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Assessment of Levels of Cadmium and Mercury of Two Estuaries in Two Regions of Ghana

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Abstract: Instrumental Neutron Analysis by Americium-Beryllium radioisotope neutron source was employed for the determination of mercury and cadmium from eight different sites of the Pra and Ayensu estuaries in Ghana. Mercury and cadmium were identified in $\mu g\ g^{-1}$ levels and values correlated with the pH values of water column and soil samples. Substrates like the Blue Tilapia and the European Green Crab were used as bioaccumulation indicators for the mercury levels in the various samples. A summary of the mean, minimum and maximum soil/water mercury and cadmium concentrations detected for the 8 sites with the widest geographical distribution; river bank at Beposo showed a maximum of 3.95×10^{-3} $\mu g\ g^{-1}$ of mercury which is far below the Environmental Protection Agency's permissible limit of 0.134 $\mu g\ g^{-1}$. Insignificantly low levels of mercury concentrations were analyzed in shoulder soils and water samples over the period of the study. A significantly high levels of concentrations of mercury existed in the riverbed sediments as compared to that for the riverbank sediments, water and the shoulder soils. The degree of concentrations of mercury showed that mercury and cadmium concentrations decreased significantly and gradually as one moved from Beposo to the Shama beach through Bosomdo and Krobo. Mercury and Cadmium residues were also recorded in both European green Crab and blue tilapia. The levels of the two elements were slightly high in the Crab as compared to the Blue Tilapia. The differences could be attributed to the fact that the Crab is a bottom-dweller and predator. The concentrations of the two elements were far below the world permissible levels.

Key words: Instrumental neutron analysis, Americium-beryllium neutron source, thermal neutrons, non-detectable, bosomdo, beposo, Shama beach, Dawukwa, Okyereko, Atakyedo, Sankor, Krobo

INTRODUCTION

Ghana is endowed with many rich mineral resources including gold, aluminum and manganese. Mining and export of gold in Ghana began long before the arrival of the Europeans. The metal was extracted from alluvial material deposited on the banks and bottoms of rivers. For example, the alluvial gold was obtained from the beds of river Offin. The use of mercury (modern technology) in the mining of gold dates from about 1880. The exploration began in Tarkwa and was extended to Obuasi in 1890. At the moment there are about fourteen mining companies in Ghana which are engaged in both surface and sub-surface mining. However, gold mining in Ghana in recent times has become unpopular as result of the environmental damages the country has experience from that sector of the economy (Kwarteng, 2003). Ghana has experienced about nine cyanide spillages between 1989-2003 (Amegbey and Adimado, 2003).

Pollution from mining activities is of major concern to governments, environmentalist and mining communities for two main reasons: it effect on human health and the environment. The processing and extraction of gold from its ore by both the small-scale miners galamsey (licensed and illegal) and the large-scale mining companies have posed serious environmental problems.

Activities at these source result in the release of toxic chemicals into the environment. Toxic chemicals pollute water, soil, terrestrial wildlife, the air and destroy biodiversity. Very often, chemicals such as mercury used in extracting gold by the small scale miners galamsey are injurious to biotic life.

Such chemical eventually get access into water systems to pollute them (Hines and Brezonik, 2007). Also, the abandoned Offin Continental Goldfields and illegal mining activities situated in the basin of river Offin and Pra, used mercury to extract gold which pollute the rivers. The mercury used in the operations just upstream of river

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Pra gets access into the river, trickling down gradually through the estuary into the sea (Asabere-Ameyaw and Anamuah-Mensah, 2004). Harmful effect of mining becomes apparent after long periods of exposure. For example, the cumulative effects of some chemicals have been seen after 10-20 years of continuous exposure. Unfortunately, these effects are irreversible. People living in and around the mines are exposed to these harmful chemicals used in mining operation. Again, certain foodstuffs absorb metals such as mercury, lead, cadmium which are eventually eaten by human beings (Essumang *et al*., 2007). These toxic chemical produces an adverse effect in a biological system. It may either alter the normal function or destroy the life of an organism. All chemicals are toxic when absorbed at certain dosages. This means that all chemicals are capable of altering some biological function or producing negative effects in some organism or can succeed in destroying certain functions of the ecosystem.

According to the Tutorial on Toxicology (Pt.I) (2004) all materials or chemicals are poisons; there is none which is not a poison. The right dose differentiates a poison from a remedy. In view of this phenomenon, it is very necessary to study the amount of mercury that are brought from upstream by these operators into the sediment, water column and the soil shoulders of the Pra basin. The harmful nature of mercury and cadmium in the environment has therefore called for this research as far as the biota and sustenance of the Pra River is concerned.

The objective of this research is to use Neutron Activation Analysis (NAA) using thermal neutrons from a low flux Am-Be radioisotope from such analysis and to determine the levels and distribution of mercury in the Pra and Ayensu estuaries. Again, the study is to assess the bioaccumulation of mercury/cadmium in blue Tilapia and European Green Crab and how pH affects the bioaccumulation.

MATERIALS AND METHODS

Sample collection, preparation and irradiation

Sampling sites: Two sampling sites were chosen and these were the Pra and Ayensu estuaries. The Pra estuary was chosen because the river Pra whose estuaries was analyzed passes through very important gold mining communities in Ghana. Sometimes some alluvial gold mining occurs right in the Pra River upstream. This makes it important to study the cadmium and mercury levels of the Pra estuary where there is lot of human activities going on there. The Ayensu river was also chosen as the control for the studies. It has a similar characteristic with the Pra river except that there is no mining activity close to the river. The river Ayensu does not pass through any gold mining community in Ghana (Fig. 1).

Sample collection: The water samples were collected in the middle of the river in camera film containers of about 10 mL capacity and stored in an ice chest with ice blocks and later in the laboratory stored in a fridge for further analysis. The samples were collected from various sites starting from Beposo and continued to Shama beach along the bank of the Pra and Ayensu rivers. The Ayensu river samples were used as control in the study. The sampling was done four times at 2 weeks intervals starting from May to the end of July, 2006 at all the sites. In all about 75 samples were collected in sampling date.

Improvise dedicated plastic scoop and bamboo stick were used for the sampling of the sediment where about 50 g of sediment samples were scooped into film containers. In the case of sediment from the middle of the river, a long bamboo stick with an open end was pushed down into the water and pressed until enough sediment was collected. The open end of the bamboo collected about 50 g of sediment which was then scooped into the film container and transported to the laboratory for further analysis. The sampling was done by the use of canoe across the river. The samples were sun/air dried.

Oreochromis niloticus (Tilapia) and *Galaxies brevipinnis* (crab crustacean species) were also used for this analysis. Four prominent fishing shores along the estuary were chosen as the sampling site. Fishing nets were set in all the four sampling site a day to the sampling date and this was left in the hands of caretakers. Fish samples (*Oreochromis niloticus* and *Galaxias brevipinnis*) caught in the net were taken by hand with gloves into a rubber container and stored in an ice chest for further treatment.

Sample preparation and analysis

Water samples: Exactly, 0.5 mL of each water sample was pipetted using calibrated Eppendorf tip ejector pipette into clean pre-weighed 1.5 mL rabbit capsule (sample vials) and reweighed and heat-sealed using soldering rod. Four of these sample vials were also placed into a 7.0 mL volume rabbit capsule and heat-sealed. Two replicates of each sample were shown.

Sediment samples: Each sediment sample was air-dried for 3 days in a clean environment. Debris such as shells and other organic materials were handpicked from the samples. The samples were crushed using an agate mortar and pestle, sieved using an 85 μm mesh size (USA

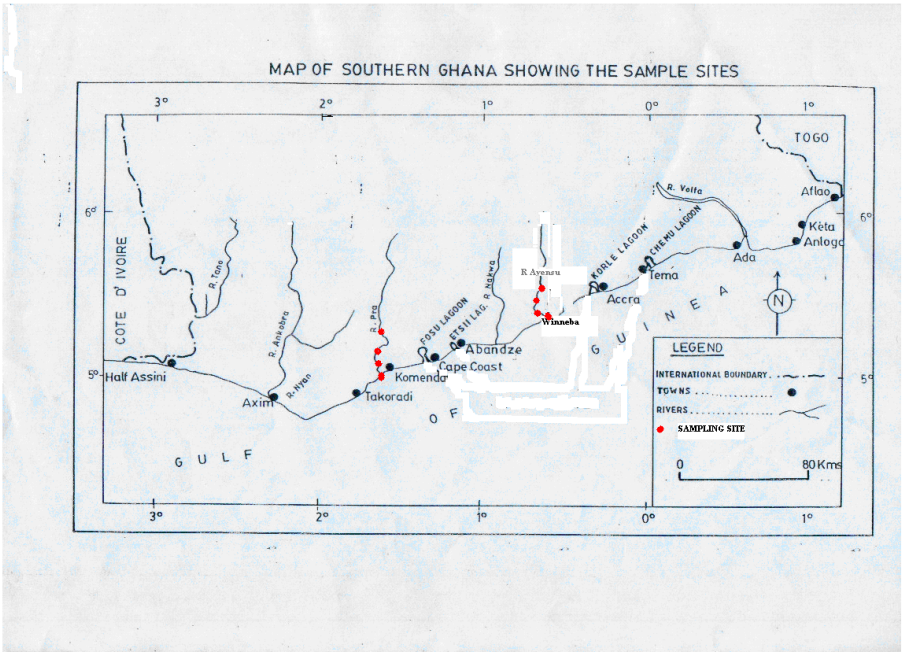

Fig. 1: Map of Southern Ghana showing the sampling sites

standard sieve) and homogenized. About 100 mg of each sample was weighed into a clean polyethylene film. The films were wrapped and heat-sealed. These samples were labeled according to their site. Two replicate sub-samples of each were shown for each sample.

The samples were again packed into 7 mL volume rabbit capsules for irradiation. Mercury reference standard material NIST-SARM 1641d with concentration 1.59 mg kg^{-1} for water and 1646a with concentration 0.04 mg kg^{-1} for sediment and standard solution Lot no. 890312 for cadmium of concentration 9.5 μg g^{-1} supplied by NIST, IAEA were used. These reference materials were treated the same way as the samples were packed into rabbit vials and heat-sealed for irradiation.

Biota samples: For the *Oreochromis niloticus* and *Galaxias brevipinnis* samples about 200 mg (wet weight) of the muscle of each were weighed using Mettler Electronic Balance AE 163-BDH into a clean polyethylene film and treated like the sediment and water sample for the irradiation. The muscles were used because that is the part that is eaten in Ghana. Shells were also used in the case of the crab and the average recorded.

Irradiation source: The irradiation source was a 20-Curie Americium-Beryllium (Am-Be) radioactive neutron source. It was cylindrically shaped and was fixed in a holder at the center of a fibre-glass tank, filled with de-ionised water.

The de-ionised water served a dual purpose of moderator and also absorber of neutrons. Concrete blocks were arranged around the tank to provide extra shielding. Transfer of the sample to and from the neutron source was done by means of a flexo-rabbit pneumatic transfer system operating under a pressure of 15 psi giving a sample transfer time of 1.3 sec (Tetteh, 1989). The thermal neutron flux at the irradiation site was 5×10^{11} ns^{-1} cm^{-2} (Osae and Amoh, 1989).

Irradiation of sample and counting: Each of the samples was sent by the pneumatic transfer system into the Am-Be source for irradiation. In this regard, 1 h was chosen for all the samples because all the metals in question were all medium lived. These samples were irradiated for 1 h and left overnight for decaying (cooling) process to take place. The samples were then counted the next day for 600 sec and their intensities saved for further analysis.

Data processing: The detector type used for the counting of signals was an ENERTEC High Germanium (HPGe) detector of 3000 (+ve) bias and a resolution of 2.55 keV for 1332 KeV photo peak of Co-60. The signals from the detector were passed through the spectroscopy amplifier and then accumulated by the Canberra Multi-Channel Analyzer (MCA) for 600 sec. The spectra from the MCA were transferred to a DEC 350 microcomputer for analysis using Gamma spectrum analysis software (Ortec multi-channel buffer (MCB) MAESTRO 32). This software

identifies the various photo peaks and works out the areas under them. By means of the equation $A = \sigma\pi (m\ M^{-1})\ N_A SDC\theta P_y\varepsilon$ (Tolgyessy and Kyrs, 1989) the concentration of each element was calculated.

Determination of pH of soil samples: For the pH of the soil samples, 1.00 g of the dried soil samples of each soil was weighed and dissolved in 100 mL of distilled water for 24 h. The supernated solution was filtered to get a clear solution and the pH of the subsequent solution was determined using the pH meter. The pH's were done for both the samples and the control samples as well. The pH meter used was properly calibrated.

Determination of pH of the water samples: The water samples from the fridge were allowed to attain the normal room temperature of about 26°C. The pH's of the water samples were determined with a pH meter. The pHs was done for both the samples and the control samples as well.

RESULTS AND DISCUSSION

Results of the validation: The performance of the analytical method: Reliability criteria and validation is shown in Table 1. The result of the validation of the method is shown in Table 1. The result of the validation as shown in Table 1 gave a percentage recovery of 99.16% for Hg in water, 92.5% for Hg in sediment and 99.70% for Cd in the sediment and water. These result shows that the NAA method of analysis employed were accurate. Interference reactions by fast neutrons may occur in the present analytical procedure however, these interference reactions were virtually negligible when we applied the interference coefficients (De Goeij, 2000; Katoh *et al*., 2003).

Results of the control samples: The result of the control samples is shown in Table 2. The result shows that all the metals of interest analyzed were below the detection limit of 0.001 μg g^{-1} of Hg and Cd of the NAA analytical method used.

The Ayensu river used does not pass through any gold mining community in Ghana. This was used to compare with the result that was obtained from the Pra estuary which passes through major gold mining community in Ghana.

The result of the Pra estuary: The mean cadmium concentrations in the shoulder soils of beposo, Bosomdo, Krobo and Shama beach proved that a significant amount of Cadmium existed. This is probably due to the gold mining activities upstream since Cd is an impurity in Zn. Also, the emissions from vehicular fall-outs may contribute to the Cd levels (Essumang *et al*., 2006).

As shown in Table 3, Beposo had a high cadmium concentration of 0.0260 μg g^{-1}. The cadmium concentration decreased as one moved from Beposo

Table 1: Validation of the method

	Concentration of mercury and cadmium						
Standard material	Standards	1	2	3	4	Mean	Recovery %
Hg/mg L^{-1} for water	1.59	1.5500	1.5800	1.570	1.579	1.5700	99.16
Hg/μg g^{-1} for sediment	0.04	0.0390	0.0380	0.035	0.037	0.0370	92.50
Cd μg g^{-1}	9.50	9.4702	9.4779	9.470	9.471	9.4727	99.70

Table 2: Results of the control samples

	Control: mean Hg and Cd concentration at the sites											
	Sh. Soil (μg g^{-1})		R. Bed (μg g^{-1})		River bank (μg g^{-1})		Water column (μg L^{-1})		Tilapia (μg g^{-1})		Crab (μg g^{-1})	
Site	Hg	Cd	Hg	Cd	Hg	Cd	Hg	Cd	Hg	Cd	Hg	Cd
Dawukwa	<0.001	<0.001	<0.001	<0.001	<0.001	<0.01	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
Okyereko	<0.001	<0.001	<0.001	<0.001	<0.001	<0.01	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
Atakyedo	<0.001	<0.001	<0.01	<0.001	<0.001	<0.01	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
Sankor	<0.001	<0.001	<0.001	<0.001	<0.001	<0.01	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001

Below detection limit of 0.001 μg g^{-1}

Table 3: Mean Hg and Cd conc. (μg g^{-1}) at the various sites

	Shoulder soil		River bed		River bank		Water column	
Site	Hg	Cd	Hg	Cd	Hg	Cd	Hg	Cd
Beposo	ND	0.026±0.004	0.004±0.00060	0.038±0.002	0.003±0.0004400	0.0133±0.002	0.0010±0.00015	0.001±0.000150
Bosomdo	ND	0.013±0.002	0.002±0.00030	0.018±0.003	0.0013±0.000015	0.0213±0.007	ND	0.0022±0.00036
Krobo	ND	0.009±0.001	0.001±0.00002	0.01±0.0020	0.00104±0.00016	0.0078±0.001	ND	0.0015±0.00020
Shama beach	ND	0.027±0.004	0.001±0.00015	0.029±0.004	ND	0.008±0.0010	0.0010±0.00015	0.0023±0.00030

Table 4: Mean Hg and Cd conc. μg g^{-1} of substrate

	Crab		Tilapia	
Site	Hg	Cd	Hg	Cd
Beposo	-	-	0.0057±0.000150	0.00293±0.000440
Bosomdo	0.00586±0.0008790	0.01009±0.00150	0.0051±0.000165	0.00274±0.000411
Krobo	0.00427±0.0006410	0.00895±0.00013	0.0047±0.000150	0.00254±0.000381
Shama. Beach	0.00323±0.0004845	0.00779±0.00120	0.0044±0.000150	0.00266±0.000399

through to Krobo. A mean of 0.0267 μg g^{-1} of cadmium was detected at Shama beach, the highest. This might be due to the average contribution of Cadmium content from the sea water which is about 0.1 μg L^{-1} or less.

Table 3 shows a significant increase of cadmium concentrations in all the sites. Much of the cadmium which entered the fresh water from industrial sources was rapidly adsorbed by particulate matter where they settled at the bed of rivers (Jain and Sharma, 2002) shown the concentration in water below detection limit in all the sites analyzed.

The cadmium concentrations in shoulder soils at Beposo were significantly high over the sampling period. At Beposo, the first sampling gave a cadmium concentration of 0.0503 μg g^{-1} with a least of 0.0254 μg g^{-1}. Subsequently sampling periods had a decreased concentrations of cadmium with the last being an exception. This trend may be due the fact that the sampling began in the dry season and entered into the raining season, hence the decrease over the sampling period. The mean mercury concentrations in the shoulder soils of Beposo, Bosomdo, Krobo and Shama beach were not detected (Table 3).

The non-detectable nature of mercury in the shoulder soils may have been caused by the volatile forms of mercury (e.g., metallic mercury and dimethyl mercury) that are expected to vaporize to the atmosphere or in solid forms, partition to particulate soil or water column and later transported down in the water column to the sediments (Stafford *et al*., 2004). Sample collected from the riverbed at various sites gave a significant mean mercury concentration.

At Shama beach 0.21×10^{-3} μg g^{-1} of mercury concentration was detected which was the least of all. Inorganic mercury sourball partiality in soil is not readily desorbed. Thus, fresh water and marine sediments are important repositions for inorganic forms of the element and leaching is a relatively insignificant transport process in soils. The values for the riverbed indicated that most of the mercury deposited on soil is absorbed onto soil sediments and does not leach down into the groundwater (Grigal *et al*., 2000). A review of the mean mercury concentration, river bank sediments showed values ranging from below detection of 0.001 μg g^{-1} -0.003 μg g^{-1} at the various sites. The highest concentration was found at Beposo, a location upstream. Mercury and cadmium concentration values in the riverbank sediment were quite low compared to concentrations in bed sediment. Shama beach which is close to the estuary had a non-detectable value with Bosomdo and Krobo giving 9.7×10^{-3} and 4.0×10^{-3} μg g^{-1}, respectively.

Table 5: Table of salinity, soil and water pH

Sample sites	Water (pH)	Soil (pH)	Salinity (%)
Beposo	7.51	5.56	0.43
Bosomdo	7.52	6.89	1.00
Krobo	7.54	7.81	1.00
Shama Beach	7.65	8.17	2.00

The study showed clearly that much of the mercury and cadmium concentration settled at the bed of the river rather than at the bank as shown in Table 3. Comparing these results with that of river Ayensu (control samples) which has similar ecosystem with that of the Pra estuary, the Pra estuary recorded elevated levels of the metals analysed. Hence, the gold mining activities upstream could be a contributory factor for the elevated levels.

The results of the biological samples: The biological concentration of *Galaxies brevipinnis* (Crabs) sampled at the various sites had a range of 0.00586-0.00323 μg g^{-1} Hg. Crabs were not found at Beposo but those of the other sites gave a substantial uptake of mercury and cadmium.

Bosomdo, a village close to Beposo had a mean mercury concentration of 5.86×10^{-3} μg g^{-1} of Hg in Crab, a value about 4 times that in the riverbed sediment as showed in Table 4. At pH 8.17, (Table 5) the average mercury concentration of Crab at Shama beach was 3.23×10^{-3} μg g^{-1} (wet weight basis). The high concentration of mercury and cadmium obtained for Crabs in the studies is an indication of their bottom-dwelling nature (Yanch, 2007).

This significant high values could also be attributed to the slow movement of Crabs which intend makes them accumulate much mercury and cadmium at the bed of the river. The mean concentration of mercury and cadmium declined significantly from 5.86×10^{-3} -3.23×10^{-3} and 0.01009-0.0078 μg g^{-1} in Crab during the survey (Table 4). This decline was presumably due to curtailed production,

use and emissions of mercury and cadmium from the erstwhile Offin Continental Goldfields and galamsey activities along the Pra River (Garg and Ramakrishna, 2006). Again compared to that of the Ayensu River (control) samples, those of the Pra estuary had higher levels (Table 2).

In Table 4, *Oreochromis niloticus* (Tilapia) samples were analysed as muscle samples and all the concentrations were shown on wet weight basis. A high mercury concentration of 5.7×10^{-3} μg g^{-1} in Tilapia was observed at Beposo. A least of 4.4×10^{-3} μg g^{-1} of mercury was shown at Shama Beach while Bosomdo and Krobo had 5.1×10^{-3} and 4.7×10^{-3} μg g^{-1}, respectively. The mercury and cadmium concentrations in Tilapia were lower than those of in the Crab.

This could be as a result of the fast movement and surficial water-dwelling habits of the Tilapia (Yanch, 2007). The most common organic form of mercury, methyl mercury is soluble, mobile and quickly enters the aquatic food chain (Davis *et al*., 2003). This form of mercury is accumulated to a greater extent in biological tissue than are inorganic forms of mercury (Riisgard and Hansen, 1990). Fishes appear to accumulate methyl mercury from both food sources and the water column. However, Hall *et al*. (1997) found that food was the predominant source of methyl mercury uptake by fish.

CONCLUSION

From the results of the analysis, it was clearly observed that despite the low neutron flux, the neutron activation technique is sensitive for the determination of mercury and Cadmium budgets of the Pra estuary. Mercury and Cadmium levels decreased from the target source (Beposo) into the sea which clearly suggest that the mercury come from upstream (Dunkwa-on-Offin) and its environs. It can also be deduce from the results that as you move downstream to Shama beach acidity decreases as salinity level increases. This effect affected the mercury and cadmium distribution in the water body. Increase in acidity and decrease in salinity decreases the concentration of the metals.

Subsistence fishermen including natives of Beposo, Krobo, Bosomdo and Shama beach who consume locally caught fish from the Pra river or consume long-lived predatory species like the European Green Crab are exposed to some elevated concentration of mercury.

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