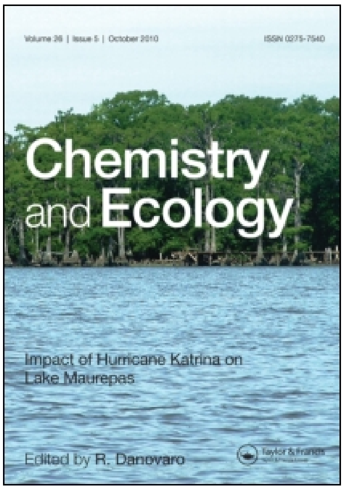

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Analysis of silver in the water column of the Pra and the Eture estuaries in Ghana

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Silver is a rare but naturally occurring metal, often found deposited as mineral ore in association with other elements like gold, manganese and aluminium. Emission from smelting and gold mining operations as well as indiscriminate disposal of waste from photographic laboratories and other chemical laboratories are some of the anthropogenic sources of silver in the environment. This work analysed the concentration of total recoverable silver in the water column of the Eture and the Pra estuaries in Ghana. The result indicated high concentrations in the two estuaries, higher than the values that should be tolerated in the estuarine environment. The high levels in the Pra estuary could be attributed to the numerous mining activities upstream and this was the same for the Eture estuary which also runs through Cape Coast and the University of Cape Coast. The higher levels of silver reported in this study area suggest that much work has to be done to monitor and improve the quality of these ecosystems in the Ghanaian environment.

Keywords: silver; Pra estuary; Eture estuary; gold mining

1. Introduction

Silver is a rare but naturally occurring metal, often found deposited as a mineral ore in association with other elements like gold. Emission from mining and smelting operations, manufacture and the disposal of certain photographic and electrical supplies, coal combustion and cloud seeding are some of the anthropogenic sources of silver in the biosphere [1]. The global biogeochemical movements of silver are characterised by releases to the atmosphere, water, and land by natural and anthropogenic sources, long-range transport of fine particles in the atmosphere, wet and dry deposition, and sorption to soils and sediments [1].

Silver in the aquatic environment has received attention in recent years because it is highly toxic to aquatic organisms even at trace levels, and widely distributed in the vicinity of industrialised areas [2–4]. The most recent measurements of silver in rivers, lakes and estuaries using clean techniques showed values about 0.01 $\mu g/l$ for pristine and unpolluted areas and 0.01–0.1 $\mu g/l$ in urban and industrialised areas [1].

In the water column, silver is readily adsorbed onto particles due to its high particle affinity (typical partition coefficient, K_d of $10^{4.5}$ to 10^6); thus, sediment constitutes the main

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repository for silver in marine and estuarine environments [5]. It has been established that trace metals in sediment can adversely affect benthic organisms [6]. Estuarine benthic invertebrates accumulate metals from their surrounding environment through two pathways. Dissolved metals are accumulated through permeable membranes, whereas particle-associated metals are assimilated after dietary ingestion [2]. In the past decade, an intensive research effort has greatly improved our understanding of the toxicity of silver in freshwater [7].

The integration of fundamental principles of physiology, aquatic geochemistry and toxicology has yielded biotic ligand models (BLMs) which are capable of site specific prediction of acute toxicity (e.g. 96-h LC_{50}s) in much particular freshwater chemistry [8–10]. BLMs hinge on the understanding that only the free ion, Ag^+ causes acute toxicity by binding to key target sites on the gills as a Na^+, K^+, ATPase, carbonic anhydrase, and Na^+ and Cl^- uptake [11,12]. Disruption of these enzymes inhibits active sodium and chloride ion uptake and therefore, osmoregulation by the fish. The greater the saturation of these sites with Ag^+, the greater the predicted toxicity. Thus freshwater organisms subjected to silver poisoning may die from ionoregulatory failure [13].

Marine and brackish fish have two potential target tissues for silver which are the gills and the gut; and in freshwater, fish gut absorbs Na^+, Cl^- and thereby, water to replace osmotic water loss across the gills [14]. Furthermore, the gills rather than performing active Na^+ and Cl^- uptake, serve to actively excrete the excess Na^+ and Cl^-. Thus, across the gradient of increasing salinity in estuaries, the function of the gills changes gradually from ionic absorption to ionic excretion as the osmotic point is crossed, and drinking begins around this point. In the water, Ag speciation is dominated by the concentration of Cl^- and as salinity increases, the free Ag^+ ions essentially disappear, being replaced with a sequence of neutral and anionic silver chloride complexes [15].

In general, silver ion is less toxic to freshwater aquatic organisms under conditions of low dissolved silver ion concentration and increasing water pH, hardness, sulphides, and dissolved and particulate organic loadings under static test conditions compared with those that flow through regiments, and when animals were adequately nourished instead of being starved [16–21]. It has now been agreed on that increasing concentrations of dissolved organic carbon afford the highest protective effects [6,19]. Among all tested species, those that are most sensitive (affected by) to silver are the poorly nourished and the younger aquatic organisms. Low water hardness or low salinity exposes the young and the poorly nourished aquatic organisms to most of the harmful effects of silver [16,22–25]. In our experiments, sensitive aquatic plants grew poorly at 3.3–8.2 μg/silver/l during exposure for 5 days and died at concentrations greater than 130 μg/silver/l.

In Ghana, gold is mined almost everywhere, from the coast to the north of the country and the fact that silver is associated with gold mining makes it very important for this study. The river Pra and Suruwe whose estuaries were analysed pass through very important gold mining communities in Ghana.

The aim of this work was to determine the concentration levels of silver in the water column of the estuaries of the rivers Pra, Kakum and Surewe (Eture) which pass through very important gold mining communities in Ghana, and to suggest some potential remedial action if alarming conditions are found.

2. Material and methods

2.1. *Sampling strategy*

Water samples were collected at 1 km from the Pra estuary and 20 metres from the Eture estuary. The sampling period spanned from January to March, 2007. This period in Ghana is the dry season with no rainfall or very little rain hence less dilution of the water bodies used in the study. The sampling sites are shown in Figure 1.

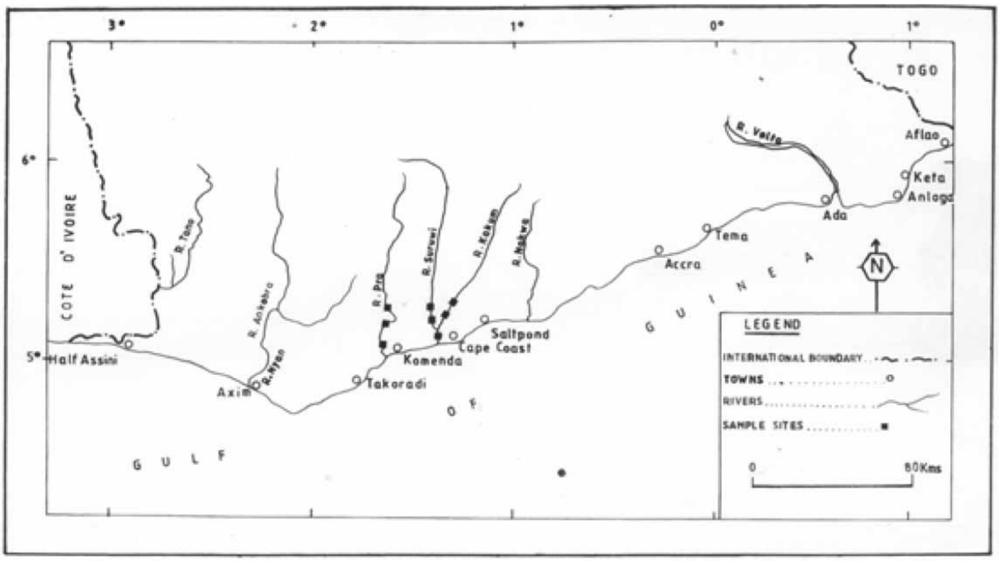

Figure 1. Map of Ghana showing the coastal belt and sampling sites.

Dark-coloured Amber glass bottles with cork caps, cleaned with detergent and thoroughly rinsed with double distilled water, were used for the sampling. After the first sampling the bottles were washed with detergent, double distilled water and nitric acid before the next sampling. For the Pra estuary, 10 sampling sites were chosen at an average distance of 100 metre intervals and increments were taken by means of a canoe. Sampling bottles of 200 ml capacity were used to collect the increments into 2.5 l Winchester bottles, out of which composite samples were made. Then 250 ml of the bulk was transferred into 300 ml capacity sample bottles and taken to the laboratory for analysis.

Three sampling sites were also chosen in the Eture estuary. The estuary consisted of two streams merging from east and west at about 50 metres from the sea. Each site was selected in the eastern stream (Kakum), the western stream (Suruwe) and the point of merger (Eture). A total of 100 samples were collected for this study. The samples were carefully collected directly by hand (in gloves) into sampling bottles by pressing them under the water surface to avoid the upper surface boundary layer, where surfactive materials accumulated. The sampling bottles were twice partially filled, well shaken and emptied before the actual filling was done. Then 5.0 ml of nitric acid was added to each of the filled bottles and they were immediately corked, properly labelled and placed in the sampling bag for further treatment and analysis in the laboratory.

2.2. *Sample treatment for total recoverable silver*

Treatment of the samples was done with aqua regia in a solubilisation process to eliminate the various complex forms of the silver [26,27]. First, 2 ml of aqua regia was added to 8 ml of sample and left overnight. The silver lamp for the analysis was set and allowed to warm up for 20 minutes. During this period, the instrument AAS was aligned by positioning the monochromator at the correct wavelength of 328.1 nm. The proper monochromator slit width was selected and the current was adjusted in accordance with the manufacturer's recommendation. Subsequently, the flame was lit and the flow of fuel and oxidant was regulated. The burner and nebulizer flow rate were adjusted for maximum percent absorption and stability. The photometer was balanced and the standards were run to construct a calibration curve by plotting the absorbance against concentrations. The treated samples were aspirated through the nebuliser into the flame and their concentrations were determined directly from the readout [27]. This was repeated for the Standards, Blank and the samples.

2.3. *Recovery and reproducibility studies*

To check the sensitivity and efficiency of the method used in the chemical analysis, recovery and reproducibility studies were conducted. In the recovery studies, known certified concentrations of Ag was determined using the flame AAS. The reproducibility studies was also carried out by analysing double distilled water containing 2 mg/l of Ag using the flame AAS. Similarly, to determine the efficiency of the digestion process, 0.5 μg/l Ag solution was used to spike the sample. The concentration in the spiked sample was determined as described above.

3. Results and discussion

3.1. *Recovery and reproducibility studies*

The percentage recoveries of silver from double distilled water are between 95 and 105% with a standard deviation of between 0.00 and 0.03. This suggests that the method used is very efficient. The results from the reproducibility studies also gave percentage recovery = >99.3; standard deviation = 0.017–0.029; standard error = 0.005–0.009; coefficient of variation = 1.6–1.7% it is clear that the methods used in analysing Ag samples is reproducible.

3.2. *Silver concentrations in the Pra and Eture estuaries*

Waste products from photographic studios, chemical laboratories and mining operations are the major sources of anthropogenic silver in the Ghanaian estuarine environment. We know this because Tim (2005) [28] reported in a study that the concentration of silver in the Atlantic Ocean which is in direct contact with these estuaries is very low [28,29].

Thus waste discharges from anthropogenic activities performed along the Kakum river and the Eture estuary strongly contributed to this high concentration reported in these ecosystems.

The results of this study suggest that the concentration of silver displayed high variability in the estuaries except for the first week of February and March, respectively (Table 1). The periodic influx of seawater due to the high tides resulted in the subsequent dilution of the mineral content and the high variability between the highest and lowest values. In order to reflect the real silver levels in the estuaries, the samples were span through the two extreme levels and the mean values were used to discuss the present results.

River Pra estuary consistently showed the least concentration of the total silver. In the Eture estuary, higher concentrations of silver were observed in the western tributary (Suruwe) than in the eastern tributary (Kakum) streams but the merging point of the two streams displayed the highest values. The highest concentrations of silver were observed in the first week of February and March, respectively. This was due to the high tides encountered during the time of sampling. This means that the sea that has very low silver concentration [28], had diluted the estuaries at the time of sampling and that represented the lowest extreme. The fairly stable high levels of silver realised on the sixth and tenth analyses (that is the fourth weeks in both February and March) represented the highest extreme.

The most recent measurements of silver in marine and fresh water ecosystems, based on clean techniques, showed levels of about 0.01 μg/l for pristine and unpolluted areas and 0.01–0.1 μg/l

Table 1. Total concentration of silver reported in the investigated estuaries.

Sampling dates	River Pra estuary/mg/l	Suruwe stream /mg/l	Kakum stream /mg/l	Eture estuary /mg/l
19 Jan 2007	0.0618 ± 0.0055	0.0711 ± 0.0008	0.0774 ± 0.0031	0.1188 ± 0.0097
26 Jan 2007	0.0565 ± 0.0002	0.0739 ± 0.0020	0.0763 ± 0.0020	0.1052 ± 0.0039
02 Feb 2007	0.0499 ± 0.0065	0.0701 ± 0.0018	0.0711 ± 0.0032	0.1001 ± 0.0090
09 Feb 2007	0.0549 ± 0.0015	0.0704 ± 0.0015	0.0741 ± 0.0002	0.1039 ± 0.0052
16 Feb 2007	0.0599 ± 0.0036	0.0709 ± 0.0010	0.0759 ± 0.0016	0.1051 ± 0.0040
23 Feb 2007	0.0621 ± 0.0058	0.0741 ± 0.0022	0.0761 ± 0.0018	0.1171 ± 0.0080
02 Mar 2007	0.0461 ± 0.0103	0.0698 ± 0.0021	0.0701 ± 0.0042	0.0998 ± 0.0093
09 Mar 2007	0.0513 ± 0.0051	0.0716 ± 0.0003	0.0713 ± 0.0030	0.1012 ± 0.0079
16 Mar 2007	0.0609 ± 0.0046	0.0714 ± 0.0005	0.0732 ± 0.0011	0.1197 ± 0.0106
23 Mar 2007	0.0601 ± 0.0038	0.0755 ± 0.0036	0.0771 ± 0.0028	0.1201 ± 0.0110
Mean of means	0.05635	0.07188	0.07426	0.1091
Standard deviation	0.0056	0.0019	0.0027	0.0087

Table 2. Recommended criteria for the protection of marine and freshwater life.

Environment	Criteria as total silver	Conditions
Freshwater	0.05 μg/l as a 30-day mean	Hardness less than or equal to 100 mg/l
Freshwater	0.1 μg/l maximum	Hardness less than or equal to 100 mg/l
Freshwater	1.5 μg/l as a 30-day mean	Hardness greater than 100 mg/l
Freshwater	3.0 μg/l maximum	Hardness greater than 100 mg/l
Marine water	1.5 μg/l as a 30-day mean	Open coast and estuaries
Marine water	3.0 μg/l maximum	Open coast and estuaries

By kind courtesy of Environmental Protection Division, Ministry of Environment, Government of British Columbia [42].

in urban and industrialized areas (Table 2). Previous studies [25,26,30–36] suggested that different ecosystems would not be conducive for organisms inhabiting these systems.

Smith and Carson (1977) [25], reported that the estimated loss of silver to the environment in the USA was 2.5 million kilograms, mostly to terrestrial and aquatic ecosystems; with the photographic industry alone accounting for about 47% of all silver lost to the environment that enters the aquatic environment from the photographic industry, mine tailing, and electroplating. Silver thiosulphate complexes in photographic developing solutions that photofinishers discarded were cited as the main source of silver contamination of water.

In Ghana the situation is not so different, from the scenario reported by Smith and Carson (1977) [25]. The leaching of silver from the soil to groundwater is evidenced in the results obtained for the Pra River as the mean value of 0.0563 mg/l silver level appears to be also very high. Thus the Pra River which meanders through many mining areas has high silver level mainly due to leaching from manganese rich lands as well as waste products from gold mining operations (mine tailings) [37].

Silver also may leach from soils into groundwater, with the leaching rate increasing with decreasing pH and increasing drainage [38]. Silver is known to adsorb onto manganese dioxide, ferric compounds and clay minerals, and these result in higher concentrations in sediments than in the water column [26].

Under reducing conditions, the adsorbed silver in sediments may be released and subsequently reduced to metallic silver, or it may combine with reduced sulphur to form the insoluble silver sulphide [26].

In marine ecosystems, where the silver concentration reached values of 0.5–4.5 μg/l, most species displayed high concentrations of silver in their tissues. Moreover, silver negatively influenced the growth of algae, clams, oysters, snails, daphnids, amphipods, and trout; moulting in mayflies and histopathology in mussels [13]. Wood et al. (1996a) [13], reported that the acute toxicity of silver for aquatic organisms varies drastically in relation to the chemical form and the availability of free ionic silver. In natural aquatic systems, ionic silver rapidly forms complexes that are absorbed by dissolved and suspended organic matter [20,39–41]. The levels obtained in this study are quite high as compared with the standards [42] and this is worrisome.

The US EPA recommends that total recoverable silver for freshwater aquatic life is 1.2 to 13.0 μg/l and a total recoverable silver of 2.3 μg/l is needed to protect marine life. With the mean total recoverable silver from the River Pra Estuary, Suruwe, (Eture west stream) Kakum (east stream) and the Eture estuary (the merging point) being 0.0563 mg/l, 0.0728 mg/l, 0.0735 mg/l and 0.1138 mg/l, respectively, indicating an elevated silver levels. Meaning that, species of younger organisms supported by these water bodies stand the chance of being endangered and urgent remedial steps therefore need to be taken to curtail the situation.

4. Conclusion

The results of this study reported that the estuaries of River Pra and Eture showed these unexpectedly high total recoverable silver content due to the strong anthropogenic activities reported along these ecosystems. This situation represents a potential risk for human health and the silver-sensitive and vulnerable species living in the Pra and Eture estuaries. These results suggest the necessity of an action plan to improve and the monitor of the quality of these important estuaries.

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