

Increased Bioavailability of Mercury in the Lagoons of Lomé, Togo: The Possible Role of Dredging

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Abstract Surface sediments of the lagoons of Lomé, Togo, were analyzed for mercury, methylmercury, and trace elements. Concentrations were greater than typical for natural lagoon sediments, and with greater variability within the Eastern lagoon compared to the Western one. The Eastern lagoon is larger and has been dredged in the past, while the Western lagoon, which also receives major waste inputs, has not been dredged and shows less tidal flushing. Accordingly, one naturally believes that the Eastern lagoon is cleaner and probably safe to use due to its natural resources, including fishes to eat. Unexpectedly, we describe here that mercury methylation was greater in the Eastern lagoon, indicating increased bioavailability of mercury, as probably facilitated by past dredging that decreased solid-phase retention of inorganic mercury. Urbanization has historically been more developed in the southern part of the lagoons, which is still reflected in contamination levels of sediment despite dredging, probably because sources of contamination are still more important there today. Such urban contamination emphasizes the need to regulate waste discharges and possible airborne contamination in growing cities of developing countries, and implements environmental and public health monitoring, especially in relation to misbeliefs systematically associated with the cleansing effect of dredging activity.

Keywords Metal contamination · Enrichment factor · Geoaccumulation index · Urban pollution · Organic chelation · Lagoon sediments

INTRODUCTION

Lagoons are typically shallow, inland yet coastal bodies of water with limited saltwater inputs. As a result of the

availability of nutrients, high productivity, and a wide array of habitats, lagoonal ecosystems are often composed of diverse and abundant fauna and flora, frequently of socio-economic importance (Dame and Allen 1996; Ray 1996). Lagoonal ecology depends on a delicate dynamic balance between a number of factors, including water circulation, sediment transport, geochemistry of sediment particles, and the life history, biology and ecology of resident species (Dame and Allen 1996; Guira 1992).

Sediment in lagoons is usually highly contaminated because lagoons are typically the site of extensive human settlement, and the end reservoir of contaminants transported by rivers and urban runoff, where the water gets filtered and the contaminants retained. Contaminants like metals thus end up in sediment where they accumulate over time up to levels where they can become hazardous to the surrounding ecosystem, making quality assessment and monitoring of lagoon sediments necessary to continuously implement (Lemly and Richardson 1997).

Several reports have been published on the effect of dredging coastal or estuarine sediment on the toxicity and bioavailability of trace metals, and both positive and negative outcome have been reported (Bloom and La-sorsa 1999; Guerra et al. 2009; Lee et al. 1982; Lewis et al. 2001). Examples of positive results are (1) plant uptake of Cd and Zn was lowered when dredged sediments were deposited in reduced (flooded) environments (Lee et al. 1982); (2) trace metal concentrations (e.g., Zn, Cr, and Hg) in periphyton were decreased after the dredging activity of estuarine sediments for Zn, Hg, and Cr (Lewis et al. 2001); and (3) dredging activity of a coastal lagoon had no dramatic effect (based on a before/after comparison) on Hg concentration and sediment toxicity (Guerra et al. 2009). Examples of negative outcome from the effect of dredging on metal

bioavailability and toxicity include (1) total Hg concentrations increased in decant water during episodic maintenance dredging (Bloom and Lasorsa 1999); and (2) uptake of Cd and Zn increased in plants when contaminated marine dredged material was reused on land (Folsom et al. 1982).

Metals are among the common contaminants found in lagoon sediment, including mercury (Hg). Mercury contamination and toxicity have long been of concerns during the last decades due to the occurrence of large-scale poisoning (Kudo et al. 1998). Natural sources of Hg comprise weathering and degassing from water surfaces and land and volcanic eruption, while anthropogenic sources include metallurgical processes, fossil fuel burning, wood pulping, paint and chloro-alkali industries, battery production, pharmaceutical waste, and agriculture (Boening 2000; Mason et al. 1994; Morel et al. 1998).

Mercury in aquatic ecosystems is present under various (bio)chemical and physio-chemical conditions, which controls bioavailability, mobility, and toxicity of Hg to organisms (Han et al. 2007; Sunderland et al. 2009). In the global biogeochemistry of Hg, it is well known that only a small fraction of Hg transported from rivers is exported to the open water ocean due to the high retention of this metal in estuarine and coastal systems by adsorption and precipitation processes (Faganeli et al. 2003; Han et al. 2006). Like other metals, once Hg enters into the estuarine ecosystems, it interacts with dissolved and particulate compounds in the water column, and a portion of it sinks to the sediments. This trapping and settling process leads to high concentrations of Hg in estuarine sediments where Hg may be retained bound to the particulate organic matter (Skylberg et al. 2006), then likely associated with Mn/Fe-oxides and/or FeS near the redox boundary of anoxic layers (Canario et al. 2003). It is in such anoxic sediments that microorganisms, such as sulfate-reducing bacteria, convert inorganic Hg into the methylated form. Methylmercury (MeHg) has higher bioaccumulation and biomagnification factors than those of inorganic Hg in the aquatic food webs, and also causes acute toxicity and long-term adverse effects to aquatic organisms (Morel et al. 1998).

Numerous studies have revealed that lagoons located in urbanized areas are easily exposed to inorganic (in particular trace elements) and organic contaminations (Tiefenthaler et al. 2008). In particular, the high trapping and settling processes characteristic of lagoons lead to high concentrations of contaminants to accumulate in sediments over time. Surface sediment then in turn appears as a contamination source, especially when physico-chemical conditions of the environment are such that contaminants associated with the sediment are able to flux into the

overlying or pore-water, and incorporate into the benthic and pelagic biota (Salomons et al. 1987).

Lomé, the capital of Togo, is a vibrant international city with >3/4 million inhabitants. Over the years, the city gained prosperity for its geographical location being the major axis of penetration to the Volta basin, a major fertile area shared by several countries. German, British, and French were among the colonists that made the city a major port and world-renowned trading and commercial center (Nilsson 2002). The city has a nearby oil refinery and phosphate mine, which bring businesses yet are associated with critical environmental issues (Gnandi et al. 2009; Gnandi and Tobschall 1999), and in addition to exporting various agricultural goods (coffee, copra, cocoa), tourism is another growing industry beneficial for the city.

Lomé has a network of narrow lagoons connected to one another that occupies an East–West shallow depression extending across various areas of the city along the coast. The lagoon system therefore receives various influxes of contaminants, including suspended loads from soil erosion, rainwater runoff, urban and hospital effluents, and leachates from the numerous waste dumps located along the lagoons shoreline, which are especially outnumbered along the southern side where the historic center of the city first developed. The two main lagoons of Lomé have for long been sites of frequent eutrophication events caused by excessive discharge of P, N, and organic carbon and decreased water circulation (Gnandi et al. 2007). The larger lagoon (East lagoon) has been dredged and deepened in the early 1970s with the goal to increase depth and water circulation (and thus cleanliness and public health safety) while keeping the same volume of lagoonal water, by extending the shoreline with the dredged sediment (Bockemühl and Triemer 1974). Despite the obvious continuous threats for these ecosystems to decline upon growing urbanization of the surrounding city, no study has assessed levels of trace elements contamination in the sediment of these lagoons. In addition, there is no information available comparing the Eastern and Western lagoons in terms of metal contamination of the sediment even though those two lagoons, technically representing the same body of water, have been subject to different sediment management practices.

Here, we aim at filling this gap by providing baseline concentration values with regards to As, Al, Cd, Fe, Mn, Pb, and Ti, and with emphasis on total Hg and MeHg. We also assess spatial distribution of these trace elements associated with surface sediment considering potential sources of contamination, and the fact that dredging activity took place in the past for one of the lagoons.

MATERIALS AND METHODS

Study Location

The lagoons of Lomé belong to a complex lagoon network in the Gulf of Guinea, which extends from Ivory Coast to Nigeria. It is locally intersected by land strips and connected to the Atlantic Ocean by two discharge inlets. In Lomé, the lagoon system consists of two main lagoons that cover a surface area of about 14 km², of which 8 km² are actual wetlands with an average depth of approximately 4 m, the remaining being a wide low depression shoreline (SGI 2003). The Eastern lagoon (Lagoon of Bè) is the larger one covering 4.9 km², while the Western lagoon (Lagoon of Nyékonakpoè) has a surface area of 3.1 km² (Fig. 1). The system comprises two channels: a large regulation channel with 2,462-m length connecting both the lagoons, and an eastward 1,200-m long channel, which is an extension of the lagoon of Bè (Fig. 1a). Furthermore, two discharge inlets connect both the lagoons to the Atlantic Ocean, the Western discharge is 2,350-m long and the Eastern one is 2,250 m. Those inlets were artificially made to increase water circulation and exchange between lagoon and oceanic waters during overflow and high tides.

Sample Collection

Sediment samples were collected at about 1–4-m water depth in December 2006, using a sediment grab (mini-Shipek type), manipulated from a motorboat. Seventeen samples of surface sediment (about top 3–5 cm) were collected from the Western Lagoon and twenty-one from the Eastern Lagoon. Acid-cleaned polypropylene bottles were used to receive the samples directly upon retrieving the grabber onto the boat; soon after collection in the laboratory, each sample was then split in two fractions; one was used for trace elements analyses and the other stored frozen for analyses of Hg and MeHg. Samples were then transported to Scripps Institution of Oceanography (San Diego, California, USA) for Hg, MeHg, and trace elements analyses, each in triplicates.

Sample Analyses

For total Hg analysis, approximately 1 g of sediment was digested overnight in Teflon[®] bottles at room temperature with 8 cm³ of 12-N HCl and 2 cm³ of 14-N HNO₃, then diluted with 500-cm³ Milli-Q water (Barnstead International, IA, USA). Methylmercury in sediments was extracted as described in Han et al. (2007). In summary, approximately 1-g wet sediment was mixed with 5-cm³ acidic KBr, 1 cm³ of 1-M CuSO₄, and 10 cm³ of CH₂Cl₂ before being exposed to vigorous shaking for 1 h using a

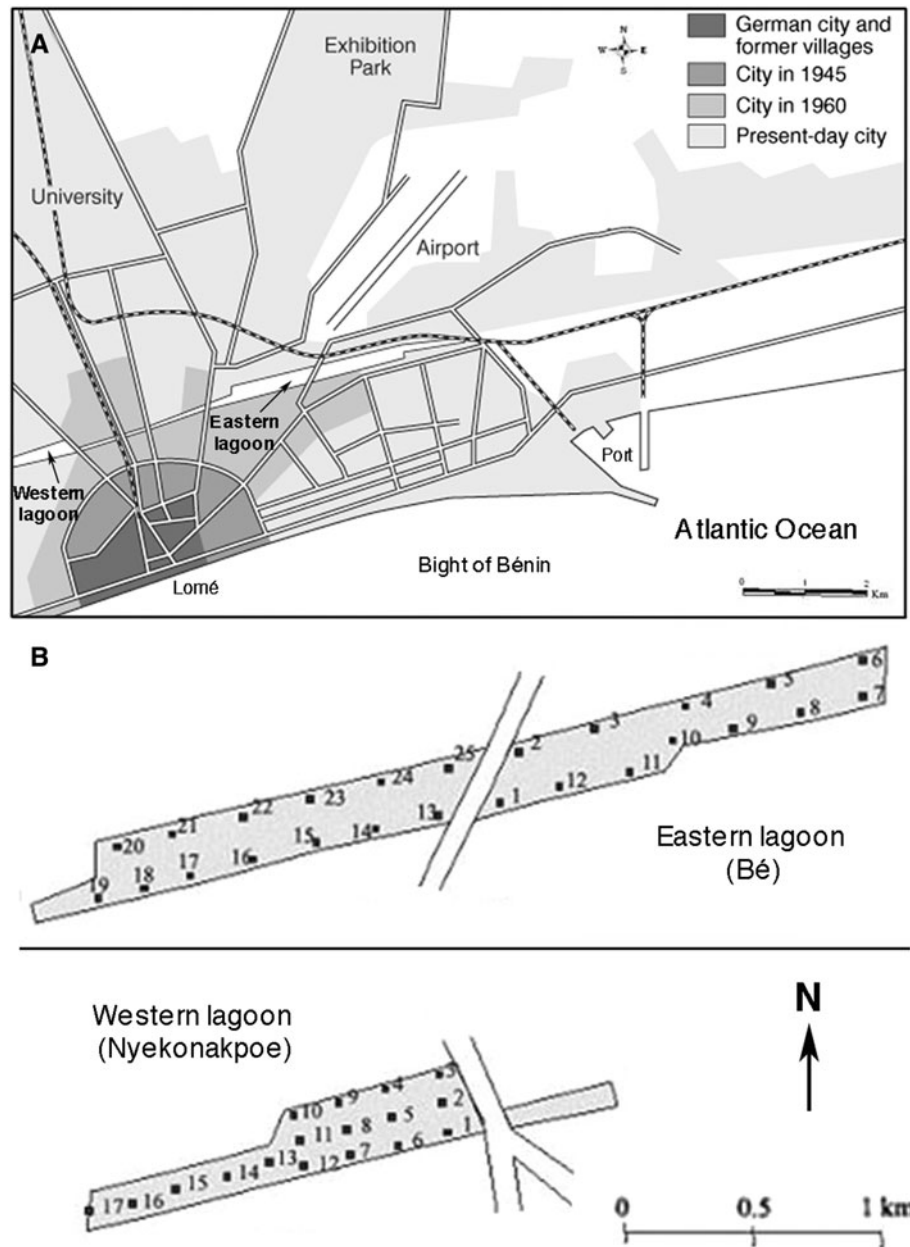
flask shaker Thermo Scientific MAXQ4000 (Terra Universal Inc, CA, USA). After centrifugation for 20 min (3,000 rpm), a 2-cm³ aliquot of the CH₂Cl₂ layer was pipetted out into acid-cleaned Teflon distillation vials that were heated (45°C) and purged with N₂ until complete volatilization of CH₂Cl₂. Total and methyl Hg were quantified by cold vapor atomic fluorescence spectrometry with recoveries of certified reference material (PACS2 for total and DORM-2 for MeHg, National Research Council Canada) being 96 and 94% for total and methyl Hg, respectively.

For determination of trace elements (adapted from Deheyn and Latz 2006), sediments freshly collected were dried at 70°C for several days before being homogeneously crushed in a silicon mortar and the <63-µm fraction separated by dry sieving. A small amount (about 0.2 g) of that fraction of sediment was weighed using a Sartorius CP225D analytical microscale (Data Weighting Systems, IL, USA) connected to a notebook computer for accurate recording of measurements. Samples were then taken in Teflon digestion vessels and partially digested during 20 min in a HCl:HNO₃ (3:1) mixture at 80°C using a Ethos EZ microwave digester (Milestones, CT, USA). Trace elements (Al, Cd, Fe, Mn, Pb, Ti, and Zn) were analyzed simultaneously using an Inductively Coupled Plasma Atomic Emission Spectrometer Optima 3000 XL (Perkin Elmer, CA, USA) with detection limits ranging from 0.05 * 10⁻⁶ to 4.0 * 10⁻⁶ mg g⁻¹ depending on the element (PerkinElmer 2000). The instrument, available at the Analytical Facility of the Scripps Institution of Oceanography, was calibrated before every run by successive dilution of a 100 µg g⁻¹ multi-element instrument calibration standard solution (Fisher Scientific, CA, USA). Recovery of the quality assurance standards was analyzed every 20 samples over the course of the run was 103%, while internal blanks were analyzed to assess any background contamination originating from the sample manipulation was negligible.

Total organic carbon (TOC) was analyzed according to the colorimetric titration process (Gaudette and Flight 1974). Approximately 200 mg of the pulverized sediment was weighted directly in an Erlenmeyer vial to which 10 cm³ of dichromate sodium solution was added. After mixing, 200 cm³ of concentrated sulfuric acid was added and allowed to react at room temperature for 30 min, after which 200 cm³ of distilled water, 10 cm³ of phosphoric acid, and 0.2 g of sodium fluoride was added to the mixture. After addition of 10 drops of indicator (biphenylamine), titration of the solution was performed using ammonium ferrous sulfate solution. The color of the indicator changes from green-brown to dark blue and dark green. The percentage of TOC in dry sediment was calculated according to the following formula:

$$\text{TOC} = 10 * (1 - T/S) * [N * (0.003) * (100/W)];$$

Fig. 1 a Location of the two main lagoons in Lomé, Togo, with indication of historical development of the city (adapted from Tonio Del Barrio image web-posted on 2/17/2006). **b** Close view indicating sediment sampling locations for the Eastern lagoon (Bè) and Western lagoon (Nyékonakpòè)



where T is the volume of ammonium solution used for the titration of the sample in cm^3 , S is the volume of ammonium solution used for the titration of the blank in cm^3 , N normality of the dichromate solution, W weight of the dry sediment in g.

Enrichment Factor (EF) and Geoaccumulation Index

EF_s

EF was calculated to evaluate natural versus anthropogenic contribution of metals concentration associated with the sediment. EF is defined as the mean concentration

ratio of a given element A relative to a standard element B in the sample, divided by the same ratio in a reference material from the Earth crust; this reference material is chosen to be the most representative of the geological characteristics of the study area (Förstner and Wittmann 1981; Li 1981):

$$EF = \frac{[\text{conc. } A \text{ sample} / \text{conc. std } B \text{ sample}]}{[\text{conc. } A \text{ ref.} / \text{conc. std } B \text{ ref.}]}$$

Average crustal shale values representative of Togo were obtained from (Turekian and Wedepohl 1961; Wedepohl 1991). Aluminum was used as a standard element since commonly considered a reference element in relation to clay content in lagoons (Durn 1996). Usually, low EF

values suggest that the trace element may be entirely from crustal materials or natural weathering processes, while high EF values would indicate that a significant portion of trace element originates from non-crustal materials or non-natural weathering processes (Feng et al. 2004). Here, the EF values were interpreted such as $EF < 1$ indicates no enrichment, $EF = 1–3$ minor enrichment, $3–5$ moderate, $5–10$ moderately severe, $10–25$ severe, $25–50$ very severe, and >50 extremely severe (Acevedo-Figueroa et al. 2006).

Geoaccumulation Index (Igeo)

Another index often used to assess level of anthropogenic contamination is the Igeo, which considers the concentration of elements from a natural baseline (pristine) environment to which sample concentration is normalized (Covelli and Fontolan 1997; Tam and Yao 1998). Geoaccumulation index was calculated using the following formula with the mean concentration of element A samples (Leopold et al. 2008):

$$I_{geo} = \log_2 (\text{conc. A sample} / k * \text{conc. A ref.})$$

with $k = 1.5$, to consider possible variations in the background data due to lithogenic effects, and background mean concentration values of element A reference from the local unpolluted environment (Calamari and Naeve 1994; Khaled et al. 2006). The geoaccumulation index consists of seven grades ranging from unpolluted to very strongly polluted: $I_{geo} < 0$ refers to unpolluted, $I_{geo} = 0–1$ unpolluted to moderately polluted, $I_{geo} = 1–2$ moderately polluted, $I_{geo} = 2–3$ moderately to strongly polluted, $I_{geo} = 3–4$ strongly polluted, $I_{geo} = 4–5$ strongly to very strongly polluted, and $I_{geo} > 5$ very strongly polluted (Müller 1981).

Statistical Analysis

Descriptive statistics (mean \pm one SE, minimum, maximum, and median values) were used to illustrate concentrations of trace elements and TOC, and were carried out using Microsoft Excel, while represented (Box Plot) using Deltagraph 5.0 software. Analysis of variance (single factor ANOVA) and post hoc multiple comparison of means (Fisher's PLSD) were used to test significance of differences between metal concentrations between lagoons and southern versus northern side of the lagoons. Pearson's coefficients analyses were used to test extent and significance of the correlation among concentrations of the various trace elements, and between concentrations of the trace elements and the ones of TOC. These analyses were completed both considering an Eastern–Western lagoons grouping (all northern, central, southern data aggregated for each lagoon), or a Southern–Northern grouping

(all Eastern and Western data aggregated for each part, excluding data from central part for the Western lagoon). All statistical analyses were performed using Statview® 5.0 software or SPSS 10.1 statistical softwares (SAS Institute, Inc.), with significance based on an α of 0.05. Unless otherwise stated, values represent means with one standard error, and stated P values are the highest encountered among all comparisons.

RESULTS

The Eastern (Bé) and Western (Nyékonakpoè) lagoons showed similar content of Hg, MeHg, and trace elements associated with sediment (Tables 1, 2), although the mean concentration values were always systematically lower for the Eastern lagoon (Fig. 2). In general, the Eastern lagoon showed greater range of concentrations when compared to the Western lagoon, thus indicative of greater variability of metal content among the samples. In most cases, each element showed mean and median concentrations values that were similar, except for MeHg that had only few samples with out-of-range concentration values (Fig. 2). Concentrations for Al, Cd, and TOC were significantly different ($P < 0.040$) between the Eastern and the Western lagoons, and greater in the Western lagoon. Such trend was also observed for Fe and Hg for which, however, the difference was not significant ($P = 0.068$), while no particular trend of variation was observed for MeHg, Mn, Pb, Ti, and Zn (Tables 1, 2).

In both the lagoons, there was a tendency for the concentrations to be greater in the southern and center (Western lagoon only) part of the lagoon when compared to the northern part (Fig. 3). This was significant in both the lagoons for Cd ($P < 0.0194$), while it was only significant in the Western lagoon for Hg, Pb, TOC, and Zn ($P < 0.0192$), only significant in the Eastern lagoon for Fe ($P = 0.008$), and not significant for MeHg and Ti ($P > 0.2801$). As for Al and Mn, they did not show any significant trend of variation between southern and northern shores of the lagoons.

Total Mercury Concentration in Sediments

Total Hg concentrations in the sediment of the Eastern Lagoon ranged from $0.01 \mu\text{g g}^{-1}$ (B3) to $3.69 \mu\text{g g}^{-1}$ (B19) with a median value of $0.54 \mu\text{g g}^{-1}$; those in the Western Lagoon ranged from $0.02 \mu\text{g g}^{-1}$ (N3) to $4.73 \mu\text{g g}^{-1}$ (N15) with a median value of $1.11 \mu\text{g g}^{-1}$ (Tables 1, 2). In both the lagoons the total Hg concentrations were greater in the southern shores ($0.84 \mu\text{g g}^{-1}$ for Eastern lagoon, $2.08 \mu\text{g g}^{-1}$ for Western lagoon) when compared to the northern shores ($0.23 \mu\text{g g}^{-1}$ for Eastern

Table 1 Eastern lagoon (Bè): concentrations (mean from triplicate) for each sampling site and descriptive statistics for the all sites, for total Hg, MeHg, trace elements, and TOC

Site	Element measured										
	Al (mg g ⁻¹)	Cd (µg g ⁻¹)	Fe (mg g ⁻¹)	Hg (µg g ⁻¹)	MeHg (ng g ⁻¹)	MeHg (%)	Mn (mg g ⁻¹)	Pb (mg g ⁻¹)	Ti (mg g ⁻¹)	Zn (mg g ⁻¹)	TOC (%)
B1(s)	21.07	0.19	13.50	0.04	0.02	0.06	0.47	0.04	2.28	0.10	1.86
B2(n)	20.54	0.21	13.77	0.02	bdl	NA	0.49	0.03	24.66	bdl	bdl
B3(n)	15.14	bdl	13.92	0.01	0.05	0.45	0.54	0.01	2.94	0.02	0.79
B4(n)	Lost during processing										
B5(n)	Lost during processing										
B6(n)	47.43	0.84	34.55	0.01	0.17	1.20	0.93	0.12	4.02	0.40	5.84
B7(s)	43.93	1.26	39.65	0.06	0.13	0.22	1.05	0.12	39.65	bdl	0.56
B8(s)	Lost during processing										
B9(s)	41.44	1.91	43.36	0.19	bdl	NA	1.24	0.09	2.94	0.24	bdl
B10(s)	Lost during processing										
B11(s)	68.02	bdl	49.52	0.54	0.07	0.01	2.01	0.16	4.98	0.35	bdl
B12(s)	52.24	1.01	46.23	0.84	0.12	0.14	1.78	0.13	3.54	0.38	8.61
B13(s)	62.03	0.84	46.16	0.57	0.18	0.03	2.17	0.14	4.92	0.33	7.06
B14(s)	61.56	bdl	41.27	0.55	0.15	0.02	1.24	0.20	4.68	0.80	5.69
B15(s)	60.71	0.80	43.50	0.58	bdl	NA	2.01	0.14	4.56	0.35	6.49
B16(s)	68.49	bdl	43.29	0.80	Bdl	NA	1.78	0.19	5.63	0.47	bdl
B17(s)	78.23	1.15	47.98	1.13	0.13	0.01	1.70	0.08	6.05	0.19	8.75
B18(s)	72.57	1.11	44.20	1.14	bdl	NA	0.78	0.36	6.29	1.18	0.78
B19(s)	61.08	bdl	41.13	3.69	8.42	0.22	0.62	0.36	5.57	1.30	bdl
B20(n)	27.74	0.38	12.66	bdl	bdl	NA	0.39	0.02	2.70	0.07	7.02
B21(n)	72.30	0.69	47.00	0.96	0.04	0.04	2.01	0.19	5.40	0.42	2.56
B22(n)	73.20	bdl	46.79	bdl	bdl	NA	2.09	0.08	5.40	0.18	bdl
B23(n)	26.25	0.31	14.69	bdl	bdl	NA	0.62	0.08	3.30	0.20	6.74
B24(n)	41.71	0.51	33.22	0.27	bdl	NA	2.32	0.21	3.30	0.55	6.02
B25(n)	33.34	0.57	21.19	0.10	bdl	NA	1.39	0.06	3.06	0.13	0.34
Descriptive statistics (whole lagoon)											
Min.	15.14	0.19	12.66	0.01	0.02	0.01	0.39	0.01	2.28	0.02	0.34
Max.	78.23	1.91	49.52	3.69	8.42	1.20	2.32	0.36	39.65	1.30	8.75
Mean	49.96	0.79	35.12	0.64	0.86	0.21	1.32	0.13	6.94	0.40	4.61
SE	4.36	0.12	3.00	0.20	0.76	0.11	0.14	0.02	1.92	0.08	0.80
BC	92.00	0.11	41.00	0.05	NA	NA	0.77	0.02	NA	0.10	1.00
EC	80.00	0.30	47.20	0.40	NA	NA	0.85	0.02	4.60	0.10	NA
EF	0.6	4.2	1.2	2.6	NA	NA	2.5	10.6	2.4	6.8	NA
Igeo	-1.5	2.3	-0.8	3.1	NA	NA	0.2	2.2	NA	1.5	1.6

Location of site in the lagoon is South (s) or North (n)

BC background concentrations, from GESAMP (1982), Salomons and Forstner (1984), and Sunderland et al. (2006); EC Earth's crust values (Turekian and Wedepohl, 1961), Igeo Geoaccumulation Index, EF enrichment factor, bdl below detection limits, NA not applicable/available

lagoon, 0.17 µg g⁻¹ for Western lagoon), being 3.6× greater for the Eastern lagoon, and 12.3× greater for the Western lagoon.

Methylmercury Concentration in Sediments

Methylmercury concentrations in the sediments of the Eastern Lagoon varied between 0.02 and 8.42 ng g⁻¹, with

a median value of 0.13 ng g⁻¹, while in the Western lagoon they varied between 0.08 and 21.46 ng g⁻¹ with a median value of 0.12 ng g⁻¹ (Tables 1, 2). In the comparison of the northern and the southern shores of the lagoons, average MeHg contents in the southern shores of the lagoons (1.21 ng g⁻¹ Eastern lagoon, 2.33 ng g⁻¹ Western lagoon) were greater than those found in the northern shores of the lagoons (0.09 ng g⁻¹ for Eastern

Table 2 Western lagoon (Nyékonakpoè): concentrations (mean from triplicate) for each sampling site and descriptive statistics for the all sites, for total Hg, MeHg, trace elements, and TOC

Site	Element measured										
	Al (mg g ⁻¹)	Cd (µg g ⁻¹)	Fe (mg g ⁻¹)	Hg (µg g ⁻¹)	MeHg (ng g ⁻¹)	MeHg (%)	Mn (mg g ⁻¹)	Pb (mg g ⁻¹)	Ti (mg g ⁻¹)	Zn (mg g ⁻¹)	TOC (%)
N1(s)	49.60	1.82	39.10	2.68	21.46	0.8	0.70	0.22	4.56	0.92	10.62
N2(c)	64.63	1.34	45.46	1.11	0.12	0.01	1.32	0.17	4.98	0.56	7.45
N3(n)	42.08	0.68	24.06	0.02	bdl	NA	0.85	0.05	3.06	0.12	2.38
N4(n)	66.48	0.75	44.06	0.08	bdl	NA	1.55	0.07	4.32	0.21	3.66
N5(c)	66.00	1.31	41.75	0.81	0.10	0.01	1.39	0.17	5.16	0.60	8.02
N6(s)	63.73	0.63	41.68	1.35	0.62	0.04	0.93	0.21	5.46	0.95	9.86
N7(s)	64.15	1.35	44.34	1.64	0.54	0.03	1.47	0.20	4.98	0.65	8.63
N8(c)	81.46	1.07	47.77	1.22	0.07	0.01	1.63	0.13	5.81	0.40	5.00
N9(n)	70.93	0.33	51.97	0.05	0.08	0.16	1.01	0.03	4.56	0.07	1.51
N10(n)	68.12	1.10	38.54	0.53	bdl	NA	1.08	0.16	5.63	0.53	6.53
N11(c)	66.90	1.32	41.82	1.40	bdl	NA	1.39	0.20	5.16	0.59	8.09
N12(s)	68.39	1.44	49.10	2.63	0.11	0.004	1.39	0.19	5.10	0.60	6.61
N13(c)	65.00	1.29	40.71	0.75	bdl	NA	1.32	0.19	4.98	0.58	8.4
N14(c)	59.49	1.58	39.59	1.59	0.12	0.01	1.16	0.11	4.68	0.29	9.45
N15(c)	78.07	1.07	41.20	4.73	0.11	0.002	1.70	0.14	5.75	0.40	5.36
N16(c)	86.33	1.13	42.73	0.54	0.09	0.01	1.08	0.18	6.23	0.52	7.46
N17(c)	89.29	1.05	37.91	0.87	2.57	0.29	0.70	0.15	7.19	0.47	5.79
Descriptive statistics (whole lagoon)											
Min	42.08	0.33	24.06	0.02	0.07	0.002	0.70	0.03	3.06	0.07	1.51
Max	89.29	1.82	51.97	4.73	21.46	0.80	1.70	0.22	7.19	0.95	10.62
Mean	67.69	1.13	41.87	1.29	2.16	0.11	1.22	0.15	5.15	0.50	6.75
SE	2.86	0.09	1.45	0.29	1.76	0.07	0.08	0.02	0.22	0.06	0.62
BC	92.00	0.11	41.00	0.05	bdl	NA	0.77	0.02	NA	0.10	1.00
EC	80.00	0.30	47.20	0.40	NA	NA	0.85	0.02	4.60	0.10	NA
EF	0.9	4.5	1.1	3.8	bdl	NA	1.7	8.9	1.3	6.2	NA
Igeo	-1.0	2.8	-0.6	4.1	NA	NA	0.1	2.4	NA	1.8	2.2

Location of site in the lagoon is South (s), center (c) or North (n)

BC background concentrations, from GESAMP 1982, Salomons and Forstner (1984), and Sunderland et al. (2006), EC Earth's crust values (Turekian and Wedepohl, 1961), Igeo Geoaccumulation Index, EF enrichment factor, bdl below detection limits, NA not applicable/available

lagoon, 0.08 ng g⁻¹ for Western lagoon), being 13.3× greater for the Eastern lagoon, and 73.9× greater for the Western lagoon.

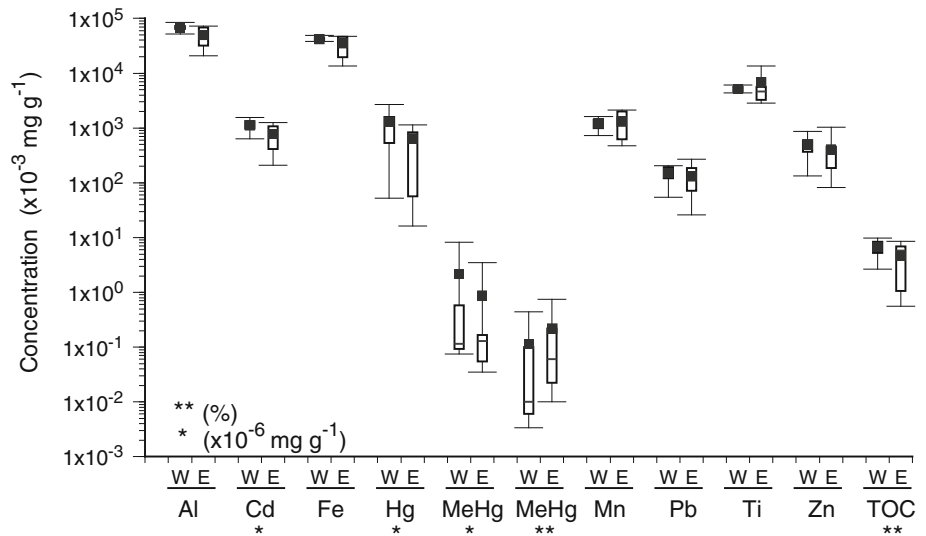
The fraction of MeHg in total Hg ((MeHg/Hg) × 100) ranged between 0.01 and 1.20%, with an average of 0.21 ± 0.36% for the Eastern lagoon, and between 0.002 and 0.8%, with an average of 0.12 ± 0.23% for the Western lagoon. When comparing the southern and the northern shores of the lagoons, northern shores generally showed greater fraction of MeHg from the total Hg. Indeed, for the Eastern lagoon, MeHg percent values were 0.56 (northern part) versus 0.09 (southern part), while for the Western lagoon the values were less different, being 0.16 versus 0.11%, respectively. Such contrasting trends indicate that inorganic Hg content in

sediment was not a limiting factor for MeHg production in both the lagoons.

Trace Elements Concentrations in Sediments

Trace elements concentrations were variable across the various shores of both the studied lagoons, thus sometimes covering a wide range of concentration values (Tables 1, 2; Fig. 2). Aluminum and iron were the most abundant elements, their concentration ranging from 15.14 to 89.29 mg g⁻¹ for Al, and 12.66 to 51.97 mg g⁻¹ for Fe. Titanium, manganese, and cadmium showed the second highest concentrations values, ranging from 2.28 to 39.65 mg g⁻¹ for Ti, 0.38 to 2.32 mg g⁻¹ for Mn, and 0.19 to 1.91 µg g⁻¹ for Cd. These elements were followed by zinc and lead with concentration

Fig. 2 Box plot representation showing variation of concentrations for trace elements and TOC in sediments for Bè, the Eastern lagoon (E), and for Nyékonakpoè, the Western lagoon (W). Error bars represent deciles (10th and 90th percentiles), boxes represent quartiles (25th, 50th, and 75th percentiles), and the filled squares represent the means



values ranging from 0.24 to 1.30 mg g⁻¹ for Zn and 0.01 to 0.36 mg g⁻¹ for Pb (Tables 1, 2).

In general, elemental concentrations from the sediment samples were also different between the southern and the northern shores of the lagoons. In the Eastern lagoon, values for the southern shores were about 1.5 × greater for Al, Fe, Mn, Ti, and TOC, and 2 × greater for Cd, Pb, and Zn. In the Western lagoon, the southern shores also showed greater concentrations, with values about 2 × greater for Cd, 2.5 × greater for Pb and TOC, and 3.5 × greater for Zn. As for concentrations of Al, Fe, Mn, and Ti, they were similar between southern and northern shores.

EF and Geoaccumulation Index

The EF was >1 in most cases indicating that significant amount of trace elements originated from anthropogenic source rather than being part of the geological matrix. The EF values were in a comparable range between the two lagoons (Tables 1, 2), except for Hg that was in a lower range bracket for the Eastern lagoon (EF_{Hg} = 2.6) when compared to the Western lagoon (EF_{Hg} = 3.8), and Pb that showed the opposite trend, being in a higher range bracket for the Eastern lagoon (EF_{Pb} = 10.6) when compared to the Western lagoon (EF_{Pb} = 8.9) (Tables 1, 2).

The geoaccumulation index was >0 in most cases (except for Al and Fe; Tables 1, 2) suggesting that most elements occurred in concentrations greater than pristine lagoons of reference. Igeo values were found in comparable range brackets between the two lagoons, except for Hg with values lower in the Eastern lagoon (Igeo_{Hg} = 3.1) when compared to the Western lagoon (Igeo_{Hg} = 4.1). Such difference was also observed for TOC with

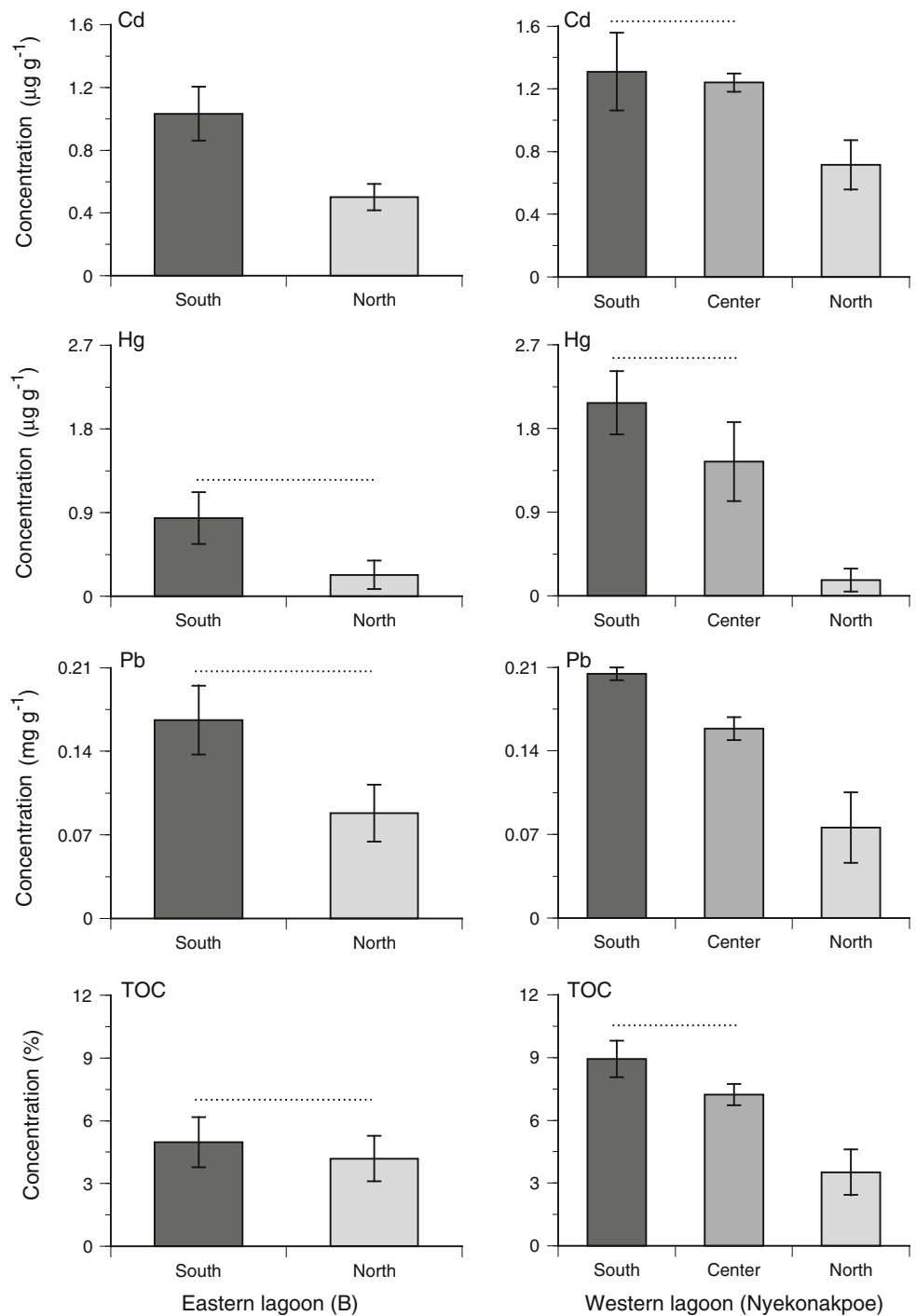
Igeo_{TOC} = 1.6 for the Eastern lagoon, and Igeo_{TOC} = 2.2 for the Western lagoon (Tables 1, 2).

Correlations Among Elements and TOC Concentrations

In general, there was some correlation among the elements, and between the elements and the TOC concentrations (Table 3A, B). The correlation coefficients, however, were often different between the two lagoons. In the Eastern lagoon, there was a high correlation (r = 0.88) between Hg and its methylated form MeHg, which was not the case in the Western lagoon (r = 0.34) (Table 3A). In the Eastern lagoon, there was often a positive correlation between the major geological elements like Al and Fe and the other trace elements which, however, was not the case for the Western lagoon (Table 3A). This was also the case between Al and Fe, with a strong correlation (r = 0.92) for the Eastern lagoon, and a much weaker one (r = 0.48) for the Western lagoon. For both the lagoons the correlation was high between Pb and Zn (r = 0.95 and 0.94) while low between Pb and Mn (r = 0.17 and -0.08). Pb otherwise showed reversed trends of correlations between the two lagoons for the other metals, having no significant correlation with Cd and Ti in the Eastern lagoon (r = 0.01 and -0.08, respectively), while these correlations are significantly greater in the Western lagoon (r = 0.64 and 0.47, respectively) (Table 3A).

In the Eastern lagoon, correlation with TOC was always low and non-significant, being positive only for Cd and Mn, while negative for MeHg and Ti; almost the reverse was found in the Western lagoon with correlation greater and significant with TOC, being positive for Hg, MeHg, Pb

Fig. 3 Concentration (mean \pm SE) of Cd, Hg, Pb, and TOC for the South–North sample locations of the Eastern lagoon (Bè), and for the South–center–North sample locations of the Western lagoon (Nyékonakpoe). Dotted lines group sites with no statistical difference in concentrations



Cd, Ti, and Zn, while non-significant for Al, Fe, and Mn (Table 3A).

Correlation analysis was also completed considering the southern and the northern parts of the lagoons. Correlation coefficients were usually distinct between the two parts, and different from the Eastern–Western lagoon analysis. Interesting correlations to highlight include the one between Hg and MeHg, being highly significant for the southern parts

($r = 0.71$) while not significant for the northern parts ($r = -0.05$) (Table 3B). In general, correlation of the analyzed elements with Fe and Al was relatively low for the southern parts while greater for the northern parts, while both Fe and Al were highly correlated with each other in both the southern and the northern parts ($r = 0.80$ and 0.96 , respectively). The element Mn showed negative correlations with Hg ($r = -0.38$), MeHg ($r = -0.43$), and Pb

Table 3 Pearson’s correlation coefficients (*r*) between the concentrations of each analyzed elements in (A) the Eastern Lagoon (Bè), and the Western Lagoon (Nyékonakpoè), and (B) the southern and northern parts of the lagoons

	Hg	MeHg	Pb	Cd	Fe	Al	Ti	Mn	Zn	TOC
(A) Eastern and Western lagoons										
Eastern lagoon										
Hg	1.00	0.88	0.75	−0.09	0.42	0.48	−0.12	0.01	0.78	−0.08
MeHg		1.00	0.55	−0.24	0.11	0.14	−0.03	−0.24	0.61	−0.21
Pb			1.00	0.01	0.56	0.61	−0.08	0.17	0.95	−0.12
Cd				1.00	0.34	0.13	0.18	0.13	−0.08	0.22
Fe					1.00	0.92	−0.02	0.72	0.45	0.05
Al						1.00	−0.11	0.65	0.52	0.07
Ti							1.00	−0.18	−0.26	−0.29
Mn								1.00	0.03	0.21
Zn									1.00	−0.06
TOC										1.00
Western lagoon										
Hg	1.00	0.34	0.40	0.46	0.05	0.07	0.22	0.34	0.37	0.39
MeHg		1.00	0.40	0.57	−0.08	−0.35	−0.09	−0.47	0.52	0.43
Pb			1.00	0.64	0.03	0.11	0.47	−0.08	0.94	0.88
Cd				1.00	−0.14	−0.14	0.13	−0.01	0.49	0.76
Fe					1.00	0.48	0.30	0.34	0.05	0.04
Al						1.00	0.88	0.18	−0.02	−0.09
Ti							1.00	−0.06	0.36	0.27
Mn								1.00	−0.22	−0.10
Zn									1.00	0.85
TOC										1.00
(B) Northern and southern parts										
Southern parts										
Hg	1.00	0.71	0.71	0.01	0.15	0.28	−0.22	−0.38	0.77	0.21
MeHg		1.00	0.34	0.29	−0.08	−0.12	−0.09	−0.43	0.45	0.29
Pb			1.00	−0.18	0.26	0.46	−0.11	−0.36	0.93	−0.14
Cd				1.00	0.19	−0.15	0.17	−0.07	−0.14	0.35
Fe					1.00	0.80	0.01	0.63	0.16	0.17
Al						1.00	−0.16	0.47	0.39	0.18
Ti							1.00	−0.12	−0.32	−0.26
Mn								1.00	−0.45	0.16
Zn									1.00	0.09
TOC										1.00
Northern parts										
Hg	1.00	−0.05	0.73	0.49	0.43	0.48	−0.04	0.49	0.64	0.13
MeHg		1.00	0.06	0.16	0.25	0.12	−0.14	−0.14	0.14	0.08
Pb			1.00	0.56	0.44	0.44	−0.18	0.69	0.97	0.44
Cd				1.00	0.31	0.40	−0.21	0.15	0.66	0.51
Fe					1.00	0.96	−0.20	0.68	0.45	−0.10
Al						1.00	−0.21	0.63	0.46	−0.02
Ti							1.00	−0.23	−0.27	−0.38
Mn								1.00	0.60	−0.11
Zn									1.00	0.57
TOC										1.00

Correlations of interest are highlighted in *bold*

($r = -0.36$) in the southern parts, while these correlations were positive with Hg ($r = 0.49$) and Pb ($r = 0.69$) and not significant with MeHg ($r = -0.14$) in the northern parts (Table 3B). Correlations for Zn were high with Pb in both the southern and the northern parts ($r = 0.93$ and 0.97 , respectively) while negative with Mn ($r = -0.45$) in the southern parts and positive ($r = 0.60$) in the northern parts (Table 3B). Correlations with TOC were positive for Al ($r = 0.18$), Fe ($r = 0.17$), Hg ($r = 0.21$), MeHg ($r = 0.29$), and Mn ($r = 0.16$) in the southern parts, and not significant in the northern parts being Al ($r = -0.02$), Fe ($r = -0.10$), Hg ($r = 0.13$), MeHg ($r = 0.08$), and Mn ($r = -0.11$). The reverse was observed for Pb ($r = -0.14$) and Zn ($r = 0.09$) in the southern parts with values increasing to Pb ($r = 0.44$) and Zn ($r = 0.57$) in the northern parts (Table 3B).

DISCUSSION

The lagoons of Lomé were found to have sediment contaminated with various trace elements. In general, sediment from the Eastern lagoon (Bè) showed the tendency to be less contaminated than that of the Western lagoon (Nyékonakpoè), yet the sediment in the Eastern lagoon was more heterogeneous as indicated by a high variability across different sampling sites. Such variability also includes the fact that for each lagoon the southern shores were more contaminated than the northern shores. Overall, the contamination levels were usually high considering that the EF values were always >1 (except for Al); for the eastern lagoon the values indicated pollution that was severe for Pb, moderately severe for Zn, and moderate for Cd. As for the Western lagoon, the EF values indicated pollution moderately severe for Pb and Zn, and moderate for Cd and Hg. Similarly, Igeo values were always >1 (except for Al, Fe, and Mn), thus also indicating anthropogenic input. For the eastern lagoon, Igeo values were indicative of strong pollution for Hg, moderate to strong pollution for Cd and Pb, and moderate pollution for Zn and TOC. Similarly for the Western lagoon, Igeo values were indicative of strong to very strong pollution for Hg, moderate to strong pollution for Cd, Pb, and TOC, and moderate pollution for Zn. Thus, in general for most elements but in particular for Cd, Hg, Pb, and Zn, the levels found associated with the sediments originated from anthropogenic sources (GESAMP 1982; Salomons and Forstner 1984; Sunderland et al. 2006).

Recent grain size analysis of the lagoons (Table 4) also suggests an important contribution of terrigenous material originating from runoff of surrounding watersheds. Indeed, the lagoons contain mainly fine silt/clay material with levels of Al similar to geological background levels, or

Table 4 Sediment seafloor characteristics of the lagoons in Lomé, Togo, measured in August 2010 from five sites across each lagoon, along an E–W axis

	Eastern lagoon	Western lagoon
Redox potential	-82.2 ± 8.9 mV	-76.8 ± 5.5 mV
pH	8.6 ± 0.3	8.4 ± 0.3
Temperature (°C)	26.7 ± 1.2	26.6 ± 1.1
Grain size		
1000–500 μm	$0.7 \pm 0.4\%$	$0.7 \pm 0.2\%$
500–250 μm	$1.1 \pm 0.6\%$	$0.9 \pm 0.3\%$
250–125 μm	$1.7 \pm 0.3\%$	$1.2 \pm 0.6\%$
125–63 μm	$3.3 \pm 0.9\%$	$1.8 \pm 0.8\%$
<63 μm	$93.0 \pm 1.4\%$	$95.1 \pm 2.0\%$

Redox potential, pH and T were measured from sediment that was collected free-diving, using a hand-core PVC tube closed at each end immediately after collection underwater. Measurements were completed at the surface in a boat using a handheld Myron L Ultrameter PS6 (Carlsbad, CA, USA)

Grain size analysis was performed on the sediment samples oven-dried at 70°C and homogenized with a piston and mortar, and weighted using a Sartorius CP224S microbalance. Size fractions of sediment were then separated through Tyler sequential sieving of increasing mesh size, and the cumulative weight percent retained/passing through each sieve expressed in relative percent to the total sediment sample before sieving

lower (EF ranging from 0.6 to 0.9 for the Eastern and Western lagoon, respectively). This suggests that most of the fine material has a geological origin, while also containing particles from anthropogenic sources, whether related to industry or urban activities. The correlation coefficients between metals and Al (considered a proxy of terrigenous material) are clearly different between the two lagoons, ranging from $r = 0.14$ to 0.92 for the Eastern lagoon, and $r = -0.35$ to 0.48 for the Western lagoon, suggesting different ratios of natural versus anthropogenic source of metals to each lagoon, considering fine-grained material as the common carrier to these metals.

Possible Sources of Contamination for the Lagoons of Lomé

There are multiple possible sources of contamination for the lagoons of Lomé, and if identification of such sources goes beyond the goal of this study, there are several historic and current site characteristics to be noted to help understand some of the observed patterns of contamination. For example, it is known that historically the city of Lomé started developing along the southern shores of the lagoon before expanding on their northern shores, which happen after the 1960s (Fig. 1). Such urbanization development from South to North could therefore contribute to the southern shores being more contaminated, as a result from older and larger exposure to possible sources of

contamination when compared to the northern shores. In addition to this historical factor, current sources include about 50 official (and innumerable illegal ones) urban channels for sewage/runoff output that are opening out directly into both lagoons from the southern side. In contrast, the northern shores of the lagoons receive only six of such channels.

As for the difference between the Eastern (Bè) and the Western (Nyékonakpoè) lagoons, it must be noted that the Eastern lagoon was subject to intense dredging activity as a clean-up program in the 1990s (Gnandi et al. 2007), which might contribute to the lower contamination observed there today. However, the Western lagoon could also show greater contamination, in particular for Cd, Zn, and Hg, because (1) this lagoon is the recipient of a major wastewater effluent from the neighboring hospital of Lomé, and (2) this lagoon appears to have different physico-chemical processes driving the partitioning (e.g., adsorption to particles versus dissolution in seawater) and spatial distribution of metals, thus affecting their fate in whether they are rendered bioavailable and enter the local fauna/flora, or reach the open ocean while adsorbed to fine particles or dissolved in seawater. Indeed, the sediment and water column in the Western lagoon clearly appear to contain high amounts of decaying organic material and a large portion of fine clay. Measurements subsequent to this study showed that sediment in the Western lagoon is highly reducing (probably due to microbial activity and organic decay) with redox potential around -80 mV, and grain size content made of $>90\%$ fine particles (Table 4), indicative of continuous accumulation of contaminants entering the lagoon. Accordingly, this was reflected in our data by greater concentrations of major elements like Al and Fe, and by increased TOC in the Western lagoon. The Al–Fe correlation was much weaker in the Western lagoon, suggesting sources other than strictly geological for these elements in that particular lagoon. The Western lagoon also seems to experience less tidal flushing circulation with the coastal waters (hence the eutrophication events repeatedly observed), which takes place through a discharge channel that is longer for the Western lagoon when compared to the Eastern lagoon, and thus less exposed to fresh oceanic seawater input. As a result, lagoonal waters from the Western lagoon might have a greater residence time when compared to the Eastern lagoon, which might contribute to the observed differences (Chadwick and Largier 1999; Chadwick et al. 2004).

Greater content in clay, and higher concentrations in Al and Fe, could favor solid phase retention of other trace elements (El Nemr et al. 2007; Preda and Cox 2002). This was not observed here as shown by the relatively low correlation coefficients found for these metals in the Western lagoon. However, the Western lagoon also

contains more decaying organic material and, accordingly, greater concentrations of TOC, which could interfere and compete with the metal adsorption on clay mineral (Clemente et al. 2006). This could explain the strong correlation between TOC and Pb–Cd–Zn in the Western lagoon (Clemente et al. 2006), while such correlation was not present in the Eastern lagoon.

Correlations of metal concentrations with that of Zn also lead to the evidence that the industry associated with automobiles and traffic (including tire abrasion, gasoline particulate exhaust, and waste batteries and oil disposal) represents an important source of metals (Sherwood 2005). Indeed, in both the lagoons the Zn enrichment was moderately severe, and the correlation high between Pb and Zn ($r = 0.95$ and 0.94 ; Table 3A), indicating their common origin from automobiles by-products entering the lagoon from surrounding watersheds. This was also evidenced by strong correlations between Pb and Zn in both the southern and the northern parts of lagoon ($r = 0.93$ and 0.97 , respectively; Table 3B). It is known that Pb can occur in aqueous ionic form as Pb^{2+} and in various hydroxides such as $Pb(OH)^+$, $Pb(OH)_2$, $Pb(OH)_3^-$, and $Pb(OH)_4^{2-}$, yet all these forms are known to have limited solubility under the common physico-chemical conditions found in natural waters (Fetter 2009). Like other metals such as Zn, Pb is actually expected to undergo cation exchange with clays in fine fraction, which is dominant in lagoon sediments. Both Pb and Zn will therefore show strong adsorption to clay particles in this environment and likely co-precipitate by chelation with organic matter (evidenced by high correlation between Pb–Zn and TOC in the Western lagoon), or by formation of carbonates or through sorption by hydrous oxides of iron and magnesium (evidenced by high correlations between Pb–Zn and Fe in the Eastern lagoon).

Correlations Between Contaminants and TOC

TOC concentrations were greater for the Western lagoon when compared to the Eastern lagoon, and in each lagoon, greater in the southern side compare to the northern side. Such variability in TOC concentration could therefore influence the physico-chemistry of trace elements, which is especially true for the methylation of mercury (Hammerschmidt and Fitzgerald 2006). Accordingly, no to little correlation was found for the Eastern lagoon and the northern parts of the lagoons between TOC and total Hg and MeHg, while these correlations were greater for the Western lagoon and the southern parts of the lagoons suggesting that particulate organic matter could be a significant sink for Hg and MeHg in these estuarine surface sediments (Skylberg et al. 2006).

In addition to TOC, reduced sulfur, and in particular acid-volatile sulfides (AVS), has been reported as a major

sink of Hg and trace elements in anoxic estuarine sediments (Niessen et al. 2003). The Eastern lagoon was subject to dredging activity about 15 years ago, and this dredging activity may have removed or weakened surface anoxic zone which in turn would have affected the chemical form of contaminants, including sulfur and its occurrence as acid-volatile sulfides. AVS usually consists of a complex mixture of reduced sulfur components that can build up in high concentrations with greater organic content and lower oxygen levels in sediment (Rickard and Morse 2005). AVS content is often dominated by Fe and Mn sulfides that can have high affinity for other metals for which they can act as strong scavengers, thus limiting their bioavailability (Cooper and Morse 1998). As for Mn, it showed positive correlation with Hg in the Western lagoon and the northern parts of the lagoons, while the correlation was negative for MeHg, indicative of Hg co-precipitation with Mn and lower bioavailability for methylation. Negative correlation with MeHg was also observed for the Eastern lagoon or the southern parts of the lagoons, despite showing positive correlation with Mn, suggesting that other elements (whether metals or organic matter) could then participate to the decrease bioavailability of Hg. In addition, high correlation between TOC, and geological metals like Al, Fe, and Zn might be caused by an organic adsorption on clay, and aluminosilicate minerals (El Nemr et al. 2007; Preda and Cox 2002). For example, the fact to have relatively less Al, Fe, Zn, and TOC in the Eastern lagoon could contribute to the greater bioavailability and methylation of Hg observed there. These processes, however, result from a complex interaction among multiple factors that is difficult to identify since changing locally for different ranges of physico-chemical conditions and/or ratios of elemental concentrations (Sherwood 2005). The greater bioavailability of Hg due to lesser concentrations of TOC indicated for the Eastern lagoon was not observed when considering northern and southern parts of the lagoons. Indeed, the correlation between Hg and MeHg was significantly positive ($r = 0.71$) in the southern parts that show higher levels of TOC while not significant ($r = -0.05$) for the northern parts with lower levels of TOC (Table 3B). This indicated that the southern parts of the lagoons, exposed to larger organic contamination, are sites of larger mercury methylation for the lagoons.

Mn is one of the only metal that often exhibited higher concentration in the Eastern lagoon when compared to the Western lagoon (Tables 1, 2). Differences in historical sources of Mn could be responsible for such discrepancy between the two lagoons. However, it must be noted that the Eastern lagoon is adjacent to the airport of Lomé, with thus the possibility that Mn originates from airborne deposition related to fuel combustion of airplanes (Fang et al. 2007, 2010). Indeed, Mn is increasingly used as a

substitute of Pb in octane and other high-power fuels, and is spread through atmospheric deposition around airports (Boyes 2010; Butcher 2002; Kaiser 2003; Winder et al. 2010). Evidences for this possibility are that Mn is one of the two metals (with Al) found equally concentrated between the northern and the southern part of the lagoons, and also one of the only few (with Pb and Zn) to have EF values greater for the Eastern lagoon when compared to the Western lagoon. Interestingly, there was no significant correlation between Pb and Mn for both the Eastern and the Western lagoons, while such correlation was positive for the northern parts (airport side) of the lagoons, possibly reflecting their common origin, while the southern parts (city side) of the lagoons showed negative correlation, indicative of different levels and sources of contamination (Table 3B).

Mercury Contamination is High in the Lagoons of Lomé

Mercury contamination was different between the Eastern and the Western lagoons, but also within each lagoon between the southern and the northern side of the lagoons, as driven by the possible sources of contamination and the TOC concentration of which mercury is highly dependent. For the southern shores of the lagoons, total Hg concentrations ($x = 842 \text{ ng g}^{-1}$ for the Eastern lagoon; $x = 1,640 \text{ ng g}^{-1}$ for the Western lagoon) were usually higher than values given in literature for urbanized and industrialized estuarine sediments (Table 5). In contrast, the average MeHg concentrations (1.2 ng g^{-1} for Eastern lagoon; 2.3 ng g^{-1} for Western lagoon) were comparable or slightly higher than the typical values found in urbanized estuarine sediments (Table 5).

Regarding the northern shores of the lagoons, the average total Hg concentrations were comparable to those found in urbanized and in industrialized estuarine sediments ($x = 230 \text{ ng g}^{-1}$ for Eastern lagoon; $x = 169 \text{ ng g}^{-1}$ for Western lagoon). In contrast, the average MeHg concentrations ($x = 0.09 \text{ ng g}^{-1}$ for Eastern lagoon; $x = 0.08 \text{ ng g}^{-1}$ for Western lagoon) were lower than typical values found in urbanized and in industrialized estuarine sediments (Table 5).

Methylation of Inorganic Hg is Variable in the Lagoons of Lomé

Despite elevated concentrations of mercury, methylation of inorganic mercury into methylmercury was clearly not very active in both the lagoons. The ranges of %MeHg/Hg in sediments ($x = 0.21\%$ for Eastern lagoon; $x = 0.12\%$ for Western lagoon) were relatively low when compared to those reported for “active” marine sediments ($x = 0.47\%$;

Table 5 Total Hg and MeHg concentrations (mean and range), and percent of methylated mercury (MeHg/Hg) in sediment for various places around the World

Location	Total Hg (ng g ⁻¹)	MeHg (ng g ⁻¹)	MeHg/Hg (%)	References
Southern New England shelf	20.2 (8.6–31.7)	0.148 (0.072–0.227)	0.74	Hammerschmidt and Fitzgerald (2006)
Bay of Fundy, Canada/U.S.	42.1 (10–140)	0.311 (0.050–1.48)	0.74	Sunderland et al. (2006)
Arctic Ocean	72.2 (34.1–116)			Gobeil et al. (1999)
Lagoon of Bizerte, Tunisia	104 (8.02–646)	0.465 (0.08–2.93)	0.45	Mzoughi et al. (2002)
San Francisco Bay, California	221 (20–702)	0.501 (0.02–3.41)	0.23	Conaway et al. (2003)
Seine River Estuary, France	461 (301–1000)	2.41 (0.1–6.02)	0.52	Mikac et al. (1999)
Venice Lagoon, Italy	1,306 (100–3,417)			Berto et al. (2006)
Venice Lagoon, Italy	729 (209–1,144)	0.96 (0.31–1.7)	0.13	Han et al. (2007)
Minamata Bay, Japan	3250 (341–4830)			Tomiyasu et al. (2006)
Bering Sea	88.26 (78.23–112.33)	0.21 (0.05–0.62)	0.24	Kannan and Falandysz, (1998)
Baltic Sea	92.27 (38.11–312.92)	0.69 (0.04–2.01)	0.75	Kannan and Falandysz, (1998)
Lagoons of Lomé, Togo	965 (11–4,726)	1.51 (0.02–21.46)	0.16	This study

Adapted from Fitzgerald et al. (2007)

(Fitzgerald et al. 2007). This suggests two possibilities: (1) activity of the microbial community associated with the sediment and capable of methylation was low, which is probably not the case considering that mercury methylation is a widespread phenomenon even more intense in the tropics; and/or (2) solubility and bioavailability of inorganic Hg was decreased probably through association with sedimentary organic carbon and/or acid-volatile sulfides, and in particular FeS, the mercury then occurring in a particulate form, which is not bioavailable to Hg-methylating organisms and inhibits MeHg production (Hammerschmidt and Fitzgerald 2004; Liu et al. 2008). In agreement with the last hypothesis, the fraction of Hg as MeHg was lower in the Western lagoon sediments when compared to those of the Eastern lagoon that contains less organic material. Similarly, MeHg concentrations were relatively more important in the northern shores of the lagoons despite relatively lower Hg concentrations. This discrepancy is probably related to the fact that TOC concentration is also lower in the northern shores of the lagoons, thus providing less retention of the mercury into a non-bioavailable form, therefore allowing more mercury methylation by microbes. Such a negative correlation between %MeHg/Hg and total Hg concentrations is similar to the one reported in the San Francisco bay-delta (Choe et al. 2004).

Despite %MeHg/Hg being generally low for both the lagoons, it was clear that its value was highly variable within each lagoon, reaching up to 1.2 and 0.8% for the Eastern and Western lagoon, respectively. Such levels can be considered relatively high when compared to other localities (Table 5), yet seem to reflect localized seafloor heterogeneity rather than being indicative of particular

lagoon-wide processes. Indeed, in the Eastern lagoon, highest levels of %MeHg/Hg were found for B3 and B6, both actually having low levels of Hg, 100× less than for some other samples (Table 1). If both B3 and B6 were located at the northern part of the lagoon, all other sites sampled in that part showed lesser %MeHg/Hg values, thus not supporting the trend that the northern part of the lagoon would systematically have greater %MeHg/Hg values. Accordingly, the highest levels of %MeHg/Hg in the Western lagoon were found for the southern to mid-part of the lagoon (N1 and N17) that also did not show the highest concentrations in Hg (Table 2). Such heterogeneity and seemingly random distribution of areas with intense MeHg production seems therefore related to localized and restricted conditions not necessarily involving high Hg concentration, and possibly related to past dumping, underground water seeping, and/or proximity to discharge channels.

Methylmercury is predominantly synthesized by sulfate-reducing bacteria (SRB) in anoxic sediments (King et al. 1999). Even though a few papers have reported that activity of SRB controls Hg methylation rate in anoxic sediments (Choe et al. 2004; Han et al. 2007; King et al. 1999), sediment speciation of Hg seems to control the MeHg production in our sampling locations. In reference to contamination thresholds proposed for MeHg in sediments (Salomons and Forstner 1984), most of the sites sampled here could produce adverse effects on biota. Despite the latter statement, fishing continues to these days by local communities in the entire lagoon systems of Lomé. Such situation highlights the need to implement environmental monitoring of the lagoons as well as regulation of the waste discharges occurring in the lagoon.

CONCLUSION

The sediments from the lagoons of Lomé contain high concentrations of trace elements, including mercury and methylmercury. Concentrations of the elements showed the tendency to be greater for the Western lagoon when compared to the Eastern lagoon, and especially greater for the southern shores when compared to the northern shores. Such variation in concentration was attributed to sources of contamination that were greater in the southern shores of the lagoon, but also to chelation with organic carbon. Indeed, trace elements can be immobilized through trapping processes involving sedimentary organic carbon and/or acid volatile sulfides, which could explain the Eastern–Western lagoons differences, but also those observed between the southern–northern parts. The increased %MeHg/Hg in the dredged Eastern lagoon sediments also suggests that clean-up dredging activity carried out there in the past could actually be responsible for increasing production of MeHg, by decreasing the amount of organic carbon, and thus the trapping capacity of the lagoon. The cleansing aspect usually associated with dredging is therefore misleading in this particular case of Hg methylation in the Eastern lagoon. Through calculation of EFs anthropogenic activities were shown to be the main source of the contamination input in the lagoons. Such activities include those associated with the nearby airport as well as about 50 official urban waste discharge channels (and innumerable illegal ones) that are located in the southern shores of the lagoons. This side of downtown Lomé is densely populated, and the numerous homes lacking modern waste collection facilities, probably also contribute to organic contamination of the lagoon.

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