

Exposure of Mercury from Gold Mining Area: A Case Study in Tendo and Aby Lagoon in Côte d'Ivoire

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Abstract

Lagoons of West African countries are seriously threatened by rapid artisanal and small-scale gold mining (ASGM) that exposes lagoons to mercury pollution. In this study, the mercury level in the sediments of the Tendo and Aby lagoons in Côte d'Ivoire had been evaluated. A total of 87 samples from 25 stations located on Tendo and Aby lagoons were analyzed by atomic fluorescence spectrometry. The mean Hg concentrations obtained in the sediments of Tendo and Aby lagoons were 0.89 ± 0.26 mg·kg⁻¹ and 0.70 ± 0.18 mg·kg⁻¹, respectively. Hg concentrations evaluated in the bays of Tendo and Aby during the dry season were 1.38 ± 0.45 mg·kg⁻¹ and 1.07 ± 0.31 mg·kg⁻¹, respectively. The minimum and maximum total Hg concentrations in the sediments from 25 stations were 0.04 and 3.56 mg·kg⁻¹, respectively. Mercury contamination in lagoons during ASGM poses risks of pollution for the lagoon ecosystem and also poses health risks for the population living near these lagoons.

Keywords

ASGM, Mercury, Sediments, Côte d'Ivoire, Pollution

1. Introduction

Mercury (Hg) is a recognized ubiquitous contaminant in aquatic ecosystems [1] [2]. Methylmercury (MeHg) is the Hg species of particular interest due to its

propensity to bioaccumulate and potentiality to cause health problems to humans and wildlife when exposed at high levels [3]. Hg pollution is a worldwide problem that has got severed during the past few decades because of its toxicity, extensive sources, non-biodegradable characteristics, and capacity to accumulate in aquatic environments [4] [5].

More than 99% of the heavy metals can be stored in the sediments indifferently [6] [7]. Sediments constitute a habitat for many aquatic organisms and often serve as effective reservoirs for pollutants [8] [9]. These toxic elements can be found in the water during favorable environmental conditions and degrade the quality of the water [10] [11]. Recent studies showed that mercury (Hg) released into the atmosphere from artisanal and small-scale gold (Au) mining (ASGM; approximately 880 Mg/year) activities is one of the primary emissions to the atmosphere [5]. It currently accounts for an estimated 37% of global Hg emissions into the atmosphere [12] [13]. Indeed, the amalgam is heated to vaporize the mercury and separate the gold. In “open burning”, all of the mercury vapour is emitted into the air [14]. After elemental mercury is vaporized, it can enter into aquatic ecosystems, where it may be biomethylated by bacteria into an organic form, methylmercury [15]. Fish and macroinvertebrates in aquatic ecosystems accumulate methylmercury in their tissues, with increasing concentrations in higher trophic levels [16].

The sediments were also contaminated by the activities of ASGM with up to 78% of THg of anthropogenic origin. This anthropogenic Hg, more than half (66% - 74%) comes from the liquid Hg (0) which is released during ASGM [17]. Gold miners usually add larger amounts of mercury to ensure that all available gold is amalgamated. Mercury is generally used in ASGM without any type of capture system to reduce chemical releases into the environment including soil and water [18]. The remaining anthropogenic Hg was from ASGM-induced erosion of Hg-rich soils in the river [17].

ASGM occurs in over 70 developing countries [13]. It is estimated to employ 13 million people globally, and an additional 80 - 100 million people are directly reliant upon or impacted by ASGM [19] [20]. This concerns most of the West African countries [21]. A recent investigation conducted in West Africa suggested in general that artisanal-mining activities are a source of trace metals to wetlands, with negative consequences on human health [22] [23].

In Côte d'Ivoire, ASGM has been increasingly developed since the outbreak of the sociopolitical crisis of September 19, 2002. Although ASGM attracts rural populations through the incomes it provides, however, documentation of its environmental consequences remains more fragmented. A few studies have investigated metal contaminations in lagoons during gold-mining activities [21] [23].

The aim of this study was to determine accumulations of mercury in the sediments during ASGM activities along Aby and Tendo lagoons in Côte d'Ivoire. Indeed, Aby and Tendo lagoons are under the influence of the Bia and Tanoe rivers, respectively, and these rivers constitute many ASGM industries. However,

ASGM activities that have gradually developed over the years around the rivers Bia and Tanoé fear that these rivers will serve as a vector of pollution for heavy metals, including mercury from gold-mining areas to the estuary of Aby and Tendo lagoons. Assessment of the exposure of Aby and Tendo lagoon sediments to pollutants resulting directly or indirectly from gold-mining activities is a relevant indicator of this pollution.

2. Materials and Methods

2.1. Study Areas

Aby lagoon ($5^{\circ}05'N$ - $5^{\circ}22'N$ and $3^{\circ}16'W$ - $2^{\circ}55'W$) is located in West Africa, on the coast of the Gulf of Guinea between Côte d'Ivoire and Ghana. Aby lagoon covers 24.5 km and 56 km, respectively, from east to west with an estimated area of 420 km^2 , while the Tendo lagoon is formed by the band from west to east and has a width of about 20 km. Aby and Tendo lagoons are located in an equatorial climate region with a climate composed of 4 successive seasons: 2 rainy seasons from May to July and from October to November and 2 dry seasons from August to September and from December to March/April [24].

We indicated successively from north to south for Aby north and south lagoons, and from west to east for Tendo (Figure 1). The 2 north and south Aby lagoons will simply be called Aby throughout this study. Several towns, villages,

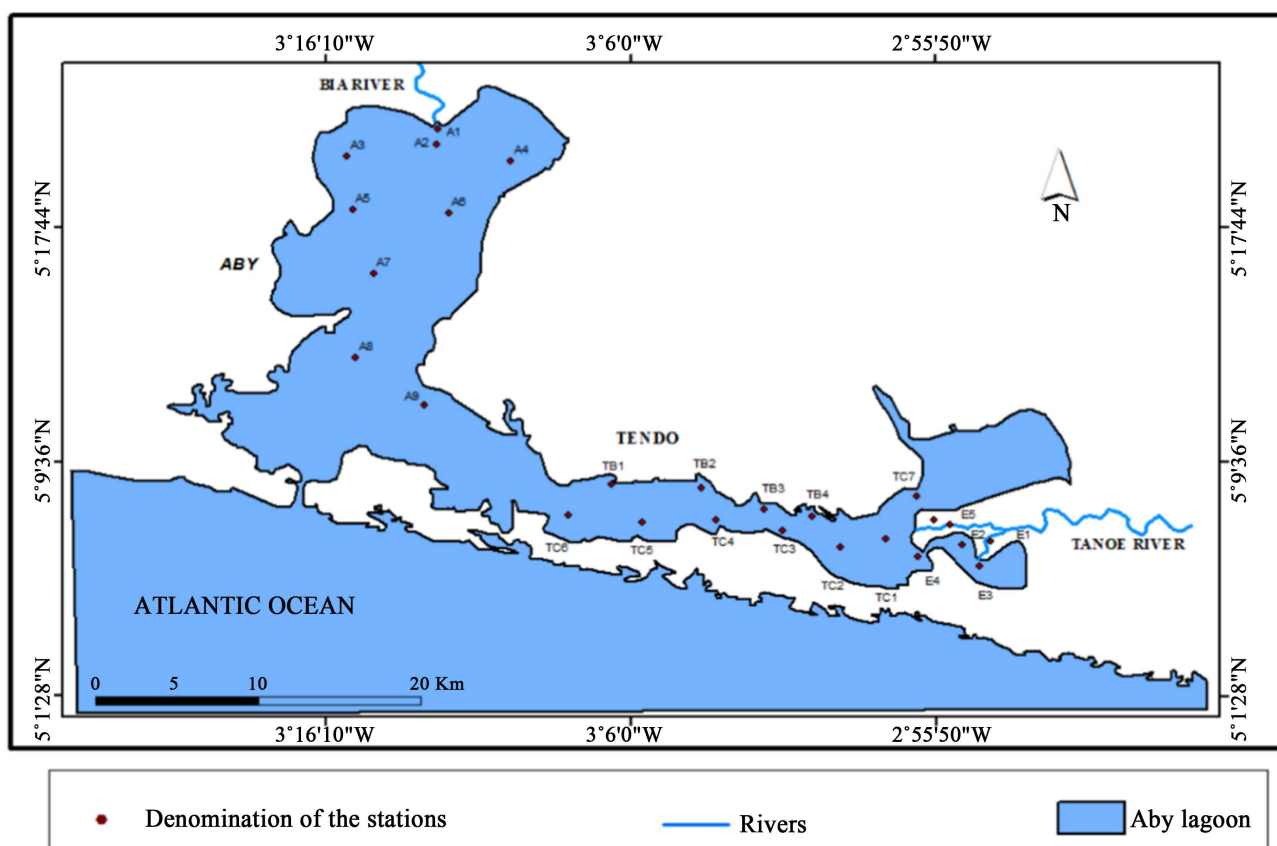


Figure 1. Locations of sampling sites.

and seasonal fishing camps exist around the Aby and Tendo lagoons. The population estimated to 30,000 [25] are living around Aby and Tendo lagoons that constitute and provide a real source of food and living for these populations through fishing activities. Aby and Tendo lagoons are under the influence, respectively, of the Bia river whose watershed is estimated at about 10,000 km² with an average flow estimated at 300 m³/s during rainy floods and the Tanoé river whose watershed is at about 16,074 km² with an average flow estimated at 142 m³/s [26]. These rivers are home to numerous gold deposits that have gradually developed over the years. Thus, artisanal and industrial gold mining around the Bia and Tanoé rivers fears that these rivers will serve as a pollution vector for heavy metals, especially mercury from gold-mining areas to its outlets in Aby and Tendo lagoons. Assessment of Aby and Tendo lagoon sediments' exposure to pollutants resulting directly or indirectly from gold-mining activities, namely, mercury, is a relevant indicator of this pollution. In addition to spatial, bathymetric, and hydrological variations, Aby and Tendo lagoons were influenced by the Bia and Tanoé rivers, respectively. Aby and Tendo lagoons are the estuaries of the Bia and Tanoé rivers, respectively.

2.2. Sampling Method

The study was conducted on the Aby and Tendo lagoons during the rainy and dry seasons. Two sampling periods were carried out during the rainy season from May to July and from October to November and two others during the dry season from August to September and from December to March/April based on the climatic variations.

Sampling was carried out on four sites: Aby lagoon where in samples A1 and A2 correspond to the estuary of Bia river, Tendo lagoon channel, bay of Tendo lagoon, and estuary of Tanoé river. Sediment samples of Aby and Tendo lagoons were sampled at stations preselected by the research team according to spatial, bathymetric, and hydrological variations. The sediment samples were collected in borosilicate glass vials (50 mL), previously washed in order to avoid adsorption and desorption reactions of the vessel walls. Wearing polyethylene gloves was adopted in all washing steps and while using equipment. Sampling equipment and sample containers were cleaned and kept clean according to the protocol described by Quémerais *et al.* [27]. A total of 87 sediment samples (0 - 5 cm) from 25 stations located on Aby and Tendo lagoons were collected (**Table 1**).

2.3. Sample Preparation and ICP Analysis

Samples were stored in polyethylene bags after being perfectly homogenized and kept in the dark in a cooler. Sediment samples were transported to the laboratory 24 hours after collection. As soon as it reached the laboratory, the sediments were immediately dried in an oven at 40°C until a constant weight was obtained and then sieved. The fraction inferior to 63 µm was collected in hermetic polyethylene

Table 1. Distribution of sampling points and locations on Aby and Tendo lagoons

Sites	n	Stations	GPS coordinates	Location
Aby lagoon	9	A1	05°21' (15.6°)N 003°12' 04.9°)W	Bia river
		A2	05°20' (59.5°)N 003°1' (08.6°)W	Estuary of Bia
		A3	05°20' (00.8°)N 003°15' (01.7°)W	Opposite side of A3
		A4	05°20' (06.8°)N 003°10' (01.9°)W	Estuary of Eholie (village of Eboué)
		A5	05°17' (31.8°)N 003°15' (07.0°)W	Channel of Adiaké
		A6	05°17' (32.0°)N 003°11' (02.9°)W	Abiaty
		A7	05°15' (06.0°)N 003°15' (00.0°)W	Ettueboué
		A8	05°12' (31.1°)N 003°15' (00.1°)W	Proximity switches to sea
		A9	05°10' (00.0°)N 003°10' (00.0°)W	Akounougbe
Tendo lagoon channel	7	TC1	05°07' (42.6°)N 002°57' (08.6°)W	Crest of Frambo
		TC2	05°06' (57.6°)N 002°57' (41.4°)W	Channel of N'zobenou
		TC3	05°07' (26.8°)N 003°00' (36.8°)W	Channel of Assué
		TC4	05°07' (33.6°)N 003°02' (29.6°)W	Channel of Tiapoum
		TC5	05°08' (20.0°)N 003°05' (00.0°)W	Eboko
		TC6	05°08' (00.5°)N 003°07' (36.0°)W	Channel of Eboinda
		TC7	05°07' (42.6°)N 002°57' (08.6°)W	Toward Ehy lagoon
Bay of Tendo lagoon	4	TB1	05°07' (46.7°)N 002°59' (55.6°)W	Bay of Mohoua village
		TB2	05°08' (11.5°)N 003°01' (42.8°)W	Bay of Tiapoum village
		TB3	05°09' (06.1°)N 003°03' (36.8°)W	Bay of N'Guiémé village
		TB4	05°08' (54.7°)N 003°06' (35.6°)W	Bay of Eboinda village

Continued

Estuaries of river Tanoé	5	E1	05°06' (16.3°)N 002°54' (19.3°)W	Tanoé river (south of Ghana's border)
		E2	05°05' (55.2°)N 002°54' (34.7°)W	South-side estuary extension 1
		E3	05°06' (32.4°)N 002°55' (03.5°)W	South-side estuary extension 2
		E4	05°06' (17.3°)N 002°55' (55.6°)W	Connection to the channel
		E5	05°07' (19.5°)N 002°56' (01.4°)W	Tanoé river (north of Ghana's border)

tubes and stored in a clean, dry place. The mineralization was made in Montpelier in the Department of Environmental Sciences and Public Health by the faculty of pharmaceutical sciences [28] [29].

The microwave mineralization method (301 Prolabo[®] and Microdigest Prolabo[®]) was used [30]. Sediment samples were taken up in a nitric acid and hydrogen peroxide (HNO₃ and H₂O₂) mixture according to the protocol described by Tseng *et al.* [31]. About 0.25 g of dried, homogenized sediment was weighed into a container added with 8 mL of concentrated nitric acid, and the container was closed using a reflux column. A plastic seal was placed between the reflux column and the container to avoid evaporation. The tube and its contents were subjected to the first phase of microwave heating at 20 W for 5 minutes; after cooling for 5 minutes, 2 mL of H₂O₂ were added; and they were subjected to the second heating phase at 20 W for 5 minutes. Finally, the mineralization was diluted with Milli-Q water in a 50 ± 0.06 mL borosilicate glass volumetric flask and then stored in the refrigerator in a polyethylene tube until analysis.

Analyses were performed on the PerkinElmer[®] ICP/MS Elan 6000 with Baffled Quartz Cyclonic Spray chamber. The methodology described by US EPA was used for instrument optimization and the analytical procedure [32]. The analysis was carried out with an RF power of 1100 W, a plasma gas flow of 15 L/min, an auxiliary gas flow of 1.2 L/min, and with nebulizer gas flow of 0.96 L/min. Peak scanning mode and an integration time are 1625.0 ms. Dwell time was fixed at 50 ms and 3 replicates were performed per sample.

2.4. Statistical Analysis

The statistical analysis of the data was carried out using the GraphPad Prism 5.0 software. Quantitative variables are summarized as the mean value and standard deviation in the tables and figures. For statistical analysis, quantitative data were analyzed using the Mann-Whitney U test, according to their distribution evaluated by the Shapiro-Wilk test. All tests were two-tailed and viewed as indicating statistical significance at a p-value of less than 0.05.

3. Results and Discussions

A total of 87 samples were analyzed in this study: 16 from Tanoe river, 23 from the channel of Tendo lagoon, 16 from bays of Tendo, and 32 from Aby north lagoon.

3.1. Mercury Accumulation in Selected Sediments

The highest concentrations of mercury were obtained in the bays of Tendo with an average of $0.89 \pm 0.26 \text{ mg}\cdot\text{kg}^{-1}$ and extremes ranging from 0.23 to $3.56 \text{ mg}\cdot\text{kg}^{-1}$ and in Aby with an average of $0.70 \pm 0.18 \text{ mg}\cdot\text{kg}^{-1}$ and extremes ranging from 0.07 to $4.16 \text{ mg}\cdot\text{kg}^{-1}$ (Table 2).

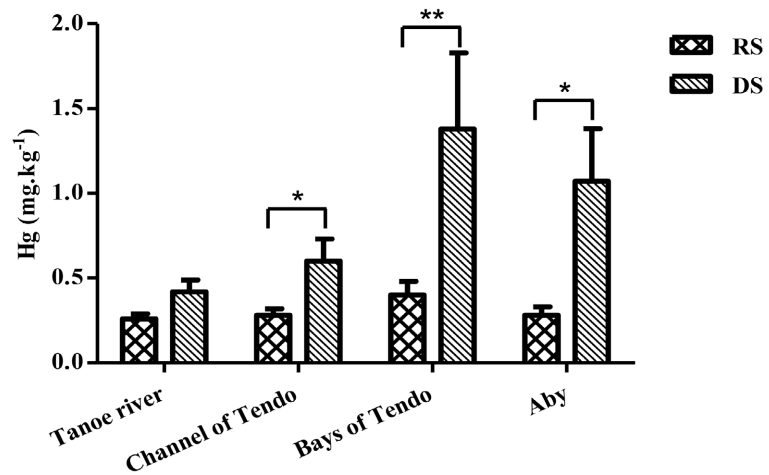
As shown in Table 2, sediment bays of Tendo and Aby north lagoons were the most contaminated. The results of the bays in this study were significantly higher than that of Bietri Bay. Indeed, in sediments of Bietri Bay, Hg ranged from 0.35 to $1.33 \mu\text{g}\cdot\text{kg}^{-1}$ [33]. The Hg concentrations in these sediments were similar to those reported by Kehrig *et al.* [34] (ranged from 0.50 to $2.38 \text{ mg}\cdot\text{kg}^{-1}$). According to Veeck *et al.* [35], mercury accumulation was reported in bays, and sediment composition in these bays is strongly influenced by fluvial inputs of mercury.

3.2. Mercury Accumulation during Seasons

In this study, two sediment collections were carried out during the rainy season (RS) and two others during the dry season (DS). RS was characterized by a significant ($p < 0.05$) mercury accumulation in bays of Tendo ($0.40 \pm 0.08 \text{ mg}\cdot\text{kg}^{-1}$) with extremes between 0.23 and $0.94 \text{ mg}\cdot\text{kg}^{-1}$ compared with other sampling sites. DS was characterized by significant ($p < 0.05$) mercury accumulation in bays of Tendo ($1.38 \pm 0.45 \text{ mg}\cdot\text{kg}^{-1}$) with concentrations ranging from 0.30 to $3.56 \text{ mg}\cdot\text{kg}^{-1}$ and in the Aby lagoon ($1.04 \pm 0.31 \text{ mg}\cdot\text{kg}^{-1}$) with extremes ranging from 0.13 to $4.16 \text{ mg}\cdot\text{kg}^{-1}$ compared with other sampling sites. However, Hg concentrations were significantly elevated ($p < 0.05$) in the dry season than the rainy season at all sampling sites as shown in Figure 2, with high rates at the bays of Tendo and Aby, precisely during the first dry season (DS1) compared with the second dry season (DS2) (Table 3 and Table 4). The results of this study were higher than those of Kouamenan *et al.* [36] in the mainland ($0.44 \pm 0.19 \text{ mg}\cdot\text{kg}^{-1}$) and maritime ($0.39 \pm 0.13 \text{ mg}\cdot\text{kg}^{-1}$) sediments of the Ebrié lagoon

Table 2. Mean sediment Hg by site.

Sites	Tanoe river	Channel of Tendo	Bays of Tendo	Aby
n	16	23	16	32
Mean ($\text{mg}\cdot\text{kg}^{-1}$)	0.35	0.44	0.89	0.70
Median	0.27	0.30	0.40	0.31
Min.-Max.	0.04 - 0.96	0.10 - 1.50	0.23 - 3.56	0.07 - 4.16
Standard Deviation	0.05	0.08	0.26	0.18



Note: RS—Rainy Season, DS—Dry Season.

Figure 2. Comparison of Hg concentrations between RS and DS for different sites of the Tanoe river, Tendo lagoon, and Aby lagoon.

Table 3. Hg concentrations between RS1/RS2 for different sites of Tanoe river, Tendo lagoon, and Aby lagoon.

Sites		RS1	RS2	p-value
Tanoe river	Mean ± SD	0.18 ± 0.02	0.30 ± 0.03	
	Median	0.19	0.27	0.04
	Min.-Max.	0.14 - 0.22	0.27 - 0.43	
Channel of Tendo	Mean ± SD	0.21 ± 0.04	0.35 ± 0.06	
	Median	0.20	0.30	0.23
	Min.-Max.	0.10 - 0.38	0.26 - 0.60	
Bays of Tendo	Mean ± SD	0.36 ± 0.02	0.43 ± 0.17	
	Median	0.36	0.28	0.69
	Min.-Max.	0.31 - 0.42	0.23 - 0.94	
Aby	Mean ± SD	0.27 ± 0.09	0.29 ± 0.05	
	Median	0.19	0.26	0.85
	Min.-Max.	0.08 - 0.59	0.07 - 0.50	

Note: RS1—First Rainy Season, RS2—Second Rainy Season.

in Côte d'Ivoire during the dry season. Like Tomiyasu *et al.* [37], a difference was observed in mercury concentrations between the rainy and dry seasons in this study. Koffi *et al.* [33] also reported higher Hg mean concentrations in the dry season. In addition, the concentrations of mercury in the sediments of the study areas exceeded the standard concentration levels ($Hg = 0.04 \text{ mg.kg}^{-1}$) as recommended by INERIS [38]. This high level of Hg in bays could be explained by the fact that in the rainy season, there was a greater distribution of Hg, while in the dry season, the predominant process was the remobilization of Hg due to

Table 4. Hg concentrations between DS1/DS2 for different sites of Tanoé river, Tendo lagoon, and Aby lagoon.

Sites		DS 1	DS2	p-value
Tanoé river	Mean ± SD	0.55 ± 0.12	0.28 ± 0.04	0.061
	Median	0.47	0.26	
	Min.-Max.	0.40 - 0.96	0.22 - 0.43	
Channel of Tendo	Mean ± SD	0.61 ± 0.18	0.58 ± 0.20	0.924
	Median	0.48	0.47	
	Min.-Max.	0.13 - 1.25	0.13 - 1.50	
Bays of Tendo	Mean ± SD	2.29 ± 0.63	0.46 ± 0.07	0.028
	Median	2.53	0.47	
	Min.-Max.	0.54 - 3.56	0.30 - 0.59	
Aby	Mean ± SD	1.77 ± 0.50	0.37 ± 0.07	0.018
	Median	1.68	0.28	
	Min.-Max.	0.13 - 4.16	0.19 - 0.70	

Note: DS1—First Dry Season, DS2—Second Dry Season.

the resuspension of bottom sediments [39]. Tendo and Aby lagoons are the estuaries of the Tanoé and Bia rivers, respectively. Mirlean *et al.* [40] reported concentrations of 0.02 - 17.84 mg·kg⁻¹ in sediments of the Patos estuarine lagoon in southern Brazil. The Tendo and Aby lagoons receive freshwater from the Bia and the Tanoé rivers, respectively. During the rainy season, these rivers are home to numerous gold deposits that have gradually developed over the years. Thus, artisanal gold mining around the Bia and Tanoé rivers fears that these rivers will serve as a pollution vector for heavy metals, especially mercury from gold-mining areas to its outlet in lagoons Aby and Tendo. Moreover, the Bia river sampling site was downstream of mining activities in Ghana [41] [42], and the samples in the other locations were all near commercial or artisanal gold-mining activities. Moreover, estuaries are the meeting place of saltwater from the sea and freshwater from rivers and are dynamic environments characterized by large fluctuations in environmental conditions [43]. The mercury is then transferred from lower to higher trophic levels, from plankton to fish [44], which consumption is the main source of exposure to mercury (Hg) through diet in humans [45]. The health risks associated with increased Hg levels in reservoir fish, which impact communities that rely on fish as a traditional food [46].

3.3. Spatial Distribution of Mercury

A wide range of Hg levels were captured in estuarine sediments with concentrations varying across regions and between sites and subsites [47]. As shown in **Figure 3**, during the DS1, all sites in the bay of Tendo exhibited the highest concentrations of mercury. However, no spatial evolution of Hg in the sediments could be observed

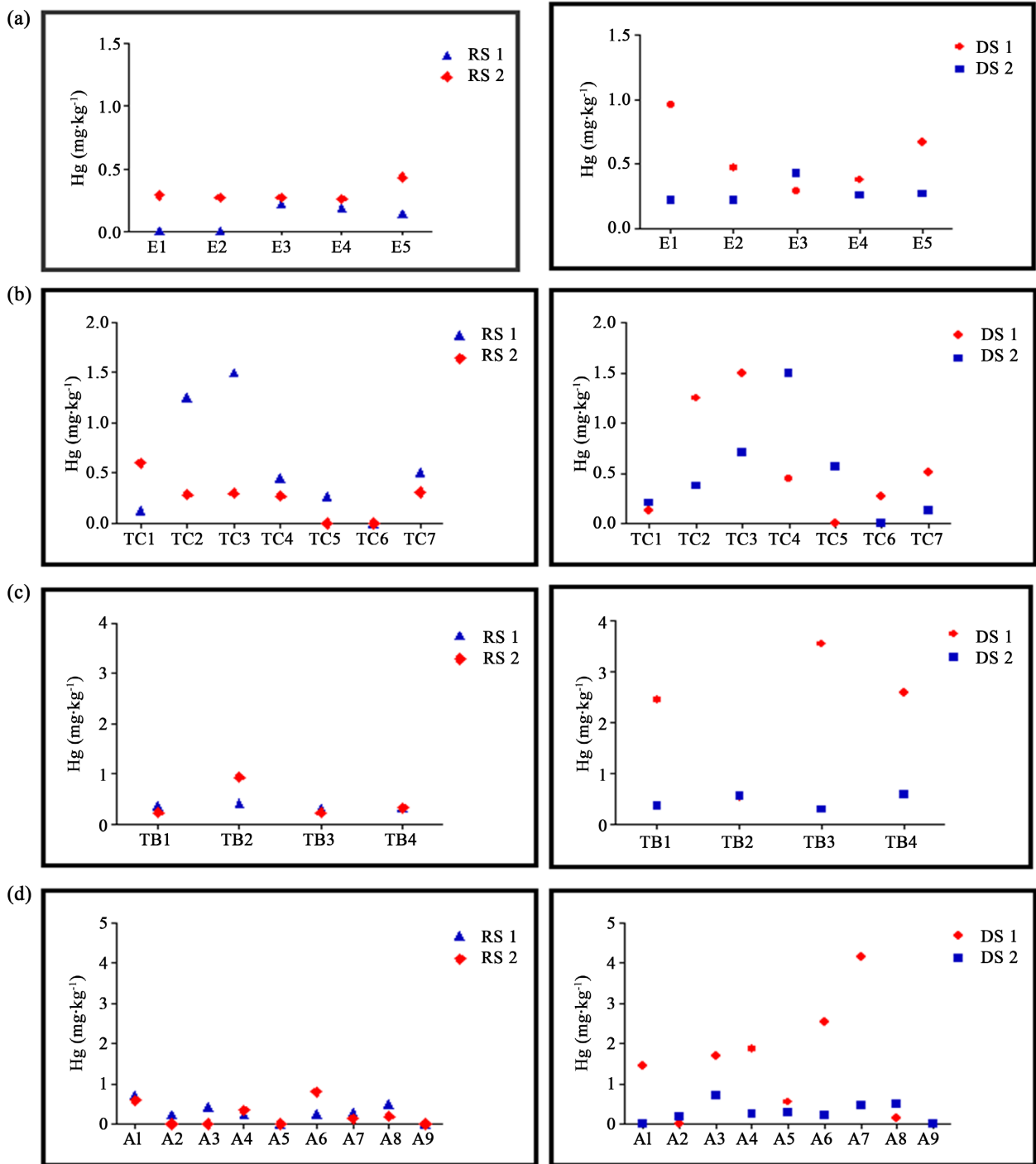


Figure 3. Hg concentrations at RS1 and RS2 and DS1 and DS2 in: (a) the Tanoe river from the Ghanaian side to the northern border; (b) the channel of the Tendo lagoon from the shore of the river (TC1) to the outlet to the sea (TC4); (c) the bays of the Tendo lagoon from the shore of the river (TB1) to the outlet to the sea (TB4); and (d) sampling points from the shore of the Bia river (A1) to the Aby lagoon (A6). Note: RS1—First Rainy Season, RS2—Second Rainy Season, DS1—First Dry Season, DS2—Second Dry Season.

between sites TB1 (close to the outlet of the river) and TB4 (near the inflow to the sea). Site TB2 was an exception to this evolution because unlike the other bays, it was the outlet of a tributary of the Tendo lagoon; the sedimentation

could have been disrupted. Moreover, mercury was accumulated along the Aby lagoon, as well as at the mouth of the Bia river during the DS1 with high concentrations at Abiaty (A6) and Ettueboué (A7). These two sampling points are located in the central part of the lake that constitutes the deepest region, so the removal of Hg may be more difficult [39]. In the DS1, Hg concentrations in the sediments of the Aby north lagoon showed a positive gradient at the Bia river estuary where it runs to the sea. This phenomenon was not observed in other sampling operations (DS2, RS1, and RS2). The distribution pattern of Hg in the bottom sediment demonstrates that an expansion of the area occurs in relation to the area of distribution of Hg in the dry season [39]. Bełdowski *et al.* [48] suggested that despite the decrease in mercury emissions to the environment in recent years, local weather conditions intensified by climate change seriously affect the bioavailability of past mercury deposits in coastal sediments. Indeed, global mineralization of the waters of the Aby lagoon caused by significant evaporation due to high temperatures during the dry season [22] [49] could explain this accumulation of mercury. Temperatures at all sites in the Aby lagoon are higher in the dry season (29°C - 31°C) than in the rainy season (25°C - 27°C) [50] [51]. Indeed, physic-chemical parameters such as temperature (T), pH, DO, salinity, conductivity (EC), total suspended solids (TSS) and total phosphorus (TP) in surface water, AVS and TOC in sediments are important factors affecting the concentrations, migration and transformation of heavy metals in rivers [52].

This accumulation of mercury could also be due to the limited exchanges between the lagoon and the sea due to the shrinkage of the Assinie pass over time [50] [51].

4. Conclusion

This study showed mercury accumulation in sediments of Aby north and Tendo lagoons. It increases in the dry season, particularly in the Aby north lagoon and in the bays of the Tendo lagoon, where its distribution in the sediments was subject to spatial variations. In this study, sediment concentrations of Hg were higher than normal with some contaminated sites. The risk to the preservation of the Aby lagoon ecosystem linked to the presence of Hg in the sediments is not negligible. The highest levels of Hg found in sediments were mainly due to the anthropogenic input around these lagoons.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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