Quality assessment and potential health risk of heavy metals in leafy and non-leafy vegetables irrigated with groundwater and municipal-waste-dominated stream in the Western Region, Ghana

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ABSTRACT
Vegetables cultivated in soil irrigated with untreated groundwater and municipal-waste-dominated (MWD) stream can elevate the concentration of heavy metals (Cd, Fe, Zn, Hg, Cr, and Ni) in edible parts of the crop, affecting food safety and public health worldwide. This study assessed the quality, sources, and distribution of heavy metals in surface soils, MWD stream and groundwater, and edible tissues of leafy and non-leafy vegetables from a major urban farm in the Sekondi-Takoradi metropolis, Ghana. Human health risk due to exposure to the metals in frequently consumed vegetables were investigated. Indigenous leafy vegetables (Corchorus olitorious and Amaranthus spinosus), exotic leafy vegetables (Lactuca sativa, Brassica oleracea, and Brassica rapa), and non-leafy vegetables (Capsicum annum, Raphanus sativus, Daucus carota, and Allium cepa) were collected from the urban farm. The mean concentration of Cd, Hg, and Fe ranged from 0.008 – 0.027, 0.001 – 0.013, and 4.517 – 36.178 mg/kg fw in edible parts of non-leafy vegetables, respectively and 0.011 – 0.035, 0.002 – 0.011, and 3.617 – 13.695 mg/kg fw in exotic or indigenous leafy vegetables. The vegetables were less impacted with the metals if compared to similar vegetables produced from other urban farms, locally and in some countries in Africa, Asia, and Europe. Water resource on the farm were not suitable for vegetable crop irrigation since mean concentration of E. coli (200 cfu/mL), Hg (0.009 mg/L), and Cd (0.019 mg/L) in the MWD stream and 80 % of the groundwater sources exceeded the safe limits recommended by the Food and Agriculture Organization. Geo-accumulation index for each metal in soil was /C20, however, enrichment factor indicated a high anthropic enriched soil for Cr and Ni. Principal component analysis-multiple linear regression of the metals in soil identified mixed household waste/fertilizer, fertilizer, and crustal material as main sources for the heavy metal load in soil for which geogenic sources accounted for 74.3 %. Preferentially, Cd and Hg accumulated in Amaranthus spinosus, Daucus carota, and Corchorus olitorious. The estimated daily intake of each metal in the vegetables were below local and international daily dietary intake levels. At the 95th percentile concentration of each metal, target hazard quotient and the hazard index was <1 for adult male or female who consume the vegetables. Finally, appropriate agri-horticultural practices must be enforced to mitigate Cd, Ni, Cr, and Hg accumulation in the soil-vegetable system since the metals have profound adverse effect on human health.

1. Introduction

Vegetables form an important component of the human diet, because it is a good source of antioxidants, proteins, fiber, carbohydrates, vitamins, bioactive compounds, and minerals (Lente et al., 2012; Drechsel and Kariate, 2014). In Ghana, exotic leafy vegetables (e.g. Lactuca sativa, Brassica oleracea, and Brassica rapa), indigenous leafy vegetables (Corchorus olitorious and Amaranthus spinosus), and non-leafy vegetables (Capsicum frutescens, Raphanus sativus, Daucus carota, Capsicum annum, Allium cepa, and Solanum torvum) are mainly cultivated on farmlands.
located in urban and peri-urban communities (Drechsel and Keriate, 2014; Amoah et al., 2016). Moreover, these vegetable varieties are also cultivated in some countries in Asia (Mahmood and Malik, 2014), Europe (Alvarenga et al., 2014), Africa (Weldegebriel et al., 2012), and South America (Mauad et al., 2016). The Food and Agriculture Organization (FAO) (2016) estimated that 800 million people practice urban farming worldwide. Despite the socio-economic benefits of urban vegetable farming, Drechsel and Keriate (2014) revealed that poor agronomic practices can cause heavy metals, and other contaminants (foodborne pathogens and pesticides) to accumulate in edible tissues of vegetables; compromising the safety and quality of edible vegetables produced from farms sited in cities of many countries of the world, including Ghana. Swartjes et al. (2007) revealed that uptake of heavy metals by plants is a complex process and is affected by the plant species, climate, agriculture soil, and irrigation water. For instance, Yang et al. (2010) determined that Cd accumulates preferentially in lettuce, spinach, carrot, and radish but not in onion, pumpkin, and sweet pea. Hamilton et al. (2007) indicated that uptake of metals by plant crops depend on the bioavailable fraction of the metal species in the soil. For instance, Cd is less soluble in water and is strongly retained in the soil whereas As and Hg sorbs less strongly to soil colloids. Meanwhile Mn, Ni, Zn, Co, and Cd sorbs loosely and are more soluble in the soil solution.

In the western part of Ghana, there is a dearth of information on the status, sources, and distribution of heavy metals, as well as local factors that control metal uptake and transport into edible tissues of vegetables. This baseline information is of critical need for strategic development of remediation technology. In addition, heavy metals that induce adverse health effects has a long latency period, chronic, and difficult to detect in the affected human (Hamilton et al., 2007). Importantly, to meet the United Nations agenda for Sustainable Development Goal 2, which provides the blueprint to end hunger and malnutrition by 2050 (United Nations, 2014), it is equally essential to also highlight on the safety and quality of edible vegetables consumed in Ghana. According to Amoah et al. (2007), 0.82 million tons of vegetable were produced in 2010, and about 1250 tons of lettuce sold on markets in Accra, Ghana. Previous study showed that extensive information exists on the occurrence of food-borne pathogens (Amoah et al., 2007) and pesticides (Ntow et al., 2006) in edible portions of vegetables produced in some cities in Ghana. Unfortunately, the few studies that focused on metal distribution in vegetables (Lente et al., 2012) were limited in scope, for which information on soil and irrigation water quality; and sources of metal load in the soil-vegetable system were poorly investigated.

In spite of the epidemiological and scientific evidence which support the protective effect of vegetables against cancers, obesity, diabetes, and other non-communicable diseases (International Agency for Research on Cancer, 2003), Cd, Fe, Hg, Cr, and Ni (Xu et al., 2013; Lente et al., 2012) and faecal coliforms (Schmidt and Gemmell, 2012) in vegetables can pose health risk to human. At normal background concentrations, the less toxic heavy metals such as Fe and Zn, which are essential nutrients for plant growth may pose reduced risk to human health. However, at elevated concentrations, these metals deplete some essential nutrients, retard intrauterine growth, cause immunological weakness, organ damage, and reproductive dysfunction in human (WHO, 1995). Chronic exposure to low doses of more toxic metals such as Cd, Hg, Ni, and Cr contribute to upper gastrointestinal cancer, lung cancers, post-menopausal cancer, renal tubular dysfunction and anaemia in human (Xu et al., 2013). Furthermore, these metals are persistent in the environment and bioaccumulate in human tissues (Hajeb et al., 2014). Sekondi-Takoradi Metropolis, the administrative capital of the Western Region is a fast-growing urban community in Ghana. With a population of 559548 as at the 2010 Population and Housing Census (Ghana Statistical Service, 2010), the city is the hub for the offshore oil drilling and exploration, industries, and export and import activities in the country (Ghana Statistical Service, 2010). In this city, Lactuca sativa, Brassica oleracea, Brassica rapa, Corchorus olitorius, Amaranthus spinosus, Capsicum frutescens, Raphanus sativus, Daucus carota, Capsicum annum, and Allium cepa are openly irrigated with both untreated MWD stream and groundwater from shallow wells, since good quality water sources are not available on the farms (Lente et al., 2012). In addition, high input of inorganic and organic fertilizers, animal manure, and pesticides applied to the soil, may elevate the background concentration of Cd, Fe, Zn, Hg, Cr, and Ni in soils and edible tissues of vegetable crops. Florkowski et al. (2014) revealed that these vegetables are purchased by residents, tourist, and expatriates from open markets, street stands, or the supermarkets. To protect public health from exposure to heavy metal, it is critical that the quality, safety, and sources of metal species in irrigation water, soil, and tissues of vegetables are evaluated. The objectives of the study are: 1) to evaluate the distribution of Fe, Zn, Cd, Ni, Hg, and Cr in irrigation water sources, soil, leafy vegetables, and non-leafy vegetables produced from an urban farmland 2) to evaluate the quality of soil and irrigation water and to compare the concentration of the metals in edible tissues of vegetables to local and international safe limits; 3) to assess the sources and the accumulation potential of the metals in the soil and vegetables, and 4) to evaluate the health risk of the metal species in edible portions of vegetables frequently consumed by the population.

2. Materials and method

2.1. Chemicals

Concentrated nitric acid (purity: 70%), hydrochloric acid (37%), and hydrogen peroxide (30 %) of trace grade from Sigma Aldrich (St Louis, USA) were used in the acid digestion. Other chemicals and reagents including orthophosphoric acid (85 %), sulphuric acid (95–98%), potassium dichromate (>99.0 %), ferrous ammonium sulphate (99 %), barium chloride (10 mesh, 99.9 %), barium sulphate (99.9 %), and ferroin indicator solution (quality level 200) were of American Chemical Society (ACS) grade from Sigma Aldrich (St Louis, USA). Standard reference material (SRM) consisting of tomato leaf 1573a (trace metals in vegetable), 1646a (trace metals in soil) and 1643e (trace metals in water) from the National Institute of Science and Technology (NIST) (Gaithersburg, Maryland, USA) were used to check accuracy and precision of the analytical procedures. Deionized water (18.2 MΩ cm) produced by Milli-Q system (Millipore Co., USA) were used to prepare working solutions used in the laboratory analysis.

2.2. Description of the study area

The vegetable farms are located at latitude 4°54’0” N to 4°54’20” N and longitude 1°46’25” W to 1°46’15” W (Figure 1). The perimeter and area of the vegetable farm are 1649 m and 91953 m², respectively. The elevation varies from 23.0 to 31.7 m (a.m.s.l.). The location of the vegetable farm is about 2 km from Takoradi market, 9 km from Sekondi market, and 50 m perpendicular from the highway which links Takoradi and Elbow townships. Also, the vegetable farm is about 300 m away from a cluster of automobile fitting shops, fuel (petrol and gasoline) station, residential communities, and 350–450 m away from a local airfield. It is important to note that extensive research work of Nabulo et al. (2006) revealed that vegetable crops tend to contain background concentrations of Cd and other metals when planted 30–33 m away from vehicular emission. During rainfall, run-off and wastewater from nearby residential areas infiltrate into the urban stream which flows through the vegetable farmland. In reference to Swartjes et al. (2007), the criteria used to select the vegetables included: (1) the frequency of occurrence on the farm; (2) the purchasing pattern and preference of consumers, and (4) the affinity of the vegetable to accumulate heavy metals. In this context, indigenous leafy vegetables (Corchorus olitorius and Amaranthus spinosus), exotic leafy vegetables (Lacuna sativa, Brassica oleracea, Brassica rapa), and non-leafy vegetables (Capsicum annum) cultivated on the farmlands in the twin-city are investigated. The vegetables are irrigated daily in the morning and evening with water from six unprotected shallow wells (20–35 m depth) and the MWD stream using buckets and cans. In the dry
seasons, groundwater is mainly used to irrigate the vegetable crops. The climate is warm and humid, and temperature ranged from 20 to 30 °C. The annual precipitation is 1380 mm (Affum et al., 2015). Rainfall occurs from May to July and peaks from September to October in each year. From November to April, harmattan wind blows dusty and dry wind from the Saharan desert into the country (Affum et al., 2015). Ferric Acrisol derived from rocks formed in early Paleozoic Sekondian Group is the dominant soil in the vegetable farm (FAO, 2015; Affum et al., 2015). The tectonic of the area indicates a sedimentary basin domain composed of five sandstone formations interbedded with two undifferentiated shales formations (Geological Map of Ghana, 2009). The Global Positioning System (GPS) location of the sampling sites are shown in Table S1 of the supplementary material.

2.3. Sample collection and field assessment

The irrigation water, cultivating soil, and edible portions of vegetable were sampled on the same day from November 2018 to January 2019. The soil and the vegetable samples were collected from nine beds in the vegetable farm. For irrigation water, duplicate sets of 300 mL of water from the five shallow wells and the MWD stream were collected at 7–9 am from the vegetable farm into pre-cleaned polyethene bottles. One set of the water samples was acidified to a pH < 2 with 60 % nitric acid, whereas the other water samples remain unacidified. For bacteriological analysis, 50 mL of the water was collected into sterile Eppendorf tubes and kept at 4 °C. Subsequently, nine duplicate composite good quality and matured indigenous leafy vegetables (Corchorus olitorius, Amaranthus spinosus), exotic leafy vegetables (Brassica rapa, Brassica oleracea, and Lactuca sativa), and non-leafy vegetables (Raphanus sativus, Daucus carota, Allium cepa, and Capsicum annuum) were collected randomly from nine beds in the vegetable farm (see Figure 1). The cost of each vegetable did not exceed GHS¢10.00 (USD $1.70). Finally, a duplicate composite surface soil sample each weighing 100 g and from a depth of 0–20 cm were collected with a stainless steel from the root zone of the vegetable crop per each vegetable bed into a zipped locked polyethylene bag. For the reference background concentration of the metals, three random soil samples were collected from a pristine site located uphill of the vegetable farm. This site has no record of ever being exposed to any industrial or agricultural activity.

2.4. Sample treatment

The soil, vegetable, and water samples were steriley packed into individual ice-chest, maintained at 4 °C, and conveyed to the Ghana Atomic Energy Commission in Accra for sample treatment, storage, and analysis. In the laboratory, the edible parts of the vegetable were separated from each of the vegetable samples with a stainless knife and washed three times with double distilled water to remove air-born pollutants, soil, metal debris from the watering can, and other contaminants. Each fresh vegetable was allowed to dry at room temperature (21 °C), diced and weighed as fresh weight (fw). Later, the samples were lyophilised using the Labconco laboratory-scale Freeze Dryer System 77545-11 (Labconco Corporation, Missouri, USA) and reweighed as dry weight (dw). A waring laboratory-scale blender (model 348L09, Waring Commercial, New Hartford, Connecticut, USA) was used to ground the dried vegetable to <0.2 mm mesh size, kept in polyethylene containers.
and preserved in a desiccator. Soil samples were air-dried at room temperature for three days. Artifacts, such as plant material and cobble stones in the soil were removed, after which the soil was passed through a nylon sieve of 2 mm mesh size. Approximately 2 g of this soil was ground to 0.150 mm mesh size for Cd, As, Hg, Zn, Fe, and Ni analysis. The remaining portion of the 2 mm mesh size soil was used to determine cation exchange (CEC), pH, and total organic carbon (TOC) in the soil samples.

2.5. Sample analysis

2.5.1. Acid digestion and instrumental analysis of samples

The water, vegetable, and soil samples were acid digested in a Milestone microwave (ETHOS 900 microwave digester, Connecticut, USA) using standard acid digestion protocol described by Affum et al. (2015). Approximately 0.5 g of the soil and 5.0 mL of the water were separately digested in 6 mL nitric acid (65 %) and 2 mL hydrochloric acid (37 %). For vegetables, 0.5 g of the sample was digested in 5 mL of nitric acid (65 %) and 1 mL of hydrogen peroxide (30 %). The digested samples were diluted with deionized water before metal analysis.

The digested samples were analyzed by Atomic Absorption Spectrophotometry (AAS) (Varian Spectra AA110/220, Agilent Scientific Instruments, California, USA). Mercury was determined by cold vapour AAS (CVAAS), while the Zn, Cd, Cr, Fe, and Ni were by the direct flame AAS (FAAS) (240FS AA System, Agilent Scientific Instruments, California, USA) using the validated standard protocol for metal analysis (APHA, 1995). Each sample was analyzed in triplicate to assess the precision and accuracy of the AAS method. Three blanks and certified reference materials SRM 1573a, NIST 1643e, and SRM 1646a (see section 2.1) were digested along with the vegetables, water, and soil samples, respectively.

2.6. Physicochemical characteristics

2.6.1. Cultivating soil

The sand, clay, and silt content of the soil samples associated with the growth of the vegetable crops were determined by the Bouyoucos hydrometer method (Bouyoucos, 1962) after pretreatment with 30 % hydrogen peroxide to remove organic matter and soluble salts. The pH of the soil samples was measured in a 1:2 ratios (w/v) of 2 mm air-dried soil sample to distilled water using a Hanna HI 9813-5 multi-parameter meter (Hanna Instruments, Rhode Island, USA). Percent total organic carbon in the soil was determined by the acidified potassium dichromate oxidation method (Nelson and Sommers, 1982). Soil organic matter (SOM) was obtained by multiplying the % TOC by the universal Van Bemmelen factor of 1,724. The cation exchange capacity (CEC) (cmol+/kg) was determined using 1.0 M ammonium acetate buffer to a pH of 7.0 (Chapman, 1965; Andrea et al., 2019).

2.6.2. Irrigation water

The in-situ parameters such as, conductivity, pH, total dissolved solids (TDS), salinity, and temperature were determined with a standardized Hanna HI 9813-5 (Hanna Instruments, Rhode Island, USA) during the time of water sampling. Using standard methods for water and wastewater analysis described in APHA (1995), the concentration of Cl−, SO42−, NO3, and PO43− ions in the non-acidiﬁed water samples were determined. For bacteriological analysis, the pour plate method described by Affum et al. (2015) was used to identify the total coliform and faecal coliform (E.coli) load in the irrigation water sources. To achieve a bacteria colony count of within 20-100 colonies, water samples were diluted 10 and 100 folds with the Ringer’s solution. In the biochemical test, a single coliform colony from the 10 fold diluted water sample was inoculated into Triple Sugar Ion Agar Slant, Sulphur Indole Motililty Agar Butt and Simmons’ Citrate Agar slant and incubated at 37 °C. Cell growth and colour in the agar after incubation was indicative of the existence of coliforms in the water. Finally, detected coliforms were confirmed using the BioMérieux's API® identification Test Kits (API – 20 E). This method was validated from negative, positive and sterility controls. The results were expressed as coliform forming units (cfu/100 mL).

2.7. Quality control and quality assurance

At a signal-to-noise ratio of 3 and 10, the limit of detection (LOD) and the limit of quantitation (LOQ) were respectively computed from the standard deviation of twenty-five injections of the blank solution and the slope of the calibration curve. The recoveries of heavy metals ranged from 70 to 102 % after spiking at 0.5 and 1 mg/L of each element in the water, soil, and vegetable samples.

2.8. Statistical analysis

Descriptive statistics and normality (Shapiro test and normal Q-Q plot) test of the physicochemical and heavy metal datasets obtained from the irrigation water, soil, and the vegetable samples were achieved by SPSS 22.0 (SPSS Inc., Chicago, USA). The Analysis of Variance (ANOVA) and the post hoc test, Tukey HSD were used to identify heavy metals whose distribution among the sampling sites was statistically significant (p < 0.05). For source apportionment of the metals in the soils, principal component analysis-multiple linear regression (PCA-MLR) model (Shi et al., 2009; Zhang et al., 2017) was applied to 9 soil samples x 7 metal species. The model identifies the potential metal source categories, the source contributions of the metal species, and metal profile in soil. Pearson correlation and hierarchal cluster analysis (HCA) were used to identify the relationship between and within metal species in the soil and vegetables. Result were considered statistically significant at a probability level of p < 0.05 or 0.01. A lognormal (mean, standard deviation) probabilistic distribution model in @Risk™ Software 7.6 (Palisade Corp., USA) was fitted to the concentrations of each heavy metal category to obtain the 95th percentile value (worst-case scenario or high-end exposure probability) used in the non-carcinogen and carcinogenic risk assessment. Where appropriate, data values < LOD were treated as half the LOD.

2.9. Pollution assessment

2.9.1. Groundwater and municipal-waste-dominated stream

Irrigation water quality indicator sodium adsorption ratio (SAR), and other models such as sodium soluble percentage (SSP); residual sodium carbonate (RSC); Kelly ratio (KR); permeability index (PI); and magnesium adsorption ratio (MAR), and toxic metal and E. coli load were used to evaluate the quality and suitability of groundwater and the MWD stream for vegetable crop irrigation (Ayers and Westcot, 1985). The SAR quantified the sodicity potential of the water resources on the farm (Hamilton et al., 2007) and is expressed by the equation:

\[ \text{SAR} = \frac{\text{Na}^+}{\sqrt{(\text{Ca}^{++} + \text{Mg}^{++})}} \]  

Modeled equations for the other irrigation models are provided in the supplementary material.

2.9.2. Cultivating soil

The geo-accumulation index (Igeo) and the enrichment factor (EF) were used to assess the status of Cd, Fe, Zn, Hg, Cr, and Ni in the soil samples (Muller, 1969; Bhuiyan et al., 2010; Yang et al., 2019). The Igeo is expressed by the equation:

\[ I_{geo} = \frac{\log C - \log C_{0}}{1.5Bn} \]
In this study, Fe in the background soil sample was used as the reference to differentiate the anthropogenic and geogenic sources of the analyzed metals. The EF values for soil sample sites were classified as unpolluted (Igeo < 1); moderately polluted (1 < Igeo ≤ 2); heavily polluted (2 < Igeo ≤ 3); extremely polluted (Igeo > 5) (Bhuian et al., 2010; Chen et al., 2014). The EF is defined by the equation:

$$EF = \frac{C_{veg}}{C_{soil} \text{background}} (3)$$

Zhang et al. (2017) used the normalized enrichment factor to differentiate the anthropogenic and geogenic sources of the analyzed metals. In this study, Fe in the background soil sample was used as the reference element for geochemical normalization (Al-Wabel et al., 2017). Iron is typically used in geochemical normalization because it is naturally abundant, natural concentration is uniform, interacts minimally with other metals, has fine solid surfaces, and its geochemistry is similar to that of many trace metals (Al-Wabel et al., 2017). Soil sample site is classified as deficient and minimally enriched for EF value < 1; moderately enriched (2–5); significantly enriched (5–20); very highly enriched (20–40); and extremely highly enriched (>40). Ye et al. (2011) reported that soils with an EF value < 2 indicates a crustal origin and are governed by natural weathering process, whereas those with an EF > 2 suggests that the element is of anthropogenic origin. An EF value < 1 suggest a possible mobilization or depletion of metals.

### 2.10. Accumulation of heavy metal species in vegetables

In reference to Zhang et al. (2017), the bioconcentration factor (BCF) establishes the relationship of elements in soil to edible parts of vegetable and is expressed by the equation:

$$\text{BCF} = \frac{C_{veg}}{C_{soil}} (4)$$

where \( C_{veg} \) and \( C_{soil} \) represent the concentration of heavy metal in edible part of vegetable (fresh weight) and in the soil (dry weight), respectively.

### 2.11. Potential human health risk assessment

Non-carcinogenic (acute and chronic) risk models developed by the US EPA (2007) were used to assess the potential human health risk of Zn, Fe, Hg, Ni, Cr, and Cd in edible portions of leafy and non-leafy vegetables. In Ghana, Brassica rapa, Brassica oleracea, and Lactuca sativa are often eaten raw as salad, whereas Corchorus olitorius and Amaranthus spinosus are cooked as stew (Amoah et al., 2007; Lente et al., 2012; Drechsel and Kariate, 2014). Meanwhile, Capsicum annum is eaten raw or added to stew. In order to reflect the consumption pattern of the vegetables in major cities, data on the consumption amount and frequency of vegetable intake were obtained from Amaoh et al. (2007), Florkowski et al. (2013) and Lente et al. (2012). Based on a 24-h dietary recall survey for 1076 adult respondents in 2013, Florkowski et al., 2013 revealed that 52–73 % of adult male or females respond consumption cabbages weekly, whereas 20–25 % the respondents consume the vegetable daily in Takoradi and Accra of Ghana. Also, Lente et al. (2012) and Amaoh et al. (2007) showed that the amount (upper end of normal consumption amount) of fresh Lactuca sativa (lettuce), Brassica rapa and Brassica oleracea (cabbages), Capsicum annum (pepper) and Corchorus olitorius (Ayoyo) or Amaranthus spinosus (Alefe) consumed by average income earning person is 12.86, 35.71, 7.14, and 57.14 g/person/day, respectively. This information was used to estimate the daily intake (EDI), target hazard quotient (THQ), and the hazard index (HI) due to the exposure to Fe, Zn, Cd, Ni, Hg, and Cr through oral consumption of the vegetables. The EDI is expressed by the equation:

$$\text{EDI} = C_{veg} \times IR_{veg} (5)$$

The EDI (µg/day) of the metals in edible tissues of the vegetables were compared to the recommended tolerable upper daily intake (UL) for an adult older than or equal 30 yrs reported by the Joint FAO/WHO Expert Committee on Food Additives (JECEA, 2011; World Health Organization (WHO, 1996)) and the Food and Nutrition Board of the National academy of Sciences, USA (Food and Nutrition Board, 2001). The THQ was used to characterize the health risk of consuming vegetables (Xu et al., 2013), and the non-cancer health risk associated with heavy metals (US EPA, 2007). The THQ is a ratio of the determined dose of a pollutant to a reference dose (RfD) (Xu et al., 2013):

$$\text{THQ} = \frac{\text{CDI}}{\text{RfD}} = \frac{C_{veg} \times IR_{veg} \times EF_{veg} \times ED \times 10^{-3}}{\text{BW} \times AT \times RfD} (6)$$

For THQ < 1, the exposed population is unlikely to experience adverse health effects from exposure to the metal species through the ingestion pathway. The RfD is an estimation of the daily exposure to the human population likely to be without an appreciable risk of deleterious effects during a lifetime (US EPA, 2007). The chronic daily intake (CDI) (mg/kg/day) is the exposure to the population over a lifetime. The values of RfD for Zn, Cr, Ni, Fe, Cd, and Hg are 0.3, 0.003, 0.02, 0.7, 0.001, and 0.0003 mg/kg/day, respectively (US EPA, 2007). The Cveg is the 95th percentile concentration value of heavy metals in the edible tissue of the vegetable (mg/kg, fresh weight). The IRveg is the amount of vegetable consumed daily. Information on the average IRveg for children and the youth is poorly reported in Ghana. EDI is the exposure frequency. The ED values for cabbages, lettuce, green pepper, and Ayoyo/Alefe are 52, 156, 261, and 104 days/year, respectively. The ED is the exposure duration (70 yrs for adult), and BW is the body weight (72.43 kg for adult > 30 yrs) (Vuvor and Harrison, 2017). AT is the average lifetime for non-carcinogens (ED x 365 days/year). The cumulative dietary exposure (hazard index) (Xu et al., 2013) which is the sum of the target hazard quotients (THQ) for all heavy metals is expressed by the equation:

$$\text{HI} = \sum_{i=1}^{n} \text{THQ}_i (7)$$

It is worth to mention that insufficient information on cancer slope factor for the heavy metals from the US EPA Integrated Risk Information System database (US EPA, 2007), did not allow for cancer risk assessment. For regulatory purposes, the acceptable or tolerable cancer risk is established for the range of 10⁻⁶ – 10⁻⁴ (US EPA, 2007).

### 3. Results

#### 3.1. Physicochemical characteristics of cultivating soil

The physicochemical characteristic of the soils which support the growth of the vegetable crops is presented in Table 1. The pH of the soil ranged from 6.60 to 8.05 (7.41 ± 0.04 units), indicative of slightly acidic to alkaline soil. Apart from the pH of the soil around the root zones of Capsicum annum (S5) and Dacucus carota (S7), the pH of the remaining soil samples was relatively higher than pH of the reference background soil (6.60) which was slightly acidic. The percent total organic carbon of the soil samples ranged from 4.71 to 6.97 % (5.75 ± 0.00%). The highest TOC occurred at S5 (6.97 %). Meanwhile, apart from soils at sites S1, S2, S3, and S6 which had lower TOC, 44 % of the soil’s samples had a TOC ranging from 4.71 to 6.97 % (5.75 ± 0.00%). Meanwhile, apart from soils at sites S1, S2, S3, and S6 which had lower TOC, 44 % of the soil’s samples had a TOC ranging from 4.71 to 6.97 % (5.75 ± 0.00%).
| Soil sample ID# | Physicochemical parameters of soil | Heavy metal concentration in soil (mean ± sd, mg/Kg) |
|----------------|-----------------------------------|-----------------------------------------------|
|               | pH    | % TOC | % SOM | CEC Cmol+/kg | Cd     | Fe     | Hg     | Cr     | Ni     | Zn     |
| S1            | 7.63 ± 0.00 | 5.63 ± 0.02 | 9.70 ± 0.00 | 27.69 ± 0.09 | <0.002 | 369.06 ± 0.02 | 0.073 ± 0.02 | 0.64 ± 0.00 | <0.001 | 1.71 ± 0.00 |
| S2            | 7.45 ± 0.07 | 4.81 ± 0.00 | 8.29 ± 0.00 | 20.22 ± 0.01 | 0.007 ± 0.00 | 294.57 ± 0.01 | 0.036 ± 0.00 | 1.47 ± 0.01 | <0.001 | 4.00 ± 0.00 |
| S3            | 7.50 ± 0.00 | 4.71 ± 0.01 | 8.13 ± 0.00 | 36.77 ± 0.00 | <0.002 | 294.24 ± 0.01 | 0.021 ± 0.01 | 2.15 ± 0.01 | <0.001 | 2.55 ± 0.01 |
| S4            | 7.65 ± 0.07 | 6.38 ± 0.00 | 10.99 ± 0.01 | 11.94 ± 0.00 | <0.002 | 293.87 ± 0.00 | 0.054 ± 0.01 | <0.001 | <0.001 | 5.05 ± 0.00 |
| S5            | 6.60 ± 0.07 | 6.97 ± 0.00 | 12.01 ± 0.00 | 10.93 ± 0.05 | <0.002 | 295.64 ± 0.02 | <0.002 | 2.51 ± 0.00 | <0.001 | 1.20 ± 0.00 |
| S6            | 7.40 ± 0.00 | 4.81 ± 0.00 | 8.29 ± 0.00 | 33.40 ± 0.00 | 0.013 ± 0.00 | 296.34 ± 0.00 | 0.120 ± 0.01 | 3.42 ± 0.00 | 0.460 ± 0.01 | 6.28 ± 0.00 |
| S7            | 6.80 ± 0.00 | 6.17 ± 0.00 | 10.64 ± 0.00 | 14.58 ± 0.01 | <0.002 | 297.00 ± 0.01 | 0.053 ± 0.01 | 3.73 ± 0.00 | <0.001 | 3.84 ± 0.01 |
| S8            | 7.65 ± 0.07 | 6.43 ± 0.00 | 11.09 ± 0.00 | 29.63 ± 0.00 | <0.002 | 294.72 ± 0.01 | 0.031 ± 0.00 | 1.58 ± 0.00 | <0.001 | 1.91 ± 0.01 |
| S9            | 8.05 ± 0.07 | 5.81 ± 0.00 | 10.02 ± 0.00 | 18.80 ± 0.00 | <0.002 | 294.99 ± 0.00 | 0.011 ± 0.00 | 2.01 ± 0.01 | <0.001 | 2.67 ± 0.01 |
| BKG soil      | 6.60 ± 0.07 | 5.86 ± 0.23 | 10.10 ± 0.38 | 20.03 ± 0.00 | <0.002 | 250.00 ± 0.00 | 0.032 ± 0.00 | <0.001 | <0.001 | 1.02 ± 0.01 |
| Min           | 6.60 | 4.71 | 8.13 | 10.93 | <0.002 | 293.87 ± 0.00 | <0.002 | <0.001 | <0.001 | 1.20 |
| Max           | 8.05 | 6.97 | 12.01 | 36.77 | 0.013 | 369.06 | 0.120 | 3.73 | 0.460 | 6.28 |
| Mean          | 7.41 ± 0.04 | 5.75 ± 0.00 | 9.91 ± 0.00 | 22.64 ± 0.02 | 0.002 ± 0.00 | 303.38 ± 0.01 | 0.044 ± 0.01 | 1.95 ± 0.00 | 0.051 ± 0.00 | 3.25 ± 0.00 |

Literature SQG
- 3a
- 50000b
- 0.07
- 100c
- 50d
- 300e

Canadian SQGf
- 1.4
- 6.6
- 64
- 45
- 200

US EPAg SQG
- 0.77
- 
- 2300

FinlandSQG
- 10.00
- 200
- 100
- 250

Soil samples S1, S2, S3, S4, S5, S6, S7, S8, and S9 were collected around the root zone of Allium cepa, Amaranthus spinosus, Brassica oleracea, Brassica rapa, Capsicum annuum, Corchorus olitorious, Daucus carota, Lactuca sativa, and Raphanus sativus respectively. N = 9. Each value represents an average of a triplicate determination. Meaning of abbreviation: Soil Quality Guideline (SQG), Background (BKG).

aEwers (1991); bKabata-Pendias and Pendias (1992); cCCME (2007); dUS EPA (2018), and eMinistry of the Environment, Finland (2007).
Table 2. Concentration of heavy metal and water content in fresh edible parts of leafy and non-leafy vegetable species produced from an urban farm.

| Plant species          | Edible part | Water Content (%) | Heavy metal concentration in edible part of vegetables (mean ± 3SD, μg/kg, fresh weight) |
|------------------------|-------------|-------------------|-------------------------------------------------------------------------------------|
|                        |             |                   | Cd                                                                                   |
|                        |             |                   | Cr                                                                                   |
|                        |             |                   | Zn                                                                                   |
|                        |             |                   | Ni                                                                                   |
|                        |             |                   | Fe                                                                                   |
|                        |             |                   | Hg                                                                                   |
|                        |             |                   | Relative standard deviation (RSD) of the concentration of Cd, Fe, Zn, Cr, Ni, and Pb | 3.2. Physicochemical characteristics of irrigation water

The physicochemical parameter of the irrigation water (groundwater and the MWD stream) is presented in Table S2 (supplementary material). Generally, the physicochemical parameters in both water sources did not exceed the recommended maximum concentration for crop irrigation, except for K⁺, pH, PO₄³⁻, E. coli, and total coliforms. For both water sources, concentration of Ca²⁺, Mg²⁺, Na⁺, and K⁺ in water sources ranged from 3.67 to 17.07 mg/L (11.16 mg/L), 7.87–97.37 mg/L (44.19 mg/L), 28.8–254 mg/L (130 mg/L), 14.3–69.6 mg/L (37.2 mg/L). Meanwhile, the concentration of Cl⁻, NO₃⁻, SO₄²⁻ and PO₄³⁻ also ranged from 37.9 to 334.35 mg/L (166 mg/L), <0.010–0.597 mg/L (0.161 mg/L), 0.6–6.5 mg/L (3.9 mg/L) and <0.01–16.8 mg/L (8.2 mg/L), respectively. The TDS and EC ranged from 150 to 1043 mg/L (626 mg/L) and 325 to 2057 µS/cm (1107 µS/cm), respectively. The hydrogen ion activity (pH) of W1 (7.8), W2 (7.5), MWD stream (8.0) and W6 (8.2) indicate a near neutral to slightly alkaline, however, W3 (9.0) and W5 (8.7) were more alkaline, exceeding the FAO recommended guideline limit of 6.5–8.4 (Ayers and Westcot, 1985), and 6.22–8.64 for wastewater used to irrigate vegetable crops in Accra (Ackah et al., 2014). The concentration of K⁺ (35.4 mg/L) and PO₄³⁻ (12.2 mg/L) in MWD stream was higher than observed in W1 (14.3 mg/L, 1.72 mg/L), W2 (18.9 mg/L, 0.001 mg/L), W5 (10.7 mg/L, and W6 (34.8 mg/L, 6.8 mg/L), respectively. An E. coli load of 200 cfu/mL was observed in only MWD stream and W3. The temperature of the water sources ranged from 28.0 to 31.2 °C.

3.3. Distribution of heavy metal species in vegetables

The concentration of Cd, Fe, Zn, Hg, Cr, and Ni in the edible parts of the vegetables is shown in Table 2. The difference in the mean concentration of the metals among the different non-leafy and leafy vegetables was statistically significant for Cd, Fe, and Zn (p < 0.05, ANOVA). A post hoc test revealed that significance differences (p < 0.05, Tukey HSD) existed for Cd in Daucus carota and Amaranthus olitorious, Fe in Raphanus sativus and Brassica rapa, and Zn in Raphanus sativus, Brassica rapa, Corchorus olitorious, Allium cepa and Amaranthus spinus with the rest of the vegetables. The mean concentration of Hg, Cd, Fe, Cr, Ni, and Zn in each leafy and non-leafy vegetable was less than guideline limits proposed by the Joint FAO/WHO Expert Committee on Food additives (FAO/WHO, 2001), and data provided by Bigdeli and Selipsespor (2008) who investigated similar metal accumulation in vegetables irrigated with wastewater in the Shahre Rey, Iran. The concentration of Cr and Ni in edible parts of the different leafy and non-leafy vegetables were less than the LOD value of 0.001 mg/kg. The mean concentration of the metals in the indigenous leafy vegetable (mg/kg fw) ranged from 0.022 – 0.027 (Cd), 27.582–36.178 (Fe), 0.009–0.013 (Hg), and <0.0001–0.202 (Zn). For the exotic leafy vegetable (mg/kg fw) the heavy metals ranged from 0.008 – 0.018 (Cd), 4.517–16.520 (Fe), 0.001–0.004 (Hg), and <0.001–0.079 (Zn). The mean concentration of Cd, Fe, Hg, and Zn in Raphanus sativus, Allium cepa, and Capsicum annum were 0.014, 4.625, 0.004, and <0.001 mg/kg fw; 0.011, 7.294, 0.002, and 0.071 mg/kg fw; and 0.024, 3.617, 0.004, and 0.093 mg/kg fw, respectively. The decreasing order of the mean concentration of heavy metal Fe > Zn > Hg value of 20.03 ± 0.00 cmol+/kg for an average soil composition of 67 % sand, 15 % clay and 18 % silt. Reference to the US soil textural classification (USDA, 2020) revealed a sandy environment categorized as sandy loam and coarse. The relative increases in TOC and CEC in soils above the background soil may be due to the high organic matter from decomposed fallen parts of the vegetable crop or the MWD stream. The mean concentration of CEC in soils was about two times lower than the mean CEC value reported for soil samples of urban vegetable farms in Akaki (50.2 cmol+/kg), Coffa and Kera (39.8 cmol+/kg), and Peacock farms (38.4 cmol+/kg) in Ethiopia (Weldegebrial et al., 2012). The clay content of soil at Akaki, Coffa and Kera, and Peacocks farms was 51, 47, and 37 %, whereas the organic carbon was 1.5, 2.4 and 1.7 %, respectively.
The concentration of heavy metals in irrigation water from various sources is shown in Table 3. Generally, Fe concentration was higher than observed in the MWD stream. Furthermore, the concentration of Ni and Zn in the wastewater (0.06 and 0.37 mg/L) and groundwater (0.13 mg/L) reported by Lente et al. (2012) exceeded the concentration measured in this study. The concentration of Fe, Hg, and Zn in the soil ranged between 293.87 to 369.06 mg/kg (303.38 ± 0.01 mg/kg), <0.002–0.120 mg/kg (0.044 ± 0.01 mg/kg) and 1.28–6.28 mg/kg (3.25 ± 0.00 mg/kg), respectively. The mean concentration of heavy metals in soil decreased in the order of Fe > N > Hg > Cd.

### 3.4. Distribution of heavy metal species in cultivating soil

In reference to Table 3 of section 3.1, the difference in the mean concentration of Cd, Fe, Zn, Hg, Cr, and Ni among the soil sampling sites was statistically significant for Fe, Hg, Zn, and Ni (p < 0.05, ANOVA). A post hoc test revealed that significance differences (p < 0.05, Tukey HSD) exist for Fe and Zn at all soil sites except S1 and S7. For Hg and Ni, the differences occurred between S1 and all soil sites (p < 0.05, Tukey HSD). Apart from the mean concentration of Hg (0.120 ± 0.01 mg/kg) in soil around the root zone of Corchorus olitorius, and Raphanus sativus (0.073 ± 0.02 mg/kg), the concentration of Zn, Cd, Cr, Fe, Ni, and Hg in soils from the root zone of the other vegetable crops were generally below the soil quality guideline limits reported by Ewers (1991) and the Environment Minister of Canada (Canadian Council of Ministers of the Environment, 2007), United States Regional Soil Screening levels (US EPA, 2018), and Ministry of the Environment of Finland (MEF, 2007).

Generally, Cd, and Ni were below the LOD values of 0.002 and 0.005 mg/kg, respectively. Furthermore, the concentrations of Cd, in soils around the root zone of Amaranthus spinosus (0.007 mg/kg) and Corchorus olitorius (0.013 mg/kg; Ni 0.460 mg/kg) were lower than the guideline values reported in the USA, Finland, and Canada. The concentration of Fe, Hg, and Zn in the soil ranged between 293.87 to 369.06 mg/kg (303.38 ± 0.01 mg/kg), <0.002–0.120 mg/kg (0.044 ± 0.01 mg/kg) and 1.28–6.28 mg/kg (3.25 ± 0.00 mg/kg), respectively. The mean concentration of heavy metals in soil decreased in the order of Fe > Zn > Cr > Ni > Hg > Cd.

### 3.5. Distribution of heavy metal species in irrigation water

The mean concentration of Cd, Fe, Zn, Hg, Cr, and Ni in the MWD stream and groundwater sources on the vegetable farm is shown in Table 3. Generally, the concentration of Cd (0.016 mg/L), Fe (2.336 mg/L), and Hg (0.007 mg/L) in 60%, 80%, and 40% of the groundwater sources, respectively, was higher than observed in the MWD stream. Furthermore, the concentration of Fe, Ni, and Cr were below the FAO safe limit of 5.0, 0.2, and 0.1 mg/L, respectively, for irrigation water (Ayers and Wescot, 1985), except for Cd which exceeded the FAO limit of 0.01 mg/L by 160–220 folds in 83% of the water samples. The mean concentration of Cd in W1, W2, and W3 was about 1.2 times higher than observed in MWD stream (0.018 mg/L). Meanwhile, Cd in MWD stream was 1 and 9 times higher than in W5 and W6, respectively. The Cd concentrations in the water sources was higher than 0.005 mg/L reported by Lente et al. (2012) for irrigable wastewater and groundwater applied to vegetable farms in Accra. Except for W2 (0.842 mg/L), the mean concentration of Fe in W1, W5, W6, and W3 were 1.97, 1.48, 2.19, and 1.0 times, respectively, higher than in MWD stream (1.622 mg/L). The concentration of Ni, Zn, and Cr in both water sources were below the LOD value of 0.001 mg/L. The concentration of Cr in both water sources was consistent with the data reported by Lente et al. (2012) who investigated Cr in groundwater and municipal wastewater applied to vegetables in Accra. In contrast, the concentration of Ni and Zn in the wastewater (0.06 and 0.14 mg/L) and groundwater (0.13 mg/L) reported by Lente et al. (2012) exceeded the concentration measured in this study. The concentration of Hg in the groundwater sources ranged from 0.005 to 0.013 mg/L, while that of MWD stream was 0.008 mg/L. The differences in the mean concentration of the heavy metals in the water sources was statistically significant for Fe and Hg (p < 0.05, ANOVA). A post hoc test revealed that significance differences (p < 0.05, Tukey HSD) exist for Fe in all groundwater sources and the MWD stream, whereas for Hg it existed in W6 and W2. Since the MWD stream and groundwater are applied to the...
crops concurrently, the mean concentration of Cd, Fe, and Hg for the combined water sources at the point of application to soil ranged from 0.002 to 0.022 mg/L (0.016 ± 0.000 mg/L), 0.842–3.552 mg/L (2.217 ± 0.005 mg/L) and <0.001–0.013 mg/L (0.001 ± 0.000 mg/L) respectively.

3.6. Geo-accumulation index (Igeo) and enrichment factor (EF)

In reference to the criteria used to assess the geochemical accumulation of the heavy metals in soil (Zhang et al., 2017), the Igeo value for Cd, Fe, Cr, Ni, and Zn at each soil sampling sites was <0, indicative of low geochemical contribution to these elements in the cultivating soil. Al-Wabel et al. (2017) and Ye et al. (2011) used the EF method to identify the origin of the metals in the environmental matrices. The EF values for Cd, Hg, Cr, Ni, and Zn ranged from 0.68 to 11.25, 0.03 to 3.17, 0.85 to 6271.73, 0.68 to 772.76, and 0.99 to 5.19, respectively. The average EF values for the individual elements in the soil samples decreased in the order of Cr (3265.52) > Ni (86.60) > Zn (2.66) > Cd (2.52) > Hg (1.13) > Fe (1.00). This indicates a relatively high enrichment for Cr and Ni, moderately enrichment for Zn and Cd, and deficient to minimal enrichment for Hg, and Fe. Considering each element per soil sample site, 78 % of the samples were deficient to minimally enriched (EF < 2), whereas soil samples at S2 (EF = 5.66) and S6 (EF = 11.25) were moderate and significantly enriched, respectively. Soil S2 and S6 support the growth of Amaranthus spinosus and Corchorus olitorious, respectively. For Hg and Ni, EF value was <2 at 89 % of the soil sample sites whereas soil sample S6 was moderately enriched (EF = 3.17) for Hg and extreme to highly enriched (EF = 772.76) for Ni. Chromium was extremely enriched at 89 % of the soil sample sites except at soil site S4 (0.85) which was deficient to minimally enriched. The S4 support the growth of Brassica rapa. Zinc was deficient to minimally enriched, moderately enriched, and significantly enriched at soil sites S1, S5 and S8; S2 -4, S7 and S9; and S6, respectively.

3.7. Quality of irrigation water

In reference to Ayers and Westcot (1985), the groundwater and the MWD stream can be used to irrigate the vegetable crops if the physico-chemical parameters and irrigation indices are within the FAO recommended safe limits (Table S3, supplementary material). Considering the period for which the groundwater and the MWD stream were analyzed, the pH, K⁺, Na⁺, Ca²⁺, SO₄²⁻, NO₃⁻, Cl⁻, TDS and EC for the water samples were within the FAO safe limits of 6.5–8.4, 2 mg/L, 930 mg/L, 400 mg/L, 952 mg/L, 45 mg/L, 1091 mg/L, 0–2000 mg/L and 0–3000 μS/cm,

Table 4. Varimax rotated factor loadings and source profile of heavy metal species in the soil obtained by the PCA-MLR model.

| Metal species | Factors | PC 1 | PC 2 | PC 3 | Communalties |
|---------------|---------|------|------|------|--------------|
|               | FC 1    | FC 2 | FC 3 |
| Cd            | 0.879   | -0.088 | 0.275 | 0.856 |
| Fe            | 0.007   | 0.952 | -0.278 | 0.984 |
| Hg            | 0.915   | 0.323 | -0.014 | 0.942 |
| Cr            | 0.160   | -0.271 | 0.925 | 0.954 |
| Zn            | 0.876   | -0.406 | -0.092 | 0.940 |
| Ni            | 0.880   | 0.029 | 0.388 | 0.925 |

Eigenvalues 3.177 1.258 1.167
% of variance 52.946 20.963 19.446
Cumulative % 52.946 73.909 93.355
Household waste and fertilizer Crustal material fertilizer

Factors have eigenvalues greater than 1. Factor loadings greater than 0.7 are highlighted.

Figure 2. PCA-MLR source contribution plot for soil used to cultivate leafy and non-leafy vegetable crops in an urban farm.
respectively. However, for W1 (1.72 mg/L) and W2 (<0.001 mg/L), the phosphate concentration in the MWD stream, W3, W5, and W6 was 2 or 3 times higher than the FAO safe limit of 6 mg/L. This is problematic because at such concentration the yield and maturity of the crops are affected since phosphorus is highly immobile in soils (Hamilton et al., 2007).

As inferred from Table 3, the irrigation index values were compared to the FAO safe limits proposed by Ayers and Westcot (1985). The calculated SSP indicated a “slight to moderate restriction” on the use of the groundwater sources for irrigation purposes. The RSC and SAR values indicated a “no restriction” on the use of the MWD stream and the groundwater sources for vegetable crop irrigation. Meanwhile, the calculated MAR showed a slight to moderate restriction for W1 and W2 and a severe restriction for W3, W4, MWD stream, and W6. The value of the Kelly ratio for each water source was greater than 1. This is indicative of a slight imbalance in magnesium and calcium ions in the groundwater and stream sources. The permeability index (59.38-80.33 %) rendered the water sources suitable for irrigating purposes. Although ESP for W3 (6.03 %) and W5 (5.71 %) marginally exceeded the 5 % limit, it was not a limiting factor for irrigation; thus, the soil samples were non sodic. An ESP of 10-15 % is generally accepted as critical level (Al-Hwaiti et al., 2016).

The exceedance of Cd above the FAO safe limits in about 80 % of the groundwater sources is a potential threat against the use of the water sources to irrigate the vegetable crops. Furthermore, the E. coli load in W3 (20000 cfu/100 mL) and MWD stream (20000 cfu/100 mL water) exceeded the WHO safe limit of 1000/100 mL (WHO 2006; Schmidt and Gemmell, 2012) by twenty folds. However, the geometric mean of E. coli load in both water sources (5.85) was higher than the value reported for groundwater (4.81) and streams (5.75) in Kumasi; and also for drains (4.89) and streams (4.99) in Accra as revealed by Amoah et al. (2007) who investigated the microbial contamination of irrigation water sources available in Accra and Kumasi. The E. coli load found in the water sources was lower than the observed for polluted Baynespruit river which is also used to irrigate vegetable crops in South Africa (Schmidt and Gemmell, 2012). Total coliforms load in the water sources ranged between 20000 and 240000 cfu/100 mL of water.

### 3.8. Source apportionment of heavy metal species in soil using PCA-MLR model

The source profile of the metal species, the contribution of the metals sources, and the association of the metals in the soil was determined from hierarchal cluster (HCA) and the classical principal component analysis-multiple regression (PCA – MLR). In the PCA, Bartlett’s Test of Sphericity (p < 0.05) and Kaiser-Meyer-Olkin (KMO = 0.703) measure of sampling adequacy were significant, indicating that dataset for Cd, Fe, Zn, Hg, Cr, and Ni in the soil was suitable for statistical analysis. In reference to the varimax rotated PCA loadings for the metals in Table 4, the three principal components (PC) resolved from the analysis explained 93.355% of the total cumulative variance of the data structure at an eigenvalue greater than 1.0. The first factor (PC1) accounted for 52.946 % of the total variance and revealed high positive loading value for Zn (0.876), Hg (0.915), Ni (0.880) and Cd (0.879). Considering the high loadings of Hg and Ni, this category of metal species is likely derived from mixed household waste and some fertilizers such as phosphate, nitrogen, potassium, and manure type fertilizers. In addition, the metals maybe transported together from specific sources. This supports the high enrichment factors (>1) observed for Zn, Hg, Ni, and Cd, indicative of an anthropic origin for the metals in the soil. Clearly, the loadings plot (Figures S1) and the dendrogram of the HCA (Figure S2a) confirmed the results of the PCA. Furthermore, Pearson correlation (Table S4) showed a

### Table 6. Estimated daily intake of heavy metal species from the consumption of leafy and non-leafy vegetables.

| Vegetable                | Estimated daily intake (EDI) of vegetable (µg/day/person) | Target hazard quotient (THQ) | Hazard index (HI) |
|--------------------------|----------------------------------------------------------|-----------------------------|------------------|
|                          | Cd    | Fe     | Hg     | Cr     | Ni     | Zn     | Cd     | Fe     | Hg     | Cr     | Ni     | Zn     |
| Amaranthus spinosus      | 1.54E+00 | 2.07E+03 | 5.33E+00 | 2.40E-03 | 2.40E-03 | 1.15E+01 | 0.01 | 0.01 | 0.07 | 0.00 | 0.00 | 0.00 | 0.09 |
| Corchorus olitorius      | 1.38E+00 | 1.58E+03 | 7.56E+00 | 2.16E-03 | 2.16E-03 | 2.16E-03 | 0.01 | 0.01 | 0.10 | 0.00 | 0.00 | 0.00 | 0.11 |
| Brassica oleracea        | 6.45E-01 | 2.10E+02 | 4.03E-02 | 1.01E+03 | 1.16E-03 | 1.04E+03 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| Brassica rapa            | 2.68E-01 | 1.61E+02 | 1.12E-01 | 6.71E+04 | 6.71E+04 | 6.71E+04 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| Lactuca sativa           | 1.82E-01 | 2.12E+02 | 2.02E-02 | 2.53E+04 | 2.53E+04 | 2.53E+04 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| Capsicum annuum          | 1.60E-01 | 2.58E+01 | 2.86E-02 | 2.86E+04 | 2.86E+04 | 2.86E+04 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| Total                    | 4.17   | 4.25E+04 | 1.31E+02 | 1.00E-02 | 1.00E-02 | 1.58E+01 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| RDA                      | 10-20× | 80000× | 40× | 25-35× | 70-260× | 8000-11000× |
| UL                       | 65×    | 45000× | 46× | 1000× | 40000× |

Meaning of abbreviations: Recommended Daily Allowance (RDA); Tolerable Upper Dietary Intake level (UL); Not available (-); The EDI and THQ for *Raphanus sativus*, *Allium cepa* and *Daucus carota* was not determined because poor data on consumption amount.

Food and Nutrition Board, Institute of Medicine (2004). Dietary reference intakes: Tolerable Upper Intake Levels, Elements. National Academy of Sciences; WHO (1993).
significant positive relationship existed for Cd vs Hg (p < 0.05), Zn vs Cd (p < 0.05), Zn vs Hg (p < 0.05), Ni vs Cd (p < 0.01), Ni vs Hg (p < 0.05), and Ni vs Zn (p < 0.05) at the different soil sites. The FCI revealed higher positive factor scores at soil sample site S6 and a relatively low positive score at S2 and S4 (Figures S2b). The second factor (FC2) and the third factor (FC3) showed a high positive loading for only Fe and Cr and accounted for 20.96 % and 19.44 % of the total variance with high factor score for soil S6 and also S3, S5, S7, S8, and S9, respectively. This is suggestive of a predominant crustal material source for Fe and a fertilizer source for Cr in the soil samples. The relationship between Cr or Fe with the remaining metal species was not significant (p > 0.05). The quantitative contribution of the three factors to the heavy metal mass (Z) at each soil sample site was achieved by applying multiple regression analysis on the absolute principal component score. A model equation from the regressed factors is defined by the equation:

\[ Z = 0.086FC1 + 0.954FC2 - 0.244FC3; \quad R^2 = 0.976, \quad p < 0.05 \]  

(8)

The percentage contribution to the mean of the three factors were 6.7 %, 74.3 %, and 19 % for the FC1, FC2, and FC3, respectively. The distribution of the three sources for metals in each sediment as shown in Figure 2 is consistent with the observed concentrations measured in the soil.

3.9. Accumulation and behaviour of heavy metal species in soil and vegetable

The bioconcentration factor (BCF) of Fe, Zn, Cr, Ni, Hg, and Cd in the different vegetable species is shown in Table 5. The BCF for the heavy metals in the leafy vegetables and non-leafy vegetables decreased in the order of Cd (5.85) > Hg (3.83) > Fe (0.31) > Zn (0.11) > Cr (0.82) > Ni (0) and Hg (2.46) > Zn (0.12) > Fe (0.1) > Cd (0) = Cr (0) = Ni (0), respectively. In general, the trend of metal uptake by the crops is consistent with the data reported by McBride (2003). The capacity of leafy vegetables and non-leafy vegetables to accumulate Fe, Cd, Zn, Cr, Ni, and Hg decreased in the order of Amaranthus spinosus > Corchorus olitorius > Brassica oleracea > Brassica rapa and Daucus carota > Raphanus sativus > Allium cepa ~ Capsicum annum. Pearson correlation between the metal species, and the controlling factors pH, CEC, and SOM in the soil was not significant (p > 0.05). This phenomenon might be due to the non-equilibrium state of the interaction between the metals, and the pH, SOM, CEC, % sand, % clay, and % silt parameters in soil at the time of sampling. Thus, the variation in uptake of the metals can best be explained from vegetable species, metal species, phytotoxic nature of metal, and their interactions (McBride, 2003). The high BCF for Cd and Hg in only Amaranthus spinosus (4.04 and 2.58), Corchorus olitorius (1.81 and 1.09) and Daucus carota (Hg = 2.06), indicate that the vegetables may have special mechanism to absorb and translocate the metal species to the leaves and root, respectively. The high uptake of Hg contrasts with Hamilton et al. (2007) who indicated that Hg is poorly bioavailable and soluble in soil. Although Fe is less soluble than Zn and Ni, its uptake is regulated as it is essential to the growth of the vegetable crops. The low BCF for Cr and Ni in all vegetable species may be due to their toxicity of the crops. The poor uptake of Cr is consistent with Hamilton et al. (2007) who indicated that at neutral pH, Cr sorbs strongly to soil and is bioavailable for plant uptake.

3.10. Human health risk assessment

The estimated daily intake (EDI) and the non-carcinogenic risk (HI and HQ) of the heavy metals in the leafy and non-leafy vegetables to adult human population in the urban community is presented in Table 6. Based on intake amounts of the edible portions of the exotic and the indigenous vegetables, the EDI of heavy metals in the leaf of Amaranthus spinosus, Brassica oleracea, Brassica rapa, Corchorus olitorius, and Lactuca sativa, and fruit of Capsicum annum through oral ingestion by an adult male or female (≥30 yrs, 70.42 kg) in the urban community was compared to the tolerable upper dietary intake level (UL) of the Food and Nutrition Board of the Academy of science, USA (Food and Nutrition Board, 2001) and the Joint FAO/WHO Expert Committee on Food Additives (JECFA, 2011). The EDI of the heavy metals decreased in the order of Fe > Zn > Hg > Cr > Ni in the exotic and indigenous leafy vegetables. For the vegetables the EDI of the metals decreased in the order of Amaranthus spinosus > Corchorus olitorius > Brassica oleracea > Lactuca sativa > Brassica rapa > Capsicum annum. For adults, the EDI (μg/day) of Cd (1.60 × 10^-4 to 1.05), Hg (2.02 × 10^-2 to 7.50), Fe (2.58 × 10^-7 to 2.07 × 10^-6), Cr (2.53 × 10^-4 to 2.40 × 10^-4), Ni (2.53 × 10^-4 to 2.86 × 10^-5), and Zn (2.16 × 10^-3 to 11.53) in different edible portions of the vegetables were lower than UL values of 65, 46, 45000, 10000, and 40000 μg/day, respectively (Food and Nutrition Board, 2001). The EDI of each metal species in the vegetables was within respective Recommended Daily Allowance (RDA) (Food and Nutrition Board, 2001; JECFA, 2011). The target hazard quotient of Cd (5.38 × 10^-6 to 6.08 × 10^-4), Zn (4.64 × 10^-5 to 1.51 × 10^-5), Fe (3.64 × 10^-4 to 1.16 × 10^-4), Cr (5.08 × 10^-7 to 3.12 × 10^-6), and Ni (7.62 × 10^-8 to 4.68 × 10^-7) indicates a low adverse risk to adult male or female resident who consume the indigenous and exotic leafy vegetables and fruit vegetables. Cumulative exposure (HI) decreased in the order of Corchorus olitorius (0.158) > Amaranthus spinosus (0.121) > Capsicum annum (0.029) > Brassica rapa (0.016) > Lactuca sativa (0.014) > Brassica oleracea (0.013).

4. Discussion

Generally, the soil was of good quality since CEC, TOC, pH, and heavy metal concentration did not exceed the FAO guideline limits and data reported for soils from other urban farms locally and in some countries. The concentration Cd, Zn, Cr, Ni, and Hg in the soils were lower than in soils from urban vegetable farms in Accra of Ghana (Akoto et al., 2015), Varanasi of India (Kumar et al., 2007), Harare of Zimbabwe (Muchuweti et al., 2006), and Dabaochan of South China (Zhuang et al., 2009).

The irrigation quality indicators for the MWD stream and the groundwater revealed a moderate to desirable water quality sources. However, the exceedance of Hg and E-coli load in the MWD stream and 20 % of the groundwater source over the FAO safe limit poses a health risk to consumers of the raw vegetables. Irrigation water sources, such as W3 may cause nutritional imbalance and ionic toxicity in the vegetable crops. Apart from Cd, the mean concentration of Ni, Zn, Fe, and Cr, in the irrigation water sources were lower than reported in urban vegetable farms in Mardan of Pakistan (Ni: 0.040; Zn: 0.360; Cr: 0.050; Fe: 2.090 μg/L) (Kumar et al., 2007) and in Varanasi, India (Ni: 0.37; Zn: 1.25; Cr: 0.82 mg/L) (Kumar et al., 2007).

The heavy metals preferentially accumulated in the edible parts of the Corchorus olitorious, Amaranthus spinosus, and Daucus carota than in Brassica rapa, Brassica oleracea, Lactuca sativa, Capsicum annum and Allium cepa. The differences in heavy metal uptake by the vegetables can be attributed to the mobility and bioavailability of the metals in the soil medium, as well as the toxicity and mechanism involved in the translocation of the metals in the crop (Hamilton et al., 2007). The poor uptake of Zn in the vegetables, may be due to the relatively alkaline nature of the soil which stimulates adsorption of Zn onto cation exchange sites of metal oxides and clay minerals, lowering the bioavailability of Zn in soil solution for uptake by the vegetables (Gupta et al., 2016). The high accumulation of Fe in the leaves of the Corchorus olitorious (27.585 mg/kg fw), Amaranthus spinosus (36.178 mg/kg fw), Lactuca sativa (16.520 mg/kg fw) than in the edible parts of the non-leafy vegetables (3.617–13.695 mg/kg fw) confirmed the leaf as important Fe sinks for leafy crops as suggested by Connorton et al. (2017). The poor accumulation of Fe edible parts of the non-leafy vegetables corroborates the findings of Connorton et al. (2017), who revealed that, in spite of Fe being abundant in the soil and essential for plant growth, its poor solubility as well as the chelated oxidized form (Fe²⁺ and Fe³⁺) render Fe inaccessible to the roots (Connorton et al., 2017). The preferential
accumulation of Hg and Cd in *Amaranthus spinosus* and *Corchorus olitorious* (BCF > 1) is consistent with the results obtained by Gary et al. (2014) who reported high uptake of heavy metals by leafy vegetables compared to root vegetables in Harayan State, India. Muchuweti et al. (2006) revealed that leafy vegetables have high translocation; high transpiration rate, fast growth rate and are prone to physical contamination from soil dust and splashed water that settles on the leaves. The presence of Cd in all vegetable species is indicative of its high bioavailability in the soil medium (Hamilton et al., 2007). Chang et al. (2014) found that the BCF of Cd was thirty times higher than Hg in vegetables produced from farms around the Pearl River in Guangdong Province, China. Hamilton et al. (2007) indicated that Cd$^{2+}$ is highly soluble in soil solution. Moreover, Cd$^{2+}$ conveniently competes with divalent elements, Ca$^{2+}$, Mg$^{2+}$, and Fe$^{2+}$ while been transported across the membrane and the binding sites of cation transporters in the crop. Singh et al. (2012) revealed that vegetable crops do not have specific uptake mechanism for Cr uptake.

The mean concentration of Zn in *Brassica oleracea*, *Brassica rapa*, *Capsicum annuum*, *Raphanus sativus* and *Daucus carota* was two-hundred times lower than reported in China (5.17 mg/kg fw) (Zhuang et al., 2009; Huang et al., 2013), Portugal (44.8 mg/kg fw) (Alvarenga et al., 2014), and Germany (23.3–122.8 mg/kg fw) (Samuel et al., 2012). Similarly, the concentration of Zn in *Allium cepa* (11.4–25.5 mg/kg) reported by Amin et al. (2013) in Mardan, Pakistan was higher than reported in this study. The concentration of Cd in edible parts of vegetable crops was similar to Cd quantified in *Brassica oleracea* (0.01–0.32 mg/kg fw), *Brassica rapa* (0.10–0.17 mg/kg fw), *Lactuca sativa* (0.02–0.33 mg/kg fw), *Capsicum annuum* (0.05–0.07 mg/kg fw), *Raphanus sativus* (0.04–0.13 mg/kg fw) and *Daucus carota* (0.14–0.38 mg/kg fw) cultivated in urban farms near Daboshan mine in South China. Locally, concentration of Cr in the vegetables was lower than reported by Akoto et al. (2015) for *Lactuca sativus* (5.2 mg/kg fw and 7.0 mg/kg fw converted at 90% moisture) irrigated with urban wastewater in Accra. The concentration of Fe in *Allium cepa* (6.15–26.5 mg/kg) produced in Mardan, Pakistan (Amin et al., 2013) was lower than reported in this study. Nickel in the vegetables was lower compared to Ni (14.5 and 15.7 mg/kg fw at 90% moisture level) in *Lactuca sativus* cultivated in Accra (Akoto et al., 2015).

The heavy metals in the vegetables appeared to originate primarily from mixed household waste with fertilizers, fertilizers (mainly organic manure from poultry, and chemical fertilizers) (Lente et al., 2012), and crustal materials. Contributions from pedogenic weathering of minerals, manure from poultry, and chemical fertilizers) (Lente et al., 2012) and Alloway (2013) showed that fertilizers such as phosphate, NPK, copper sulphate, iron sulphate, and animal manure commonly introduce Hg, Cd, Ni, Zn, and Cd into the soil.

The low EDI of the Cd, Fe, Cr, Hg, Ni, and Zn in the indigenous leafy vegetables (*Amaranthus spinosus* and *Corchorus olitorious*), exotic leafy vegetables (*Brassica rapa*, *Brassica oleracea*, and *Lactuca sativa*) and *Capsicum annuum* suggests a lower adverse health effect to the adult male and female population that frequently consume the vegetables. The THQ of the metals in the different vegetables being <1 is indicative of a low human health risk. The potential health risk of Hg through intake of the vegetable is far lower than reported for vegetables (HI = 1.01) from an urban farm in Zhejiang Province of China (Pan et al., 2014). The cumulative dietary exposure to metals (HI) indicates that consumption of *Amaranthus spinosus*, *capsicum annuum*, *Corchorus olitorious* *Brassica oleracea*, *Lactuca sativus*, *Brassica rapa*, and *Capsicum annuum* present a minimal health risk to the human in their lifetime. Nevertheless, the fraction of heavy metal which bioaccumulate in the body tissues and organs may eventually be detrimental to human health (Horiguchi et al., 2004).

5. Conclusion

Despite the soils being moderately enriched with Fe, Zn, Hg, Cr, and Ni, primarily from mixed household waste with fertilizer, fertilizer and crustal materials, the uptake and accumulation of the metals in the edible tissues differed for indigenous and exotic leafy vegetables and non-leafy vegetables. The mean concentration of heavy metals in the edible portions of the vegetables decreased in the order Fe > Zn > Hg > Cr = Ni. For the individual vegetables, the bioaccumulation of the metals in the edible parts decreased in the order of *Amaranthus spinosus* > *Corchorus olitorious* > *Daucus carota* > *Raphanus sativus* > *Lactuca sativa* > *Brassica oleracea* > *Allium cepa* = *capsicum annuum* > *Brassica rapa*. Heavy metal load in edible parts of the vegetables was lower than recommended safe limits by FAO, as well as values reported for vegetables cultivated from other urban vegetable farms in Ethiopia, Ghana, India, Pakistan, Germany, China, and Portugal. Irrigation water sources with elevated concentration of Cd, Hg, and E. coli present a health risk to human that consume the vegetables. At the worst-case scenario, the EDI levels, THQ, and the HI of heavy metals in the different vegetables showed minimal adverse effect for the adult human population that consume *Capsicum annuum*, *Amaranthus spinosus*, *Corchorus olitorious*, *Lactus sativa*, *Brassica oleracea*, and *Brassica rapa*. More importantly, *Amaranthus spinosus* and *Corchorus olitorious* must be planted among the exotic vegetable crops to mitigate the elevated concentration of Cd and Hg in the soil. Finally, the study provides valuable information essential in developing agric- horticultural policy and local regulatory guidelines to mitigate toxic metal load in edible vegetables produced from urban farmlands nationwide and in sub-Saharan Africa.

Declarations

Author contribution statement

Andrews Obeng Affum: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

Shiloh Dede Osae: Contributed reagents, materials, analysis tools or data.

Edward Ebow Kwaansa-Ansah, Michael K Miyittah: Conceived and designed the experiments.

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Data availability statement

Data will be made available on request.

Declaration of interests statement

The authors declare no conflict of interest.

Additional information

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