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## Distribution and level of arsenic in selected environmental indicators

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In this study, total arsenic was determined in soil, common grass (*Cenchrus ciliaris*), plant leaf (Dogwood; *Cornus florida*) and an invertebrate (Stag beetle; *Rhinotia hemistictus*). This was with a view of investigating its distribution and level in the environment. Samples were randomly collected from stratified sections of the study area, processed and analysed using validated acid extraction technique. Detection of arsenic was by use of ICP-OES. Percentage recovery range of 78-92% was obtained and can be adjudged acceptable for application. Overall mean concentration of arsenic ranged from  $0.35 \pm 0.12$  to  $2.52 \pm 1.85$  mg kg<sup>-1</sup>;  $0.01 \pm 0.03$  to 0.34 mg kg<sup>-1</sup>;  $0.02 \pm 0.03$  to 0.46 mg kg<sup>-1</sup> and  $0.04 \pm 0.02$  to  $0.72 \pm 0.54$  mg kg<sup>-1</sup> across sampling sections 1 to 4 for soil, grass, leaf and insect samples respectively. Arsenic was detected in all samples, however levels obtained were below prescribed toxicity limits. Samples were highly contaminated based on contamination factors of > 6. The strong correlation coefficients (> 0.9) showed association between arsenic and analysed samples. The study revealed widespread distribution of arsenic in analysed samples which portend serious health implications across the food chain.

Key words: Pollution, trace metal, grass, arsenic, environment, health, invertebrate, Namibia.

#### INTRODUCTION

Arsenic (As) is a toxic trace metal that is non-essential and do not play any physiological role in human system even in low doses (Tchounwou et al., 2012; Chung et al., 2014). It has also been reported to be of no benefit to plants and animals (Roggeman et al., 2013). Arsenic has been implicated in the inhibition of proper functioning of important enzymes in human body (Le et al., 2013; Le et al., 2015). Arsenic also exists naturally in the earth's crust (Jang et al., 2016) just as other metals such as Cd, Pb and Mn. The level has however increased tremendously in the past decade because of anthropogenic activities (Chung et al., 2014; Chen et al., 2016). Industrial processes such as the production of herbicides, pesticides, electronic components, pharmaceuticals,

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Author(s) agree that this article remain permanently open access under the terms of the <u>Creative Commons Attribution</u> <u>License 4.0 International License</u> metal alloys and others have been found to contribute to elevated level of arsenic in the environment (Jang et al., 2016; Vimercati et al., 2017). Prevalence and elevated level of arsenic in groundwater have also been reported in groundwater in India where significant correlation between arsenic contamination in groundwater with depth and distance from river Ganga was found (Kumar et al., 2016). Similar detection of high level of arsenic in groundwater and the health effects in Bangladesh was also reported (Islam et al., 2017).

The ecosystem that is most susceptible as the recipient of the toxic pollutants is the soil. The soil has generally been described as the reservoir for pollutants including toxic trace metals (Han et al., 2017). This unenvious attribute however has serious implications on the aquatic and atmospheric ecosystems. This is due to the fact that soil can act as a conduit for toxic trace metals into other ecosystems. Erosional process of surface soil can significantly increase the metallic load of the aquatic body (Issaka and Ashraf, 2017). In addition, wind dispersal can massively mobilise and introduce metal-bound particulate matters into the atmospheric sphere (Craw and Pacheco, 2002; Martin et al., 2014). These particles can be dispersed far beyond the point of source or generation and deposited on water, on plant leaves, soil and other media.

In soil, arsenic commonly associates with minerals such as arsenopyrite (FeAsS) and inorganic arsenopyrite. In contaminated soil however, it exists mostly as inorganic arsenic (V) and (III) but can also bind to some organic compounds (Lim and Goh, 2005). Chemical conversions between the inorganic and organic forms of arsenic are usually dictated by the oxidation-reduction, and precipitation-adsorption biomethylation and volatilisation processes (Jang et al., 2016). Generally, the availability of arsenic in soil is usually influenced by some factors such as the source that is whether natural or anthropogenic, soil clay content and redox potential (Manning et al., 2002; Cai et al., 2005). However, anthropogenic activities, low clay content and high pH play significant role in the availability of arsenic in soil (Wuana et al., 2011). Arsenate, being the predominant form of As present in most soils, means that plants take up As mostly as arsenate. As such, studies on the kinetics of plant As uptake have focused almost entirely on arsenate (Meharg et al., 2002).

Some of the activities through which arsenic find its way into the ecosystems include the use of arsenical liquid in the removal of parasitic ticks from animals such as cattle through a process commonly referred to as arsenic deep. The metal is also utilised in the preservation of wood, in the medical and electronic fields as well as in several industrial processes (Sharma et al., 2011). Hence, arsenic can find its way into the soil through atmospheric deposition of metal-bound particulate matters (PMs), indiscriminate dumping of electronic components on soil and dump sites. Transfer of trace metals from contaminated soil to plant and uptake by lower animals (van der Fels-Klerx et al., 2016) and ruminants has been reported (Roggeman et al., 2013; Mandal, 2017) with possible bioconcentration and bioaccumulation across the food chain.

Prevalence of toxic level of arsenic in the ecosystems has serious health implications across the food chain. Hence, environmental monitoring of its' trend, distribution and level are usually carried out. Of interest in this study is the prevalence and distribution of arsenic in living organisms that depend on the terrestrial ecosystem in view of close and direct interaction with the ecosystem (soil) and the high toxicity of arsenic.

Hence, the study aimed at investigating the level and distribution of arsenic in selected environmental samples in a local municipal area of Namibia. This was with a view of investigating the prevalence of the metal as a result of anthropogenic activities in environmental samples of soil, grass, plant leaf and insect through their interactions and dependence on soil. Possible implications on the outcome on human and environmental health will also be reflected.

#### MATERIALS AND METHODS

#### Description of the study area

The study was conducted within the municipal township of Tsumeb, located in the Oshikoto region in the Northern part of Namibia. It has a population of about 22, 500 (NSA, 2011) and covers an area of about 271 km<sup>2</sup>. The study area is notable for its dynamic agricultural practices including food crops farming and animal husbandry as well as industrial activities. The area lies within an altitude of 1, 266 m, latitude 19° 13' 59.88" S and longitude 17° 43' 0.12" E. In view of the size of the study area, stratification was adopted, and the area was stratified into four sections for sample collection purposes. Replicate samples were randomly collected within each section here in referred to as sampling section (SS). Hence each section, designated as SS1, SS2, SS3 and SS4 and their coordinates are presented in Table 1.

#### Samples and sample collection

Selected samples utilised in this study were the soil, common grass (*Cenchrus ciliaris*) that grow widely, plant leaf (Dogwood; *Cornus florida*) and an invertebrate (Stag beetle; *Rhinotia hemistictus*) as shown in Figure 1. All samples were randomly collected from each sampling section. Soil samples were collected to a depth of about 100 mm using clean stainless-steel soil trowel. The trowel was adequately washed and rinsed with distilled water after each sampling to prevent cross contamination of soil samples which may lead to concentration augmentation across sampling areas and influence the final results obtained. Plant samples (grass and leaf) as well as the invertebrate were also collected randomly from each section.

Samples were collected from each of the four stratified sampling sections represented as SS1, SS2, SS3 and SS4 between the periods of July to November 2015. Six (6) set of samples were

 Table 1. Sample collection sections of the study area and their coordinates.

Sample Point	Coordinates			
SS1	S19°13'58,8; E 017°42'35.7′			
SS2	S19°14'41.7; E017°43'12.0			
SS3	S19°15'21.6; E017°42'08.5′			
SS4	S19°15'38.5; E017°42'43.2'			
CS	S22°34'00.1; E017°04'42.5			

\*SS=Sampling Section; CS= Control Site.

collected within this period which falls within late winter and beginning of summer period in Namibia. This was with a view of evaluating possible differences in arsenic level across sampling periods (months) and within sampling sections but not necessarily due to weather variation. Control samples were also collected within the Namibia University of Science and Technology environment. The institution is located 400 km away from the study area and is devoid of anthropogenic activities that may introduce the metal under investigation to soil. Samples were placed in transparent plastic zipper bags, labelled and taken to the laboratory for further treatment and analysis.

#### Sample treatment

All soil samples were gently dried in oven overnight for about 12 at  $30^{\circ}$ C and then ground using acid washed mortar and pestle. These were passed through a 0.63 µm pore size sieve to obtain very fine particles following similar soil pre-treatment protocol (Aziza et al., 2015; Chowdhury et al., 2016). Determination of arsenic in samples was based on the final fine powdery samples. The plant samples were rinsed with water and then distilled water to remove any attached soil particles that may cause metallic concentration augmentation. They were cut into smaller pieces with the aid of stainless steel scissors, placed in clean crucible and dried in the oven at 120°C for 24 h. Dried plant samples were also ground in clean acid-washed mortar and pestle and passed through 0.63 µm sieve to obtain fine particles on which all metallic analysis was based. This process was also applied to the invertebrate samples.

#### Quality assurance and analysis

All reagents used were of analytical grades and metal standard solutions prepared from 1000 ppm stock solution was of high purity (>99.98%) and purchased from Merck Germany. All glass ware used was properly washed, rinsed and soaked in dilute acid for 12 h and then rinsed with distilled water. Working standards were prepared from the stock solution with good linearity of calibration curve. Arsenic in samples was extracted through acid digestion process following a previously described method of Awofolu (2005). Total arsenic concentration in all samples was determined using Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES)- Optima 7000 DV from Perkin Elmer.

Quality assurance of the analytical process was by addition of arsenic standard. To 5 g of pre-digested soil sample in 100-ml beaker, 0.5 ppm of arsenic (As) was added. This was followed by 3 ml of 30 %  $H_2O_2$  and the contents were allowed to stand for 1 h until the vigorous reaction ceased. Thereafter, about 75 ml of 0.5 M solution of HCl was added and the content heated gently at low heat for about 2 h on hot plate. The content was allowed to cool,

filtered into 50 ml standard flask and made up with deionised water. Triplicate digestion together with blank was carried out to verify the precision and bias of the process respectively. The process was also applied to invertebrate sample. For grass and leaf samples, 0.5 ppm of standard As was added to 0.5 g of pre-digested sample of each in a 100-ml beaker and digested with 5 ml of conc. HNO<sub>3</sub> and 2 ml of HClO<sub>4</sub> on low heat until the volume was about 2 ml. The content was allowed to cool, filtered into 50 ml standard flask using 0.45  $\mu$ m Millipore Filter paper and then made to volume with deionised water. Triplicate digestion was also carried out as previously described (Awofolu, 2005)

#### Statistical analysis

Pearson Correlation Coefficient (r) using Microsoft Excel version 2010 was used to evaluate possible relationship between arsenic load and the analysed samples across SS1-SS4, using the overall mean concentration as presented in Table 7. The extent of contamination of these samples by arsenic was also assessed through the contamination factor (CF) as applied in a previous study (Likuku et al., 2013). The CF was calculated as the ratio of the overall mean concentration of arsenic in samples across SS1-SS4 to that obtained from the control site (CS).

That is: CF = X/CS

Where X = overall mean concentration and CS = metal concentration at control site. Classification of the degree of contamination is as shown in Table 8. Two-factor analysis of variance (ANOVA) at p = 0.05 using Microsoft Excel version 10 was applied for possible statistical significance between the metal and analysed samples.

#### RESULTS

#### Method quality assurance

Results of the quality assurance process of experimental protocol utilised in the analysis of environmental samples are presented in Table 2. Percentage recoveries of arsenic through the standard metal addition protocol for soil, grass, leaf and invertebrate samples ranged from 82  $\pm 0.15$ ; 92  $\pm 0.12$ ; 84  $\pm 0.17$  and 78  $\pm 0.25$  respectively. This range can be adjudged acceptable for application in the analysis of environmental samples.

#### Level of arsenic in environmental samples

Results of the analyses of arsenic in a total of 96 samples of soil, grass, plant leaf and invertebrate across sampling sections (SS) and sampling periods (SPs) are presented in Tables 3 to 6. The concentration of arsenic in analysed samples at SS1 and across the sampling period during the first period of sampling that is month of July is as presented in Table 3. Arsenic concentration varied from  $0.67 \pm 0.05$  (SP5) to  $5.50 \pm 0.27$  mg kg<sup>-1</sup> (SP1);  $0.11 \pm 0.03$  mg kg<sup>-1</sup> (SP2) to  $0.48 \pm 0.13$  mg kg<sup>-1</sup> (SP1);  $0.20 \pm 0.04$  mg kg<sup>-1</sup> (SP5) to  $1.36 \pm 0.24$  mg kg<sup>-1</sup> (SP1) and 0.28



Figure 1. Pictures of environmental samples: Stag Beetle (L), Common grass (M) and Dogwood Plant Leaf (R).

Sample	Spiked concentration (µg/ml)	% Recoveries
Soil	0.5	$82.0\ \pm 0.15$
Grass	0.5	$90.0\ \pm 0.12$
Leaf	0.5	$84.0\ \pm 0.17$
Invertebrate	0.5	$78.0\ \pm 0.25$

Table 2. Percentage recoveries  $(\pm SD)$  of arsenic (n = 3) from spiked samples.

**Table 3.** Total arsenic concentration (mg/kg, dry wt.), ( $\pm$  SD) in environmental samples collected in July 2015; n = 24.

Parameter	SP	Soil	Grass	Leaf	Insect
004	SP1	5.50 (0.27)	0.48 (0.13)	1.36 (0.24)	0.47 (0.02)
	SP2	3.35 (0.18)	0.11 (0.03)	0.37 (0.06)	1.56 (0.12)
	SP3	1.19 (0.07)	0.32 (0.16)	0.25 (0.05)	0.28 (0.05)
SS1	SP4	3.21 (0.14)	0.12 (0.05)	0.27 (0.04)	0.10 (0.03)
	SP5	0.67 (0.05)	0.62 (0.12)	0.20 (0.04)	0.79 (0.01)
	SP6	1.18 (0.04)	0.39 (0.18)	0.22 (0.03)	1.10 (0.03)

SS = Sampling section; SP = sampling period.

 $\pm$  0.05 (SP3) to 1.56  $\pm$  0.12 mg kg<sup>-1</sup> (SP2) respectively for soil, grass, leaf and invertebrate samples.

The outcome of arsenic analysis at SS2 during the second period (August) of sampling is as shown in Table 4. Concentration of arsenic varied from  $0.33 \pm 0.05$  (SP5) to  $1.06 \pm 0.15 \text{ mg kg}^{-1}$  (SP1) in soil samples;  $0.11 \pm 0.05$  (SP1) to  $0.57 \pm 0.08 \text{ mg kg}^{-1}$  (SP4) in grass samples;  $0.10 \pm 0.04$  (SP6) to  $0.42 \pm 0.13 \text{ mg kg}^{-1}$  (SP1) in plant leaf sample and  $0.12 \pm 0.02$  (SP2) to  $1.47 \pm 0.04 \text{ mg kg}^{-1}$  (SP5) in invertebrate samples.

The level of arsenic across the sampling periods at SS3 is as shown in Table 5. The concentration ranged from 0.15  $\pm$  0.04 (SP5) to 0.60  $\pm$  0.03 mg kg<sup>-1</sup> (SP1); 0.04  $\pm$  0.03 (SP2) to 0.21  $\pm$  0.05 mg kg<sup>-1</sup> (SP4); 0.04  $\pm$  0.02

(SP6) to 0.12  $\pm$  0.04 mg kg<sup>-1</sup> (SP1) and 0.13 0.06  $\pm$  0.03 (SP5) – 0.91  $\pm$  0.02 mg kg<sup>-1</sup> (SP2) for soil, grass, leaf and invertebrate samples respectively. The concentration of arsenic in analysed samples at SS4 during the fourth sampling period is as presented in Table 6. The concentrations of arsenic obtained varied from 0.20  $\pm$  0.06 (SP5) to 0.56 mg kg<sup>-1</sup> (0.05) for soil; 0.02  $\pm$  0.01 (SP2) to 0.10  $\pm$  0.03 mg kg<sup>-1</sup> (SP5) for grass; 0.02  $\pm$  0.04 (SP5) to 0.21  $\pm$  0.05 mg kg<sup>-1</sup> (SP1) for leaf samples and 0.18  $\pm$  0.05 mg kg<sup>-1</sup> (SP1) to 1.14  $\pm$  0.06 mg kg<sup>-1</sup> (SP6) in the invertebrate.

The overall mean concentration of arsenic across the sampling sections (SS) within the study area as well as the mean concentration of arsenic in samples from the

Parameter	SP	Soil	Grass	Leaf	Insect
SS2	SP1	1.06 (0.15)	0.11 (0.05)	0.42 (0.13)	0.17 (0.04)
	SP2	0.63 (0.04)	0.04 (0.01)	0.25 (0.05)	0.12 (0.02)
	SP3	0.64 (0.07)	0.13 (0.02)	0.13 (0.07)	0.68 (0.07)
332	SP4	0.34 (0.05)	0.57 (0.08)	0.23 (0.03)	0.47 (0.02)
	SP5	0.33 (0.05)	0.13 (0.03)	0.12 (0.02)	1.47 (0.04)
	SP6	0.76 (0.11)	0.09 (0.03)	0.10 (0.04)	0.93 (0.06)

**Table 4.** Total arsenic concentration (mg/kg, dry wt.), ( $\pm$  SD) in environmental samples collected in August 2015; n = 24.

SS = Sampling section; SP = sampling period.

**Table 5.** Total arsenic concentration (mg/kg, dry wt.), (± SD) in environmental samples collected in September 2015; n = 24.

Parameter	SP	Soil	Grass	Leaf	Insect
000	SP1	0.60 (0.03)	0.08 (0.02)	0.12 (0.04)	0.24 (0.03)
	SP2	0.35 (0.05)	0.04 (0.03)	0.07 (0.02)	0.91 (0.02)
	SP3	0.55 (0.03)	0.09 (0.01)	0.08 (0.03)	0.22 (0.05)
SS3	SP4	0.26 (0.13)	0.21 (0.05)	0.06 (0.02)	0.27 (0.04)
	SP5	0.15 (0.04)	0.09 (0.02)	0.05 (0.03)	0.13 (0.06)
	SP6	0.35 (0.06)	0.10 (0.03)	0.04 (0.02)	0.45 (0.03)

SS = Sampling section; SP = sampling period.

Table 6. Total arsenic concentration (mg/kg, dry wt.), ( $\pm$  SD) in environmental samples collected in October 2015; n = 24.

Parameter	SP	Soil	Grass	Leaf	Insect
	SP1	0.26 (0.08)	0.04 (0.02)	0.21 (0.05)	0.18 (0.05)
004	SP2	0.56 (0.05)	0.02 (0.01)	0.05 (0.02)	0.53 (0.03)
	SP3	0.36 (0.04)	0.05 (0.03)	0.04 (0.01)	0.38 (0.10)
SS4	SP4	0.32 (0.03)	0.09 (0.02)	0.03 (0.02)	0.24 (0.04)
	SP5	0.20 (0.06)	0.10 (0.03)	0.02 (0.04)	0.80 (0.05)
	SP6	0.37 (0.05)	0.05 (0.01)	0.04 (0.03)	1.14 (0.06)

SS = Sampling section; SP = sampling period.

control site (CS) are presented in Table 7. The overall mean level of arsenic across SS1-SS4 ranged from 0.35  $\pm$  0.12 to 2.52  $\pm$  1.85 mg kg<sup>-1</sup>; 0.01  $\pm$  0.03 to 0.34  $\pm$  0.20 mg kg<sup>-1</sup>; 0.07  $\pm$  0.03 to 0.46  $\pm$  0.45 mg kg<sup>-1</sup> and 0.37  $\pm$  0.29 mg kg<sup>-1</sup> to 0.72  $\pm$  0.54 mg kg<sup>-1</sup> for soil, grass, plant leaf and invertebrate samples respectively.

#### **Statistical applications**

The extent of contamination of the study area by arsenic was evaluated and presented in Table 8. The CF at SS1-SS4 ranged from 6.5 to 34, 1.6 to 16, 1 to 10 and 0.9 to

13.8 respectively across soil, grass plant leaf and invertebrate samples. In terms of correlation coefficient, r values of 0.96 (soil/grass), 0.97 (soil/leaf) and 0.99 (leaf/grass) were obtained. However, result of the analysis of variance at p < 0.05 generated a p value of 0.11.

#### DISCUSSION

#### Method quality assurance

The outcome of the quality assurance process revealed

Parameter	Samples							
Farameter	SS	Soil	Grass	Leaf	Insect			
Sampling sections	SS1	2.52 (1.85)	0.34 (0.20)	0.46 (0.45)	0.72 (0.54)			
	SS2	0.63 (0.27)	0.18 (0.09)	0.21 (0.12)	0.64 (0.51)			
	SS3	0.38 (0.17)	0.10 (0.06)	0.07 (0.03)	0.37 (0.29)			
	SS4	0.35 (0.12)	0.06 (0.03)	0.07 (0.07)	0.55 (0.37)			
Control site (CS)		0.39 (0.07)	0.01 (0.03)	0.02 (0.03)	0.04 (0.02)			
Threshold values		7.4	0.1-0.9	2.1-9.5	100 - 1,000			

Table 7. Overall mean concentration (mg/kg, dry wt.), ( $\pm$  SD) of arsenic across SS and threshold values in environmental samples.

SS = Sampling section.

Table 8. Metal contamination factor (CF) in samples and contamination criteria.

	S	amples	Classification d	lagrap of contomination			
SS	Soil	Grass	Leaf	Insect	Classification degree of contamination		
SS1	6.5	34	23	18	CF < 1	Low	
SS2	1.6	18	11	16	1 ≤CF < 3	Moderate	
SS3	1	10	3.5	9.3	3≤CF < 6	Considerate	
SS4	0.9	6	3.5	13.8	CF > 6	High	

SS = Sampling section.

applicability of the analytical process based on the relatively high percentage recoveries obtained. Generally, in quality assurance evaluation processes, the amount of recovered analyte(s) either through standard addition or use of Standard Reference Material (SRM) is utilised as an indication of the efficiency and applicable of the analytical method for the intended experimental process. In a related study, metal recovery range of 75 -125 percentage was obtained and considered acceptable (Leshe and Tessema, 2014). This recovery is similar to the range of 78-92 percentage obtained in this study. In addition, recovery range of 80 to 120 was also considered acceptable for metals and metalloids (Simpson and Batley, 2016).

#### Level of arsenic in environmental samples

Arsenic is a ubiquitous metalloid of significant environment importance in view of its' toxicity and health implications. One of the exposure pathways of arsenic into the food chain has been through ingestion of food that emanates from arsenic contaminated soil or soil irrigated with arsenic-contaminated water (Hong et al., 2014). In this study, arsenic was detected in all the analysed environmental samples across the sampling sections which perhaps support the assertion of ubiquity of the metalloid. Highest concentration of 5.50 mg kg<sup>-1</sup> of arsenic in soil samples was obtained at SS1 when compared to 0.62 mg kg<sup>-1</sup>, 1.36 mg kg<sup>-1</sup> and 1.56 mg kg<sup>-1</sup> obtained in grass, leaf and insect samples respectively.

This result possibly supports the general assertion of soil as a sink for heavy metals (Han et al., 2017). In terms of the sampling period, highest level of arsenic was also obtained during SP1, SP5 and SP2 in soil and leave, grass and insect respectively. This might be due to higher level of anthropogenic activities in this section of the study area. Many petrochemical operators are located within this section of the study area. There was a decreasing trend in the distribution of arsenic across the sampling periods in soil and leaf samples from SP1-SP3 followed thereafter by irregular pattern. The decreasing trend could be due to lesser contribution of arsenic into the environment by impactors. There was no specific trend in the level of the metal in grass and invertebrate samples.

At SS2 however, the highest level of 1.47 mg kg<sup>-1</sup> arsenic was obtained in insect during SP5 while the lowest value of 0.04 mg kg<sup>-1</sup> in grass was obtained during SP2. This lower level in grass might be due to lower amount of arsenic in the sampled grass. The high level of arsenic obtained in insect might have occurred through bio-augmentation and bio-accumulation processes over a long period of time. Invertebrates especially the beetle

insect are known to feed on particles of leaves and organic matter that may also contain trace metals (Chiarelli and Roccheri, 2014). Hence, ingestion of these materials over a long period of time might account for the high level obtained in this study. Generally, there was no observable pattern of metallic trend across the sampling periods as well as analysed samples within this sampling section of the study area.

At sampling section 3 (SS3), highest level of 0.91 mg kg<sup>-1</sup> arsenic was obtained in the invertebrate sample during SP2 while the lowest (0.04 mg kg<sup>-1</sup>) was obtain in grass and leaf samples during SP2 and SP6 respectively. Concentration of arsenic at this section also did not show any peculiar pattern or trend either across the SP or in the analysed environmental samples. In the leaf samples however, the value decreased across the sampling period (SP1-SP6) except during SP3 where it rose slightly before continuing the decreasing trend. The non-peculiarity of concentration pattern could be due to relative difference in the level of uptake of arsenic by analysed samples.

At sampling section 4 (SS4), highest metallic value of 1.14 mg kg<sup>-1</sup> of arsenic was obtained in insect during SP6 while the lowest concentration of 0.02 mg kg<sup>-1</sup> was obtained in grass and leaf samples during SP2 and SP5 respectively. Also, at this section, there was no defined distribution pattern of arsenic in the analysed samples except in leaf where decreasing trend was observed from SP1-SP5 with a slight increase during SP6. Similar study with irregular metallic distribution and trends in analysed samples has been reported (Raulinaitis et al., 2012).

In environmental pollution studies, control sites are expected to be devoid of or are very low in anthropogenic influence relative to the area under investigation. The overall mean range of 0.35 to 2.52-mg kg<sup>-1</sup> of arsenic obtained in soil samples from this study was lower than the prescribed limit of 7.4 mg kg<sup>-1</sup> of arsenic in uncontaminated soil (Dudas, 1984). In addition, the highest value of 5.50 mg kg<sup>-1</sup> (SS1, SP1) arsenic obtained in this study was also lower than the prescribed limit. However, continual anthropogenic contributions on soil over a long period of time may exacerbate the pollution situation. Some of these contributions include atmospheric deposition of metal-carrying particulate matters (Qian and Wan, 2013), deposition of metal containing wastes (Wuana et al., 2011) and use of metal containing sludges as soil enrichment during agricultural activities (Karczewska et al., 2013).

With respect to the overall mean of arsenic in grass samples, the range obtained in this study (0.01 to 0.34 mg kg<sup>-1</sup>) was also found to be lower than the prescribed range of 0.1 to 0.9 mg kg<sup>-1</sup> (dry wt.) in grass in non-treated area (NAS, 1977). At this range, the plant may not experience serious toxicity issues. Although, these values did not represent the bioavailable fraction of the metal for toxicological inferences on plant, tolerance level

of 2 mg kg<sup>-1</sup> of arsenic has been reported to disrupt enzyme function and impair phosphate flow in the plant system (Kabata-Pendias and Mukherjee, 2007). In plant leaf, the overall mean concentration range of 0.07 to 0.46 mg kg<sup>-1</sup> arsenic obtained in the study was also lower than the prescribed range of 2.1 to 9.5 mg kg<sup>-1</sup> of total arsenic level in plant leaf of White spruce, Picea alba (Jenkins, 1980). At this lower concentration range, serious impact on proper leaf functioning was not expected. Hood (1985), prescribed a range of 100 to  $1,000 \text{ mg kg}^{-1}$ as fatal arsenic concentration in including beetles. pestiferous species Overall concentration range of 0.37 to 0.72 mg kg<sup>-1</sup> arsenic in invertebrate samples obtained in this study was much lower than the prescribed range. Hence, no serious impact of the metal on the insects would be expected.

# Contamination factor (CF), metal inter-sample correlation and analysis of variance

Based on the contamination assessment criteria, CF range of 6.5 to 34 obtained at SS1 reflects a section that can be regarded as highly contaminated. At SS2 with CF range of 1.6 to 16, high contamination was also recorded soil except for samples having moderate in contamination. At SS3, the CF values ranged from 1 to 10. At this section, the grass and insect samples were highly contaminated while the leave and soil recorded considerable and moderate contamination respectively. At SS4, low and considerable contamination was observed in soil and leave samples respectively while the grass and insect samples were highly contaminated. The contamination trend in the analysed samples followed the pattern grass > leave > insect > soil. From this pattern, higher CF in grass relative to others may be related to wider contact with soil and exposure to possible atmospheric deposition of arsenic laden particulate matter when compared to other samples. The pattern could also have been influenced by the level of arsenic obtained in analysed samples from the CS. Metal accumulation by the grass from soil is highly possible.

Generally, all sampling sections (SS1 to SS4) of the study area can be adjudged to be contaminated by arsenic in view of the high CF values obtained across the samples. Similar high CF in environmental samples and sites have been reported (Rahman et al., 2012). Although, all the sampling sections might be regarded as contaminated by arsenic based on the CF values, there are differences in contamination level across the sections. SS1 reflected the most contaminated site which could be as a result of higher level of anthropogenic activities such as petrochemical occupations as earlier mentioned. Differences in contamination level across sampling sections might also be due to chemical phenomenon such as volatilisation. The chemical

	Soil	Grass	Leaf	Insect
	1	0.956*	0.970*	0.722
Soil		0.044	0.030	0.278
	4	4	4	4
	0.956*	1	0.991**	0.759
Grass	0.044		0.009	0.241
	4	4	4	4
	0.970*	0.991**	1	0.823
Leave	0.030	0.009		0.177
	4	4	4	4
Insect	0.722	0.759	0.823	1
	0.278	0.241	0.177	
Ν	4	4	4	4

Table 9. Correlation of arsenic between environment	al samples	5.
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\*Correlation is significant at the 0.05 level (2-tailed); \*\*Correlation is significant at the 0.01 level (2-tailed).

properties of soil as well as soil bacterial activities in methylation and volatilisation of arsenic in soil may play a role in the level of arsenic obtained at respective sampling sections (Chirenje et al., 2002).

Detection of arsenic in analysed environmental samples has serious implications on human and environmental health as well as across the food chain. Arsenic has been classified as environmental carcinogen (Duker et al., 2005) and enter the food chain through edible plants that might have accumulated high level of arsenic which eventually poses health problems to human. Ruminants are known to feed on road side grass and plant leaves (Roggeman et al., 2013). Consumption of arsenic laden grasses by livestock such as cows and goats may have health implications indirectly on human through the consumption of milk and meat of these animals (Chung et al., 2014). In the same manner, consumption of insects with high level of arsenic by birds and other terrestrial lower animals such as chickens will seriously affect the trophic balance of the ecosystem. Hence, possible transfer of arsenic across the food chain may occur through the sequence of soil to grass, to ruminant and to human. Long-term exposure to arsenic may result in skin lesions, lung and kidney cancer (Mondal et al., 2006).

High correlation (r > 0.9) was obtained between soil and grass and soil and leaf with moderate correlation between soil and insect at p < 0.05. Strong positive correlation (r > 0.99) was recorded between the leaf and grass at p < 0.01 while moderate correlation was obtained between the insect, grass and leaf (Table 9). These significant correlations indicate common association between arsenic and analysed samples. Result of the analysis of variance of possible association between the metal and the analysed samples revealed a p value of 0.11, hence there was no statistical significant difference between the focus (arsenic) and the analysed samples.

#### CONFLICT OF INTERESTS

The authors have not declared any conflict of interest

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