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Public health assessment of Kenyan ASGM communities using multi-element biomonitoring, dietary and environmental evaluation

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ABSTRACT

The Kakamega gold belt's natural geological enrichment and artisanal and small-scale gold mining (ASGM) have resulted in food and environmental pollution, human exposure, and subsequent risks to health. This study aimed to characterise exposure pathways and risks among ASGM communities. Human hair, nails, urine, water, and staple food crops were collected and analysed from 144 ASGM miners and 25 people from the ASGM associated communities. Exposure to PHEs was predominantly via drinking water from mine shafts, springs and shallow-wells (for As>Pb>Cr>Al), with up to 366 μg L^{-1} arsenic measured in shaft waters consumed by miners. Additional exposure was via consumption of locally grown crops (for As>Ni>Pb>Cr>Cd>Hg>Al) besides inhalation of Hg vapour and dust, and direct dermal contact with Hg. Urinary elemental concentrations for both ASGM workers and wider ASGM communities were in nearly all cases above bioequivalents and reference upper thresholds for As, Cr, Hg, Ni, Pb and Sb, with median concentrations of 12.3, 0.4, 1.6, 5.1, 0.7 and 0.15 μg L^{-1}, respectively. Urinary As concentrations showed a strong positive correlation (0.958) with As in drinking water. This study highlighted the importance of a multidisciplinary approach in integrating environmental, dietary, and public health investigations to better characterise the hazards and risks associated with ASGM and better understand the trade-offs associated with ASGM activities relating to public health and environmental sustainability. Further research is crucial, and study results have been shared with Public Health and Environmental authorities to inform mitigation efforts.

1. Introduction

Globally, studies have linked various health effects to human exposure to various environmental hazards associated with artisanal and small-scale gold mining (ASGM) (Allan-Blitz et al., 2022; Landrigan et al., 2022; WHO, 2016). Across Africa, the poor and most inefficient ASGM technology used in ore crushing, milling, wetting, sluicing, panning, Hg-Au amalgamation, and Hg-Au amalgam burning to recover gold facilitates the introduction of Hg and other potentially harmful elements (PHEs) into the wider environment, food chains and, subsequently, human systems affecting health (Dooyema et al., 2012; Ondayo et al., 2023b; Plumlee et al., 2013). Consequently, around nine million ASGM workers and those living near ASGM activities in Africa are heavily exposed to PHEs that are both geogenic and anthropogenic. Despite the known risks associated with ASGM, coordinated, interdisciplinary environmental characterisation coupled with public health investigations are uncommon in Africa, with fragmentary snapshot studies often reported. Furthermore, studies are frequently disconnected from attempts to holistically reduce and eliminate hazards in ASGM (Ondayo et al., 2023b). For instance, extensive biomonitoring studies across Africa report exposure to Hg and related health effects among ASGM workers and nearby populations, while few report exposures to multiple hazards or PHEs other than Hg (Basu et al., 2015; Bose-O'Reilly et al., 2020; Dooyema et al., 2012; Ondayo et al., 2023b).

Exposure to most PHEs, even at low doses, can adversely affect human health, well-being, and life quality at a broad scale (Axelrad et al., 2022; Ortega et al., 2021; Simões et al., 2021). Following exposure, PHEs are absorbed, distributed, metabolised, accumulated and eliminated from the human body, ultimately determining the total PHE burden (Mitra et al., 2022). Elimination of PHEs from the human body occurs via urine, faeces, saliva, or breastmilk, while storage of PHEs occurs in keratin tissues (hair, nails, and skin), body fat or bone (Lehman-McKeeman, 2010). Human biomonitoring (HBM) is the most appropriate approach to characterise exposure of the ASGM

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communities to environmental and dietary hazards by directly quantifying their concentrations, metabolites, or reaction products in human tissues or specimens (Schmidtkunz et al., 2019; WHO, 2021a). Blood and urine are the most widely used biological matrices for monitoring human PHE exposure. However, blood is an invasive matrix, has many practical and ethical downsides, and is often limited by poor volunteer response (Alves et al., 2014). For this reason, other biomarkers, notably human hair and nails, which are non-invasive, easy to sample, and with less risk of disease transmission, are preferred (Veiga et al., 2004; Watts et al., 2021). Biomonitoring data directly reflects the total body burden of the hazard or biological effects resulting from multiple routes of exposure over time and spatial trends (Mitra et al., 2022).

Nationwide HBM outside occupational settings are rare across the African continent (Godebo et al., 2019; Watts et al., 2021), unlike Europe (Apel et al., 2017; Černá et al., 2017; Schmidtkunz et al., 2019) and North America (CDC, 2015; Haines et al., 2017; Saravanabhavan et al., 2017), where they are routinely undertaken. Difficulties usually experienced in interpreting HBM data in the context of a health risk assessment need to be fully addressed by developing biomonitoring equivalents (BE) (Hays and Aylward, 2009; Watts et al., 2021) based on the African population data. In Kenya, single-event studies with 49–200 participants commonly reported human exposures to various PHEs in Kisumu, Kendu-Bay, Karungu (Oyoo-Okoth et al., 2010), Mombasa (Etiang'' et al., 2018), Nairobi City, and rural Bungoma (Were et al., 2008). Studies linking HBM data to adverse outcomes in Kenya are still scarce; thus, only a few health-related HBM values have been determined. Previous studies established urinary biomonitoring reference values for PHEs and micronutrients covering Western Kenya (Watts et al., 2021; Watts et al., 2020). However, reference values (RVs) and biomonitoring equivalents (BE) for PHEs and micronutrients specific to the Kenyan population are lacking. Reliable risk assessment and management of ASGM is therefore still complex in Kenya as the country lacks data and harmonised nationwide information concerning exposures, including ASGM-related exposures, of citizens to single and multiple hazards and their interplay with other concurrent environmental and dietary exposures and effects on human health.

In Kenya, the ASGM sector employs over 250,000 people, supports one million dependent family members, and accounts for 60 % of the 0.134–3.6 tons of gold produced annually from 2010 – 2020 (Barreto et al., 2018; CEIC, 2021; planetGOLD, 2021). Kenyan health and safety laws and regulations in the mines were repealed in 2007 when the Occupational Safety and Health Act, 2007 (No. 15 of 2007) was enacted (Tampushi et al., 2021). Significant steps towards legalising and streamlining ASGM countrywide were made by enacting the Kenyan Mining Act (No. 12 of 2016) (Fritz et al., 2018; GoK, 2016). However, both Acts lack specific and comprehensive regulations on protecting the health of ASGM workers and local communities within ASGM settings (GoK, 2016; Ondayo et al., 2023a). Like other developing countries, the ASGM sector in Kenya currently lacks proper enforcement, licensing, management, and monitoring systems (Tampushi et al., 2021), resulting in most of the activities being largely illegal and informal. Even so, like most African countries, Kenya has not ratified several international treaties on health and safety. A recent study found high concentrations of major and trace elements, including potentially harmful elements (PHEs) such as arsenic (As), chromium (Cr), nickel (Ni), and lead (Pb), among others, in the geologically-enriched Kakamega gold belt. The rapidly expanding ASGM activities disperse these elements and introduce mercury (Hg) and cyanide (CN), used in gold recovery, to the broader environment, causing widespread contamination of air, soils, water sources, and sediments. These findings have led to concerns about the possible relationship between the reported environmental pollution, human exposure, and potential health effects among the ASGM communities (Ondayo et al., 2023a). Countrywide, limited studies report ASGM-related human exposures to PHEs in Migori county and Lake Victoria (Ngure and Kinuthia, 2020; Ngure et al., 2017; Oyoo-Okoth et al., 2010). No study has examined human exposure to major and trace elements, including PHEs, among ASGM communities in the Kakamega gold belt. Additionally, studies linking environmental contamination, HBM data, and adverse health outcomes in ASGM communities are scarce in Kenya.

Therefore, this study aimed to investigate the public health risk associated with PHEs dispersed into the environment by ASGM activities in Kakamega and Vihiga counties by the following objectives: (1). biomonitoring of ASGM communities using nail, hair and urine matrices, (2) evaluate asssociation between PHEs for human biomonitoring data versus environmental data for water, soil and sediment published in Ondayo et al., 2023 and dietary data, (3) evaluate overall hazard associated with ASGM for exposed communities.

2. Materials and methods

2.1. Approvals

Ethical approvals for the study protocol were obtained from the AMREF Ethics and Scientific Review Committee (AMREF ESRC P871/2020) on 16/12/2020 and 19/09/2022, and the British Geological Survey Research Ethics Committee (BGSREC-2021–003) on 23/02/2021 and research permits from the National Commission for Science, Technology and Innovation (NACOSTI/P/21/8536 and NACOSTI/P/22/20356) on 22/01/2021 and 22/09/2022. Additional permissions and assistance were requested from the Departments of Medical Services and Public Health in Kakamega and Vihiga counties before engaging with ASGM communities.

2.2. Study setting

This study was conducted in key ASGM villages in Kakamega and Vihiga counties in western Kenya, referred to as the Kakamega gold belt in this paper (Fig. 1). The number of control residents was limited, at 12 people, yet the data will be presented for comparison. Within the discussion, we refer to Watts et al. (2021) data for the authors' wider study of 357 volunteers for urinary elemental concentrations and Watts et al. (2019) for a survey of 450 properties including soil, water, vegetation from across Western Kenya who were not associated with ASGM activities and can provide a robust control group to supplement the discussion.

2.3. Study design, subject recruitment and sample collection

This cross-sectional, observational, community-based study was conducted in 19 ASGM villages in Kakamega and Vihiga counties in western Kenya from October 2021 to October 2022. Participants were recruited on-site by the study team randomly and through word of mouth. Active pre-determined ASGM sites and households were approached through in-person visits. All participants identified as ASGM workers (those involved in ASGM operations), residents of ASGM villages (people residing in ASGM villages but not working in ASGM) and residents of non-ASGM villages (control) were informed about the study. The study included only participants who gave their prior informed consent. For children below 18 years, prior informed consent was obtained from their parents or guardians. Pretested questionnaires with translation into Kiswahili were used to obtain biodata and evaluate exposure situations and risk factors in ASGM, potential confounders, and the general nutrition and health status of the ASGM communities and the control. Unique identifiers were used to identify each participant and match them with their samples, and high confidentiality was maintained.

Hair was cut from the nape of the participant's head using clean, sterilised stainless steel scissors and packed in a kraft bag. Fingernails were collected once using cleaned, sterilised clippers. Spot urine samples were collected in 30 mL Nalgene low-density polyethene (LDPE) bottles and stored in ice in a coolbox (~4°C). The urine samples were

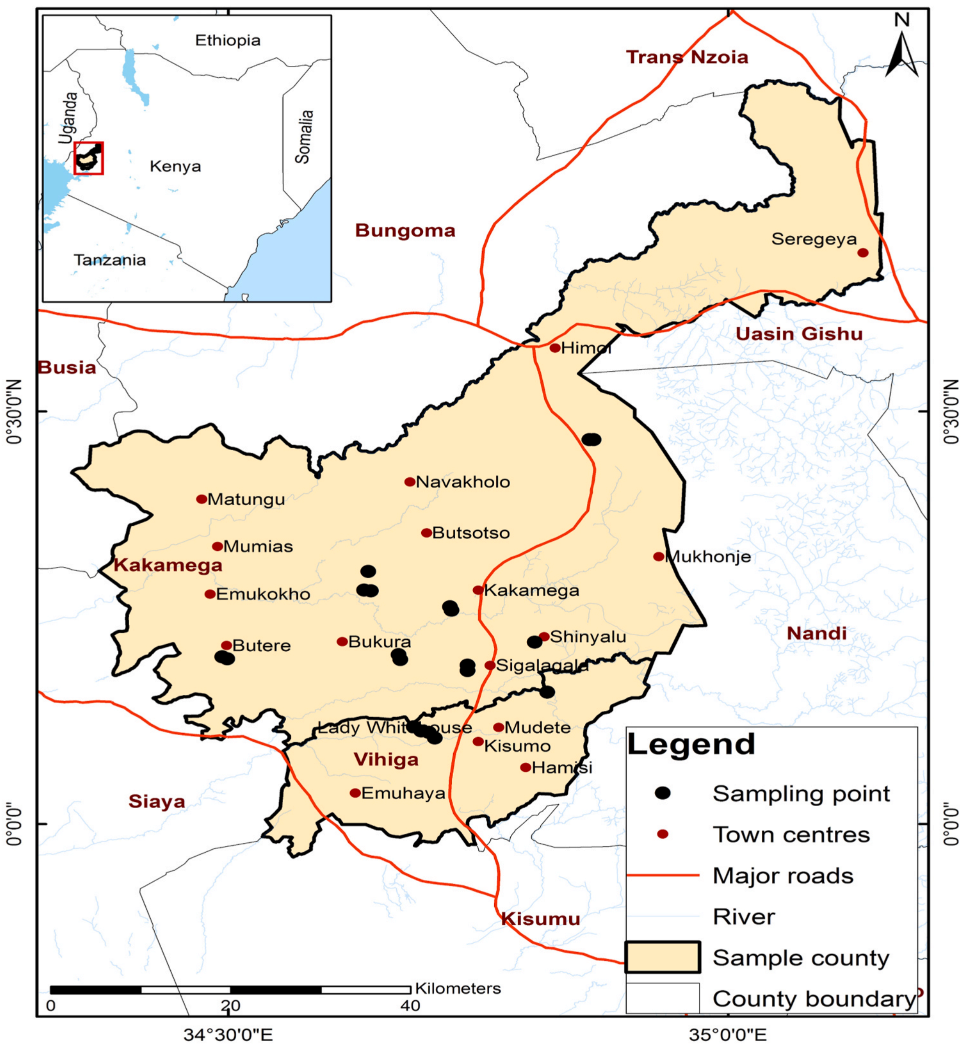

Fig. 1. Sampling points in Kakamega and Vihiga counties.

filtered into 15 mL Nalgene bottles using a nylon 0.45 µm syringe filter and frozen (~-20°C) at the end of each day. Samples of staple food crops (cereals, pulses, vegetables, tubers, and fruits) were collected from each participant's household. Collected fresh vegetable samples were thoroughly washed with distilled water to remove foreign material and dust adhering to the surfaces and later on air-dried to eliminate excess moisture. Food crop samples were freeze-dried and ground to fine powder using kitchen blenders. In order to remove oils and other external pollutants, each hair and nail sample was washed in the following sequence: de-ionised water, double rinsed with acetone, and de-ionised water, then freeze-dried (Middleton et al., 2016a). Hair and nail samples >0.2 g were freezer-milled (6850 Freezer/Mill, SPEX CertiPrep UK) with liquid nitrogen.

2.4. Sample dissolution and elemental analyses

Each urine sample was thawed and diluted ten-fold with 1 % v/v conc. HNO_3 and 0.5 % v/v HCl. Each milled hair and nail sample material (nominally 0.1 g, but in some cases 0.05 g due to low sample material) was weighed into Perfluoroalkoxy alkanes (PFA) vessels and subjected to microwave-assisted dissolution. Before digestion, sample contents in each vessel were soaked in 4 mL conc. HNO_3 and 1 mL H_2O_2 for 30 min at room temperature to enable hair and nails to submerge in the acid fully. The vessels were placed in the microwave digestion unit (MDU) (CEM^R model MARS 5). Microwave digestion was performed under the following conditions: temperature, 200 °C; pressure, 800 psi; ramp, 15 minutes; hold, 30 minutes; and microwave power, 1600 W. The digested samples were cooled down for 30 minutes. To each, 10 mL MΩ water with a resistivity of 18.2 MΩ (Millipore, UK) was added, swirled, and then transferred to a 30 mL Nalgene LDPE bottle. The PFA vessel was rinsed with 15 mL water, making the final digest volume 30 mL.

From each of the 102 food crop samples, 0.5 g was weighed into PFA vessels and 10 mL conc. HNO_3 was added, left for 30 minutes, microwaved under temperature 200 °C; pressure, 800 psi; ramp, 15 minutes, cooled, vented, and 1 mL H_2O_2 added to the contents, left for 30 minutes, then microwaved by ramping to temperature, 200∘C; pressure, and 800 psi for 15 minutes. After cooling, 30 mL MΩ water was added, swirled, and poured into a 60 mL Nalgene LDPE bottle. Each vessel's residue was rinsed with 9 mL of MΩ water, with a final volume of 60 mL.

Total major and trace element concentrations were determined using inductively coupled plasma mass spectrometry (ICP-MS) (Agilent 8900 ICP-QQQ). The detection limits were calculated as 3×SD of blanks x dilution factor and presented with the certified reference material (CRM) data to evidence the uncertainty on measurements for urine (Supplementary Table 1), hair (Supplementary Table 6), nail (Supplementary Table 11), and staple food crops (Supplementary Table 16). Mercury analysis was performed on solid materials of hair, nail, and food crop samples, using a DMA-80 Thermal Decomposition-Atomic

Absorption Spectrometer (TD-AAS) (DMA-80, Milestone Inc., Italy). A subset of hair, nail, food crop samples and CRMs were additionally analysed for Hg using ICP-MS and results of the two methods compared well (*F-ratio=0.0; p > 0.98*) (Supplementary Tables 6 and 11). In the field, collected urine samples were preserved in ice in a cooling box and frozen in the laboratory without adding conc. HCl or conc. HNO_3 acids.

2.5. Urine dilution correction

Specific gravity (SG) was measured using a PAL-10-S digital refractometer (Atago, Japan). In order to account for intra- and interindividual variations in hydration status in the study populations, correction was performed using (Equation 1); $UC_{cor}=UC_{vol}$ x [(SGm −1) / SG −1)], where UC_{cor} is dilution-corrected urinary concentration using specific gravity; UC_{vol} is the measured volume-based urinary element concentration (in μg L^{-1}); SGm is the median specific gravity value amongst the study population; and SG is the measured specific gravity of individual urine sample (Abuawad et al., 2022; Middleton et al., 2016b; Watts et al., 2021). Median specific gravity values were 1.023, 1.017, and 1.015 for ASGM workers, residents of ASGM villages, and non-ASGM villages, respectively.

2.6. Hazard associated with the Foods consumed

To evaluate the risk of health effects from the foodstuff consumed, the hazard indices for each type of the major foods (maize, beans, cowpeas, bananas, and African nightshade) were compared with and without factoring the bioaccessibility of the key study trace elements (Al, As, Cr, Hg, Ni, Pb, Sb, and Cd) in the foodstuff. The Health Index was deduced as the summation of the hazard quotients of each trace element contaminant in each of the food types i.e

$$HI = \sum HQ$$

and

$$HQ = \frac{C_p \quad x \quad ADI \quad x \quad F_{wc}}{RfD \quad x \quad BW}$$

Where where Cp is the trace element concentration in the edible portion of food (mg kg^{-1} dry weight-DW), ADI is the average daily intake (fresh weight) of food and F_{wc} is a dry-to-fresh weight conversion factor and BW is the body weight in kg. The RfD for Al, As, Cr, Hg, Ni, Pb, Sb, and Cd are 1, 0.0003, 1.5, 0.0003, 0.02, 0.0035, 0.0004, and 0.001, respectively and the Bioaccessiblity factors for As, Cr, Ni, Pb, and Cd are 0.214, 0.002, 0.065, 0.106, and 0.518, respectively (Mwesigye et al. 2016).

2.7. Statistical analysis

All the data was stored in Microsoft Excel (2016) and analysed using R and RStudio (Rstudio Team, 2022, Version 4.2.2) except for near-distances analysed using Arc Map version 10.4. The Shapiro-Wilk one-sample test tested the fitness of all the data to the normal distribution, and the null hypothesis was rejected ($p<0.05$). Thus, non-parametric tests were used for further analyses. Descriptive summaries of the laboratory data, participant characteristics, exposure risk factors, reported health effects by county administrative boundaries, and vulnerabilities such as self-ASGM activities, age, and gender were generated (Supplementary Tables 1–22). Outliers were identified and removed, and respective boxplots were generated for various exposed groups (ASGM workers, residents in ASGM villages and residents in non-ASGM villages). Associations between observed elemental exposure in biomonitors (hair, nails, urine) and their concentrations in the diet and environment (water and soil) were examined using Spearman's correlation.

The average daily intake of respective food crops (Supplementary Table 19; Table 19.1) and drinking water (1.9±1.3 L) per person were derived from the questionnaire data (n=180 participants). Respective mean daily element intakes via food crop consumption (Supplementary Tables 19.2) were calculated by the formula: daily intake of an element via food crop consumption = (element concentration in a given food crop (mg kg^{-1}) x daily average weight of food consumed by an individual (kg $person^{-1}$ day^{-1})) / the median body weight for the studied population (66.1 kg bw) (Equation 2), calculation steps fully detailed in Supplementary Table 19.3. Mean daily element intakes via drinking water (Supplementary Table 22.1) were calculated by the formula: daily intake of an element via drinking water = (element concentration in drinking water (μg L^{-1}) x daily average volume of water consumed by an individual (L $person^{-1}$ day^{-1})) / the median body weight for the studied population (66.1 kg bw) (Equation 3), calculation steps fully shown in Supplementary Table 22.2.

3. Results and discussion

3.1. Study participants' characteristics and demographics

A total of 180 participants were recruited and grouped as ASGM workers (n=144), non-ASGM workers residing in ASGM villages (n=25), and non-ASGM workers residing in non-ASGM villages (control, n=11) (Table 1). The majority of the respondents (ASGM workers, n=121/144, 84 %; residents of ASGM villages, n=14/25, 56 %; and residents of non-ASGM, n=9, 82 %;) were in their reproductive ages (16–49 years) with a general male predominance (n=83; 58 %). Most participants attained primary (n=79; 45 %) and secondary (n=78; 44 %) education, while few attained tertiary level (n=17; 10 %) or had no formal education (n=3; 2 %).

3.2. Human biomonitoring

3.2.1. Urinary elemental concentrations

3.2.1.1. Influence of urinary hydration correction. Urinary biomarkers for chemical exposures are highly susceptible to measurement errors. This is because measured urinary PHE concentrations depend on sample dilution, primarily influenced by urine flow rate and hydration status, which vary from individual to individual (Abuawad et al., 2022). Specific gravity correlates with urinary creatinine concentration (Abuawad et al., 2022). However, a strong agreement between correction methods was reported using specific gravity, compared to creatinine and osmolality (Abuawad et al., 2022; Watts et al., 2021). This is because specific gravity has a lower resolution than creatinine correction and is less influenced by demographic factors such as age, sex, and body mass index (BMI), and is thus employed more frequently (Abuawad et al., 2022). However, it is normalised on the median specific gravity of the study population rather than a constant value, which may hinder the ability to compare concentrations across populations across different literature sources (Abuawad et al., 2022; Watts et al., 2021). In the present study, urine specific gravity in 42 % (n=49) of the participants was within the normal range (1.001–1.02), while 57 % (n=67) of the participants had over-range specific gravities (>1.021≤1.33) that indicated dehydration (Abuawad et al., 2022). As presented in Fig. 2, population-level differences for this study were found between uncorrected and specific gravity-corrected urine PHE data indicating influence of hydration correction. The changes included up to −16 % Al and −15 % Cd reduction, and up to +26 % As, +210 % Cr, +35 % Hg, +14 % Ni, +229 % Pb, +174 % Sb, and +50 % Cs increase following correction, as similarly reported in previous studies (Middleton et al., 2019; Watts et al., 2021). The full urine dataset, including the hydration correction (specific gravity), is provided in Supplementary Table 1. Descriptive statistics for each administrative county area (Supplementary Table 2), ASGM worker versus non-worker (Supplementary Table 3), gender

Table 1
Profiles of study participants in ASGM and non-ASGM villages.

Sampling area		ASGM villages (ASGM workers and residents, n=169)				Non-ASGM villages (n=11)	
Participants		ASGM workers (n=144)		Non-ASGM workers (residents) (n=25)		Residents (n=11)	
Variable		Total	%	Total	%	Total	%
Female/male		57/87	40/60	13/12	52/48	5/6	45/55
Age (mean, median, range) (years)		35.5, 33.3, 16–73.6		35, 40, 2.5–77		38, 33.6, (19–78.6)	
Age group (years)	0–7	nil		3	12	nil	nil
	7–17	4	3	4	16	nil	nil
	18–49	116	81	11	44	9	82
	≥50	19	13	7	28	2	18
Education	No school	2	1	1	4	nil	nil
	Primary	61	42	9	36	6	55
	Secondary	63	44	11	44	4	36
	Tertiary	12	8	4	16	1	9

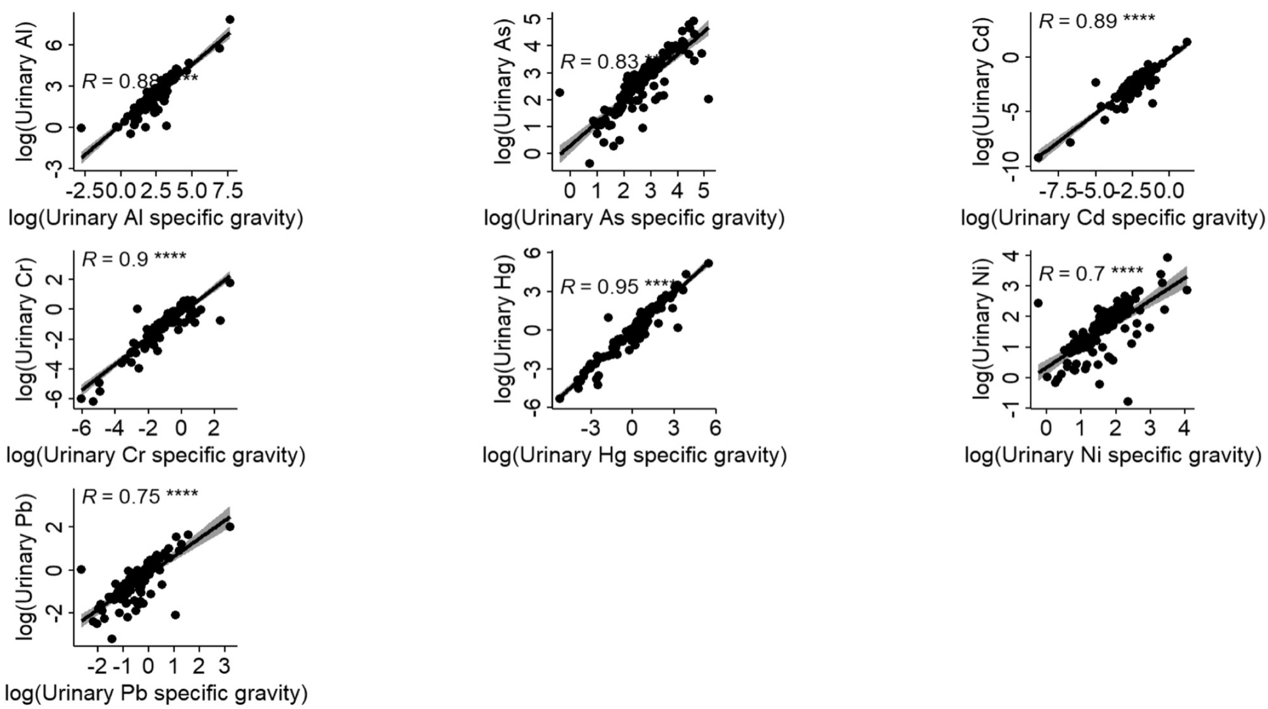

Fig. 2. Influence of urinary hydration correction, Urinary Al, As, Cd, Cr, Hg, Ni and Pb concentrations were measured in μg L^{-1}.

(Supplementary Table 4), and age category (Supplementary Table 5) are provided both with and without hydration correction (specific gravity) to allow for comparisons with existing literature in the discussion. There are no existing countrywide bioequivalents and standards for urine PHEs in Kenya. Therefore, this study's urinary findings were interpreted in a health-based context by comparing them to urine reference values found among the general population in the USA (CDC, 2015, 2021) and Canada (Saravanabhavan et al., 2017) and globally derived health-based bioequivalents (Hays et al., 2010; Hays et al., 2008; Poddalgoda et al., 2021; Poddalgoda et al., 2017), and reference values developed in western Kenya and Kinshasa, Congo (Tuakuila et al., 2015; Watts et al., 2021).

3.2.1.2. Potentially harmful elements (PHEs) with published urinary thresholds based on Bioequivalence (Al, As, Ba, Cd). Urinary total As is the most reliable indicator of recent As exposure since As is excreted predominantly by glomerular filtration (Balali-Mood et al., 2021; Rana et al., 2018). For 79 % of the ASGM workers (n=75 out of 95) and 77% of the residents of ASGM villages (n=13 out of 17), urinary As were above the biomonitoring equivalent (BE) upper threshold representing toxicity for As (6.4 μg L^{-1}) (Hays et al., 2010), with a range of 6.8 – 138 μg L^{-1}. By contrast, all the urinary As concentrations (range, 0.7 – 4.2 μg L^{-1}) were below 6.4 μg L^{-1} - excess BE value for As in urine (Hays et al., 2010) for residents of non-ASGM villages. The 95th percentiles (for ASGM workers, As 55.2 μg L^{-1}; for residents in ASGM, As 100 μg L^{-1}) and median (for ASGM workers, As, 12.2 μg L^{-1}; for residents in ASGM, As 13.9 μg L^{-1}) urinary As in ASGM villages were significantly higher than non-ASGM villages As (*p=0.02*) as illustrated in Fig. 3, and those reported in a bigger baseline group of 357 volunteers for Western Kenya by Watts et al. (2021) (median As, 3.9 μg L^{-1}; **$RV_{95}S$** As, 22.1 μg L^{-1}). Total urinary As in 21 % of samples from 20 ASGM workers and 3 ASGM residents exceeded the ACGIH occupational biological exposure index (BEI) threshold of 35 μg L^{-1} (ACGIH, 2009). The self-reported fish consumption did not affect urinary As. The Kakamega gold belt ASGM communities have no access to the open sea and seafood, and 95 % of them reported that they commonly consume freshwater fish from Lake Victoria (Supplementary Table 19.1), which contain relatively low amounts of As (Marriott et al., 2023). The elevated urinary As values may be explained by diet, drinking water patterns, or a generally elevated As in air, soil and dust due to mining activities. This study's As results compare well with findings in active ASGM areas in Tanzania (As median 9.4 μg L^{-1}, p75 15.1 μg L^{-1}),

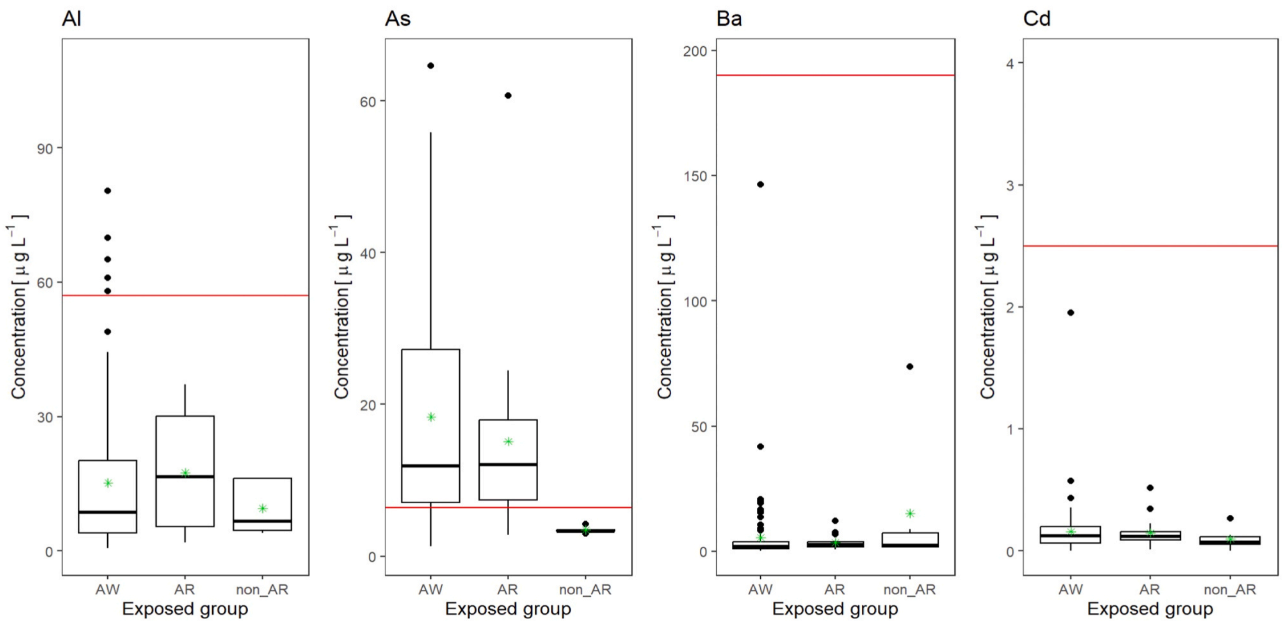

Fig. 3. Boxplots of urinary PHEs with published thresholds, above green asterisk, shows the mean concentration, the black line denotes the median, the red line indicates excess biomonitoring equivalent (BE) from the most conservative values for Al (57–137 μg L^{-1}), As (6.4 μg L^{-1}), Ba (190 μg L^{-1}), and Cd (2.5–6.38 μg L^{-1}). AW=ASGM workers, AR=residents of ASGM villages and non_AR=residents of non-ASGM villages.

Zimbabwe (As median 9.7 μg L^{-1}; p75 11.1 μg L^{-1}), and legacy mining areas in Mexico (As median 16.5 μg L^{-1}, p75 19.4 μg L^{-1}) (Moreno et al., 2010; Nyanza et al., 2019; Rakete et al., 2022), but are ten times less than those reported in Ghanaian ASGM (As median 100 μg L^{-1}; p75 135 μg L^{-1}) (Basu et al., 2011).

Cadmium concentrations in the urine samples (n=117) were below the lowest threshold for excess BE (2.25–6.38 μg L^{-1}) (Hays et al., 2008) except for a single sample drawn from a cyanidation plant worker/amalgam burner that had 4 μg L^{-1}. This indicates a generally low Cd exposure (Fig. 3), with median urinary Cd concentrations for ASGM workers at 0.45 μg L^{-1} compared to a baseline of 0.20 μg L^{-1} for western Kenya reported by Watts et al. (2021). Levels of Cd are generally elevated in smokers (Balali-Mood et al., 2021). However, there was no difference between urinary Cd in smokers and non-smokers (p=0.27) in this study. Similarly, low urinary Cd levels were found in ASGM communities in Ghana (Basu et al., 2011), Zimbabwe (Rakete et al., 2022), and other areas across Africa (Ondayo et al., 2023b), an indication that Cd exposure in ASGM was generally below BE thresholds.

As shown in Fig. 3, all urinary Ba concentrations indicated low exposure for ASGM workers and residents with a median of 1.8 and 2.8. μg L^{-1}, respectively. These values indicate low exposure well below the BE upper threshold for Ba (190 μg L^{-1}) (Poddalgoda et al., 2017), but comparable to a baseline study for Western Kenya at 1.7 μg L^{-1} (Watts et al., 2021).

3.2.1.3. Potentially harmful elements (PHEs) in urine without threshold values based on Bioequivalence (Cr, Cs, Hg, Li, Ni, Pb, Rb, Sr, Sb, Ti, Tl, V). Urinary Cr biomonitoring is reliable in determining exposure since Cr is principally excreted in the urine (Balali-Mood et al., 2021). This study's urinary Cr in ASGM workers (median 0.4 μg L^{-1}) and residents (median 0.4 μg L^{-1}) of ASGM villages was up to 22 times greater than the value for urinary Cr (0.22 μg L^{-1}) considered for healthy persons (ATSDR, 2012) for 76 % of volunteers, with a range of 0.03–5.8 μg L^{-1} (Fig. 3). Chromium occurs in valence states (−2 to + 6), with hexavalent (Cr^{6+}) and trivalent (Cr^{3+}) forms being the most prevalent forms. Upon exposure to biological tissues, Cr^{6+}, a group 1 carcinogen, is usually converted to Cr^{3+}, the only Cr species found in biological specimens and is essential in trace amounts as a cofactor for insulin action and natural protein and lipid metabolism (Balali-Mood et al., 2021; IARC, 2021; Mitra et al., 2022). This study's urinary Cr results were far lower than the 25 μg L^{-1} reported for comparable ASGM activities in the Talensi-Nabdam ASGM area in Ghana (Basu et al., 2011).

The highest urinary Hg concentrations were recorded among ASGM workers, followed by residents of ASGM and residents of non-ASGM villages (p=0.02), which is clearly illustrated in Fig. 3. No significant difference (p=0.11) between females (n=43) and males (n=75) was observed. Recorded median urinary Hg concentration for ASGM workers (1.7 μg L^{-1}) was 12 times higher than the National Health and Nutrition Examination Survey (NHANES) 2015–2016 median total Hg, 0.14 μg L^{-1} for the adult US population (ATSDR, 2022). Urinary Hg levels predominantly reflect acute or chronic elemental or inorganic Hg exposure, which primarily occurs through inhalation (with a 75–85 % absorption rate) and, to some extent, ingestion (with a 10–30 % absorption rate) and dermal (with almost 0 % absorption rate) routes (Balali-Mood et al., 2021; Mitra et al., 2022; WHO, 2021b). In the present study, recent individual involvement in amalgamation, amalgam-burning activities, direct Hg contact, and Hg spillages strongly influenced total urinary Hg concentrations ($p<0.001$). Urinary Hg ranged between 11.1 and 178 μg L^{-1} among active ASGM workers (12%, n=11) living within a 100 m radius from ASGM activities and had direct contact with Hg. Besides, the 11 ASGM workers reported having chest pains, headaches, stroke, epilepsy and fertility problems, which necessitate further inquiry into their causes. Urinary Hg among ASGM workers involved in amalgam burning (n=79; 0.21–178 μg L^{-1}; median Hg 6.8 μg L^{-1}) significantly exceeded Hg recorded among ASGM workers that did not conduct amalgam burning (n=39; 0.05–22 μg L^{-1}; median Hg 1.8 μg L^{-1}). These urinary Hg results are higher than the 0.29–76 μg L^{-1} reported for southwestern Ghana (Abrefah et al., 2011) but below urinary Hg levels found in ASGM workers in Burkina Faso (3–3493 μg L^{-1}; median Hg 123 μg L^{-1}), and, Tanzania and Zimbabwe (up to 440 μg L^{-1}; median 8.5 μg L^{-1}) (Bose-O'Reilly et al., 2020; Tomicic et al., 2011).

Urinary Pb excretion mainly reflects recent exposure and is limited in assessing long-term Pb exposure or body burden (Balali-Mood et al., 2021; Mitra et al., 2022). Lead concentration in urine reportedly increases exponentially with increased blood Pb levels (Mitra et al., 2022). In the present study, most (83 %) of the ASGM workers and residents had urinary Pb above the NHANES 2013–2014 median urinary Pb concentration (0.277 μg L^{-1}) reported for the general US population based on 2664 individuals of all ages (CDC, 2021). The Pb exposure could be attributed to airborne dust due to ore mining and milling across the communities and contaminated drinking water and food - although the uptake of Pb in the gastrointestinal tract of adults is considered relatively low. This study's Pb results illustrated in Fig. 4 show a similar range of urinary Pb for ASGM workers (median 0.7 μg L^{-1}) and residents (median 0.7 μg L^{-1}), but elevated from the control group (Fig. 4) and wider baseline median for western Kenya of 0.5 μg L^{-1} reported by Watts et al. (2021). These figures are comparable with previous findings for ASGM in Ghana at 1.1 μg L^{-1} (range 0.18–2.8 μg L^{-1}) (Basu et al., 2011). Similar to urinary Pb, median urinary Cs for ASGM workers (Cs, 5.7 μg L^{-1}) and residents of ASGM villages (Cs, 5.2 μg L^{-1}) exceeded the Western Kenya baseline urinary Cs concentration of 2.1 μg L^{-1} reported by Watts et al. (2021).

Urinary Ni concentrations in 97% of ASGM workers and residents in ASGM villages and 86 % of residents in non-ASGM villages significantly exceeded the upper limit of the ATSDR's reference urinary Ni values for healthy adults (1–3 μg L^{-1}) (ATSDR, 2005) (Fig. 4). Median urinary Ni concentrations for ASGM workers (5.2 μg L^{-1}) and ASGM residents (4 μg L^{-1}) was above the baseline median for Western Kenya 3.5 μg L^{-1} reported by Watts et al. (2021). The urinary Ni followed a decreasing order of ASGM workers>residents of ASGM>residents of non-ASGM villages and were similar to urinary Ni concentrations reported in ASGM in Ghana (median 5.4 μg L-1; 1.4–25.7 μg L^{-1};) (Basu et al., 2011). The median urinary Sb concentration was higher than the Western Kenya baseline of 0.08 μg L^{-1} (Watts et al., 2021), for ASGM workers (0.14 μg L^{-1}), ASGM residents (0.18 μg L^{-1}), whilst non-ASGM residents in the small control grop were higher than all three categories (0.28 μg L^{-1}) for whom the pathway of exposure may require further study.

The urinary results revealed that a high percentage of the ASGM residents and workers had elevated levels of PHEs, especially As, Cr, Hg, Ni, Pb and Sb, above health-based bioequivalence, toxicologically derived exposure limits and background concentrations as can be seen in Fig. 4. Even though urine is relatively easy to collect, store and prepare, urine monitoring for PHEs may only show recently accumulated PHE levels within 24–48 hours (Alves et al., 2014). This factor should be considered when interpreting the urinary results and associated health risks. Overall, biomonitoring studies regarding the ASGM communities' health are urgently needed to provide tools for better risk assessment and implementing actions to reduce PHE exposure.

3.2.2. Hair and nail elemental concentrations

Results of selected PHE concentrations in human hair drawn from 68 study participants are summarised in Table 2. The complete human hair laboratory dataset is detailed in Supplementary Table 6. Statistical descriptions of the hair data by county administrative areas (Supplementary Table 7), self-ASGM activities and villages (Supplementary Table 8), gender (Supplementary Table 9), and age category (Supplementary Table 10) are all provided.

Hair As in 50 % of ASGM workers (median, 4.92 mg kg^{-1}; 1–786 mg kg^{-1}) and 36 % of residents of ASGM villages (median, 7.2 mg kg^{-1}; 1–73 mg kg^{-1}) were higher than the WHO threshold representing potential As toxicity (1 mg kg^{-1}) (FAO/WHO, 2011; WHO, 1996). Concentrations of As in male hair were often much higher (range, 0.032–786 mg kg^{-1}; mean, 33.8 mg kg^{-1}; median 2.2 mg kg^{-1}) than female ASGM workers (range, 0.012–10.4 mg kg^{-1}; mean, 3.1 mg kg^{-1}; median 1.6 mg kg^{-1}) (Supplementary Table 9). Thus, male ASGM workers were more exposed to As than their female colleagues. This can be explained by the fact that males are more involved in rudimentary ore extraction, crushing and milling compared to females since As was mainly associated with the soils, ores and mine tailings in a previous study in the ASGM villages (Ondayo et al., 2023a). The As results in this study were higher than those in Migori-Kenya ASGM areas, in which hair As ranged from 0.01 to 0.015 mg kg^{-1} with a mean of 0.01 mg kg^{-1}

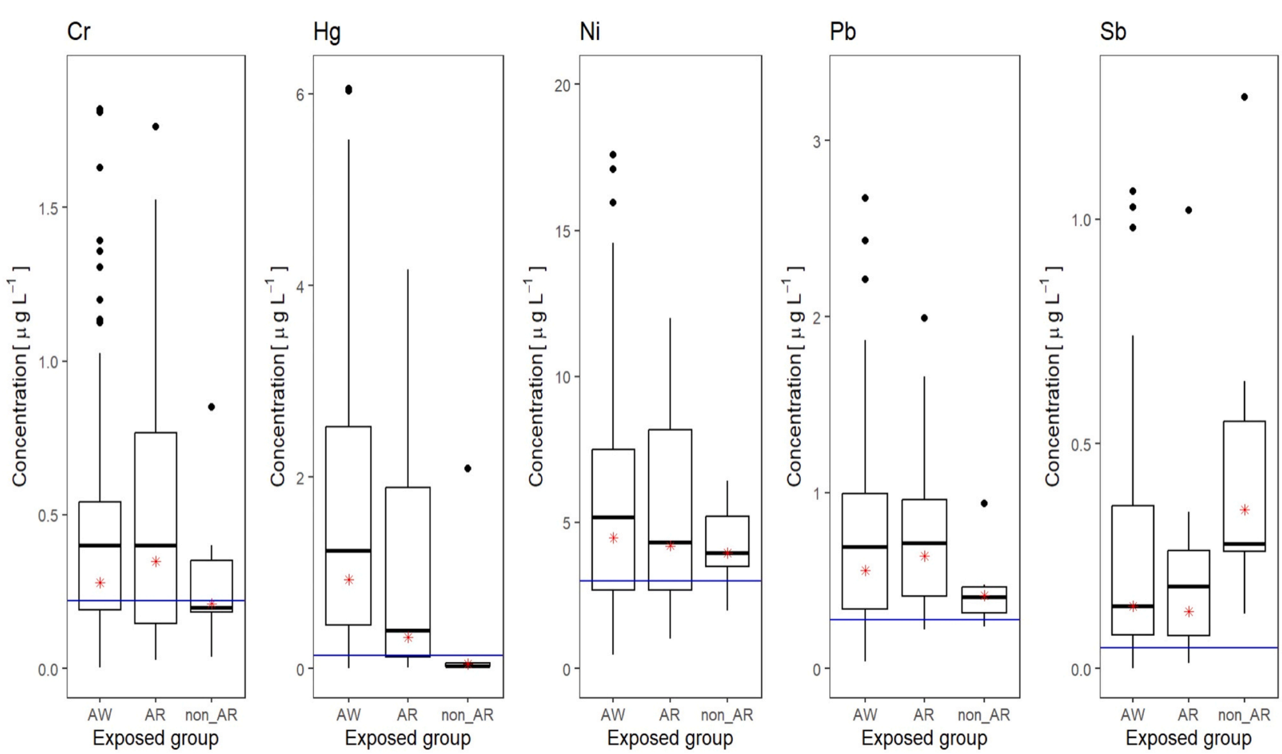

Fig. 4. Boxplots of urinary PHEs without published thresholds for Bioequivalence, above red asterisk shows the geometric mean concentration, the black line denotes the median, the blue line indicates standard urinary concentration based on the most conservative values for urinary Cr (0.22 μg L^{-1}), Hg (0.14 μg L^{-1}), Ni (1–3 μg L^{-1}), Pb (0.277 μg L^{-1}) and Sb (0.046 μg L^{-1}) concentrations for the general healthy US population. AW=ASGM workers, AR=residents of ASGM villages and non_AR=residents of non-ASGM villages.

Table 2
Concentrations of selected potentially harmful elements in human hair and nails in ASGM villages.

Element	Biomonitor	Exposed group		maximum	minimum	p25	p50	p95	Published/ Reference ranges
		units	*n samples*	$mg\ kg^{-1}$		$mg\ kg^{-1}$	$mg\ kg^{-1}$	$mg\ kg^{-1}$	$mg\ kg^{-1}$=$\mu g\ g^{-1}$
Al	Hair	ASGM workers	56	5285	119	367	597	2430	
	Hair	ASGM residents	11	1024	111	351	489	1021	
	Hair	non-ASGM residents	1	475	475	475	475	475	
Al	Nail	ASGM workers	26	4054	468	945	1455	2722	
	Nail	ASGM residents	2	1528	333	632	931	1468	
	Nail	non-ASGM residents	3	1245	824	881	938	1215	
As	Hair	ASGM workers	56	786	0.03	0.3	1.6	29.3	1[a]
	Hair	ASGM residents	11	73.4	0.1	0.2	1	48.7	1[aa]
	Hair	non-ASGM residents	1	0.15	0.15	0.15	0.15	0.15	1[a]
As	Nail	ASGM workers	26	129	0.07	0.73	3.7	73	0.07–1.09b
	Nail	ASGM residents	2	1.43	0.12	0.45	0.78	1.37	0.07–1.09b
	Nail	non-ASGM residents	3	0.36	0.05	0.12	0.19	0.34	0.07–1.09b
Cd	Hair	ASGM workers	56	0.58	0.01	0.03	0.04	0.15	0.25–1b
	Hair	ASGM residents	11	0.66	0.004	0.03	0.06	0.46	0.25–1b
	Hair	non-ASGM residents	1	0.12	0.12	0.12	0.12	0.12	0.25–1b
Cd	Nail	ASGM workers	26	0.11	0.007	0.02	0.03	0.09	0.01–0.44b
	Nail	ASGM residents	2	0.01	0.01	0.01	0.01	0.01	0.01–0.44b
	Nail	non-ASGM residents	3	0.031	0.014	0.015	0.015	0.03	0.01–0.44b
Hg	Hair	ASGM workers	56	12.8	0.1	0.2	0.37	7.4	1c
	Hair	ASGM residents	11	1.4	0.09	0.12	0.2	1	1c
	Hair	non-ASGM residents	1	0.14	0.14	0.14	0.14	0.14	1c
Hg	Nail	ASGM workers	26	3.3	0.04	0.14	0.27	1.2	0.03–0.31b
	Nail	ASGM residents	2	0.07	0.06	0.06	0.07	0.07	0.03–0.31b
Ni	Hair	ASGM workers	56	27.7	0.4	1.4	1.8	9.9	0.02–0.2a
	Hair	ASGM residents	11	35.9	0.31	0.8	3.1	32	0.02–0.2a
	Hair	non-ASGM residents	1	0.77	0.77	0.77	0.77	0.77	0.02–0.2a
Ni	Nail	ASGM workers	26	646	2	3.5	6.3	102	0.14–6.95b
	Nail	ASGM residents	2	4.4	3.4	3.6	3.9	4.3	0.14–6.95b
	Nail	non-ASGM residents	3	3.8	3	3	3	3.7	0.14–6.95b
Pb	Hair	ASGM workers	56	61	0.4	1.3	2.5	11.5	0.2a
	Hair	ASGM residents	11	11.1	0.2	2.5	2.9	10.9	0.2a
	Hair	non-ASGM residents	1	3	3	3	3	3	0.2a
Pb	Nail	ASGM workers	26	14.2	0.23	1.1	1.7	5.9	0.27–4.75b
	Nail	ASGM residents	2	1.3	0.84	1	1.1	1.3	0.27–4.75b
	Nail	non-ASGM residents	3	0.38	0.22	0.25	0.27	0.37	0.27–4.75b

[a] Published/reference ranges obtained from World Health Organization (WHO) (FAO/WHO, 2011; WHO, 1996).
[b] Published/reference ranges obtained from Coelho et al. (2013).
[c] Reference limits obtained from the United States Environmental Protection Agency (USEPA) (WHO, 2008).

(Ngure and Okioma, 2020). Median hair Cd concentrations were 0.04 $mg\ kg^{-1}$ for ASGM workers was well below the maximum permissible range of 0.25–1 $mg\ kg^{-1}$ (Coelho et al., 2013).

For all subjects, hair Ni concentrations exceeded the upper threshold range for Ni in human hair 0.02–0.2 $mg\ kg^{-1}$ (WHO, 1996), with median of 1.9 and 2.8 $mg\ kg^{-1}$, respectively for ASGM workers and ASGM residents significantly higher than the reported threshold. Additionally, hair Pb concentrations in all the ASGM workers (median 2.6 $mg\ kg^{-1}$) and residents of ASGM villages (median 2.7 $mg\ kg^{-1}$) were above the 0.2 $mg\ kg^{-1}$ threshold (WHO, 1996), showing that communities were equally exposed to environmental Pb as the miners and ore processors. The hair Pb results (0.22–60.7 $mg\ kg^{-1}$) in this study far exceed those reported in the Migori gold belt (Pb 0.61–0.86 $mg\ kg^{-1}$) (Ngure et al., 2017). During formation, hair sequesters MeHg (methyl mercury) and is reliable for MeHg measurement (WHO, 2008). Hair Hg directly relates to blood Hg concentrations, yet hair provides information on long-term average exposure (WHO, 2008). This study found elevated hair Hg concentrations that exceeded the USEPA 1 $mg\ kg^{-1}$ threshold (WHO, 2008) in 21 % of ASGM workers that mainly participated in ore washing, sluicing, amalgamation and amalgam burning (range, 1.1–13 $mg\ kg^{-1}$; median 2.6 $mg\ kg^{-1}$). This study's hair Hg concentrations exceeded those found in the Migori gold belt in Kenya (0.12–0.32 $mg\ kg^{-1}$) (Ngure et al., 2017), but were, however, lower than those reported (up to 27.8 $mg\ kg^{-1}$, median 1.5 $mg\ kg^{-1}$) among nursing mothers that conducted panning and amalgam smelting in ASGM areas in Kadoma, Zimbabwe (Bose-O'Reilly et al., 2020). Females were often involved in the amalgamation and burning of Hg, which would be of concern for future generations as Hg in maternal scalp hair is a proxy of fetal Hg exposure (McDowell et al., 2004).

Nails collected from ASGM workers contained similarly significant Al, As, Hg, Ni, and Pb concentrations. Human nail (n=31) analysis results for selected PHEs are summarised in Table 2, whilst the full toenail laboratory dataset is presented in Supplementary Table 11. Descriptive statistics summaries by county administrative areas (Supplementary Table 12), self-ASGM activities and villages (Supplementary Table 13), gender (Supplementary Table 14), and age category (Supplementary Table 15) are provided. Concentrations of As in 65 % (n=17 out of 26) of nails drawn from ASGM workers (1.4–129 $mg\ kg^{-1}$; median, 8.5 $mg\ kg^{-1}$; mean 22.9 $mg\ kg^{-1}$) exceeded the global reference range (0.07–1.09 $mg\ kg^{-1}$) by up to 118 times (Coelho et al., 2013). The ASGM workers with over-reference nail As were spread across seven villages (Rosterman, Viyalo, Lunyerere, Indete, Malinya–Shikoye, Malinya–Shitoli, and Bushiangala). Moreover, 77 % of them undertook gold ore excavation; ore crushing; ore washing, sluicing and amalgamation; and amalgam burning. This study's nail As concentrations for ASGM workers (mean As) are much higher than nail As concentrations reported in the Migori gold belt, Kenya (mean As, 0.002–0.04 $mg\ kg^{-1}$) (Ngure et al., 2017). All nail Cd values were lower than the maximum permissible range of 0.01–0.44 $mg\ kg^{-1}$ (Coelho et al., 2013) and previous findings in the Migori gold belt, Kenya 0.08–0.82 $mg\ kg^{-1}$ (Ngure et al., 2017).

Mercury in human toenails reflects long-term exposure via food consumption, especially fish (Salcedo-Bellido et al., 2021). Toenails from 46 % of the ASGM workers had Hg concentrations

(0.4–3.3 mg kg^{-1}; median, 0.87 mg kg^{-1}; mean 0.98 mg kg^{-1}) that exceeded a published reference range of 0.03–0.31 mg kg^{-1} up to 11 times (Coelho et al., 2013). These ASGM workers were involved in multiple ASGM processes across seven villages. These data were however, lower than Hg concentrations measured in nails from smelter miners (0.4–12.7 mg kg^{-1}; median 3.3 mg kg^{-1}) and non-smelter miners (0.1–7.4 mg kg^{-1}; median 2.1 mg kg^{-1}) from Amansie West in Ghana (Kwaansa-Ansah et al., 2019) but significantly higher than reported from the Migori gold belt in Kenya (0.24–0.5 mg kg^{-1}) (Ngure et al., 2017). Forty-two per cent of the ASGM workers in this study had nail Ni concentrations (7.4–646 mg kg^{-1}; median, 14.4 mg kg^{-1}; mean, 88.4 mg kg^{-1}) above the published reference range of 0.14–6.95 mg kg^{-1} (Coelho et al., 2013). Nail Pb concentrations for 15 % of ASGM workers exceeded the published reference of 0.27–4.75 mg kg^{-1}, with Pb concentrations of 5.3–14.2 mg kg^{-1} and a median of 5.9 mg kg^{-1} (Coelho et al., 2013). Nail Cd, Ni and Pb have been reported to reflect between 7 and 12 months' exposure (Salcedo-Bellido et al., 2021). This study's nail As (0.1–129 mg kg^{-1}; median 5 mg kg^{-1}), Cd (0.01–0.11 mg kg^{-1}; median 0.03 mg kg^{-1}), Ni (2–646 mg kg^{-1}; median 6.9 mg kg^{-1}), and Pb (0.2–14.4 mg kg^{-1}; median 1.6 mg kg^{-1}), concentrations were generally higher than the findings of Mwesigye et al. (2016) in the Kilembe Copper mine, Uganda, where nails from children and adults had similarly high As (0.05–5.2 mg kg^{-1}), Cd (0.01–0.07 mg kg^{-1}), Ni (0.9–40 mg kg^{-1}) and Pb (0.25–8.8 mg kg^{-1}). Further work in the field of HBM is required because reliable reference ranges based on bioequivalence for hair and nail PHE are lacking due to the absence of baseline data.

Hair and nail matrices, unlike urine and blood, give indication of long-term exposures to the PHEs for over weeks, rather than hours and days (Hamouda and Felemban, 2023). They are non-invasive and easy to collect, transport, and store (Alves et al., 2014). The hair and nail findings (Table 2) provide significant evidence indicating that the majority of the ASGM workers and residents of ASGM villages in the Kakamega gold belt were exposed to PHE levels above reference values. However, the levels of PHEs in these external tissues could also arise from environmental contamination by dust and dirt (Button et al., 2009; Middleton et al., 2016a).

3.3. Elemental concentrations in drinking water and participants' intake

Analytical results for selected PHEs in drinking water are summarised in Table 3, while the whole dataset is provided in Supplementary Table 20, with further categorisation by use in Supplementary Table 21. The concentrations of Al in 9 % of the community drinking water sources, which ranged from 96 to 13,909 μg L^{-1} were up to 9 times greater than the WHO-health-based guideline for Al of 90 μg L^{-1} in drinking water (WHO, 2017). Aluminium concentrations in this study were higher than similarly elevated Al values reported in drinking water ASGM-impacted areas in Adola, Ethiopia were 0.8–139 μg L^{-1} (Getaneh and Alemayehu, 2006) and Lake Victoria, Kenya 2–3324 μg L^{-1} (Marriott et al., 2023).

Total As concentrations in 18 % of the community drinking water sources (13–366 μg L^{-1}) were up to 36 times greater than the WHO health-based guideline for As in drinking water at 10 μg L^{-1} similar to the Adola mines of Ethiopia (As 0.6–337 μg L^{-1}; Getaneh and Alemayehu, 2006), but exceeding those reported in ASGM-impacted Kenyan Gucha, Migori and Nzoia (0.06–23 μg L^{-1}; Ngure et al., 2014).

Table 3
Concentrations of selected PHEs in drinking water in ASGM villages.

Element	maximum	minimum	p50	p95	WHO guideline
	μg L^{-1}	μg L^{-1}	μg L^{-1}	μg L^{-1}	μg L^{-1}
Al	***13909****	1	10.7	***106****	90
As	***366****	0.06	1.3	***37.7****	10
Cd	0.46	0.003	0.03	0.09	3
Cr	***103****	0.08	0.5	2.7	50
Ni	64.4	0.32	3	17.2	70
Pb	***100****	0.02	0.2	0.84	10
Sb	9.5	0.002	0.1	0.34	20

Values in bold italics font with (*) exceed World Health Organization (WHO) guidelines for elements in drinking water (WHO, 2022).

Similarly, concentrations of Cr and Pb for this study in spring water (Pb 12.4 μg L^{-1}) and mine shaft water (Cr 103 μg L^{-1}, Pb 100 μg L^{-1}) were above WHO guidelines at 50 μg L^{-1} for Cr and 10 μg L^{-1} for Pb in drinking water. In contrast, throughout the ASGM villages, Cd, Ni, and Sb in the waters were within respective WHO permissible limits (WHO, 2022) shown in Table 3. Previous environmental investigations attributed the elevated PHE (As, Cr, Cd, Ni, and Pb, among others) concentrations in waters and soils in the studied ASGM villages to natural geological enrichment, with the unregulated ASGM activities as the primary modes of dispersion across various environmental matrices (Ondayo et al., 2023a).

The average daily water intake per person in the studied ASGM villages was 1.9 ± 1.3 L, with a median body weight of 66.1 kg for adults, compared to WHO (2022) 2 L for 60 kg adults. The mean daily elemental intakes via drinking water are presented in Supplementary Table 22.1, with their full calculation details in Supplementary Table 22.2. The questionnaire data showed that rivers and streams (58 %, n=104); groundwater from boreholes, shallow wells, mine shafts, and ponds (25 %, n=43); springs (46 %, n=80), rainwater (36 %, n=63) and piped and bottled water (16 %, n=28) were the primary drinking water sources in the studied ASGM villages. Overlaps in drinking water sources per participant were found for rivers and streams and springs (n=16), rivers and streams, and groundwater (n=21), and groundwater and springs (n=4). Seemingly, rain, bottled, and piped water only supplemented drinking water from rivers and streams, groundwater, and springs since only 6% (n=10) of the participants relied entirely on piped and bottled water for drinking. The piped water is sourced from boreholes operated by the County government. Mean daily intakes of As and Al via consumption of spring water (As 0.003 mg kg^{-1} bw day^{-1} and Al 0.4 mg kg^{-1} bw day^{-1}) and mine shafts (As 0.005–0.012 mg kg^{-1} bw day^{-1}) exceeded the FAO/WHO tolerable daily intake (TDI) values (As 0.0021 mg kg^{-1} bw day^{-1}; Al 0.1 mg kg^{-1} bw day^{-1}) (WHO, 2022). Besides, the participants' average daily intake of Cr via drinking spring (0.00014–0.0005 mg kg^{-1} bw day^{-1}) and mine shaft water (0.003 mg kg^{-1} bw day^{-1}) in Shikoye village in Malinya was above the FAO/WHO permitted TDI for Cr (0.0001 mg kg^{-1} bw day^{-1}) (WHO, 2022). In contrast, calculated mean daily Cd, Ni and Pb intakes via drinking water were within respective FAO/WHO tolerable daily intake (TDI) guidelines for all the participants (FAO/WHO, 2011) (Supplementary Table 22.1). The study results reveal that drinking water contributes to As, Al and Cr exposure among the ASGM communities. Despite the recorded low daily Cd, Ni and Pb intakes via drinking water, investigating chronic exposure of the ASGM workers and residents to these and other PHEs is crucial, considering their respective recorded concentrations in primary community drinking water sources (Table 3; Supplementary Tables 20–21.1).

Concentrations of As in 32 % (23.2–846 μg L^{-1}) of the ore washing and Hg-Au amalgamation-tailing pond waters were up to 43 times higher than the WHO threshold (20 μg L^{-1}; Supplementary Table 21.2). The PHE's are dispersed by the practice of irrigation using the mining effluent and disposal of mine wastewater into the rivers and streams in Kakamega gold belt. Similarly, in ASGM sites along the River Nile and the Red Sea coastline, wastewaters released from amalgamation-tailing ponds were found to raise total Hg concentrations in irrigation waters to 0.05–0.1 μg L^{-1} and drainage canals to greater than 0.2 μg L^{-1}, and aquifers to 0.08–0.5 μg L^{-1} (Abdelaal et al., 2023).

3.4. Dietary elemental concentrations and their hazards

Results for selected PHEs in staple food crops in the studied ASGM villages are summarised in Table 4, whilst the whole dry weight and

Table 4
Concentrations of selected potentially harmful elements in staple food crops in ASGM villages.

Element	Food crop *(n samples)*	maximum	minimum	p50	p95	JECFA (FAO/WHO) guidelines	% over-standard by samples and range
	units	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	%, (mg kg^{-1})
Al	Beans **(15)**	8755	3.9	50	3238		
	Cowpeas **(26)**	273	13.7	37.8	213		
	African nightshade **(13)**	154	10.9	57.2	136		
	Maize ***(42)***	196	0.18	10.4	26.2		
	Tubers **(6)**	95.5	4.2	38.2	85.7		
As	Beans **(15)**	5.7	0.0004	0.039	2.3	0.5	**20**, (0.6–5.7)
	Cowpeas **(26)**	0.93	0.003	0.25	0.23	0.5	**4**, (0.93)
	African nightshade **(13)**	0.58	0.005	0.024	0.53	0.5	**8**, (0.58)
	Maize ***(42)***	0.12	0.00009	0.003	0.05	0.2	nil
	Tubers **(6)**	0.013	0.0008	0.003	0.012	0.5	nil
Cd	Beans **(15)**	0.012	0.0003	0.002	0.01	0.1	nil
	Cowpeas **(26)**	0.011	0.0003	0.001	0.005	0.2	nil
	African nightshade **(13)**	0.06	0.005	0.013	0.05	0.2	nil
	Maize ***(42)***	0.01	0.0001	0.002	0.003	0.1	nil
	Tubers **(6)**	0.004	0.001	0.003	0.004	0.1	nil
Cr	Beans **(15)**	97.5	0.03	0.69	33.6	2.3	20, (2.5–97.5)
	Cowpeas **(26)**	1.6	0.035	0.21	1.32	2.3	nil
	African nightshade **(13)**	0.52	0.05	0.2	0.44	2.3	nil
	Maize ***(42)***	2.1	0.002	0.13	1.8	2.3	nil
	Tubers **(6)**	0.32	0.07	0.15	0.31	2.3	nil
Hg	Beans **(15)**	0.02	0.0001	0.002	0.02	1	nil
	Cowpeas **(26)**	0.97	0.002	0.01	0.1	1	nil
	African nightshade **(13)**	0.1	0.004	0.03	0.1	1	nil
	Maize ***(42)***	0.01	0.00003	0.002	0.01	1	nil
	Tubers **(6)**	0.009	0.0003	0.001	0.008	1	nil
Ni	Beans **(15)**	20.2	1.4	6	18.4	2.7	**87**, (2.8–20.2)
	Cowpeas **(26)**	2	0.04	0.3	0.9	2.7	nil
	African nightshade **(13)**	1.4	0.12	0.2	0.8	2.7	nil
	Maize ***(42)***	1.8	0.1	0.47	1.4	2.7	nil
	Tubers **(6)**	2.95	0.08	0.4	2.4	2.7	**17**, (2.95)
Pb	Beans **(15)**	8.4	0.0005	0.04	2.7	0.1	**27**, (2.1–8.4)
	Cowpeas **(26)**	0.2	0.01	0.02	0.1	0.3	nil
	African nightshade **(13)**	0.12	0.007	0.03	0.08	0.3	nil
	Maize ***(42)***	0.2	0.0001	0.006	0.05	0.2	nil
	Tubers **(6)**	0.17	0.007	0.01	0.14	0.1	**17**, (0.17)

Cowpeas = cowpeas leaves; African nightshade = African nightshade leaves; JECFA guidelines = Joint FAO/WHO Expert Committee for Food Additives guidelines (FAO/WHO, 2011).

fresh weight-based laboratory datasets are provided in Supplementary Tables 16 and 17 respectively, with additional summaries by crop type (Supplementary Table 18), food crop consumption and element intakes (Supplementary Table 19). Concentrations of As in beans, cowpeas, and African nightshade; Cr in beans; Ni in beans and cassava, and Pb in cassava were above respective FAO/WHO guidelines (Table 4). Commonly eaten food types in the study location were Maize, Beans, Cowpeas, African nightshade and Banana with mean ADIs of 0.276, 0.12, 0.099, 0.104 and 0.133 kg.day^{-1}, respectively. Risk potential for consumption of the food items are as follows in a decreasing order importance Beans (Phaseolus vulgaris; HI = 5.383) > African nightshade (Solanum scabrum; HI = 1.199) > Cowpeas (Vigna unguiculata; HI = 0.817) > Maize (Zea mays; HI = 0.457) >Tubers (Potatoes and Cassava; HI = 0.15) (Supplementary Table 24). Hazard index integrates the various concentrations of trace element in the food item, takes into consideration their toxicity (RfD), accounts for the respective quantities of the food consumed and therefore presents a more comprehensive measure of how hazardous a food item could be. In this case, the beans had the potential to be the most hazardous and maize the least of the compared food items. Concentrations of PHEs varied widely among the investigated food crops due to their different element absorption capacities (Khan et al., 2015). Our findings also corroborate earlier studies that shows that maize grains poorly accumulate PHEs (Mohammed et al., 2021; Kaninga et al., 2020b), while beans, leafy and non-leafy vegetables are accumulators of PHEs.

Supplementary Table 19.1 shows the quantities (kg/person/day) of food consumed for each category (cereals, pulses, leafy vegetables, tubers, fruits, and animal proteins) and specific subtypes by the interviewees. From these, respective mean daily element intakes via food crop consumption were generated (Supplementary Tables 19.2) for cowpeas, African nightshade, beans, maize and tubers. These form up to 85 % of the diet in the studied ASGM villages. Full details of the calculations of the mean daily element intake via food crop consumption are shown in Supplementary Table 19.3. The results of the questionnaire on dietary consumption (Supplementary Table 19.1) indicated that by quantity of daily intake, maize is the major staple food crop, and leafy vegetables have the second-greatest consumption rates in the studied ASGM villages, followed by bananas and beans. Most food crops are locally grown in the ASGM villages (Supplementary Table 19.1).

Daily intakes of Al via consumption of 31 % (n=12/39) of the leafy vegetables (range, 0.103–0.41 mg kg^{-1} bw^{-1}), beans (47 %; n=7/15) (range, 0.103–15.9 mg kg^{-1} bw^{-1}) and 17% of the maize (range, 0.103–0.82 mg kg^{-1} bw^{-1}) exceeded the TDI limit (0.1 mg kg^{-1} bw^{-1}) (Supplementary Table 19.2) (FAO/WHO, 2011). Intakes of As per day from the consumption of 7 % of the beans (0.01 mg kg^{-1} bw^{-1}) exceeded the TDI limit (0.0021 mg kg^{-1} bw^{-1}) (FAO/WHO, 2011) (Supplementary Table 19.2). Similarly, calculated average daily intakes of Cr via consumption of 74 % (n=29/39) of the leafy vegetables (0.00011–0.008 mg kg^{-1} bw^{-1}), tubers (0.0002–0.001 mg kg^{-1} bw^{-1}), 93 % of the beans (0.0003–0.18 mg kg^{-1} bw^{-1}), and 93 % of the maize (0.000103–0.0009 mg kg^{-1} bw^{-1}) significantly surpassed the TDI limit (Cr 0.0001 mg kg^{-1} bw^{-1}) (FAO/WHO, 2011) (Supplementary Table 19.2). Cadmium intakes via consumption of all the food crops were below FAO/WHO's TDI (0.025 mg kg^{-1} bw^{-1}) (Supplementary Table 19.2). Consumption of 4 % cowpeas in the ASGM villages resulted in significant daily Hg intakes (0.0015 mg kg^{-1} bw^{-1}) above the

0.0007 mg kg^{-1} bw^{-1} FAO/WHO's TDI for Hg (FAO/WHO, 2011). Ingestion of Ni in the ASGM villages via 40 % of the beans (range, 0.015–0.0.037 mg kg^{-1} bw^{-1}) significantly exceeded the FAO/WHO TDI for Ni (0.013 mg kg^{-1} bw^{-1}). Lead intake from eating 7% of beans (up to 0.015 mg kg^{-1} bw^{-1}) in the ASGM villages was higher than FAO/WHO TDI for Pb (0.01 mg kg^{-1} bw^{-1}) (Supplementary Table 19.2) (FAO/WHO, 2011).

3.5. Risk factors for PHE exposure among the studied ASGM communities

These were evaluated in 180 respondents (ASGM workers, n=144; residents of ASGM, n=25; residents of non-ASGM villages, n=11) as detailed in Supplementary Table 23.1. The ASGM activities are conducted near residences, within or along water sources and agricultural fields. Tailings contaminated with Hg and other PHEs are haphazardly disposed of in residential areas, agricultural fields, and water sources (Ondayo et al., 2023a). In the present study, the median distance between participants' households and ASGM activities was ≤100 m (range ≤100–15,000 m). Most of the ASGM workers and residents of ASGM villages (78 %; n=131/169) live within ≤100 m radius of ASGM operations, namely, mine excavation; ore crushing; ore washing, sluicing, panning and amalgamation; amalgam burning; gold buying and selling; dumping of mine tailings/ waste at home or in the farm, and in some cases cyanidation plants. Thus, most residents of ASGM villages have similar risk factors to PHE exposures as the ASGM workers. Conversely, some of them live 300–500 m (2 %; n=4/169) and >500 m-15,000 m (20 %; n=34/169) from ASGM activities. Most ASGM workers and ASGM residents (72 %; n=122/169) reported that ASGM operations, including ore milling and Hg-Au amalgam burning, were being carried out inside their homes and households (Supplementary Table 23.1).

Inhalation of airborne PHEs, especially Hg (Black et al., 2017; Gyamfi et al., 2020), and respirable PHE-ladden dust are essential pathways in the studied ASGM villages (Ondayo et al., 2023a; Ondayo et al., 2023b), as observed by the authors and reported by 90 % of the subjects. Many homesteads, playgrounds, yards (53 %) and house floor material (46 %) were bare soil. The walls, floors, and roof surfaces had crevices on which PHE-containing dust was embedded. As observed by the authors, most of the dirt-filled house floors are dry-swept, with children nearby. The subjects further reported that powdered ores, gold extraction chemicals (Hg and CN), and tailings are stored in peoples' living rooms (76 %; n=110/144), bedrooms (71 %; n=103/144), kitchens (35 %; n=51/144), other rooms in the house (25 %; n=36/144), at work (86 %; n=125/144), and outside (4 %; n=6/144). During the interviews, the authors observed powdered ores, mine tailings, and gold extraction chemicals in 83% (n=140/169) of the homes and households. Specifically, these were observed in children's playgrounds (69 %; n=117/169), crop farms (71 %; n=120/169), driveways (69 %; n=117/169), along fences (67 %; n=113/169), and inside houses where people live (72 %; n=122/169) as presented in Supplementary Table 23.1.

Individuals overall risk of PHE exposure comes from the different workplace points as 90 % of the ASGM workers conducted in multiple ASGM activities, notably, mine excavation, ore crushing and milling, washing, sluicing and amalgamation, amalgam burning, and gold buying and selling, except for cyanidation (10 %). For each ASGM worker interviewed, an average of two of their family members (range, 1–14 family members) were similarly involved in ASGM activities and had primarily conducted ASGM for a median time of 5 years (mean, 7.6 years; range 0.08–39 years). They also reported having had direct contact with Hg (90 %; n=130/144), CN (4 %; n=6/144), and both Hg and CN (3 %; n=4/144). While at work, the ASGM workers had Hg (83 %; n=121/144), CN (4 %; n=6/144), and both Hg and CN (9 %; n=13/144) spillages. Similarly, they reported accidental spillages of Hg (51 %; n=87/144), CN (3 %; n=5/144), and both Hg and CN (4 %; n=7/144) while in their homes. Furthermore, ASGM workers potentially track PHEs to their homes and immediate family members as they store ASGM work clothes in their living rooms (55 %; n=80/144), bedrooms (43%; n=63/144), kitchens (15 %; n=22/144), other rooms inside their houses (4 %; n=6/144), outside (55 %; n=80/144), and at work (7 %; n=7/144). The interviews additionally revealed geophagy among 27 % of the subjects (children (n=35) and pregnant women (n=3) (Supplementary Table 23.1).

3.6. Linking environmental data, food and biomonitoring results

Various PHEs from ASGM activities have been shown to contaminate surface and groundwater sources, soils, sediments and food, accumulate in human body systems, and cause injury, disease and deaths across Africa (Ondayo et al., 2023b) and globally (Allan-Blitz et al., 2022; Landrigan et al., 2022). Previous research in the studied ASGM villages found high PHE levels, especially As, Cd, Cr, Hg, Ni, and Pb in soils, waters and sediments. Besides, the PHEs were generally highly gastric bioaccessible, with up to 72 % of the respective total PHEs solubilized in 20 agricultural; gold ore mining and processing sites; and schools, playgrounds and residential soil samples analysed (As,1–60% of the total; Cd, 2–56 % of the total; Cr, 0–18% of the total; Hg, 1–72 % of the total; Ni, 0.4–14% of the total; and, Pb, 1–35 % of the total) (Ondayo et al., 2023a). The highest percentage of bioaccessibility was measured in moderate to heavily polluted agricultural soils (Hg 35–51 % of the total; Pb 24–25 % of the total); schools, children's playgrounds and residential soils (Hg 2–39 % of the total; As 2–60 % of the total); and, ore mining and processing sites (Hg 1–72 % of the total).

This present study's chemical analyses of drinking water (Table 3) and 102 edible African nightshade, beans, cowpeas, maize and tuber samples found appreciable Al, As, Cd, Cr, Hg, Ni and Pb contamination (Table 4). The same suites of Al, As, Cd, Cr, Hg, Ni and Pb containing minerals were found in the community drinking water sources and staple food crops as the ores and soils previously reported (Ondayo et al., 2023a). This mineralogical fingerprint confirms transfers of various PHEs from soils and natural waters to locally grown crops. Additionally, leafy vegetables, stored cereals, and pulses were contaminated by ore processing dust. Elevated concentrations of Hg in cowpeas leaves (0.002–0.97 mg kg^{-1}) (Table 4; see also Supplemental Tables 16–17) found in this study indicate processing-related contamination, possibly from airborne Hg, and food crops storage in rooms where Hg is stored and amalgam burning is done. Inhalation of gaseous PHEs and crystalline respirable soil dust; incidental ingestion of soils via hand-to-mouth transmission, and direct ingestion of soil dust trapped in the mucus lining of the respiratory tract and drinking water were previously reported as the key PHE exposure pathways that posed a growing risk of non-cancer health effects (98.6) and cancer among adults (4.93×10^{-2}) and children (1.75×10^{-1}) in the studied ASGM villages (Ondayo et al., 2023a). Additionally in the present study, ASGM workers and local community members are exposed to multiple PHEs principally through consumption of untreated contaminated drinking water (for Al, As and Cr) (Table 3; Supplementary Table 22.1) and locally grown staple food crops, notably cowpeas (for Al, As, Cr and Ni), African nightshade (for Al, As, Cd, Cr and Hg), beans (for Al, As, Ni and Pb) and, maize and cassavas (for Cr) (Table 4; Supplementary Table 19.3).

The HBM (nails, hair, and urine) findings (Figs. 2–3; Table 2) confirm human exposure to the same suite of PHEs as those found in the dietary and environmental matrices (ores, soils, water and sediments). Spearman correlation analysis between investigated staple food crops, drinking water, nails, hair, urine, and soils revealed that there are strong positive linear relationships which point to drinking water (R = 0.958 for As in drinking water and urine), soil, and farm-to-fork pathways for Al, As, Cr, Ni, Hg and Pb as presented in Supplementary Table 23.2. Weak positive correlations were found between As in food crops and hair (R = 0.26), nails (R = 0.32) and urine (R = 0.32). These findings are similar to findings in ASGM in Nigeria, which linked geological enrichment, environmental contamination (soil, food, water, and dust), human exposure and health effects, including deaths (Dooyema et al., 2012; Plumlee et al., 2013).

3.7. Health effects among the studied ASGM communities

Following absorption, PHEs accumulate in human matrices and cause various health effects (Balali-Mood et al., 2021; Mitra et al., 2022; Zeng and Zhang, 2020), as documented in ASGM as well (Basu et al., 2015; Landrigan et al., 2022; Ondayo et al., 2023b). Primarily, PHEs have similar toxic mechanisms, notably reactive oxygen species (ROS) generation, inactivation of enzymes, and antioxidant defence suppression that lead to various health effects (Balali-Mood et al., 2021; Mitra et al., 2022). Conversely, some PHEs additionally bind selectively to specific macromolecules, for instance, the interaction of Pb with ALAD and ferrochelatase and the substitution of Ca^{2+} by Cd and Pb in neural cells (Balali-Mood et al., 2021; Mitra et al., 2022). Globally, neurological disorders among human populations are strongly linked to exposure to As (Thakur et al., 2021), Cd, Pb (Axelrad et al., 2022; Gonçalves et al., 2021; Mitra et al., 2022; Simões et al., 2021), CN (Isom and Borowitz, 2015), and Hg (Zhu et al., 2022).

Typical urinary Hg concentrations in unexposed individuals are less than 10 μg L^{-1} (Ye et al., 2016). Urinary Hg concentrations ranging from 30 to 100 μg L^{1} have been linked with subclinical neuropsychiatric symptoms and tremors, while those greater than 100 μg L^{-1} can be associated with overt neuropsychiatric disturbances and tremors (Ye et al., 2016). This study's urinary Hg ranged between 11.1 and 178 μg L^{-1} among active ASGM workers (12 %, n=11) living within a 100 m radius from ASGM activities and had direct contact with Hg. Similar findings were also reported among children and adult ASGM workers and community members in Nigeria (Dooyema et al., 2012), Tanzania, and Zimbabwe (Bose-O'Reilly et al., 2010; Bose-O'Reilly et al., 2017; Bose-O'Reilly et al., 2008a; Harada et al., 1999).

Human exposure to Al, Be, Cd, CN, Cr, Cu, Fe, Hg, Mn, Ni, Pb, Ti, and Zn causes a wide range of respiratory problems (Isom and Borowitz, 2015; Mitra et al., 2022; Zhou et al., 2022). In the studied ASGM villages, the suspension of PHE-contaminated respirable dust particles in the air exposed mine excavators, ore crushers and millers, and the nearby community members to the risk of respiratory infections (Ondayo et al., 2023a). Similar exposure risks were reported for ASGM sites in Burkina Faso, Cameroon, and Ghana, where respiratory ailments were linked to As, CN, Hg, and Pb exposure. High prevalence of silicosis and tuberculosis associated with inhalation and ingestion of contaminated dust exposure in ASGM have been reported in Tanzania and Zimbabwe (Mbuya et al., 2023; Moyo et al., 2022; Moyo et al., 2021).

Signs and symptoms that may be associated with As poisoning reported in this study include headaches, ulcers, stomach cancer among other cancers, hypotension, fever, and neurological effects (Balali-Mood et al., 2021; Ondayo et al., 2023b). Global literature links various cancers to As, Cd, Cr, (hexavalent), Ni, Pb, and SiO_2 exposures (Chou and Harper, 2007; IARC, 2021; Mitra et al., 2022). Growing risk of non-cancer health effects (97) and cancer among adults (4.93×10^{-2}) and children (1.75×10^{-1}) potentially exposed to As, Cd, Cr, Ni, and Pb were previously estimated in the studied ASGM villages (Ondayo et al., 2023a). Further investigations, including clinical examination and mapping of cancer cases in ASGM, are crucial. These health effects, alongside injuries, can be fatal or result in disabilities, as found in ASGM in Burkina Faso, Nigeria, and Cameroon (Dooyema et al., 2012; Ralph et al., 2018; Tomicic et al., 2011). Baseline health data in the villages studied is lacking as the record-keeping system used in nearby hospitals was still paper-based, contrary to computerised hospital systems.

Various findings that reported diseases in ASGM communities in more than 30 African countries support this study's results. The health problems found in the studied ASGM sites result from poor knowledge of health, safety and pollution controls by the ASGM communities and a lack of regulation and or enforcement of public health and occupational safety and health measures in the field by responsible authorities. Most ASGM workers (62 %; n=89/144) in this study reported not using PPE while working. As observed, 38 % (n=56/144) of the ASGM workers who reported using dust masks, boots, helmets, or gloves did not use complete PPE sets or regularly, which is the case in most ASGM activities across Africa (Afrifa et al., 2017; Ondayo et al., 2023b).

Other hazards that were non-specific to the workplaces included alcohol consumption, cigarette smoking, use of skin-lightening products, child labour (n=3), the labour of expectant women (n=3), and labour of nursing mothers (n=16). In this study, 42 % of the workers in their reproductive ages were female (42 %). Female active involvement in ASGM activities is highly likely to increase PHE exposure to fetuses, neonates and older children living in these communities (Bose-O'Reilly et al., 2020; McDowell et al., 2004). Women in Kenya and across Africa work with their children in mining sites, thus exposing them to PHEs (Bose-O'Reilly et al., 2020; Ondayo et al., 2023a; Ondayo et al., 2023b).

3.8. Limitations and strengths of this study

Our study did not conduct PHE speciation, especially As, Hg, and Cr, to account for PHE sources and toxicities to better predict binding phases in soils, bioavailability and leaching into groundwater by incorporating such data into modelling methods such as the Windermere Humic Aqueous Model (WHAM). Such data and use if models may help to inform transfer of PHEs from tailings to soils and uptake by plants (Kaninga et al., 2020a; Kaninga et al., 2020b; Kaninga et al., 2021). The second limitation was the small sample size of residents of non-ASGM villages and the relatively smaller nail sample size compared to the urine sample size – although this study had a much bigger sample size than many studies reported in the literature. The reasons for this are common among community-based studies, including families not home when visited, refusal to participate, resource constraints, and the commonly observed and self-reported chronic nail loss among ASGM workers working with bare hands. Our sample size restricted this study's efforts to evaluate effect modification between exposure and health effects and demographic variables among residents of non-ASGM villages. The CRM recoveries for hair and nail Cr concentrations were, thus were not reported.

Nonetheless, this study demonstrates that ASGM activities are important sources of multiple PHE exposure for ASGM workers and residents of ASGM villages, with severe health consequences. Using randomized multiple biomonitors (hair, nails, and urine), interviews, and dietary and environmental evaluation allowed for better characterisation and understanding of the extent of exposure and hazard, including estimation of both long and short-term distribution of PHE concentrations in the population of interest and can assist policymakers in designing holistic exposure remediation and prevention interventions. The holistic multidisciplinary approach in this study informs other researchers to improve the quality of exposure and risk assessments and be more transparent with research data.

3.9. Opportunities for further research

Given the PHE findings in the locally grown staple food crops, additional research to understand the mechanisms of environment (water/soil)-food crop element transfers and conversion factors is crucial for developing effective remediation technologies. Further study to test PHE exposure in human blood matrices and clinically examine the ASGM communities is crucial. Finally, a further study that enrolls a larger number of children, pregnant women, and nursing mothers to provide more detail on their exposure status, behavioural and nutritional status, and general health would provide additional information for policy and decision-makers and public health professionals to put in place population protective measures.

4. Conclusion

This is the first comprehensive multidisciplinary public health assessment of ASGM communities that employs multi-element biomonitoring using multiple matrices that complement each other (hair, nails, and urine), dietary and environmental evaluation, and exposure risk factors assessment. The hair, nails and urine findings revealed a high body burden of multiple PHEs, especially Al, As, Cr, Cd, Hg, Ni and Pb, among ASGM workers and residents of ASGM villages, most of whom (80 %) were in their reproductive ages (16–49 years), with multiple health implications, including, fatalities. Urinary As, Cr, Hg, Pb and Ni in over 76 % of ASGM workers and residents were up to 22, 3, 42, 17 and 7 times greater than respective upper health-based thresholds and bioequivalents. Aluminium, As, Cd, Cr, Hg, Ni and Pb concentrations higher than the FAO and WHO maximum allowable limits were found in community drinking water sources and locally grown staple food crops in the studied ASGM villages. Leafy vegetables (cowpeas and African nightshade) and pulses (beans) bioaccumulated most PHEs, and maize was the least bioaccumulator, though not safe from Cr. The PHE concentrations in drinking water, locally grown staple food crops, and environmental media were positively correlated hair, nails and urine, pointing to an environment-farm-fork pathway. Kenya has no national regulatory standards for all the PHEs in drinking water and food and health-based bioequivalents for hair, nails, urine and other human biomonitoring matrices. Thus, developing specific biomarkers for monitoring PHEs will be a significant achievement in Kenya. Regularly monitoring PHEs in food crops and drinking water within the ASGM villages is essential to prevent potential health risks. Safe drinking water alternatives, like piped water, should be provided to the ASGM communities.

The study recommends decreasing and or eliminating PHE exposure in ASGM communities and implementing safer ASGM alternatives sustainably. This includes explicitly relocating residences and schools far from ASGM activities; encouraging regular 100 % PPE use among ASGM workers; reducing dust by more efficient crushing, for instance, use of wet milling; safer alternatives to Hg and use of retorts; stopping irrigation of crops using PHE-contaminated waters; and safer cyanidation processes, which could reduce PHE exposure. All ASGM processes should be adequately regulated, and the ASGM workers' and residents' health should be protected. There is a need for training and making the communities fully aware of the environmental and health hazards associated with ASGM and the need for stringent safeguards. Study results have been shared with the respective Health Services, Agriculture and Environment county departments to inform mitigation efforts, including follow-up of the exposed study participants.

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CRediT authorship contribution statement

Odipo Osano: Writing – review & editing, Writing – original draft, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Conceptualization. **Maureene Ondayo:** Writing – original draft, Methodology, Investigation, Data curation, Conceptualization. **Michael Watts:** Writing – review & editing, Writing – original draft, Validation, Supervision, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization. **Olivier Humphrey:** Writing – review & editing.

Declaration of Competing Interest

Michael Watts reports financial support was provided by Natural Environment Research Council. Michael Watts reports financial support was provided by The Royal Society. Michael Watts reports financial support was provided by The British Academy. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2024.116323.

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