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Widespread and Persistent Mercury Contamination Beyond Disposal Sites: Case study on Challenges for Remediation in Artisanal Gold Mines of Tanzania

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Abstract

Many national and international initiatives aim to control and limit the use of mercury (Hg) in gold extraction. However, the feasibility of Hg eradication from the environment depends on understanding the extent of its distribution. This case study focuses on the spatial and vertical distribution of total mercury (THg) residues at five artisanal mining sites in Tanzania namely, Mgongo, Sekenke, Nyarugusu, Rwangasa, and Mugusu to assess the feasibility of remediating the problem. The trend showed presence of THg residues in surface layers (0–20 cm), decreasing with depth but still detectable in deeper layers (> 20–100 cm). A horizontal distribution in surface-layer concentrations was also observed along the Mabubi River, which drains across Mugusu mine into Lake Victoria. Among all sites, the highest surface-layer (20 cm) total mercury (THg) concentration was 1.48 ± 0.02 mg/kg, measured from a sample collected at the Nyarugusu mine site, with a moderate decrease to 0.12 ± 0.001 mg/kg at a depth of 100 cm. Other soil samples from the Rwangasa mine site showed THg concentrations of 0.048 ± 0.012 mg/kg and 0.082 ± 0.01 mg/kg at depths of 70 and 80 cm, respectively. These findings suggest that significant THg residues are detected from surface to deeper layers and wide area of river sediment distribution, mediated by physical, environmental, biological and chemical processes that support simultaneous Hg transport and suspension. The THg residues in soils and sediments challenge the feasibility of remediation efforts in areas with similar wide contamination extents. They demonstrate a long-term legacy of contamination that will continue to impact environmental quality in many regions affected by artisanal mining.

Introduction

Analyses of soil, water, and biota in areas surrounding mines have generally indicated widespread metal residues (Mariki et al. 2024; Sanga et al. 2023; Pu et al. 2019; Koleleni and Mbike 2018; Song et al. 2018; Demková et al. 2017; Ikingura et al. 2006 etc.). This highlights the inadequately controlled spread of mine-related contaminants into the surrounding environment. Mercury (Hg) is a heavy metal with well-documented hazards to human health (Park and Zheng 2012; Ha et al. 2017). Consequently, reducing ongoing Hg contamination sources and minimizing the impacts of historical Hg contamination are crucial. These goals are key objectives of the international Minamata Convention, which came into force in 2017, aiming to protect human health and

the environment from anthropogenic emissions and releases of Hg and its compounds (UN 2013). Even with reductions in current releases, addressing the historical legacy of Hg contamination remains a significant challenge. Substantial quantities of historical Hg contamination persist in ecosystems worldwide (Al-Sulaiti et al. 2022).

Efforts to mitigate mercury environmental contamination have also focused on the remediation of contaminated sites, employing methods such as chemical oxidation, reduction, adsorption, and desorption (Wang et al. 2020). However, the speciation of Hg and other physicochemical parameters and the contaminated medium significantly influence the effectiveness of these approaches (Gai et al. 2019). In general, the effectiveness and implementation costs of remediation

techniques vary widely, particularly when addressing large, contaminated sites, such as those associated with mining activities (Khalid et al. 2017).

Currently, Artisanal and Small-Scale Gold Mining (ASGM) activities in developing countries are reported as the leading global sources of Hg in the environment, primarily due to the widespread use of mercury and poor handling practices during gold extraction processes (Calao-Ramos et al. 2021). Globally, emissions associated with ASGM activities are estimated to contribute about 38% of total mercury emissions (UN Environment 2019). Historical records indicate that the large quantities of mercury that were released into the environment since the Spanish and Portuguese colonial periods (Brown 2017), still persist as residues in the environment (Alpers and Hunerlach 2000). In Tanzania, a significant number of artisanal and small-scale gold miners have been registered, estimated at approximately 700,000–1,500,000 (URT 2017).

Although trace amounts of mercury in gold ore have been identified (URT 2017), Tanzania lacks a geochemical baseline to indicate the natural occurrence of Hg in the environment. Nevertheless, the well-documented elevated Hg concentrations in areas of significant anthropogenic Hg releases are likely primarily attributable to these activities. According to the Tanzania National Action Plan for Artisanal and Small-Scale Gold Mining 2020–2025 (URT 2020), the annual utilization of Hg for gold extraction ranges from 13.2 to 24.4 tonnes. Consequently, environmental protection efforts and Hg residue detection initiatives in Tanzania have focused on regions influenced by ASGM.

Artisanal and small-scale gold mining has been practiced in Tanzania for more than fifty years. ASGM sites have evolved into permanent human settlements with growing populations, sometimes exceedingly high, such as Rwamgasa and Mugusu mines, with approximately 16,000 and 15,000 residents, respectively (pers. communication). These sites are officially recognized as settlement villages, supported by essential social services like schools and places of worship. However, they have also become areas contaminated by the accumulation of mercury-polluted soil and mine effluents within their ecosystems.

Historically, in Tanzania, ASGM sites and their vicinities have been recognized for having elevated concentrations of Hg (Sanga et al. 2023; Campbell et al. 2003; Van Straaten 2000; Ikingura and Akagi 1996; etc.). Research findings have revealed the presence of Hg in all environmental components at mining sites, including water, sediments, fish, cattle, other domestic animals, and various plant-based foods (Machiwa 2009; Ikingura et al. 2006; Chibunda et al. 2008; Chibunda and Janssen 2009; Tungaraza et al. 2011; Mariki et al. 2024). Notably, crucial food items, such as Nile perch from Lake Victoria, have demonstrated values

nearing the WHO limit of 0.2 µg/g (Campbell et al. 2003; Campbell 2001). However, observations by Ikingura et al. (2006) on total Hg (THg) concentrations in Nile perch indicated contamination levels ranging from 2 to 35 µg/g, far exceeding the WHO recommended limit.

In the most recent study by Nipen et al. (2022), conducted in Dar es Salaam, Tanzania's largest city, which does not have any ASGM activities, relatively low THg concentrations were found in soil samples (ranging from 0.0067 to 0.098 mg/kg). These concentrations were much lower than the general background soil concentrations of 0.03–1.0 mg/kg (Wang et al. 2020). However, in other parts of the world, higher levels of gaseous elemental Hg (as high as 2.41 ng/m³) have been reported as typical of urban environments (Lin et al. 2023). This suggests the potential significance of atmospheric Hg transport impacting the region from diverse sources. Atmospheric Hg influences are beyond the scope of this investigation but are being evaluated in ongoing work. Nonetheless, it is unlikely that the locally elevated environmental concentrations found near ASGM sites are due to regional or long-range atmospheric transport. To address the critical issue of environmental pollution, Tanzania has undertaken various initiatives, including becoming a signatory to the international Minamata Convention. Other efforts to curb mercury use include policy implementations such as the National Environmental Policy (URT 1997), a comprehensive countrywide inventory of mercury releases (URT 2012), and the Minamata Initial Assessment (URT 2017).

The purpose of this investigation was to characterize and quantify the extent of mercury contamination in areas affected by ASGM (artisanal and small-scale gold mining) activities by measuring mercury distribution in vertical soil and sediment profiles. This study provides a deeper examination of the distribution of mercury residues in mining villages and their surroundings. Given the need for concrete actions, particularly regarding the remediation of already contaminated sites and the associated challenges, it is essential to present the extent of mercury residue distribution.

Methodology

Study Area

Soil samples were collected from five artisanal and small-scale gold mines in Tanzania located in the central region (Sekenke and Mgongo) and the Lake Victoria region (Rwamgasa, Nyarugusu and Mugusu). Sediment samples were taken along the Mabubi River that crosses adjacent to Mugusu mine to Nungwe floodplain close to Lake Victoria (Fig. 1). The Mabubi River is characterized by a shallow, clear, and continuously surface flowing water with aquatic

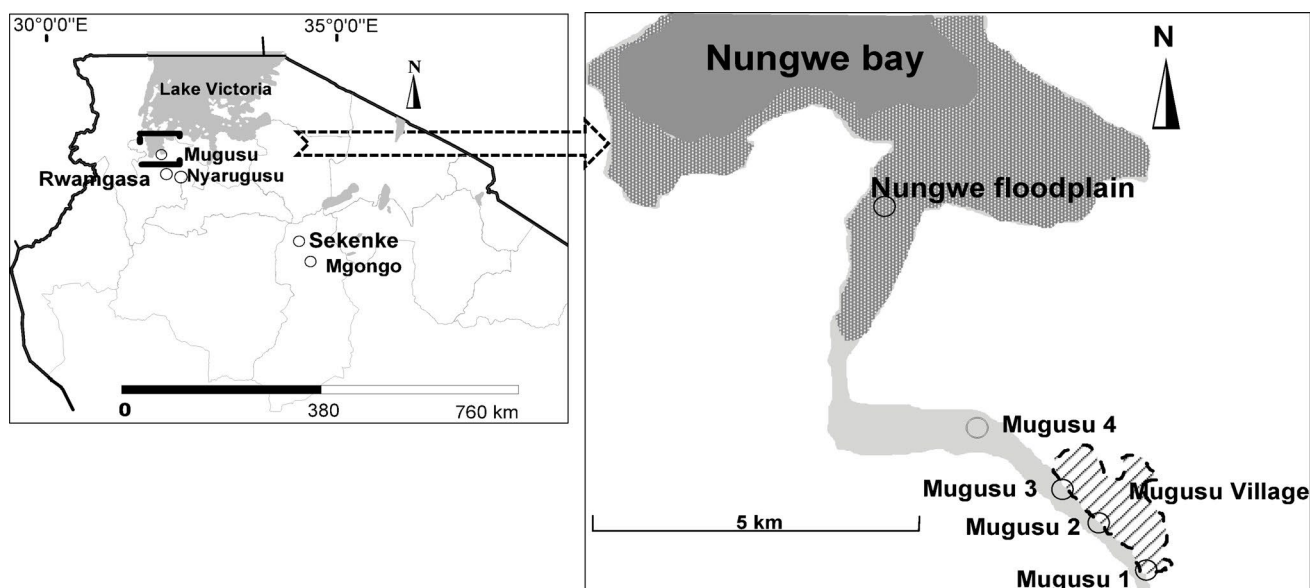


Fig. 1 Locations of ASGM sites in the Lake Victoria region and part of central Tanzania. A redrawn map from Google Earth image shows the details of the sampling points along the Mabubi River next to the Mugusu village

and riverine vegetation. All mines are actively using Hg-amalgam method for gold extraction, which involves mixing finely ground gold-bearing ore with liquid mercury to form a gold–mercury amalgam. The amalgam is then heated in open air (roasting), which vapourize mercury at its low boiling point of about 357 °C, leaving behind metallic gold. The characteristics of the mines are summarised in Table 1. Mining activities take place outside the human settlement area at all mining sites studied here, but all gold extraction processes take place within the settlement premises.

Sampling

The sample collection took place during a dry period characterized by intensified mining and processing activities. Soil samples were collected from each artisanal mining village, while at the Mugusu mining village, sediment samples were taken from the Mabubi River and the floodplain at Nungwe Bay. For soil sampling in the mining villages, the central point of each village was selected as the sampling location. At each site, three sampling points were established at intervals of approximately 20 m along the middle section of the riverbanks, covering depths in 10 cm increments from the surface to a maximum depth of 100 cm. The sampling intervals were chosen to maximize the likelihood of capturing the true distribution of mining impacts across the site. However, it is acknowledged that this approach does not guarantee full representativeness of pollutant distribution across the entire mining environment. Additionally, certain depth intervals were inaccessible due to the presence of underlying rocks, which posed limitations to the sampling process. Samples

were collected using a stainless steel Edelman hand auger with graduated depth marks.

As noted before, sediment samples were collected from the Mabubi River and the Nungwe floodplain. The Mabubi River is a perennial water body that flows adjacent to the Mugusu mine and eventually reaches Lake Victoria at Nungwe Bay through a floodplain, located approximately 10 km from the Mugusu mining village. Using a similar sampling technique, points along the river were selected at intervals of at least 1 km, starting at a point upstream, at the beginning of the mining village. At each sampling point, three samples were collected at a spacing of about 60 cm. This was necessitated by the narrow riverbanks, physical barriers such as vegetation, and potential interference from local human activities. This approach enabled the collection of a broad range of sediment samples to assess the extent of mercury contamination associated with mining activities. All samples were stored in amber bottles with Teflon caps and transported to the laboratory within 24 h of collection.

Sample Preparation and Analysis

Sample Preparation

Soil and sediments were freeze-dried for 24 h, followed by sample pulverization before chemical treatment. The digestion of samples was conducted following EPA method 1631 (USEPA 2001), a chemical digestion method. For each sample, approximately 0.5 g was weighed into a 40 ml reagent bottle. Subsequently, 8 ml of high-purity analytical-grade

Table 1 Characteristics of sampling locations where vertical soil and sediment samples were collected during the investigation

Sampling sites	Type of sample	Site characteristics
Sekenke (Singida, central region)	Soil	Mining and processing activities conducted across the site Sample(s) collected from processing locations Sparse population with semi-permanent settlements Area $\approx 0.5 \text{ km}^2$
Mgongo (Singida, central region)	Soil	Previously known as Sekenke Mine during colonial times (established in 1909 under industrial mining; Henckel et al. 2016) Dense human settlement with permanent housing Highly active with mining and processing activities spread throughout the village Samples collected from mining and processing areas Area $\approx 2.5 \text{ km}^2$
Mugusu #1–4 (Geita, Lake Victoria region)	Sediment	Sampling points along Mugusu river that runs adjacent to Mugusu mining village, empties into Lake Victoria Mining is conducted on a hill overlooking the village, which lies downhill Processing activities are carried out across Mugusu village ($\approx 1.5 \text{ km}^2$), with effluents draining into the Mabubi River, especially during periods of high runoff Water depths at sampling sites: Mugusu #1 $\approx 10 \text{ cm}$, Mugusu #2 $\approx 15 \text{ cm}$, Mugusu #3 $\approx 15 \text{ cm}$, Mugusu #4 $\approx 30 \text{ cm}$ Sampling depth at Mugusu #4 was restricted by underlying rocks
Nungwe floodplain (Geita)	Sediment	A floodplain connecting the Mabubi River to Nungwe bay of Lake Victoria The floodplain is utilized for rice cultivation Samples were collected from the floodplain (rice farm) Water depth at sampling site $\approx 15 \text{ cm}$
Nyarugusu (Geita)	Soil	The oldest artisanal mine, in operation since the 1970s Currently with reduced activity, but extraction continues using previously mined drainage soil Samples collected from the central processing area, within the former mining site Area $\approx 6 \text{ km}^2$
Rwamgasa (Geita)	Soil	The mine has transitioned into a permanent human settlement (village) Mining and processing activities occur within the village Samples were collected from the mining and settlement area Area $\approx 3.0 \text{ km}^2$

concentrated HCl (37% Sigma-Aldrich) and 2 ml of concentrated HNO_3 (70% Sigma-Aldrich) were added within a fume chamber. The samples and reagents were allowed to digest at room temperature overnight. Following digestion,

40 ml of 0.07 M (35%) BrCl solution was introduced into each digested sample and allowed to settle overnight to facilitate the oxidation of all mercury species in the sample.

Sample Analysis

Each sample was analysed by transferring 1.25 mL subsamples from the digested and oxidized samples into 25 mL analytical glass vials. The subsamples were then diluted to 25 mL with deionized water (18.2 M Ω), and 0.1 mL of 30% hydroxylamine hydrochloride was added to neutralize the BrCl. Subsequently, 0.1 mL of 20% stannous chloride (sigma-Aldrich) in 10% HCl was added to each subsample in the analytical vials, which were immediately sealed with Teflon-lined caps.

Sample quantification was conducted using the Brooks–Rand "MERX" automated mercury analytical system, equipped with a Brooks–Rand MERX III CVAFS detector model III Atomic Fluorescence Spectrophotometer. The analysis was carried out at the Department of Chemistry and Physics, Sokoine University of Agriculture (SUA). Mercury concentrations were determined using calibration curves based on a certified mercury standard (VWR–BDH 10 mg/L stock solution in 5% HNO_3). The method involved purging volatile mercury from the sample with argon gas to the detector. Quality control was conducted by triplicate analysis of equipment blanks and calibration blanks to discriminate the influence of both analytical equipment and chemicals and reagents used. The analyses of samples were also simultaneously conducted in triplicates together with prepared certified reference estuarine sediment (ERM–CC580; certified value: $132 \pm 3 \text{ mg/kg}$). A recovery rate of 98.73% was achieved, with a mean concentration of 130.33 mg/kg and a standard deviation of $\pm 0.87 \text{ mg/kg}$ ($n = 4$).

Results

A summary of the results is presented graphically in Figs. 2, 3 and 4. In Fig. 2, total mercury (THg) concentrations at both the Sekenke and Mgongo mines were higher in the surface (20 cm) soil samples. At Sekenke, the average concentration slightly decreased from $0.77 \pm 0.02 \text{ mg/kg}$ at 20 cm to $0.73 \pm 0.02 \text{ mg/kg}$ at 30 cm, and further decreased to $0.32 \pm 0.09 \text{ mg/kg}$ at 40 cm. At 50 cm, the recorded average concentration was $0.14 \pm 0.09 \text{ mg/kg}$, with the minimum concentration recorded in the deeper sampled layer (60 cm), which showed an average concentration of $0.106 \pm 0.02 \text{ mg/kg}$. At the Mgongo mine, the surface (20 cm) concentration was higher ($2.1 \pm 0.06 \text{ mg/kg}$) compared to Sekenke mine. The concentration decreased to $0.85 \pm 0.05 \text{ mg/kg}$ at 30 cm and further to $0.25 \pm 0.008 \text{ mg/kg}$ at 40 cm. Between 40 and

Fig. 2 Concentrations (mean \pm s) of THg in Vertical soils samples at Sekenke and Mgongo mines in Singida, central region, Central Tanzania

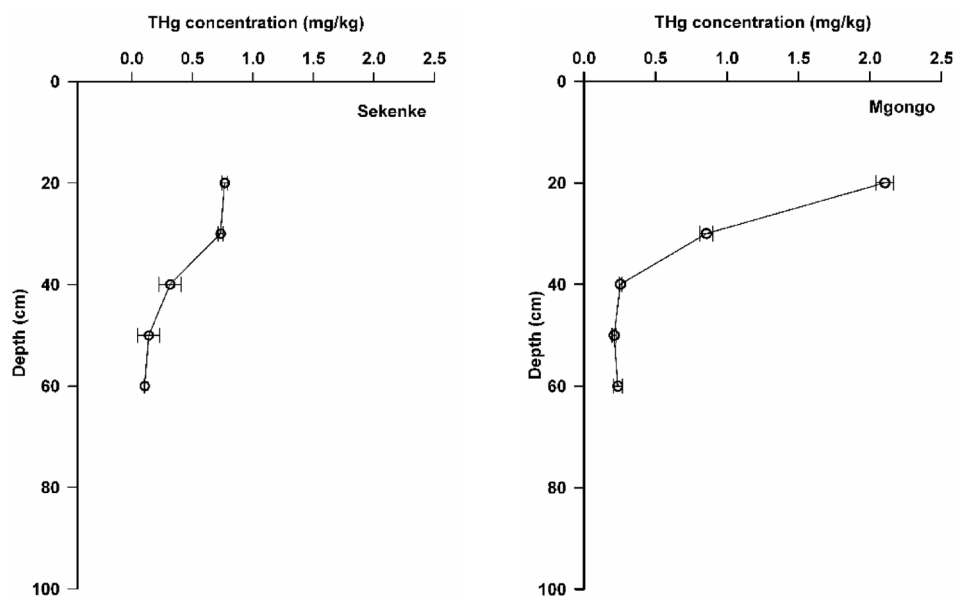
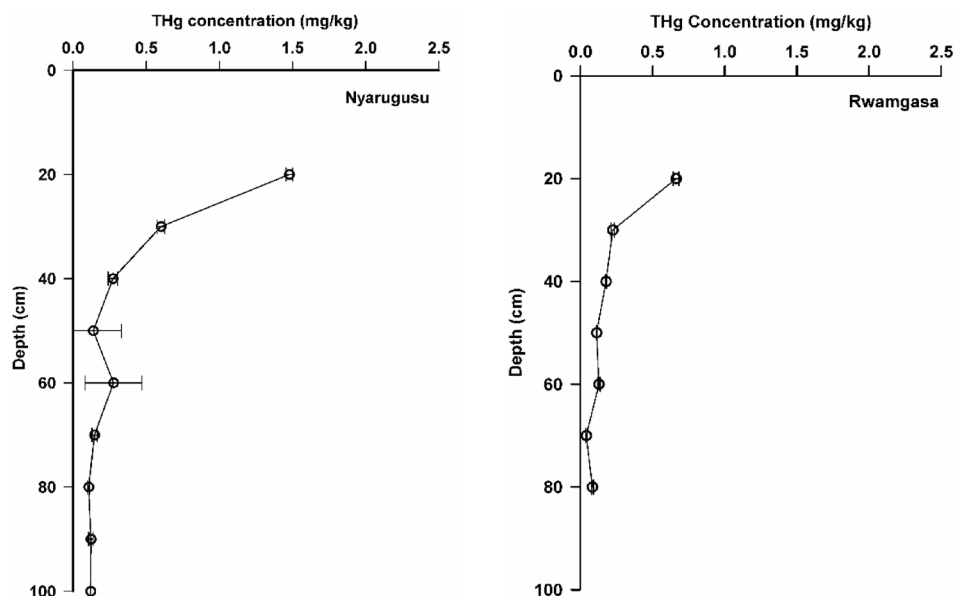


Fig. 3 Vertical concentrations (mean \pm s) of THg in soil vertical samples from Nyarugusu and Rwamgasa mines (in Geita region, Tanzania). Samples collected from 20 to 100 cm depth



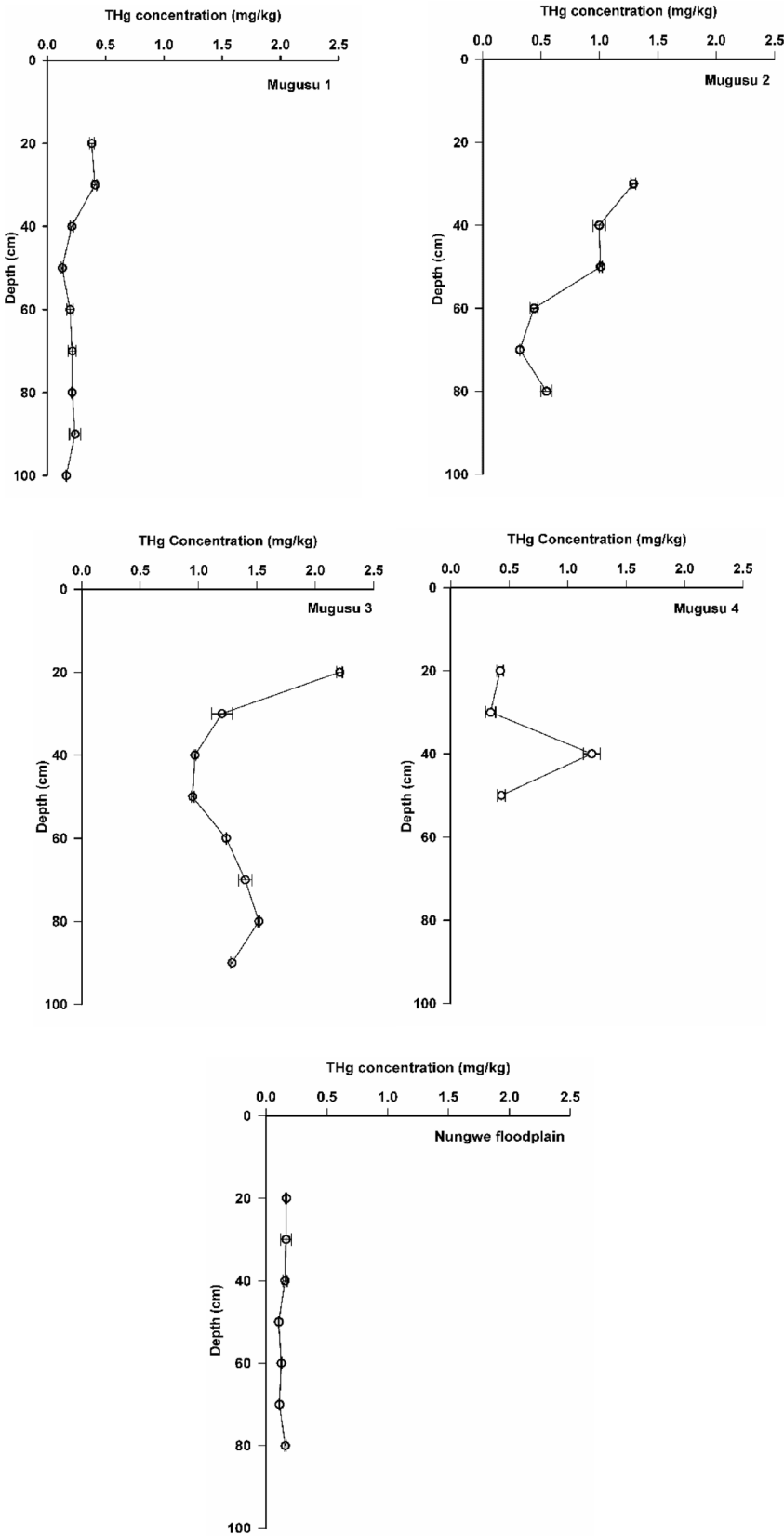
60 cm, the variation in concentration was not significant compared to the surface layers. Soil sampling at these mines was limited to a depth of 60 cm due to the rocky substrate at deeper levels. The general trends in the figure for both mining sites indicated a decrease in concentration from the surface to deeper soil layers. However, at 60 cm depth, the Mgongo mining area's soil samples still showed a significantly higher average THg concentration (0.24 ± 0.03 mg/kg).

In Fig. 3, the trend of THg concentration at the Nyarugusu mine mirrors that of Fig. 2 but exhibits a more pronounced decreasing trend extending deeper to 100 cm. The average concentration decreased from 1.48 ± 0.02 mg/kg in the surface (20 cm) soil sample to less than 50% (0.60 ± 0.03 mg/kg)

kg) at a depth of 30 cm. Similarly, at 40 cm (0.27 ± 0.03 mg/kg) and 50 cm (0.28 ± 0.19 mg/kg), the concentration was less than half of that observed at 30 cm. Between 60 and 100 cm depth, the concentrations showed no statistically significant difference ($p = 0.19$). Overall, the trend indicated a general decrease from the surface layers, with THg still detectable at 100 cm depth.

The vertical distribution of THg in the soil from the Rwamgasa artisanal mining village (a highly active mining area with other mixed human activities) showed the highest average concentrations in the surface layers. At 20 cm, the concentration was 0.66 ± 0.014 mg/kg, decreasing to 0.24 ± 0.02 mg/kg at 30 cm. This trend continued with depth, reaching 0.176 ± 0.003 mg/kg at 40 cm, 0.11 ± 0.003 mg/kg

Fig. 4 THg vertical concentrations ((mean±s)) of sediment samples at various sampling points (Mugusu 1–4 and Nungwe floodplain) along the Mabubi River, which flows adjacent to the Mugusu artisanal mine to Lake Victoria



at 50 cm, and the lowest concentrations observed at 70 cm (0.048 ± 0.01 mg/kg) and 80 cm (0.082 ± 0.006 mg/kg). This general trend of decreasing THg concentrations with depth was consistently observed across all mining sites.

River sediments (Fig. 4) sampled along the Mabubi River exhibited varying trends depending on the sampling points. Downstream of the Mugusu mine (Mugusu 1 to 4 and Nungwe floodplain), the area was characterized by a muddy and boggy environment. Mugusu 1, a point located at start of the mining village, showed relatively elevated average concentrations in the 20 cm (0.38 ± 0.02 mg/kg) and 30 cm (0.41 ± 0.02 mg/kg) surface sediments. From 40 to 100 cm depth, average concentrations ranged from 0.21 ± 0.01 mg/kg at 40 cm to 0.16 ± 0.003 mg/kg at 100 cm. However, slightly higher average concentrations were observed at specific depths, such as 70 cm (0.21 ± 0.03 mg/kg) and 90 cm (0.24 ± 0.05 mg/kg).

Mugusu 2 was a sampling point adjacent to the mining village, close to the village portable water source. It is influenced by the riverbank slope from mine processing activity sites. Due to challenges in segregating the top 20 cm of sediment, the surface layer from a depth of 30 cm was used for analysis. The layer showed elevated concentrations of $\sim 1.29 \pm 0.02$ mg/kg, which then decreased to about 1.0 ± 0.05 mg/kg at both depths of 40 cm and 50 cm respectively. The decrease was also observed at 60 cm (0.44 ± 0.03 mg/kg) and 70 cm (0.32 ± 0.002 mg/kg). At 80 cm there was a slight increase to 0.50 ± 0.05 mg/kg. At 100 cm, the concentration was 0.34 ± 0.03 mg/kg. The general trend was a decrease of concentration with increase of depth, although the surface concentration decrease was gentle.

Mugusu 3, like Mugusu 2 was a sampling point adjacent to the mining village, downstream. It exhibited a decrease in concentration similar to many of the soil samples discussed earlier. The surface sediment layer (20 cm) showed an average THg concentration as high as 2.2 ± 0.02 mg/kg. However, this concentration decreased by about half to 1.2 ± 0.09 mg/kg at the 30 cm depth. From 40 to 100 cm, the THg concentrations was relatively constant with concentration value of about 1.0 mg/kg, except at 80 cm, where 1.5 mg/kg was recorded. In contrast, at Mugusu 4, the concentration trend was more consistent, with relatively equal values observed from a depth of 20 cm to 50 cm, but a spike was observed at 40 cm.

Nungwe floodplain showed a nearly constant (~ 0.1 mg/kg) distribution of THg concentrations in sediments from 20 to 80 cm in depth. The highest recorded average value for the profile was approximately 0.17 ± 0.006 at 20 cm depth, while the slightly lower average value of 0.10 ± 0.003 was recorded at 50 cm depth. At 80 cm, the deepest sediment sample the concentration was 0.16 ± 0.006 , which was

almost identical to the concentration value recorded in the surface sediment (20 cm).

Discussion

The observed spatial and vertical distributions of total mercury (THg) in soil and sediment samples across the mines provide crucial insights into the complexities of mitigating the long-term impacts of mercury residues in the environment. The distribution highlights the challenges of reducing the environmental burden of mercury residues where mercury's spatial dispersion, extends far from the point of introduction. The typical example is the influence of Mugusu mine influencing Mabubi River for approximately 10 km downstream through a vast floodplain.

From this investigation it is revealed that although soils and riverine sediments in all areas associated with artisanal gold mining activities exhibit mercury residue contamination, there are exceptions. For example, it is observed that at the Rwamgasa mine (Fig. 3), THg levels in deeper soil layers (70 cm: 0.048 ± 0.01 mg/kg; 80 cm: 0.082 ± 0.006 mg/kg) were within the global average background concentration of < 0.1 mg/kg, (Erickson et al. 2006). This shows that deeper soils at Rwamgasa are less contaminated compared to other artisanal mining sites.

The relatively higher Total Mercury (THg) concentrations in surface (20 cm) soil samples supports the long-time persistence emanating from previous intense mercury use which has also been observed by different authors (Loewen et al. 2005; Bugmann et.al, 2022 and Dhiman 2023). At all mines sites, surface layers (20 cm) had relatively higher Total Mercury (THg) concentrations. For instance, at Mgongo mine (Fig. 2), showing the concentration of 2.1 mg/kg, Nyarugusu mine (1.5 mg/kg), Sekenke (0.8 mg/kg), and Rwamgasa (0.7 mg/kg). They all indicate the direct deposition from gold processing activities where open air heating of Hg-gold amalgam vapourize mercury. The released mercury vapor disperses into the atmosphere and, because of its high density (≈ 13.5 g cm⁻³), it is likely to condense and settle easily. A significant portion of mercury remains in the residues and tailings, which are often discharged into the environment as has been reported by other investigators (e.g. Schwartz, et al. 2023; Esdaile and Chalker 2018, etc.). This trend also reflects the varying levels of mining activity at the sites, as detailed in Table 1. Mgongo mine, in particular, was observed to have exceptionally active mining and processing operations, accompanied by a large human settlement.

Among the five mines investigated, the Nyarugusu artisanal mine is the oldest, with mercury use dating back to the 1970s. As presented in Table 1, the mine is no longer a

"gold-rush" site; however, mercury residues persist at significant levels. The THg concentration, approximately 1.5 mg/kg in surface soil (sampled at 20 cm), may be influenced by ongoing reprocessing activities of previously mined drainages and slurries. Notably, THg residues remain detectable at deeper layers, extending up to 100 cm. This persistence likely reflects the long-term environmental impacts of historical mining and processing activities at the site. Such long-lasting impacts are a common pattern associated with mining activities, as demonstrated by the legacy of the California gold rush (1840s–1880s), investigated between 1999 and 2001 (Alpers et al. 2004). These findings emphasise this study's prediction of the enduring presence of mercury residues in the environment at all mines, further compounded by the continued release of mercury.

The distribution of mercury in rivers has been correlated with total suspended solids concentrations in another study e.g. Balogh et al. (1998). However, there has been no comprehensive analysis of mercury vertical distribution in river sediments. In this study, river sediments in Mabubi River (at Mugusu 1–4 and Nungwe floodplain) show a lack of specific pattern of THg concentration (Fig. 4) implied the influence of river dynamics on the mixing of sediment and mercury residues transport. This has also been reported by Cui et al., (2024) in sediments of Wuliangshuai Lake in Mongolia. A good example is shown by surface sediments (sampled at 20 cm depth) concentration pattern of 0.3 mg/kg, 5.8 mg/kg, 2.2 mg/kg, and 0.4 mg/kg for successive downstream sampling points, respectively. This was an indicator of the dominance of river dynamics, which are non-specific in determining the THg distribution along the river. The consistently higher concentrations at Mugusu 2 and Mugusu 3, both at the surface and in deeper sediments, can also be explained to be due to the impact of tailings from gold processing activities (uphill). From field observation, Mugusu mining village terrain facilitates the flow of sediment into the river. The proximity of the two sampling stations (Mugusu 2 and Mugusu 3) to intense processing activities likely influenced the higher mercury spikes.

The results indicating THg ~0.2 mg/kg at Nungwe floodplain (~10 km from the mine and processing site), highlights the strong transport of mercury residues over long distances, facilitated by sediment flow and mixing processes. Results from the Nungwe floodplain reveal evidence of vertical mixing, likely driven by ongoing physical activities, particularly extensive rice farming in the area. The bay, which is utilised as a rice paddy and animal grazing area, is continuously replenished with water and sediments from the Mabubi River, maintaining a near-constant mercury concentration (~0.2 mg/kg) that points to ongoing sediment mixing.

Mercury is released into the aquatic environment, where biological activities may influence its vertical distribution. In dynamic river and floodplain systems like Nungwe floodplain, numerous biological, chemical, and physical processes, though not directly evaluated in this study, may facilitate interconversions between mercury species. For instance, Brown et al. (2015) observed in an estuarine environment that surface layers (samples from 10 cm deep) showed active mixing, while deeper layers had lower concentrations of inorganic and organic (MeHg) mercury due to interactions with Fe(III) hydroxide. Although mercury speciation was not assessed in this study, the consistent THg concentrations observed in Nungwe floodplain suggest the presence of similar mixing processes within this environment.

A general observation of THg concentration in the Mabubi River channel, can explain long-term transformation between Mugusu 1 and Nungwe floodplain. Although hydrological dynamics (flooding, sedimentation, and water-level fluctuations) was not assessed, literature information can be used to explain the likely processes that may affect the environment through THg mobility and methylation risks. Lake Victoria's ecological health, in particular, is at higher risk due to the transport of mercury from mining sites, such as the Mugusu mine, which is interconnected through the Mabubi River. Given Lake Victoria's critical role in food security and the economic stability of the region, monitoring the dynamics of mercury pollution and its ecological impacts is essential.

Results from Mabubi River channel sampling points provide key observations on Hg distribution and hydrological influence. These results are explained in the summary in Table 2, where Mugusu 1–4 are characterized as sites exposed to the erosive forces of mining activities, which can allow Hg suspension and transport downstream. Although hydrological dynamics such as flooding and sediment resuspension were not directly measured, previous studies (De Lacerda and Salomons 1998; Grigal 2003) suggest that these processes may explain the observed downstream persistence and redistribution of THg. This process can also lead to the washing and mixing of buried Hg, as observed at Mugusu 2 (30–50 cm) and Mugusu 3 (60–80 cm). In the Nungwe floodplain, low THg concentrations suggest limited retention of contaminated sediments, likely because most THg is transported further downstream into Lake Victoria. Although the floodplain shows low THg concentrations, it remains a potential methylation hotspot due to waterlogged soil conditions.

Table 2 Trend of key observations from Mabubi River sediment THg distribution, likely influenced by hydrological processes from upstream (Mugusu 1) to the Nungwe floodplain, which connects to Lake Victoria

Site	Observed THg trends	Interpretation
Mugusu 1	Peaks at 30 cm (0.41 ± 0.0209) Declines with depth	Recent deposition from upstream mining; possible erosion of surface Hg during floods
Mugusu 2	High Hg at 30–50 cm (1.29 ± 0.02 – 1.01 ± 0.05 mg/kg) Drops at 60 cm	Legacy contamination buried under newer sediments; redox cycling likely and resuspended sediments
Mugusu 3	Extremely high Hg at 20 cm (2.20 ± 0.02 mg/kg) Stays elevated at depth	Major contamination source (resuspension); Hg retained due to organic matter
Mugusu 4	Spike at 40 cm (1.2 ± 0.073 mg/kg) Lower above/below	Possible physical activities (Gardening) spill or flood deposit
Nungwe Floodplain	Very low Hg (0.1 ± 0.003 – 0.17 ± 0.006 mg/kg), uniform with depth	Acts as a Hg sink (traps particles during floods), human activities influence (rice farming, and <u>animal grazing</u>)

Conclusion

From this study, it can be concluded that mercury residues are likely to persist long-term in ASGM mining areas and other contaminated environments. Since all forms of mercury are known to have negative environmental consequences, the transport of its residues from disposal sites and their transformation into different chemical forms contribute to its widespread spatial distribution. The example of the California gold rush-related mercury residues cited herein, along with the observed scenarios of this study, highlights not only spatial distribution of total mercury (THg) residues, but also deep into soils and sediments, and the sizes of the mining villages indicate challenges in remediating mercury-contaminated sites. From Table 1, the surface areas of some mines are substantial enough to limit the feasibility of soil mobilization. For example, Nyarugusu, the oldest of the artisanal mines, covers the largest area (~ 6 km²), with THg residues of 0.2 mg/kg recorded at depths of up to 100 cm. Mgongo mine spans approximately 2.5 km², while Rwamgasa is estimated at 3.0 km². Mobilizing THg-contaminated soil is crucial for ensuring safe human settlements, given the residues recorded at various depths. However, implementation of large-scale soil mobilization remains impractical. This indicates that mercury residues are likely to have long-lasting impacts on mining sites and areas influenced by the mining activities, even with the enforcement of the Minamata Convention in the United Republic of Tanzania.

Consequently, mercury contamination is expected to continue affecting human health for the foreseeable future.

This investigation indicated that remediation of contaminated sites are highly limited by various factors cited in this work. Meanwhile there are other recognised limitations. Worldwide, efforts to remediate metal-contaminated sites have faced significant challenges, with persistent technological limitations and capacity gaps hindering effective solutions. Several tested remediation methods, such as those by Xu et al. (2019) employing a recirculating soil washing system and electrochemical filtration, as well as studies by Hidayati et al. (2009), Moreno et al. (2004), and Tiodar et al. (2021), have explored the use of plants (phytoremediation). However, limitations such as long operation times, high chemical costs, large energy consumption, secondary pollution, and soil degradation have been reported, challenging the viability of these remediation technologies. While remediation has mostly been successful at the experimental scale, field applications for large areas and deeper soils, as observed in this investigation, cannot yet be considered a viable alternative approach.

The general observation is that the Mugusu mine, whose mercury residues are carried into Lake Victoria via the Mabubi River, will continue to serve as a source of contamination for the lake. Further investigations are urgently needed to reveal the safety of consumers of fish from Nungwe bay and rice cultivated within the Nungwe floodplain.

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Data Availability The data used in this publication are available for review and can be shared to enhance knowledge upon request.

Declarations

Conflict of interest The publication of this manuscript is not associated with any agency employment benefits or losses, nor does it have any financial impact on shares or other benefits to any party upon its publication. No undisclosed conflicts of interest exist.

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