



Research article

Mercury contamination of soil and water media from different illegal artisanal small-scale gold mining operations (galamsey)

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ABSTRACT

Environmental media contamination with mercury, because of illegal artisanal small-scale gold mining (popularly called galamsey), is a major concern in Ghana; yet specific details as to how such contaminations are influenced or distributed across different galamsey operations have been lacking. We monitored mercury levels across nine different galamsey operations (Washing Board, Washing Plant, Anwona, Dig and Wash, Dredging, Underground Abandoned Shaft, Underground Sample Pit, Chamfi and Mill House) in three hotspot assemblies (Tarkwa Nsuaem, Amenfi East and Prestea Huni Valley) of the Western Region of Ghana. Triplicate samples each of background soil, surface water/drainage, slurry/sludge and galamsey waste materials (totaling 160) were obtained and analyzed using Atomic Absorption Spectrophotometer (AAS) to determine total Hg concentrations. From the comparison of mean ranked concentration of mercury, using the Kruskal-Wallis Test, it was realized that the observed differences in ranking was significant for all four environmental media considered. Thus, the poor handling, usage and disposal of mercury from the different galamsey activities did result in elevation of harmful quantities of mercury into the environment. Overall, the highest median value obtained for mercury was recorded at the Mill House galamsey sites and within slurry/sludge medium. This was followed by Chamfi, Washing Board, Washing Plant, Anwona, Dig and Wash and Dredging in descending order, with the Underground Abandoned Shaft and Underground Sample Pit galamsey types recording values below detection limit. In terms of their contribution to mercury contamination to the environment, Mill House, Chamfi, Anwona, Washing Board and Washing Plant galamsey types recorded the highest mean rankings. Overall, key priority information required for influencing reclamation and cleanup policy decisions for mercury, for the many affected wastelands across the country, can be derived from this paper.

1. Introduction

Artisanal small-scale gold mining (ASGM), the source of approximately 20% of the world's gold, is the largest source of mercury (Hg) pollution on earth; contributing approximately 20–30% (Veiga et al., 2006; UNEP, 2005). Roughly 10 to 19 million people use mercury to extract gold in more than 70 countries, making mercury pollution from ASGM a global issue. In addition, 13 million people worldwide (including children), work in artisanal gold mining and use elemental mercury to extract gold from ore (IPEN, 2013). To extract 1 g of gold, for instance,

miners need 1.5–2 g of mercury. Some 650–1000 tonnes of mercury are released annually, with an estimated 350 tonnes entering the atmosphere directly and the rest released into water systems. In addition, mercury release from tailings and vaporized mercury exceed 1000 tonnes each year from ASGM (Balifokus, 2012).

Galamsey, a popular term for illegal artisanal small-scale gold mining (ASGM), is widespread in Ghana. It is operationally diverse in its mining and beneficiation processes, has varying scale, footprints and complexity (Owusu-Nimo et al., 2018; Mantey et al., 2016; Hinton et al., 2003; Veiga, 1997). Thus, galamsey can be small, medium or large scale,

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typically employing either rudimentary, semi-mechanized or fully mechanized techniques. In other words, the conventional techniques of geological exploration, drilling, proven reserves, or engineering studies are not employed (Mantey et al., 2016; Villegas et al., 2012; Walrond, 2012; Aryee et al., 2003; Hinton et al., 2003; Amegbey and Eshun, 2003; Hilson, 2001; Ntibrey, 2001; Veiga, 1997).

This illegal gold mining business is widespread in its distribution; often ranging from forests, water bodies, underground and within urban centers (Owusu-Nimo et al., 2018). In a study by Mantey et al. (2016) within eleven (out of the 22 assemblies) municipal and district assemblies (MDAs) in the Western Region of Ghana, eleven (11) operational forms of galamsey (under five (5) broad categories) were unearthed. Some of the very traditional and purely artisanal alluvial galamsey operations like Panning, Selection and Dig and Wash, as well as those from underground mining types (Abandoned Shafts and Sample Pits/Hole) are reported to make limited or no use of mercury in their processing or beneficiation activities (Owusu-Nimo et al., 2018; Mantey et al., 2016). On the other hand, the commercially, mechanically and chemically driven types like Mill House, Chamfi, Washing Board, Washing Plant, Dredging and Anwona galamsey operations involves the use of substantial volumes of mercury in their gold extraction (Mantey et al., 2016).

Throughout Ghana, where artisanal gold mining is an important economic activity for many rural communities, mercury amalgamation and gold recovery through heating has not only been practiced for decades, but has also increased in terms of both intensity, scale and scope. Frequent inappropriate discharge of mercury into the environment, especially from galamsey, has reportedly led to elevated levels of the metal in various media such as soils, surface waters, sediments, sludge and slurry, crops, fish, plants and humans (Mantey et al., 2016; Tschakert and Singha, 2007; Cortes-Maramba et al., 2006; Hilson, 2006; Hinton, 2005; Spiegel and Veiga, 2005; Amegbey and Eshun, 2003; Aryee et al., 2003). The use of mercury is the favored practice by galamsey operators because it is easy to use, highly effective in the capture of gold, very accessible and easy to transport as well as cheaper compared to other methods (Telmer and Veiga, 2009).

In the artisanal mining beneficiation process, mercury combines with the gold to form an amalgam, which facilitates its separation from other minerals of no immediate financial interest. Thus, separation process can be accomplished using mainly two techniques; each one releasing different quantities of mercury into the environment (Telmer and Veiga, 2009; Hylander et al., 2007; Veiga et al., 2006). In the first technique, which is least practiced by galamsey operators due to the high quantities of mercury used and cost implications, 100% of the material is amalgamated during the crushing, grinding and washing process. This method is considered the most polluting use of mercury as it requires 3–50 units of mercury to produce one unit of gold, on average about 5 units. The second technique, which is employed by the different galamsey operations at different scales relative to the business size and operational style, involves either the crushing, grinding, washing, screening or gravimetric concentration of gold bearing material whereby the auriferous mineral collects with the heavier particles in a pan and the water washes away the lighter particles. Mercury is then added to the leftovers in the pan to amalgamate the finest particles of gold. The amalgam is typically isolated by hand and then heated often with a torch or over a stove to distill the mercury and isolate the gold (Telmer and Veiga, 2009; Hylander et al., 2007; Veiga et al., 2006). In this case, between 1 and 2 units of mercury is used to produce one unit of gold. In some cases, amalgams are processed near the home or in gold shops in villages or cities, so the mercury vapor generated in the process affects non-miners living in these areas. Practices such as whole ore amalgamation, open heating of amalgams, heating amalgams in residential areas, direct addition in trammel etc. are employed by different galamsey operations.

Mercury contamination is widely known to be a pressing problem in the galamsey sector in Ghana and has been demonstrated to be widespread, especially in biota as well as many of the human populations residing within the country's gold belts (Golow and Mingle, 2003; Golow

and Adzei, 2002; NSR, 1994). Indeed, the environmental and health-related impacts of mercury exposure have been widely documented in literature for over 40 years (Chan et al., 2003; Eisler, 2003; Wolfe et al., 1998; D'Itri, 1972; Friberg and Vostal, 1972; Klein and Goldberg, 1970; Johnels et al., 1967), popularized initially by accidents in Minamata (Japan). Galamsey activities are reported to release a significant quantity of mercury to the air, causing severe damage to soils, water and wildlife near the operating sites, and results in heavy mercury exposures to the miners and their families (Amegbey and Eshun, 2003; Aryee et al., 2003).

The health effects on the miners are dire, with inhaled mercury leading to neurological damage and other health issues. The communities near Galamsey sites are also affected due to mercury contamination of water and soil and subsequent accumulation in food staples, such as fish—a major source of dietary protein. The risks to children are also substantial, with mercury emissions from ASGM resulting in both physical and mental disabilities and compromised development. Mercury vapor inhaled by illegal mine operators could result in impaired cognitive function, neurological damage, kidney damage and several health problems. Mercury released into the atmosphere by the roasting of amalgam causes respiratory tract problems upon exposure (Cordy et al., 2011). In spills, mercury is deposited directly into the soil or water bodies (Telmer and Veiga, 2009). Mercury in the soil is absorbed by plants and makes its way to the fruits and seeds consumed by herbivorous species (Eisler, 2004). In water, mercury is methylated by the bacteria in sediments producing methylmercury, which bio accumulates along the food chain and accumulates, in high concentrations, in organisms that predate on fish (Liu et al., 2012). By consuming the fish contaminated with methylmercury or the plants grown from contaminated soils, humans become the final recipients of the mercury (Liu et al., 2012; UNEP, 2005; Eisler, 2004).

Although acknowledged widely that the extent of environmental damage caused by mining is dependent on the mining method, the beneficiation processes, the operational scale, location and characteristics of the receiving environmental media (Lein, 2012; Hinton and Holstelle, 2012; United States EPA, 2000), the degree of mercury contaminations caused to water and soil media have not been adequately related to galamsey types. So far, research efforts to assess the effects of galamsey on the environment from mercury have been very generic and failed to relate the levels of contaminations to specific galamsey types. Thus, the extent or degree to which various types of galamsey influences mercury pollution levels within surface drainages, soils, slurry and solid wastes is largely unknown.

This research work therefore aimed at assessing the extent to which nine (9) types of galamsey influences mercury concentrations within surface drainages, soils, slurry/sludge and wastes in three hotspot assemblies in the Western Region of Ghana.

2. Materials and methods

2.1. Site selection study

The following assemblies in the Western region of Ghana, West Africa; Tarkwa Nsuaem Municipal Assembly, Prestea Huni Valley Municipal Assembly and Amenfi East District Assembly, were selected for the study (see Figure 1). These three assemblies have been identified by Owusu-Nimo et al. (2018) and Mantey et al. (2016) as the three hotspot locations for galamsey in the Western Region; having the highest number, distribution diversities, operational scale and significant visual environmental impacts of the illegal operation.

Triplicate samples each of background soil, surface drainage, slurry/sludge and galamsey waste materials for nine galamsey types (Washing Board, Washing Plant, Anwona, Dig and Wash, Dredging, Underground Abandoned Shaft, Underground Sample Pit, Chamfi and Mill House) were collected from two randomly selected galamsey host village or

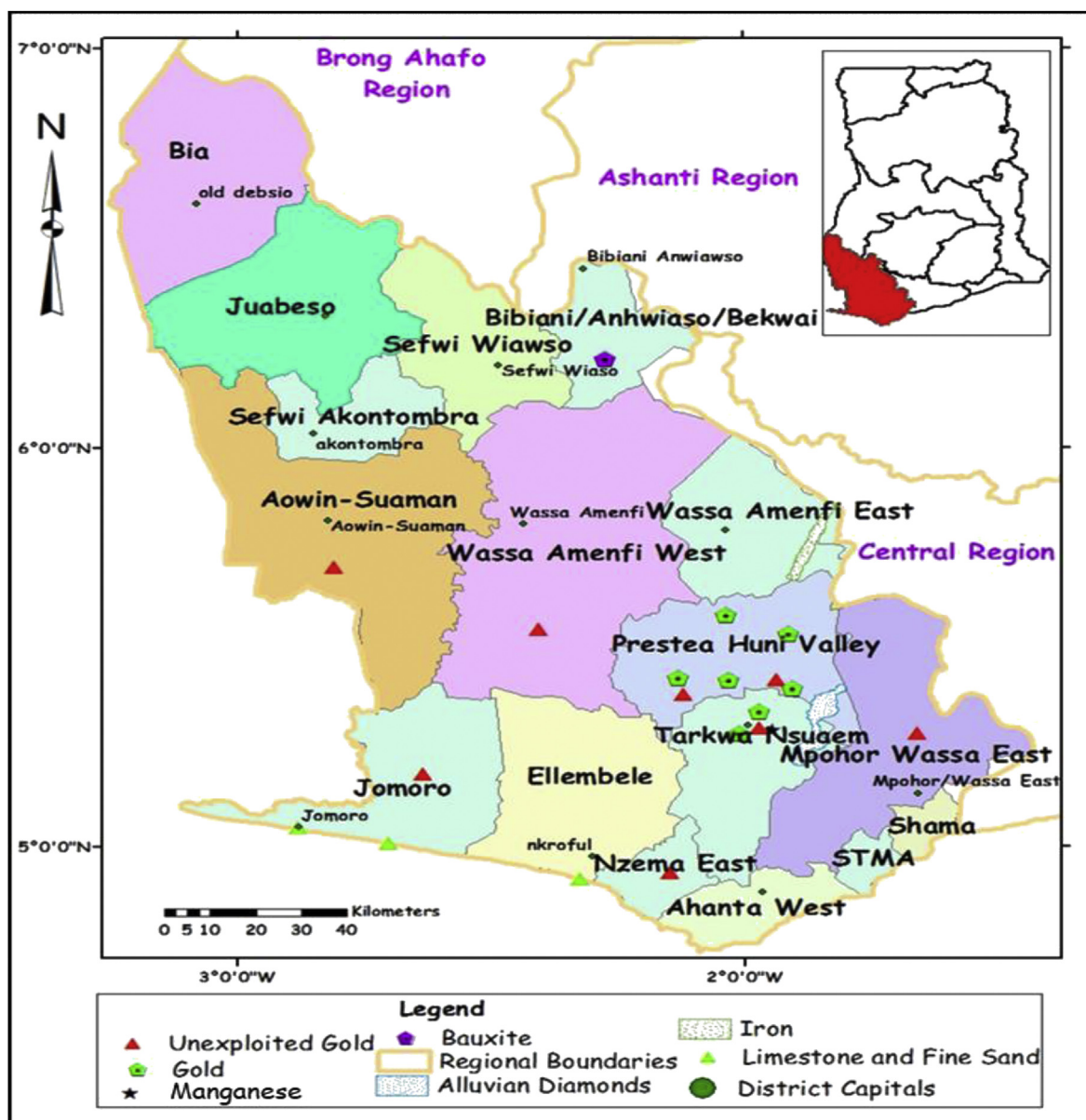


Figure 1. Map of the Western Region-Ghana showing the study sites (district assemblies of interest) and mineral deposits (Western Regional Coordinating Council, 2016).

communities in each of the three galamsey hotspots assemblies (Tarkwa Nsuaem, Amenfi East and Prestea Huni Valley).

At each sampling point, for background soils, galamsey wastes and slurry/sludge, three composite samples each at depths of between 0-50 cm and a mass of 1500 g, using a hand auger and polyethylene shovel, were collected from 30 different spots on impacted areas that were identified to be the main mining and or gold extraction/ore processing locations and other generally impacted areas on the galamsey site.

The sampled soil, slurry/sludge and waste materials from the galamsey sites were stored in adequately labelled 'Ziploc™ bags, which had been washed and rinsed with distilled water and 20% HNO₃ before sampling. The samples were preserved by adding 1.5 mL of conc. HNO₃ and stored in a refrigerator at about 4 °C for subsequent analysis, as per standard procedure (U.S. Geological Survey (USGS), 2006; Voutsas et al., 2001; Voutsas et al., 1999; APHA, AWWA & WPCF, 1985). Control or reference samples obtained in a similar vein from randomly selected communities (Moseaso, Huni Ano and Esaman Kakrabawere) where no galamsey operations are known to be carried out within each of the three-galamsey hotspot assemblies. It is worth stating that, due to the

absence of galamsey in the reference sites, no waste samples were obtained; only slurry/sludge, base soils and surface drainages were obtained.

Surface drainage/water samples were collected from any of the following surface drainages found within the active footprint of the nine different galamsey sites; ponds, streams, run-off water, wetlands, surface pooling and pit lakes. They were sampled into 330 mL pre-cleaned high-density polyethylene bottles. In situations where almost all of the above drainages or more than one of them exist, samples were collected using the worst-case scenario principle, which is, the visually most polluted water was collected. The bottles were conditioned by washing with 5% HCl, and then rinsed several times with de-ionized water. This was done to ensure that the sampling bottles were free from contaminants (U.S. Geological Survey (USGS), 2006; Voutsas et al., 2001; Voutsas et al., 1999; APHA, AWWA & WPCF, 1985).

Preservation of mercury metal in the samples were achieved by acidification with 4 mL of concentrated HNO₃ (70%) to 1 L of sample. In all, a total of 160 surface drainage/water (40), soil (40), slurry (40) and wastes (40) samples were obtained and transported to the Nuclear

Chemistry and Environmental Research Laboratory of Ghana Atomic Energy Commissions for mercury analysis using Atomic Absorption Spectrophotometer (AAS). The geographical location (latitude and longitude) of each sampling point was recorded with a mobile device, using Open Data Kits (ODK) as described by Anokwa et al. (2009) and a map generated (as depicted in Figure 2).

2.2. Distribution of the types of galmsey and samples

Of the nine (9) types of galmsey considered in this study, their presence in the three-galmsey hotspot assemblies was seen to vary. While the Tarkwa Nsuaem and Amenfi East study areas had four (4) each

of galmsey types from which samples were collected, the Prestea Huni Valley study area had 5 types of galmsey. Due to the spatial distribution dynamics observed for the various galmsey types across the study areas (see Owusu-Nimo et al., 2018; Mantey et al., 2016 for details) and the random sampling approach used (for each of the 9 type of galmsey with at least one MDA and two host villages randomly sampled), no single type of galmsey could feature and provide samples for all three study areas. However, each of the following, Anwona, Dig and Wash, Washing Board and Underground Abandoned Shaft, were common to two different combination of the study areas (Figure 3).

For each of the nine (9) types of galmsey, six (6) each of environmental media types (slurry/sludge, soil, waste and surface drainage/

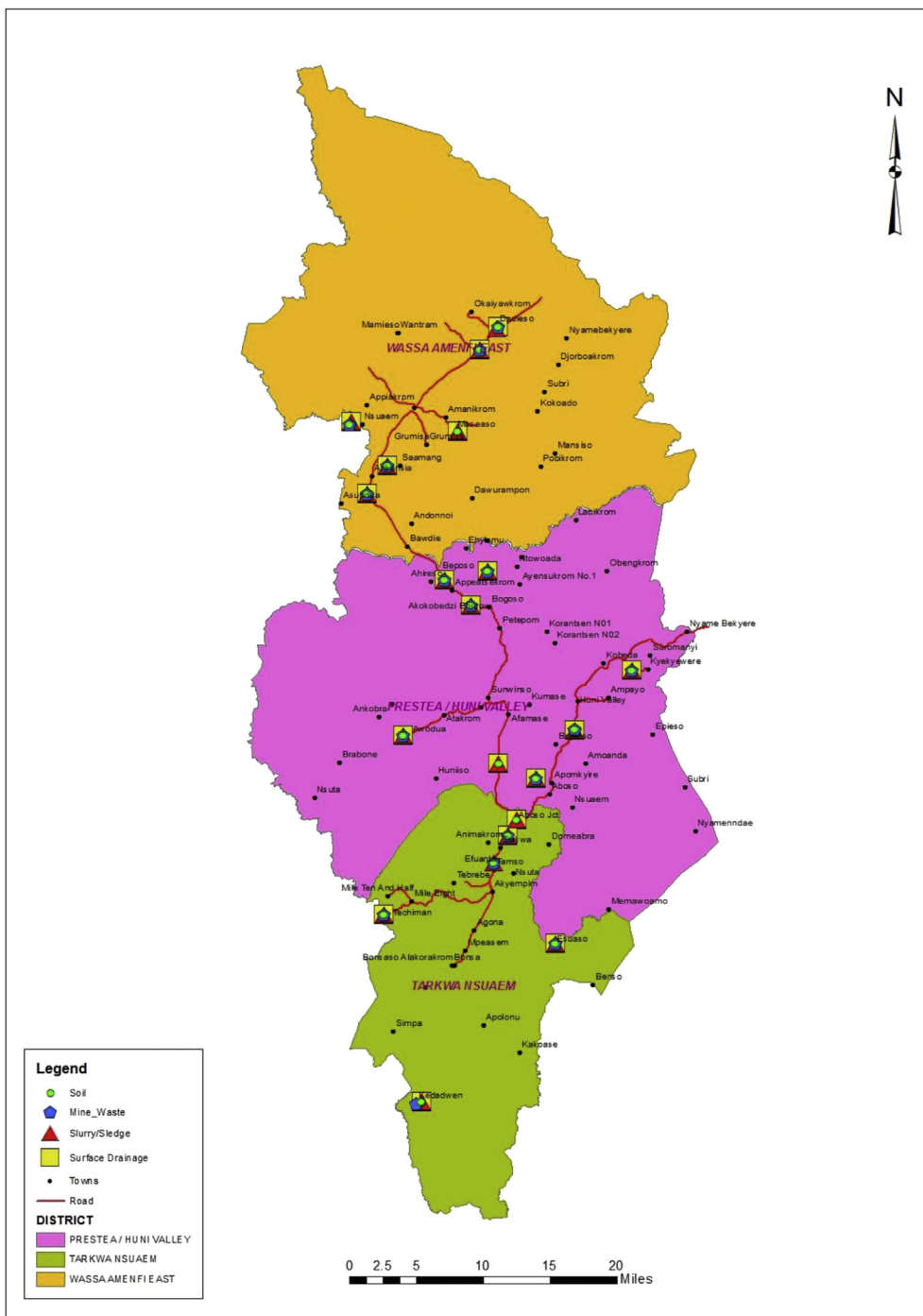


Figure 2. Sampling Stations for all four environmental media within the study sites (three main Assemblies/Districts of interest) in the Western Region of Ghana.

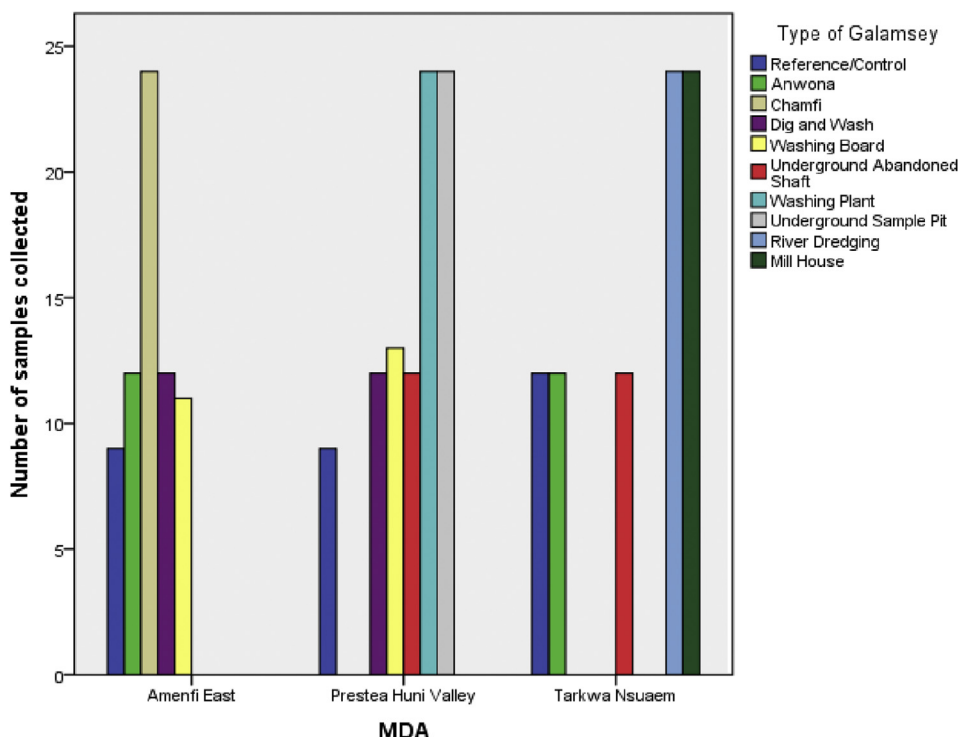


Figure 3. Distribution of the number of samples collected stratified by the type of galamsey activity in each MDA.

water) were sampled. Corresponding reference/control samples (areas having no associations with galamsey) were also collected, with nine (9) slurry, soil each and 12 water samples. The equal number of samples ensured that selection bias was reduced to the minimum.

2.3. AAS analysis of metals (Hg)

Hg analysis of the samples were analyzed by Varian model AA 240 FS electrothermal Atomic Absorption Spectrometer (purchased from Varian Inc., California, USA) which is one of the most extensively used techniques for determining various elements with significant precision and accuracy. This analytical technique is remarkable for its selectivity, speed and fairly low operational cost. The AAS was equipped with quartz T tube and GTA 3000 graphite furnace, with deuterium arc background corrector using the following analytical conditions: a high intensity Hg hollow cathode lamp was used as the source of radiation with a lamp current of 4.5 mA and the resonance line of Hg analytical line adjusted on 253.7 nm, with a spectral band (slit) width of 0.5 nm was used. Flow rate of high purity argon gas and reading time were 50 ml/min and 10 s were

Table 1. Operational conditions employed in the determination of Hg.

Parameter	Set value
	Hg
Wavelength (nm)	253.7
Source Lamp (mA)	4.5
Slit Width (nm)	0.5
Background Correction	-
Purge gas	Argon
Argon flow (mL/min)	50
Calibration Algorithm	Linear thru Zero
Sample Volume	20
Matrix Modifier Volume	-
Graphite furnace	High intensity hollow cathode lamp

used for the determination of Hg. Table 1 presents the operational conditions employed in the determination of Hg.

2.3.1. Digestion of slurry/sludge and surface water samples

To ensure the removal of organic impurities from the samples and thus prevent interference in analysis. Triplicate sample each of slurry and surface water samples totaling 80 samples were digested (using Multi-wave 7000 microwave digester purchased from Anton Paar GmbH, Austria) each and separately with concentrated nitric acid. 10 mL of nitric acid was added to 50 mL of each samples in a 250 mL conical flask. The samples were then digested following five-steps program: (i) 5 min at 150 °C and 50 % power; (ii) 5 min at 220 °C and 70 % power and (iii-iv) 5 min at 100 °C and 10 % power after which it was allowed to cool and then filtered. Finally, the filtrate was adjusted to pH 3.0–4.5 with dilute HNO₃ before it was made to volume in a 10-ml calibrated flask. A reagent blank was prepared in parallel. The dilute sample (10 mL) was analyzed immediately after preparation. For the slurry preparation and Hg determination, 5.0-mL aliquot of the slurry was transferred to the reaction flask of the hydride generator. 1.0 mL of 7 mol L⁻¹ HCL and 0.25 mL of isoamyl alcohol were added, and deionized water was added until the total volume reached 10 mL. The sodium tetrahydroborate solution was added to the reaction flask over a period of 10 s, and the vapor that was generated was carried into the quartz T tube, which was coupled to the AAS spectrometer.

2.3.2. Digestion of soil and galamsey waste samples

In order to determine the total concentrations of mercury in the soil and galamsey waste, the samples were dried using a freeze-drier (-50-60 °C) for 3–4 days and homogenized by thorough mixing. The homogenized dried samples for both soil and galamsey waste were sieved through a 0.2 mm sieve and were digested using EPA method 6010 (Roy-Keith, 1998). Triplicate sample each of soil and galamsey waste totaling 80 samples were digested separately and replicate results for each sample were obtained by repeating the same process on two different days. The final suspended mixture was filtered through a 0.45 μm membrane filter. The same procedure was performed with a blank

and a standard reference material (SRM 2711 Montana Soil II, a moderately contaminated soil) in each batch of digestion. The concentrations of total mercury in the leachate were determined using atomic absorption spectrometer (AAS).

2.3.3. Sample analysis

Sample volumes of 20 μL were injected into the GTA 3000 graphite furnace. The resultant signal of atomic absorption was determined as a peak height mode against an analytical curve. Then, the temperature program was run and integrated atomic absorbance was measured under the conditions shown in Table 2 for Hg determination. Background-corrected integrated absorbance was used as the analytical signal. The light source was an electrodeless discharge lamp (EDL) for the Hg operating at their respective lamp current with their respective spectral bandwidths to isolate the individual resonance lines (Table 2).

2.3.4. Accuracy of analytical method and validation

Mercury calibration solutions at concentrations from 0.03 to 25.00 $\mu\text{g L}^{-1}$ were prepared daily by the serial dilution of a stock solution (1000 mg/L Hg, Merck, Darmstadt, Germany) with a 0.05% (v/v) HNO_3 solution. The reductant was a 1% (w/v) sodium tetrahydroborate solution that was stabilized with 0.05% (w/v) sodium hydroxide which was prepared as in Silva et al. (2012) using analytical grade reagents from Merck, Darmstadt, Germany and was filtered through a 0.45- μm filtration membrane. The 1.0% (w/v) thiourea solution was prepared by dilution of the reagent with high-purity water. The certified reference material used in the accuracy evaluation was CRM 580 (estuarine sediment) from Joint Research Center in Belgium.

To determine the matrix effect and to choose proper calibration technique, the analyte addition technique was employed as in Silva et al. (2012). Mercury concentrations of 1.0, 1.5, 2.0, 2.5, 3.0, 3.5 and 4.0 $\mu\text{g L}^{-1}$ were added to the samples. The slope of the analytical curve (expressed at the 95% confidence level) was (0.0826 ± 0.0024) . A calibration curve was determined using aqueous standards at the same concentrations; the slope was (0.0873 ± 0.0029) . These results show that the external calibration technique can be used with this method to determine the mercury content in samples.

The accuracy of the method was confirmed by analyzing the certified reference material, CRM 580, which was provided by Joint Research Center in Belgium. The certified mercury concentration was $(0.005 \pm 0.0003 \text{ mg/kg})$, and the concentration determined by this method was $(0.0060 \pm 0.0004 \text{ mg/kg})$ (Table 3).

Moreover, the known concentration of Hg solution is measured periodically with the measurement of sample solution. Recalibration was done when the measured value of the standard Hg solution shown deviation more than ten percent (10 %) from its known concentration per standard procedures prescribed by Assignment Point (2013). Hence, the accuracy and precision of the analytical data were strictly followed throughout the present study and this method has limit of detection and precision comparable with other procedures as proposed in Su et al. (2008); Collasiol et al., (2004); Flores et al., (2001).

The detection limit of the Hg for the instrument and the spike recovery rates of Hg analyzed under the experimental conditions were determined (Table 4).

2.4. Comparison of findings to established threshold and statistical analysis

The median and data ranges obtained for surface drainages were compared with the Ghana EPA Effluent Discharge Guideline (Ghana EPA, 1999) whilst the earth samples (slurry/sludge, soils and wastes) were compared to the New Dutch List (The New Dutch List, 2000) to establish exceedances (if any).

Following the determination that the data was not normally distributed by both the Kolmogorov-Smirnov and Shapiro-Wilk tests, the data generated was statistically assessed and the mean values ranked using the Kruskal-Wallis Test at a confidence level of 95% (McDonald, 2014; D'Agostino and Stephens, 1986; Shapiro and Wilk, 1965). Differences in mercury levels due to galamsey types under the different environmental media (soil, waste, surface drainage and sludge/slurry) was evaluated.

3. Results

3.1. Concentrations of mercury recorded for each galamsey type across the four environmental media

3.1.1. Slurry/sludge

From Table 5, the mercury concentrations within sampled slurry/sludge substrates from the nine (9) galamsey sites generally showed extensive differences. Specifically, the median of mercury associated with Anwona was recorded as 59.00 mg/kg (IQR = 45.80–68.20 mg/kg) and found to be above the median of the reference/control sample recorded as 0.0004 mg/kg (0.0004–0.065 mg/kg). In addition, the median recorded for Chamfi 81.40 mg/kg (IQR = 73.70–87.10 mg/kg), Washing Board 47.15 mg/kg (IQR = 36.100 to 55.83) Washing Plant 22.30 mg/kg (IQR = 20.47–23.97 mg/kg) and Mill House 218.76 mg/kg (IQR = 210.250–225.490 mg/kg) was above the median of the reference sample. Additionally, the median of mercury recorded for Dig and Wash was 2.00 mg/kg (IQR = 1.86–2.10 mg/kg), River Dredging 1.35 mg/kg (IQR = 1.20–1.50 mg/kg) and Underground Abandoned Shaft 0.1125 (IQR = 0.07–0.19 mg/kg) was above the median of the Reference/Control sample.

The median value obtained for the following galamsey operations exceeded the Action value of the New Dutch List (The New Dutch List, 2000). Mill House, Chamfi, Anwona, Washing Board, Washing Plant, and River Dredging (in descending order), with the highest value being 210.25 from a Mill House operation. The two underground operations and the reference/control site recorded no exceedances.

3.1.2. Background soils

As could be inferred from Table 6, the median value for Chamfi 2.45 mg/kg (IQR = 1.90–2.70 mg/kg), Dig and Wash 2.00 (IQR = 1.70 to 2.20) and Washing Board 2.65 mg/kg (IQR = 2.50–2.90 mg/kg) within soil medium are above the median of the reference sample 0.0004 mg/kg (IQR = 0.0004 to 0.0004 mg/kg). In addition, the median recorded for Washing Plant 1.80 (IQR = 1.60 to 1.80) and Mill House 195.26 mg/kg (IQR = 155.22–215.24 mg/kg) are above the median of the reference sample. The median recorded for Underground Sample Pit 0.0004 mg/kg (IQR = 0.0004–0.01 mg/kg) was same as the median of the reference sample. Additionally, the median recorded for Underground Abandoned

Table 2. Temperature program for Hg determination.

Step	Temperature ($^{\circ}\text{C}$)	Ramp Time (sec)	Hold Time (sec)
Injection	20	–	–
Drying	90	30	15
Pyrolysis	200	30	40
Atomization	1100	1500	10
Cleaning	1700	200	4
Cooling	40	25	5

Table 3. Accuracy of the analytical data with reference to the CRM 580 standard.

Name of the metal	Hg
Certified Value (mg/kg)	0.005 ± 0.0003
Measured Value (mg/kg) ± SD	0.0060 ± 0.0004

SD – Standard deviation.

Table 4. Detection limit (DL) and Spikes recovery of Hg.

Metals	Hg
DL (ppm)	0.001
Recovery (%)	90–99

Table 5. Distribution (lower quartile, median and upper quartile) of contaminants in slurry/sludge samples for each galamsey type.

Contaminants	Galamsey Types	Lower Quartile	Median	Upper Quartile	New Dutch Lists' Action & Intervention Values	
					Action	Intervention
Mercury (mg/kg)	Reference/Control	0.00	0.00	0.07	0.3	10
	Anwona	45.80	59.00	68.20	0.3	10
	Chamfi	73.70	81.40	87.10	0.3	10
	Dig and Wash	1.86	2.00	2.10	0.3	10
	Washing Board	36.10	47.15	55.83	0.3	10
	Underground Abandoned Shaft	0.07	0.11	0.19	0.3	10
	Washing Plant	20.47	22.30	23.97	0.3	10
	Underground Sample Pit	0.01	0.02	0.03	0.3	10
	River Dredging	1.20	1.35	1.50	0.3	10
	Mill House	210.25	218.76	225.49	0.3	10

Note: Galamsey types with values less than the detection limit were excluded.

Shaft 0.0502 mg/kg (IQR = 0.0004–0.11 mg/kg) and River Dredging 0.40 mg/kg (IQR = 0.30–0.50 mg/kg) was above the median of the reference sample.

Also, the median value obtained for the following galamsey operations exceeded the Action value of the New Dutch List (The New Dutch List, 2000): Mill House, Washing Board, Chamfi, Dig and Wash, Washing Plant and River Dredging (in descending order) with the highest value being 195.24 from a Mill House operation. The two underground operations, Anwona and the reference/control site recorded no exceedances.

3.1.3. Galamsey waste

From Table 7, the median value for Anwona 2.15 mg/kg (IQR = 1.60–2.60 mg/kg), Chamfi 1.80 (IQR = 1.40–2.20 mg/kg), Dig and Wash 1.75 (IQR = 1.30–2.10 mg/kg) and Washing Board 1.70 mg/kg (IQR = 1.30–1.80 mg/kg) within waste medium are above the New Dutch Lists'

Value Action Value of 0.3 mg/kg (The New Dutch List, 2000). In addition, the median recorded for Washing Plant is 1.35 mg/kg (IQR = 1.20–1.70 mg/kg) whilst the concentrations recorded for the two Underground operations (Sample Pit and abandoned shaft) were negligible and showed no exceedance. The Mill House galamsey recorded the highest concentration of mercury with a median and interquartile range of 206.44 mg/kg (IQR = 198.45–209.25 mg/kg) whereas the River Dredging galamsey recorded a median value of 0.30 mg/kg (IQR = 0.30–0.40 mg/kg).

In addition, the median value obtained for the following galamsey operations exceeded the Action value of the New Dutch List (The New Dutch List, 2000): Mill House, Anwona, Chamfi, Dig and Wash, Washing Board, Washing Plant and Dredging (in descending order) with the highest value being 206.44 from a Mill House operation. The two underground operations and the reference/control site recorded no

exceedances.

3.1.4. Surface drainage/water

As could be seen from Table 8, the mercury profiles within surface drainage samples for the different types of galamsey showed extensive differences. Specifically, the median and IQR of mercury associated with the Anwona and Washing Plant galamsey types 12.400 mg/L (IQR = 10.3–15.2 mg/L) and 18.450 mg/L (IQR = 17.90–19300 mg/L) were above the median of the reference sample 0.000 mg/L (IQR = 0.00040–0.0007 mg/L) and the IQR. The Mill House (209.12 mg/L, 149.51 mg/L to 215.25 mg/L), Chamfi (82.20 mg/L, 76.50 mg/L to 88900 mg/L) and Washing Board (48.6 mg/L, 39.8 mg/L to 55.4 mg/L), being the three galamsey types with the highest median and IQR values of mercury in that order, were far above the median of the Reference Sample and the median range. Additionally, the median recorded for Dig

Table 6. Distribution (lower quartile, median and upper quartile) of contaminants in soil samples for each of the nine Galamsey types.

Contaminants	Galamsey Types	Lower Quartile	Median	Upper Quartile	New Dutch Lists' Value Action & Intervention Values	
					Action	Intervention
Mercury (mg/kg)	Reference/Control	0.00	0.00	0.00	0.3	10
	Chamfi	1.90	2.45	2.7	0.3	10
	Dig and Wash	1.70	2.00	2.20	0.3	10
	Washing Board	2.50	2.65	2.90	0.3	10
	Underground Abandoned Shaft	0.00	0.05	0.11	0.3	10
	Washing Plant	1.60	1.80	1.80	0.3	10
	Underground Sample Pit	0.00	0.0	0.01	0.3	10
	River Dredging	0.30	0.40	0.50	0.3	10
	Mill House	155.22	195.24	215.24	0.3	10

Note: Galamsey types with values less than the detection limit were excluded.

Table 7. Distribution (lower quartile, median and upper quartile) of contaminants in waste samples for each of the nine Galamsey type.

Contaminants	Galamsey Types	Lower Quartile	Median	Upper Quartile	New Dutch Lists' Value Action & Intervention Values	
					Action	Intervention
Mercury (mg/kg)	Anwona	1.60	2.15	2.60	0.3	10
	Chamfi	1.40	1.80	2.20	0.3	10
	Dig and Wash	1.30	1.75	2.10	0.3	10
	Washing Board	1.30	1.70	1.80	0.3	10
	Washing Plant	1.20	1.35	1.70	0.3	10
	Underground Sample Pit	0.00	0.00	0.00	0.3	10
	River Dredging	0.30	0.30	0.40	0.3	10
	Mill House	198.45	206.44	209.25	0.3	10

Note: Galamsey types with values less than the detection limit were excluded.

and Wash and River Dredging (0.780 mg/L, IQR = 0.550–0.911 mg/L) and 0.0055 mg/L, IQR = 0.0046–0.006 mg/L), although being the least among the galamsey operations) are respectively above the median and IQR values recorded for the reference sample.

In addition, the median value obtained for the following galamsey operations exceeded the Action value of the New Dutch List (*The New Dutch List, 2000*): Mill House, Chamfi, Washing Board, Washing Plant and Anwona (in descending order) with the highest value being 209.12 mg/L from a Mill House operation. The two underground operations, Dredging, Anwona and the reference/control site recorded no exceedances.

3.1.5. General profile of mercury across the nine galamsey operations in all four environmental media

The highest median value recorded for mercury across all nine galamsey types and four environmental media (soil, surface drainage, slurry/sludge and wastes) is 218.76 mg/kg and was within slurry samples at a Mill House operational site. For all water samples analyzed across the nine galamsey types, the Mill House again recorded the highest median value of 209.13 mg/L, followed by 82.20 mg/L from Chamfi and 48.55 mg/L from Alluvial Washing Board; with River Dredging (0.0055 mg/L) recording the least. For soil medium, the Mill House (195.24 mg/kg), Washing Board (2.65 mg/kg) and Chamfi (2.45 mg/kg) recorded the three highest median values. Similarly, slurry samples recorded 218.76 mg/kg (for Mill House), 81.40 mg/kg (Chamfi) and 59.00 mg/kg (Anwona) as the three very high median values. The waste media recorded 206.44 mg/kg for Mill House, 2.15 mg/kg for Anwona and 1.80 mg/kg for Chamfi galamsey types.

Operationally, the Anwona galamsey had its highest median value (59 mg/kg) within a slurry sample whilst the least (2.15 mg/kg) is from waste samples. The Chamfi had its highest median (82.2 mg/L) from surface drainage and least (1.8 mg/kg) from wastes. Washing Board and Washing Plant respectively had their highest median (48.55 mg/L and 18.45 mg/L) from surface drainages whilst their least value (1.70 mg/kg and 1.35 mg/kg) were recorded from waste samples. Dig and Wash, River

Dredging, Underground Sample Pit and Abandoned Shaft galamsey types recorded very low median values within the range of 0.0004 mg/kg to 2.00 mg/kg and are slightly elevated than the median recorded for reference or control (non-galamsey areas) sampled.

Also presented in *Figure 4* is a box-plot comparison of the distribution of mercury for each type of galamsey activity and from a reference/control location in all four media aggregated.

3.2. Comparison of mean ranked concentration of mercury within each galamsey type and environmental media

Generally, the ranking of galamsey types (*Table 9*) in respect of their contribution to mercury shows that the Mill House contributed the highest mercury to soil, surface drainage and slurry media, with a mean ranking of 21.5. Chamfi followed suit with a mean ranking of 19.5 in both slurry and in surface drainage. Washing Board in soil was the third highest contributor (18.8) of mercury followed by Mill House in waste (17.5) and Washing Board in surface drainage (17.5). The reference (ungalamseyed) sites unsurprisingly recorded the least mercury levels in slurry/sludge samples, but was interestingly ranked higher than Anwona in soil media and the two underground operations in surface drainage/water medium.

For individual environmental media, Mill House contributed the highest mercury to slurry/sludge. Next to this are Chamfi, Anwona, Washing Board, Washing Plant, Dig and Wash, River Dredging, Underground Abandoned Shaft and Underground Sample Pit in the descending order. The reference or control (“non-galamseyed” areas) was the least contributor of mercury to slurry/sludge. The observed differences in the ranking was significant (p = 0.015). For soils, the Mill House again contributed the highest mercury, with Washing Board, Chamfi, Dig and Wash, Washing plant, River Dredging, Underground abandoned shaft and Underground Sample Pit galamsey types following suit in descending order. Although very low in ranking, the reference/control (“non-galamseyed” areas) ranked better, in terms of contributions to mercury

Table 8. Distribution (lower quartile, median and upper quartile) of contaminants in surface drainages for each of the nine Galamsey type.

Contaminants	Galamsey Types	Lower Quartile	Median	Upper Quartile	Ghana EPA Effluent/Water Discharge Guide
Mercury (mg/L)	Reference/Control	0.00	0.00	0.00	0.005
	Anwona	11.40	12.40	14.10	0.005
	Chamfi	77.50	82.20	87.50	0.005
	Dig and Wash	0.55	0.78	0.91	0.005
	Washing Board	41.10	48.55	54.20	0.005
	Washing Plant	17.90	18.45	18.90	0.005
	River Dredging	0.00	0.01	0.01	0.005
	Mill House	172.62	209.12	211.21	0.005

Note: Galamsey types with values less than the detection limit were excluded.

contamination, than the Anwona Galamsey. The observed differences in the ranking was significant (with $p = 0.019$).

In respect to the waste medium, the Mill House galamsey type once again contributed the highest mercury concentrations. This was followed by Anwona, Chamfi, Dig and Wash, Washing Board, Washing plant, River dredging, Underground Sample Pit and Underground Abandoned Shaft in that order. The observed differences in the ranking was not significant ($p = 0.063$). The Mill House galamsey type contributed the highest concentration of mercury to surface drainage media. Next to this are, Chamfi, Washing Board, Washing Plant, Anwona, Dig and Wash and River Dredging in the descending order. Surface drainage samples from the non-galamseyed areas interestingly recorded a higher mercury contribution than the two underground types of galamsey. The observed differences in the ranking was significant ($p = 0.014$).

3.3. Discussion of results

3.3.1. Mercury trend across the nine galamsey types

Characteristically, the usage intensity and quantity of mercury in galamsey is dependent on the operational scale, commerciality and level of mechanization involved (Mantey et al., 2016). Hence, the different levels of exceedances recorded by Mill House, Chamfi, Anwona, Washing Board, Washing Plant, Dig and Wash and Dredging, with the highest median value of mercury (across all nine galamsey types and four environmental media) being 218.76 mg/kg and was within slurry samples at a Mill House operational site. The Mill House, Chamfi, Anwona, Washing Board and Washing Plant galamsey types unsurprisingly recorded the highest mean rankings in terms of their contribution to mercury contamination to the environment due to their wide operational footprints (scale and scope), commerciality and operational intensities.

According to Wuana and Okieimen (2011), the specific type of metal contamination found in a contaminated media is directly related to the operation that occurred at the site. The very high mercury levels recorded and mean ranking (21.5) across soil, surface drainage and slurry/sludge media by the Mill House galamsey operation is therefore a testament of the intensity of the gold processing activities and usage of mercury. Chamfi followed this in both slurry/sludge and in surface drainage, and Washing Board in soil medium.

The Mill House is seen as a concentration camp for processing high-grade ore mined from underground ghettos or other sources. It is a commercially oriented type of galamsey, which typically involves ore milling or processing. Mill House structures are normally erected along roadsides and positioned adjacent to water sources or wetlands for receiving ore for processing and extraction of gold. This type of galamsey is the preferred choice for processing and extracting of gold from high-grade ore normally mined from Underground Sample Hole, Selection or Abandoned Underground Shafts galamsey sites. The use of mercury is very predominant and could in the raw state or residual form find its way into surrounding soils either directly or indirectly through depositions from amalgamation and smelting activities. Received rocks or sandy materials (ore) are either crushed, smoothed or sluiced intensively using mercury. The processed wastes from Mill House operation are popularly called “shump” (tailings) and tend to be pretty high in gold grade due to the relatively low recovery methods employed. These processing wastes are either reprocessed by the galamseyers themselves (more especially during the rainy seasons where mining is seen as challenge) or sold out to some interested LSM companies for reprocessing or retreating (Mantey et al., 2016).

Also, the Chamfi, Alluvial Washing Plant, Alluvial Washing Board, Anwona and Stream Dredging operations are highly commercial businesses with very wide footprints and make extensive use of mercury in

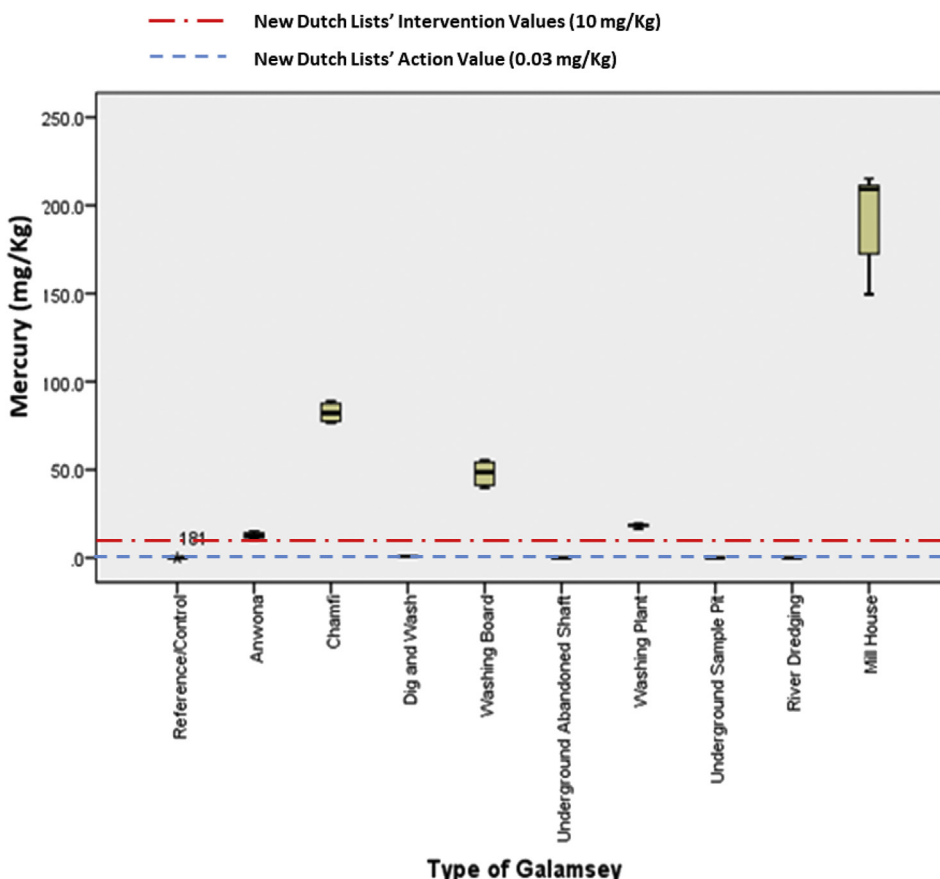


Figure 4. A box-plot comparison of the distribution of mercury for each type of galamsey activity and from a reference/control location in all four media aggregated.

Table 9. Comparison of Mean ranked concentration of mercury for each medium type and across the different Galamsey types.

Sample type		Galamsey Type	Mean Rank	P value			
Slurry	Mercury (mg/kg)	Mill house	21.5	0.015			
		Chamfi	19.5				
		Anwona	17				
		Washing board	16				
		Washing plant	13.5				
		Dig and Wash	11.5				
		River dredging	9.5				
		Underground abandoned shaft	7.5				
		Underground sample pit	3.7				
		Reverence/control	3.3				
		Soil	Mercury (mg/kg)		Mill house	21.5	0.019
Washing board	18.8						
Chamfi	16.3						
Dig and Wash	14						
Washing plant	13.5						
River dredging	10.5						
Underground abandoned shaft	6.3						
Underground sample pit	5.3						
Reverence/control	5						
Anwona	3.5						
Waste	Mercury (mg/kg)			Mill house	17.5	0.063	
		Anwona	13.8				
		Chamfi	12.3				
		Dig and Wash	11.3				
		Washing board	11				
		Washing plant	9.3				
		River dredging	5.5				
		Underground sample pit	3				
		Underground abandoned shaft	2				
		Water	Mercury (mg/l)	Mill house	21.5		0.014
				Chamfi	19.5		
Washing board	17.5						
Washing plant	15.5						
Anwona	13.5						
Dig and Wash	11.5						
River dredging	9.5						
Reference/control	5						
Underground abandoned shaft	4						
Underground sample pit	4						

their gold extraction (Mantey et al., 2016). They involve simultaneous mining and gold extraction, and are mechanically inclined with very significant footprints. These operations make extensive use of mercury in their operations and could result in the deposition or contamination of soils from poor handling of mercury and its waste materials.

Any of the four stages of the small-scale gold production process, namely amalgamation, separation of amalgamation, removal of excess mercury, and burning of the remaining amalgam, release mercury into the environment. Due to mercury's intrinsic properties, it readily forms alloys with other metals such as gold called amalgams, which facilitate the recovery of gold from its ores. To recover the gold, the amalgam is heated to evaporate the mercury, leaving the gold behind to recover it from the 50% mercury and 50% gold amalgam. The vaporized mercury then condenses back into its solid form, falling to earth and washing into streams.

What appears a bit surprising is the relatively high value of mercury recorded or ranking position chalked by Dig and Wash galamsey, as this type of galamsey is mostly known to be poverty driven, rudimentary and subsistence and make very limited or no use of mercury in its operations. The presence of mercury in the base soil perhaps signals the usage of

mercury in the operation of Dig and Wash. Concerning Dredging galamsey, the presence of mercury in the storage areas and on the banks of the host rivers or water bodies is a possibility due to the involvement of mercury in the gold operation and high intensity of mining and processing involved in this galamsey business.

The underground operations, which involves only mining of ore (gold bearing rock or soil), make limited or no use of mercury since they involve no gold extraction or processing. Ore mined are transported to processing centers or the Mill House areas for gold extraction, hence very high mercury levels recorded in the processing centers (Mantey et al., 2016).

The control/reference (natural and non-galamseyed) site unsurprisingly recorded the least mercury levels in slurry samples, but were interestingly ranked higher than Anwona in soil media and the two underground operations in surface drainage media. This occurrence might be due to other phenomenon such as atmospheric deposition which is very dominant sources of mercury over most landscapes. Once in the atmosphere, mercury is widely disseminated and can circulate for years (Hilson, 2003; Boening, 2000; Lodenius and Malm, 1998), accounting for its widespread distribution within the non-galamseyed areas like

Samahu, Moseaso, HuniAno and Esaman Kakraba where samples were obtained.

3.3.2. Mercury trend across environmental media

Environmental medium, which generally include soil, water, air, biota, slurry, sludge or any other parts of the environment, entails a material found in the physical environment that surrounds or contacts organisms through which chemicals, pollutants can move and reach the organisms, or that can contain contaminants (ATSDR, 2016; USEPA, 1996; USEPA, 1994). Once pollutants are released into a media, they are subject to a range of transport and transformation processes, including dispersal through bulk transport either in the surroundings or by diffusion or particles, down the concentration gradient. As contaminants migrate, they are diluted by mixing within the transport medium and transformed by both chemical reactions and physical processes such as abrasion, and selectively removed by gravitational settling, scavenging and filtration. Due to this, levels tend to decline with both distance and duration of transport (Duraes et al., 2018; Zayed and Paleologos, 2018).

The consequences in terms of pollution concentrations, depends heavily on the nature of the environmental medium and of the pollutants concerned. Characteristically, how far and how quickly pollutants spread is governed by the rate of movement of the medium in which they occur, either by water velocity, windspeed or mechanical pumping. Many chemical processes are also temperature (and often moisture) and local micro-environment dependent, so, rates of transformation are affected by climate and weather. The needed information on the fate of a pollutant as it runs through the environment can be obtained from direct measurement such as monitoring pollutant concentrations at a sample of locations.

From Tables 5 and 9, the highest median value recorded for mercury across all nine galamsey types and four environmental media were 218.76 mg/kg and 209.12 mg/kg for slurry/sludge and surface water/drainage samples respectively at a Mill House operational site. Statistically, the ranking of galamsey types in respect of their contribution to mercury shows that the Mill House contributed the highest mercury to soil, slurry/sludge, surface drainage and soil media, with a mean ranking of 21.5. The crushing, smoothening and sluicing processes implemented at the Mill House galamsey operation, results in the generation of tailings or slurry/sludge wastes that consist of a variety of solids and liquids with significant quantities of mercury and other contaminants. Galamsey is water-intensive activity, and is therefore not surprising to record very high mercury levels in surface water/drainage medium across the various galamsey types.

Slurry is a semi-liquid mixture, typically of fine particles of manure, cement, or coal suspended in water. A sludge is on the other hand a semi-solid slurry that can be produced from a range of industrial processes, from mining and mineral processing, water treatment, wastewater treatment or on-site sanitation systems. The difference between sludge and slurry is that sludge is a generic term for solids separated from suspension in a liquid while slurry is any flowable suspension of small particles in liquid. Slurry and sludge are characterized as being soft, wet, and thick. They may consist of a wide range of harmful substances such as dioxins and furans, polychlorinated biphenyls, organochlorine pesticides, absorbed and extracted chlorine derivatives, polycyclic aromatic hydrocarbons, phenols and their derivatives, phthalate, heavy metals, concentrated organic matter, nitrogen, inorganic salts, bacteria, O/G (Demirbas et al., 2017; Woodruff and Macnamara, 2013).

Soils are the major sink for heavy metals released into the environment by mining activities (Kirpichtchikova et al., 2006; Adriano, 2003) and was therefore unsurprising to record a high mean rating from the commercial galamsey types into soils. Indeed, mercury pollution from gold processing and land degradation has been lauded as the principal environmental problems caused by small-scale mining activity (Donkor et al., 2006a,b; Ntengwe and Maseka, 2006). Mercury, the only liquid metal, has been used in the mining industry to amalgamate and concentrate precious metals since the Phoenicians and Carthaginians

applied it around 2700 B.C. (Eisler, 2003). Today, small-scale gold miners in more than 50 developing countries (Veiga et al., 2006) use it. Therefore, it is not surprising that its use is common in Ghana, and that exceedances in six out of the nine galamsey types/operations analyzed is observed and confirmed by the results of this study.

The soil and rock, which is removed to gain access to buried ore, and the material left behind after the ore has been processed to remove the valuable commodities, are considered to be waste materials. Mine waste is a general term for material, which currently has little or no economic value (Pramoda, 2017). From the above tables, the levels of mercury recorded in galamsey wastes were generally found to be the least, with the Mill House recording the highest median value of 206.44 mg/kg. Generally, the type, amount and properties of mine waste produced at different mines vary depending on the resource being mined, process technology used, and geology at the mine site (Ziebarth and Wood, 2014). The wastes obtained from Mill House are processing wastes ("shump" or tailings), whilst those from the underground operations are mining wastes (over burden). On the other hand, those from operations comprising the simultaneous mining of ore and gold extraction/processing, are a mixture of both mining and processing wastes.

The results obtained by Bonzongo et al. (2003) for their work on Hg contamination in selected mine-impacted Ghanaian watersheds suggested a major environmental problem with Hg in Ghana, with Total-Hg concentrations in hundreds of mg/kg for both soils and sediments. The generally high levels of Hg recorded across the various galamsey operations is a direct manifestation of the heavy reliance of galamsey operators on mercury to overcome gold extraction difficulties (Rambaud et al., 2000) and so these levels may persist for a long time in those environments. According to GEF, UNDP, UNITAR & EPA (2018), Ghanaian minerals, rocks and fossils do not contain naturally occurring Mercury (Cinnabar or Mercury Sulphide - HgS) and could as such be concluded that the amount of mercury recorded in this research are purely from anthropogenic galamsey activities. Hg in the various oxidation states is known to be released into both terrestrial and aquatic systems (Hilson, 2003) and disperses very effectively through the atmosphere with long residence times of about two years, and it is normally transported from likely sources of emission (Hilson, 2003; Boening, 2000; Lodenius and Malm, 1998). According to Veiga and Baker (2004), mercury losses occur at various stages during gold production: (1) during amalgamation, where mercury may be washed out during the gravity washing; and (2) during burning, where mercury, with its high volatility, is released into the atmosphere. After burning, a sponge-like gold ore' stays behind in the can. When the gold has cooled, it is weighed and ultimately sold. Due to impurities and trapped mercury, the gold often undergoes a refining process off site that involves additional heating steps and the use of acid, borax, and soda ash. It is estimated that one or two grams of mercury is lost for every gram of gold produced in ASM (Veiga and Baker, 2004).

4. Conclusion and recommendations

4.1. Conclusion

The concentrations of mercury have been successfully measured across the nine (9) types of galamsey. In addition, the extent to which nine (9) types of galamsey influences mercury levels within surface drainages, soils, slurry/sludge and solid wastes in three hotspot assemblies of the Western Region of Ghana have been assessed.

Exceedances were recorded by the Mill House, Chamfi, Anwona, Washing Board, Washing Plant, Dig and Wash and Dredging galamsey types, with the highest median value of mercury respectively being 218.76 mg/kg and 209.12 mg/kg for slurry/sludge and surface water/drainage samples at a Mill House operational site. In terms of their contribution to mercury contamination, the Mill House, Chamfi, Anwona, Washing Board and Washing Plant galamsey types recorded the highest mean rankings due to their wide operational footprints (scale and scope), commerciality and operational intensities.

Indeed, differences exist in mercury levels due to galamsey types under different environmental media. The extent or degree to which the nine types of galamsey influences pollution levels within surface drainages, soils, slurry and solid wastes is now known.

4.2. Recommendations

1. Considering the levels of mercury recorded by the nine galamsey types across the four environmental media, the Government of Ghana to should consider the ban of the Mill House, Anwona, Washing Board, Washing Plant, Dredging and Chamfi galamsey types.
2. The galamsey operations categorized as “processing only and simultaneous mining/processing types” involve the use of mercury in substantial quantities. The underground galamsey, which involves mining type, makes no use mercury and have very limited impacts on the environment. Therefore, a decision should be made to construct and centralized a processing plant and an accompanying tailings storage facility (TSF), for lode/vein galamsey operators to use. Thus, the plant being in the shape of the Mill House operation, would serve all galamsey operators in a particular district or assembly.
3. Clean ups should be observed in affected galamsey areas for public health protection and general environmental protection.

Declarations

Author contribution statement

Jones Mantey: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

Kwabena Nyarko Biritwum, Frederick Owusu-Nimo: Conceived and designed the experiments; Analyzed and interpreted the data.

Richard Kwasi Amankwah, Crentsil Kofi Bempah, Wisdom Eli Akatu, Eugene Appiah-Effah: Contributed reagents, materials, analysis tools or data; Wrote the paper.

Adolf Kofi Awua: Conceived and designed the experiments; Analyzed and interpreted the data; Wrote the paper.

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The authors declare no conflict of interest.

Additional information

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