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Assessment of Mercury Contamination in Water and Soil from Informal Artisanal Gold Mining: Implications for Environmental and Human Health in Darmali Area, Sudan

Ahmed Elwaleed [1,2], Huiho Jeong [3], Ali H. Abdelbagi [2], Nguyen Thi Quynh [1], Willy Cahya Nugraha [3,4], Tetsuro Agusa [3], Yasuhiro Ishibashi [3] and Koji Arizono [3,5,*]

[1] Graduate School of Environmental and Symbiotic Sciences, Prefectural University of Kumamoto, Tsukide 3-1-100, Higashi-Ku, Kumamoto 862-8502, Japan; g2175005@pu-kumamoto.ac.jp (A.E.)
[2] Mining Engineering Department, Faculty of Engineering, University of Khartoum, Gamma Ave., Khartoum P.O. Box 321, Sudan; ali.abdalbagi@uofk.edu
[3] Faculty of Environmental and Symbiotic Sciences, Prefectural University of Kumamoto, Tsukide 3-1-100, Higashi-Ku, Kumamoto 862-8502, Japan; huiho.jeong@health.ny.gov (H.J.)
[4] Research Center for Environmental and Clean Technology, Nasional Research and Innovation Agency, Kawasan Sains Teknologi BJ Habibie Gd.720, Setu, Tangerang Selatan 15314, Indonesia
[5] School of Pharmaceutical Sciences, Kumamoto University, 5-1 Oe-honmachi, Chuo-ku, Kumamoto 862-0973, Japan
* Correspondence: arizono@kumamoto-u.ac.jp

Abstract: Mercury contamination stemming from artisanal and small-scale gold mining (ASGM) operations poses significant environmental and health concerns. This study focuses on the Darmali area in River Nile State, Sudan, where the reprocessing of amalgamation tailings has led to mercury contamination. This study assessed the mercury content in soil and tailings samples, as well as in tap and groundwater, to evaluate the human health risks from ASGM activities and assess contamination levels within the study area. Soil and water samples were collected from various locations, including agricultural, residential, and tailings sites, as well as groundwater and tap water from the Nile. Mercury analysis was conducted using MA-3000 (NIC), and geo-accumulation index analysis revealed extreme pollution levels in areas with tailings and moderate pollution levels in agricultural and residential areas. Hazard quotients were applied to assess health risks, with inhalation of mercury vapor identified as the primary exposure route. The results indicated that tailings pose significant health risks, particularly for children, while water samples and soil from agricultural and residential areas did not pose significant risks. These findings underscore the urgent need for authorities and local communities to address mercury contamination by removing and treating tailings from affected areas to mitigate health risks.

Keywords: mercury; ASGM; hazard quotient; health risk assessment; tailings; geo-accumulation; Sudan

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1. Introduction

Mercury is widely recognized as the most hazardous heavy metal found in the environment [1]. While it occurs naturally, human activities have significantly increased its presence in aquatic and terrestrial ecosystems [2,3]. Artisanal and small-scale gold mining (ASGM) stands as the primary anthropogenic source of mercury emissions, accounting for about 38% of total global emissions [4].

Different attempts were made to reduce mercury usage in ASGM, one of these strategies is to formalize the ASGM activities as recommended by the Minamata Convention on Mercury [5]. Artisanal gold mining in Sudan operates within a legal framework established by certain regulations, where production sites are formalized and monitored by authorities. However, the processing of ASGM ore is prohibited at these production sites. Instead, miners are required to transport their produced ores to ASGM processing facilities known as Mining Markets, which are developed by the miners themselves and regulated by the state [6]. Here, the use of mercury is permitted. Following the amalgamation process, the remaining gold in the tailings is of interest to cyanide facilities, which purchase these tailings and treat them with sodium cyanide before disposing them into tailing dams in accordance with guidelines. Cyanide is widely used as the leaching agent in gold mining due to its high efficiency in gold extraction, reliability, and comparatively low costs [7].

However, in the study area, Darmali and neighboring villages, miners engage in informal activities by smuggling tailings from the mining markets. They then treat them using sodium cyanide or gold dressing agents (GDA) (Figure 1), along with cyanidation tanks (Figure A1). Despite being marketed in Sudan and various parts of the world as environmentally friendly materials, GDAs have been found to contain about 17–27% of sodium cyanide according to the Department of Energy, Mines, Industry Regulation, and Safety, Government of Western Australia [8]. The coupling leads to increased environmental and health repercussions [9].

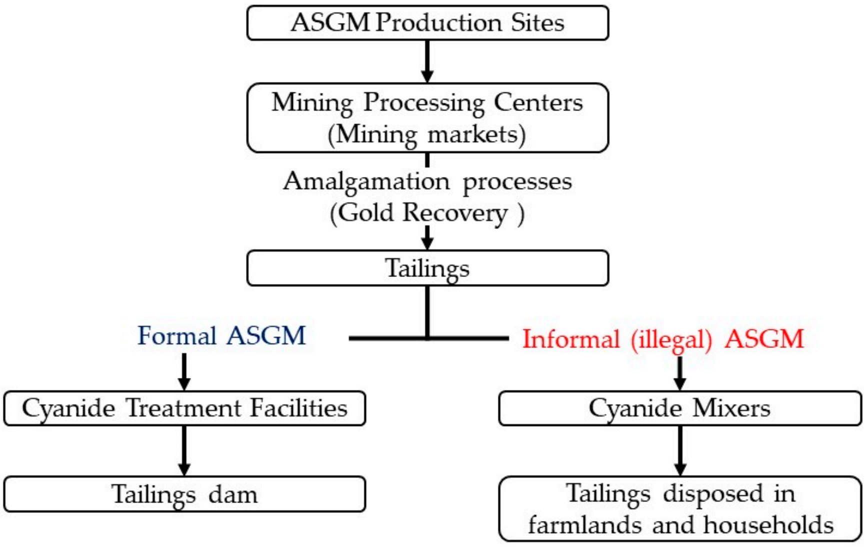

Figure 1. ASGM ore journey, from ore to tailings. Source: flowchart generated based on authors' process observation.

Improper tailings management may lead to the formation of mercury cyanide complexes [10]. When mercury combines with cyanide, it forms a soluble complex known as $Hg(CN)_2$, which has demonstrated potential bioavailability to aquatic organisms. Furthermore, these compounds exhibit remarkable stability and have the potential to cause harm and fatalities in mammalian organ systems [11]. $Hg(CN)_2$ is a robust and persistent cyanide complex, remaining stable even at pH levels below 9. Consequently, removing this complex from effluents can be challenging [12,13].

Moreover, conventional cyanide methods are not strictly followed in the study area (Figure 2). Due to the high cost of electrowinning cells, miners resort to activated carbon burning to recover gold without further processes [14]. Activated carbon also absorbs mercury [15–17]. Consequently, mercury evaporates into the atmosphere, contaminating nearby areas and posing risks to a larger population and a wider geographical area [18].

Atmospheric mercury is not the sole pathway through which mercury is transported across the environment [19]. It circulates and is recycled between major environmental compartments, including air, soil, and water [20]. In the Darmali study area, tailing piles are dispersed across the landscape. They are stockpiled on farms and even within residential properties, subjecting the local population to direct exposure. These tailings are vulnerable to wind and water erosion [21], facilitating their dispersal over a wider geographic area.

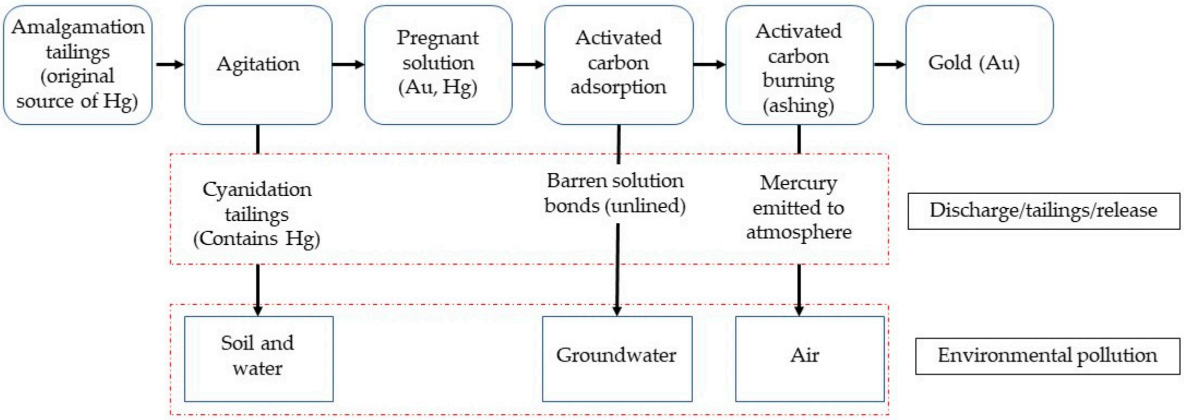

Figure 2. Cyanidation method used in the study area and possible Hg contamination. Source: flowchart generated based on authors' process observation.

The presence of these tailings that are poorly managed creates additional contamination pathways [22]. For instance, soil contamination can lead to the spread of pollutants into other environmental compartments, such as surface water [23]. When surface water is contaminated, it can subsequently lead to groundwater contamination through the focused recharge process [24]. Given this interconnectedness, addressing the tailings and their environmental impact is critical for mitigating the broader spread of mercury and reducing health risks.

In the study area, the groundwater is widely used for irrigation [25]. The shallow aquifers are unconfined and have high permeability [26]. This makes them prone to contamination from various sources, including ASGM activities. The area is prone to seasonal floods [27], which exacerbates the risk of contamination. Poor infrastructure contributes to flood and rainwater accumulation on the eastern side of the area until it evaporates or is mechanically pumped to the eastern side, potentially draining into the River Nile, west of the study area. These floods can lead to the spread of mercury and other pollutants over a wider region [28].

These circumstances highlight the need for comprehensive studies to evaluate the impact of ASGM activities on both the environment and human health. However, research in this field in Sudan is limited, with several gaps still unaddressed. The Minamata Convention on Mercury's Initial Assessment in Sudan identified the lack of data on mercury concentrations in tailings as a research gap [6]. Although some studies have analyzed mercury in ASGM tailings [29], the issue of reprocessed amalgamation tailings remains largely unexplored. This underscores the importance of continued research to fill these gaps and develop effective mitigation strategies.

In the study area, research into ASGM-related pollution has been limited. One study evaluated mercury levels in soil samples but did not include tailings or water samples for mercury analysis [30]. Another study assessed sediment and water samples for heavy metals (As, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Ti, and Zn), but it did not focus on mercury levels. Furthermore, no study has yet evaluated the human health risks in the area [31].

Given these gaps, this study seeks to assess the mercury concentration in reprocessed amalgamation tailings, as well as mercury levels in water samples from the study area. Thus, the research aims to determine the contamination levels and evaluate the associated human health risks in Darmali Area. The goal is to address the existing research gaps and offer insights into the impact of ASGM, ultimately contributing to a better understanding of the environmental and health implications in the region.

2. Materials and Methods

2.1. Study Area

The study area encompasses Darmali town and its neighboring areas north of Atbara City, situated in River Nile State, Sudan (Figure 3). The ASGM operations in the area, particularly known as Darmali Mills, have gained nationwide recognition, although they extend beyond Darmali; the area is characterized by a semi-desert climate, with long summers and low rainfall intensity, averaging 84 mm/year, along with cold, dry winters [32]. The main drainage system comprises the River Nile, Atbara River, and seasonal wadis [32].

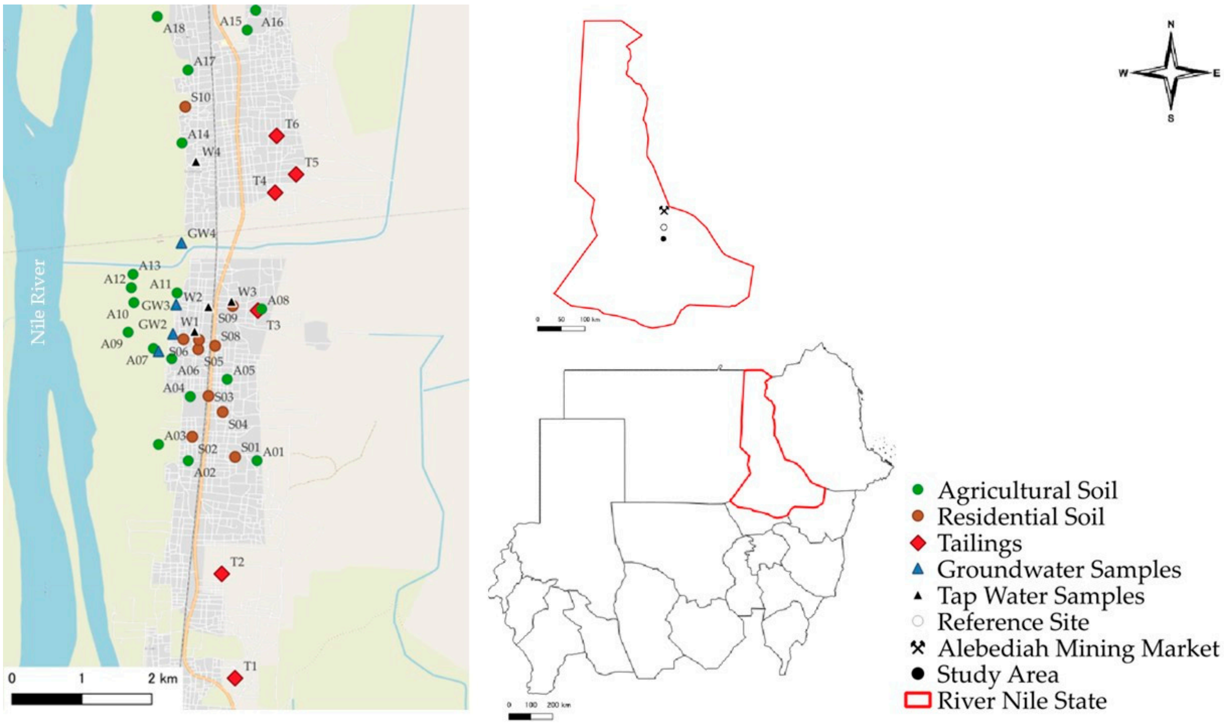

Figure 3. Sampling locations. Source: authors' data.

The study area primarily relies on agricultural production and livestock farming. Additionally, the area is home to cement production facilities and extensive agricultural schemes. A significant feature of the area is the presence of a major artisanal and small-scale gold mining processing center known as Alebediah Mining Market (Figure 3), which supplies most of the amalgamation ore to the Darmali Area. The majority of the population resides along the banks of the River Nile, as it serves as the main lifeline in the area.

Geologically, the study area contains several formations, including the Pre-Cambrian Basement Complex, upper Cretaceous Nubian sandstone formation, Oligocene Hudi Chert, and Quaternary superficial deposits, arranged in ascending order [33]. It features two primary aquifers: a shallow or upper aquifer within the alluvial deposits, ranging from 5 to 37 m thick, and a deep or lower aquifer within the Cretaceous Nubian sandstone, ranging from 17 to 60 m thick. While the upper aquifer is semi-confined, the lower aquifer is nearly confined [34]. Groundwater extraction serves as the primary discharge source, with recharge primarily originating from the main Nile and Atbara rivers. Additionally, direct precipitation at a rate of 41.9 mm per year and natural base flow contribute to recharge [26].

2.2. Sampling

Sampling was conducted in January 2023, during which a total of 35 samples were collected from various locations representing different land uses. These samples consisted of 18 agricultural soil samples (depicted as A01–A18), 10 samples from residential areas (depicted as S01–S10), 6 samples from tailings sites (designated as T01 to T06), and 1 reference sample (R01) within the study area, as depicted in Figure 3.

Soil samples were chosen randomly to encompass a broad geographic area, while tailings samples were selected based on accessibility to the tailings' areas. The soil samples were collected from a depth of 0–15 cm.

Additionally, eight water samples were collected, consisting of four groundwater samples (as depicted in GW1–GW4) and four tap water samples (as depicted in W1–W4) drawn from the River Nile via the water network distribution.

2.3. Analytical Procedure

2.3.1. Sample Preparation

All water samples were initially treated on-site by adding HNO_3 to achieve a pH level below 2. Following acidification, the samples were carefully sealed, placed in plastic containers, and covered with a protective film before being stored in a cooler for transportation to the laboratory. The samples were transported to Japan for analysis after obtaining special permission from the Ministry of Agriculture, Forestry and Fisheries of Japan (MAFF). Upon arrival at the laboratory, the samples were stored at temperatures below 4 °C until analysis.

For soil samples, an initial sieving process was conducted on-site to remove any debris and non-soil particles. The sieved soil samples were then air-dried at room temperature and subsequently sieved again using a 150 µm mesh to ensure uniformity.

2.3.2. Total Mercury (THg) Analysis

The analysis of total mercury concentration in water samples followed the standardized procedure outlined in method no. 245.1 of the United States Environmental Protection Agency (USEPA) [35]. The samples were analyzed using a Mercury Analyzer (MA-3000) by Nippon Instruments Corporation, Tokyo, Japan.

For soil samples, around 30 mg of air-dried samples were placed in sample boats in triplicate and analyzed using the same mercury analyzer (MA-3000).

2.3.3. Quality Assurance

To ensure the accuracy and reliability of mercury analysis, rigorous quality control procedures were implemented. For water samples, the limit of detection (LoD) was determined using the method detection limit (MDL) method, resulting in an MDL value of 0.00570 µg/L. The subsequent limit of quantification (LoQ) was established at 0.018 µg/L, calculated as ten times the standard deviation (SD). Calibration curves were meticulously prepared for both low and high concentrations, demonstrating exceptional linearity with coefficients of determination (R^2) of 0.9995 and 0.9931, respectively.

To verify the accuracy of the analytical method, recovery tests were conducted by spiking known amounts of mercury into the samples, followed by conducting duplicate analyses. The recovery tests yielded an average recovery rate of 88.5%, with a coefficient of variation of 2.3%. Additionally, individual water samples were analyzed in triplicate, with all samples exhibiting coefficients of variation below 5%.

For soil samples, analyses were performed in triplicate, resulting in a coefficient of variation of less than 9%. Certified reference material (CRM) was utilized, specifically NMIJ CRM 7302-a from the National Metrology Institute of Japan, designed for trace elements in marine sediment. The obtained value of 0.511 ± 0.011 mg/kg corresponded to a recovery rate of $98.45 \pm 2.19\%$, further validating the accuracy and precision of the analytical method employed in this study.

2.4. Geo-Accumulation Index

The quantitative evaluation of mercury contamination in soils involved calculating geo-accumulation indexes (I_{geo}) using Equation (1) as proposed by Muller [36]. This calculation utilized background values established for the continental earth crust (Hg = 0.04 mg/kg) [37] along with the total mercury concentration observed in both soil and tailings samples. The results can be divided into the classes shown in Table 1.

$$I_{geo} = \log_2 C_{Hg}/1.5 \times B_{Hg}, \quad (1)$$

where,

I_{geo}: geo-accumulation index for Hg;
C_{Hg}: Hg concentration in the soil samples and tailings;
B_{Hg}: mercury background level (0.04 mg/kg);
1.5: the factor used to correct lithogenic effects.

Table 1. The geo-accumulation indexes (I_{geo}) and level of contamination [38].

Class	Value	Soil Quality
0	$I_{geo} \leq 0$	Unpolluted
1	$0 < I_{geo} \leq 1$	Unpolluted to moderately polluted
2	$1 < I_{geo} \leq 2$	Moderately polluted
3	$2 < I_{geo} \leq 3$	Moderately to heavily polluted
4	$3 < I_{geo} \leq 4$	Heavily polluted
5	$4 < I_{geo} \leq 5$	Heavily to extremely polluted
6	$5 < I_{geo}$	Extremely polluted

(Source: see References).

2.5. Risk Assessment

Risk assessment is an essential procedure that entails assessing the probability and potential scale of unfavorable occurrences on health, safety, or the environment over a defined period [39].

The USEPA Exposure Assessment Model [40] was utilized to determine the Average Daily Intake (AvDi) of mercury in soil and water samples (mg-Hg/kg-body weight/day). This assessment accounts for exposure through various routes for soil, including ingestion, dermal contact, inhalation of particulates containing mercury, and inhalation of mercury vapors (volatile mercury) [41]. For water samples, ingestion and dermal contact were considered as the primary exposure routes, as outlined in Equations (2)–(5). Detailed input parameters for this study can be found in Table 2.

To assess the population's potential exposure to mercury in comparison to a standard exposure level (reference dose RfD), the hazard quotient (HQ) is calculated using Equation (6). An HQ exceeding 1 signifies an unacceptable risk of adverse non-carcinogenic health effects, whereas an HQ below 1 indicates an acceptable level of risk. When assessing multiple exposure routes, the Hazard Index (HI) is utilized (Equation (7)). An HI greater than 1 indicates an unacceptable risk, while an HI below 1 suggests an acceptable risk level [42,43].

$$\mathrm{AvDi}(\mathrm{ing\_soil}, \mathrm{ing\_water}) = \frac{\mathrm{Cs,w} \times \mathrm{IRs,w} \times \mathrm{ED} \times \mathrm{EF} \times \mathrm{CF}}{\mathrm{BW} \times \mathrm{AT}} \quad (2)$$

$$\mathrm{AvDi}(\mathrm{der\_soil}, \mathrm{der\_water}) = \frac{\mathrm{Cs,w} \times \mathrm{SA} \times \mathrm{AF} \times \mathrm{ABS} \times \mathrm{ED} \times \mathrm{EF} \times \mathrm{GI} \times \mathrm{CF}}{\mathrm{BW} \times \mathrm{AT}} \quad (3)$$

$$\mathrm{AvDi}(\mathrm{vap\_soil}) = \frac{\mathrm{Cs} \times \mathrm{IRa} \times \mathrm{ED} \times \mathrm{EF}}{\mathrm{VF} \times \mathrm{BW} \times \mathrm{AT}} \quad (4)$$

$$\mathrm{AvDi}(\mathrm{inh\_soil}) = \frac{\mathrm{Cs} \times \mathrm{IRa} \times \mathrm{ED} \times \mathrm{EF}}{\mathrm{PEF} \times \mathrm{BW} \times \mathrm{AT}} \quad (5)$$

$$\mathrm{HQ} = \mathrm{AvDi}/\mathrm{RfD} \quad (6)$$

$$\mathrm{HI} = \Sigma(\mathrm{HQ1} + \mathrm{HQ2} + \mathrm{HQ3} + \mathrm{HQ4}) \quad (7)$$

Table 2. Input parameters for evaluating the Average Daily Intake (AvDi) and hazard quotient (HQ).

Parameters		Unit	Children	Adults	Reference
Cs	Mercury concentration in soil samples	mg/kg	-	-	This Study
Cw	Mercury concentration in water samples	mg/L	-	-	This Study
BW	Body weight	Kg	15	70	[44]
AT	Averaging time	Days	2190	10,950	[44]
EF	Exposure frequency	days/year	350	250	[44]
ED	Exposure duration	Years	6	30	[44]
IRs	Ingestion rate of soil	mg/day	200	100	[44]
IRa	Ingestion rate of air	m^3/day	10	10.4	[44]
IRw	Ingestion rate of water	L/day	2	3.45	[45]
GI	Gastrointestinal adsorption factor	-	1	1	[44]
PEF	Particulate emission factor	m^3/kg	1.30×10^9	3.22×10^8	[44]
VF	Volitilisation factor	m^3/kg	8028.297	8028.297	[44]
SA	Surface area exposed skin	cm^2	2100	13,110	[44]
AF	Skin adherence factor	mg/cm^2/day	0.2	0.07	[44]
ABS	Dermal absorption factor	-	0.1	0.1	[44]
CF	Conversion factor	-	0.000001	0.000001	[44]
RfD_o	Reference dosage (Oral)	mg/kgbw/day	0.0003	0.0003	[44]
RfD_i	Reference dosage (Inhalation)	mg/kgbw/day	0.000086	0.000086	[44]
RfD_d	Reference dosage (Dermal)	mg/kgbw/day	0.0003	0.0003	[44]

(Source: see References).

2.6. Statistical Analysis

Descriptive statistics were employed to assess the mercury content of the samples. The Shapiro–Wilk and Kolmogorov–Smirnov tests were used to check for the normality of the mercury concentrations, supplemented by graphical methods of histograms and Q-Q plots. To compare the median values across different groups, the Mann–Whitney U test was utilized. Statistical analyses were performed using IBM SPSS 26 (SPSS Inc., Chicago, IL, USA). Microsoft Excel 2019 was utilized in the calculations of the geo-accumulation index and health risk assessment. Graphs were generated using OriginPro 2024 (version 10.1.0.170), and the map was produced using Quantum GIS (QGIS 3.32).

3. Results and Discussion

3.1. Mercury Analysis

3.1.1. Mercury Concentration in Soil and Tailings

The mercury concentration varied among the samples from different land uses, with residential areas exhibiting the least median mercury concentration (0.044 mg/kg), followed by agricultural areas (0.057 mg/kg), and tailings (9.6 mg/kg), as depicted in Figure 4.

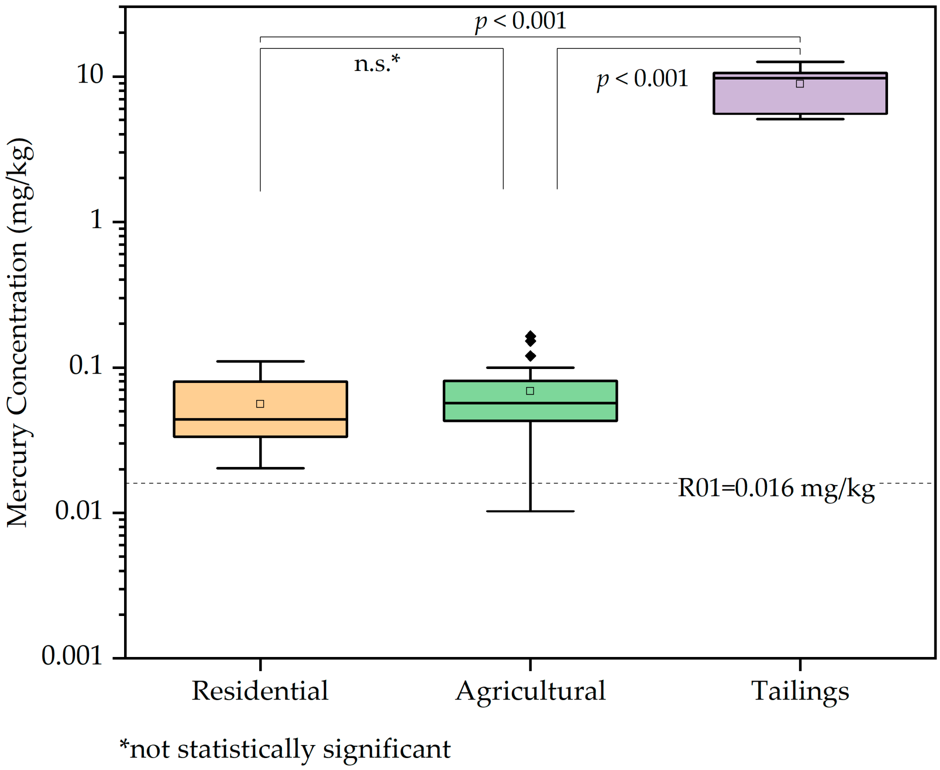

Figure 4. Mercury concentration in soil samples categorized by different land uses. Source: authors' data.

The median concentrations of all sample groups exceeded the reference point concentration (0.016 mg/kg). In residential areas, every individual sample surpassed this threshold, while 94.4% of farmland samples (17 out of 18 samples) also exceeded it. Since the mercury concentration in the reference sample is considerably low, another benchmark for comparing median mercury concentrations is the value of mercury in the continental crust (0.04 mg/kg) [37]. In the residential group, 60% of the individual samples surpassed this threshold, while in the agricultural areas group, 83.3% of the individual samples exceeded this benchmark, and all individual samples in the tailings group surpassed this value.

The median concentrations between the agricultural areas group and the residential group were not statistically different (p = 0.43). However, statistically significant differences were observed between the tailings group and both the agricultural areas (p = 0.005) and residential groups (p = 0.002). This implies that residential areas and farmlands are not influenced by the tailing dumps as a pollution source. It appears that there is almost no effect of mercury pollution in both groups, which is further confirmed by the results of the geo-accumulation index analysis (I_{geo}) (Section 3.2). Consequently, there seems to be no significant spatial variation in mercury contamination among these groups.

In the study area, previous research that investigated mercury pollution due to ASGM revealed that the maximum mercury concentration detected in the discharge ponds reached 2.62 mg/kg [30]. Interestingly, this concentration contains nearly 50% of the lower end of the range of mercury concentrations found in the tailings in the current study (5.01 mg/kg). Another study in the Abu Hamad Mining Market [29] exhibited higher mercury concentrations in tailings samples, measuring 19.0 mg/kg, which is relatively higher than the concentration found in this study. This difference may be attributed to the nature of the tailings in the present study, which primarily consist of reprocessed amalgamation tailings. In the reprocessing process adopted for this study area, mercury can be captured and adsorbed in activated carbon [13], leading to the potential loss of mercury to the atmosphere when the activated carbon is burnt.

In Sudan, specific guidelines for mercury concentration in soil have not been established. To account for this, the Canadian soil quality guidelines for the protection of environmental and human health have instead been adopted, setting the limit at 6.6 mg/kg for agricultural and residential areas [46]. The presence of tailings with concentrations ranging from 5.1 to 12.6 mg/kg in residential and agricultural areas exceeds this established limit. It is worth noting that mercury concentration limits in soil vary from one country to another. For example, the United Kingdom has established levels of 1 (mg/kg) for elemental mercury in soil [47], which is significantly lower than the concentrations found in the tailings' samples. Conversely, the Australian national guidelines for contaminated sites allow up to 15 (mg/kg) of elemental mercury in residential properties [48], indicating that the tailings samples fall below this threshold.

Soil serves not only as a pathway for plant growth or a site for waste disposal, but also as a channel for various pollutants to migrate into surface water, groundwater, atmosphere, and food chains [23]. Consequently, the ongoing presence of these tailings in agricultural areas and residential areas will continue to sustain environmental and health risks.

3.1.2. Mercury Concentration in Water Samples

The mean mercury concentration in both groundwater samples and water from the River Nile remained consistent, measuring 0.26 μg/L, closely resembling the level observed in the control sample (0.27 μg/L). Importantly, all individual samples remained below the established safe drinking water limits set by the WHO and Sudan (1 μg/L) [49], as well as those set by other countries like Japan (0.5 μg/L) [50] (Figure 5).

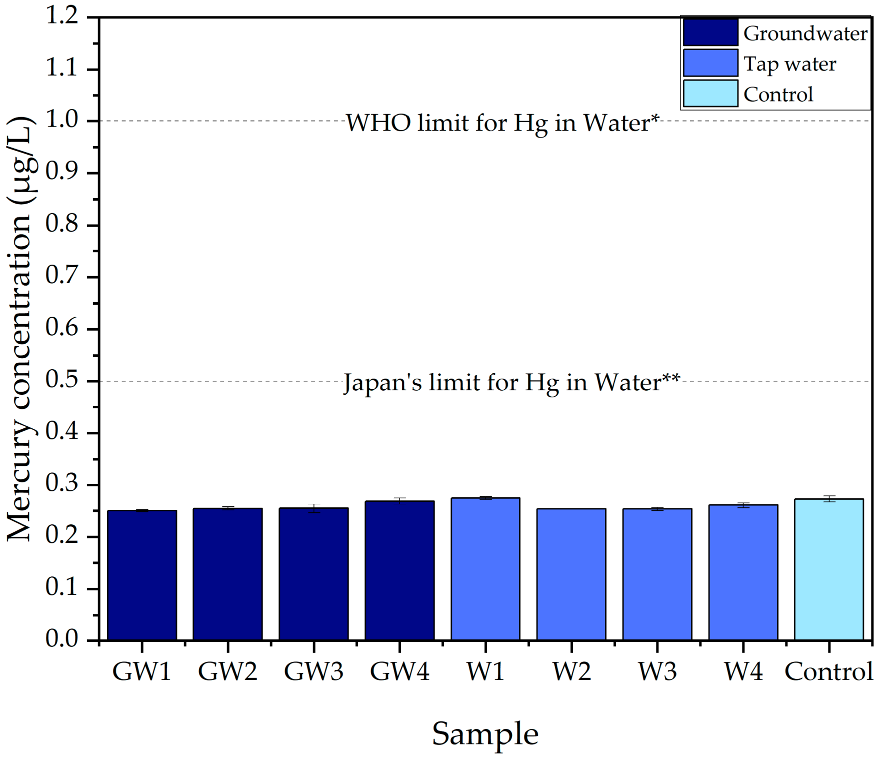

Figure 5. Mercury concentration in water samples. * Based on World Health Organization Guidelines for Drinking Water Quality [48]; ** Based on Drinking Water Quality Standards in Japan [49]. Source: authors' data (see references for additional information).

These findings were previously anticipated, as the water resources are regularly consumed, and sampling was conducted following the prohibition of ASGM activities in the study area. The groundwater resources in the study area, which are utilized for agricultural purposes, are protected by concrete slabs. Additionally, water sourced from the

Nile is pumped to a centralized water tank in the area before being distributed through the water network to households. Therefore, these water resources are not easily susceptible to tailings contamination, further reducing the risk of mercury exposure.

Furthermore, since operations were halted several months prior to sampling, the risk of atmospheric mercury contaminating these resources is significantly diminished, contributing to the overall reduction in contamination concerns among local communities. However, this does not imply that the water resources are entirely risk-free.

Previous research conducted in Darmali Area or its vicinity focused on the heavy metal content in water samples from the River Nile but did not specifically address mercury content [21]. Limited studies have compared water sample concentrations in ASGM areas in Sudan. However, one study conducted in the Abu Hamad Mining Market, where mercury is applied to the whole ore, unlike the amalgamation tailings being reprocessed by leaching as in the study area, found elevated mercury concentrations in water samples collected within the ASGM area. The levels reached a maximum of 3.26 µg/L near the mercury amalgam sites, surpassing thresholds established by global organizations [29].

3.2. Geo-Accumulation Index (Igeo)

The mercury concentration in the continental crust (0.04 mg/kg) served as a reference to assess contamination levels in the study area [37]. The geo-accumulation index (Igeo) classes were utilized to categorize individual samples based on their contamination levels.

As depicted in Figure 6, the cumulative distribution chart of the geo-accumulation Index (Igeo) reveals the distribution of contamination levels across different sample types. Among agricultural soil samples, 12 samples (66.67%) fell within class 0, indicating practically unpolluted conditions. Additionally, three samples (16.67%) were classified as class 1, representing low pollution levels, while three samples (16.67%) fell into class 2, indicating moderate pollution levels. Conversely, among residential soil samples, seven samples (70%) were categorized as class 0, signifying practically non-polluted conditions, and three samples (30%) were classified as class 1, suggesting low to moderate pollution levels. In contrast, the entirety of the six tailings samples were classified as class 6, indicating very strong pollution levels, as expected.

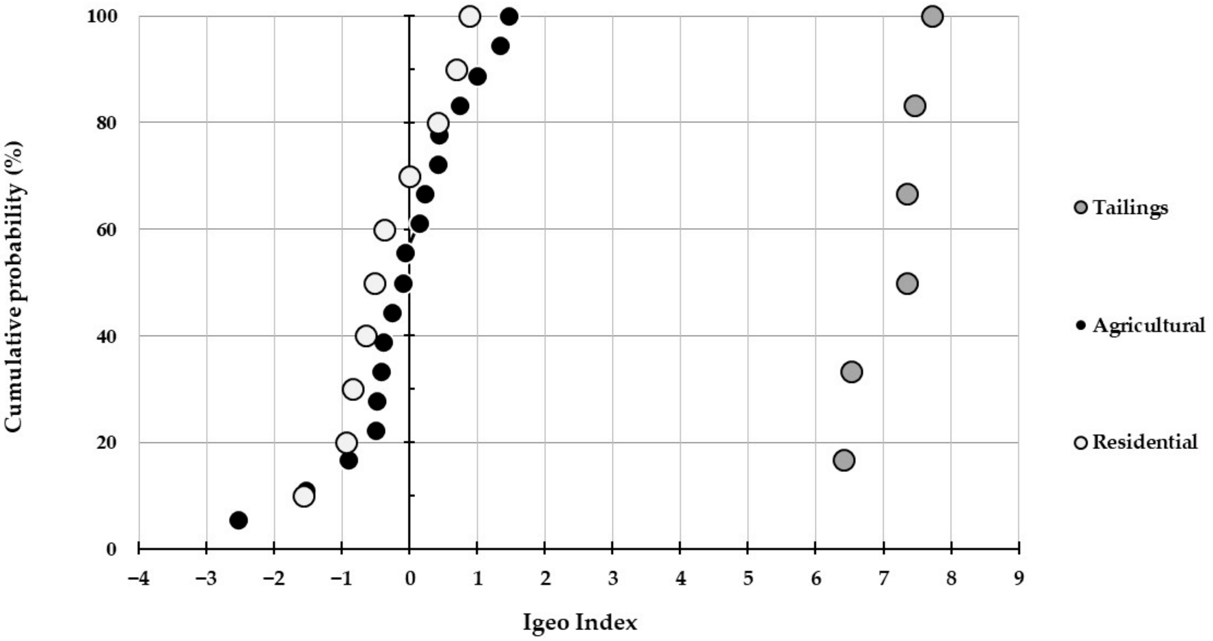

Figure 6. Cumulative distribution chart of the geo-accumulation index (I_{geo}) for the different groups. Source: authors' data.

Overall, the agricultural soil samples exhibited contamination levels ranging from practically unpolluted (class 0) to moderately polluted (class 2), while the residential soil samples ranged from practically non-polluted (class 0) to unpolluted to moderately polluted (class 1). In contrast, entire tailings samples were found to be very strongly polluted (class 6), consistent with expectations.

3.3. Health Risk Assessment

The agricultural and residential soil groups did not pose significant human health risks for both adults and children. However, in the tailings group, except for samples with concentrations of 5.01 and 5.5 mg/kg, the Hazard Index (HI) values exceeded 1 for the adults' group. Furthermore, all tailings samples reported HI values higher than 1 for children (Table 3).

Table 3. HQ and HI values for soil and tailings (adults and children).

Land-Use	**Statistics**	**Adults**					**Children**				
		HQ_{ing} *	**HQ_{Der} ***	**HQ_{Inh} ***	**HQ_{Vap} ***	**HI**	**HQ_{ing} ***	**HQ_{Der} ***	**HQ_{Inh} ***	**HQ_{Vap} ***	**HI**
Tailings (n = 6)	Min	0.017	0.015	1.87×10^{-5}	0.75	0.78	0.217	0.045	2.91×10^{-5}	4.70	4.97
	Max	0.041	0.04	4.62×10^{-5}	1.86	1.93	0.536	0.112	7.19×10^{-5}	11.6	12.29
	Median	0.032	0.03	3.58×10^{-5}	1.44	1.5	0.42	0.087	5.58×10^{-5}	9.03	9.53
Agricultural areas (n = 18)	Min	3.36×10^{-5}	3.08×10^{-5}	3.78×10^{-8}	0.0015	0	0.0004	9.22×10^{-5}	5.89×10^{-8}	0.01	0.01
	Max	0.0005	0.0005	6.04×10^{-7}	0.024	0.03	0.007	0.0015	9.4×10^{-7}	0.15	0.16
	Median	0.0002	0.0002	2.09×10^{-7}	0.008	0.01	0.002	0.0005	3.25×10^{-7}	0.053	0.06
Residential (n = 10)	Min	6.61×10^{-5}	6.07×10^{-5}	7.45×10^{-8}	0.003	0	0.0008	0.0002	1.16×10^{-7}	0.019	0.02
	Max	0.0004	0.0003	4.06×10^{-7}	0.017	0.02	0.0047	0.001	6.32×10^{-7}	0.10	0.11
	Median	0.0001	0.0001	1.62×10^{-7}	0.007	0.01	0.0019	0.0004	2.51×10^{-7}	0.04	0.04

* (ing = ingestion, Der = Dermal, Inh = inhalation, Vap = vapor inhalation). (Source: authors' data based on the US EPA model [40], utilizing Equations (2)–(7)).

The primary exposure pathway was through the inhalation of mercury vapors, aligning with the well-documented findings [51,52]. In contrast, the dermal and ingestion pathways exhibited relatively low risks. The inhalation of solid particles (dust) was almost negligible for both adults and children, similar to the studies [53,54].

Since the water samples were below the safe limit for drinking water, there were no potential health risks posed by mercury contamination for both adults and children. However, the potential human health risks from water contaminated by mercury were a primary concern, particularly considering that the water resources in the study area are shared by a large population. Additionally, the proximity of the River Nile to this area further accentuates the significance of this issue, as it serves as a lifeline for populations downstream [55]. Even the shallow deep aquifers sampled in this study raised concerns for these populations.

It is crucial to recognize that the population exposed in this study area differs from the typical setting of ASGM sites in Sudan. In ASGM processing centers or Mining Markets in Sudan, only men are permitted, with women and children generally not allowed to participate, although there are exceptions where women and children do engage in ASGM activities in some areas of Sudan [56]. However, the setting of ASGM operations in this particular area exposes even more vulnerable groups, such as pregnant women, infants, and elderly people [57,58], to potential mercury risk.

To contextualize the results within the ASGM context in Sudan, a study in the Abu Hamad ASGM center revealed that all individuals in the ASGM center are at risk, primarily from the ingestion and inhalation of mercury vapors [29], which is consistent with the findings of this study regarding the inhalation of mercury vapors. Water samples collected from ASGM processing centers posed significant human health risks, especially for children, even though they are not allowed to be present in those areas [29]. However, individuals in ASGM centers are directly or indirectly involved in ASGM activities. In contrast, the population in this study area is not entirely engaged in ASGM, and many of the ASGM workers were outsiders according to the local community. Hence, it is crucial to educate this population about the repercussions of irresponsibly discarding tailings containing mercury and potentially other contaminants to mitigate exposure to these sources of pollution.

3.4. Recommendations

The continued presence of tailings in the study area poses ongoing environmental and health risks for the local population and resources, particularly concerning land and water [59]. Given that these tailings have been depleted of gold, there is no economic incentive for miners to transport them, thereby placing the responsibility for their management on local communities and authorities.

Identification and intervention in areas affected by ASGM activities and tailings dumping are essential, given that these sites surpass intervention thresholds established by multiple organizations [46–48]. This is vital for sustaining the livelihoods of the population in these areas, as they rely heavily on farming and animal husbandry [60]. Any degradation of the environment will have far-reaching consequences that affect a large number of people and potentially jeopardize future generations.

Miners' financial motivations play a crucial role in how they perceive risk, often leading them to prioritize earning higher incomes over health considerations [61]. This tendency to focus on financial gain over personal safety is a significant challenge in regions where ASGM generates much higher income compared to other occupations [62]. Simply prohibiting ASGM and relying solely on regulations will not be sufficient [63,64].

The study area does not have proven gold ores, yet it has seen significant ASGM processing activity. Farming is the main economic activity, but many miners who operated in the area were outsiders. They often rented farmland or residential spaces to process their ores. Given this, along with the authorities' prohibition of ASGM activities, it is essential to explore alternative economic opportunities [65]. Introducing more sustainable farming practices could provide a viable source of income for the local community, thereby reducing dependency on ASGM [59].

However, while this recommendation presents possible alternatives, other options should also be explored. The involvement of local communities is crucial to ensure the effectiveness of any solution [66].

3.5. Limitations and Future Directions

While this study offers important insights into mercury pollution resulting from ASGM activities in Darmali and its associated health risks, it exclusively focuses on mercury as the primary pollutant, as is typical in ASGM settings. Other potential pollutants commonly associated with ASGM in Darmali, such as cyanide and various heavy metals, were not specifically targeted in this study. However, our future work will explore the presence of cyanide, as well as the potential impact of other heavy metals, within the study area.

4. Conclusions

The findings of this study underscore the critical issue of mercury contamination in the Darmali area, primarily due to ASGM activities. It is the first study to assess mercury levels in reprocessed amalgamation tailings and the first to evaluate mercury content in water samples from underground sources and tap water. The results revealed significantly elevated mercury concentrations in tailings samples, while the soil from agricultural and residential areas had considerably lower levels of contamination. Notably, mercury concentrations in water samples were below the safety thresholds set by Sudanese standards, though this does not guarantee complete safety, as contamination can still occur through other pathways or changing conditions. Using the geo-accumulation index (I_{geo}), the study found that tailings samples exhibited extreme pollution levels, indicating the need for immediate intervention. Soil samples from agricultural and residential areas, on the other hand, showed low to moderate pollution levels. In the first-ever mercury human health risk assessment for the study area, the inhalation of mercury vapor was identified as a significant exposure route, with children being particularly vulnerable. Given the high contamination levels in tailings and the associated health risks, the study recommends urgent remediation, focusing on the removal of tailings dumped throughout the area. Collaboration between local communities and authorities is crucial to ensure the successful identification and cleanup of contaminated sites. These efforts will help mitigate the environmental and health risks and safeguard the well-being of future generations in the Darmali area.

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Appendix A

Figure A1. Tank agitators used in the study area. Source: authors' data. Photo taken in January 2023.

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