputs of heavy

Table 2. Contamination factor of soil sampled from oil filling and service stations.

Location	Cr	Hg	Cu	Zn	Mn	Fe	Pb	Cd	Ni	As
Naagamni	0.03	0.03	0.1	0.02	0.01	0.05	0.2	6.63	0.06	0.03
Sky Petrol	0.06	0.05	0.21	0.02	0.01	0.04	0.2	0.12	0.06	0.03
Petrol Bay Oil Ltd	0.14	0.03	0.1	0.04	0.01	0.04	0.2	0.12	0.07	0.03
Frimps Oil	0.11	0.03	0.05	0.02	0	0.04	0.1	<0.002	0.08	0.01
Total Oil	0.08	0.05	0.07	0.02	0.01	0.04	0.7	<0.002	0.05	0.04
LP Oil	0.11	0.03	0.3	0.04	0.01	0.04	0.3	<0.002	0.08	0.04
Shell Kunain Road	0.11	0	0.04	0.02	0	0.04	0.1	<0.002	0.06	0.03
Deliman Oil	0.13	0.03	0.08	0.03	0.01	0.04	0.2	<0.002	0.07	0.02
Shell Hospital Rd	0.2	0.03	0.1	0.03	0.01	0.05	0.1	<0.002	0.13	0.02
Total Kum Avenue	0.11	0.03	0.07	0.03	0	0.04	0.2	<0.002	0.07	0.05
Goil Hsp Road	0.11	0	0.07	0.03	0	0.04	0.1	<0.002	0.09	0.02
Goil Tamale Cent.	0.1	0	0.05	0.02	0	0.04	0	<0.002	0.05	0.01
Shell Yendi Road	0.09	0	0.07	0.02	0	0.04	0.1	<0.002	0.08	0.01
Quantum Oil	0.05	0	0.11	0.02	0.01	0.04	0.1	<0.002	0.06	0.01
Star Oil	0.1	0.03	0.12	0.04	0.01	0.05	0.2	<0.002	0.08	0.02
Gbanzaba Sev. Sta	0.06	0	0.21	0.04	0.01	0.04	0.1	<0.002	0.06	0.02
Sky Petrol No 2	0.11	0	0.12	0.05	0.01	0.05	0.3	<0.002	0.07	0.02
Nasona	0.08	0.05	0.1	0.03	0.01	0.04	0.2	<0.002	0.06	0.02
Shell	0.11	0	0.11	0.03	0.01	0.04	0.4	<0.002	0.12	0.02
Total	0.78	0	0.06	0.02	0	0.04	0.1	<0.002	0.05	0.01
Goil	0.08	0	0.07	0.02	0	0.04	0.1	<0.002	0.05	0.01
Mean	0.13	0.02	0.1	0.03	0	0.04	0.2	2.29	0.07	0.02

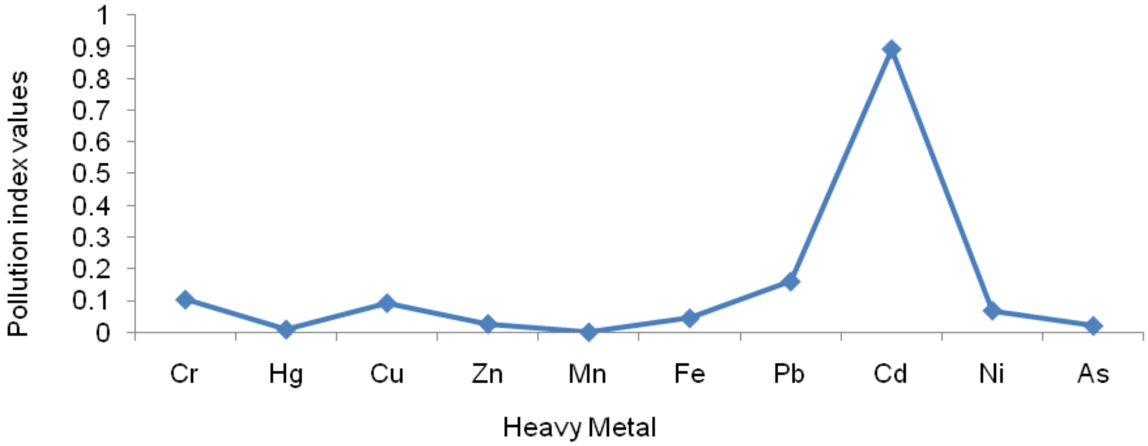

Figure 3. Pollution index of soils within oil filling and service stations.

metals from anthropogenic sources such as oil leaking from underground storage tanks, rusted storage tanks and corroded engines from vehicles that take fuel from the oil filling and service stations were less within the soil samples from the oil filling and service stations using the continental crust average where Fe (Iron) was used as reference element for normalization. The results of the enrichment factor calculations shows enrichment of some elements in the soil samples from the oil filling and service stations. Based on the results presented in Table 3, the mean enrichment factor of the heavy metals such as Hg, Mn, Fe, Zn, Ni and As gave values less than 2 signifying deficiency to minimal enrichment (Taylor and Meclenan, 1985). Hence, oil filling and service station activities and other human actions contribution to the release of these metals into that environment were minimal. Elements such as Cr, Cu, Pb and Mg gave enrichment factor values from 2–5 signifying moderate

Table 3. Enrichment factor of soils within oil filling and service stations.

Location	Cr	Hg	Cu	Zn	Mn	Fe	Pb	Cd	Ni	As
Naagamni	0.67	0.54	2.08	0.50	0.15	1	3.90	138	1.29	0.67
Sky Petrol	1.46	1.23	4.80	0.50	0.14	1	4.90	2.73	1.39	0.71
Petrol Bay Oil Ltd	3.19	0.61	2.26	0.84	0.16	1	5.00	2.79	1.70	0.63
Frimps Oil	2.47	0.61	1.07	0.42	0.09	1	2.50	0.05	1.77	0.23
Total Oil	1.91	1.26	1.58	0.54	0.12	1	17.0	0.05	1.19	0.91
LP Oil	2.57	0.59	6.86	1.00	0.14	1	6.50	0.05	1.71	0.82
ShellKunain Road	2.63	0.05	1.02	0.37	0.07	1	2.10	0.05	1.37	0.65
Deliman Oil	2.98	0.61	1.79	0.79	0.12	1	4.90	0.05	1.56	0.56
Shell Hospital Rd	4.44	0.58	2.27	0.60	0.13	1	2.90	0.04	2.93	0.33
Total Kum Avenue	2.54	0.61	1.44	0.58	0.09	1	4.80	0.05	1.51	1.09
Goil Hsp Road	2.63	0.05	1.67	0.61	0.09	1	3.30	0.05	1.98	0.47
Goil Tamale Cent.	2.26	0.05	1.09	0.37	0.07	1	1.10	0.05	1.09	0.21
Shell Yendi Road	2.16	0.05	1.61	0.42	0.09	1	1.50	0.05	1.95	0.28
Quantum Oil	1.21	0.05	2.58	0.49	0.12	1	2.40	0.05	0.14	0.33
Star Oil	0.05	0.58	2.64	0.82	0.13	1	3.60	0.04	1.82	0.53
Gbanzaba Sev. Sta	1.44	0.05	4.89	1.02	0.12	1	2.70	0.05	1.30	0.37
Sky Petrol No 2	2.47	0.05	2.58	1.16	0.13	1	5.80	0.04	1.58	0.47
Nasona	1.93	1.26	2.28	0.74	0.12	1	3.90	0.05	1.40	0.40
Shell	2.41	0.05	2.59	0.59	0.11	1	9.50	0.05	2.68	0.48
Total	18.2	0.05	1.49	0.49	0.09	1	2.60	0.05	1.16	0.28
Goil	1.95	0.05	1.61	0.54	0.00	1	2.60	0.05	1.05	0.33
Mean	2.93	0.42	2.39	0.64	0.11	1	4.45	6.88	1.55	0.51

enrichment (Table 3). Basically, as the EF values increase the contribution of anthropogenic origins also

the metal is entirely from crusted material or natural processes, whereas EF greater than 1.5 suggests the source is more likely to be anthropogenic. This indicates that the concentration of some of the elements in the soil is as a result of the oil filling, service stations activities and other human action close to the stations. Cadmium (Cd) recorded enrichment factor from one of the sampling site to be greater than 5 indicating very extreme enrichment (Taylor and Meclenan, 1985). The high level is attributed to the fact that storage materials might have rusted and/or leakages of some amount of oil from underground storage tanks on the soil and perhaps due to runoff of agricultural waste to the station site.

Index of geoaccumulation

Table 4 shows the index of geoaccumulation of the soil samples from the various oil filling and service stations. The decreasing trend of averages of metal levels was as follows: Pb > Cr = Cu >Ni > Fe > Zn> As > Hg > Mn = Cd. The result from Table 4 shows that all the heavy metal parameters recorded index of geoaccumulation less than 0.00 which signifies that the samples from the oil filling and service stations were practically unpolluted. Only Cd recorded geoaccumulation index of 2.14 from only one oil filling station which indicates a moderately to strongly increase (Surthland et al., 2000). According to Zhang and Liu (2002), the EF value between 0.5-1.5 indicates polluted site (Huu et al., 2010; Muller, 1969). This can be attributed to oil leaking from underground storage tanks, corroded metallic tanks and from the exhaust of vehicles that come to the stations or ply roads close to them. Most of these stations are located close to major roads in the metropolis.

Correlation matrix

Table 5 shows the correlation matrix of heavy metals in soil samples from various oil filling and service stations. The study observed strong positive correlations between some elements such as Cr/Ni (0.65), Cu/Zn (0.59), Mn/Fe (0.58) and Pb/As (0.58) at 1% significant levels. The strong positive correlations signify that each paired elements have common contamination sources.

The study observed strong positive correlation between Zn and Mn (0.51) at 5% significant levels indicating common contamination sources. While Pb and Cd shows a strong negative correlation of -1.0 at 1% significant level. The correlation analysis indicates that soil contamination by the metals originated from a common anthropogenic source such as leaking of diesel, petrol, brake wear, tyres wear and corroded vehicles engine materials since these sources are noted to be contributing one or the pair correlated metals to the

Table 4. Index of geoaccumulation of soils within oil filling and service stations in the Tamale metropolis.

Location	Cr	Hg	Cu	Zn	Mn	Fe	Pb	Cd	Ni	As
Naagmi	-5.6	-5.9	-3.9	-6.0	-7.8	-5.0	-3.0	2.1	-4.6	-5.6
Sky Petrol	-4.5	-4.8	-2.8	-6.1	-8.0	-5.1	-2.8	-3.6	-4.6	-5.6
Petrol	-3.4	-5.9	-4.0	-5.4	-7.8	-5.1	-2.8	-3.6	-4.4	-5.8
Frimps Oil	-3.8	-5.9	-5.0	-6.4	-8.4	-5.1	-3.8	-9.6	-4.3	-7.3
Total	-4.2	-4.8	-4.5	-6.0	-8.3	-5.1	-1.0	-9.6	-4.9	-5.3
Lp Oil	-3.7	-5.9	-2.3	-5.1	-8.1	-5.1	-2.4	-9.6	-4.3	-5.4
Shell Kum Rd	-3.7	-9.6	-5.1	-6.6	-8.8	-5.1	-4.0	-9.6	-4.7	-5.8
Deliman Oil	-3.5	-5.9	-4.3	-5.5	-8.1	-5.1	-2.8	-9.6	-4.5	-6.0
Shell Hsp Rd	-2.9	-5.9	-3.9	-5.8	-8.0	-5.1	-3.5	-9.6	-3.5	-6.6
Total Kum. Rd	-3.8	-5.9	-4.5	-5.9	-8.5	-5.1	-2.8	-9.6	-4.5	-5.0
Goil Hsp Rd	-3.7	-9.6	-4.4	-5.9	-8.5	-5.1	-3.4	-9.6	-4.1	-6.3
Goil Tam Cen	-4.0	-9.6	-5.0	-6.5	-8.8	-5.1	-4.9	-9.6	-5.0	-7.4
Shell Yen Rd	-4.0	-9.6	-4.4	-6.3	-8.4	-5.1	-4.6	-9.6	-4.1	-7.7
Quantum Oil	-4.9	-9.6	-3.8	-6.2	-8.3	-5.1	-3.9	-9.6	-4.6	-6.7
Star Oil	-4.0	-5.9	-3.7	-5.3	-8.0	-5.1	-3.2	-9.6	-4.2	-6.0
Gbanzaba Ss.	-4.6	-9.6	-2.8	-5.1	-8.2	-5.1	-3.7	-9.6	-4.7	-6.6
Sky Pet No 2	-3.8	-9.6	-3.7	-4.8	-7.9	-5.1	-2.5	-9.6	-4.4	-6.2
Nasona	-4.2	-4.8	-3.9	-5.6	-8.2	-5.1	-3.2	-9.6	-4.6	-6.5
Shell	-3.8	-9.6	-3.7	-5.9	-8.4	-5.1	-1.8	-9.6	-3.7	-6.1
Total	-4.3	-9.6	-4.6	-6.2	-8.6	-5.1	-3.8	-9.6	-4.9	-7.0
Goil	-4.2	-9.6	-4.4	-6.1	-12	-5.1	-3.7	-9.6	-5.1	-6.7
Mean	-4.0	-7.5	-4.0	-5.8	-8.4	-5.1	-3.2	-8.4	-4.5	-6.3

Table 5. Correlation matrix of heavy metals in soils around oil filling and service stations in the Tamale metropolis.

	Cr	Hg	Cu	Zn	Mn	Fe	Pb	Cd	Ni	As
Cr	1									
Hg	-0.42	1								
Cu	-0.13	0.08	1							
Zn	0.17	-0.31	0.59**	1						
Mn	0.06	-0.17	0.41	0.51*	1					
Fe	-0.16	-0.35	0.24	0.21	0.58**	1				
Pb	-0.02	0.50	0.13	0.17	0.20	0.03	1			
Cd	-0.74	-0.50	-0.48	-0.36	0.80	0.98	-1.0**	1		
Ni	0.65**	-0.47	0.08	0.12	0.33	0.25	0.05	-0.41	1	
As	-0.04	0.10	0.27	0.18	0.29	0.29	0.58**	0.63	-0.09	1

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

natural environment. However, their contribution is minimal.

Conclusion

The study employed four contamination indexes namely, EF, Igeo, CF, degree of contamination and PLI in the assessment of level of metal contamination in the metropolis. The results of all the contamination indexes used agreed well in explaining the contaminated levels and possible sources of the metals present in the oil filling and service station soil samples. For instance, enrichment factor proved to be an effective tool in differentiating a natural origin from anthropogenic source of contamination for the various elements investigated under the study. The mean value of the enrichment factor places the elements in a decreasing order as Cd > Pb > Cr > Cu > Ni> Fe > Zn > As > Hg > Mn. The degree of contamination, pollution index and mean contamination factor provided the same trend of contamination levels as in the case of the enrichment factor. While index of geoaccumulation placed the metals in a decreasing order of Pb > Cr = Cu >Ni > Fe > Zn> As > Hg > Mn = Cd. The study revealed that there were elevated concentrations of certain metals (Cr, Cu, Pb and Cd) in terms of enrichment of the soil in some of the oil filling and service stations. For instance, the cadmium recorded enrichment factor of 138 from one of the filling station indicating very extreme enrichment. At the same oil station, geoaccumulation index for cadmium (Cd) was recorded as 2.14 signifying a moderate to strongly polluted. The correlation analysis indicates that soil contamination by the metals originated from a common anthropogenic source such as leaking of diesel, petrol, brake wear, tyres wear and corroded vehicles engine materials since these sources are noted to be contributing one or the pair correlated metals to the natural environment. However, their contribution is minimal. Hence, there is a need for concern since some of these metals pose potential threat to humans and critical environmental media such as water bodies. Based on the findings of this study, it is recommended that Environmental Protection Agency (EPA) should regularly monitor the oil filling and service stations to check the levels of heavy metals in the metropolis. The EPA should also ensure that, oil filling and service stations owner's use proper storage tanks or facilities to avoid leaking of oil from tanks.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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