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Analysis of Heavy Metals in Tea Soil, Tea Leaves and Seagim Dam Water across the Seasons in Kakara District of Taraba State, Nigeria

Dr. Charles M

Associate Professor, Department of Chemistry,
Modibbo Adama University of Technology, Nigeria

Dimas BJ

Lecturer, Department of Science Education,
Taraba State University, Nigeria

Uwaisu M M

Researcher, Department of Chemistry,
Modibbo Adama University of Technology, Nigeria

Abstract:

The levels of heavy metals were assessed in tea soil, tea leaves and Seagim dam water samples collected from Kakara tea estate, Taraba State. The concentration of heavy metals was determined by the use of AAS. The concentration of Ca, Cd, Co, Cu, Fe, K, Mn, Ni, Pb, and Zn in all the soil samples collected across the season ranged from [6.102-11.00],[0.023-0.873],[0.029-0.058],[0.241-0.410],[0.900-1.958],[1.366-20.80],[0.263-0.496],[0.023-0.496],[0.018-0.068]and[0.110-0.275] mg/kg, the concentration of Ca, Cd, Co, Cu, Fe, K, Mn, Ni, Pb, and Zn in all the leaves samples collected across the season ranged from [0.982-3.566],[0.014-0.029],[0.010-0.071],[0.059-0.204],[0.066-0.755],[19.98-38.66],[0.106-0.369],[0.008-0.031], [0.006-0.099] and [0.080-0.199] mg/kg, the concentration of Ca, Cd, Co, Cu, Fe, K, Mn, Ni, Pb, and Zn in all the water samples collected across the season ranged from [9.029-13.74],[0.014-0.151],[0.063-0.114],[0.090-0.204],[1.604-2.069],[3.013-6.711],[0.262-0.429],[0.058-0.112],[0.043-0.113] and [0.128-0.225] mg/dm³ respectively. The result analysis showed that the mean concentration of heavy metal was higher in wet season than in dry season and among the metals analyzed K, Ca and Fe were the most abundance in all the samples. The concentrations of heavy were found to be lower than the maximum permissible level.

Keywords: Heavy metal, Tea leaves, tea soil, Seagim dam, season

1. Introduction

Recently, the toxicity and effect of trace heavy metals on human health and the environment has attracted considerable attention and concern. Among the heavy metals lead (Pb), cadmium (Cd), Arsenic (As), and mercury (Hg) are especially toxic and are harmful to humans even at low concentration. Heavy metals can be introduced into the environment and consequently, living organisms through air, water, food or soil, but the degree of their concentrations depends on the type of heavy metals and the activities that occur in a particular area (Zaharaddeen et al., 2015). Several attempts have been made by many researchers to assess the quality of tea using chemical analysis. Metallic constituents of tea leaves are usually different according to the type of tea (green or black) and geographical sources (Fernandez-caceres et al., 2001; Sahito et al. 2005).

Tea is one of the most widely consumed beverages across the globe and is produced from the dried leaves of *Camellia sinensis*. Drinking tea might help reduce serum cholesterol, provide antiaging activities, and decrease the risks of both cardiovascular disease and cancer. However, heavy metal contaminants might accumulate during tea growth, transportation, packaging, and processing. Heavy metals are harmful to human health. These risks are recognized by the International Agency for Research on Cancer and the National Toxicology Program (Zhong et al., 2016).

To increase the optimum quantity of tea, tea planters applied sufficient amount of fertilizers on soil, and also used fungicides to control the herbs in the tea soil. But fertilizers and fungicides itself contain sufficient amount of heavy metals. Soil is a good medium, acting as a sink for natural and anthropogenic pollutants (Chakraborty et al., 2004). Humans are responsible for introducing heavy metals into the environment. Anthropogenic contamination with heavy metals is a worldwide problem that causes massive water and soil pollution (Caussy et al., 2003). Heavy metals that have contaminated industrial areas, roadside soils, riverbanks and urban areas are among the most serious environmental hazards (Magrisso et al., 2009).

Heavy metal contamination in tea leaves has been documented. Lead concentrations in Chinese tea were found in a study with 32% of samples exceeding the national maximum permissible concentration (MPC) of 2.0 mg/kg. An increasing trend in lead concentration on tea leaves was documented from 1989 to 2000. Proximity to highway and surface dust contamination was found to cause these elevated concentrations, as well as uptake of lead in soil by the roots of the tea plant. 44 up to 83% of teas have lead levels considered unsafe for consumption during pregnancy and lactation, as well as excessive levels of manganese and aluminum (Naik, 2015),

Sahito *et al.* reported the contents of 15 essential trace and toxic elements in some green tea samples and their infusion. Karimi *et al.* worked on the concentrations and health risks of metals in tea samples that were marketed in Iran. Zaharaddeen *et al.* (2015), determine the elemental composition of different brands of tea in Zaria markets in Nigeria and compare the result with the standard and provided both researchers and consumers with information on the mineral contents of different tea brands.

This paper aims to assess the levels of heavy metals in tea soil, tea leaves and Seagim dam water samples collected from Kakara tea estate, Taraba State. The modern agricultural practices which employed the use of organic fertilizers and pesticides residues (both herbicides and insecticides) generally exposes to the environment to heavy metals. Several attempts have been made by many researchers to assess tea quality by chemical analysis with reference to pigmentation and flavorings characteristics in other places. However, little work has been done to comprehensively screen the extent of bioaccumulation of heavy metals in tea leaves, tea soil and water in Kakara village of Sardauna Local Government Area of Taraba State, Nigeria.

2. Materials and Methods

2.1. Study Area

The study was carried out in the Mambilla plateau in Sardauna Local Government Area of Taraba State (Figure 1) which has an altitude of 1,800m above sea level' the plateau forms part of the chain of the Adamawa and Mandara mountains is located between altitude 5o30o to 7o18oN and longitudes 10o18o to 11o to 37oE has a land mass of 8,3865km. Kakara Dam or tea factory dam is a shallow fresh water dam situated on the floor of North east left valley in Nigeria The Kakara tea factory dam has a capacity of 10 million cubic meters and it is used to irrigate the tea farms, supplies untreated piped water to the local community, factory and electricity is also generated from the dam. The plateau has a near temperate climate with rainfall almost throughout the year Agbongiarnouyi *et al.*, (2010).

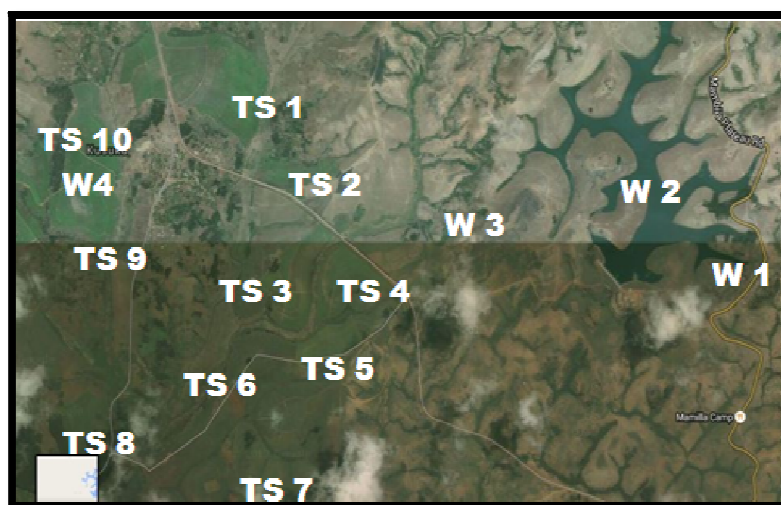


Figure 1: Map of a Study Area

Source: Google, Wikipedia

Key Table

W 1: Mouth of the Dam

W 2: Mid of the Dam

W 3: Outlet of the Dam

W 4: Tap water (Irrigation Farm)

T S. 1-10 are Tea Farms

2.2. Materials /Instruments

Digger, hammer and chisel, field bags for collection of samples, pestle and mortar, Bunsen burner, pH meter, tune cupboard, beakers, spatula, volumetric flask pipette, fitter paper, crucible, micrometer, mesh size sieve, stirring rod, multi water quality checker U-50 series, cat ion exchange AAS (Atomic absorption spectroscopy) buck scientific model 210 and oven etc.

2.3. Reagents/ Chemicals

Hydrochloric acid (HCl) perchloric acid (HClO_4) de-ionies water, aqua regia (3:1) Hydrochloric acid; Nitric acid, sodium Hydroxide (NaOH) calcium Hydroxide ($\text{Ca}(\text{OH})_2$, EDTA Nitric acid, sulphuric acid (H_2SO_4), Hydrogen peroxide (H_2O_2).

2.4. Sample Collection

2.4.1. Water Sampling

Water sample was collected monthly from September, 2015 to February, 2016. Sampling was conducted at four pre-defined stations. One site in the plume area, one site was at the central part of the dam (mid dam), one site was at the outlet of the dam while other one site was at the consumption point (Tap) or irrigated farm site. Water temperature, PH, conductivity and turbidity was measured in situ during each sampling occasion using PH Wagtech meter, turbidity meter (Ndugu, 2013).

2.4.2. Sample Preparation

The methods describe by Joseph *et al.*, 2009 was adopted for the sampling and treatment of dam water, tea leaves and tea soil for elemental analysis of heavy metals using AAS.

2.4.3. Water Sampling and Treatment for AAS

One hundred (100ml) of highland Tea Dam water was collected and transferred into clean poly ethylene bottles. The water sample was filtered through filter paper and was kept at 4°C until analysis. 100ml of water was transferred into a large beaker (750ml capacity), and 15ml of conc. HNO_3 was added to the mixture, and was allowed to evaporate on a steam bath to approximately 25ml. Then transferred to 50ml acid washed volumetric flask and brought up to the volume with deionized water.

2.5. Treatment/Digestion of Green Tea Leave for AAS

The samples that were collected from the various farms were washed with tap water, followed by distilled water. Each sample was ground into a fine powder, sieve and were finally store in a capped plastic jar with an appropriate labeled. 1g of air-dry leave was accurately weighed into 150ml beaker, 20ml conc. HNO_3 was carefully added and allowed to stand for 1 hour; then 15ml of conc. HClO_4 was carefully added and the mixture was allowed to stand on hot plate at about 200-225°C till the mixture turns to yellow or white. Digest was dissolved in 0.1M HCl and then filtered, and transferred into 150ml volumetric flask and made up to the mark. The sample solution was then filtered with a filter paper into polyethylene bottle and was taken for elemental analysis of heavy metals by using AAS.

2.6. Digestion of Soil Sample for AAS

One (1)g of air-dried soil, was accurately weighed into 150ml beaker, 20ml conc. HNO_3 was carefully added and allowed to stand for 1 hour; then 15ml of conc. HClO_4 was carefully added and the mixture was allowed to stand on hot plate at about 200-225°C till the mixture turns to yellow or white. Digest was dissolved in 0.1M HCl and then filtered, and transferred into 150ml volumetric flask and made up to the mark. The sample solution was then filtered with a filter paper into polyethylene bottle and was taken for elemental analysis of heavy metals and pesticide residue by using AAS and flame photometric analysis.

2.7. Elemental Analysis

The heavy metals (Cd, Fe, Pb, Ni, Zn, Mn, Co, Cu, Ca, and K) were analyzed on each final solution by using buck scientific model 210 Atomic Absorption spectroscopy (AAS) as described Joseph *et al.*, 2009.

2.8. Data Analysis

The Result that was obtained were analyzed statistically by using Excel, students' test and analysis of variations (ANOVA) for level of significance of variation between samples that were used to analyze the data.

2. Results and Discussion

2.1. Results

The mean Seasonal variation of heavy metals, some parameters and concentration across the locations are as presented below. Figure 2 shows the summary results of various metals concentration in tea soil samples collected during wet and dry Season from different tea estate. For wet season the concentration of Ca ranged from 6.756 to 11.000 mg/kg, Cd ranged from 0.035 to 0.087 mg/kg, the concentration of Co ranged from 0.042 to 0.058 mg/kg. Cu concentration ranged from 0.288 to 0.410 mg/kg, the concentration of Fe ranged from 1.343 to 1.958 mg/kg, for K 2.470 to 20.80 mg/kg, for Mn 0.338 to 0.496 mg/kg, for Ni 0.047 to 0.056 mg/kg, for Pb ranged from 0.026 to 0.068 mg/kg and for Zn 0.149 to 0.259 mg/kg. While for dry season the concentration of Ca ranged from 6.102 to 9.403 mg/kg, Cd ranged from 0.023 to 0.049 mg/kg, the concentration of Co ranged from 0.029 to 0.046 mg/kg. Cu concentration ranged from 0.241 to 0.388 mg/kg, the concentration of Fe ranged from 0.882 to 1.802 mg/kg, for K 1.366 to 7.212 mg/kg, for Mn 0.263 to 0.455 mg/kg, for Ni 0.042 to 0.042 mg/kg, for Pb ranged from 0.018 to 0.029 mg/kg and for Zn 0.110 to 0.225 mg/kg.

Figure 3 shows the result of heavy metals in tea leaves samples analyzed during wet and dry seasons at different locations. For wet season the concentration of Ca, Cd, Co, Cu, Fe, K, Mn, Ni, Pb, and Zn ranges from 2.311 to 3.566 mg/kg, 0.017 to 0.029 mg/kg, 0.022 to 0.071 mg/kg, 0.087 to 0.204 mg/kg, 0.139 to 0.755 mg/kg, 16.02 to 38.666 mg/kg, 0.127 to 0.369 mg/kg, 0.017 to 0.031 mg/kg, 0.006 to 0.099 mg/kg and 0.088 to 0.199 mg/kg for different farms respectively. And for dry season the concentrations of Ca, Cd, Co, Cu, Fe, K, Mn, Ni, Pb, and Zn ranges from 0.982 to 2.680 mg/kg, 0.014 to 0.021 mg/kg, 0.010 to 0.022mg/kg, 0.059 to 0.132 mg/kg, 0.066 to 0.409 mg/kg, 19.989 to 23.066 mg/kg, 0.106 to 0.360 mg/kg, 0.008 to 0.012 mg/kg, 0.008 to 0.022 mg/kg and 0.080 to 0.170 mg/kg for different farms respectively.

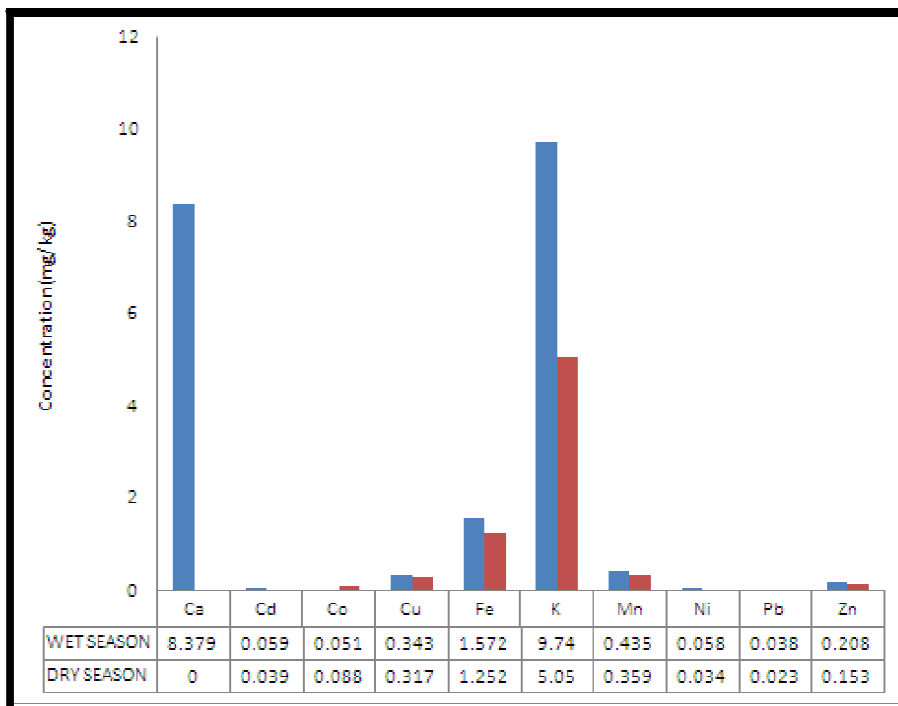


Figure 2: Seasonal Variation of Heavy Metals in Soil Samples Collected from Tea Farm, of Kakara Estateq

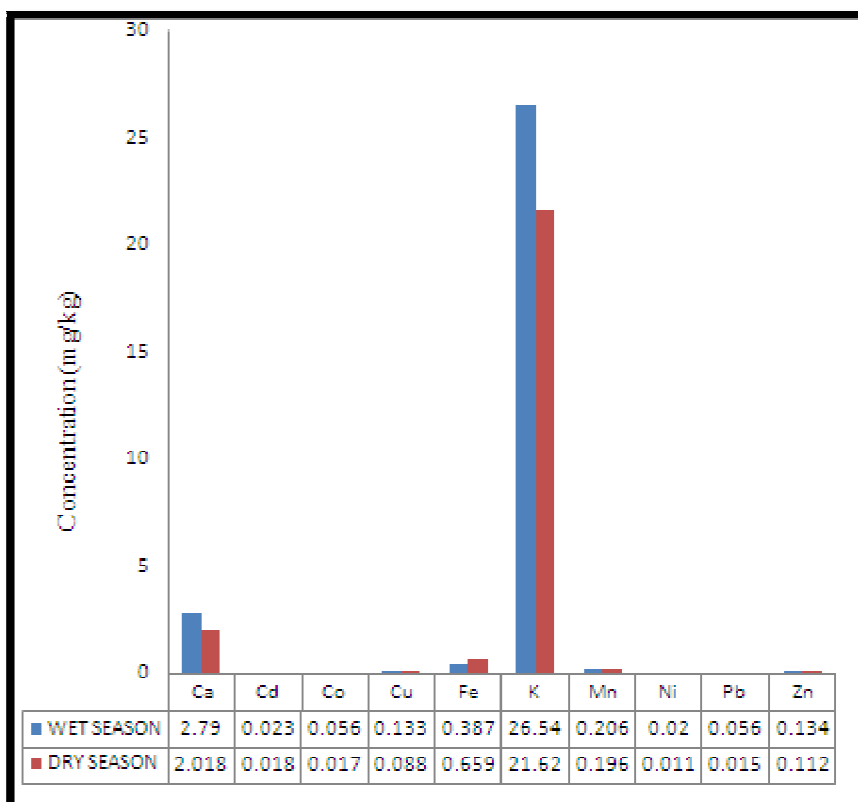


Figure 3: Seasonal Variation of Heavy Metals in Tea Leaves Samples Collected From Kakara Estates

Figure 4: presents summary results of heavy metals concentration in Seagim dam water analyzed during the seasons. For wet season the concentration of Ca, Cd, Co, Cu, Fe, K, Mn, Ni, Pb and Zn ranges from 9.730 to 13.743 mg/dm³, 0.061 to 0.151mg/dm³, 0.092 to 0.114mg/dm³, 0.177 to 0.204mg/dm³, 1.790 to 2.069mg/dm³, 3.036 to 6.904mg/dm³, 0.373 to 0.429mg/dm³, 0.070 to 0.112mg/dm³, 0.021 to 0.113mg/dm³ and 0.125 to 0.225mg/dm³.

For dry season the concentration of Ca, Cd, Co, Cu, Fe, K, Mn, Ni, Pb and Zn ranges from 9.029 to 12.580mg/dm³, 0.014 to 0.148mg/dm³, 0.063 to 0.102mg/dm³, 0.090 to 0.211mg/dm³, 1.604 to 2.048mg/dm³, 3.012 to 6.794mg/dm³, 0.262 to 0.401mg/dm³, 0.070 to 0.112mg/dm³, 0.043 to 0.102mg/dm³, and 0.128 to 0.214mg/dm³.

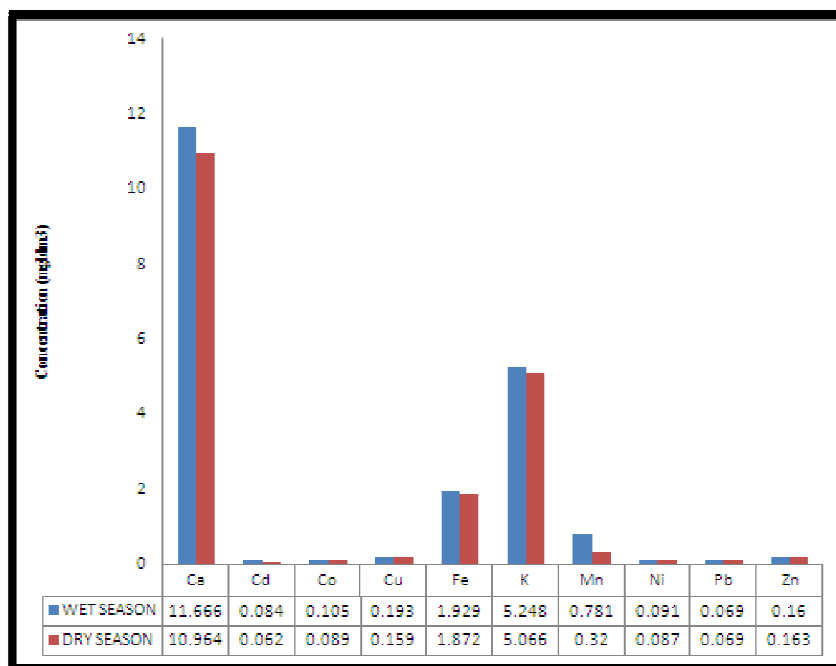


Figure 4: Seasonal Variation of Heavy Metals in Water Samples Collected from Seagim Dam Water Kakara Estate

From the result obtained the concentration of Ca is higher in water with a mean value of 11.666 ± 1.643mg/dm³, and then followed by Tea soil with Mean of 8.379 ± 1.904mg/kg and in tea leaves with a mean of 2.790 ± 0.550 mg/kg, during wet season. Also, in dry season from the result obtained Ca in water has a mean value of 10.964±1.51 mg/dm³ and less in tea leaves with a mean of 2.018±0.050 mg/kg. These concentrations were lower as reported by Kazi *et al.*, (1999) in tea leaves.

Similarly, for Cd the highest concentration was recorded in water with a mean 0.084±0.045mg/dm³, 0.059±0.023mg/kg for tea soil and 0.018 ± 1.276 mg/kg was recorded in tea leaves during wet season, also in dry season, the value recorded for seagim dam water has a mean 0.62±0.059 mg/dm³, 0.039 ± 1.012 mg/kg for tea soil and 0.023 ± 2.105 mg/kg for tea leaves. It is similar to the findings of Bansal, (2004) where concentration of these heavy metals is lower in dry season than that of wet season. According to the MOA standard (NY 659-2003), the Cd limit in tea is 1 mg/kg. The concentration of Cd in all the samples satisfied this criterion.

The level of Co in the samples were shown, from the result obtained Co has the highest concentration in wet season with means concentration of 0.105±2.944 mg/dm³ in water, 0.05± 1.487 mg/kg in tea soil and 0.017 ± 2.025 mg/kg in tea leaves. Similar for dry season Co recorded 0.089±0.018 mg/dm³ for water, 0.088±0.059 mg/kg in tea soil and 0.017±0.025 mg/kg tea leaves. The result obtained is lower than Xing and Chen, (2004) finding and also lower than the maximum permissible unit of heavy metals concentration in soil (ECDG, 2010). According to the WHO, the maximum permissible limit of cobalt is 1.5 mg kg⁻¹. The concentration of cobalt in all the tea samples in our study was ranged from 0.005 to 0.032 mg kg⁻¹. By comparing results from our study with the maximum permissible limit in plants, cobalt was in values which correspond to the literature data.

The result show that the concentration of Cu was highest in wet season with a mean concentration of 0.193± 0.00mg/dm³ in water, 0.343±0.051mg/kg in tea soil, 0.133 ± 0.001mg/kg leaves. In dry season the mean concentration of sample is 0.159 ± 0.52 mg/dm³ in water, 0.0317±1.061 mg/kg in tea soil and 0.088 ± 0.031 mg/kg in tea leaves. The result obtained showed lower value than those values reported by Saud and Al-Qud, (2003). Although Cu is an essential element for human health, excessive intake can impair organs and systems in the human body; hence the maximal content of Cu in tea is limited in some regulations. According to the Industrial Standard (NY/T 288-2012) of China's Ministry of Agriculture (MOA), the Cu content is limited to 30 mg/kg. The determined levels of Cu in this work were all below the limits.

The result obtained show that the mean concentration of Fe in the samples during a wet season 1.929 ± 0.123mg/dm³ for water, 1.253 ± 0.445mg/kg in Tea Soil and 0.387 ± 0.279mg/kg in Tea leaves. However, in dry season the mean concentration of Fe is 1.872 ± 0.207mg/dm³ in water, 1.252 ± 0.445mg/kg in Tea soil and 0.659 ± 0.113mg/kg in tea leaves. From the result obtained in comparing the value between the wet and dry season, the heavy metals are higher

in wet season with exception of tea leaves during dry season which is higher than that of wet season and result obtained in this research is lower than reported by (Ansari et al., 2007).

Potassium which is the most common from these finding, K has mean concentration of $5.248 \pm 1.879\text{mg/dm}^3$ in water, $9.74 \pm 2.021\text{mg/kg}$ in tea soil and $26.54 \pm 2.35\text{mg/kg}$ in tea leaves during wet season, while the mean concentration of K in dry season are $5.066 \pm 1.558\text{mg/dm}^3$ in water, $5.050 \pm 2.560\text{mg/kg}$ in tea soil and $21.623 \pm 1.82\text{mg/kg}$ in tea leaves. From the result obtained, comparing the value obtained between the wet and dry season, the heavy metals concentration is higher in wet season than in dry season.

The result obtained from this analysis show that Mn has a mean concentration of $0.781 \pm 0.439\text{mg/dm}^3$ in water, $0.435 \pm 0.022\text{mg/kg}$ in tea soil and $0.206 \pm 0.113\text{mg/kg}$ in tea leaves during wet season. Also, the mean concentration of Mn is $0.320 \pm 0.059\text{mg/dm}^3$ in water, $0.359 \pm 0.086\text{mg/kg}$ in tea soil and $0.196 \pm 0.112\text{mg/kg}$ in tea leaves during dry season. Comparing the values of Mn in wet and dry season from this research it was found that the concentration is higher in wet season and also when compared with (Ansari et al., 2007), it is lower. Manganese is an essential metal nutrient for humans and other living organisms. Moreover, manganese is normally present in many kinds of foods and deficiency of manganese in humans appears very rare. The population of tea drinkers may have an increased intake of manganese than the general population because tea is an important source of manganese. Side effects of manganese can be from both overexposure or deficiency of this element. It can cause neurotoxic side effects when exceeding the homeostatic range^{28, 29}. According to Shah et al., the permissible limit of manganese in medicinal plants is 200 mg

The average concentration of Ni was determined in this research as $0.091 \pm 0.023\text{mg/dm}^3$ in water, $0.058 \pm 0.004\text{mg/kg}$ in tea soil and $0.020 \pm 2.676\text{mg/kg}$ in tea leaves was analyzed during wet season, while in dry season the average concentration of Ni observed are $0.087 \pm 0.027\text{mg/dm}^3$ in water, $0.034 \pm 2.7\text{mg/kg}$ in tea soil and $0.011 \pm 1.916\text{mg/kg}$ in tea leaves. The comparison of Ni in two seasons, show that the average of Ni in wet season is higher than that of dry season. Results obtained were lower than those described in the literature (Nath, 2013). Ni is nutritionally essential as a trace element for several animal species, microorganisms, and plants excessive Ni intake by consuming tea with Ni contents above a certain threshold is harmful to humans (Zhong et al 2016). Ni obtained in this research area is lower than those reported by Nookabkaew et al (2006) of black tea from Thailand which has value ranged from 2.28 mg/kg to 9.19 mg/kg. However, there are not enough regulations for the Ni levels in tea.

The level of Pb in the samples were observed at a mean concentration of $0.069 \pm 0.039\text{mg/dm}^3$ in water, $0.039 \pm 0.02\text{mg/kg}$ in tea soil and $0.056 \pm 0.045\text{mg/kg}$ in tea leaves during wet season, while during dry season the mean concentration of Pb in water is $0.069 \pm 0.029\text{mg/dm}^3$, $0.023 \pm 1.59\text{mg/kg}$ in tea soil and $0.015 \pm 2.318\text{mg/kg}$ in tea leaves. The Pb contents obtained in this study were significantly lower than those determined by Al-Othman et al(2012), which ranged from 3.9 mg/kg to 8.7 mg/kg in the tea samples from Jazan and Jeddah, Saudi Arabia. According to the limit for the Pb content in tea leaves stipulated by the Chinese National Standards (GB 2762-2012), the Pb maximum residue limits is (5 mg/kg). According to the WHO, the permissible limit of lead in plants is 10 mg kg

The result of Zn is $0.160 \pm 0.045\text{mg/dm}^3$ in water, $0.208 \pm 0.04\text{mg/kg}$ in tea soil and $0.134 \pm 0.052\text{mg/kg}$ in tea leaves during wet season while in dry season the average concentration of Zn is $0.163 \pm 0.042\text{mg/dm}^3$ in water, $0.153 \pm 0.05\text{mg/kg}$ tea soil and $0.112 \pm 0.041\text{mg/kg}$ in tea leaves. Zinc is considered as a relatively non-toxic metal, but with extremely high-level of intakes can cause nausea, vomiting, fatigue, lethargy, and epigastric pain According to the WHO, the permissible limit of zinc in plants is 50 mg kg.

However, the average concentration of all metals was determined as mean value which appears to be quite high in water, then in tea soil and lower in tea leaves, with exception of K which is higher in the leaves and PH value of the tea estate soil was found to be acidic. Generally, increase in heavy metals levels were observed in the samples during the wet season. The study shows that the level of heavy metal analyzed seasonally are within a safe limits or permissible limit as specified by the U.S FDA for edible plants part similar to food, soil and water from other part of the world (ECDG, 2010).

Water	Tea leaf	Tea soil	
Water	1		
Tea leaf	0.550	1	
Tea soil	0.396	0.985	1

Table 1: Correlation Value of Heavy Metals in Relation between Water, Tea Leaf and Tea Soil

Table 1 shows the correlation result between the heavy metals in tea leaves and seagim water, it was found that the heavy metals in tea leaves and seagim water correlated negatively due to correlation value of 0.396. The correlation of heavy metals in tea soil and tea leaves correlated positively and significantly with tea soil and tea leaves due to the correlation value of 0.985 and finally, the correlation of heavy metals in seagim water and tea soil was positively and significantly with seagim water and tea soil due to the correlation value of 0.550. Therefore, there was positive correlation between seagim water and tea soil. The result shows the ability of these samples accumulating with heavy metals, the concentration of pollutants was maximum in wet season, thus indicating an increase in heavy metals levels in the rainy season over those in the dry season. This may be attributed to the possibility of the runoff from the surrounding farm containing metal salts being washed into the sample site, or application of fertilizes, animal wastes and herbicides to improve the tea productivity during rainy season.

3. Conclusion

Potassium, Calcium and Iron were most abundant metals analyzed in the samples. The result shows that the heavy metals concentration in almost all the samples analyzed were found to be higher in wet season than in dry season. The level of heavy metals in the sample analyzed was below the permissible limit but since there can be bioaccumulation of these elements resulting in toxicity, routine monitoring is advisable.

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