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Research Article

Sediment baseline study of levels, distributions and potential ecological risks of heavy metals in Bahiret El Bibane Lagoon (Tunisia, Southwestern Mediterranean Sea)

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Conflict of Interests:

The authors declare no conflict of interests.

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Abstract

The study aimed to providing a baseline on the state of the sediments contamination of the Bahiret el Bibane Lagoon by heavy metals, which is one of the most productive lagoons and one of the most important active commercial fish traps in the southern Tunisia. The sediment samples collected from 10 sites were subjected to a digestion with aqua regia and analysed for major (Mn, and Fe) and trace (As, Cd, Cr, Cu, Pb, Zn, Ni and Co) elements by inductively coupled plasma-atomic emission spectroscopy (ICP-AES). Results showed that heavy metal concentrations in the sediments are generally low compared to other coastal areas of the Mediterranean Sea. Geochemical approaches (enrichment factor and geo-accumulation index) indicated a widespread pollution by Cu, Pb and Zn and show that these heavy metals originate from anthropogenic sources. According to established sediment quality guidelines (SQGs), the risk for the benthic biota was insignificant.

Key words: surface sediments; metals; geochemical approaches; sediment quality criteria; Bahiret el Bibane lagoon; Mediterranean Sea

Introduction

Coastal lagoons, are among the most heavily stressed ecosystems, mainly due to growing industrial and urban development (1). They provide many important ecological and economic benefits such as flood storage, ecotourism, habitats for plants and animals (2,3). As a consequence of their proximity to the coast (between land and sea), these ecosystems receive continuously, complex mixtures of anthropogenic pollutants, thereby reducing water quality and imposing severe restrictions on pelagic and benthic organisms (4). An example of such areas is the Bahiret El Bibane Lagoon, a major coastal lagoon on the south coast of Tunisia. It is characterized by an economic based mainly on agriculture and fishing. This area is chronically polluted with industrial waste water, pesticides and chemical fertilizers, through soil erosion, water runoff and sea currents (5). This has led to a decrease in fish production over the lastfew decades. To put this into perspective, only 250 tons/year of fish were produced currently compared with 600 tons/year in the 1980s. The Bahiret El Bibane Lagoon is classified as a Ramsar site since 2007 and requires more attention (6).

Among numerous anthropogenic pollutants, heavy metals are of particular concern globally due to their environmental persistence, poor biodegradability, their accumulation in the food chain that have a significant effect on human health in the long term (7-9). Sediments are considered as an optimal medium to evaluate the environmental quality in estuarine and coastal systems, because they are the main sink for several pollutants, including heavy metals (10-14). Heavy metals can enter into aquatic environments through atmospheric deposition, sewage outfalls, storms, waste incineration, urban storm water and agricultural and industrial runoff (15-17). The toxicity, mobility and accumulation of heavy metals in sediments are influenced by complex factors, such as sediment texture, chemical forms, mineralogical composition, reduction/oxidation state, adsorption and desorption processes and hydrodynamic conditions (18,19).

Numerous indices (e.g., enrichment factor (EF), contamination factor (CF), geoaccumulation index (Igeo), toxicity indices and ecological risk indices) have been developed and utilised worldwide to assess extent and hazard of anthropogenic contamination by heavy metals (20-23). Empirical sediment quality guidelines (SQGs), such as the threshold/probable effect level (TEL/PEL) and the effect range low/median (ERL/ERM), are also commonly used worldwide as informal benchmarks to aid in the interpretation of sediment chemistry data (24-27). These SQGs evaluate the degree to which the sediment-associated chemical status might adversely affect aquatic organisms and are designed to assist the interpretation of sediment quality (28).

To our knowledge, studies examining heavy metals pollution in the Bahiret El Bibane Lagoon have never been documented, except of a preliminary study in 1988. Thus, the objectives of this study were (i) to determine concentrations and spatial distributions of heavy metals in surface sediments of the Bahiret El Bibane Lagoon; (ii) to explore the degree of heavy metal contamination in the lagoon using geochemical approaches, and (iii) to assess environmental risks of these metals in the studied area using sediment quality guidelines.

Materials and Methods

Study area

The Bahiret el Bibane lagoon is situated on the southern coast Tunisia (33°15'N; 11°15'E) near the Libyan border (Fig. 1) in an arid climatic setting (30). The lagoon has a surface area of about 230 km^2 and is separated from the sea by a calcareous cordon about 2.5 Km long divided into 9 small islands (31). It is the second largest lagoon in Tunisia with a mean depth of 4 m. The lagoon is characterized by high salinity reaching more than 50‰ during the summer (32,33). The average water temperature is 19 °C with a significant evaporation, in the order of 1,400 mm/year. The average pluviometry is 200 mm/year. Bahiret el Bibane is subjected to easterly winds during the summer and to winds from the west during the winter (30). The prevailing sea breeze is globally oriented toward the direction East-West. The main freshwater courses flowing into the lagoon is the wadi Fessi (10 million m^3/year) (5). The human activities and demographic pressure are low around the Bahiret el Bibane lagoon, its importance as a fishing center in this part of Tunisia is well known (33,30,34).

Sediments sampling

A total of ten surface sediments samples (0-10 cm) located by GPS were collected with a stainless steel grab in April 2011. Details of the sampling sites are shown in Fig. 1 and Table 1. Sites were selected randomly and well distributed all over the lagoon. After collection, samples were packed into polyethylene plastic bags and cool transported to the laboratory, where they were immediately stored in a freezer at -20 °C. All sediment samples were freeze-dried, then sieved using a 2 mm mesh to eliminate debris, homogenized and stored at 4°C until analysis.

Physico-chemical characterisation of sediments

All sampling sites were subjected to physical and chemical analyses in order to determine their natural characteristics (Table1). Water temperature, pH, and

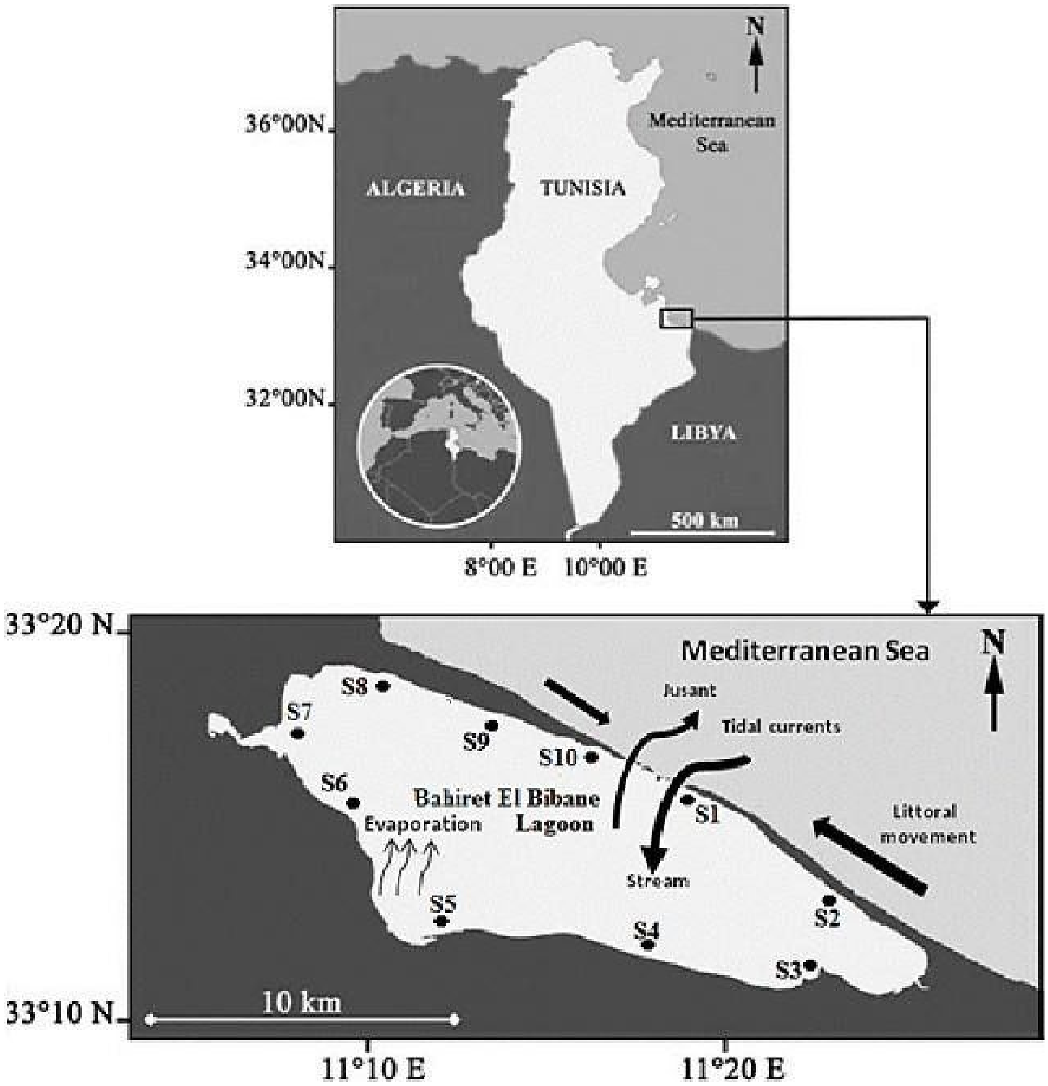

Fig 1: Map of the studied area with location of sampling stations and the direction of water circulation

salinity were determined *in situ* at the water/sediment interface using a handheld multi-parameter system (WTW Multi-197i). Redox potential (Eh) was measured using a 7-cm long microprobe (Hanna Model HI3190T Platinum Redox Electrode). The finer grain size fraction (<63 ìm) of each sediment sample was determined by wet sieving according to Walter et al. (35). Total organic carbon (TOC) was made by coulometry method using a 702 Coulomat, after decarbonisation of sediments with 2N HCl at 60 °C overnight (36).

All chemicals used for the determination of heavy metals were analytical grade (Carlo Erba). The water used was from a MilliQ system (Milford, MA, USA). Certified reference material, IAEA-SL-1 (Lake Sediment), was purchased from the International Atomic Energy Agency (Vienna, Austria).

Potentially hazardous trace elements (arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), zinc (Zn), nickel (Ni) and cobalt (Co)) and major elements (manganese (Mn) and iron (Fe)) contents in the sediment were determined after digestion of the samples with aqua regia according to ISO 11466 (37). Samples (0.5 g dw) were digested with a mixture of HCl (37 %) and HNO_3 (69 %) in a ratio of 3:1 (v/v) at room temperature during 16 h and after, at 130 °C for 2 h under reflux conditions. The obtained solution was filtered through a membrane of 0.45 µm, then diluted to 100 mL with HNO_3 0.5 M, and analyzed using a Jobin-YvonUltima-C Inductively-Coupled Plasma Atomic Emission Spectroscopy (ICP-AES). The reagent blanks were monitored throughout the analysis and were used to accredit the analytical results. Calibration standards were regularly performed to evaluate the accuracy of the analytical method. A certified reference standard of sediments, IAEA-SL-1, was analyzed at the same time to ensure quality control and accuracy of the analysis. Recoveries were above 90% for all the heavy metals measured.

Table 1: Geographical localization of the sampled stations and main physicochemical features of the sediments of Bahiret El Bibane Lagoon.

Station	Location Latitude	Location Longitude	Depth (m)	Temperature (°C)	Salinity (psu)	pH	Redox potential (mV)	TOC (%)	%<63 µm
S1	33°15.986' N	11°18.748' E	5.8	20.3	46.1	8.3	215	0.4	58.1
S2	33°14.430' N	11°21.815' E	1.8	20.5	47.9	8.2	190	0.4	30.7
S3	33°11.156' N	11°23.115' E	0.5	26.9	48.1	8.2	195	5.8	7.4
S4	33°11.376' N	11°19.906' E	0.4	26.7	48.3	8.1	210	1.3	8.1
S5	33°12.494' N	11°12.458' E	1.6	30.5	48.1	8.4	234	2.2	41.1
S6	33°14.712' N	11°09.981' E	4.2	32.2	47.0	8.2	216	0.5	23.9
S7	33°18.853' N	11°07.831' E	0.5	23.5	51.0	8.3	200	1.1	11.3
S8	33°18.578' N	11°11.315' E	6.3	26.0	48.4	8.3	254	0.7	89.3
S9	33°17.538' N	11°14.113' E	7.3	23.3	48.0	8.3	160	0.9	66.0
S10	33°16.623' N	11°17.531' E	6.0	20.7	47.0	8.2	215	2.0	50.6

Table 2: Pearson correlation coefficients between metals and geochemical variables in Bahiret El Bibane Lagoon

	As	Cd	Cr	Cu	Pb	Ni	Zn	Co	Mn	Fe	<63 µm
As	1										
Cd	0.239	1									
Cr	0.079	-0.142	1								
Cu	0.366	**0.713***	0.240	1							
Pb	0.147	0.458	0.187	**0.835****	1						
Ni	-0.097	0.462	0.177	0.561	0.547	1					
Zn	0.148	0.444	0.219	**0.918*****	**0.883*****	**0.776****	1				
Co	**0.934*****	0.245	0.300	0.420	0.174	-0.011	0.218	1			
Mn	0.531	-0.235	-0.254	-0.195	0.088	-0.314	-0.227	0.352	1		
Fe	0.548	0.433	**0.654***	**0.656***	0.369	0.298	0.509	**0.700***	-0.189	1	
<63 µm	0.031	0.603	-0.150	0.074	-0.118	0.062	-0.074	0.122	-0.198	0.206	1
TOC	-0.092	-0.317	-0.406	-0.379	-0.436	-0.303	-0.389	-0.364	0.054	-0.373	-0.409

Numbers in bold indicate significant correlations. Asterisks indicate the signification of the correlations: *$p<0.05$; **$p<0.01$; ***$p<0.001$

Geochemical approach

The geochemical approaches including geo-accumulation index and enrichment factor were employed to identify the geochemical characteristics of the measured heavy metals. The key for the calculation of these geochemical data is the choice of background values (38). In this study, the average shale values obtained by Turekian and Wedepohl (39) were used as reference baseline, since there were no data on background concentrations for the Bahiret el Bibane sediments.

Geo-accumulation index (Igeo)

The assessment of the heavy metal enrichment of the sediment was carried out using the calculation of the geo-accumulation index (*Igeo*). This index (*Igeo*) was originally used with bottom sediment by Muller (40), and is computed with the following equation:

$$Igeo = \frac{\mathrm{Log}_2(C_n)}{1.5(B_n)}$$

Where, C_n is the measured concentration of element n in the sediment and B_n is the geochemical background for the element n. Factor 1.5 is the background matrix correction factor due to lithospheric effects.

Enrichment factor (EF)

Enrichment factor (EF) is an effective method to determine the pollution status and the sources of heavy metals in sediments (41-43). Aluminum (Al) and iron (Fe) are the most commonly used normalizing reference metals because of their abundance in nature and the general lack of anthropogenic source enrichment (44,23). Aluminum concentration was not analyzed in the studied sediments. Therefore, Fe was used to calculate metal enrichment factor. According to Ergin et al. (45), the EF is computed using the relationship below:

$$EF = \frac{\left(\frac{Metal}{Fe}\right) Sample}{\left(\frac{Metal}{Fe}\right) average\ crust}$$

As suggested by Wang et al. (46), EF values of 0.5–1.5 indicates metal input from natural weathering process, whereas EF values >1.5 indicates metal input from anthropogenic sources or a greater percentage from non-natural weathering process. Generally, seven categories are recognized on the basis of the EF values. Samples having an enrichment factor of <1 indicates no enrichment and an EF between 1–3, 3–5, 5–10, 10–25, 25–50, and >50 is considered evidence of minor, moderate, moderate to severe, severe, very severe and extremely severe enrichment, respectively (47).

Sediment quality guidelines

To predict detrimental biological effects of metals on benthic organisms, sediment quality assessment guidelines (SQGs) and mean probable effect level quotients (PELq) were called. Two sets of SQGs developed for freshwater ecosystems (24) were applied in this study: the effect range low (ERL)/effect range median (ERM) and the threshold effect level (TEL)/ probable effect level (PEL) values (48). ERL and TEL (low range values) are concentrations below which adverse effects are not expected to occur. In contrast, the ERM and PEL are concentrations above which adverse effects are expected to occur more often than not. The PELq was calculated according to Chapman and Mann (49) by the following equation:

$$PELq = \sum \frac{\left[\frac{C}{PEL}\right]}{n}$$

where C is the measured metals concentration in sediments, PEL is the guideline value for each metal and n is the total number of metals (n = 7).

Statistical analysis

STATISTICA 6.0 software (version 5) was used for statistical treatment of data. Pearson's correlation coefficient was calculated to investigate possible relationships between parameters.

Results and discussion

General characteristic of sediments

The physic-chemical characteristics of studied sediments were measured to describe the environmental conditions at each sampling site (Table 1). The water depth and temperature ranged from 0.4 to 7.3 m and from 20.3 to 32.2 °C, respectively. High levels of salinity were recorded throughout the lagoon (46.1–51.0 psu), suggesting that the supply of fresh water in the environment is extremely low. The same pattern was observed in the pH data recorded in the water/sediment interface, which were maintained at around 8.2. The redox potential values ranged from 160 to 254 mV, showing that the collected sediments were mainly under aerobic conditions.

For grain size analysis, the fine fraction percentages in surface sediments from the Bahiret El Bibane Lagoon ranged from 7.4 to 89.3 % (Table1). With the exception of stations S1, S8, S9 and S10 that show a predominantly mud (silt and clay) composition, all other stations were composed of particles above 63 µm in size. Sediment grain size varies with the water depth of the lagoon. As shows in Table 1, a high percentage of sand was found at stations (S2, S3, S4, S5, S6 and S7) which have a low water depth (between 0.4 and 4.2 m). However, mud (silt and clay) percentages are dominant in the stations (S1, S8, S9 and S10) where the water depth is high (between 5.8 and 7.3 m). The same finding was observed by Pilkey et al. (30) in the same lagoon. Coarser grained sediment in the shallow water is expected since tidal and wave activity can disturb the fine particles. The abundance of fine particles was assumed to come from anthropogenic inputs of the Jdéria city located in the NNW part of the lagoon.

Total organic carbon concentrations were relatively uniform between stations (0.4–2.2%) except S3 that shows the highest content of TOC (5.8%). A range of sources other than urban and industrial wastewater discharges may provide important organic carbon inputs, including: autochthonous production by macrophytes, benthic or epiphytic micro- or macroalgae, and local water column production by phytoplankton (50). The bottom of the Bahiret el Bibane lagoon is covered by a vast meadow of *Cymodoceanodosa* (Ucria) Ascherson (1869), mixed locally with the *chlorobionta Caulerpaprolifera* (Forsskal) Lamouroux (1809) (51).

No apparent relationship was found between TOC content and grain size among stations ($p > 0.05$), suggesting that the TOC variations at these stations were influenced by factors other than the relative abundance of mud and sand (Table 2). Also, no statistically significant correlations were observed between TOC content, grain size and all analyzed metals, suggesting that the distributions and concentrations of metals were not governed by sedimentary characteristics such as TOC contents and the grain size, but, it might be due to other factors which are not analyzed in this study.

Heavy metals concentrations in sediments

Heavy metal concentrations in sediment samples from Bahiret el Bibane lagoon are shown in Table 3. The total mean concentrations are in the order Fe>Cu>Zn>Mn>Pb>Ni>Cr>As>Co>Cd, but huge differences are observed between individual locations. The highest concentration of Cu, Pb, Zn, Co, and Fe (0.05, 135.20, 24.05, 122.80, 0.60 and 1989.35 mg kg^{-1}, respectively) were found in the station S1. The other elements presented the maximum values at stations S5 (2.05), S2 (2.75), S4 (16.65), and S5 (72.30) for As, Cr, Ni and Mn, respectively. The lowest concentration of all the trace metals except Cr, Ni, Co and Fe was observed at station S9. Cadmium was not detected in the majority of the stations except S1 and S8.

No significant anthropogenic source of heavy metals is present in the study area. However, metals in the lagoon can originate from untreated urban sewages, and from farm activity - mainly poultry ranching and cereal cultivation. Because of the significant exchanges with the sea, the waters of the lagoon are also subject to a risk of contamination by pollutants inputs from industrial sites located outside the study area, on the seaboard or offshore. The high concentrations of trace metal detected at station S1 situated in the sea inlet may indicate that the pollution caused by the marine environment is considerable. The major sea currents or littoral drift have an east-west direction, which makes pollution by metals from the Libyan petrochemical complex located 30-40 km east is important (5) (Fig. 1). Gulf of Gabes, which was reported to be the most exposed to pollution through the huge industrial activities, may be another source of metals pollution. Large quantities of phosphogypsum (calcium sulphate) from the phosphoric acid and chemical product industry of Gabes are released into the Gulf of Gabes (52,53). The volume of water entering the Bahiret el Bibane lagoon from this Gulf is on the order of 0.675 million m^3 per tidal cycle (5). The mixing of lagoon water and sea water causing a change in salinity may be another possibility for the higher metal concentrations at station S2. Although station S9, is located near an urban area (Jdéria city) and a poultry farm, it presents the lower concentration for the majority of the analyzed trace metals, which indicated that the observed distribution of metals was not governed by the localized sources inputs, but, it is due to the hydrodynamic conditions. The waves and currents at station S9 are highly active and can induce sediments dispersion, therefore influencing metals levels. In addition, the dilution effects of seawater should not be ignored. The hydrodynamic conditions are important factors that affect the physical properties of sediments; furthermore, they affect the concentration and spatial distribution of heavy metals (54).

The highest concentrations were found for Fe, Cu, Zn, Mn and Pb. The elements Pb and Zn were generally regarded as the identified elements of traffic pollution (55,56). The commercial ships and small boats which use diesel as fuel are among the main potential sources of heavy metals in the Bahiret el Bibane lagoon, since, this area is characterized by an economy based mainly on agriculture and fishing. In addition, antifouling paints used in boat maintenance in the south sector (El MARSA) of the lagoon could be another source of Cu and Zn in surface sediments. Turner (57) has demonstrated that antifouling paint

Table 3: Trace metal contents (mg kg^{-1}) in the sediments from Bahiret El Bibane Lagoon.

Sites	As	Cd	Cr	Cu	Pb	Ni	Zn	Co	Mn	Fe	PELq[a]
S1	1.95	0.05	2.50	135.20	24.05	12.10	122.80	0.60	18.50	1989.35	0.32
S2	1.70	<lod	2.75	38.95	2.65	6.40	28.95	0.55	21.40	1233.25	0.10
S3	1.10	<lod	1.90	25.05	2.80	3.65	18.30	0.25	21.00	614.90	0.06
S4	0.85	<lod	2.10	55.55	15.50	16.65	97.55	0.30	14.70	754.80	0.20
S5	2.05	<lod	1.65	11.10	4.70	2.35	4.70	0.55	72.30	729.85	0.04
S6	0.95	<lod	2.50	45.25	20.20	7.65	53.05	0.30	46.30	728.35	0.14
S7	1.30	<lod	1.90	62.30	14.90	0.50	53.55	0.40	28.75	607.45	0.14
S8	1.00	0.05	1.55	57.00	10.15	10.25	53.25	0.30	16.75	507.85	0.16
S9	0.75	<lod	2.30	3.20	0.75	1.80	1.60	0.30	9.85	973.80	0.02
S10	0.95	<lod	2.15	18.70	2.35	5.75	12.35	0.35	12.55	750.00	0.06
Min	0.75	<lod	1.65	3.20	0.75	0.50	1.60	0.25	9.85	507.85	0.02
Max	2.05	0.05	2.75	135.20	24.05	16.65	122.80	0.60	28.75	1989.35	0.32
Mean ± SD	1.26 ± 0.47	0.01 ± 0.02	2.13 ± 0.39	45.23 ± 37.58	9.81 ± 8.39	6.71 ± 5.09	44.61 ± 40.23	0.39 ± 0.13	26.21 ± 19.21	888.96 ± 437.75	

[a] Mean PEL quotients calculated according to Chapman and Mann (49), see Materials and method section for explanation.

<lod: below the limit of detection.

Table 4: Comparison between heavy metal concentrations (mg Kg^{-1} dw) obtained in this study (minimum–maximum) with those obtained by other authors in Mediterranean Sea.

Location	As	Cd	Cr	Cu	Pb	Ni	Zn	Co	Mn	Fe	Reference
Bahiret el Bibane lagoon (Tunisia)	0.75–2.05	<lod–0.05	1.65–2.75	3.20–125.20	0.75–24.05	0.50–16.65	1.60–122.80	0.25–0.60	9.85–28.75	507.85–1989.35	Present study
Puerto Rico southwest	1.73	0.01	–	5.21	1.93	–	7.99	–	–	0.52%	(61)
Pearl River Estuary (China)	–	–	–	54.6	36.1	33.7	139.0	–	–	–	(62)
Bizerte lagoon (Tunisia)	–	0.4	33.7	21.2	52.7	8.3	465.6	–	145.1	15,519.5	(63)
Sfax–Kerkennah plateau (Tunisia)	–	36.0	196.9	38.4	125.2	348.0	2,077.3	–	116.9	8,835.6	(64)
Jamaica Montego Bay	1.4–7.03	nd–10.0	–	3.5–73.8	6.4–185	–	7.9–147	–	–	–	(65)
Tetouan coast (North of Morocco)	–	0.14–0.27	88.40–160.97	4.09–29.12	33.11–47.97	34.19–79.89	64.82–110.77	18.06–31.70	256.56–651.66	–	(66)
Mediterranean sea (Turky)	–	0.06–3.94	65–264	20–703	91.3–751	28.240	86–970		283–1192	–	(67)
Bni Bousra massif (Morocco)	–	–	2738–4048	–	–	1906–2569	–	78–107	–	–	(68)
Ivra complexe (Italy)	–	–	2568–2984	–	0.25	2040–2438	–	100–117	968–1053	–	(69)
Thermaikos Gulf (Greece)	–	–	–	19–165	10–218	–	74–358	–	–	29,200–45,360	(70)

<lod: below the limit of detection, nd: not detected.

Table 5: Geo-accumulation index Muller's classification and ranges of heavy metals in surface sediments of the Bahiret El Bibane Lagoon.

Muller scale (40)			This study			
I_{geo} Value	Class	Quality of sediment	Heavy metal	*Igeo* range	Mean *Igeo*	Quality of studied sediment
≤ 0	0	Unpolluted	As	−4.5 to −3.2	−4.0	Unpolluted
0–1	1	From unpolluted to moderately polluted	Cd	−3.2 to 0.0	−3.2	Unpolluted
1–2	2	Moderately polluted	Cr	−6.4 to −5.6	−6.0	Unpolluted
2–3	3	From moderately to strongly polluted	Cu	−4.4 to 1.0	−1.1	From unpolluted to moderately polluted
3–4	4	Strongly polluted	Pb	−5.3 to −0.3	−2.3	Unpolluted
4–5	5	From strongly to extremely polluted	Ni	−7.7 to −2.6	−4.5	Unpolluted
> 5	6	Extremely polluted	Zn	−6.5 to −0.2	−2.5	Unpolluted
			Co	−6.8 to −5.6	−6.3	Unpolluted
			Mn	−6.7 to −4.1	−5.9	Unpolluted
			Fe	−6.9 to −5.2	−6.4	Unpolluted

can be a source of Cu, Cd, Pb and Zn in sediments and emphasizes that release of those metals is generated during boat maintenance and cleaning. Tire particles, motor oil, and hydraulic oil are also major sources of Zn (58). The EL Ketf harbor situated in the east sector of the lagoon (Ben Gardane city), in the axis of littoral currents may contribute to the lagoon pollution by Cr, Ni and Co. This harbor contains about 100 to 150 boats, which poses solid and liquid wastes (5). Similar result was observed by Sprovieri et al. (59), which showed that Naples harbor (southern Italy) is the primary source for Cr, Ni, V and Co in surface sediments. Mn and Fe are a natural component of sediments. Their high concentrations can be related to continental inputs of matter from urban drainages. It is also important to note that atmospheric deposition and some anthropogenic diffuse sources located around the lagoon like highway traffic, waste incineration, discharges of abattoirs, sewage disposal and domestic solid residues may be contributing to pollution of the lagoon by heavy metals.

Correlations between metals indicate whether there is any common source of pollution. Table 2 showed the matrices of Pearson correlation coefficient values between analyzed pollutants. According to results of analyses for samples, high positive correlations were found for Pb–Cu, Zn–Cu, Zn–Pb, Zn–Ni, and Co–As elements. Moderately positive correlations were obtained for Cu–Cd, Fe–Cr, Fe–Cu, and Fe–Co elements. According to Suresh et al. (60), if the correlation coefficient between the metals is high, metals have common sources, mutual dependence and identical behavior during their transport. A high positive correlation was found between Zn and Ni elements, indicating a possible common source that could be the harbor activities. The same assumption can be applied to the high positive correlations for Pb–Cu, Zn–Cu, Zn–Pb elements, which indicating a possible common source that could be the traffic pollution or antifouling paints. Throughout the sites, there were no significant correlations for Mn either with other trace metals, or with TOC and grain size, which is likely due to its origins and dispersion processes.

To gain a comprehensive understanding of the degree of contamination of the Bahiret el Bibane lagoon by heavy metal, a comparison with other Mediterranean coastal areas was performed (Table 4). Generally this method is viewed as quick and practical for tracking heavy metal enrichment. However, the environmental differences may lead to a misinterpretation of the results. Metal levels in our study were similar to concentrations found in Puerto Rico southwest, and Pearl River Estuary (China). However, these levels were lower than those found in Bizerte lagoon (Tunisia), Sfax–Kerkennah plateau (Tunisia), Jamaica Montego Bay, Tetouan coast (North of Morocco), Mediterranean sea (Turky), Bni Bousra massif (Morocco),

Table 6: Metal enrichment factors (EFs) for sediments of all studied sites in the Bahiret El Bibane Lagoon.

Sites	Metal EFs								
	As	Cd	Cr	Cu	Pb	Ni	Zn	Co	Mn
S1	3.6	4.0	0.7	71.3	28.5	4.2	30.7	0.7	0.5
S2	5.0	0.0	1.2	33.1	5.1	3.6	11.7	1.1	1.0
S3	6.5	0.0	1.6	42.7	10.7	4.1	14.8	1.0	1.9
S4	4.1	0.0	1.5	77.2	48.5	15.3	64.2	1.0	1.1
S5	10.2	0.0	1.2	16.0	15.2	2.2	3.2	1.9	5.5
S6	4.7	0.0	1.8	65.2	65.5	7.3	36.2	1.0	3.5
S7	7.8	0.0	1.6	107.6	57.9	0.6	43.8	1.6	2.6
S8	7.1	15.5	1.6	117.7	47.2	14.0	52.1	1.5	1.8
S9	2.8	0.0	1.2	3.4	1.8	1.3	0.8	0.8	0.6
S10	4.6	0.0	1.5	26.2	7.4	5.3	8.2	1.2	0.9
Mean	5.6	1.9	1.4	56.0	28.8	5.8	26.6	1.2	1.9
Min	2.8	0.0	0.7	3.4	1.8	0.6	0.8	0.7	0.5
Max	10.2	15.5	1.8	117.7	65.5	15.3	64.2	1.9	5.5

Ivra complexe (Italy), and Thermaikos Gulf (Greece). These results show that contamination of metals in sediments of Bahiret el Bibane lagoon were low relative to other metal-polluted aquatic systems.

Geo-accumulation Index (Igeo) and Enrichment Factor (EF)

The results of computed geo-accumulation index (*Igeo*) values are regrouped in Table 5. This index consists of seven classes in relation to pollution extent (Table 5). According to *Igeo* values, sediments of Bahiret el Bibane lagoon can be considered as unpolluted with metals, except sediments from station S1 which are moderately polluted with Cu. The mean *Igeo*, decrease in the following order: Cu>Pb>Zn>Cd>As>Ni>Mn>Cr>Co>Fe. Copper was the main environmental pollution factor.

Pollution status and the sources of heavy metals in surface sediment of the Bahiret el Bibane lagoon was also assessed using enrichment factor (EF) (Table 6). The mean EF values for all metals studied except Cr and Co were >1.5, suggesting anthropogenic discharges of metal in the lagoon. The highest EF values were found at station S8. This result is not surprising, since this station is located near the Jdéria city, which is characterized by various anthropogenic activities like waste incineration, sewage disposal and domestic solid residues. The EF values for Cu and Zn in the sediments of station S8 were 117.7 and 52.1, respectively, indicating "extremely severe enrichment". The EF value for Pb was between 25 and 50, indicating "very severe enrichment". The EF values for Cd and Ni were 15.5 and 14.0, respectively, indicating "severe enrichment". However, minor enrichment (1<EF<3) was found for Cr, Co and Mn. Copper at all stations except S6 had the highest EF values among the eight metals studied, followed by Pb and Zn. The EF values for other studied metals indicated "minor to very severe enrichment" of sediments. The mean EF, decrease in the following order: Cu>Pb>Zn>Ni>As>Cd=Mn>Cr>Co.

Considering these geochemical approaches, it can be deduce that surface sediments of Bahiret El Bibane Lagoon are polluted with Cu, Pb and Zn and therefore require urgent action from appropriate quarters. The sources of these heavy metals are probably located outside the lagoon and are transported by water currents and atmospheric deposition, since there is no significant source of heavy metals around this lagoon.

Sediment toxicological aspects

The method of Sediment Quality Guidelines (SQGs) was applied to assess the ecotoxicological sense of metals concentrations in sediments. Three ranges in chemical concentrations were designed, where adverse effects are observed rarely (<ERL or <TEL), occasionally (ERL–ERM or TEL–PEL) and frequently (>ERM or >PEL). The distribution of metals in these ranges was presented in Table 7. Except Cu, all metals are below ERL values in all stations, indicating that the toxicity affected by these metals would be rare. TEL/PEL analysis showed that Cu concentration was higher than PEL value only in station 1, and between TEL–PEL values in 70 % of stations (S2, S3, S4, S6, S7, S8, S10), which indicated a potential harm for benthic organisms by this metal.

Table 7: SQGs values for metals (mg Kg^{-1} dw) and relative percentage of stations including in the range of Sediment Quality Guidelines.

Metals	SQG ERL–ERM	% of stations			SQG TEL–PEL	% of stations		
		<ERL	ERL–ERM	>ERM		<TEL	TEL–PEL	>PEL
As	8.2–70	100	0	0	7.24–41.6	100	0	0
Cd	1.2–9.6	100	0	0	0.68–4.21	100	0	0
Cr	81–370	100	0	0	52.3–160	100	0	0
Cu	34–270	40	60	0	18.7–108.2	20	70	10
Pb	46.7–218	100	0	0	30.2–112.2	100	0	0
Ni	20.9–51.6	100	0	0	15.9–42.8	90	10	0
Zn	150–410	100	0	0	124–271	100	0	0

Furthermore, the potential acute toxicity of contaminants in sediment samples can be estimated using

PELq. They can be used to portray the sediment as non-toxic (PELq<0.1), slightly toxic (0.1<PELq<1.5), medium toxic (1.5<PELq<2.3) and highly toxic (PELq>2.3) (71). The mean PEL quotients calculated for the studied sites (based on metals As, Cd, Cr, Cu,Pb, Ni and Zn) were in the range of 0.02–0.32 (Table 3). In this context, only two stations (S1 and S2) have PELq<0.1 and are categorized as non-toxic, whereas the other stations have PELq between 0.1 and 1.5, and are categorized as slightly toxic. Similar results were found by Nasr et al. (72) in sediments of the Southeastern Mediterranean sea, Egypt. Although, this approach (PELq) are frequently used to predict sediments toxicity (73), it presents some disadvantage such as limitation only to chemicals include in the SQG list, but not all the contaminants present in sediments (74).

Conclusion

The concentrations of heavy metals have been determined in surface sediment from 10 stations in the Bahiret El Bibane Lagoon, Tunisia. The results show that spatial distribution of metals concentrations is not uniform and it might be controlled by the hydrodynamic conditions. No significant correlation was found between metal concentrations and the physicochemical parameters of sediments (total organic carbon and grain size) suggesting that these parameters did not play an important role in controlling the metals distributions in the sediments. Iron, Cu, Zn, Mn and Pb are the most abundant elements with higher concentrations in the lagoon. Both enrichment factor (EF) and geoa-ccumulation index (*Igeo*) values suggested the elevation of Cu concentration in the region. When compared with SQGs, all the metals tested would lie below the TEL and PEL values except for Cu, indicating that the toxicity affected by these metals would be rare. Based on the mean PEL quotients (PELq), sediment of the Bahiret el Bibane lagoon can be categorized as slightly toxic. To the best of our knowledge, this is the first study to report heavy metals contamination in surface sediments of the Bahiret El Bibane Lagoon. Thereby, present results can serve as baseline information for monitoring future concentrations of metals in Tunisian coastal waters. Because we only investigated heavy metal levels in spring, more studies during the whole year with increasing the samples size is needed to fully understand levels of heavy metals and ecological risk assessment of this area.

Author contributions

Dr. Badreddine Barhoumi designed the study, performed the analysis and wrote the manuscript. Dr. Soufiane Jouili collected the samples. Dr. Abdelkader Derouiche and Ghzela Mahfoudhi involved in the design and organization of the study. Dr. Anis Elbarhoumi and Atef Atyaoui performed metals analysis. Dr. Sondes Bouabdallah, Soufiane Touil and Mohamed Ridha Driss revised the manuscript. All authors approved the final manuscript.

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