

Mercury Pollution Studies of Some Rivers Draining the Bibiani-Anwiaso-Bekwai Mining Community of South Western Ghana

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Abstract: The project assessed the extent of mercury pollution of some rivers that drain the Bibiani-Anwiaso-Bekwai district which is a typical mining community in the south western part of Ghana. In the study, surface-water and sediment samples were collected from seven streams that drain this mining community and analyzed for total mercury, organic mercury and elemental mercury. Mercury concentrations of non-filtered water was determined using the ICP-OES after reduction with stannous chloride (SnCl₂). Physico-chemical parameters were also determined for the water samples. Sediment samples were pretreated and their mercury content determined using the same instrumental technique as for the water. Total mercury content of the water ranged between 0.125 to 1.341 µg/L while sediment values ranged between 0.169 to 1.739 mg/kg. In all cases except for one site, mercury levels in the sediment have been found to be significantly higher than the corresponding water column. Except for site SW7 (1.341 µg/L), total mercury in water has also been found to be lower than 1.0 µg/L, the WHO guideline value for drinking water. On the contrary, except for site ASUS (169 mg/kg), all sampled sediments recorded values that were above 0.2 mg/kg, the US-EPA guideline value for soils. Since sediments serve as sink for mercury and release the metal into water column with time, it can be concluded that these streams are polluted with mercury.

Key words: Elemental mercury, gold mining, organic mercury, surface-water, sediments, total mercury

INTRODUCTION

It is now generally recognized that the various chemical forms (or species) of an element in natural waters can behave differently, thereby controlling its biogeochemical behaviour and toxicity to organisms. Therefore chemical speciation information is particularly important from a geochemical standpoint for those elements, like Hg, whose solution forms may include species resulting from thermodynamic equilibrium due to biological interactions. For example, non-equilibrium, methylated species of arsenic, selenium, and sulphur play a significant role in their cycling in natural waters (Andreae, 1986; Cooke and Bruland, 1987).

Substantial field evidence suggests that Hg in many natural water systems exists in organic forms, although the nature of these forms has often not been clearly identified (Mantoura *et al.*, 1978; Davies *et al.*, 1979; Minagawa *et al.*, 1980; Goulden and Anthony, 1980; Olafsson, 1983; Amdurer *et al.*, 1983). Two general classes of organic-Hg associations can occur in natural waters and these are highly toxic organomercurials

(compounds where Hg is covalently bonded to carbon; e.g., methylmercury and dimethylmercury) and complexes of Hg²⁺ with natural organic ligands. Considerable evidence suggests that organomercurials are produced biotically, although abiotic methylation processes have been reported (Akagi *et al.*, 1977; Hayashi *et al.*, 1977; Nagase *et al.*, 1982).

Mercury is readily absorbed into/onto sediments and slowly released to the water forming a reservoir capable of causing chronic pollution long after the original source of mercury has been removed. The concentration of mercury in bottom sediments is therefore a very good indicator of water polluted with this element. On the one hand, bottom sediments are where mercury accumulates as a result of simple sedimentation, and released from the sediments becoming available for further biogeochemical transformations. The rates of these two processes depend significantly on the specific environmental conditions in a given water system (Boszke *et al.*, 2003).

The gold mining sector is an important geological and anthropogenic source of mercury emission (Kim *et al.*, 2005). The use of mercury amalgamation

technology in gold mining has been known since ancient times and was first used in Spain as early as 700 BC (Lacerda and Salomons, 1998). Due to lack of innovations in prehistoric times, the amalgamation technique was a generally accepted procedure for gold extraction (Nriagu, 1994; Nriagu and Wong, 1997). The methods for gold extraction have varied during the years, starting with small hand operations such as gravity panning and diverse sluicing techniques to the presently used amalgamation and cyanide leaching technique (McDonald, 1983). As a result of the inexpensiveness of the amalgamation technique and efficiency of gold recovery, it is the method of choice for current small scale gold mining nations in the developing world. The use of this technique, despite the socio-economic importance of gold recovery, faces strong opposition by the international community. This is as a result of the toxicity of alkyl mercury, specifically, methyl mercury (MeHg) which bioaccumulates and results in deleterious effects on humans and ecosystem functions (Nriagu, 1994; Nriagu and Wong, 1997).

Ghana is made up of five geological provinces. The south-eastern province which includes the Dahomeyan system; Togo series and the Buem formation, the western province within which are the Birimian with in-folded Tarkwaian rocks; the central province which includes mainly the Voltaian System; the coastal basins which include the Sekondian Series, the Accraian Series, the Amissain Formation and the Apollonian Formation; and tertiary to recent deposits. The country's major minerals are gold, diamond, bauxite and manganese which are mainly associated with Birimian and Tarkwaian rocks (Kesse, 1985; Grubaugh, 2002). Gold is the predominant mineral produced in Ghana. Records indicate that alluvial gold mining was practised as early as the 4th century and indigenous population of Ghanaians got more involved during the European times (Akabzaa and Darimani, 2001).

There are both large-scale and small-scale gold mining in Ghana (Nartey *et al.*, 2004). The general processing techniques involve handpicking, amalgamation, cyanidation, floatation, electrowinning and roasting. The techniques differ between large and small scale mining and also vary depending on the type of deposit and its location (Akosa *et al.*, 2002; Ntibery *et al.*, 2003).

Gold mining operations began in the Bibiani area in 1902 making it an area with a long history of mining operations with its subsequent danger of possible mercury pollution of the environment.

MATERIALS AND METHODS

Sampling area: The study was carried out at Bibiani-Anwiaso-Bekwai district, a gold mining community in the Western Region of Ghana during the dry season of

October 2008 to March 2009. This area is known to be one of the areas where gold mining is usually done using mercury amalgamation. A map of the sampling sites and the surrounding is shown in Fig. 1.

Sample site description: With reference to Table 1, sample code, SW3 is surface water that runs through the mining area. This river is not very large and therefore not of much domestic and economic importance in the area. Nonetheless it contains fishes which may be caught and eaten by members of the community. Kyereyar river which is coded SW7, Table 1 is a large stream which runs through the large scale mine site and serves as a source of drinking water for "pipeline" village. As indicated in Table 1, SW10 is Pamonu River, this is a stream that serves as source of drinking water for Bibiani township. Lake Amponsah, which is coded SW14, Table 1 is also a large collection of water that is close to galamsey sites. As a result, water from pits due to galamsey activities discharge into this lake. During the rainy season, the lake equally overflows its banks thereby reaching the galamsey sites. The water serves as source of drinking water for inhabitants of old Bibiani town. From Table 1, GM is a galamsey tailing site where small scale miners are known to have channelled their process waste water through. As shown in Table 1, Asusuo and Abubayenyunu coded, Asus and Abuba, respectively are two streams with no known records of mining activities around them. Fig. 2 is a picture of lake Amponsah which is coded SW14, Table 1.

Climate and topology: The mining concession falls within the wet semi-equatorial climate zone of Ghana and is characterised by an annual double maxima rainfall pattern which is under the influence of the dust laden 'Harmattan' winds from December to February. The mean annual rainfall at Bibiani ranges from 888 mm to 2253 mm. The annual mean air temperature is about 26.9°C with maximum air temperature values observed in March and August. Mean annual pan evaporation is 1511 mm or 4.14 mm/day (<http://www.nobleminres.com.au/bibiani.html>).

In general, the area consists of two main land forms. These are the mountainous areas reaching heights of 340 m and occupying the western portion of the concession. They are of the Birimian geological formation and consist of very steep-sided hills with V-shaped narrow valleys dissecting them. The remaining area consists of low penepplain with elongated ridges which are about 260 m in height with wide broad valleys.

Sample collection and treatment: Water and sediment samples were collected in the dry season between October 2008 and March 2009. These samples were derived from water bodies that drain sites known to be mine tailing

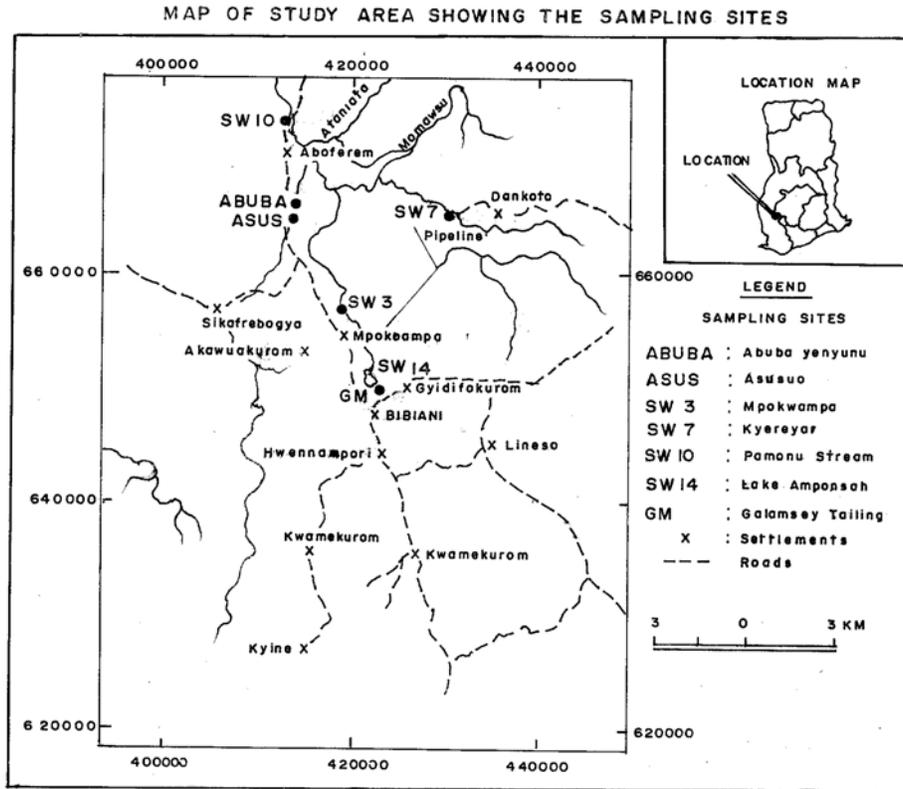


Fig. 1: Map of study area showing sampling sites.



Fig. 2: Lake Amponsah(SW14)

Table 1: Sample coding and identification

Sample code	Sample ID	GPS coordinates
SW3	Mpokwampa	002.32328W; 06.47192N
SW7	Kyereyar	057.7878W; 07.18878N
SW10	Pamonu Stream	002.34131W; 06.52191N
SW14	Lake Amponsah	057.5705W; 07.14381N
GM	Galamsey Tailings damp	057.5707W; 07.14327N
ASUS	Asusuo	071.8667W; 05.22825N
ABUBA	Abubayenyunu	071.8821W; 05.73162N

damp sites, abandoned mine sites, small scale mining (Galamsey) and large scale mine sites. In all, about 60 samples were taken and analysed for levels and species of mercury.

A survey was carried out on the whole area to locate and identify the important mine waste dumping sites. Geological setting and sites with record of previous usage of mercury in mining were also surveyed in order to choose the sampling location and site. Samples were collected from seven sites namely; Mpokwampa, Kyereyar, Pamonu Stream, Lake Amponsah, Galamsey Tailings damp, Asusuo and Abubayenyunu. Water samples were collected into teflon-bottles using ultra-clean free-metal sampling protocol (Gill and Fitzgerald, 1985; 1987) and acidified immediately with HCl to a pH less than 2. This was to prevent loss of analyte by adsorption of metal ions onto the walls of the vessel. This is very essential because it is very difficult to remobilize into solution previously adsorbed trace metals. Samples were kept in ice chest at all times during collection and transportation and stored in a refrigerator at 4°C until analysis was performed. Surface sediments (0 to ~10 cm) were taken from spots of water collection into pre-cleaned polyethylene containers by manual scooping using Teflon spoons.

Analyses of water samples: Preliminary tests were conducted by measuring in-situ physico-chemical parameters that affect mercury speciation. Parameters determined were temperature, pH, conductivity, total dissolved solids, alkalinity and dissolved oxygen. Mercury species were later determined.

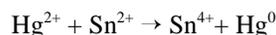
The pH meter used for this determination was calibrated using a pH 7.0 buffer powder pillow dissolved in 50 mL of water. The pH of the sample was then determined by pouring the sample into a 50 mL plastic beaker and with the tip of the electrode immersed 2.5-3.5 cm into the sample, the sample was stirred gently. The stable pH reading was recorded to the nearest 0.1 pH unit. The electrode tip was rinsed with deionised water and wiped with a tissue paper before continuing with the next sample.

The HACH section 5 conductivity meter was used for this determination and the instrument was calibrated using NaCl standard with known electrolytic conductivity (1000 $\mu\text{s}/\text{cm}$ at 25°C). The conductivity of the sample was determined by pouring 60ml of sample into a beaker; placing the probe into the sample and agitating for 5-10 sec to remove bubbles trapped in the slot. The conductivity of the sample was then determined and recorded.

The TDS pocket PAL tester was used to determine the TDS content of water samples. This instrument was calibrated using a standard solution of concentrated NaCl. TDS was determined by pouring 50 mL of sample into a

plastic beaker and the stainless steel probe immersed into the sample solution with gentle stirring. The stable reading was recorded and the value multiplied by 10 to determine the TDS value in mg/L.

Mercury determination in water samples: The total mercury concentration of non-filtered water was determined using the ICP-OES after reduction with stannous chloride (SnCl_2). The SnCl_2 reduces all reactive mercury present into elemental mercury according to the chemical reaction equation:



In the determination of organic mercury, 50ml of non-filtered water sample was passed through a column of activated charcoal. The organic mercury concentration of the eluent was determined using the ICP-OES. The activated charcoal was meant to trap all elemental mercury present in the sample. The difference between concentrations of the total and organic mercury gave the concentration of the elemental mercury.

Mercury determination in sediment samples: The sediment samples were air dried in the laboratory for 24 days. The dried samples were ground using mortar and pestle and then sieved into fine powder using laboratory test sieve of 125 μm diameter and 120 mesh size.

Qualitative detection of mercury (Amalgamation): The copper foil amalgamation technique was used for the qualitative determination of mercury in the sediment samples. The surface of a 2 cm copper foil was cleaned by washing with a 0.1M HNO_3 and rinsed with distilled water. 1.0 g of ground sediment sample was weighed into a 250 mL beaker. The copper foil was placed in the beaker and 30 mL of 0.5M HCl solution was added to it. The content was heated to boiling for about 30 min after which the copper foil was taken out with the aid of forceps. The copper foil was washed with distilled water and examined for any grey deposition. The grey deposition if present indicates the presence of mercury in the sediment samples. The deposition was then scrapped off to reveal the shining copper surface. When copper foil with the deposit was warmed in a test tube, globules were formed on the tube which confirmed the presence of mercury in the samples.

Quantitative determination of mercury in sediment: An Inductively Coupled Plasma-Optical Emission Spectrophotometer (ICP-OES) was used to determine the concentrations of species of mercury at the Ghana Standards Board chemistry laboratory.

To determine total mercury, sediment was digested with 1:1:1 solution of HCl, HNO_3 and HF in a round

Table 2: Physico-chemical Parameters of Water Samples

Site	pH	Temp. (°C)	Cond. (ms/cm)	TDS (mg/L)	Alkalinity (mg/L)	Eh (mV)	Do (mg/L)
SW3	7.1	22.7	2.25	135.0	580	13	5.8
SW7	7.2	23.6	0.22	111.8	140	7	5.2
SW10	7.4	23.5	0.21	114.1	140	23	4.8
SW14	8.4	30.6	0.48	168.9	180	54	5.3
GM	8.4	31.6	0.43	185.9	2.49	200	52
ASUSWater	7.3	25.1	2.77	132.6	130	25	5.5
ABUBA	7.5	24.2	2.75	134.3	190	37	5.4

Table 3: Sampling sites, total-Hg, organic-Hg and elemental-Hg concentrations in aqueous phase(in mg/L) and sediment(in mg/g)

Sample sites	Unfiltered Water(µg/L)			Sediments(mg/kg)		
	Total-Hg	Organic-Hg	Elemental-Hg	Total-Hg	Organic-Hg	Elemental-Hg
SW3	0.127	0.099	0.028	0.264	0.185	0.079
SW7	1.341	0.170	1.171	0.351	0.068	0.089
SW10	0.124	0.095	0.029	0.198	0.128	0.123
SW14	0.222	0.102	0.120	1.115	0.693	0.346
GM	0.446	0.346	0.125	1.739	0.123	1.616
ABUBA	0.203	0.105	0.098	n.d	n.d	n.d
ASUS	0.125	0.107	0.018	0.169	0.235	0.021

*n.d = not determined

bottom flask under reflux at 110°C for twelve hours (Bertsch and Bloom, 1996; Hossner, 1996). The mixture was diluted on cooling with saturated solution of H₃BO₃ to dissolve the fluorides and then filtered. Total mercury content of the solutions were then determined using Inductively Coupled Plasma-Optical Emission Spectrophotometer (ICP-OES).

In the case of Organic mercury, approximately 10g samples of sediment were placed in a 250 mL beaker in an oven and heated at 150°C for 48 hours to liberate elemental Hg (Biester, 1994). A Petri dish containing activated charcoal was placed over the beaker, covering approximately half of its mouth. This was to minimize the transfer of Hg vapour from one sample to another during heating and also capture Hg vapour that would otherwise be released to the laboratory environment. Samples were then digested and treated as outlined for the total mercury detection. The difference in the concentration between the total and organic mercury was reported as the concentration of the elemental mercury in the samples.

RESULTS AND DISCUSSION

Physico-chemical parameters and mercury species in water as can be seen in Table 2, the highest pH of 8.4 was recorded at both SW14 and GM while the lowest of 7.1 occurred at SW3. With respect to temperature, the water samples can be regarded as warm with the highest temperature of 31.6°C recorded at GM while the lowest occurred at SW3 with a value 22.7°C. In the case of conductivity, the highest value of 2.77 ms/cm was recorded at ASUS with the lowest of 0.21 ms/cm occurring at SW10. Finally, redox potential, another parameter that affects mercury speciation, showed the highest value of 200 mV at GM and the lowest value of 7 mV at SW7.

Total, elemental and organic mercury in water: With reference to Table 3, it is observed that the highest concentration of total mercury in water was 1.341 mg/L and this was recorded at SW7. The lowest value of this species was 0.124 mg/L and occurred at SW10. SW7 happens to be a river that runs through mining sites with records of gold amalgamation while SW10 is surface water which runs close to an abandoned galamsey site at Bibiani North. With regards to elemental mercury, SW7 still recorded the highest concentration of the metal with a value of 1.171 mg/L while the lowest concentration of 0.018 mg/L of this species was recorded at ASUS. ASUS is a site with no known records of mining activity. The highest concentration of 0.170 mg/L for organic mercury was recorded at SW7 while the lowest value of 0.095 mg/L occurred at SW10.

Correlation between organic mercury species and physico-chemical parameters of water: Mercury transformation in aquatic environments are known to take place through biotic or abiotic processes. The efficiency of microbial mercury transformation for instance is generally known to be influenced by parameters such as, temperature, pH, redox potential and the presence of inorganic and organic complexing agents (Ullrich *et al.*, 2001). In the light of this, the influence of the measured physico-chemical parameters; pH, temperature, conductivity and redox potential on mercury speciation have therefore been discussed. A look at how these parameters correlate with organic mercury content of the water samples was therefore undertaken.

Correlation between pH of water and organic mercury species: With reference to Fig. 3, it is observed that organic mercury concentration in water exhibited a negative correlation with pH where correlation

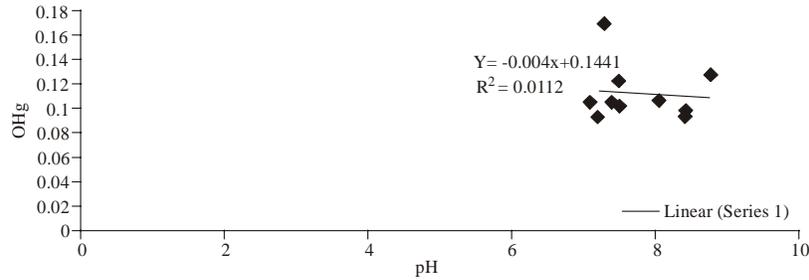
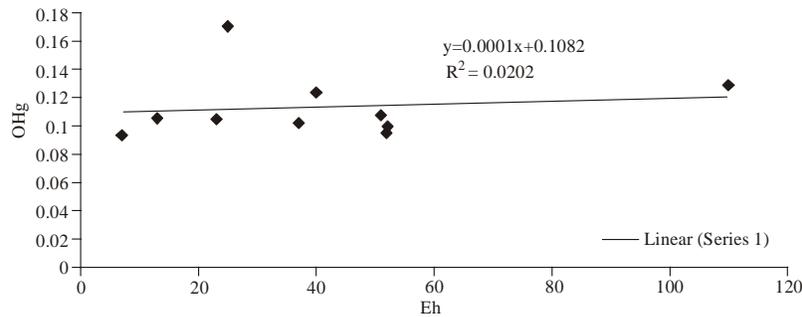


Fig. 3: Correlation of organic mercury in water with pH



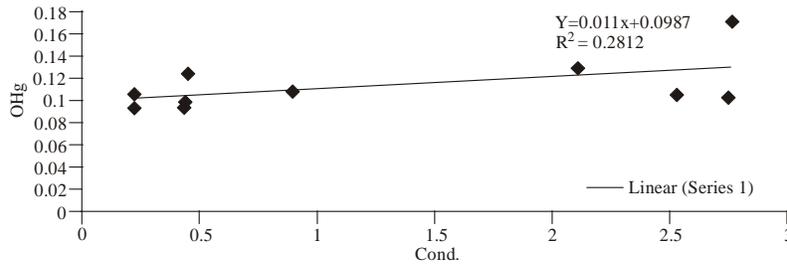


Fig. 6: Correlation of organic mercury in water with conductivity

was 0.281. Conductivity seems to have a direct bearing on the presence of the species of mercury. With the very strong correlation between the two, it is very likely the presence of certain ionic species in the water is influencing mercury transformation in the water. This trend can be seen in Fig. 6.

As can be seen from Fig. 7, total mercury levels in all the water samples fall below the WHO guideline value of 1.0µg/L (WHO, 2006) for drinking water except SW7 where the value of 1.341µg/L was recorded.

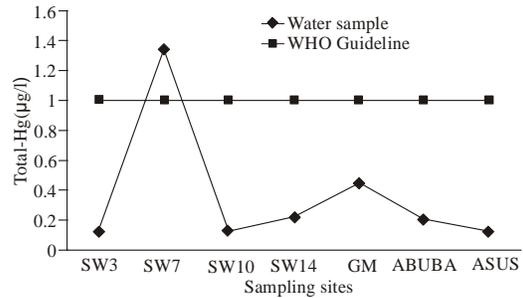


Fig. 7: Comparison of total mercury levels in water samples with WHO guideline values

Total, elemental and organic mercury in water and sediment: With reference to Table 3, total mercury values for the water ranged from 1.341 to 0.124 µg/L. The highest value occurred at SW7 while the lowest occurred at SW10. In case of elemental mercury, the highest value of 1.171 µg/L was recorded at SW7 while the lowest value of 0.018 µg/L occurred at ASUS. With organic mercury, the values were relatively low and ranged from 0.346 to 0.095 µg/L and recorded at GM and SW10, respectively.

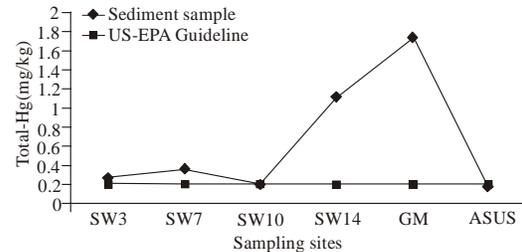


Fig. 8: Comparison of total mercury levels in sediment samples with US-EPA guideline values

Total mercury values for sediments ranged between 1.739 and 0.169 mg/L for GM and ASUS respectively while elemental mercury levels also for sediments at the sites recorded the highest value of 1.616 mg/L at GM and lowest value of 0.021 mg/L at ASUS. With regards to the organic mercury levels, the highest value of 0.693 mg/L was recorded for SW14 (Fig. 2) which is a very large lake that covers many gold mining sites. The lowest for the species occurred at SW7 with a value of 0.068 mg/L.

that sediments usually serve as sinks for mercury, it is not surprising recording higher values of the metal in sediments in relation to the values in water column.

Mercury is readily adsorbed unto sediments and slowly released into the water thereby forming a reservoir capable of causing chronic pollution long after the original source of mercury has been removed. The concentration of mercury in bottom sediments is therefore a very good indicator of water polluted with this element. On the one hand, bottom sediments are where mercury accumulates as a result of simple sedimentation. Mercury is usually released from the sediments becoming available for further biogeochemical transformations. The rates of these two processes; accumulation and release depend significantly on the specific environmental conditions in a given water system (Boszke *et al.*, 2003). Given the fact

In this study, it has been observed that total mercury in sediments is above 0.20 mg/kg, the US-EPA guideline value (US-EPA, 2007) for soils, Fig. 8. It is also obvious from Fig. 9, that, total mercury levels in sediments from all sampled sites are higher than observed for the corresponding water columns except at SW7 where total mercury in the water column is found to be higher than that observed for the sediment. SW7 is a large stream which runs through many mine sites. It is likely mercury bound to bulk organic matter in soils is susceptible to wash out in runoffs finding their way into this stream. With regards to both organic and elemental mercury, the same trend as observed for total mercury in water and sediments is obvious and can be seen in Fig. 10 and 11.

Table 4: Sampling sites, total - mercury, arsenic, iron, lead, copper, and zinc concentrations in sediment (in mg/kg)

Sample sites	Other Metal Species in surface sediments(mg/kg)					
	Total-Hg	Arsenic	Iron	Lead	Copper	Zinc
SW3	0.264	235.114	3342.730	111.239	17.786	235.114
SW7	0.351	1960.480	3107.380	62.723	20.042	80.611
SW10	0.198	1185.900	3141.650	39.250	12.614	46.686
SW14sed	1.115	550.391	3438.730	111.992	32.024	38.961
GMsed	1.739	4.135	3562.320	68.706	29.765	36.719
ASUSsed	n.d	9.210	2689.580	12.242	6.081	21.289

Table 5: Pearson correlation coefficients (r) for sediment data

	Total - Hg	Arsenic	Iron	Lead	Copper	Zinc
Total-Hg	1.000					
Arsenic	- 0.580	1.000				
Iron	0.872	- 0.100	1.000			
Lead	0.170	- 0.061	0.779	1.000		
Copper	0.877	- 0.022	0.887	0.729	1.000	
Zinc	- 0.504	- 0.046	0.231	0.575	- 0.050	1.000

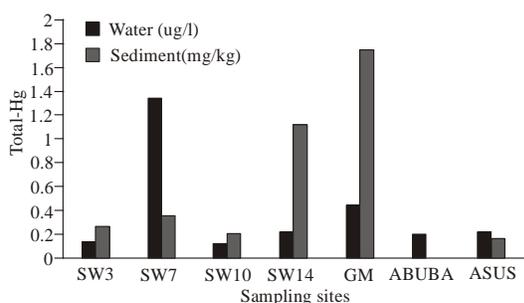


Fig. 9: Comparison of total mercury levels in water and sediment

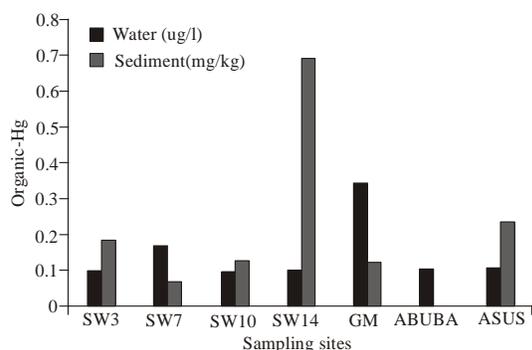


Fig. 10: Comparison of organic mercury levels in water and sediment

Concentrations of other metals namely, arsenic, iron, lead, copper, and zinc were determined in the sediment samples, Table 4. These results were correlated with total-mercury to ascertain how their concentrations affected the level of mercury in the sediments.

Arsenic was negatively coordinated with total mercury as seen in the computed Pearson correlation coefficients, Table 5. The observation suggests that the mercury and arsenic are coming from different sources.

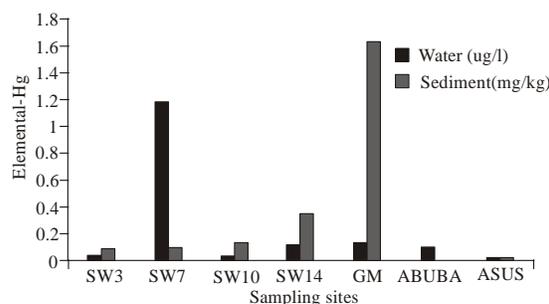


Fig. 11: Comparison of elemental mercury levels in water and sediment

Arsenopyrites are the main gold bearing ores found in the area. Arsenic is therefore likely to be becoming from natural source of gold ore while mercury is from anthropogenic sources due to gold amalgamation.

The other metals, iron, lead, and copper are positively correlated with mercury. While iron and copper are strongly correlated with coefficients, 0.872 and 0.877, respectively, lead is though positive, weakly correlated with mercury with coefficient value 0.170, Table 5. These metals may be coming from similar sources and fed into the rivers by runoffs. Zinc, just as arsenic is negatively correlated with mercury suggesting the two may be coming from different sources.

CONCLUSION

The study revealed that metallic mercury released into the environment persists for a long time and subsequently transformed into other species of mercury. Total mercury determined in water and surface sediments are found to be in the range; 0.124 to 1.739 µg/L and 0.169 to 1.115 mg/g, respectively. It is obvious from this study that sediments serve as a sink for mercury species. The sediment is more polluted among the environmental

matrices studied as it is observed that sediments carry greater mercury content than the water. From this observation, it can be ascertained that runoffs carry mercury species during the rainy season and deposit them in the rivers in the mining area where the metal accumulates as a result of simple sedimentation. .

Elemental mercury concentrations range between 0.018 to 1.616 µg/L in water, and 0.021 to 0.346 mg/g for surface sediments. The water samples contain higher mean value of elemental mercury than the sediments. The high volatility of elemental mercury may account for the release of the metal from the sediment into the water column which may eventually vaporise into the atmosphere.

With regards to organic mercury, recorded values are in the range, 0.093 to 0.170 µg/L for water and 0.068 to 0.693 mg/g for surface sediments. The surface sediments recorded the highest concentration of the species while the water recorded the least concentration. It is likely the factors responsible for mercury transformation are more active in the sediment and subsequent release of the species into the water column is not vigorous.

With regards to the influence of physical parameters of water on mercury species, the study showed a decrease in mercury species as pH increased. It is likely mercury speciation occurred more in an acidic environment. It has also been established that as redox potential and conductivity increased, mercury speciation also increased as the species have all shown positive correlation with these two parameters. Mercury speciation is therefore promoted under these conditions.

The study has also revealed that, while mercury, iron lead and copper in the sampled sites may be from anthropogenic sources, arsenic and zinc may be coming from natural sources. It is also likely some species of these metals are aiding mercury speciation in water.

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