Elevated total mercury (THg) levels in water sources under the influenced of artisanal and small-scale gold mining (ASGM) in Tanzania.

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Abstract

This study investigated the presence and distribution of mercury in water bodies under the influence of artisanal and small-scale mining (ASGM) activities in Tanzania, which continue to predominantly rely on mercury for gold extraction. Various water bodies available for domestic and animal use in mining communities were sampled from surface water sources in ASGM settlements during the rainy and dry seasons. Water samples were analysed using cold vapour atomic fluorescence spectrophotometer (CVAFS). The results indicate that most of water sources had THg levels above the WHO guideline of 1.0 µg/L (1000 ng/L) for safe drinking water. The levels were significantly higher during the wet season ranging from 3.4 to 96.3 µg/L, whereas the range was from 0.84 to 2.12 µg/L during the dry period. The higher THg values during the wet season are likely a result of increased lateral transport (e.g. via enhanced runoff) and physical properties of the waterways. Transportation and resuspension of matrix-bound mercury from surface soils and inflow of contaminated water from unprotected tailings were also observed to be potential means of lateral mercury transport.

The lowest concentrations (0.846 µg/L) were observed in water samples from the Mabubi River, upstream of a mining village. Downstream of the mining village in the same river, higher concentrations were observed in the Nungwe Bay region of Lake Victoria. In other surveyed mining settlements on the impacts of mercury to organisms as part of the strategies to mitigate mercury pollution.

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Introduction

Artisanal and small-scale gold mining (ASGM) is the world's largest anthropogenic source of mercury emissions to the environment (UNEP, 2012). This is the result of direct emissions occurring during ore amalgamation and remobilization of mercury accumulated in soils and sediments surrounding ASGM settlements over time. Specifically, contaminated gold mine tailings are a major route of environmental contamination (Abdelaalet al., 2023; Esdaile & Chalker, 2018; Gerson et al., 2018; Selin & Selin, 2022; Spiegel & Veiga, 2010) together with vaporized mercury when the gold-mercury amalgam is burned to separate gold from mercury (Van Straaten, 2000). It is estimated that for every gramme (g) of gold produced, there is between 1 and 2 g of mercury lost to the environment (Van Straaten, 2000; Tschakert & Singha, 2007; Yoshimura et al., 2021; Timmins, 2003;). In 2015, ASGM introduced about 1220 tonnes of mercury into the terrestrial and freshwater environments, while other sources of mercury, like waste treatment, ore mining and processing, and energy production, introduced about 580 tonnes of mercury (UNEP, 2019). ASGM activities also represent the largest global source of emissions of mercury to the air, estimated to be 838 (range, 675–1000) tonnes per year (UNEP, 2019). In the air, mercury can be transported and transformed (e.g. oxidized) through atmospheric processes and be incorporated into food webs after deposition primarily via aqueous ecosystem processes (Kim and Zoh, 2012; Nyanza et al., 2014).

Gold mining activities in Tanzania are carried out by both large-scale mining operations and ASGM activities. The ASGM sector is a growing employer, involving a significant number of miners who play an important role in improving community livelihoods (Mutagwaba et al.,

2018). Gold is the leading export mineral from Tanzania, with more than 300 ASGM sites in the country, including both licensed and informal ones, employing between 0.5 and 1.5 million people (Mdee, 2015; Rwiza et al., 2023; URT, 2020). A major concern is the continued use of mercury and the growing number of ASGM activities throughout the country, which is accompanied by the widespread release of mercury (Hg) into the environment. The mercury amalgamation gold extraction process is predominantly preferred by miners due to its simplicity and assumed low cost. However, it results in longlasting impacts on the environment and human health. ASGM-associated environmental degradation and the likely impacts on human health have been described elsewhere (Sanga et al., 2023; Steckling et al., 2017; Zvarivadza & Nhleko, 2017).

A major source and most likely the predominant source of mercury in the Tanzanian environment is ASGM. According to the government report (URT, 2020), ASGM contributes more than 80% of the total mercury used in Tanzania while the remaining 20% is from dental amalgam and imported mercury-added products. Although Tanzania has no routine mercury monitoring system, there have been sporadic assessments of mercury in water, sediments, and food (e.g. Sanga et al., 2023; Nyanza et al., 2014; Tungaraza, et al., 2011; Chibunda, et al., 2010, etc.), which have characterized the extent of mercury contamination in the country. The Tanzanian government reported that between 2017 and 2020, the average annual production of gold by ASGM was 15.3 tonnes and that 13.2–24.4 tonnes of mercury were consumed per year (URT, 2020). This is consistent with an estimate by Reid et al. (2019) in which the annual 2013 gold production in Tanzania by small-scale miners was 20 metric tonnes.

In Tanzania, ASGM activities are prevalent in the Lake Victoria region, which contains large deposits of gold. However, the southern highlands region has also experienced significant ASGM activities, particularly at the Makongolosi mine. In the Lake Victoria region, mercury contamination has been detected in nearly all environments at mining settlements, including biota, water, soil, and sediments (Ikingura et al., 2006; Nyanza et al., 2014; Taylor et al., 2005, etc.). Elevated levels of mercury in human blood and urine have been reported around ASGM sites (Nyanza et al., 2014, 2019). More concern has been raised after realizing that some fish species, such as Nile perch samples collected from Lake Victoria, have mercury concentrations exceeding the WHO threshold limit of 0.2 μg/g (Campbell, 2001; Campbell et al., 2004). Ikingura et al. (2006) noted THg concentrations ranging from 2 to 35 μg/g, with the highest concentrations again recorded in Nile perch. Contaminated environments and

foodstuffs pose a significant risk to human health, as reported by Tungaraza et al. (2011), who found that among different categories of food intakes, fish contained the highest content of total Hg (0.16 μg/g wet weight). The average dietary intake of Hg was higher (about 41 μg/ day) than the daily intake limit of 15.5 μg/day recommended by FAO and WHO as a safe limit for pregnant women to protect a developing foetus.

Communities in ASGM areas may be exposed to mercury through water, soil, air, and subsequent accumulation in foodstuffs such as fish, cassava, and vegetables. Transformation of inorganic mercury to methylmercury—a significantly more toxic form of mercury—exacerbates the public health consequences of mercury contamination. Several strategies have been employed to protect public health and the environment. Such strategies include containment of mercury-contaminated water in ponds or recovery of mercury during gold-mercury separation using retorts (Jønsson et al., 2009). The extent and/or success of such strategies to safeguard the environment and human health appears to be minimal in most ASGM communities (Rwiza et al., 2023) as high concentrations of mercury are found in the environment and biota. While humans are still exposed to mercury contamination, and communities around ASGM in Tanzania are at a higher relative risk of chronic or acute mercury poisoning, there is no routine monitoring and protection against mercury. The fate and distribution of mercury around ASGM areas and beyond, therefore, need to be identified. Since water runoff plays a major role in mercury transport from mining sites to rivers, plants, and large water bodies, this study investigated the distribution of mercury in different water bodies in Tanzania. The objective of this study was to evaluate the extent of THg pollution in the aquatic environment by measuring its concentration in water bodies in the vicinity of several gold recovery operations in Tanzania.

Methodology

Study Area

Water samples were collected from six active ASGM settlements located in three administrative regions in Tanzania. These include the Mgusu and Nyarugusu mines in the Geita region, the Nyakavangala and Itengulinyi mines in the Iringa region, and the Makongolosi and Itumbi mines in the Mbeya region (Fig. 1). The water sources sampled were rivers, ponds, and shallow wells located near and within the mining settlements. At the Mgusu mine, samples were collected from the surface water of the Mabubi River, a shallow running water that originates from an upstream perennial spring and flows into Lake Victoria's Nungwe bay. At Nyarugusu mine, samples were collected from surface water ponds less than 30 cm deep within the floodplain which at some areas dries up during dry period. Rivers and springs are the major water sources for domestic use, livestock needs, irrigation (observed specifically at Mgusu mine), and gold extraction activities at all mines. The Mgusu mine is situated near a permanent river (Mabubi River) that discharges water into Lake Victoria at Nungwe bay, while other mining settlements have temporary water streams that dry up or pond during dry periods. In terms of the intensity of mining activities, Mgusu and Nyarugusu mines in the Lake Victoria region are more active with larger populations compared to the other mines in this study. Table 1 summarises the locations and characteristics of the sampling points in the study areas.

Table 1: Physical location and characteristics of sampling points at six different artisanal gold mines in Tanzania

Mine	Sampling point and characteristics	Mine	Sampling point and characteristics
Mgusu Mine	Mgusu 1: -Upstream spring, upper river point	Nyarugusu mine	Nyarugusu 1: - Pond water within the floodplain and mine tailing disposal
	Mgusu 2: -Downstream, 180 m from Mgusu1 adjacent to the mining settlements		Nyarugusu 2: - Floodplain water at mine tailing disposal (Dry during the dry period)
	Mgusu 3: -Downstream, 400 m from Mgusu 2 adjacent to the mining settlements	Nyakavangala Mine	Mine point: -Stream adjacent to the mines (Dry During the Dry Season)
	Mgusu 4: -Downstream, 390 m from Mgusu 3 outside the mine.		Mbungu river: 3 km from a mining settlements

	Mgusu 5: - Rice farm, floodplain,~5 km downstreame from Mgusu 4	Itengulinyi & Itumbi mines	Itengulinyi mine Ditch: -Stagnant shallow water at the mine
	Mgusu 6: -2nd site within the rice farm		Itumbi: -Stagnant pond water at the mine
	Mgusu 7: - Close to Lake Victoria Nungwe bay ~8 km	Makongolosi mine	River water ~800 m from the mine

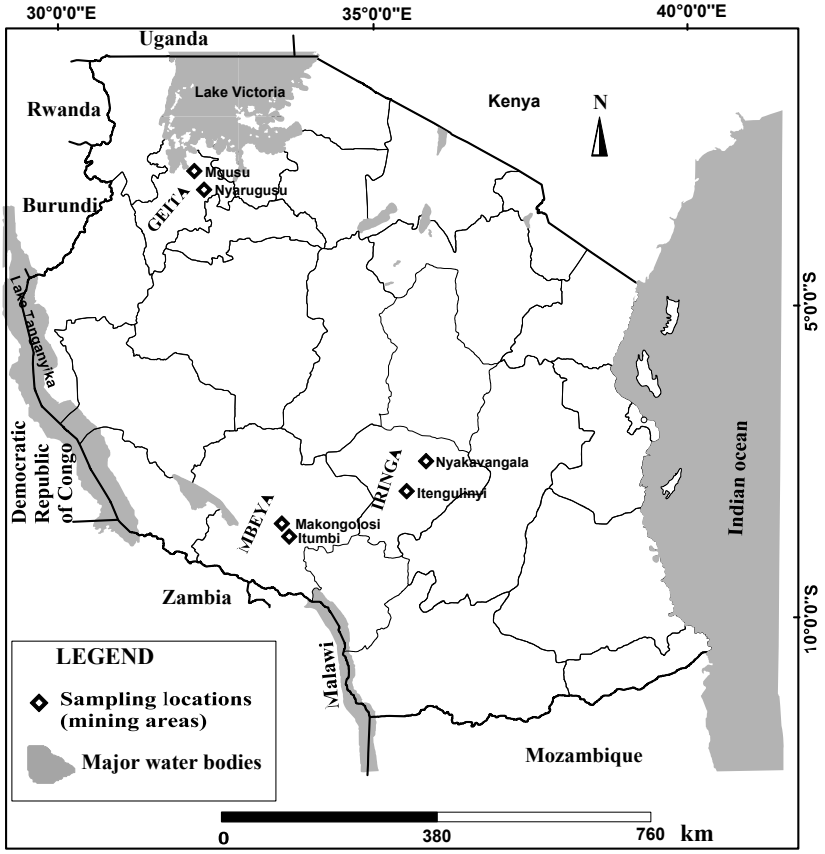

Fig. 1 Study sampling locations in three regions Geita region (Mgusu and Nyarugusu), Iringa region (Nyakavangala and Itengulinyi) and Mbeya region (Makongolosi and Itumbi) in the United Republic of Tanzania.

Sample collection and pre-treatment

Duplicate surface water samples were collected from different sources around active ASGM sites into 250 ml amber glass bottles with Teflon-lined caps. The sample bottles were pre-cleaned by acid following the procedures outlined in EPA Method 1631. At the sampling sites, the bottles were rinsed three times using sample water. From each sampling point,

duplicate sets of water samples were collected during the dry period (July 2020) and at the end of the wet season (May 2021). During the dry period, some surface water sources were dry; therefore, samples were collected from fewer locations at mining settlements. Each water sample was preserved by adding 2 mL of bromine monochloride (BrCl) immediately after sampling.

Sample Preparation and analysis

The samples were filtered using pre-heated GF/F filters (at 450 °C). Analysis followed EPA Method 1631, Revision E, for cold vapour atomic fluorescence spectrometry (CVAFS). 25 mL water subsamples in analytical glass vials were treated with 100 µL of bromine monochloride (BrCl) and left to rest overnight to release matrix-bound mercury and oxidize all forms of mercury to Hg^{2+}. Prior to analysis, the BrCl in the 25 mL analytical vials was reduced by adding at least 100 µL of hydroxylamine hydrochloride ($NH_2OH.HCl$), ensuring complete decolourization of the samples from the brown colour of BrCl. Stannous chloride ($SnCl_2$) was added to convert Hg^{2+} to volatile Hg°, followed by immediate analysis of the samples. Sample analysis was done by using the Brooks-Rand MERX™ Total-Hg direct purge, equipped with Atomic Fluorescence spectrophotometer, model III detector. Purging was done using ultra-pure argon gas. The analysis was carried out at the Department of Chemistry and Physics, Sokoine University of Agriculture (SUA).

RESULTS AND DISCUSSION

Figure 2 and Tables 2 and 3 summarize the total mercury (THg) concentrations in the water samples collected from surface water sources in the study areas. Figure 2 and a sampling station in Table 2 provide concentrations for both dry and wet seasons, while Table 3 indicates that no water samples were collected from the southern highlands during the dry period. This was due to the drying up of water sources at the mining settlements; therefore, water samples were collected only during the wet season in these areas.

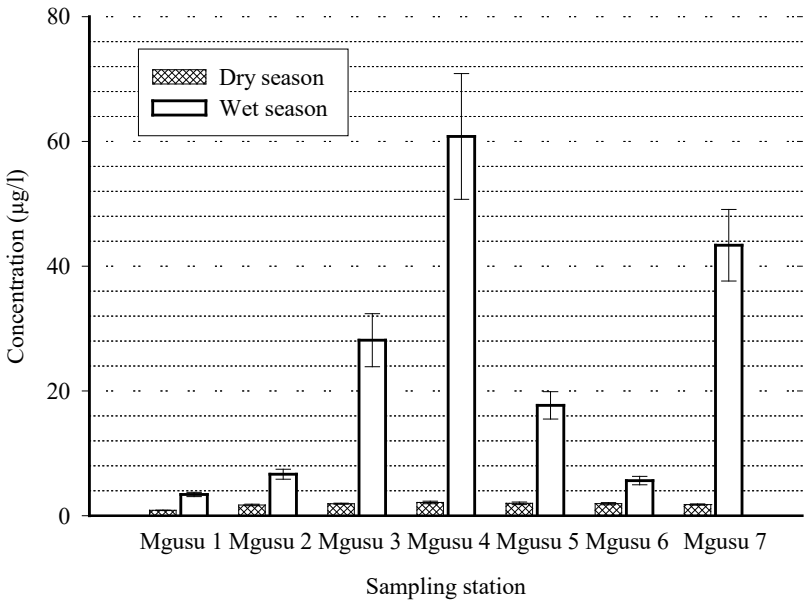

Fig. 2 Concentration of THg in water samples collected from sources for domestic use and livestock consumption around Mgusu mine (ASGM) areas downstream of the Lake Victoria gold field region during wet and dry seasons. Error bars represent standard deviation of triplicate analyses of samples.

Table 2a: Concentration of THg in water samples collected from ponds at Nyarugusu mine in the Lake Victoria gold field. The water sources are mainly for livestock.

Sampling location	Concentration (µg/L)							
	Dry period samples				Wet Season samples			
	1	2	$\bar{x}$	$s(\pm)$	1	2	$\bar{x}$	$s(\pm)$
Nyarugusu 1	1.21	1.40	1.31	0.13	17.06	15.28	16.17	1.26
Nyarugusu 2	ND	ND	-	-	14.21	12.50	13.35	1.22

ND= Not determined due to absence of water samples from dried ponds

Table 2b: Concentration of THg in water samples collected from sources within and close to mining settlements in the Southern highland regions during the wet season

Sampling location	**Concentration (µg/L)**				
	Dry period	**Wet Season**			
		1	2	$\bar{x}$	$s(\pm)$
Nyakavangala Mine	ND	27.55	31.99	29.77	3.15
Nyakavangala Bridge	ND	1.74	1. 90	1.82	1.87

Itengulinyi Ditch	ND	108.82	83.70	96.26	17.76
Itumbi Ditch/Pond	ND	55.81	44.29	50.05	8.139
Makongolosi Bridge	ND	21.38	19.36	20.37	1.425

ND= Not determined. No water samples could be collected due to the drying of water sources.

From Fig. 2 and Tables 2 and 3, the results indicated that all sampled water sources had THg above the background levels (0.04–0.05 μg/L) established by Taylor et al. (2005). Similar elevation of THg concentration in the river water surrounding the ASGM over the background concentration has been reported elsewhere (Abdelaalet al., 2023; Gerson et al., 2018). Such elevation of THg concentration in water above the background concentration is a consequence of ASGM activities. At Mgusu (Fig. 2) during the dry period, the THg concentrations ranged from 0.84 to 2.12 μg/L where the highest concentration was detected at point 4, downstream before the Mabubi River exits the Mgusu mine area. Similarly, during the wet season, upstream point (Mgusu1) had the lowest THg concentration, and the river exit point (Mgusu 4) had the highest concentration. In general, concentrations in the wet season were higher by about 30-fold when comparing the river exit point values and were about fourfold when comparing the lowest detected concentration value (at the upstream site). Samples taken from the Nyarugusu mine (Table 2) do not show a defined trend. This is due to the characteristics of the sampling location, which is a valley, seasonally flooded by slowmoving, river-like water. This flooding likely leads to a spatial distribution of samples with no noticeable horizontal pattern. The water movement and varying flow conditions contribute to the irregular distribution of the sampled materials and contaminants. Nevertheless, samples from Nyarugusu 1 reflect similar variation between the wet season and dry period at the mine where the wet season had the highest concentration (about 12-fold) compared with the dry period season. In Table 3, results indicated the concentration range of mines with similar water source characteristics. The four mines all use water sources from seasonal water pools, which are available only during the wet season. Nevertheless, there were higher concentrations (average 96.26 μg/L) of THg at the Itengulinyi mine sampling site, more than any of the analysed samples. Higher concentrations (average 50.05 μg/L) were also observed at the Itumbi mine water pool. These results indicate that the majority of all samples during the dry season and all samples collected during the wet season were above the WHO (2004) guideline value (1.0 μg/L) for safe drinking water.

The observed higher THg values were consistent with previous findings by other authors (Moreno- Brush et al., 2019; Rytuba, 2000, etc.) who ascribed the concentration differences to stream flow characteristics. During low stream flow (dry period), soil characteristics like fine-grained sediments have been considered to have adsorbed mercury, which is deposited at the bottom of the stream. However, during the wet season when stream flow is high and turbulent, mercury-enriched sediments are transported (Moreno- Brush et al., 2019; Rytuba, 2000). Similar results were reported by van Straten (2000) where water quality data indicate very low mercury transport rates during the dry period. Given the nature of mercury use at mining sites, mobilization of surface soils by running water during the wet season contributes to the desorption of adsorbed mercury, as shown in Fig. 2, which presents concentration trend of a defined river flow all the way to Lake Victoria (Nungwe Bay). Nungwe Bay is a floodplain with reduced surface water movement, possibly enhancing the accumulation of THg-rich sediment and subsequent elevated desorbed Hg. However, Nungwe Bay is observed to be a landing station for a number of fishermen, and these activities may have also influenced the mercury distribution. What also needs to be noted is that even though the concentration at the upstream site (Mgusu 1, Fig. 2) was lower than at other sampled locations at this mine, the concentration is above the WHO guideline of 1.0 μg/L for safe drinking water. More serious is the fact that this is the main source of water for the Mgusu mining population. In a previous study on dietary intake in the area (Tungaraza et al., 2011), adult daily water intake was estimated at about 1 L. Given the current observed elevated levels of mercury residues, it implies that mercury intake through water is a risk to human health. The higher mercury residues at the water source can be speculated to be attributed to deposition from short-distance atmospheric transport because there are no nearby activities related to mercury use further upstream. This issue warrants further investigation.

These results indicate that the gold amalgamation process contaminates all water sources at mining settlements mainly through surface runoff, as exemplified by the higher concentration (28.13 μg/L) at Mgusu 3 (Fig. 2), where the influence on the concentration is facilitated by the wet season conditions. Mgusu mining and processing locations are at higher altitude, close to the banks of the Mabubi River. The higher levels of mercury observed during the wet season imply a significant mercury residue transport through runoff to the river. This shows how weather conditions influence the distribution of mercury in the mining settlements. In contrast, the reduced runoff during the dry period explains the generally reduced concentrations during that period (Tables 2 and 3). In addition to the impact of weather on the

distribution of mercury residues in surface water, there is potential of atmospheric mercury transport and farreaching consequences associated with artisanal and small-scale gold mining (ASGM) from mining sites. ASGM activities are the largest source of anthropogenic mercury emissions to the air, and the atmospheric transport and deposition of emitted mercury have been discussed in previous studies (e.g. Cheng et al. (2013) and Cohen et al. (2016)). One concerning aspect highlighted in this study is the consistent occurrence of ASGM activities near important water sources. For example, the Mgusu mine is situated in close proximity to Lake Victoria, which serves as a water source for the region's public water utility.

At Mgusu where there is a permanent river (Mabubi River), THg concentration increased as the river flows across the mining village and reached the maximum at Mgusu 4 point where the river exits the immediate region of mining activities. The increasing trend in THg concentration from Mgusu points 1 to 4 may be due to cumulative contamination along the river downstream in both seasons and inflow of contaminated water from uncontained tailings and amalgamation ponds during the wet season. At the end of the village boundaries, downstream, there is a less defined river pathway and thus reduced water flow rate, resulting in relatively muddy conditions. This is probably reducing flushing but more deposition leading to elevated THg concentrations. From Mgusu 4, THg values decreased to Lake Victoria at Nungwe Bay. Mgusu 5 and 6 is a point where the river passes a wetland where rice is cultivated. The decrease in concentration in Mabubi River downstream is likely a result of the settling of mercury-sorbed sediments down the stream. It has been reported that, in streams affected by mercury pollution, THg transport is affected by many physicochemical processes including adsorption onto particulate phases, e.g. iron oxyhydroxide (Moreno-Brush et al., 2019; Nyanza et al., 2014, 2019; Rytuba, 2000; Sanga et al., 2023; Taylor et al., 2005) and reaction of soluble mercury with inorganic sulphur compounds to insoluble mercury compounds (López et al., 2008). However, a more complete understanding of the spatial concentration trend found in Mgusu 5, 6, and 7 will require further investigation.

The mines in the Southern Highlands, namely Nyakavangala, Itengulinyi, Itumbi, and Makongolosi, have less intensive mining activities compared to the Lake Victoria region in terms of the population at the settlements. However, it has not been established why generally higher concentrations of mercury residues were found in these areas compared to the Lake Victoria region. One possible factor that may have contributed to this finding is the fact that the mines are located in a region that mostly experiences a unimodal rainfall pattern, whereas

the Lake Victoria region experiences a bimodal rainfall pattern (Agrawala et al., 2003). Due to limited surface water sources at the mines, water for mining activities is primarily obtained from shallow wells within the mine premises. The water used for gold extraction was observed being recycled, which may have enriched the concentration of mercury residues in the water samples.

CONCLUSION AND RECOMMENDATIONS

This investigation found that surface water around the studied artisanal and small-scale mining areas was contaminated by varying concentrations of mercury, in many cases higher than the WHO safe level for drinking water. Since the processes and conditions at the studied sites have similarities, it is likely that other mining sites in the country have a similar mercury contamination problem within and around ASGM sites. This poses health risks to the populations, especially those who have resettled in these mining areas as permanent residents, as well as those downstream, as shown at the Mgusu mine. Higher total mercury (THg) concentrations in surface water during the wet seasons than during the dry period provide evidence of higher mercury loading into surrounding water bodies, aided by weather phenomena. The terrain of the mining environments studied was observed to facilitate soil erosion, particularly during the rainy season, thereby flooding water bodies with mercury residues through tailings. Additionally, atmospheric deposition may have contributed to elevated concentration of THg in the surface water. Meanwhile, all ASGM sites are located in places with reliable water sources for processing activities. The findings of this study need to be suggested that water reservoirs at mining settlements be restricted from human and animal consumption unless shown to be safe, e.g. after protection measures are introduced to safeguard the water supply from mercury contamination.

Although this study focused on ASGM-related mercury contamination in six mining settlements, it is important to note that mining activities in Tanzania are carried out in numerous other areas not covered by this research. According to the National Action Plan for Artisanal and Small-Scale Gold Mining 2020–2025 (URT, 2020), Tanzania, with a surface area of approximately 945,087 km^2, is estimated to have 300 known sites involved in ASGM activities across the country, whereas they are estimated to utilize approximately 24.4 metric tonnes of mercury per year. This exposes a significant portion of water resources in the country to mercury contamination.

The observations reported here are based on water bodies influenced by runoff from artisanal and smallscale gold mines, which facilitate the transport of mercury residues through surface water and soil. It is recommended that future studies investigate how mercury is distributed among different media, particularly water and sediments, along various pathways. Additionally, it is important to assess the spatial and temporal variations of different species of mercury in the biosphere under different weather conditions, as these conditions can also influence the growth, feeding, breeding, etc., of organisms.

DECLARATION

All authors have read, understood, and have complied as applicable with the statement on "Ethical responsibilities of Authors" as found in the Instructions for Authors.

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