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Statistical Evaluation of Environmental Contamination, Distribution and Source Assessment of Heavy Metals (Aluminum, Arsenic, Cadmium, and Mercury) in Some Lagoons and an Estuary Along the Coastal Belt of Ghana

C. K. Adokoh · E. A. Obodai · D. K. Essumang · Y. Serfor-Armah · B. J. B. Nyarko · A. Asabere-Ameyaw

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Abstract An environmental pollution investigation was carried out to determine the concentrations of aluminum (Al), arsenic (As), cadmium (Cd), and mercury (Hg) (heavy metals) in the surface water and river water bed sediments of lagoons and estuaries along the coastal belt of Ghana. The study assessed the environmental pollution situation and evaluated their sources and distribution of these metals. The total concentrations of Al, As, Cd, and Hg were determined by the neutron activation analysis technique. Water and sediment samples were collected from the Benya, Fosu, and Narkwa lagoons in the Central Region and from the Pra estuary in the western part of Ghana. Some indices, such as contamination factor, pollution-load index, contamination degree, and geoaccumulation index, were used to assess eco-environmental quality of the sampling sites. The analysis indicated that the Fosu lagoon was fairly polluted with Cd. The investigation indicated a highly localized distribution pattern closely associated with the two pollution sources (garbage/solid waste dumps and industrial activities) along the coastal belt. The resulting environmental deterioration required a concerted evaluative effort by all stakeholders.

C. K. Adokoh (✉)
Chemistry Department, University of Johannesburg, Auckland Park Campus, Johannesburg 2006, South Africa
e-mail: christattom@yahoo.com

C. K. Adokoh · D. K. Essumang
Chemistry Department, School of Physical Sciences, University of Cape Coast, Cape Coast, Ghana

E. A. Obodai
Department of Fisheries, University of Cape Coast, Cape Coast, Ghana

D. K. Essumang
Department Biotechnology, Chemistry and Environmental Engineering, Aalborg University, Alborg, Denmark

Y. Serfor-Armah · B. J. B. Nyarko
Ghana Atomic Energy Commission Ghana (GAEC), Legon Accra, Ghana

A. Asabere-Ameyaw
University of Education Wineba, Winneba, Ghana

Heavy-metal contamination in the environment causes serious water-quality problems in many fast-growing cities. Improvements in water and sanitation infrastructure have not kept pace with population growth and urbanization in most developing countries (Mintz and Baier 2000). The quality of river water is affected by daily, seasonal, and climatic rhythms. The purity of ground water is also dependent on the physical, chemical, and biological cycles of its surrounding environment (Svobodová et al. 1993).

Heavy metals occur as natural constituents of the earth's crust and are known persistent environmental contaminants that cannot be degraded or destroyed. These contaminants enter biological systems through food, air, and water and subsequently bioaccumulate over time (Lenntech 2004; United Nations 2004). The metals enter water bodies from a variety of sources, such as rocks and soils, decomposing organic matter, fallout of atmospheric particulates, and anthropogenic activities, including the discharge of various treated and untreated liquid wastes (Olayinka 2004). Some of these heavy metals are mercury (Hg), arsenic (As), and cadmium (Cd).

The demand for adequate quality water to satisfy domestic, industrial, and agricultural needs in the developing world has increased during the last decade. As a result, it has become imperative to identify the various sources of contaminants and assess the quality of the lagoons, estuaries, river, and ground waters by quantifying the amount

of pollutants deposited in these water bodies. This will provide the scientific basis for finding appropriate remedies to the contamination problems that confront the basin and their inherent impacts on the ecosystem. A number of factors influence water chemistry: as proposed by Gibbs (1970), rock weathering, atmospheric precipitation, evaporation, and crystallization control the chemistry of water. The influence of geology on water quality has been widely recognized (Gibbs 1970; Langmuir 1997; Lester and Birkett 1999). The influence of soils on water quality is very complex and can be ascribed to the processes controlling the exchange of chemicals between the soil and water (Hesterberg 1998).

The coastline of Ghana, which forms part of the Gulf of Guinea, is dotted with approximately 98 lagoons and 10 estuaries (Intergovernmental Oceanographic Commission [IOC] 1998; Obodai 1997). There are two main types of lagoons: the "open" and the "close." The former is always opened to the sea and is associated with rivers that flow year round, whereas the latter remains closed to the sea for most of the year. The quality of both types, particularly the "closed," of which Fosu and Narkwa lagoons are two examples, are often influenced by inland activities. These serve as traps for materials brought from inland as a result of flocculation, precipitation, and sedimentation or by careless direct waste disposal into the lagoon (IOC 1998; Gilbert et al. 2006).

Recent research by Gilbert et al. (2006) on heavy-metal pollution in sediments of the Fosu Lagoon observed significantly high concentrations at certain locations on the lagoon adjacent to human settlements, but only copper (Cu) and zinc (Zn) were measured. Another study by Essumang (2009), reported a high concentration of heavy metals in parts of manta ray caught along the coastline of Ghana. This demonstrated the extent of pollution in those bodies of water. Despite the potentially direct negative impact, current chemical data pertinent to this crisis are still lacking on these lagoons and estuaries. The limited available data in the literature are valuable. This study investigated the levels of aluminium (Al), As, Cd, and Hg pollution of four water bodies identified to be the main heavy-metal pollution pathway to the sea in the central and western regions of Ghana. Some of these water bodies pass through mining communities, whereas others pass through heavily populated areas coupled with busy commercial activities and industrialized towns, thereby given these water bodies higher pollution loads.

Methodology

Study Areas

The Pra Estuary is the westernmost and the largest of the three principal rivers that drain into the area south of the Volta divide. The river rises in the Kwahu Plateau near Mpraeso and flows 240 km southward to enter the Gulf of Guinea, east of Takoradi at Shama. The Pra Estuary is discharges into atlantic ocean at 5°1′N, 2°22′E (Fisheries and Aquaculture Department 1990). The area provides the second largest discharge of fresh water into the gulf of Guinea from Ghana. The flow of water is double maximum as a result of the rainfall pattern at the catchments area. The bay curves an average of 2.5 km^2 and has an average depth of 1.5 m; hence, the volume of the system is 3.75 × 106 m^3.

Benya Lagoon is one of the many lagoons located along the Gulf of Guinea in the city of Elmina. It is approximately 62 m and is located in Ghana (N 5°5′ 0″ W 1°22′ 0″ 5.08333/–1.36667; GeoNameId: 2303164) and is hydrographic (Fisheries and Aquaculture Department [FAD] 1990). *Fosu Lagoon* is one of the many lagoons located along the Gulf of Guinea in the city of Cape Coast, Ghana, in West Africa. Cape Coast lies along the Atlantic Ocean and is on the longitude 1°15′ west of the Greenwich Meridian and latitude 5°5′ north of the Equator (Gilbert et al. 2006). *Narkwa Lagoon* is also located in Ghana –5°12'N; 0°55'W and is found in central region close to Saltpong Beach (Fisheries and Aquaculture Department 1990) (Fig. 1).

Sample Collection

Water Samples

Two water samples, A and B, were collected from each of the four habitats previously described. Each "A" sample was extracted approximately 1 km distant from each "B" sample. Samples contents were placed into 1 litre polyethylene bottles. Before use, the bottles were washed with acetone to remove any grease and with dilute nitric acid to dissolve any metal. Finally, they were rinsed with distilled water. Each bottle was labeled and kept in a refrigerator. Four sets of samples were gathered during each of the two major seasons in Ghana (*i.e.*, dry season and wet season). In all, 32 water samples were collected for this study. These were subsequently transported to Ghana Atomic Energy Commission (GAEC), Accra, for analysis.

Sediment

In addition to water samples, 32 sediment samples were collected for heavy-metal determination. Collection points for samples A and B were separated by approximately 1 km. Samples were extracted from the rivers and lakes at 100 meters from the bank or shore. Approximately 200 g sediment were collected per sample by way of a dedicated plastic scoop. These were placed into black polyethylene containers. The containers were labeled according to the

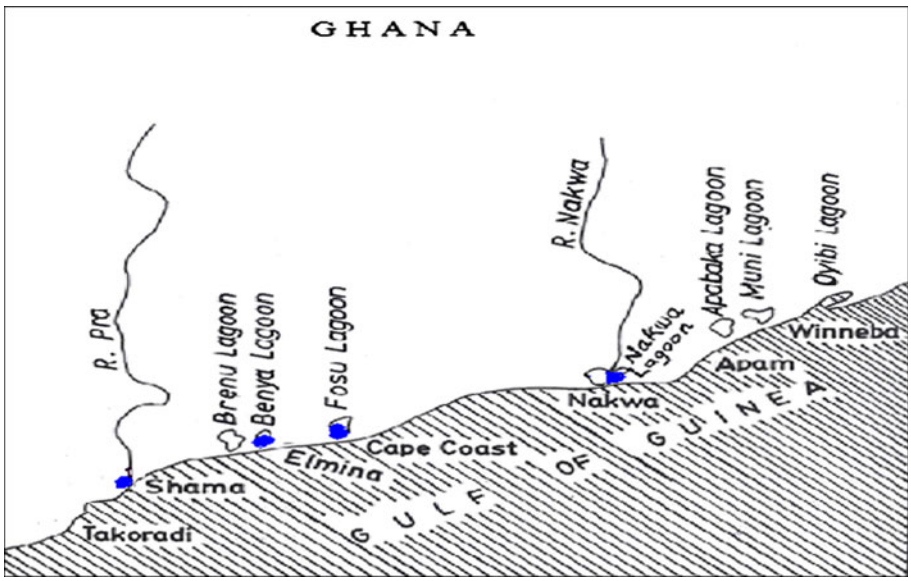

Fig. 1 Map of the principal lagoons of Ghana. Sampling sites are marked in blue (Color figure online)

name of the sampling site. Samples were further prepared for laboratory analysis by air drying.

Sample Preparation

Exactly 0.5 ml of each water sample was pipetted (using calibrated Eppendorf tip ejector pipette) into clean, pre-weighed 1.5-ml rabbit capsules (i.e., sample vials), reweighed, and heat-sealed using a soldering rod. Four of these sample vials were also placed into a 7.0-ml rabbit capsule and heat-sealed. Two replicates of each sample were prepared.

Each sediment sample was air-dried for 3 days in a clean environment. Other organic debris, such as shells, stones, and microorganisms, were hand-picked from the samples. The samples were crushed using an agate mortar and pestle, sieved using an 85-μm mesh size USA standard sieve, and homogenized. Approximately 100 mg of samples were weighed into a clean polyethylene film. The films were wrapped and heat-sealed. These samples were labeled according to the sampling site. The samples were packed into 7-ml rabbit capsules for irradiation. Certified reference material IAEA-405 estuarine sediment was treated the same as the samples. The bagged samples were then packed into rabbit capsules and heat-sealed for radiation (Essumang et al. 2008a).

Sample Analysis

Neutron activation analysis (NAA) was used to quantify the concentration of heavy metals using thermal neutrons from a low-flux Am-Be radioisotope. Theoretically, NAA is based on the measurement of characteristic gamma-rays from a radionuclide formed from the specific neutron reaction, which can be used to measure the quantity of an element using the usual radioactive decay law (Tolgyessy and Kyrs 1989; Adomako et al. 2008).

Irradiation Source

The irradiation source was a 20-curie Am-Be radioactive neutron source. The thermal neutron flux at the irradiation site was 1.124×10^5 ns^{-1} cm^{-2} (Nyarko et al. 2004; Essumang et al. 2008a, b).

Sample Irradiation

Each of the samples was sent by the pneumatic transfer system into the Am-Be source and irradiate for 1 h. This was followed by a 12-h cool off period (Nyarko et al. 2004; Essumang et al. 2008a, b). The samples were then tested for 600 s to determine remaining radiation intensities. This information was saved for further analysis.

Data Processing

Counting of signals was performed by an ENERTEC high-germanium detector of 3000 (+ve) bias and a resolution of 2.55 keV for a 1332 keV photo peak of cobalt-60. Micro-soft window based software MAESTRO-32 provided by ORTEC® was used for spectrum analysis (Qualitative and quantitative analysis) (Serfor-Armah et al. 2001; Adomako et al. 2008; Essumang et al., 2008a, b).

Validation of Analytical Method

The accuracy of the analytical method was evaluated using the standard reference material (SRM) IAEA-405 (trace

elements and methyl mercury in estuarine sediment) obtained from the International Atomic Energy Agency (IAEA). IAEA-SOIL-7 was also weighed and sealed into polyethylene bags for the quantitative analysis of the elements in the soil and water samples. Two replicates of the SRM were prepared and packaged in a similar manner. The analytical values of the reference material obtained from this study were compared with the recommended values (in ppm).

Statistical Analysis

Pollution-load indices (PLIs), contamination factor (CF), and geoaccumulation index (Igeo) analysis were used to assess and quantify the pollution levels of the monitored elements in sediments of the study area. In the case of water samples, PLIs, CF, and contamination degree (CD) were also used to assess and quantify the pollution levels of the element in the sampling sites. The mean concentrations, SDs, and correlation matrices for sediment data were determined using SPSS version 16 software provide by IBM cooperation. According to Tomlinson et al. (1980), indices enable the quality of the environment to be easily understood by a nonspecialist. Descriptions of CF, PLI, and Igeo classes and their pollution intensities are listed in Table 1. CF, which gives an indication of the presence and concentration of particular contaminants, was computed for the soils using their average elemental concentrations and the maximum corresponding values in the world shale values of abundance metal in the earth crust (0.4, 0.3,13, and 80,000 ppm for Hg, Cd, As, and Al, respectively) (Turekian and Wedepohl 1961). The CFs was calculated using the following equation:

$$\mathrm{CF} = \text{element concentration in soil}/\text{background value in the earth crust.}$$

The resultant CFs of the elements were used to compute the PLIs as a measure of the mutual pollution effect on the soils (Tomlinson et al. 1980; Cabrera et al. 1999; Adomako et al. 2008) as follows:

$$\text{PLI sampling site} = \sqrt{[3]}\mathrm{CF_{Pt}} \times \mathrm{CF_{Pd}} \times \mathrm{CF_{Rh}}.$$

The Igeo indexing approach was used to quantify the degree of anthropogenic contamination in soils and to compare the different elements (Yaqin et al. 2008; Okweye et al. 2009). The Igeo for each element was calculated using the following formula:

$$\mathrm{Igeo} = \log_2(\mathrm{Cn}/1.5 \times \mathrm{Bn}),$$

where Igeo is geoaccumulation index, Cn is measured element concentration in soil sample, and Bn is the geochemical background value in world average shale or maximum corresponding values in the world average abundance of metal in the earth crust reported (Greenwood and Earnshaw 1984; Karbassi et al. 2008).

Table 1 CF, PLIs, and Igeo values with their pollution grade and intensities

	Grade	Intensity
CF value		
<1.2	I	Unpolluted area
1.2–2	II	Lightly polluted area
2–3	III	Moderately polluted area
>3	IV	Heavily polluted area
PLI value		
<0	1	Unpolluted
1	2	Baseline levels of pollutant present
>100	3	Progressive deterioration of environment
Igeo value		
<0	1	Unpolluted
>0–1	2	Unpolluted to moderately polluted
>1–2	3	Moderately polluted
>2–3	4	Moderately to strongly polluted
>3–4	5	Strongly polluted
>4–5	6	Strongly to very strongly polluted
>5	7	Very strongly polluted

Source: Nyarko et al. (2004) for CF Yaqin et al. (2008); Lokeshwari and Chandrappa (2007); Grzebisz et al. (2002) for Igeo and Tomlinson et al. (1980); Nyarko et al. (2006); Angulo (1999) for PLI

For water samples, CD was used to assess the pollution level of monitored elements. The approach of Teng et al. (2004) was followed using this equation:

$$\mathrm{CD} = \sum \mathrm{Cf}_i,$$

where CD is the contamination degree, $Cf_{\mathrm{i}} = (C_n/C_b) - 1$ (where Cf_i is the contamination factor for the ith element), C_n is the analytical value of the ith element, and C_b is the upper permissible limit of element in water by WHO (0.001 mg/l/d for Hg, 0.003 mg/l/d for Cd, 0.01 μg/g/d for As, and 0.2 mg/l/d for Al), which was selected for calculation of the CD of the water samples (World Health Organization 2004).

EF Calculations as Evidence of Anthropogenic Origin of Heavy Metals

Because metal loads from natural and anthropogenic sources accumulate together to differentiate the fraction of the element concentration originating from natural sedimentary sources from the anthropogenic fraction, normalizing tools are needed (Green-Ruiz and Páez-Osuma 2001). To decrease heavy-metal variability caused by grain size and mineralogy of the sediments and to

identify anomalous metal contributions, geochemical normalization has been used with various degrees of success by employing various conservative metals, including Al (Gilbert et al. 2006). Heavy-metal concentrations in the various lagoons and estuary sediments are being normalized using Al as a conservative element to evaluate anthropogenic sources of the metals. The conservativeness of Al in bed sediments received confirmation in this study for the various water bodies studied (see previous text) despite the possible anthropogenic contributions to the lagoons and estuaries. The EF, which is the index of contamination formula for the sediment samples, was calculated using the heavy-metal average concentrations at sites Pra to Keta (target sites), with levels at the Narkwa site taken as baseline. Narkwa is expected to receive the least anthropogenic metal load to the lagoon because it is remote from industrial activities, is not a recreational center, and is the least populated and apparently most environmentally clean settlements in the vicinity of the lagoon.

$$\mathrm{EF} = \frac{[M]/[\mathrm{Al}]_{\mathrm{target-site}}}{[M]/[\mathrm{Al}]_{\mathrm{baseline}}}$$

where $[M]$ is metal concentration studied.

Results and Discussion

Quality Assurance

The accuracy and precision of the analytical technique neutron activation analysis (NAA) was assessed by simultaneous activation of CRM IAEA-405 (trace elements and methyl mercury in estuarine sediment). Table 2 lists the analytical results obtained at GHARR-1 laboratory for the reference material compared with the recommended values. The values compared favourably well with the recommended values for Hg, Cd, As, and Al, with bias <6%. Precision was calculated as a percentage relative SD of two replicate samples of the prepared standard and was found to be <5% with a percentage recovery of approximately 97% (Table 2).

Element Concentrations in Soil and Water Samples

The average concentrations of the examined elements are listed in Table 3. Hg concentration ranged from 0.002 and 0.017 μg/g, with the highest value being recorded at Pra estuary (0.017 μg/g). Cd concentration ranged between 0.02 and 0.526 μg/g; As concentration between 0.132 and 0.728 μg/g; and Al concentration between 16.678 and 60.407 μg/g. The highest concentration of Cd (0.526 μg/g) was recorded in Fosu Lagoon sediment. Al concentration was highest at Benya Lagoon followed by Fosu Lagoon. In general, the concentration of metals did not show any clear difference among the four sites.

The concentrations of these elements fall into the category regarded as good soil quality according to the reference ministerie van volkshuisvesting Ruimitelijke Ordening Milieubewer (VROM 1983) and The Canadian interim cadmium marine/estuarine sediment guideline for the protection of aquatic lives (0.7 mg/kg); all of the Cd levels in the present study were lower than that. The increased concentration of Hg (0.017 μg/g dry weight) recorded at Pra estuary may be attributed to the small-scale mining activities around the estuary upstream.

Water samples generally had lower values compared with sediment from the same sampling point. Hg concentration ranged between 0.0002 and 0.005 μg/l; Cd concentration between 0.022 and 0.051 μg/l; As concentration between 0.002 and 0.231 μg/l; and Al concentration between 0.964 and 1.706 μg/l (Table 3). Hg concentration across the studied sites was generally low compared with Al, which had the highest concentration followed by As and then Cd. This is not surprising because Al is the third most common element in the earth crust; thus, it is a major constituent of most soils. As ranks 52nd and Cd in trace form in the earth crust. In contrast, Hg ranks 67th (trace) in the list of naturally occurring elements in crustal rocks. Appreciable concentrations found in the environment typically originate from anthropogenic sources.

In general, the concentrations of elements in sediment samples were greater than those in water column. This might be due to the fact that metals can be either

Table 2 Result of quality-control analysis of the method

Analyte	This work		Recommended			
	1	2	Mean	Value	95% CI	% Recovery
Hg	0.79 ± 0.02	0.789 ± 0.02	0.789	0.81	0.77–0.85	97.41
Cd	0.70 ± 0.01	0.71 ± 0.02	0.71	0.73	0.68–0.78	97.26
As	23.1 ± 0.15	22.87 ± 0.15	22.98	23.6	22.9–24.3	97.37
Al	75641 ± 25.1	76142 ± 25.35	75891.5	77900	72700–83100	97.42

1 and 2 values are two repricates of standards runed for quality control analysis

Table 3 Average concentration of elements from the sampling sites

Sampling		Season	Heavy-metal concentration (±SD) site			
			Hg	Cd	As	Al
Pra Estuary (1)	*Sediment (μg/g dry weight)*					
	A	Dry	0.005 ± 0.000	.056 ± 0.009	0.106 ± 0.00	40.02 ± 0.989
		Wet	0.008 ± 0.006	0.020 ± 0.010	0.378 ± 0.021	33.155 ± 1.52
		Mean	0.007 ± 0.002	0.038 ± 0.025	0.378 ± 0.192	36.588 ± 4.854
	B	Dry	0.032 ± 0.00	0.069 ± 0.017	0.007 ± 0.002	39.842 ± 0.398
		Wet	0.001 ± 0.00	0.140 ± 0.000	0.025 ± 0.011	44.762 ± 59.451
		Mean	0.017 ± 0.022	0.105 ± 0.050	0.016 ± 0.013	42.302 ± 3.479
	Water (μg/l)					
	A	Dry	ND	0.033 ± 0.000	0.020 ± 0.014	2.292 ± 1.015
		Wet	ND	0.006 ± 0.000	0.019 ± 0.002	0.579 ± 0.028
		Mean	ND	0.019 ± 0.019	0.019 ± 0.001	1.436 ± 1.211
	B	Dry	0.003 ± 0.002	0.025 ± 0.033	0.020 ± 0.000	1.316 ± 0.063
		Wet	0.001 ± 0.000	0.006 ± 0.000	0.011 ± 0.001	0.611 ± 0.041
		Mean	0.002 ± 0.001	0.016 ± 0.013	0.016 ± 0.006	0.964 ± 0.498
Benya Lagoon (2)	*Sediment (μg/g dry weight)*					
	A	Dry	ND	0.017 ± 0.00	0.010 ± 0.00	77.707 ± 5.58
		Wet	0.012 ± 0.008	0.043 ± 0.00	0.067 ± 0.010	43.114 ± 42.199
		Mean	0.012 ± 0.008	0.030 ± 0.018	0.039 ± 0.040	60.407 ± 24.456
	B	Dry	0.002 ± 0.00	0.030 ± 0.00	0.122 ± 0.00	38.876 ± 9.115
		Wet	0.003 ± 0.00	0.027 ± 0.018	0.308 ± 0.00	48.954 ± 7.645
		Mean	0.003 ± 0.001	0.029 ± 0.002	0.215 ± 0.131	43.915 ± 7.126
	Water (μg/l)					
	A	Dry	0.004 ± 0.001	0.024 ± 0.026	0.125 ± 0.038	2.653 ± 1.691
		Wet	0.002 ± 0.001	0.038 ± 0.027	0.086 ± 0.015	0.759 ± 0.082
		Mean	0.003 ± 0.001	0.031 ± 0.009	0.106 ± 0.028	1.706 ± 1.339
	B	Dry	0.005 ± 0.000	0.051 ± 0.021	ND	2.323 ± 0.999
		Wet	ND	0.050 ± 0.006	ND	0.703 ± 0.006
		Mean	0.005 ± 0.000	0.051 ± 0.001	ND	1.513 ± 1.146
Fosu Lagoon (3)	*Sediment (μg/g dry weight)*					
	A	Dry	0.006 ± 0.000	0.782 ± 0.028	0.08 ± 0.007	66.512 ± 41.24
		Wet	0.012 ± 0.001	0.27 ± 0.019	0.735 ± 0.986	22.74 ± 19.16
		Mean	0.009 ± 0.004	0.526 ± 0.362	0.408 ± 0.463	44.626 ± 30.951
	B	Dry	0.019 ± 0.002	0.754 ± 0.139	0.910 ± 0.000	63.808 ± 39.8
		Wet	0.008 ± 0.000	0.227 ± 0.037	0.148 ± 0.004	49.098 ± 46.43
		Mean	0.014 ± 0.008	0.491 ± 0.373	0.529 ± 0.539	56.453 ± 10.402
	Water (μg/l)					
	A	Dry	0.002 ± 0.000	0.024 ± 0.017	ND	2.242 ± 1.051
		Wet	0.001 ± 0.00	0.026 ± 0.019	ND	0.712 ± 0.044
		Mean	0.002 ± 0.001	0.025 ± 0.001	ND	1.477 ± 1.081
	B	Dry	0.002 ± 0.001	0.052 ± 0.023	ND	1.533 ± 0.166
		Wet	ND	0.019 ± 0.006	ND	0.812 ± 0.063
		Mean	0.002 ± 0.001	0.036 ± 0.023	ND	1.173 ± 0.509

Table 3 continued

Sampling		Season	Heavy-metal concentration (±SD) site			
			Hg	Cd	As	Al
Narkwa Lagoon (4)	*Sediment (μg/g dry weight)*					
	A	Dry	0.004 ± 0.003	ND	0.023 ± 0.00	19.604 ± 1.20
		Wet	0.012 ± 0.00	0.041 ± 0.049	1.183 ± 0.00	13.752 ± 0.139
		Mean	0.008 ± 0.006	0.041 ± 0.049	0.603 ± 0.820	16.678 ± 4.138
	B	Dry	0.001 ± 0.00	0.038 ± 0.00	0.069 ± 0.00	24.452 ± 11.315
		Wet	.005 ± 0.00	0.011 ± 0.00	0.112 ± 0.00	11.398 ± 3.468
		Mean	0.003 ± 0.003	0.0245 ± 0.019	0.091 ± 0.030	17.925 ± 9.231
	Water (μg/l)					
	A	Dry	0.002 ± 0.00	0.036 ± 0.004	ND	1.411 ± 0.001
		Wet	0.001 ± 0.00	0.049 ± 0.002	0.002 ± 0.00	0.813 ± 0.036
		Mean	0.002 ± 0.001	0.0425 ± 0.009	0.002 ± 0.00	1.112 ± 0.423
	B	Dry	0.003 ± 0.00	0.001 ± 0.00	0.068 ± 0.011	1.293 ± 0.334
		Wet	ND	0.047 ± 0.011	0.019 ± 0.007	0.875 ± 0.033
		Mean	0.003 ± 0.00	0.024 ± 0.033	0.044 ± 0.035	1.084 ± 0.296

ND not determined

transported with the water or suspended within the riverbed bottom sediment (Garbarino et al. 1995; Adomako et al. 2008). Also, suspended metallic chemical solids are stored in riverbed sediments after they aggregate to form large particles that are denser than water. These settle from the water when the river's flow is not sufficient to keep them suspended (Garbarino et al. 1995, Adomako et al. 2008). Cd concentrations (0.526 to 0.491 μg/g dry weight) were highest in samples from Fosu Lagoon. The relatively high prevalence of Cd distribution at this site seems to be mainly attributable to leaching of the metal from garbage and solid waste dumps. Atmospheric deposition from garbage incineration, volatile petroleum products from nearby filling stations, and smoke from vehicles may also contribute to Cd enrichment in sediments. Predominant industrial activities include car fittings, welding, painting, *etc*. Careless disposal of kinds of waste from these activities into the lagoon is the norm in the locality. Disposal of waste from nickel/cadmium batteries, Cd-containing alloys, foils, oils, and electrical equipment are all possible sources of Cd to the lagoon. In addition, most drivers use this site as their car-washing base, and that may be another likely source of Cd to the lagoon because most detergents contain Cd. Gilbert et al. (2006), reported a similarly high average Cd level of 0.70 mg/kg.

The highest Al concentrations were found in Benya Lagoon (60.407 μg/g dry weight) followed by Fosu Lagoon (56.453 μg/g). Benya Lagoon is one of the busiest fishing harbors in Ghana, and it is believed that the numerous activities on the lagoon contribute immensely to the high Al concentrations. Leaching of Al metal from garbage and solid waste dumps may contribute strongly to this effect.

Pollution Load Assessment

CF, PLI, Igeo, and EF, as well as CD analysis, were used to assess and quantify pollution levels of the monitored elements in soils and water of the study area. All of the sampling points had CFs less than unity for almost all of the elements studied. An exception was the Cd CF value at Fosu Lagoon (1.694) (Table 4). This value is an indication of a lightly polluted area. The sampling points show variations in the PLI values. However, all of the sampling points have PLI values < 1.0 (Table 4). The highest and lowest PLI values occurred at Fosu lagoon and Benya Lagoon: 0.032 and 0.010 for sediment, respectively. The water samples generally had low PLI values, indicating an unpolluted status of the water bodies. Generally, Al PLI values were small even though their concentration was the highest (Table 4).

Igeo values (Table 5) recorded for all the elements studied were <1.0 or negative, indicating practically unpolluted soils, except for Fosu Lagoon, where the CF Cd value was 1.694, which recorded a positive Igeo value (0.176), and was even less than unity.

EF values of the studied metals followed same the trend seen with other pollution indices as previously described, i.e., almost all of study areas had values <3, indicating depletion to minimal enrichment. The only exception was the Cd EF value at Fosu Lagoon, which was slightly >3 (Table 5).

Table 4 Water bodies and their CFs and PLIs

Sampling site	Sample	CFs				
		Hg	Cd	As	Al	PLI
Pra Estuary	Sediment	0.029	0.237	0.010	0.001	0.014
	Water	0.005	0.058	0.001	1.5E−05	0.002
Benya Lagoon	Sediment	0.014	0.097	0.010	0.001	0.010
	Water	0.009	0.136	0.008	2.01E−05	0.004
Fosu Lagoon	Sediment	0.028	1.694	0.036	0.001	0.032
	Water	0.004	0.108	0	1.7E−05	0.009
Narkwa Lagoon	Sediment	0.014	0.1	0.027	0.0002	0.023
	Water	0.005	0.111	0.002	0.00001	0.009

Table 5 Igeo and EFs of the sediments for heavy metal at the study areas

Sampling site	Hg	Cd	As	Al
Igeo				
Pra estuary	−5.705	−2.659	−7.239	−11.571
Benya Lagoon	−6.726	−3.943	−7.265	−11.167
Fosu Lagoon	−5.737	0.176	−5.380	−11.213
Narkwa Lagoon	−6.769	−3.907	−5.813	−12.759
EFs				
Pra estuary	1.009	0.558	0.534	
Benya Lagoon	0.376	0.173	0.397	
Fosu Lagoon	0.770	3.104	1.512	
Narkwah Lagoon	1.100	0.535	1.271	
Water CD				
Pra estuary	1	4.833	0.75	4.997
Benya Lagoon	2.667	12.583	9.55	7.048
Fosu Lagoon	0.667	9.778	0	5.624
Narkwah Lagoon	1	10.083	1.967	4.49

CD values for water samples were generally low. Benya Lagoon water samples had the highest CD value for all of the metals analyzed (Table 5). The highest Cd CD value recorded (12.583) was at Benya Lagoon followed by Narkwa Lagoon, which had a Cd CD value of 10.083. Comparing CD values, it is clear that the water bodies are polluted with Cd more than any other metal studied, followed by Al, Hg, and As respectively. This situation may certainly be associated with the increasing population growth and developmental activities in the municipality.

Elmina is one of such coastal cities in the country where small-scale informal-sector activities are denominating its economic base. However, these informal ventures are poorly regulated and usually dispose carelessly waste. Most of the activities occur within the vicinity of the Benya Lagoon. The fishing harbor is one of the busiest harbors in Ghana, and commercial activities take place on the lagoon, which directly affects the impact of the heavy-metal accumulation in the lagoon. The high Cd concentration may be due in part to nearby automobile pollution. The other metals may arise from anthropogenic, nonpoint sources, especially through the local fishing industry.

Intersite Correlations

Correlation analyses were performed to determine interrelationships between the various sampling sites and heavy-metal concentrations (Table 6). There was a strong correlation between all sites, with the exception of Pra Estuary and Benya Lagoon (0.501) ($p < 0.01$). The strongest correlation was between Fosu Lagoon and Narkwa Lagoon (0.907) ($p < 0.01$). These correlations suggest a common sink of contamination (Adomako et al. 2008). A common sink of contamination does not imply that the heavy elements all originate from a common source. Hg is not an abundant element in the earth's crust, so its environmental source is almost surely anthropogenic. In contrast, Al is a common crustal element, which indicates it is probably of natural origin. There is weak correlation coefficient for elements in streams of the studied areas, suggesting a poor relationship between water and sediment element concentrations in the lagoons and estuaries (Table 7). All of the elements, except Cd, had positive correlation coefficients (with 90% showing weak positive correlation coefficients). Cd did not correlate with any of the metal studied (Table 7) in riverine water-sediment. The fairly good correlation between the sites is expected in each case because of the similarities of anthropogenic activities

Table 6 Correlation coefficients between the four studied sites ($N = 64$)

	Pra	Benya	Fosu	Narkwa
Pra	1			
Benya	0.501**	1		
Fosu	0.592**	0.697**	1	
Narkwa	0 .761**	0.709**	0.907**	1

** Correlation significant at $p = 0.01$ (one-tailed)

Table 7 A correlation matrix of elements in the soil and water of the study area ($N = 114$)

	Hg	Cd	As	Al
Hg	1			
Cd	0.144	1		
As	0.321**	0.172*	1	
Al	0.266**	0.318**	0.276**	1

Correlation significant at * $p = 0.05$ and ** $p = 0.01$ (one-tailed)

Table 8 Interrelationships between elements in soil and water of the sampling sites ($N = 57$)

	Hg sed	Hg W	Cd sed	Cd W	As sed	As W	Al sed	Al W
Hg sed	1							
Hg W	0.001	1						
Cd sed	0.045	−0.001	1					
Cd W	0.07	0.251*	0.084	1				
As sed	0.242*	−0.006	0.107	0.06	1			
As W	−0.167	0.024	−0.245*	−0.106	−0.171	1		
Al sed	−0.004	0.323**	0.203	−0.165	0.061	0.238*	1	
Al W	−0.069	0.558**	0.158	0.082	−0.231*	0.134	0.392**	1

Sed sediment, *w* water

Correlation significant at * $p = 0.05$, ** $p = 0.01$ (one-tailed)

in these areas. Although these areas are not adjacent to each other, sites in these areas are close to public recreational activities and appear to offer similar heavy metal input into the lagoons and estuaries (mainly attributable to leaching of metal from garbage and solid waste dumps). Similar correlations between water and sediment levels have been reported from several global locations (Karbassi et al. 2008; Gilbert et al. 2006). Cd seems to have a different source because it did not correlate with any of the metals. It may be strongly related to automobile emissions because almost all of the sampling sites are situated along highways. Other minor potential sources of Cd are atmospheric deposition from garbage incineration and volatile petroleum products from nearby filling stations (Gilbert et al. 2006).

There are weak correlation coefficients for elements in water-sediment in streams of the study area as follows: Al in water correlated with Al in sediment (0.392) at all sites ($p < 0.01$); As in water correlate with Al in sediment (0.238) ($p < 0.05$) at all sites; and Al in sediment correlated with Hg in water (0.242) all at sites ($p < 0.05$). These values suggest a fairly strong relationship between water and sediment element concentrations in the lagoons and estuaries (Table 8). It can be inferred that water current aided in the interactions between water column and bed sediments. The correlation between element concentrations in water and sediment indicates a possible action of sediments as a secondary pollution source. Two negative correlations were recorded between As in water and Cd in sediment and between Al in water and As in water all at ($p < 0.05$). This suggests that there is a different anthropogenic source of Al and As in the water column (Table 8).

Conclusion

The results of this study demonstrate that the levels of certain heavy metals, as quantified in river and lagoon sediments, are greater than those found naturally in the earth's crust. The concentrations of Hg, Cd, and As in all of the soil samples were lower than reference values by VROM (1983) and within the normal soil range reported by Bowen (1979), which is reflected in the overall pollution load indices (<1.0), except for Cd in Fosu lagoon, which showed moderately pollution status. Strong positive site correlations were observed for sediment samples from all of the lagoons and the estuary, indicating a common source of pollution.

Generally, the water quality of the studied water bodies, with respect to the monitored elements, can be described as good as demonstrated by CD values. The results demonstrate that river sediment is the secondary anthropogenic source and that it accumulates heavy metals.

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Appendix

See Table 9.

Table 9

Sampling site		Season	Heavy-metal concentration (±SD)			
			Hg	Cd	As	Al
Pra Estuary	*Sediment (μg/g)*					
	A	Dry 1	ND	0.062	0.106	40.72
		Dry 2	0.055	0.049	ND	39.32
		Mean	0.005 ± 0.00	0.056 ± 0.009	0.106 ± 0.00	40.02 ± 0.989
		Wet 1	0.012	0.027	0.393	34.231
		Wet 2	0.003	0.013	0.363	32.079
		Mean	0.008 ± 0.006	0.020 ± 0.01	0.378 ± 0.021	33.155 ± 1.52
	B	Dry 1	ND	0.081	0.008	39.56
		Dry 2	0.032	0.057	0.005	40.123
		Mean	0.032 ± 0.00	0.069 ± 0.017	0.007 ± 0.002	39.842 ± 0.398
		Wet 1	0.001	0.140	0.033	2.723
		Wet 2	ND	ND	0.017	86.80
		Mean	0.001 ± 0.000	0.140 ± 0.000	0.025 ± 0.011	44.762 ± 59.451
	Water (μg/l)					
	A	Dry 1	ND	0.033	0.030	3.010
		Dry 2	ND	ND	0.010	1.574
		Mean	ND	0.033 ± 0.000	0.020 ± 0.014	2.292 ± 1.015
		Wet 1	ND	0.006	0.021	0.598
		Wet 2	ND	ND	0.018	0.559
		Mean	ND	0.006 ± 0.000	0.019 ± 0.002	0.579 ± 0.028
	B	Dry 1	0.001	0.002	ND	1.271
		Dry 2	0.004	0.048	0.020	1.360
		Mean	0.003 ± 0.002	0.025 ± 0.033	0.020 ± 0.000	1.316 ± 0.063
		Wet 1	0.001	0.006	0.012	0.582
		Wet 2	ND	0.006	0.010	0.640
		Mean	0.001 ± 0.000	0.006 ± 0.000	0.011 ± 0.001	0.611 ± 0.041
Benya Lagoon	*Sediment (μg/g)*					
	A	Dry 1	ND	ND	0.010	81.653
		Dry 2	ND	0.017	ND	73.761
		Mean	ND	0.017 ± 0.00	0.010 ± 0.00	77.707 ± 5.58
		Wet 1	0.017	0.043	0.0745	72.953
		Wet 2	0.006	ND	0.060	13.274
		Mean	0.012 ± 0.008	0.043 ± 0.00	0.067 ± 0.010	43.114 ± 42.199
	B	Dry 1	0.002	0.030	0.122	45.321
		Dry 2	ND	ND	ND	32.43
		Mean	0.002 ± 0.00	0.030 ± 0.00	0.122 ± 0.00	38.876 ± 9.115
		Wet 1	0.003	0.014	ND	43.548
		Wet 2	0.003	0.040	0.308	54.360
		Mean	0.003 ± 0.00	0.027 ± 0.018	0.308 ± 0.00	48.954 ± 7.645
	Water (μg/l)					
	A	Dry 1	0.004	0.042	0.152	3.848
		Dry 2	0.003	0.005	0.097	1.457
		Mean	0.0035 ± 0.001	0.024 ± 0.026	0.125 ± 0.038	2.653 ± 1.691
		Wet 1	0.003	0.057	0.096	0.701
		Wet 2	0.001	0.018	0.075	0.817
		Mean	0.002 ± 0.001	0.038 ± 0.027	0.086 ± 0.015	0.759 ± 0.082
	B	Dry 1	0.005	0.036	ND	3.029
		Dry 2	0.005	0.066	ND	1.616
		Mean	0.005 ± 0.000	0.051 ± 0.021	ND	2.323 ± 0.999
		Wet 1	ND	0.054	ND	0.698
		Wet 2	ND	0.045	ND	0.707
		Mean	ND	0.050 ± 0.006	ND	0.703 ± 0.006

Table 9 continued

Sampling site		Season	Heavy-metal concentration (±SD)			
			Hg	Cd	As	Al
Fosu Lagoon	*Sediment (μg/g)*					
	A	Dry 1	0.006	0.802	0.085	95.671
		Dry 2	ND	0.762	0.075	37.352
		Mean	0.006 ± 0.000	0.782 ± 0.028	0.08 ± 0.007	66.512 ± 41.24
		Wet 1	0.011	0.040	1.432	36.290
		Wet 2	0.012	0.213	0.037	9.191
		Mean	0.012 ± 0.001	0.27 ± 0.019	0.735 ± 0.986	22.74 ± 19.16
	B	Dry 1	0.020	0.852	0.910	91.952
		Dry 2	0.017	0.656	ND	35.665
		Mean	0.019 ± 0.002	0.754 ± 0.139	0.910 ± 0.000	63.808 ± 39.8
		Wet 1	ND	0.200	0.150	81.933
		Wet 2	0.008	0.253	0.145	16.262
		Mean	0.008 ± 0.000	0.227 ± 0.037	0.148 ± 0.004	49.098 ± 46.43
	Water (μg/l)					
	A	Dry 1	0.002	0.012	ND	2.985
		Dry 2	ND	0.036	ND	1.499
		Mean	0.002 ± 0.000	0.024 ± 0.017	ND	2.242 ± 1.051
		Wet 1	0.001	0.039	ND	0.681
		Wet 2	ND	0.012	ND	0.743
		Mean	0.001 ± 0.00	0.026 ± 0.019	ND	0.712 ± 0.044
	B	Dry 1	0.003	0.036	ND	1.415
		Dry 2	0.001	0.068	ND	1.650
		Mean	0.002 ± 0.001	0.052 ± 0.023	ND	1.533 ± 0.166
		Wet 1	ND	0.015	ND	0.767
		Wet 2	ND	0.023	ND	0.856
		Mean	ND	0.019 ± 0.006	ND	0.812 ± 0.063
Narkwa Lagoon	*Sediment (μg/g)*					
	A	Dry 1	0.006	ND	ND	20.455
		Dry 2	0.002	ND	0.023	18.753
		Mean	0.004 ± 0.003	ND	0.023 ± 0.00	19.604 ± 1.20
		Wet 1	0.012	0.076	1.183	13.850
		Wet 2	ND	0.006	ND	13.654
		Mean	0.012 ± 0.00	0.041 ± 0.049	1.183 ± 0.00	13.752 ± 0.139
	B	Dry 1	ND	ND	ND	32.453
		Dry 2	0.001	0.038	0.069	16.451
		Mean	0.001 ± 0.00	0.038 ± 0.00	0.069 ± 0.00	24.452 ± 11.315
		Wet 1	0.005	ND	ND	13.850
		Wet 2	0.005	0.011	0.112	8.945
		Mean	0.005 ± 0.00	0.011 ± 0.00	0.112 ± 0.00	11.398 ± 3.468
	Water (μg/l)					
	A	Dry 1	ND	0.039	ND	1.410
		Dry 2	0.002	0.033	ND	1.412
		Mean	0.002 ± 0.00	0.036 ± 0.004	ND	1.411 ± 0.001
		Wet 1	0.001	0.048	0.002	0.838
		Wet 2	0.001	0.051	ND	0.787
		Mean	0.001 ± 0.00	0.049 ± 0.002	0.002 ± 0.00	0.813 ± 0.036
	B	Dry 1	ND	0.001	0.060	1.056
		Dry 2	0.003	ND	0.075	1.529
		Mean	0.003 ± 0.00	0.001 ± 0.00	0.068 ± 0.011	1.293 ± 0.334
		Wet 1	ND	0.039	0.023	0.898
		Wet 2	ND	0.054	0.015	0.852
		Mean	ND	0.047 ± 0.011	0.019 ± 0.007	0.875 ± 0.033

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