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Original Article

Concentration of Heavy Metals in Water, Soil, and Vegetables Irrigated with Industrial Wastewater in Oromia Special Zone Surrounding Finfinne, Ethiopia

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Abstract

Background: This study sought to determine the spatial distribution of heavy metal concentrations in water, soil, and vegetables that were irrigated with industrial wastewater in Ethiopia's Oromia Special Zone. We aimed to make recommendations for corrective actions that would reduce the negative environmental effects of untreated waste.

Methods: Five sampling sites were meticulously chosen within the Sululta, Laga Tafo Laga Dadi, Galan, Sabata, and Burayu regions, taking into account the varying stressors across the upper stream, middle stream, and downstream segments of the rivers. These sampling sites were strategically selected to capture a comprehensive understanding of heavy metal aggregation within the area. The sampling encompassed the collection and analysis of 25 water and wastewater samples, 13 soil samples, and 8 vegetable samples. The distribution of sampling efforts was tailored to reflect the availability of irrigation sites within the respective areas, resulting in a harmonized amalgamation of 5, 3, and 1 samples for water and wastewater, soil, and vegetable matrices, respectively.

Results: The lettuce in the Gelan area had the greatest quantity of lead, suggesting higher cancer risk. The sites of Gelan and Burayu had significant levels of chromium contamination in their lettuce, followed by their cabbage, according to the WHO/FAO and USEPA. As a result, those who eat vegetables that have high levels of heavy metal contamination may be at risk for developing cancer.

Conclusion: The concerned institutions and stakeholders must work to mitigate the high concentration of heavy metals in water, soil, and vegetables by the installation of treatment plant.

Keywords: Bioaccumulation, Carcinogenic, Detection, Heavy metal, Pollution

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Introduction

Environmental contamination caused by heavy metals has significantly impacted ecosystem functions and biodiversity services.[1,2] Industrialization and urbanization have also contributed to environmental degradation through the release of heavy metals.[3] Heavy metals are non-biodegradable and pose significant threats to the environment, food chains, and the health of both animals and humans.[4,5] Exposure to heavy metal pollution poses a serious threat to living organisms.[6] In developing countries, heavy metal pollution of agricultural land has resulted in severely adverse ecological conditions in recent decades.[7] Irrigation with industrial wastewater has led to significant heavy metal toxicity in soils and plants. The assimilation of trace elements in agriculture through contaminated substances has resulted in their accumulation in edible plant parts.[8] Therefore, the concern over trace element pollution in irrigation water, soil, and vegetables has increasingly posed a serious threat to biodiversity and ecosystems as a consequence of industrialization and urbanization

In developing countries, industries such as metal smelting, beverages, textiles, chemicals, paints, paper, cement, plastic, and tanneries have been established near residential areas and in close proximity to riverbanks. The discharge of industrial wastewater has led to significant absorption of trace metals, resulting in serious ecological disturbances.[1,9-12] In the past few decades, various types of industries in the Oromia Special Zone Surrounding Finfinne (OSZSF) towns have been discharging untreated industrial wastes into nearby rivers. This practice has resulted in heavy metal pollution in irrigation water. As a consequence of irrigating vegetables with this wastewater, the concentration of heavy metals in the edible parts of plants becomes significantly elevated.[8] Therefore, this research aimed to assess the levels of trace metals in water, soil, and plants irrigated with industrial effluents in Ethiopia's OSZSF (including Sululta, Laga Tafo, Galan, Sabata, and Burayu). The goal was to formulate recommendations for corrective actions to address the pollution resulting from industrial activities in the area.

Materials and Methods

Study Area

The OSZSF comprises eight towns: Burayu, Dukam, Galan, Holota, Laga Tafo, Sabata, Sandafa, and Sululta. For this study, five towns—Burayu, Sululta, Laga Tafo, Galan, and Sabata—were purposively selected. These towns were chosen due to their rapidly expanding industries that support economic development and their quickly growing populations, which have led to increased environmental stressors.

The altitude of the study area ranges from 1500 to 3443 meters above sea level. The OSZSF is situated between latitudes 8°34'25" to 9°32'41"N and longitudes 38°25'50" to 39°07'53"E. The temperature in the area varies from 10 °C to 26 °C, and the annual rainfall ranges from 1043.87 to 1316.6 mm.

Sampling Sites

The analysis of the spatial distribution and impact of untreated industrial waste disposal on the environment revealed significant heavy metal pollution in the water, soil, and vegetables in the selected study towns of OSZSF. Five sampling sites—Sululta, Laga Tafo Laga Dadi, Galan, Sabata, and Burayu—were selected from the eight main towns based on pollution levels along the rivers' upper, middle, and downstream sections (Table S1, Supplementary file 1). Both primary and secondary data were utilized in the study. Primary data were collected through field observations and laboratory analyses of heavy metals in water, soil, and vegetable samples from various sites with recorded coordinates. Secondary data were obtained from journals, articles, and research papers.

Sampling Design

The water sampling was taken following the procedures of the American Public Health Association.[13] Furthermore, the vegetable and soil samplings were conducted based on the methodologies of Jones et al[14] and Tan,[15] respectively. Composite samples were collected from streams, land, and vegetable sites. The coordinate points of water, soil, and vegetable sampling locations in Sululta, Laga Tafo, Galan, Sabata, and Burayu were recorded using GPS and plotted using ArcGIS (Figure 1).

Water, soil, and vegetable samples were collected to analyze the aggregate distribution of heavy metals, specifically targeting copper (Cu), lead (Pb), zinc (Zn), nickel (Ni), manganese (Mn), chromium (Cr), and cadmium (Cd), from January 2022 to March 2023. Soil and vegetable samples were collected in clean polyethylene plastic containers and transported in an icebox at 4 °C to the Oromia Environmental Laboratory Center of the Oromia Environmental Protection Authority (OEPA) in Burayu town. The samples were delivered within eight hours, with full information labeled on the test material for traceability.

Sampling Method

Vegetable samples were collected from various sites using polyethylene plastic bags cleaned with HNO_3 and purified water before sample collection. The samples included cabbage, lettuce, and Swiss chard irrigated with wastewater, which constituted the treatment group, while garden cabbage served as the control group. Soil samples were collected by plowing the soil with a shovel. One kilogram of surface soil was taken from a depth of 20 cm in rain-fed farms, irrigated areas, and wetlands. Water samples were collected from upstream (control group) and middle and downstream sites (treatment group), where industrial waste is in immediate contact.

The sample collection was conducted daily from 7:00 to 11:00 A.M. between January 2022 and March 2023. Samples were analyzed for the aggregation of heavy metals (Cu, Pb, Zn, Ni, Mn, Cr, and Cd). A total of 25 water and wastewater samples, 13 soil samples, and 8 vegetable samples were collected, corresponding to the five, three, and one irrigation sites available in the area, respectively. Water samples were gathered from Sululta, Laga Tafo, Galan, Sabata, and Burayu using clean polyethylene plastic bags, which were rinsed three times with purified water before sample collection, and sealed with nitric acid. The concentration of heavy metals in water and wastewater was analyzed using a ultra-violet (UA) atomic absorption spectrophotometer (AAS) (NovAA 400P AAS) at the Oromia Environmental Laboratory Center of the OEPA.

Digestion Processes for Soil Samples

The collected soil samples were positioned, sieved, and oven-dried at 105 °C for 24 hours. Then, 1000 g of dried soil was transferred into a 100 mL Erlenmeyer flask and moistened with 2-3 mL of water. Subsequently, 7.5 mL of concentrated hydrochloric acid and 2.5 mL of concentrated nitric acid were added under a fume hood. The flask was covered with a watch glass and left overnight at room temperature under the fume hood. The soil mixture was then gently heated for 2 hours on a hot plate at 100 °C. After cooling to room temperature, the sample was washed with 30 mL of water. The extract was filtered through acid-resistant filter paper into a 100 mL volumetric flask. The digestion container and the residue

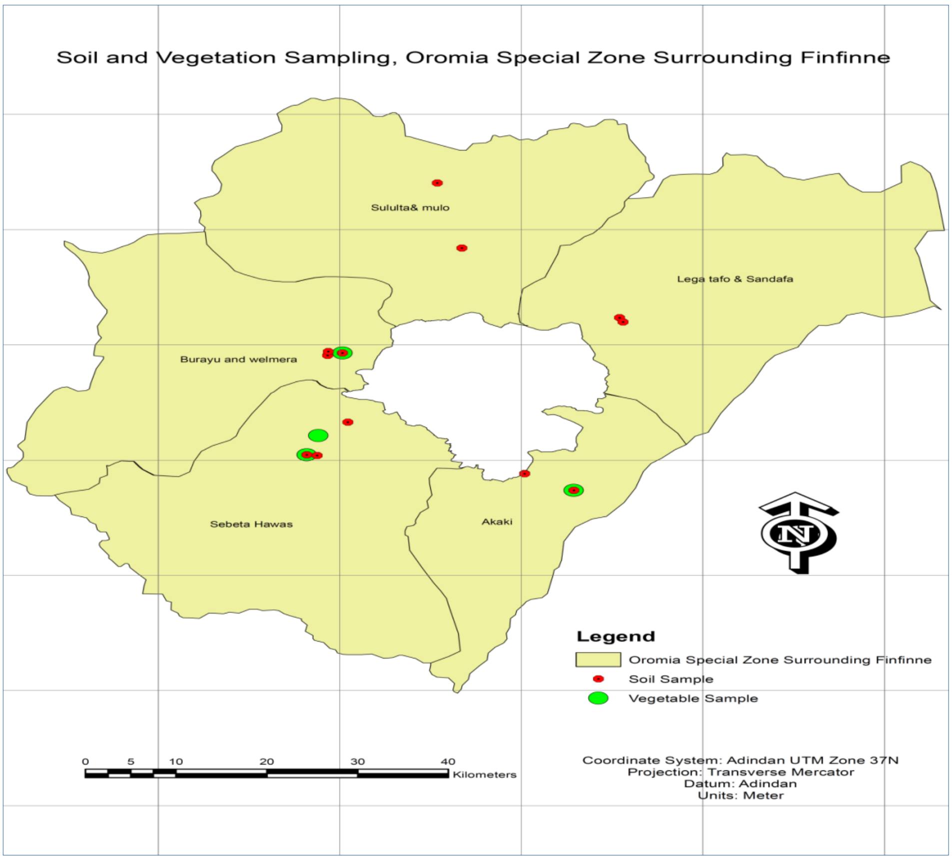

Figure 1. Geographical Location of Soil and Vegetation Sampling Sites

on the filter paper were repeatedly washed with small amounts of warm (±50 °C) 2M HNO_3. The solution was allowed to settle and then diluted to volume with the 2M nitric acid solution, including a blank sample. The flask was boiled in a water bath at 100 °C for two hours. Finally, the prepared soil solutions were analyzed for heavy metal concentrations using a UV AAS.

Digestion Processes for Vegetable Samples

The vegetable samples were digested using nitric acid (HNO_3) to extract heavy metals (Cu, Pb, Zn, Ni, Cr, and Cd). Approximately 1000 g of the vegetable samples was dried at 105 °C and transferred into a porcelain crucible. The samples were pre-ignited on a hot plate or in a muffle furnace at 200 °C, with adequate ventilation, until fully mineralized, typically for at least 2 hours at 450 °C. The crucibles were removed from the furnace using tongs and placed on a hot plate. Subsequently, 5 mL of 6M HNO_3 was added and evaporated by gentle boiling until only about 1 mL remained. Then, 5 mL of 3M HNO_3 was added, and the solution was reheated for 30 minutes. After cooling, the solution was transferred into a 100 mL volumetric flask, and the volume was adjusted with a glass rod. The crucible and glass rod were rinsed several times with 1% HNO_3, and the residues were collected on a filter paper. The filtrate was cooled, diluted to 100 mL with water, and the flask was stoppered for further analysis.

The concentration of heavy metals from the solutions prepared from the vegetable samples was analyzed using an AAS (Analytic Jena, Germany, model NOVAA 400).[10] The analytical instrument was equipped with a 100 mm burner, HCL lamp, and single-beam optical configuration. It utilized the flame technique with a detection limit of 10^{-4} mg/L. The quantification limit was determined from samples containing a low concentration of analyte near the expected limit of quantification (LOQ) for the NOVAA 400, which is 10^{-4} mg/L or slightly above its baseline. The validity of the soil analysis method was assessed based on the guidelines provided by the Certified Manual of the Plant Analytical Laboratories Network of Ethiopia (SPALNE) and the National Soil Research Laboratory (NSRL). Quality control for the analysis included blank samples with zero concentration readings, ensuring that all measured values were corrected to zero, as well as national standard solutions of heavy metals used for calibration. The concentration of heavy metals in water, soil, and vegetable samples was determined using flame atomic absorption spectrophotometry with a detection limit of 1 part per billion. The method detection limit (MDL) and LOQ were established following the standard operating procedures for measuring heavy metals with the environmental laboratory version of the AAS, as detailed in the laboratory instrument operating manual developed by the OEPA.

Data Calculation

Bioaccumulation Factor

The bioaccumulation factor is the proportion of heavy metal aggregation in part of a plant sample to heavy metal aggregation in a soil sample.[16] Thus, transfer of heavy metals from soil to plant was calculated as equation 1[17]:

$$\mathrm{BCF} = \frac{\mathrm{C\ plant}}{\mathrm{C\ soil}} \tag{1}$$

where C_{plant} is the amount of heavy metal in part of the plant and C_{soil} is the amount of heavy metal in soil. A bioconcentration factor (BCF) greater than one indicates that a plant accumulates or absorbs heavy metals from its environment.

Hazard Analysis

Predictable Ingestion of Toxic Substances

Estimated daily intake (EDI) of trace metals is computed by multiplying their respective average concentrations in vegetable samples by the weight of the vegetables consumed, divided by a person's body weight. According to Chen et al[18] the EDI values of vegetables can be calculated using equation 2:

$$EDI = \frac{Ef \times ED \times FIR \times CM \times Cf}{BW \times TA} \tag{2}$$

Where, *Ef* is exposure occurrence (365 days/year), *ED* is the exposure interval (65 years), which is equal to an average life time.[19] *FIR* is the vegetable consumption (cabbage, lettuce, and Swiss chard for each 240 g/individual/day which was introduced by the World Health Organization (WHO) for low fruit and vegetable intake, C_M is the heavy metal concentration (mg/kg dry weight), C_f is the concentration conversion factor for fresh vegetable weight to dry weight (0.085).[8,20,21] BW is reference body weight for adult, which is 70 kg; *TA* is the average exposure time (65 years × 365 days).[19]

Target Hazard Quotient

a. Non-carcinogenic and Carcinogenic Risk Assessment

The non-carcinogenic risks associated with heavy metals in edible vegetables were assessed using the target hazard quotient (THQ). The THQ values for the consumption of contaminated vegetables were calculated using equation 3[18,22,3,23]:

$$THQ = \frac{\mathrm{EDI}}{\mathrm{RfD}} \tag{3}$$

EDI represents the estimated daily intake of heavy metals, measured in mg/day/kg body weight. RfD denotes the reference dose (mg/kg/day) values for each heavy metal. A THQ value of less than 1 indicates safety from non-carcinogenic effects, whereas a THQ value greater than 1 suggests a potential risk of non-carcinogenic effects.[18,24]

b. Hazard Index

The health risk of heavy metals hoard vegetables are analyzed using the risk indicator.[18,21-23,25-28] Thus, the hazard index (HI) of heavy metals in vegetables was computed using equation 4[29,30]:

$$HI = \sum_{n=1}^{i} \mathrm{THQn}; \mathrm{i} = 1, 2, 3, \ldots,\ \mathrm{n} \tag{4}$$

Where: HI < 1 implies no likelihood or possibility of adverse health impacts. An HI value greater than 10 indicates a severe and persistent health hazard.[29,30]

c. Carcinogenic Risk

According to the study by Kamunda et al[26], the target cancer risk (TCR) of heavy metal intake (Pb, Ni, Cr, and Cd) evaluates the carcinogenic risks based on the exposure dose. Sharma *et al*[16] stated that the Carcinogenic Risk (CR) is an individual's health risk resulting from the intake of carcinogenic heavy metals which can be computed using equations 5, 6.

$$\mathrm{CR} = \mathrm{EDI} \times \mathrm{CPSo} \tag{5}$$

$$TCR = \sum_{n=1}^{i} \mathrm{CR}; \mathrm{i} = 1, 2, 3, \ldots, \mathrm{n} \tag{6}$$

Where CR represents the cancer risk over a lifetime due to individual intake of heavy metals, EDI denotes the estimated every day intake of heavy metals (mg/day/kg body weight), carcinogenic potency slope (CPSo) refers to the oral cancer slope factor (mg/kg/day)-1, and n is the number of heavy metals considered for cancer risk calculation. Some studies have reported the carcinogenic potency slope factors for Pb, Ni, Cr, and Cd as 0.0085, 1.7, 0.5, and 0.38, respectively.[31]

Statistics Study

Trace metal concentrations in streams, soils, and vegetables were statistically analyzed using one-way ANOVA and descriptive statistics to assess variances across different sites. International standards from WHO, FAO, US EPA, and others were employed to evaluate water, soil, and vegetable quality. HI, cancer target risk, and Pearson correlation coefficient (PCC) were calculated using STATA version 14.

Results and Discussion

Validation of the Method

The validity of the heavy metal concentration analysis was assessed using the limit of quantification (LOQ = 3 × SD) and the method detection limits (MDL = 10 × SD). The values of LOQ and MDL are presented in Table 1. An UA AAS was employed to obtain valid results for the samples with zero blank calibration. The precision and accuracy of the heavy metal concentration results are detailed in Table 1. Accordingly, the proportions of heavy metals in water, soil, and vegetables, along with the potential health hazards, were calculated using the relative standard deviations (% RSD) for samples collected from the study area.

Amount of Trace Metals in Land and Plant

The findings on heavy metal concentrations in soil from different land-use areas revealed that the maximum

Table 1. The Precision and Accuracy of the Heavy Metal Measurements

Element	Wave Length (nm)	Lamp Current (mA)	MDL (mg/L)	LOQ (mg/L)	% RSD	R^2
Mn	279.5	5.0	0.0054	0.018	0.1-4.4	0.9980
Ni	232.0	3.0	0.0072	0.024	1.2-9.0	0.9978
Cu	324.8	2	0.0051	0.0170	1.3-7.8	0.9982
Zn	213.9	0.2	0.00039	0.0013	0.1-8.6	0.99791
Pb	283.3	2.0	0.0096	0.032	0.1-8.7	0.99813
Cr	357.9	4.0	0.02946	0.0962	0.5-9.7	0.99819
Cd	228	2.0	0.0042	0.0140	1.3-10.2	0.99801

concentration of Mn (27.48 mg/kg) was found in the wetland, followed by irrigated vegetables at the Gelan site, indicating no significant difference ($P=0.06$). The minimum concentration of Zn (0.00054 mg/kg) was observed at the Sabata farmland site (Table 2).

The variability in heavy metal concentrations in vegetables across various locations was detected spatially (Table 2). The highest absorption of heavy metals was recorded for Pb (40.14 µg/kg) in lettuce from the irrigated area at Gelan, followed by Swiss chard (34.91 µg/kg) at the Burayu site, which was statistically significant (P = 4.13E-20). Lettuce (29.13 µg/kg) and cabbage (27.21 µg/kg) from the Sabata sites also showed statistically significant concentrations (P = 1.26E-28). The lowest mean concentration of heavy metals was found for Ni (0.01 µg/kg) in Swiss chard at the Sabata site.

Relationship Analysis

A linear relationship was employed to analyze the association between concentrations of heavy metals in soil across different spatial locations (Table S2). Specifically, the concentration of Cd displayed a strong positive correlation with Cu, while the concentration of Zn showed a strong positive correlation with Ni. Conversely, the concentration of Mn exhibited a slight correlation with the concentrations of Pb, Cr, and Cd. Similarly, the concentration of Zn showed a slight correlation with the concentrations of Pb and Cd, and the aggregation of Cu with Ni, Mn, and Zn revealed a small correlation as well.

The concentration of Cd with Pb, the concentration of Mn with Pb and Cr, and the concentration of Zn with other heavy metals, except for Ni, showed a strong positive relationship with the concentrations of other elements.

Concentration of Heavy Metal in Water

The mean concentrations of trace metals in stream water at various locations are presented in Table 3. Different aggregations of heavy metals in water samples across these locations were observed (Table 3). The highest concentrations of heavy metals were detected for Mn (10.65, 14.76, and 16.17 mg/L) at the SbS5, SbS3, and SbS4 sampling sites, respectively, for the Sabata site, which was statistically significant (P = 2.8968E-22). This was followed by Mn concentrations (5.83 and 11.77 mg/L) at the GS3 and GS1 sites for the Gelan site, which were also statistically significant (P = 3.26416E-27). The lowest mean concentration of heavy metal was recorded for Mn (0.0009 mg/L) at the LLD5 site in the Laga Tafo Laga Dadi site, which was statistically significant (P = 8.89847E-10).

Pearson Correlation Coefficient of the Metal Concentration in Water

Pearson correlation was used to analyze the relationship between concentrations of heavy metals in water at different sites spatially (Table S3). Ni showed a strong positive correlation with the concentration of Cd (Table S3). Additionally, the concentration of Zn with Cr, Cu with Zn, Cd with Cr and Cu, Ni with Cr and Cu, and Pb with Cr displayed a slight negative correlation. Conversely, the concentrations of Mn with Zn, Cd, Ni, and Pb revealed a slight negative correlation. On the other hand, the concentrations of Cu with Cr, Cd with Zn, Ni with Zn, Pb with Zn, Cu, Cd, and Ni, and Mn with Cr and Cu showed a slight positive correlation.

Bioaccumulation Factor

The bioaccumulation factors for different heavy metals at various sites are presented in Table 4. The maximum Bioaccumulation Factor was recorded as 95.000 for Zn in lettuce at the Burayu site, followed by 45.870 and 46.304 for Zn in cabbage and lettuce, respectively, at the Gelan site. Conversely, the minimum bioaccumulation factor was recorded as 0.000 for Pb in garden cabbage, Swiss chard, and cabbage, and for Mn in garden cabbage at the Sabata and Burayu sites, respectively.

Predicated Everyday Ingestion

EDI values of trace metals at various sites are presented in Table S4. The highest EDI values were recorded as 0.0242 for children and 0.0222 for adults for Pb in lettuce at the Gelan site. Additionally, EDI values of 0.0211 for children and 0.0193 for adults were recorded for Pb in lettuce at the Burayu site, followed by 0.0188 for Pb in cabbage for both children and adults at the Gelan site. The lowest EDI values were recorded at different sites with varying concentrations of heavy metals (Table S4).

Target Risk Ratio

The THQ is calculated using equation 3. For a child consuming lettuce exposed to high levels of Pb at the Gelan site, the THQ was found to be 6.0517. Similarly, adults and children consuming lettuce exposed to Pb at

Table 2. Result of Trace Metals (Mean ± SD) by the Study Sites

Study Site	Vegetable	Parameter						
		Pb (µg/kg)	**Cr (mg/kg)**	**Cd(mg/kg)**	**Ni (mg/kg)**	**Mn (mg/kg)**	**Zn (mg/kg)**	**Cu (mg/kg)**
Sabata	Cabbage	27.21 ± 0.009	0.211 ± 0.000	0.19 ± 0.002	0.034 ± 0.001	0.134 ± 0.012	0.015 ± 0.007	0.113 ± 0.007
	Garden Cabbage	ND	0.217 ± 0.000	0.016 ± 0.005	0.013 ± 0.000	0.013 ± 0.008	0.013 ± 0.008	0.132 ± 0.005
	Swiss Chard	ND	0.275 ± 0.000	0.031 ± 0.009	0.012 ± 0.003	0.034 ± 0.002	0.021 ± 0.009	0.12 ± 0.001
	Lettuce	29.13 ± 0.019	0.297 ± 0.000	0.21 ± 0.000	0.032 ± 0.007	0.212 ± 0.005	0.141 ± 0.003	0.212 ± 0.002
Burayu	Cabbage	ND	0.317 ± 0.000	0.017 ± 0.006	0.013 ± 0.004	0.012 ± 0.002	0.029 ± 0.012	0.172 ± 0.003
	Lettuce	34.91 ± 0.048	0.317 ± 0.000	0.34 ± 0.009	0.042 ± 0.002	0.121 ± 0.002	0.19 ± 0.008	0.121 ± 0.010
Gelan	Lettuce	40.14 ± 0.002	0.316 ± 0.000	0.067 ± 0.0089	0.013 ± 0.007	0.11 ± 0.003	0.211 ± 0.001	0.310 ± 0.007
	Cabbage	34.09 ± 0.023	0.5693 ± 0.000	0.27 ± 0.00153	0.023 ± 0.010	0.34 ± 0.001	0.213 ± 0.006	0.131 ± 0.004
	Soil	**Pb (µg/kg)**	**Cr (mg/kg)**	**Cd(mg/kg)**	**Ni (mg/kg)**	**Mn (mg/kg)**	**Zn (mg/kg)**	**Cu (mg/kg)**
Sabata	Irrigated land	3.91 ± 0.001	10.79 ± 0.006	0.19 ± 0.03	4.88 ± 0.003	ND	0.004 ± 0.006	1.8 ± 0.004
	Wet land	2.76 ± 0.001	2.02 ± 0.004	0.13 ± 0.0008	1.73 ± 0.001	ND	0.003 ± 0.001	0.485 ± 0.001
	Farm land	3.09 ± 0.001	3.27 ± 0.000	0.17 ± 0.002	4.62 ± 0.003	27.04 ± 0.001	0.001 ± 0.004	1.215 ± 0.003
Sululta	Wet land	2.18 ± 0.001	4.612 ± 0.001	0.17 ± 0.004	1.61 ± 0.001	26.82 ± 0.002	0.001 ± 0.005	1.943 ± 0.005
	Farm land	ND	0.559 ± 0.001	0.12 ± 0.0001	4.62 ± 0.001	25.53 ± 0.002	0.004 ± 0.017	0.030 ± 0.002
Laga Tafo	Wet land	4.56 ± 0.004	4.53 ± 0.001	0.22 ± 0.004	8.83 ± 0.001	26.97 ± 0.001	0.004 ± 0.001	1.916 ± 0.004
	Farm land	2.21 ± 0.006	3.73 ± 0.003	0.15 ± 0.004	7.85 ± 0.007	23.52 ± 0.005	0.005 ± 0.009	1.85 ± 0.005
Gelan	Farm land	5.04 ± 0.000	5.128 ± 0.000	0.26 ± 0.0007	9.95 ± 0.007	25.53 ± 0.002	0.005 ± 0.000	1.944 ± 0.006
	Wet land	2.26 ± 0.001	7.56 ± 0.005	0.11 ± 0.0005	14.36 ± 0.01	27.48 ± 0.001	0.031 ± 0.005	0.9802 ± 0.002
	Irrigated land	4.32 ± 0.001	2.53 ± 0.006	0.076 ± 0.0004	10.74 ± 0.006	27.09 ± 0.001	0.005 ± 0.007	0.5141 ± 0.001
Burayu	Farm land	4.45 ± 0.000	5.277 ± 0.000	0.22 ± 0.001	3.78 ± 0.002	ND	0.003 ± 0.004	2.018 ± 0.005
	Irrigate land	2.27 ± 0.001	3.35 ± 0.001	0.14 ± 0.0003	2.89 ± 0.001	23.64 ± 0.015	0.002 ± 0.006	1.189 ± 0.004
	Wet land	3.33 ± 0.000	3.29 ± 0.001	0.18 ± 0.001	4.2 ± 0.001	26.71 ± 0.003	0.003 ± 0.008	1.554 ± 0.004

the Gelan and Sabata sites showed THQ values of 5.5451 and 5.2632, respectively (Table S5).

Carcinogenic Risk Analysis

The TCR is computed according to the method outlined in the study. The result of CR for Pb, Cr, Cd, and Ni in cabbage and lettuce in Sabata sites showed 1×10^{-4} and 1×10^{-4} (Pb), 1×10^{-4} and 1×10^{-4} (Cr) and 0 (Cd and Ni), respectively. The TCR for other vegetables and sites have been presented in Table S6.

The spatial distribution of water, soil, and vegetables was examined to assess the levels of heavy metals, specifically Cr, Cd, Ni, Zn, Mn, and Cu, in the study area. The concentrations of Pb, Cr, Cd, Ni, Zn, Mn, and Cu in samples of soil, surface water, and vegetables irrigated with industrial wastewater were analyzed and compared to WHO/FAO and USEPA guidelines.

The mean concentrations of Cr, Cd, and Cu in cabbage and lettuce at the Sabata, Gelan, and Burayu sites were above the safe limits set by WHO/FAO, which are 0.1–0.2 µg/g for Cr, 0.1 µg/g for Pb, 0.1 µg/g for Cu, 0.1 µg/g for Zn, 0.1 µg/g for Ni, 0.02 µg/g for Cd, and 0.3 µg/g for Mn. Furthermore, the mean concentration of Mn in cabbage at Gelan and Zn in lettuce at the Sabata, Burayu, and Gelan sites exceeded the permissible limits. In contrast, the levels of Ni in all vegetables at all sites and Mn levels, except in cabbage at the Gelan site, were within the maximum permissible limits set by WHO/FAO. Elevated concentrations of Pb pose a cancer risk if daily ingestion surpasses the safe value limit.[32-34] Based on vegetable samples, lettuce (0.316, 0.317, and 0.297 µg/g at Gelan, Burayu, and Sabata sites, respectively) was highly polluted by Cr, followed by cabbage (0.569, 0.317, and 0.211 µg/g at Gelan, Burayu, and Sabata sites, respectively). This pattern was also observed for the mean concentration of Cu in vegetables at all study sites. According to a study by Woldetsadik et al[19] a high concentration of Pb above the allowable limit in green vegetables was found in the Akaki area of Addis Ababa city. The concentration levels of Cr and Cu in plants in the recent study are associated with the effects of industrial wastewater irrigation in the study area.

Ingestion of vegetables with high levels of Pb significantly impacts public health, as Pb's propensity for bioaccumulation in body tissues leads to toxicological outcomes. Pb is a well-known toxic metal that causes kidney dysfunction and neurological harm even at low blood concentrations.[24,29,35]

The mean concentrations of heavy metals Cr, Cd, and Ni in water samples from nearly all sites, as well as Mn at the Sabata and Gelan sites, exceeded the limits set by US EPA guidelines. However, the mean concentrations of the other heavy metals were below the permissible limits established by the US EPA. This indicates that the

Table 3. Trace Metals Concentration (Mean ± S.D.) of Stream Waters by the Study Sites

Places	Trial Site	Heavy Metal						
		Cr (mg/L)	Zn (mg/L)	Cu (mg/L)	Cd (mg/L)	Ni (mg/L)	Pb (mg/L)	Mn (mg/L)
Sabata	SbS1	0.26 ± 0.003	0.09 ± 0.007	0.34 ± 0.001	0.05 ± 0.001	0.03 ± 0.005	0.028 ± 0.016	2.03 ± 0.001
	SbS2	0.19 ± 0.001	0.081 ± 0.002	0.07 ± 0.001	0.02 ± 0.001	0.03 ± 0.001	0.010 ± 0.015	0.90 ± 0.002
	SbS3	0.37 ± 0.001	0.91 ± 0.001	0.15 ± 0.006	0.08 ± 0.005	0.53 ± 0.001	ND	14.76 ± 0.015
	SbS4	0.40 ± 0.006	0.004 ± 0.002	0.09 ± 0.000	0.07 ± 0.001	0.38 ± 0.001	ND	16.17 ± 0.018
	SbS5	0.30 ± 0.008	ND	0.08 ± 0.001	0.03 ± 0.005	0.06 ± 0.001	ND	10.65 ± 0.011
Gelan	GS1	0.34 ± 0.002	ND	0.31 ± 0.003	0.05 ± 0.005	0.09 ± 0.002	0.028 ± 0.061	11.77 ± 0.007
	GS2	0.41 ± 0.001	0.008 ± 0.006	0.69 ± 0.001	0.04 ± 0.001	0.12 ± 0.001	0.50 ± 0.003	0.85 ± 0.001
	GS3	0.54 ± 0.002	0.005 ± 0.001	0.012 ± 0.006	0.08 ± 0.0006	0.21 ± 0.001	ND	5.83 ± 0.007
	GS4	0.47 ± 0.001	0.003 ± 0.01	0.016 ± 0.005	0.05 ± 0.006	0.21 ± 0.000	ND	1.38 ± 0.001
	GS5	0.46 ± 0.001	ND	0.02 ± 0.001	0.04 ± 0.001	0.15 ± 0.001	ND	0.25 ± 0.000
Sululta	SS1	0.004 ± 0.02	0.002 ± 0.008	0.006 ± 0.004	0.09 ± 0.004	0.65 ± 0.000	0.011 ± 0.02	0.008 ± 0.005
	SS2	0.45 ± 0.004	ND	0.009 ± 0.007	0.16 ± 0.002	0.48 ± 0.004	ND	0.013 ± 0.00
	SS3	ND	0.005 ± 0.006	0.007 ± 0.004	0.10 ± 0.001	0.25 ± 0.000	ND	0.023 ± 0.001
	SS4	0.008 ± 0.01	0.004 ± 0.001	0.009 ± 0.002	0.07 ± 0.001	0.24 ± 0.001	ND	0.016 ± 0.008
	SS5	ND	0.056 ± 0.004	ND	0.07 ± 0.002	0.43 ± 0.009	0.008 ± 0.10	0.012 ± 0.001
Laga Tafo	LLD1	0.023 ± 0.05	ND	ND	0.06 ± 0.001	0.36 ± 0.002	0.033 ± 0.05	0.054 ± 0.004
	LLD2	0.004 ± 0.43	0.005 ± 0.003	0.065 ± 0.002	0.10 ± 0.009	0.55 ± 0.001	0.039 ± 0.43	0.017 ± 0.007
	LLD3	0.36 ± 0.001	0.07 ± 0.001	0.029 ± 0.001	0.12 ± 0.004	0.37 ± 0.000	0.47 ± 0.003	0.021 ± 0.005
	LLD4	0.27 ± 0.003	0.14 ± 0.000	0.055 ± 0.004	0.13 ± 0.001	0.43 ± 0.001	ND	0.02 ± 0.003
	LLD5	0.14 ± 0.001	0.039 ± 0.006	ND	0.09 ± 0.001	0.32 ± 0.001	ND	0.001 ± 0.002
Burayu	BuS1	0.027 ± 0.05	0.007 ± 0.006	0.013 ± 0.007	0.13 ± 0.003	0.55 ± 0.000	0.022 ± 0.003	ND
	BuS2	ND	0.002 ± 0.007	0.000 ± 0.000	0.11 ± 0.000	0.46 ± 0.001	0.50 ± 0.001	ND
	BuS3	ND	0.007 ± 0.007	0.004 ± 0.02	0.13 ± 0.001	ND	0.05 ± 0.003	ND
	BuS4	ND	4.31 ± 0.000	0.023 ± 0.001	0.15 ± 0.008	0.52 ± 0.001	0.23 ± 0.004	ND
	BuS5	0.029 ± 0.003	0.005 ± 0.005	ND	0.12 ± 0.001	0.73 ± 0.006	0.25 ± 0.0001	ND

Table 4. Bioaccumulation Factor of the Vegetables by the Study Sites (BCF)

Site	Vegetable	BCF of Trace Metals						
		Pb	Cr	Cd	Ni	Mn	Zn	Cu
Sabata	Cabbage	6.959	0.020	1.000	0.007	1.000	4.167	0.063
	Garden cabbage	0.000	0.066	0.094	0.003	0.000	23.704	0.109
	Swiss chard	0.000	0.025	0.163	0.002	0.254	5.833	0.067
	Lettuce	7.450	0.028	1.105	0.007	1.582	39.167	0.118
Burayu	Cabbage	0.000	0.095	2.429	0.005	0.001	14.500	0.145
	Lettuce	0.0193	0.060	0.118	0.015	0.005	95.000	0.102
Gelan	Cabbage	7.891	0.225	3.553	0.002	0.013	46.304	0.255
	Lettuce	9.292	0.125	0.882	0.001	0.004	45.870	0.603

continuous use of industrial wastewater for agricultural purposes can increase the load of heavy metals in the soil.[36]

The BCF values for Zn and Pb in cabbage and lettuce at the Sabata and Gelan sites, Cd and Mn in lettuce at Sabata, and Cd in cabbage at the Burayu and Gelan sites were above 1.0. The high BCF of Zn indicates its elevated mobility from soil to vegetables, followed by Pb. Conversely, Gupta et al[31] reported that the Pb transfer factor was very low compared to Fe, Cd, Mn, and Cr. The transfer factor of heavy metals from soil to vegetables may be influenced by the nature of the soil, the composition of the parent material, vegetable species, and the metals' solubility.[37,38]

Individuals are encouraged to consume more plants due to their high-quality sources of macronutrients, micronutrients, fibers, and other beneficial components for overall well-being. Furthermore, according to WHO/FAO, the essential feature of nutritional value is regulated by the concentration of trace metals within food to protect humans from the harmful effects of toxic substances. The results of the EDI for Pb at Sabata in cabbage were 0.015 and 0.016 for adults and children, respectively, while in lettuce, the values were 0.016 and 0.0176 for adults and children, respectively. At the Burayu site, the EDI values for Pb in lettuce were 0.019 and 0.021 for adults and children, respectively. In contrast, at the Gelan site, the EDI values for Pb in lettuce were 0.022 and 0.024 for adults and children, respectively, followed by cabbage at 0.0188 for both adults and children. Thus, the EDI results for heavy metals at different study sites for both adults and children were within the Provisional Maximum Tolerable Daily Intakes (PMTDI) set by FAO/WHO.[39,40]

The non-carcinogenic individual health risks from the consumption of vegetables contaminated with heavy metals were assessed using the THQ as described in the Methods section, employing equation "Y". The THQ values for cabbage at the Sabata site were ordered as follows: Pb > Cd > Cr > Ni > Cu > Mn > Zn. At the Burayu site, the order was Cr > Cd > Cu > Ni > Mn > Zn > Pb, and at the Gelan site, the order was Pb > Cd > Cr > Cu > Ni > Mn > Zn. For lettuce, the THQ values were ordered as Pb > Cd > Cr > Cu > Zn > Mn > Ni at the study site; Pb > Cd > Cr > Cu > Ni > Mn > Zn at the Burayu site; and Pb > Cr > Cd > Cu > Mn > Zn > Ni at the Gelan site. The estimated THQ values for lettuce were 8.416, 10.086, and 11.597 for Pb at the Sabata, Burayu, and Gelan sites, respectively. For cabbage, the THQ values for Pb were 7.86 and 9.42 at the Sabata and Gelan sites, respectively, with all these values exceeding 1. Therefore, there is a significant probability of exposure to non-carcinogenic health effects.[18,25] Except for Pb, the THQ values for other heavy metals considered in this study were below 1, indicating that they are generally considered safe from non-carcinogenic effects.

The HI, which reflects the cumulative risk from the intake of various potentially hazardous heavy metals through the consumption of different vegetables, has also been computed. The HI values for cabbage, lettuce, and Swiss chard are reported in Table S5. At the Sabata site, the HI values for cabbage and lettuce were 8.1677 and 8.784, respectively. For lettuce at the Burayu site, the HI was 10.609, while for cabbage and lettuce at the Gelan site, the values were 9.9587 and 11.8122, respectively. An HI value greater than 1 indicates a potential health risk, and HI values greater than 10 suggest a severe and persistent health effect. Thus, the HI values imply significant potential health impacts for lettuce grown at the Burayu and Gelan sites.[25,30]

In accordance with the study by Sharma et al[16] the carcinogenic hazard to individual health was calculated using equation 6. The TCR for the intake of Pb, Ni, Cr, and Cd was assessed based on the estimated quantities. The TCR values for Pb in lettuce at the Sabata, Burayu, and Gelan sites were 1×10^{-4} (Table S6), indicating that the TCR value of Pb in lettuce is within the safe limit (1×10^{-4} to 1×10^{-6}) set by Health Canada.[41] The TCR values for Pb in cabbage at the Sabata and Gelan sites were 1×10^{-4} and 2×10^{-4}, respectively, with the TCR for cabbage at the Gelan site falling within the threshold value.

This implies no cancer risk from Pb for the adult population in the vicinity of the study site from consuming cabbage and lettuce grown in the area. The TCR values for Cr in cabbage and lettuce at Sabata (1×10^{-4}), Burayu (1×10^{-4}), and Gelan (2×10^{-4} and 1×10^{-4}, respectively) were within the safe limit, indicating a potential exposure to cancer risk from Cr for the adult population consuming these vegetables in the study area. Moreover, the TCR values for Cd and Ni in these study areas were within the threshold value, indicating a potential cancer risk from Cd and Ni for those consuming vegetables from the study area.

Conclusion

In sites 4 and 5 of the Sabata area, the concentrations of heavy metals in irrigation water and vegetables exceeded the permissible limits. Conversely, in the soil across all sampling sites and study areas, the concentrations were predominantly below the allowable levels. Significant effects of various industrial activities on heavy metal concentrations were observed in irrigation water at each sampling and study site, but not in soil and vegetables. The presence of heavy metals in vegetables was attributed to soil and water contamination from the discharge of untreated industrial effluents. The concentrations of heavy metals in irrigation water at industrial sampling locations were higher than those at control sites. Similarly, elevated levels were also detected in vegetable and soil samples from these locations.

In Sabata, vegetables cultivated with irrigation water and contaminated by industrial effluents exhibited heavy metal concentrations exceeding permissible levels. Some concentrations remained within permissible limits, but the high bioaccumulation factor in vegetables was a significant concern. To prevent the introduction of heavy metals into the irrigation area, untreated public or industrial waste must not be released into water bodies and farmlands. Furthermore, the treatment of industrial waste before its release into irrigation areas should be implemented as a mitigation measure to clean previously polluted water bodies of toxic trace metal substrates. Continuous monitoring of soil, plants, and water quality is essential to prevent the accumulation of heavy metals in vegetables, thereby reducing risks to human health and environmental disruption. Therefore, relevant institutions and stakeholders must work together to mitigate high concentrations of heavy metals in water, soil, and vegetables through the installation of treatment plants, awareness campaigns, and the use of phytoremediation methods.

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Formal analysis: Milkessa Jagemma.
Investigation: Milkessa Jagemma.
Methodology: Milkessa Jagemma.
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Resources: Milkessa Jagemma.
Supervision: Hailu Worku.
Validation: Hailu Worku.
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Writing–review & editing: Hailu Worku.

Competing Interests

The authors declare no conflicting interests.

Ethical Approval

Not applicable.

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Supplementary Files

Supplementary file 1 contains Table S1-S6.

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