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Bioaccumulation and Biota-Sediment Accumulation Factor of Metals and Metalloids in Edible Fish: A Systematic Review in Ethiopian Surface Waters

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ABSTRACT: Metals and metalloids pollution of Ethiopian surface water is becoming an environmental issue. Bioaccumulation and Biota-Sediment Accumulation Factor (BAF and BSAF) are used to quantify the bioaccumulation of contaminants from water and sediment to biota. The present study aimed to determine the BAF and BSAF of metals and metalloids in different surface waters of Ethiopia. Furthermore, the ecological and human health risks were also examined. Generally, 902 peer-reviewed papers from 2005 to 2022 were searched using search engines. The most common types of edible fish species in studied surface waters of Ethiopia were *Oreochromis niloticus*, *Clarias gariepinus*, and *Barbus intermedius*. The concentration of metals and metalloids were higher in sediment than in water and in carnivorous fish than in herbivorous fish. The BSAF of Se in all fish species was greater than 1. *Oreochromis niloticus* was a bio-concentrator of As and Se. The dissolved concentration of Cu, Cd, Pb, and Ni were higher compared to the Ethiopian Environmental Protection Authority and the European Union; Organization of Economic and Co-Operation Development environmental water quality standard guidelines for inland surface freshwater bodies. Likewise, in sediment, the concentration of Cu, Pb, Ni, Zn, and Cr were above Tolerable Effect Concentration values and Cd, Ni, and Cr were above Probable Effect Concentration levels when compared to the United States Consensus-Based Sediment Quality Guidelines for freshwater which indicates that these metals may pose risk to aquatic organisms. Consumption of raw water and fish contaminated with the detected metals and metalloids is not associated with any diseases. However, local consumers who live close to each freshwater ecosystem may become more exposed to health risk hazards. The findings of this study will provide baseline information on BAF and BSAF of metals and metalloids in surface waters and will contribute to the effective monitoring of environmental quality.

Keywords: Bioaccumulation, Ethiopia, freshwater ecosystem, environmental pollution, metal(loid)s

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Introduction

Surface water has been exposed to numerous pollutants. Because of their persistence, non-biodegradability and bio-magnification potential, water pollution by metals and metalloids become a serious environmental problem worldwide.^{1,2} In the aquatic environment, metals and metalloids are partitioned among various media such as biota, water, and sediment.3 In the water phase, metals and metalloids are partitioned in dissolved form and adsorbed to suspended matter.4 However, they preferentially accumulate in sediments due to their adsorption capacity, from where due to desorption and remobilization processes, they are later retransferred to the water column and food chain.⁵⁻⁹

The main possible uptake routes of metals and metalloids for fish are through food ingestion and the permeable epidermis in the gills.10 Fish can accumulate metals and metalloids from their diet, water and sediment and these accumulated concentrations can be transferred to humans through fish consumption.11 However, the toxicity of metals and metalloids to aquatic organisms is linked to the bioavailable fraction.12 Metals and metalloids can magnify when they accumulate in aquatic biota via the food chain.13 Because of their relatively large body size, long life cycle, position in the aquatic food chain and value for human food, fish are involved as biological

indicators for the assessment of the effects of metals and metalloids and the quality of aquatic environment.¹⁴

Bioaccumulation factor (BAF) is used to quantify the bioaccumulation of metals and metalloids from water to biota due to uptake through all possible routes of exposure which is expressed as a quotient of the concentration of a specific metal or metalloid in biota and the concentration of that metal or metalloid in the water (ambient media).15 Likewise, Biota-Sediment Accumulation Factor (BSAF) quantifies the bioaccumulation of sediment-associated metal or metalloid into biota.16 In many Ethiopian freshwater ecosystems, metals and metalloids pollution is increasing.17 Metals and metalloids can originate from 2 main sources, that is natural causes such as weathering of the earth's crust or mineral ores weathering, soil runoff, soluble salts dissolution in water); and anthropogenic causes include withdrawing of minerals, wastewater, industrial processes, irrigation activities, runoff from urban areas, and pesticide spray.4 Moreover, most industries in Ethiopia are discharging their untreated water to the nearby surface waters.18,19

Previous studies have explored higher levels of metals and metalloids in sediment than in water, high levels of Hg in carnivorous fish (Barbus intermedius),17,19,20 and none of the detected metals was hazardous to humans.17,21,22 These studies

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Figure 1. A personal creation shows the flowchart representing the methodology of the review.

also mostly determined the total concentrations of metals and metalloids in water, sediment and fish tissue. However, only one study17 has determined BAF and BSAF of metals and metalloids. Therefore, this study aims to determine BAF and BSAF of metals and metalloids in different surface waters of Ethiopia. Furthermore, the ecological and human health risks associated with metals and metalloids concentration were also examined. Hence, the findings of this study will provide baseline information on the levels of BAF and BSAF of metals and metalloids in Ethiopian surface waters, and will contribute to effective monitoring of environmental quality. Later, it will also enable the evaluation of future trends in surface water pollution by metals and metalloids.

Materials and Methods

Searching strategy and the study protocol

The study has been conducted between 19 March, 2022 and 23 August, 2022. Google Scholar, PubMed, Web of Science, Embase and SCOPUS were mainly used for web-searching of peer-reviewed articles focusing on the distribution and concentration of metals and metalloids in water, sediment and fish in surface waters of Ethiopia. Accordingly, 902 papers from 2005 to 2022 were searched and collected (Figure 1). Important key words mainly "metals," "trace metals," "heavy metals," and "metals in water," "metals in sediment," "metals in fish tissue," and "metals in the aquatic environment" were used in the search engine (databases). The study period of peer-reviewed articles was left closeended to search for updated papers.

Inclusion and exclusion criteria

For the inclusion of peer-reviewed papers in the study, basically, the following considerations were taken such as all factors related to metals and metalloids in surface water of Ethiopia. The focus of the study was on published and peer-reviewed papers. However, books, reviewed papers, conference articles, and master's and PhD thesis were excluded from the study. Throughout the review, all searches may have a limitation according to the following considerations: (1) Published literatures may be omitted due to a lack of a linkage with the important keywords, (2) all used literatures were only in the English versions, (3) some may not available due to closed access, (4) others were not cataloged in the electronic databases, and (5) all included studies are cross-sectional studies.

Because of the variation of metals and metalloids concentration and their multitude in different surface water of the region, the collected data was analyzed from numerous points of view including the spatial distribution of metals and metalloids in the aquatic environment. All the units of concentration are expressed as µg/L and mg/kg for metal and metalloid levels in water, sediment and fish tissue respectively. Concentrations that were below the limit of quantification (detection) (<LOQ/ LOD), were substituted with a value of LOQ/2.23,24

Bioaccumulation -and biota-sediment accumulation factor

The most common types of edible fish species found in many studies were Nile tilapia (*Oreochromis niloticus*), African sharp

tooth catfish (*Clarias gariepinus*), and African big barb (*Barbus intermedius*). Following, the bioaccumulation-and Biota-Sediment Accumulation factor (BAF and BSAF) of metals and metalloids were calculated as follows using equations.16

$$
BAF = C_b/C_w
$$

$$
BSAF = C_b/C_s
$$

Where: BAF=bioaccumulation factor (L/kg) , BSAF=biotasediment accumulation factor (mg/kg/dw), C_b = concentration of metal in fish (μ g/g dw), C_s =concentration of metal in sediment (μ g/g dw), and C_w =concentration of metal in water (mg/L) .

Ecological risk assessment of metals and metalloids in water and sediment

To determine whether the measured dissolved metals and metalloids in the surface waters pose adverse effects on the benthic aquatic fauna, the average concentrations of metals and metalloids were compared to the water quality standards of the Ethiopian Environmental Protection Authority (EEPA)25 and the European Union water quality standard guidelines for inland surface freshwater bodies.26 Likewise, to determine whether the detected metals and metalloids in the lake sediment have potential adverse effects on the benthic fauna, the investigated concentrations were compared to the United States Consensus-Based Sediment Quality Guidelines (SQGs) for freshwater. These values represent Threshold/ Tolerable Effect Concentration (TEC) and Probable Effect Concentration (PEC) values.27

Human health risk assessment

To estimate the potential non-carcinogenic human health risk associated with metals and metalloids in the surface waters of Ethiopia, the detected metals and metalloids concentrations were compared with water quality standards for drinking water by the World Health Organization (WHO)²⁸. Likewise, to estimate human health risks associated with metal or metalloid contaminated fish consumption, estimated daily intake (EDI in mg/kg/day), average daily intake (ADI in mg/kg/ day), and hazard quotient (HQ) values for metals and metalloids were calculated using the corresponding reference dose (RfD in mg/kg/day).29 A 60 kg body weight person (ie, an average body weight of Ethiopian adults) and a national fish consumption of 0.19 kg/week (~0.027 kg/day)30 were used for the calculations. The maximum edible amount (MEA in kg/ day) of fish consumption per person per day, without causing a potential human health risk, was calculated using the reference dose (the maximum tolerable daily intake of a metal or metalloid that does not pose any adverse health effect) of metals and metalloids established by United States Environmental Protection Authority,³¹ Agency for Toxic Substances and

Disease Registry,³² World Health Organization,³³ and United States Food and Drug Administration.³⁴ The used concentrations of metals and metalloids in fish muscle were the average of the measured concentrations in each edible fish species in Ethiopia. EDI, HQ, and MEA were calculated using the following equations^{29,35}:

$$
EDI = C * DFC/BW
$$

ADI = [EDI * ED * EF) / (AT)] * 10⁻³
HQ = ADI/RfD
MEA = RfD * BW/C

With *C* is the concentration of metals or metalloids in μ g/g ww, DFC is fish consumption in kg/day, BW is adult Ethiopian body weight (60kg), ED is the exposure duration (30years for an adult),³⁶ EF is the exposure frequency (365 days/year),³⁷ AT is the average exposure time for non-carcinogens (365days/ year \times ED),³⁷ and RfD is the reference dose for Cd (0.00001 mg/ kg/day), Cu (0.01mg/kg/day), Pb (0.004mg/kg/day), Cr (0.0009mg/kg/day), Co (0.01mg/kg/day), As (0.00003mg/kg/ day),³² Ni (0.02 mg/kg/day), Zn (0.3 mg/kg/day),³¹ Se (0.17 mg/ kg/day), and Hg (0.00023mg/kg/day).33 EDI, HQ, and MEA values were also calculated based on the maximum concentrations detected in each edible fish species in Ethiopia, to determine the worst-case scenario.

Result and Discussion

Data accessibility

After searching, 902 peer-reviewed articles focusing on the distribution and accumulation pattern of metals and metalloids; that is Arsenic (As), Cadmium (Cd), Chromium (Cr), Cobalt (Co), Copper (Cu), Mercury (Hg), Lead (Pb), Nickel (Ni), Selenium (Se), and Zinc (Zn) in water, sediment, and fish muscle from 2005 to 2022, 15 articles were used for the analysis of the result. After screening all the reports in the Ethiopian aquatic environment, a majority (73.68%) of reports were from the Rift Valley Lakes (RVLs). For instance, from the RVLs, more (47.37%) was done in Lake Hawassa followed by Lake Ziway (26.32%).

Detection method of metals and metalloids

Observing the collected data, different methods (instrumental analysis) were used for the detection of metals and metalloids in water, sediment and fish. The most frequently used detection method (Figure 2) for metals and metalloids analysis was Atomic Absorption Spectrophotometer (AAS= 66.7%) followed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) (26.7%). However, this was unexpected, even though both are the most common detection methods of metals and metalloids, AAS detects metals and metalloids serially while ICP-MS did concurrently. Moreover, ICP-MS is faster and has cheaper processing costs and better detection limits (LOD) compared to ASS.38

Metals and metalloids in water, sediment, and fish muscle

The concentration of 10 metals and metalloids (ie, As, Cd, Cr, Co, Cu, Hg, Pb, Ni, Se, and Zn) were collected from previously done literatures. An overview of the concentration of metals and metalloids in water (µg/L), sediment (mg/kg dw), and fish muscle (mg/kg dw) of Ethiopian surface waters are presented in Table A1 to A3 (see in Appendix). Zn has the highest concentration (575.90µg/L) of metals and metalloids in water (Figure A1 in Appendix) which has shown a similar pattern as compared to previous studies.17,18,39-41 The average concentration of Cu, Cd, Pb, Ni, Cr, Co, Hg, As, and Se in water were 176.43, 66.60, 59.07, 191.84, 35.32, 109.04, 0.32, 2.10, and 0.82µg/L. In terms of specific location, more data were collected from Rift Valley Lakes, Lake Hawassa, with the highest concentration of Zn 1059.76µg/L and was similar with other studies.17,18,39,41 In sediment, Figure A2 (see in Appendix) the concentration of Cr (4529.21mg/kg dw) and Ni (4422.56mg/kg dw) was higher when compared with other metals and metalloids which is similar compared to Kassaye et al⁴¹ in Lake Ziway and Hawassa. The concentration of Cu, Cd, Pb, Zn, Co, Hg, As, and Se were 32.93, 24.49, 16.10, 259.30, 23.17, 8.15, 2.08, and 0.18mg/kg dw. The highest concentration of Cr in sediment may be ingested by fish and later accumulated. Overall, the concentration of metals and metalloids in sediment was higher than in water.

Analysis of the results also revealed metals and metalloids levels in fish muscle (Table A3). For instance, in all fish species, the concentration of Zn was highest which was 15.40, 18.48, and 31.00mg/kg dw in *C. gariepinus*, *O. niloticus*, and *B. intermedius* respectively. This shows a similar pattern with previous studies,17,19,22 which may be due to the essentiality of Zn for physiological role and uptake from the aquatic environment by fish preferentially. Overall, similar to previous studies, 17,19,22 the finding of this study also revealed that the concentration of most metals and metalloids were relatively higher in carnivorous fish than in herbivorous fish.

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Bioaccumulation-and biota sediment accumulation factors

The average concentration of detected metals and metalloids in water (µg/L), sediment (mg/kg dw), fish muscle (mg/kg dw), and their BAF and BSAF are summarized in Table 1. For instance, the BAF of Hg in *O. niloticus* and *B. intermedius* and As in *O. niloticus* was greater than 1. The BSAF of Se in all fish species and As for *O. niloticus* and Co for *B. intermedius* was also greater than 1. Moreover, BSAF of As and Se for *O. niloticus* was greater than 2. In contrast, previous studies have shown a higher BAF for Cd⁴⁰ and Cr and Hg¹⁷ and BSAF of all metals or metalloids was also less than 1.17 Overall, BAF and BSAF value greater than 1 indicates that metal or metalloid may be accumulated in fish muscle, while a value less than 1 indicates that a metal or metalloid is not accumulated in fish tissue directly from water and is associated with sediment respectively.42 In addition, an organism tissue with $BSAF > 2$, $1 < BSAF < 2$, and $BSAF < 1$ can be considered as a macro-concentrator, micro-concentrator, and de-concentrator respectively.43 Therefore, the results of this study illustrated that *O. niloticus* is a bio-concentrator of As and Se. While, authors like Arnot and Gobas⁴⁴ suggest that BAF values <1000, 1000 < BAF < 5000, and >5000 are also considered as having no probability of accumulation, bioaccumulative, and extremely accumulative respectively.

Ecological risk assessment

Based on data analysis, in surface waters of Ethiopia, the dissolved concentration of Cu, Cd, Pb, and Ni were higher compared to the water quality standards of the Ethiopian Environmental Protection Authority25 and the European Union water quality standard guidelines for inland surface freshwater bodies²⁶ (Table A1). Likewise, in sediment of surface waters (Table A2), Cu, Pb, Ni, Zn, and Cr were above TEC values and Cd, Ni, and Cr were above PEC levels when compared to the United States Consensus-Based Sediment Quality Guidelines (SQGs) for freshwater which represent Threshold/Tolerable Effect Concentration (TEC) and Probable Effect Concentration (PEC) values,²⁷ and this was similar to a previous study¹⁷ in Lake Hawassa. This indicates that these detected metals in water and sediment may pose risk to aquatic organisms or benthic fauna.

Human health risk assessment

The concentration of detected metals and metalloids in water is not associated with any disease. Fish muscle is consumed by people in most countries.17,45 Analysis of human health risk (hazard quotient of metals and metalloids) associated with metal or metalloid-contaminated fish consumption (Table A4), HQ for all metals and metalloids for all edible fish species

Table 1. The average concentration of detected metals and metalloids in water (µg/L), sediment (mg/kg dw), fish tissue (*O. niloticus*, *C. gariepinus*, and *B. intermedius*) (mg/kg dw)), and respective BAF and BSAF values.

was <1. This was as expected because previous studies found, except Hg, metals, and metalloids were not hazardous to human consumers.^{17,21,22} A HQ > 1 is considered to be hazardous and a HQ<1 is non-hazardous.29,46,47 Similarly, the MEA of most metals and metalloids in all edible fish species was higher than the average daily national fish consumption of Ethiopians. This indicates that a consumer (a 60 kg body weight person) is not at risk or will not be poisoned by the measured metals or metalloids. Accordingly to Dsikowitzky et al,22 a regional fish consumption of 1.05 kg/week was recorded. Therefore, local consumers who live close to each freshwater ecosystem may be more exposed to health hazards.

Recommendations

Consumption of raw water and fish contaminated with the detected metals and metalloids is not associated with any diseases. However, to give a comprehensive inference, analysis of metals and metalloids in other organs of the fish like kidney, gills, intestine, bone, and heart is required. It is highly recommended to characterize some physicochemical parameters of water (pH, temperature, conductivity, redox potential, hardness, divalent cations (eg, Mg^{2+} and Ca^{2+}), metal and metalloid chemical speciation, total alkalinity, total suspended, and dissolved solids) to investigate their effect in the distribution, bioavailability and bioaccumulation of metals and metalloids in the aquatic environment. Further investigators should also detect the effect of seasonal variation on BAF and BSAF of metals and metalloids.

Conclusion

Metals and metalloids have been detected in different surface water ecosystems of Ethiopia. Data on metals and metalloids in water, sediment and fish muscle were collected. The most used detection method for metals and metalloids analysis was Atomic Absorption Spectrophotometer (AAS). The average concentration of metal and metalloids in sediment were higher than in water. The average concentration of Zn was highest in all fish species muscle. The results of the study reveal that the concentration of most metals and metalloids were relatively higher in carnivorous fish than in herbivorous fish. BAF of Hg in *O. niloticus* and *B. intermedius* and As in *O. niloticus* was greater than 1. Likewise, BSAF of Se in all fish species was greater than 1. *O. niloticus* is considered as a bio-concentrator of AS and Se. Cu, Cd, Pb, and Ni in water and Cd, Ni, and Cr in sediment may pose risk to aquatic organisms. Consumption of raw water contaminated with the measured metals and metalloids is not associated with any disease. Similarly, eating fish contaminated with the measured metals and metalloids may not pose risk to consumers. The results of this study may also be a good database for government, risk managers, and researchers on metals and metalloids pollution of an aquatic environment. It will also enable the evaluation of future trends in surface water pollution.

Author Contributions

Conceptualization: BAM. **Developing methods**: BAM, SME, TSA. **Data analysis**: BAM, SME, TSA. **Writing and editing**: BAM, SME, TSA.

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Appendix

Table A1. The average concentration of metals and metalloids (μ g/L) in water of Ethiopian surface waters from 2005 to 2022.

Table A2. The average concentration of metals and metalloids (mg/kg dw) in sediment of Ethiopian surface waters from 2005 to 2022.

 (Continued)

STUDY AREA CONCENTRATION OF METALS AND METALLOIDS (MG/KG DW) REFERENCE Cu Cd Pb Ni Zn Cr Co As Se Hg Lake Hawassa, Ethiopia 32.5 6 14.5 14 119.5 4-9 25.5 - - - - Nigussie et al³⁹ Lake Ziway, Ethiopia 36.5 20 15.5 19.5 142.5 6.5 20 - - - Lake Hashenge, Ethiopia 56 208 3 39424 1129 87 34 - - - Gebrekidan Asgedom et al⁴⁰ Lake Hawassa, Ethiopia - 0.1 1.1 - 3.5 0.55 - - - 0.05 Amare et al¹⁸ Tendaho Reservoir, Ethiopia 10 1 6 5 22 2.5 3 - - - Asefa and Beranu²¹ TEC values 31.6 0.99 35.8 22.7 121 43.4 - 9.8 - 0.18 MacDonald et al²⁷ PEC values 150 5 130 49 460 110 - 33 - 1.1

Table A2. (Continued)

Table A3. The average concentration of metals and metalloids (mg/kg dw) in 3 edible fish species (*O. niloticus*, *C. gariepinus*, and *B. intermedius*) muscle of Ethiopian surface waters from 2005 to 2022.

Table A4. Estimated Daily Intake (EDI in mg/Kg/day) levels, the average daily intake (ADI in mg/Kg/day), Hazard Quotient (HQ), and Maximum Edible Amount (MEA in mg/Kg/day) of metals and
metalloids when consuming the 3 comm **Table A4.** Estimated Daily Intake (EDI in mg/Kg/day) levels, the average daily intake (ADI in mg/Kg/day), Hazard Quotient (HQ), and Maximum Edible Amount (MEA in mg/Kg/day) of metals and metalloids when consuming the 3 commercially important fish species muscle (*O. niloticus, C. gariepinus*, and *B. intermedius*) from 2005 to 2022.

Water Sediment

Figure A1. The average concentration of metals and metalloids in water (µg/L) and sediment (mg/kg dw) in Ethiopian surface waters.

Figure A2. The average concentration of metals and metalloids in 3 edible fish species muscle (mg/kg dw) in Ethiopian surface waters.