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Spatial and Seasonal Trends of Trace Metals Diffusive Fluxes in Ebrié Lagoon, Côte d'Ivoire

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ABSTRACT

The aim of the present study was to determine the fluxes of five trace metals (Pb, Cd, Ni, Mn and Zn) in the Ebrié lagoon. Physicochemical parameters were measured *in situ* in both water and interstitial waters of sediment at each sampling station. The daily diffusive fluxes of metals were determined according to Fick's First Law of Diffusion. Pb, Cd, Ni, Mn and Zn fluxes were in the following respective ranges: -24.75 - 8.23, -3.41 - 0.16, -7.55-0.93, -6.36-0.31 and -529.98-1053.22 $\mu g.m^{-2}.d^{-1}$ in the flood season. In the dry season, respectives ranges of -25.50 to 10.27, -2.22 to 0.90, -10.76 to 3.62, -16.63 to 7.63 and -2028 to 671.53 $\mu g.m^{-2}.d^{-1}$ were observed for Pb, Cd, Ni, Mn and Zn. The following ascending rank were observed for metals fluxes: Cd<Ni<Mn<Pb<Zn. Significant differences ($p<0.05$) were observed between sampling stations in the two seasons. Apart from Cd, metal fluxes observed in the dry season were higher than the flood ones. The results of this study have reavealed that, at present, the major flux of metals was from water to sediments, with a release of a small part upward to the overlying waters. In Ebrié Lagoon, sediments act as a sink and as a source of metals. Sediments contamination with metals can be harmfull for marine organisms that are sensitive to physicochemical properties fluctuations in their living environment.

INTRODUCTION

Metals, because of their wide societal use and applications, are among the widespread environmental contaminants and are hazardous to human and non-human biota on one hand and on the other hand, are also persistent and bio-accumulated and magnified *via* food web and finally reach to human, fish consumers posing public health risks [1-3]. Thus, due to their hazardous effects on the equilibrium of aquatic systems and on human health, metals are studied worldwide in waters, in sediments and in marine organisms [3-14]. Metals are accumulated in marine organisms through a variety of pathways such as respiration, adsorption and ingestion [15]. Some epidemiological and experimental evidences indicate a range of toxic effects of Cd including carcinogenicity, mutagenicity and teratogenic effects in human and animals [16], while chronic environmental exposures to low levels of lead still reported in many developing and industrializing countries, particularly in urban areas [17]. In aquatic systems, sediments can occasionally act as a sink and/or source of metals where their fate and toxicity are greatly dependent on their partitioning (speciation) or their remobilization between solid phase and the dissolved one [7] [11], with an influence of diagenetic mechanisms [18]. Following the example what occurs in soils, diagenetic mechanisms are also influenced with several physicochemical properties such as pH, salinity, dissolved oxygen concentration, amount of suspended solids, redox potential, organisms activities, clay minerals content, sediment organic matter, Fe and Mn oxides, calcium carbonate, etc.[5], [19-22]. In aquatic systems, metals can reach the sediment by several mechanisms including adsorption, biogenic sedimentation of carbonate particles and sedimentation of particulate organic matter [11]. The release of metals from contaminated sediments to the water column occur by desorption, dissolution of biogenic carbonate particles and disintegration of particulate organic matter [11]. Due to the necessity of long term management of aquatic systems in a context of blue economy, the diffusive fluxes of pollutants in general, and particularly of metals, across the sediment-water interface are studied worldwide [3], [9], [11], [14], [23-24]. Metals diffusion is also one of the dominant transport mechanisms across the sediment-water interface in marine, coastal, and lake systems. Several studies conducted in Ebrié lagoon have revealed high concentrations of metals in waters and sediments collected at stations closed to Abidjan city, compared to samples collected farther, with spatial and seasonal variations [12], [25-26].

However, existing literature does not provide information regarding metals fluxes in the urban area of the Ebrié lagoon. The purpose of this study was to determine spatial and seasonal variations of the diffusive fluxes of lead (Pb), cadmium (Cd), nickel (Ni), manganese (Mn) and zinc (Zn) across the sediment-water interface in the urban area of the Ebrié Lagoon, around Abidjan city in the flood and dry seasons. It focuses on dynamics of transport of metals between the sediments and the overlying waters.

MATERIALS AND METHODS

Water and sediments samples were collected in 18 stations located in the Ebrié Lagoon, the largest lagoon in Côte d'Ivoire with an estimated area of 566 km^2 and stretches on 125 km along the coast, between 3°40' and 4°50' West, at latitude 5°50' North (Table 1) [10], [27]. Table No.1 presents the names and GPS coordinates of the different sampling stations. Surface and bottom waters and sediment (20 cm depth) samples were collected at each sampling station, using respectively a Niskin bottle and a stainless steel Van Veen Grab Sampler [10], [12]. Exact position of each sampling station was recorded using Global Positioning System (GPS) (Table 1). Organic matter contents were determined in sediments according to the chromic acid titration method [29]. In the laboratory and for each sediment sample collected, a first part was treated by centrifugation at a rate of 4000tr/min for 20 min. Then, samples were filtrated through Whatman GF/F (0.45µm) for interstitial water sample; while the second one was used to determine sediment porosity after drying at 60°C in an oven, in 48 h.

Table No. 1: Names and GPS coordinates of the sampling stations.

Bay	N°	Station	Latitude	Longitude	Bay	N°	Station	Latitude	Longitude
Banco	1	CARENA	5°20.031'	4°01.946'	Biétri	7	SIR	5°16.149	3°59.358'
	2	Sebroko	5°20.358'	4°02.183'		8	Bidet	5°15.598'	3°58.511'
	3	Bolibana	5°21.750'	4°02.548'		9	Abattoir	5°15.890'	3°58.333'
Cocody	4	Hôtel-Ivoire	5°19.396'	4°00.565'		10	Unilever	5°16.970'	4°00.203'
	5	BNETD	5°19.598'	4°00.898'		11	SIVOA	5°17.284'	3°59.540'
	6	Stade FHB	5°19.774'	4°01.025'		12	Marina	5°17.119'	4°00.071'
Milliardaires	15	Milliardaires1	5°15.989'	4°04.638'	Marcory	13	Grand C.	5°18.666'	4°00.101'
	16	Milliardaires2	5°15.998'	4°04.744'		14	Biafra	5°18.898'	4°00.383'
	17	Milliardaires3	5°16.989'	4°04.896'	Bingerville	18	Ana Ext.	5°19.500'	3°53.590'

Grand C.: *Grand caniveau,* Ana Ext: *Ana Extension,* CARENA : Compagnie Abidjanaise de Réparation et de Travaux Industriels ; SIVOA: *Société Ivoirienne d'Oxygène et d'Air Liquide* ;

BNETD : Bureau National d'Etudes Techniques et de Développement ; SIR : Société Ivoirienne de Raffinage ; FHB : Félix Houphouët-Boigny.

Trace metals concentrations were determined in both waters and interstitial waters with an Atomic Absorption Spectrophotometry flame technique (AAS) using a 3030 PERKIN ELMER instrument. Statistical analyses and graphs were performed with STATISTICA Software (2005, 7.1 Version) and Microsoft Office Excel 2007. Trace metals fluxes were calculated according to the following formulation [9], [11], [23] [30]:

$$J_D = - Ø \times Ds \times \Delta C/\Delta z \qquad (1)$$

Where:

- J_D = diffusive flux of trace metal (calculated in $\mu g.m^{-2}.d^{-1}$);

- Ø = sediment porosity (dimensionless);

- Ds = effective diffusion coefficient (in $m^2.d^{-1}$);

- $\Delta C/\Delta z$= metal concentration gradient across the *SWI ($\mu g.m^{-4}$);

*SWI: Sediment-Water Interface.

- Ds is the coefficient of diffusion of the sediment corrected to the temperature and tortuosity in the sediment;

- $Ds = Do/Ø^2$, with Do the molecular coefficient of diffusion of water [30]. Do values were calculated using the empirical formula by [31] corrected to the water temperature and sediment porosity [32], [23]. The value of $Ø^2$ is equal to (1 - ln ($Ø^2$) according to [32]. Thus, a positive flux of a metal indicate a release of the metal from the sediment to the overlying water, while a negative one represent the metal trap in sediment.

RESULTS AND DISCUSSIONS

Data relative to physicochemical parameters observed in waters and sediments are presented in Table No. 2. Temperature values observed in waters were higher than the sediments ones. pH values were in alkaline ranges in both waters and sediments samples and were found favorable for metallic complexes formation and metals precipitation from water to sediment. Temperature, pH, salinity and conductivity values were found safe for marine organisms

[33], [10]. Salinity and conductivity values observed in sediments and waters have confirmed the fact that the Ebrié Lagoon was diluted with waters from the Atlantic Ocean, through the Vridi Channel [10], [26]. The highest values of salinity was observed in sediment, while the conductivity one was observed in waters (Table No. 2). Organic matter contents observed in sediments were ranged from 8.05±7.32 (Ana Extension) to 67.37±14.07 % (SIVOA). Urban bays (Biétri, Banco, Cocody and Marcory) recorded high contents of organic matter in sediments, due to domestic, industrial and agricultural effluents inputs [10], [12].

Table No. 2: Physicochemical parameters ranges observed in Ebrié Lagoon.

Matrix	**Temperature (°C)**	***pH***	**Salinity (%)**	**Conductivity (mS.cm^{-1})**	**DO (mg/L)**	**Organic matter (%)**
Water	28.24-28.24	7.30-8.10	1.00-1,78	17.80-31.50	1,62-7,26	ND
Sediment	21.70-26.25	7.04-7.90	0.35- 2.11	8.24-30.90	ND	8.05- 67.37

ND: Not Detected

In sediment, organic matter can react as a binding agent with trace metals for the formation of aggregates. Highest organic matter contents in sediment can produce strong binding of trace metals according to the metal speciation and the overlying water physicochemical properties. The organic matter contents observed for this study were higher than 3.5%, the lowest in soil to avoid its erosion [34-35]. High contents of organic matter of sediments, associated with alkaline values of pH observed in Ebrié lagoon can lead to a loss of metals in the water column, increasing their concentrations in the sediments. High contents of organic matter can also increase metals complexation and/or their adsorption on sediments particles, influencing their speciation around the sediment-water interface.

Long term accumulation of metals in sediments may be harmful for benthic organisms such as mussels, in association with their speciation, in relation with physicochemical parameters such as pH, temperature, salinity, dissolved oxygen contents, etc. on one hand, and on the overhand, their release rate from sediment to the overlying water [5], [20-21]. Therefore, physicochemical parameters of the overlying waters may conduce to metals partitioning and/or remobilization between solid and dissolved phases, increasing hazardous effects on marine organisms. Study conducted by [10] showed high metals concentrations in waters collected in Ebrié Lagoon compared to the [33] recommended respective limits of 10, 3, 20, 400 and 3000 µg.L^{-1} for Pb, Cd, Ni, and Zn. No Ecological risk was observed for Mn that recorded low concentrations compared to the recommended limit value of 400 µg.L^{-1}.

According to these recommended values, and apart from Mn, waters were found to have hazardous effects on aquatic organisms [33]. Pb and Cd were of public concern due to their harmful ranges of concentrations on one hand, and on the overhand, to their no beneficial effect regarding the metabolism of living [10]. Even if Zn is known to be essential for the metabolism of livings beings, it becomes toxic at high concentration in aquatic systems. Ecological risks associated with metal contamination in sediments collected in Ebrié Lagoon were assessed with a pair of empirically derived numerical Sediment Quality Guidelines (SQGs), namely the Effects-Range Low (ERL), effects-range medium and Effects-Range Severe (ERS) [12]. Results of sediments contents in metals and details relative to effects (ERL and ERS) are available in a paper published by [12]. Results showed Pb, Cd, Ni, Mn and Zn concentrations above ERL values, and several samples contained concentrations higher than ERS values for Pb, Ni and Zn [12]. Thus, Pb, Cd, Ni and Zn may pose a high ecological risk for marine organisms and long-term management of the Ebrié Lagoon.

Average diffusive fluxes of metals across the sediment-water interface are presented in Table No. 3. Pb, Cd, Ni, Mn and Zn average fluxes were in the respective following ranges: -25.12-9.25, -2.50-0.53, -7.58-2.28, -11.49-3.84 and -1063.04-332.68 $\mu g.m^{-2}.d^{-1}$. Station SIR recorded highest average values of release rate of Pb, Cd, Ni and Mn from sediments to the overlying waters. Respective highest sedimentation values of Pb, Cd, Ni were observed at stations Bolibana, Bidet, Marina. Station Hotel-Ivoire recorded the highest trend of Mn and Zn to be trapped in sediments (Table No. 3).

Station Ana Extension (the farest station from Abidjan) recorded the lowest average of fluxes for the five metals studied (Tables 1&2). Highest values of metals fluxes were generally oberseved at stations located in Bietry and Cocody Bays, two of the most polluted ones of the urban area of the Ebrié Lagoon (Tables No. 1 & No. 3). According to the data regarding average fluxes (Table No. 3), a trend to a trap of metals from waters to sediment was observed in 2/3 of the all stations for Pb, Ni and Zn, against 1/3, where metals tended to be released from sediment to the overlying waters (Table No. 3). For the present study, at the 18 stations studied, Cd was tended to be trapped in the sediment at 13 locations against 5 with a trend of release from sediment to the overlying waters (Table No. 3).

Table No. 3: Daily mean flux of metals ($\mu g.m^{-2}.d^{-1}$) determined in the different stations.

	1	2	3	4	5	6	7	8	9
Pb	-1.06	-0.51	-4.27	-6.39	-9.65	-2.87	-25.12	4.6	4.04
Cd	-0.07	-0.08	0.26	0.43	-0.64	-0.67	-2.50	0.53	0.43
Ni	-0.31	-1.22	-4.36	-2.52	1.06	-2.12	-7.58	0.91	1.15
Mn	-0.07	0.24	-3.24	3.84	0.72	0.87	-11.49	1.81	-0.40
Zn	-64.91	32.88	-111.69	332.68	-1063.04	-392.56	-91.38	127.36	5.42
	10	**11**	**12**	**13**	**14**	**15**	**16**	**17**	**18**
Pb	-3.77	9.25	7.61	-3.61	-6.31	-0.19	-0.29	1.09	0.17
Cd	-0.06	-0.13	-0.28	-0.42	-1.56	-0.01	-1.59	-1.26	0.01
Ni	-0.98	1.46	2.28	-0.58	-5.5	-0.13	-3.66	-2.1	0.27
Mn	-0.12	-1.47	1.97	-0.49	0.42	0.04	-1.31	-2.48	0.11
Zn	-168.93	-567.84	-83.41	-428.6	-681.04	-60.67	82.73	16.6	-2.21

Mn was released from sediment to the water column at 50% of the stations against 50% with a trend of Mn to be trapped in sediment. Highest release rates were observed at station SIR for Pb, Cd, Ni and Mn, while Zn one was observed at station BNETD. One-way ANOVA F-variance analysis was used to capture respective levels of similarities and/or differences between the sampling stations (Table No. 4). Differences between stations were found significant ($p<0.05$) for the five metals, with strongest ones observed for Pb ($p=0.007$) and Ni ($p=0.008$) compared to the three other metals (Table No. 4). The following ascending rank of the metals fluxes observed at the different stations was observed: Cd<Ni<Mn<Pb<Zn. The fluxes of metals in Ebrié Lagoon were characterized by large spatial fluctuations, which may be attributed to localized metal inputs and dynamics of sediments under spatial variations in physicochemical parameters that occurred in overlying waters and into sediments [10-12; 19-22]. In aquatic ecosystems, variations of metals concentrations are commonly attributed to variations in silt-clay, Fe and Mn oxides and hydroxides as well as organic matter content in sediments, as these tended to concentrate metals into sediment. The mayor trend of Pb to be loss from the water column to the sediment is due to its strong affinity to Fe and Mn oxides and in particular to Mn oxides [36]-[38].

Table No. 4: One-way ANOVA of metals fluxes in differents locations.

Parameters	**Pb**		**Cd**		**Ni**	
	F	**p-value**	**F**	**p-value**	**F**	**p-value**
Locations	1.38	0.007	1.102	0.013	1.164	0.008
Parameters	**Mn**		**Zn**			
	F	**p-value**	**F**	**p-value**		
Locations	1.154	0.012	1150.678	0.034		

$p < 0.05$: Significant difference.

The spatial trends in metals fluxes in the Ebrié lagoon observed in the dry and flood seasons are presented (Figures No. 1-5). Pb, Cd, Ni, Mn and Zn fluxes were in the following respective ranges: (-24.75) - 8.23, -3.41 - 0.16, -7.55-0.93, -6.36-0.31 and -529.98-1053.22 $\mu g.m^{-2}.d^{-1}$ in the flood season. In the dry season, respectives ranges of -25.50 to 10.27, -2.22 to 0.90, -10.76 to 3.62, -16.63 to 7.63 and -2028 to 671.53 $\mu g.m^{-2}.d^{-1}$ were observed for Pb, Cd, Ni, Mn and Zn. Pb was trapped into the sediments at stations BNETD-SIR and Unilever, with highest values observed at station SIR (Figure No. 1). For the same period, a release of lead from sediment to the water column was observed at stations Bidet, SIVOA and Marina. Fluxes observed in dry season were higher than those observed in the food one, probably due to the waters dilution with those of continental origins followed with the metal desorption. Highest fluxes of Cd were observed in the flood season at stations Milliardaires 1&2 (located in Millardaires Bay) and at station SIR, with a trend to Cd trap into sediments (Figure No. 2). In the dry season, differences were observed between locations, marked with a release rate of Cd at stations Bolibana, Hotel-Ivoire, Bidet and Abattoir. Cd was lost from water to sediment at stations from BNETD to SIR in the dry season (Figure No. 2). In flood season, low rates of Ni were observed, apart from stations SIR, Milliardaires1&2, where the Ni was lost in the water (Figure No. 3). Ni fluxes observed in dry season were higher than those of the flood one, with highest values observed at stations located in Bietri Bay (Biafra, SIR and Bolibana), followed with stations into Cocody Bay (Hotel-Ivoire and Stade FHB). The release of Ni from the surficial sediment to the overlying water was only observed in the dry season, and at stations BNETD, and Bidet-Marina (Figure No. 3). Mn fluxes observed in the dry season were higher than those observed in the flood one. In the two seasons, a trend to Mn loss from water to sediments were observed at station SIR, where higest fluxes were also recorded (Table No. 2, Figure No. 4).

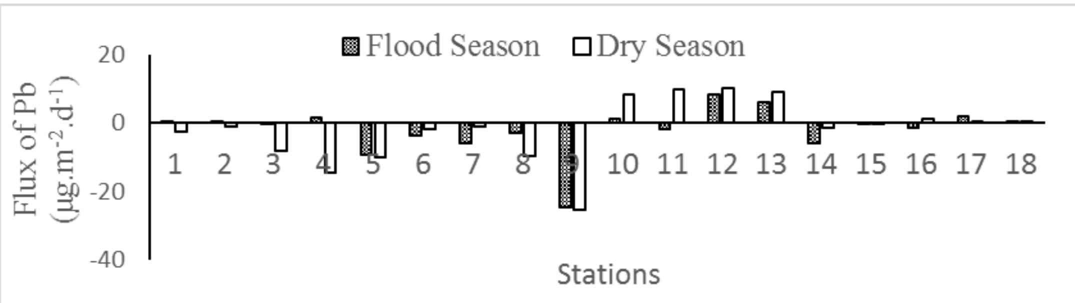

Figure No. 1: Flux of lead observed in the different stations for the two seasons.

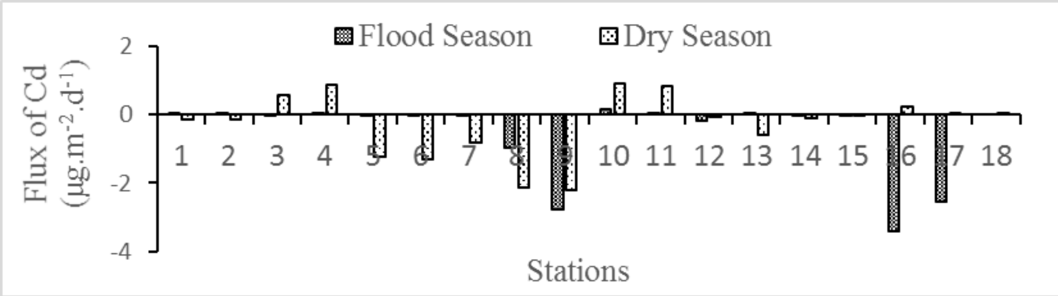

Figure No. 2: Flux of cadmium observed in the different stations for the two seasons.

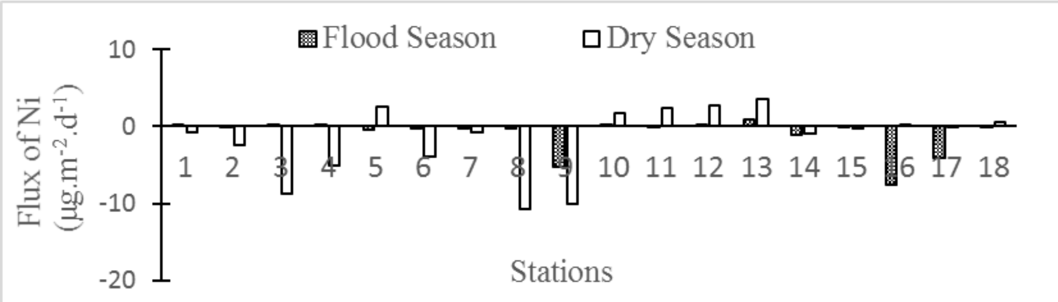

Figure No. 3: Flux of nickel observed in the different stations for the two seasons.

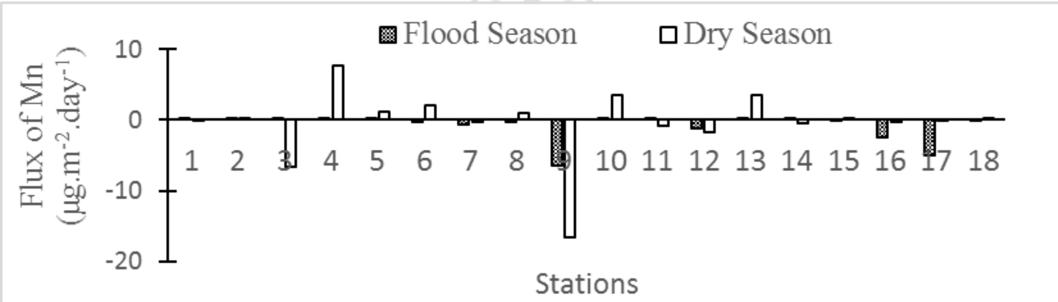

Figure No. 4: Flux of manganese observed in the different stations for the two seasons.

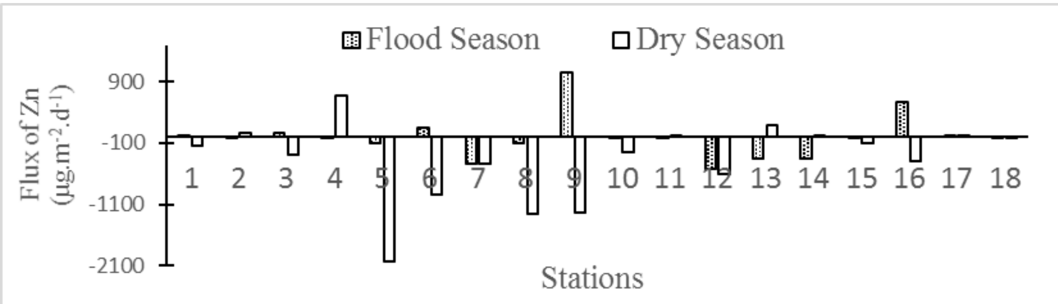

Figure No. 5: Flux of zinc observed in the different stations for the two seasons.

Apart from a few stations in dry season (Bolibana, BNETD, SIR, Bidet, Marina and Milliardaires 1&2 (Flood season), flux rates of Mn were found insignificant elsewhere (Figure No. 4). Zn fluxes were higher than the thourth (Pb, Cd, Ni and Mn) previous ones (Figure No. 5). In the flood season, apart from stations SIR, Grand caniveau, SIVOA, Marina, Unilever and Milliardaires2, fluxes rates of Zn were low elsewhere, with a release from sediment to the overlying waters at stations SIR and Milliardaires2 (Figure No. 5). Dry season recorded high fluxes of Zn, compared to the flood ones, with a general trendancy to the metal sedimentation, apart from station Hotel-Ivoire at wich a release rate was observed. Pb is finally stored in sediments due to its affinities to the formation of stable forms with solid particles [39]. Pb enters the Ebrié Lagoon waters through several pathways such as domestic and industrial effluents (the use of Pb in batteries) [40]. Pb accumulation in sediments can be harmful for benthic biota on one hand, and on the overhand to the lagoon equilibrium regarding the biota diversity such as oysters, disappeared from the polluted areas of the Ebrié Lagoon. High fluxes of Cd oberved in the flood season are due to agricultural effluents, industrial wastewaters (e.g., batteries), and runoff from upstream mining or smelting activities through the Comoé River waters that dilute the Ebrié Lagoon during this wet season [10], [12]. A mayor part of metals in Ebrié Lagoon may be of anthropogenic origin due to the fact that, in aquatic systems, anthropogenic metals are predominantly found in the most labile sediment fraction [41]. Metals such as Ni, Pb and Cd are considered to be carcinogenic to human by International Agency for Research on Cancer [42-43]. Therefore, these elements are likely to pose a direct and significant threat to the surrounding environment. Ebrié lagoon waters and sediments contamination with trace metals was attributed to the discharge of industrial and domestic effluents. In coastal areas, roof runoff may be one of the major sources of Zn contamination in aquatic systems [44].

Anthropogenic Zn input in sediments related to a recent pollution event and could have an ecological risk, particularly in the case of resuspension. Fujiyoshi and Katayama [45] showed a high affinity between Zn and carbonates, hydrous iron oxides, and silicate minerals. Metals, such as Zn and Pb can reach the sediment by sedimentation of detrital particles and co-precipitation with metal oxy-hydroxides [11]. High fluxes of metals observed in Ebrié Lagoon are the result of rapid urbanization associated with intensive industrial activities along the lagoon and uncontrolled dumps and several effluents that are finally diluted into the lagoon waters. Fluxes of metals observed in Ebrié Lagoon were compared to those of other authors worldwide (Table No. 5). Oueslati and Added [9] reported fluxes values of Pb, Cd, Ni and Mn higher in Ghar El Melh Lagoon than the Ebrié Lagoon ones (Table No. 4). Zn fluxes observed in Ebrié Lagoon were higher than the in Ghar El Melh Lagoon ones. However, apart from Cd that released from sediment to the overlying water, a trend to the sedimentation occurred for the other metals [9].

Table No. 5: Metal fluxes observed in different aquatic systems worldwide (in µg.m^{-2}.d^{-1}).

Pb	Cd	Ni	Mn	Zn	Reference
-25.49 – 10.27	-3.41 – 0.90	-10.76 – 3.62	-16.63 – 7.63	-2028.00 – 1053.22	**This study**
80.80 – 428.88	-34.84-10.12	1.17-48.73	315.91-6592.8	47.07-119.63	[9]
-0.92 - (-0.09)	-	-1.37 – 2.63	-	-180.26 – 7.51	[46]
0.41 – 0.68	0.003 – 0.027	0.027 – 0.301	-	0.41 – 0.63	[47]
0.01	-	0.015	-	0.005	[48]

Fuyang River recorded a release and sedimentation rates of Ni and Zn [46]; the same trend occurred in Ebrié Lagoon (Table No. 5). At the opposite, [47] and [48] have reported positive fluxes for all metals. The Conwry Estuary recorded the lowest fluxes of all the metals [48].

CONCLUSION

Spatial and seasonal variations were effective for the diffuse fluxes of the five studied metals in Ebrié Lagoon. The spatial variations of fluxes were significant ($p<0.05$) due to their individual physicochemical properties and pollutants inputs. Our results showed that sediments acted as a sink as well as a source of all of the studied metals, independently of the season. Thus, metals stored in sediments returned to the overlying waters with physicochemical properties fluctuations. Regarding the dynamics of metals in Ebrié Lagoon, metal enrichment at the surficial sediments have to not be interpreted as evidence of recent natural and/or anthropogenic inputs alone, but may also be a consequence of the deposition of metals previously released from the sediments through diagenesis. The present study have also provided significant data regarding diffusive fluxes of trace metals in the most polluted area of the Ebrié Lagoon around Abidjan City. Trace metals fluxes presented in this paper revealed a high bioavailability of trace metals contents in the surficial sediments of Ebrié Lagoon. At long term, such situation will pose an ecological risk in the studied area in general, and particularly to Milliardaires Bay with the high fluxes of Cd and Ni.Consequently, further investigation with long time assessment is required for spatial and seasonal variations of pollutants fluxes in Ebrié Lagoon for its protection and management.

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