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Trace metal contamination by phosphogypsum discharge in surface and core sediments of the Gabes coast area (SE of Tunisia)

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Abstract

The purpose of the present study is to ascertain the extent of the effect that phosphate fertilizer industrial waste has on the surface and bottom sediments of the Ghannouch-Gabes coast, off the Tunisian Mediterranean Sea. To achieve this, 44 surface sediments and 3 core sediments were studied for mineralogy, trace metals (Cd, Cu, Pb, and Zn), F, CaO, and SO_3. For all the analyzed elements, the spatial distribution in surface sediments showed that the area located between the commercial and the fishing port of Gabes is the most polluted zone. The ranking of metal contents was found to be Zn > Cd > Cu > Pb. The vertical distribution of trace metals indicated that the highest levels were found in the uppermost segment of the sediment cores compared to the lower depth subsurface due to a continuous input of phosphogypsum (PG) release and confirmed that the area between the two harbors suffered from several types of pollutants compared to reference core C1, collected from other non-industrialized areas. This spatial and vertical distribution is probably due to the harbor piers which acted as barriers and limited the dispersion of PG discharge. The contamination factor, the geoaccumulation index, and the pollution load index were determined. The results obtained confirm the anthropogenic impact on the levels of metal, on the fluorine, calcium, and sulfate concentrations in the area, located between the commercial harbor of Ghannouch and the fishing harbor of Gabes, whereas the concentrations of elements analyzed tends to decrease on both sides of this sector. Statistical analyses (principal component analysis) showed trace metals, fluoride, sulfate, and a large amount of calcium resulting from the same anthropogenic source.

Keywords Metals · Surface sediment · Core sediment · Geoaccumulation index · Contamination factor · Statistical analyses · Phosphogypsum

Introduction

In recent decades, urban and industrial activities located in coastal areas have often been considered a factor in the pollution of marine environments (Varol 2011; Gonzalez-Fernandez et al. 2011; Jiang et al. 2014; Gao and Chen 2012; Armstrong-Altrin et al. 2015a). Trace metals in marine sediments may have a natural (physical and chemical weathering of parent rocks) and anthropogenic sources such as wastewater discharges, urban, and industrial effluents. The distribution and the accumulation of heavy metals is influenced by mineralogical composition, sediment texture, reduction/oxidation state, hydrolysis and co-precipitation of metal ions, and adsorption and desorption process (Buccolieri et al. 2006; Armstrong-Altrin et al. 2015b; Armstrong-Altrin and Machain-Castillo 2016). A large part of the free metal ions are deposited in the sediments. Marine sediments have the ability to retain metals or release them to the aquatic column (Buccolieri et al. 2006; Bastami et al. 2014). The growth of industrialization and urbanization along coastal regions and littoral zones are increasingly being exposed to heavy metals pollutants. For these reasons, the sediments are commonly chosen as environmental indicators of the quality and potential risk with biota (Jiang et al. 2014; Ramos-Vasquez et al. 2017).

Sediment cores contain information about the events that occurred in preindustrial times in the marine area and the metal concentrations evolved with depth in sediment cores, which can inform us about the long-term trends in the evolution of

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metals which imply possible pollution. The background concentrations of the cores can be considered as a reference for evaluating pollution by the trace metals (Atalar et al. 2013).

The Gulf of Gabes located at the southeast of Tunisia in the eastern Mediterranean Sea is characterized by high marine productivity, and it is the most important fishing zone in Tunisia (Ben Amor and Gueddari 2016). However, The Gulf of Gabes has experienced, since the past few decades (1972), major environmental problems due to the industrial and economic growth in this zone, especially after the installation of the biggest industrial complex of Tunisia, in the Gabes coast area, between the fishing port of Gabes and the commercial port situated in Ghannouch City (at 5 km from fishing harbor). These industries treat phosphate ore to obtain phosphoric acid and discharge at sea, among others, phosphogypsum (PG) which is a waste by-product of the phosphate fertilizer industry, which is usually released into the environment because of its restricted use in industrial applications (Papanicolaou et al. 2009).

The aim of this study is (1) to ascertain the extent of the effect of the phosphate fertilizer industrial waste on the surface and bottom sediments of the Ghannouch-Gabes coast (GG); (2) to describe the spatio-temporal distribution of trace metals in the surface and core sediments; (3) to determine the degree of trace metal pollution using different contamination indices; and (4) to determine relationship among trace metals, fluoride, calcium, and sulfate and specify their origin using multivariate statistical techniques.

Site description

The GG coast, located in the central part of the Gulf of Gabes (lat. 33° 56′–33° 52′ N and long. 10° 04′–10° 07′ E) (Fig. 1), is limited by the Ettine River in the north and the El Fard River in the south and 12.5 m isobaths contours. The hydrographic network is dense but with a low flow. The river originates in the Medenine District and drains 500 km^2 of catchment areas before reaching the Gulf of Gabes. The climate in the region is Mediterranean with a mild winter and an average temperature of 20 °C (INM 2015). The Gabes coast area is characterized by irregular rainfall with stormy character. The lithology of the study area consists of Mio-pliocene (gypsum clay), Pleistocene (limestone crust), and Holocene (alluvium) (Ben Ouezdou 1987; Jedoui 2000). In addition, the studied area is distinguished by a low sandy coast. The submarine morphology is monotonous showing slight slope due to the continental shelf extension. Indeed, the isobath − 10 m is 6.5 km from the coastline (Ben Amor 2001). The tide is semidiurnal and is the highest in the Mediterranean Sea. The tidal amplitude can reach 1.7 m (Sammari et al. 2006). The principal swell is in the eastern sector where it induces a principal littoral drift of the N-S sector. Swells of W to SW sectors, which are less important, induce a secondary littoral

drift directed to the S-N sector (Oueslati 2004). The GG coast receives, since the past few decades, a continuous untreated industrial waste from the Tunisian chemical complex, situated in front of the sea, between the fish and the commercial harbor. These industrial wastes are mainly represented by phosphogypsum (PG), which is an acidic by-product of the phosphate fertilizer industry. Previous studies indicate that the PG is composed mainly of gypsum (Rutherford et al. 1994) and has a similar structure to that of natural gypsum. It is in the form of a very fine wet powder, with a water content ranging between 20 and 30% (Rutherford 1994). The size of the PG particles is generally between 0.01 and 0.5 mm, with a mean diameter (D_{50}) of the order of 0.05 mm.

In addition, PG contains impurities, like high amounts of fluoride, and considerable levels of heavy metals such as lead (Pb), zinc (Zn), chromium (Cr), copper (Cu), and cadmium (Cd) (Arocena et al. 1995; Ghafoori and Chang Wen 1986; Nordstron and Jenne 1977; Ben Amor and Gueddari 2016; Gouidera et al. 2009; Garrett et al. 2011; Zaghden et al. 2014). Since 1972, almost 12,000 tons of PG are daily released into the open sea without any treatment (GCT 2003).

Through the years, many authors (Bradai et al. 1995; Bejaoui et al. 2004; Aloulou et al. 2011) reported a degradation of the aquatic environment shown by the decrease in animal and plant production, which is mainly due to chemical industry releases. Therefore, the study will attempt to evaluate the extent of heavy metal contamination from the surface to the bottom sediments and the degree to which trace metals are influenced by other sedimentological parameters such as grain size, sediment composition, and hydrodynamic parameters.

Materials and methods

Sampling

Knowing that the top few centimeters of the sediments reflect the continuously changing present-day degree of contamination, whereas the bottom sediments record its history, a 44-surface sediment samples and 3-sediment cores were collected along the coastal area of GG (southeast of Tunisia).

Surface samples were collected through 13 profiles parallel to the beach (Fig. 1) in which the depth varied between − 0.5 and − 12.5 m. The surface sampling stations were divided into two site groups: (1) group I (G1–G21), located in the south of fishing port and the El Fard River, and (2) group II (G22–G44), located between the two harbors (commercial and fishing ports) and the north of the study area. Samples from each station were collected using a Peterson grab. The top 5 cm of the surface sediment was taken with an acid washed spatula to prevent contamination. These sediments were immediately placed in polyethylene bags, refrigerated at 4 °C until analyses
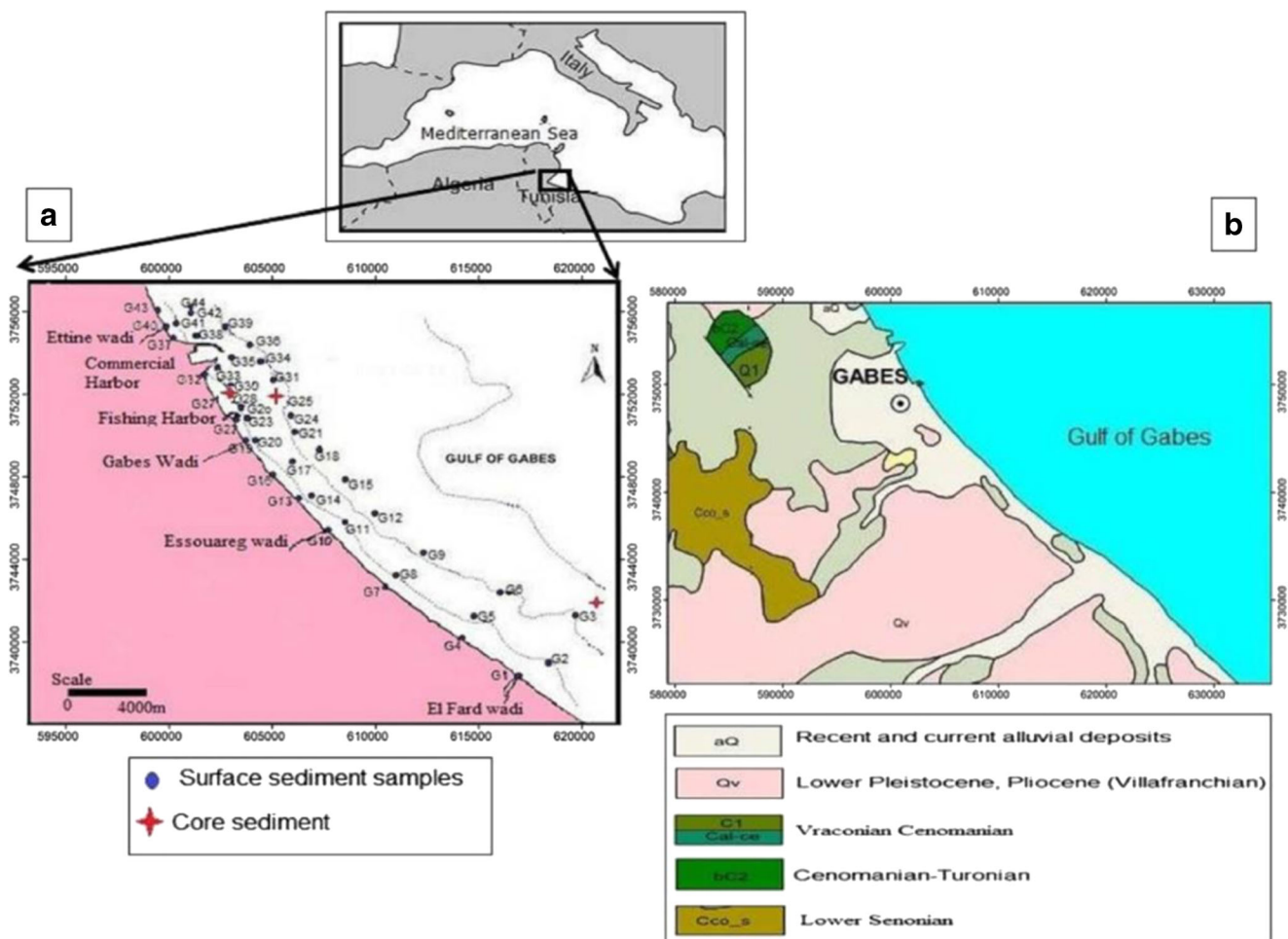
Fig. 1 **a** Location of sediments samples (surface and core) along the Ghannouch-Gabes coast. **b** Simplified geological map (source: Geological map of Tunisia 1/500000)

in a laboratory. Three sediments cores were collected from the GG coast by scuba divers using PVC tubes 120 cm long and 6.5 cm in diameter. Core C1 was sampled at the south of the study area at − 12.5 m depth; its length is 51.5 cm. This core will be considered as a reference because of its distance from the PG discharge area (Fig. 1). C2 was sampled in the zone located between the commercial and fishing harbor at − 3.5 m depth (length of C2 52 cm). C3 was sampled between the fishing harbor and Gabes wadi at − 7.5 m depth (length 57 cm). In the laboratory, sediment cores were subsampled at 5 cm intervals for the top 20 cm, then to 10 cm intervals for the rest of core. Grain size analysis of the surface samples and core subsamples was done using a set of eight sieves ranging from 1000 mm to 63 μm for 20 mn using Rotop Sieve Shaker (Folk and Ward 1957).

Geochemical analysis

For the determination of the trace metals, the samples (44 surface samples and 21 subsamples) were digested by adding a mixed solution of concentrated of 15 mL of $HClO_2$, 15 mL

HF, and 20 mL HNO_3 to 1 g sediment in Teflon bombs. The resulting solutions were analyzed for Pb, Zn, Cu, and Cd by atomic absorption spectrometry (Thermo Scientific, ICE 3000 series). The procedures used for trace metals analysis were checked for accuracy using the BCR-032 certified reference material (Table 1), obtaining good concurrence (< 12%); the certified values and the relative standard deviations obtained from five replicates of one sample were typically (< 2.49%).

For the fluoride determination, only surface sediments were analyzed: 1 g of sediment sample was dissolved in 3 mL of 6 M HCl and diluted to 100 mL with distilled water, and an

Table 1 Accuracy of trace metal analysis (%)

Element	BCR-032	This study	Error (%)	RSD (%)	Limit of detection
Zn	0.253	0.281	11.07	2.49	0.008
Pb	–	–	–	–	0.05
Cu	33.7	34.1	1.19	1.97	0.05
Cd	0.02	0.024	20	0.22	0.005

aliquot of the extract was used for the determination of fluoride by the specific ion electrode method (Abu Hilal 1985).

In order to analyze major element contents (CaO%, SO$_3$%), 100 mg of sample sediments were dissolved in a strong acid mixture (5.0 mL of Milli-Q water for hydrolysis, 1.5 mL of 40% HF, and 3% of HNO$_3$-HCl l:3 v/v to digest silicates and for complete oxidation) (Cha et al. 2006). For fluorine, CaO, and SO$_3$, each sample was passed three times and was expressed as an average of the three values.

Mineralogical analysis

The sediment's mineral composition of 44 total surface sediment samples and 21 core sample were identified using X-ray diffraction according to "the powder method" which consists of irradiating the finely ground sample in the angular space between 2° and 52° using copper Kα radiation. The different minerals were identified with the X'pert HighScorePlus Software consisting on the d-spacing of height main picks such as quartz (3.34 A°), calcite (3.03 A°), gypsum (7.60 A°), and fluorine (3.15 A°) of each mineral with those of reference mineral standards.

Sediment quality index

In an aquatic ecosystem, contamination of a sediment by trace elements is a relative concept. It must be defined in relation to the natural contents in the sediments. The anthropogenic impact of the considered trace elements in marine sediments can be evaluated from the metal enrichment relative to unpolluted background levels. Since the use of background values is important for interpretation of geochemical data (Wang et al. 2015), the area of Teboulbou (the bottom of C1, samples C1–7 at 52 cm from surface, at 12.5 m depth) was chosen as a reference for the background of GG sediments. This site was first chosen for its remoteness from the discard zone (the farthest and deepest point), then because the concentration of all metals in this station are lower than those noted in the other sampling station. The pollution level was evaluated at each of the samples taken.

To assess a toxic metal pollution, geoaccumulation index (Igeo) (Muller 1980), contamination factor (C_f) (Hakanson 1980), and pollution load index (PLI) (Tomlinson et al. 1980) were calculated. Details of quality sediment index classes are given in Table 2.

Statistical analysis

The obtained data of chemical analysis of the GG surface sediments was performed using principal component (PCA) techniques (XLSAT 2013 for Windows) in order to establish the relationship among the investigated variables in the samples and to identify the most common pollution sources.

Results

Grain size distribution

The results of grain size distribution are shown in Table 3 for surface sediments and in Table 4 for core sediments. The sediment composition reveal that the surface sediments are mainly sandy in the south of the study area with content varied from 54.4 to 85.5% and mean grain size (Mz expressed in phi units) varied from 2.09 and 3.55. In the inter-harbor zone, the grain size distribution was dominating by clay and silt with proportion oscillated respectively from 32.9 to 65.85% and from 21.4 to 41.3%. The same trend was observed for core subsamples. In fact, sand was dominating in core C1 (reference core located at the south of study area) with sand content varied from 87.1% in surface and tend to decrease with depth to reach 37.5%. The clay was present only at about 6.8–12.4%. In core C2 and C3, there was a variation in trend such that silt and clay was present at higher percent ranging from 36.1 and 86.4 for clay and silt content dropped from 18.3 to 41.8%.

Mineralogical analysis

Mineralogical analyses by X-ray diffraction performed on all surface sediment samples revealed the presence of quartz, calcite, gypsum, and fluorine. The most abundant mineral was quartz, followed by calcite with proportions varying from 10.22 to 100% and from 3.1 to 47.59%, respectively. Gypsum and fluorine were present only in few samples which are located in the area between the fishing and commercial harbors. As for surface sediments, mineralogical analysis shows that the main non-clay minerals are for the core C1 quartz (16.57–79.59%) and calcite. At the core C2 sampled in front PG release, only gypsum was identified on full length. The XRD shows for C3 that quartz is the most abundant mineral with proportions that reached 55% in the surface and tend to decrease with depth. Calcite has a homogenous evolution; gypsum was identified on full length and its proportions are very heterogeneous, 18% in the surface and 50% in the bottom layer.

Geochemical analysis

The proportion of calcium and sulfur are shown in Tables 6 and 7. The CaO (%) contents vary from 3.3 to 45.1% in the surface sediments and from 28 to 54.54% in bottom sediment layers. The total sulfur content SO$_3$ (%) vary between 0.1 and 49.43% for surface sediments (Fig. 3) and from 0.12 to 54.77% for core sediments (Fig. 4). Fluoride showed a high accumulation in superficial sediments

Table 2 Geoaccumulation index (Igeo), contamination factor (C_f), and pollution load index (PLI) for contamination levels in sediments

Sediment quality index	Classes
Geoaccumulation index	Igeo ≤ 0 uncontaminated
Igeo = log$_2$ (Cn/1.5 Bn)	0 < Igeo ≤ 1 uncontaminated to moderately contaminated
Muller (1980)	1 < Igeo ≤ 2 moderately contaminated
	2 < Igeo ≤ 3 moderately to strongly contaminated
	3 < Igeo ≤ 4 strongly contaminated
	4 < Igeo ≤ 5 strongly to extremely contaminated
	Igeo > 5 extremely contaminated
Contamination factor	C_f < 1 low factor
$C_f = C_{heavy\ metal}/C_{background}$	1 ≤ C_f < 3 moderate factor
Hakanson (1980)	3 ≤ C_f < 6 considerable factor
	C_f ≥ 6 very high factor
Pollution load index	PLI ≤ 1 no pollution
PLI = $(C_{f1} * C_{f2} * C_{f3}....)^{1/n}$	PLI > 1 pollution
Tomlinson et al. (1980)	

between the commercial and the fishing harbor whose contents varied between 0.11 and 12.19%.

Trace metal composition of surface sediments from the GG coast is shown in Table 6. The concentrations of metals in the surface sediments showed significant differences among the sampled locations. Ranges of heavy metals in sediments are 3–59 μg/g for Cd, 2–13 μg/g for Cu, 0.1–3.8 μg/g for Pb, and 18–57 μg/g for Zn (Table 3). The mean concentration in sediment collected in the zone II (located between the commercial harbor and the fishing port) exceed the upper continental crust (UCC) for Cd and Zn (Table 5). The mean concentration in sediments of all metals (except for Cd) were below UCC average (Martin and Whitfield 1983) in zone I.

For core subsamples, the trace metals composition is shown in Table 7. The concentrations of lead (Pb) vary from 0.5 to 0.7 μg/g along the three cores (Table 6). In all cores, the lead contents are homogenous; no notable variation was observed with depth. The cadmium contents vary from 1 to 63 μg/g in core sediments (Table 7). The vertical distribution of these elements is heterogeneous depending on the sample location. For C1 (reference core), vertical distribution is homogenous, remained fairly stable throughout the core. For C2 and C3, the maximum accumulation of Cd was found in the upper layer at 15 cm. In C3, Cd decrease from 63 to 21 μg/g in the first 15 cm. Both C2 and C3 showed high levels of Cd (collected between the commercial and fishing harbors) compared to other sites in the Mediterranean Sea (Table 5), and they exceeded largely crust average vertical line. Copper (Cu) varied from 2.2 and 12.7 μg/g; for C1, a peak was recorded at 10 cm and tended to slightly increase with depth. For C3 core, the highest values characterize the surface sediments. Concentrations profile is characterized by slight decrease

in low sediments. For C4 core, the highest levels are observed between 10 and 20 cm depth, whereas in the lower sediment, the levels are almost constant. The contents of Zn vary from 3.9 and 52.1 μg/g. The vertical distribution shows no specific trend emerges. Levels of Zn contents remain relatively low. The reference core C1 is homogeneous. The concentrations tend to decrease with depth in C4 with the highest value at the first 5 cm. For C3, the lowest values are observed between 30 and 40 cm depth and increase in the bottom sediments (Fig. 7).

Assessment of trace metals

The different assessment indexes for surface and core sediments are summarized in Tables 6 and 7, respectively. The C_f of lead varied from 1 to 38 for surface sediments and from 4 to 7 for core sediments. The C_f of Zn varied from 0.5 to 7.2 for surface sediments and for 0.5 to 6.6 for bottom sediments. The C_f values of copper are between 0.3 and 1.8 for surface sediments and between 0.3 and 1.6 for core sediments. The calculated values of contamination factor of cadmium are the most elevated and varied between 3 and 59 and between 1.1 and 63, respectively, for surface and bottom sediments.

The geoaccumulation index varied between −0.6 and 4.7 for surface sediments and between 1.4 and 2 for Pb; similarly, the Igeo for Zn varied from 0.6 to 2.3 for surface sediments and from −1.6 to 2.1 for core sediments. Igeo values of Cu ranged from −2.6 to 0.2 and from −2.4 to 0.1 for surface and core sediments, respectively. Finally, Igeo for Cd are the most elevated values varied between 0.4 and 5.3 for surface sediments and between −0.4 and 5.4 for core sediments.

Table 3 Grain size and textural parameters for surface sediments of the Ghannouch-Gabes coast

Stations	Latitude (° N)	Longitude (° E)	Depth (m)	Sand (%)	Silt (%)	Clay (%)	Mz (Φ)
G1	616410.83	3737477.99	0.5	91.7	8	0.3	2.58
G2	617803.22	3738155.37	5	60.6	39.2	0.2	3.59
G3	619045.07	3740526.18	12	84	13.4	2.6	3.59
G4	613776.59	3739359.59	0.5	90.3	9	0.7	2.88
G5	614341.07	3740488.55	6	54.6	35.3	10.1	3.59
G6	615582.93	3741692.77	10	50.3	35.3	14.4	3.57
G7	610276.82	3741956.2	0.5	86.2	13.8	0	2.22
G8	610728.4	3742558.31	5	63.1	31.2	5.7	3.55
G9	612007.89	3743724.9	11	57.2	37.6	5.2	2.09
G10	607567.31	3744853.86	0.5	75.9	21.5	2.6	2.8
G11	608395.22	3745230.18	5	86.4	11.3	2.3	2.27
G12	609787.6	3745681.77	12	88	11	1	2.05
G13	606250.19	3746434.41	0.5	85	14.6	0.4	3.17
G14	606852.31	3746584.93	6	91.2	6.9	1.9	3.34
G15	608395.22	3747375.21	11	81.2	11.1	7.7	3.55
G16	605045.97	3747638.63	0.5	88.7	11.1	0.2	3.32
G17	605949.14	3748278.37	7	98.5	1	0.5	2.65
G18	607228.63	3748918.12	12	69.1	22.4	8.5	2.96
G19	603841.75	3749369.7	0.5	73.2	21.1	5.1	2.62
G20	604293.33	3749369.7	5.5	67.3	27.4	5.3	3.28
G21	606062.03	3749821.29	10	71.7	21.5	6.8	3.21
G22	603352.53	3750461.03	0.5	82.5	15.8	1.7	2.47
G23	603917.01	3750536.29	5	79.1	16.4	4.5	2.05
G24	605873.87	3750649.19	11	68.4	12.8	18.8	2.01
G25	605610.45	3751213.67	10	54.8	32.5	12.7	2.34
G26	603390.16	3750423.4	7	57.2	38.2	4.6	1.9
G27	603917.01	3750498.66	0.5	51.9	22.7	25.4	1.18
G28	603615.96	3751025.51	5	49.5	31.4	19.1	1.12
G29	602524.63	3751439.46	9	37.8	29.3	32.9	1.79
G30	603126.74	3752116.84	5.5	19.3	41.3	39.4	0.33
G31	605083.6	3752417.89	10.5	18.7	24.1	57.2	1.22
G32	601884.88	3752718.95	1	12.8	21.3	65.9	1.93
G33	602524.63	3753057.64	8	14.9	22.9	62.2	2.2
G34	604519.12	3753358.69	10	15.1	30	54.9	2.34
G35	603164.37	3753584.49	22	18.4	41.3	39.8	2.15
G36	604029.91	3754186.6	10	22.8	29	48.2	1.59
G37	600454.87	3754562.92	0.7	25.3	37.9	36.8	2.25
G38	601546.2	3754675.81	7	25.6	36.2	38.2	0.35
G39	602863.32	3755127.4	10	42.8	35.1	22.1	1.11
G40	600116.18	3755127.4	0.5	43.1	37.2	19.7	0.22
G41	600605.4	3755277.93	5	57.2	36.1	6.7	2.69
G42	601336.47	3755842.41	8	64.7	33.1	2.2	3.42
G43	599702.23	3755955.3	1	74.2	19.2	6.6	1.82
G44	601395.67	3756181.01	9	84.9	14.3	0.8	1.3

The PLI which provides a simple comparative mean for assessing the level of metal contamination showed that the values oscillated between 1.1 and 13 and between 1 and 6.5 for surface and core sediments, respectively.

Table 4 Grain size and textural parameters (Folk 1966) for core sediments of the Ghannouch-Gabes coast

Core		Latitude (° N)	Longitude (° E)	Depth (m)	Sand (%)	Silt (%)	Clay (%)	Mz (Φ)
C1	C1-1	6181722.12	3740296.96	12.5	87.1	6.3	6.6	1.62
	C1-2				68.3	22.3	9.4	2.93
	C1-3				69.3	20.8	9.4	3.03
	C1-4				63.8	24.2	12	1.76
	C1-5				71.2	20.7	8.1	2.46
	C1-6				58.2	13.5	28.3	2.38
	C1-7				37.6	50	12.4	2.16
C2	C2-1	603615.52	3750923.15	3.5	8.8	13.8	77.4	2.02
	C2-2				1.2	13.6	85.2	3.34
	C2-3				1.8	11.8	86.4	3.18
	C2-4				3.8	17.1	79.1	2.96
	C2-5				5.3	21.3	73.4	2.47
	C2-6				5.7	22.4	71.9	2.27
	C2-7				11.1	21.6	67.3	2.86
C3	C3-1	604519.36	3753057.85	7.5	66.4	18.3	15.3	3.82
	C3-2				58.6	31.6	9.8	3.17
	C3-3				47.6	16.5	35.9	3.96
	C3-4				41.8	22.1	36.1	3.42
	C3-5				30.3	36.8	32.9	3.53
	C3-6				25.3	41.9	32.8	4.01
	C3-7				28.3	41.8	29.9	3.96

Discussion

Grain size distribution

The sediment texture was characterized by silt and clay in the inter-harbor area with a small sand fraction. This is may be due to the fineness of the phosphogypsum grain ($d_{50} = 0.05$ mm) and to the limited exchange with the open sea in this area. In the south of the study area, between Gabes wadi and El Fard wadi, there was a variation in trend, whereas the sediments are sandy with a low percent of clay and silt. This suggested that the recent deposition of sediments was more of sandy nature

and the probably source of these coarser sediments are not only from terrigenous supply but also from the open sea.

Mineralogical analysis

The mineralogical analysis was undertaken with the aim of characterizing the impact of anthropogenic releases on the mineralogical composition of surface sediments. The highest levels of quartz were detected in front of wadi Gabes and wadi El Fard which could be the main origin of terrigenous sediments. Thus, it appears that these two streams play an important role in transporting this mineral to the coast. Calcite was

Table 5   Pollutants concentrations in sediment samples from the Ghannouch-Gabes coast and other selected areas, UCC, and composition of Tunisian PG

	References	Major elements			Metal			
		CaO (%)	SO$_3$ (%)	F (%)	Pb µg/g	Zn µg/g	Cu µg/g	Cd µg/g
Range surface sediments	This study	3.3–45.1	0.06–32.04	0.11–12.19	0.1–3.8	18–57	2–14	2–59
Range core sediments	This study	26.76–47.34	0.12–54.77	–	0.4–0.7	3.9–52.1	2.1–12.7	1–63
Composition Tunisian PG	GCT (1992)	30.3	43.9	1.6	< 5	4	18	8
Crust average	Martin and Whitfield (1983)				16	127	32	0.2
Gulf of Tunis	Rais (1999)				12–112	70–226	–	–
Barcelona coast	Lopez-Sanchez et al. (1996)				91–1046	108–955	47–392	0.17–5.08
Algeria Bay	Boudjellal et al. (1993)				16–93	60-256	23–79	0.16–1.1

Table 6 Pollutants concentrations, geoaccumulation index (Igeo), contamination factor (C_f), and pollution load index (PLI) values in surface sediments from the Ghannouch-Gabes coast

Stations	Elements			Metal				PLI	Igeo Pb	Igeo Zn	Igeo Cu	Igeo Cd	C_f (Pb)	C_f (Zn)	C_f (Cu)	C_f (Cd)
	CaO %	SO$_3$ %	F (%) %	Pb µg/g	Zn µg/g	Cu µg/g	Cd µg/g									
G1	3.3	0.16	0.12	0.1	18	2	3	1.1	−0.6	0.6	−2.6	1.0	1.0	2.3	0.3	3
G2	4.3	–	0.12	–	27	2	4	–	–	1.2	−2.6	1.4	0.0	3.4	0.3	4
G3	7.8	–	0.13	–	27	3	3	–	–	1.2	−2.6	1.0	0.0	3.4	0.4	3
G4	9.4	0.14	0.17	0.3	35	4	3	2.1	1.0	1.6	−1.6	1.0	3.0	4.4	0.5	3
G5	3.6	0.51	0.11	0.4	36	5	3	2.4	1.4	1.6	−1.2	1.0	4.0	4.6	0.6	3
G6	10.4	0.59	0.18	0.4	28	5	2	2.1	1.4	1.2	−1.2	0.4	4.0	3.5	0.6	2
G7	9.2	–	0.14	0.5	24	–	3	–	1.7	1.0	–	1.0	5.0	3.0	0.0	3
G8	5.9	–	0.11	–	–	5	3	–	–	–	−1.2	1.0	0.0	0.0	0.6	3
G9	3.5	–	0.11	–	–	3	3	–	–	–	−2.0	1.0	0.0	0.0	0.4	3
G10	13.6	0.18	0.13	–	22	3	2	–	–	0.9	−2.0	0.4	0.0	2.8	0.4	2
G11	18.4	0.15	0.17	0.5	22	5	4	2.4	1.7	0.9	−1.2	1.4	5.0	2.8	0.6	4
G12	8	0.32	0.16	0.8	21	4	4	2.6	2.4	0.8	−1.6	1.4	8.0	2.7	0.5	4
G13	10.5	0.13	0.13	0.8	24	4	4	2.6	2.4	1.0	−1.6	1.4	8.0	3.0	0.5	4
G14	6	–	0.11	0.9	–	–	3	–	2.6	–	–	1.0	9.0	0.0	0.0	3
G15	8.7	–	0.12	0.9	32	5	4	3.1	2.6	1.4	−1.2	1.4	9.0	4.1	0.6	4
G16	10.7	–	0.13	0.9	34	5	4	3.1	2.6	1.5	−1.2	1.4	9.0	4.3	0.6	4
G17	14.6	–	0.14	–	34	6	4	–	–	1.5	−1.0	1.4	0.0	4.3	0.8	4
G18	10	0.86	0.21	0.9	35	6	13	4.5	2.6	1.6	−1.0	3.1	9.0	4.4	0.8	13
G19	18.6	0.06	0.15	1	32	7	4	3.5	2.7	1.4	−0.8	1.4	10.0	4.1	0.9	4
G20	2.4	–	0.22	1	32	–	10	–	2.7	1.4	–	2.7	10.0	4.1	0.0	10
G21	5.7	–	0.2	1	34	7	10	4.4	2.7	1.5	−0.8	2.7	10.0	4.3	0.9	10
G22	21.5	0.07	4.25	1	33	–	22	–	2.7	1.5	–	3.9	10.0	4.2	0.0	22
G23	–	–	4.25	–	34	8	21	–	–	1.5	−0.6	3.8	0.0	4.3	1.0	21
G24	33	1.47	4	1	33	8	24	5.6	2.7	1.5	−0.6	4.0	10.0	4.2	1.0	24
G25	25.9	1.1	3.72	1	34	8	22	5.6	2.7	1.5	−0.6	3.9	10.0	4.3	1.0	22
G26	37.9	–	3.75	1	–	–	29	–	2.7	–	–	4.3	10.0	–	–	29
G27	25.4	7.81	3.75	1.4	32	9	28	6.5	3.2	1.4	−0.4	4.2	14.0	4.1	1.1	28
G28	36.4	0.19	4	1.4	34	11	28	7.0	3.2	1.5	−0.1	4.2	14.0	4.3	1.4	28
G29	41.1	0.35	5.79	1.9	38	14	31	8.4	3.7	1.7	0.2	4.4	19.0	4.8	1.8	31
G30	45.1	32.04	10.25	–	38	14	33	–	–	1.7	0.2	4.5	–	4.8	1.8	33
G31	39.46	0.78	11.2	2.5	34	13	39	9.1	4.1	1.5	0.1	4.7	25.0	4.3	1.6	39
G32	22.2	0.51	12.19	3.8	57	14	59	13.0	4.7	2.3	0.2	5.3	38.0	7.2	1.8	59
G33	41.4	–	10.89	3.2	53	14	41	11.2	4.4	2.2	0.2	4.8	32.0	6.7	1.8	41
G34	40.3	0.44	10.25	–	41	12	27	–	–	1.8	–	4.2	–	5.2	1.5	27
G35	21.6	4.02	10.75	–	39	12	32	–	–	1.7	–	4.4	–	4.9	1.5	32
G36	15.9	0.57	9.66	1	39	11	25	6.4	2.7	1.7	−0.1	4.1	10.0	4.9	1.4	25
G37	15.2	0.67	8.95	1	38	10	25	6.2	2.7	1.7	−0.2	4.1	10.0	4.8	1.3	25
G38	13	6.06	6.19	1	31	10	25	5.9	2.7	1.4	−0.2	4.1	10.0	3.9	1.3	25
G39	13.5	0.44	5.79	0.9	30	7	26	5.3	2.6	1.3	−0.8	4.1	9.0	3.8	0.9	26
G40	3.9	–	5.62	0.6	29	4	20	3.9	2.0	1.3	−1.6	3.7	6.0	3.7	0.5	20
G41	6.5	0.28	5.54	–	–	3	22	–	–	–	−2.0	3.9	–	–	0.4	22
G42	11.9	0.57	5.34	0.7	–	2	22	–	2.2	–	−2.6	3.9	7.0	–	0.3	22
G43	–	–	5.34	–	–	–	32	–	–	–	–	4.4	–	–	–	32
G44	–	–	–	0.1	21	2	18	1.9	−0.6	0.8	−2.6	3.6	1.0	2.7	0.3	18

Table 7 Pollutants concentrations, geoaccumulation index (Igeo), contamination factor (C_f), and pollution load index (PLI) values in core sediments from the Ghannouch-Gabes coast

Stations	Elements			Metal			PLI	Igeo Pb	Igeo Zn	Igeo Cu	Igeo Cd	C_f (Pb)	C_f (Zn)	C_f (Cu)	C_f (Cd)
	CaO %	SO$_3$ %	Pb µg/g	Zn µg/g	Cu µg/g	Cd µg/g									
C1-1	36.96	0.12	0.7	3.9	2.2	1.2	1	2.2	−1.6	−2.4	−0.3	7	0.5	0.3	1.2
C1-2	30.09	0.12	0.6	4.1	5.4	1.1	1.2	2.0	−1.5	−1.1	−0.4	6	0.5	0.7	1.1
C1-3	32.11	0.13	0.5	6.3	3.2	2.1	1.4	1.7	−0.9	−1.9	0.5	5	0.8	0.4	2.1
C1-4	31.78	0.26	0.6	5.2	4.8	1.5	1.4	2.0	−1.2	−1.3	−	6	0.7	0.6	1.5
C1-5	29.32	0.34	0.6	4.6	5.7	2.5	1.6	2.0	−1.4	−1.1	0.7	6	0.6	0.7	2.5
C1-6	36.28	0.35	−	6.1	6.3	1.1	−	−	−1.0	−0.9	−0.4	−	0.8	0.8	1.1
C1-7	37.09	0.66	0.1	7.9	7.9	1	−	−	−0.6	−0.6	−0.6	−	1.0	1.0	1
C2-1	29.11	51.02	0.5	48.5	12.7	37	6.5	1.7	2.0	0.1	4.6	5	6.1	1.6	37
C2-2	30.42	51.01	−	48.7	8.9	34	−	−	2.0	−0.4	4.5	−	6.2	1.1	34
C2-3	28.19	51.16	0.5	41.3	7.4	33	5.3	1.7	1.8	−0.7	4.5	5	5.2	0.9	33
C2-4	27.86	50.21	0.6	31.1	6.3	37	5.1	2.0	1.4	−0.9	4.6	6	3.9	0.8	37
C2-5	26.76	49.89	0.4	10.5	4.7	41	3.4	1.4	−0.2	−1.3	4.8	4	1.3	0.6	41
C2-6	29.56	52.54	−	10.4	4.3	33	−	−	−0.2	−1.5	4.5	−	1.3	0.5	33
C2-7	28.36	54.77	−	52.1	4	28	−	−	2.1	−1.6	4.2	−	6.6	0.5	28
C3-1	30.01	0.12	0.7	43.5	2.1	63	5.0	2.2	1.9	−2.5	5.4	7	5.5	0.3	63
C3-2	47.25	0.12	0.6	23.8	4.1	46	4.6	2.0	1.0	−1.5	4.9	6	3.0	0.5	46
C3-3	46.18	0.12	0.5	25.8	11.9	34	5.4	1.7	1.1	0.0	4.5	5	3.3	1.5	34
C3-4	47.25	0.14	0.6	17.5	5.7	21	3.8	2.0	0.6	−1.1	3.8	6	2.2	0.7	21
C3-5	40.22	0.25	0.6	11.2	3.6	21	3.0	2.0	−0.1	−1.7	3.8	6	1.4	0.5	21
C3-6	42.45	0.37	−	7.7	4.6	13	−	−	−0.6	−1.4	3.1	−	1.0	0.6	13
C3-7	47.34	0.65	−	7.8	6.1	14	−	−	−0.6	−1.0	3.2	−	1.0	0.8	14

identified essentially in the south of the study area, but almost absent in the area between the two harbors; this is due to its instability and its dissolution in the acidic marine water in this zone (pH = 4) (Ben Amor and Gueddari 2016), and calcite probably has a biogenic origin. For core 1, the proportions of calcite and quartz remained fairly stable throughout the core column and showed no significant variation. They have the same origin as surface sediments. Gypsum was identified only in some samples picked in the area between the fishing and commercial harbors and in the full length of core 2. The diffractogram shows an offset from the main peak, signifying that the gypsum was derived from PG discharge (anthropogenic origin) (Fig. 2). The fluorine was identified only in few surface samples located in the area II, taken in front of discharge of PG. Fluorine has been shown by polarizing microscope. This mineral is in the form of cubic crystals, extinguished under polarized light. Fluorine may have two possible origins, either solid discharges from the fluoride chemical industry, which processes fluorinated ores, or the precipitation from the elements dissolved in water, since the waters taken from the zone of influence of the PG discharges have relatively high Ca^{++} and F$^-$ concentrations and that the saturation test has shown that these waters are supersaturated

with respect to this mineral (Ben Amor and Gueddari 2016). The mineralogical composition of the surface and core sediments are highly influenced both by the anthropogenic release and by terrigenous transport and biogenic precipitation.

Geochemical analysis

Calcium (CaO%) and sulfur (SO$_3$%) are known to be enriched in the Tunisian PG (Table 5) and can be taken as its geochemical identity. The CaO contents are, for the most part, associated with gypsum and carbonate minerals (calcite, dolomite, aragonite, and magnesium calcite). The spatial distribution showed that the highest concentrations of calcium were found in the sediments taken in the area limited by Ouadi Gabes and Ouadi Ettine (Fig. 3), from the coast to the open sea for superficial sediments; while for the core subsamples, the highest contents of CaO were recorded for cores 2 and 3 located in the same area. Calcium at C1 generally tended to decrease towards the deep layers (Fig. 6). The CaO (%) may have different origins such as the water supply, and it alters limestone and the gypsum land; the solid phases contain 31.3% (GCT 2003) and/or the carbonated tests. The highest levels of total sulfur contents were measured in the area between the two harbors
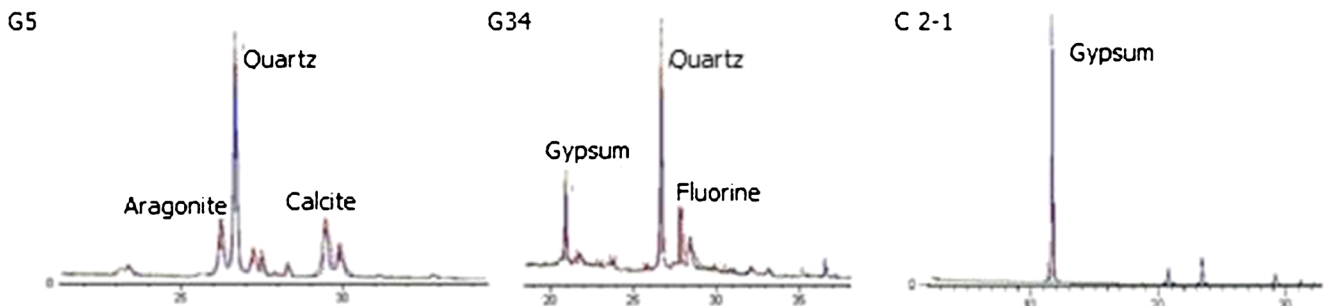
Fig. 2 X-ray diffraction data for surface and bottom sediments. Sample number G5 (south of the study area), sample number G34 (between commercial and fishing harbor), and core sample C2-1 (PG discharge area)

and tend to decrease towards the open sea. The similar trend was observed for core samples where only the core 2 showed a high concentration of sulfur. The highest content of sulfur was exhibited in the sediments where gypsum was identified. The sulfur would mostly be in the form of sulfate in gypsum (Table 5). The spatial distribution of fluoride showed that the concentrations tend to decrease with increasing distance (Fig. 4). The comparison of fluoride contents with non-contaminated superficial marine sediments (0.055%) shows that all sediments taken are contaminated by fluoride (Da Lazzari et al. 2004).

Spatial distribution of trace metals (Cd, Cu, Pb, and Zn) showed higher values at near-shore sediments, in the zone II, located between the commercial harbor and the fish harbor of Gabes (Fig. 5). Lower trace metal concentrations in the south of study area were observed, probably due to low anthropogenic activities and a limited population density and urbanization in this area. The abundance in the order of these metals was found to be Zn > Cd > Cu > Pb with mean concentrations of 27.88, 16.2, 6.06, and 0.22 μg/g, respectively, in surface sediments (Fig. 6). The comparison of heavy metal contents measured from the GG coastline with those reported in sediments from different Mediterranean regions (Table 5) showed that Cd and Zn concentrations in surface and core sediments are high (Fig. 7). The PG impact is clearly established in the surface and lower sediments which suggests an important and continuous input of cadmium, in particular, by PG release. Cu and Pb concentrations are below those reported in previous studies. These elements do not appear to be problematic here when compared to another marine environment or to crust average (Table 5). The observed low concentrations of lead in surface and bottom sediments are probably due to a low concentration of Pb in the Tunisian PG (< 5 μg/g) (GCT 2003).

Assessment of trace metals

According to the classification adopted by Hakanson (1980), the contamination factor (C_f) values for surface sediments were found to be "moderate" for Zn in all study area, between "moderate" and "considerable" for Cu, and "considerable" to "very high" for Pb. The C_f values of cadmium were identified
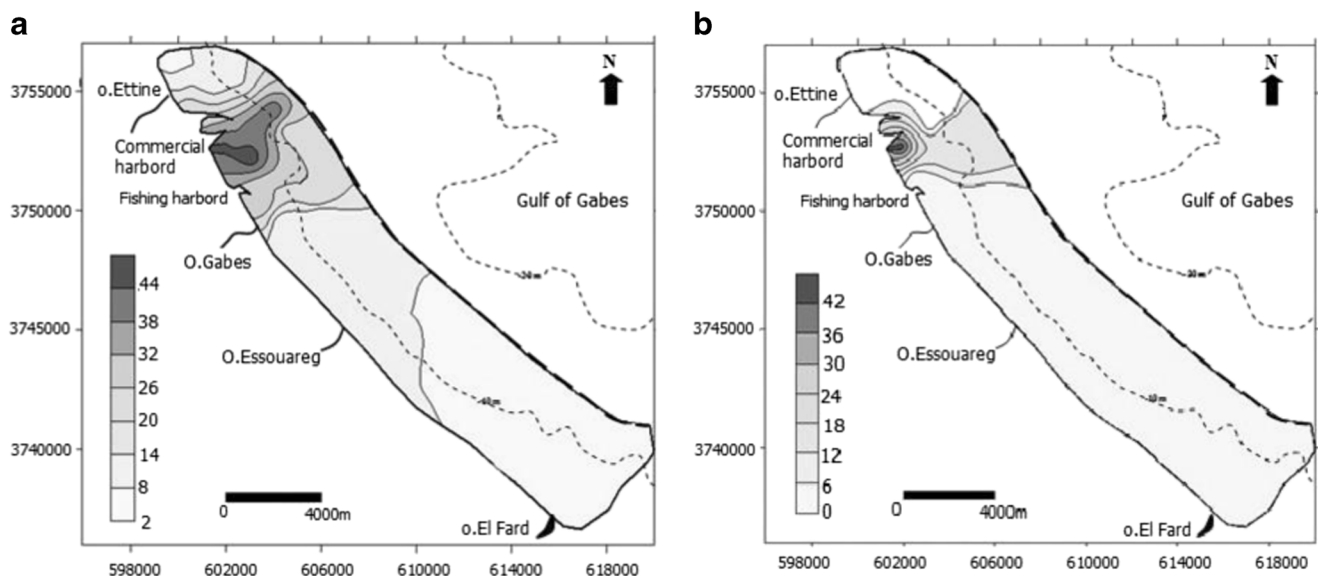
Fig. 3 Spatial distribution of **a** CaO (%) and **b** SO₃ (%) in the surface sediments of the Ghannouch-Gabes coast
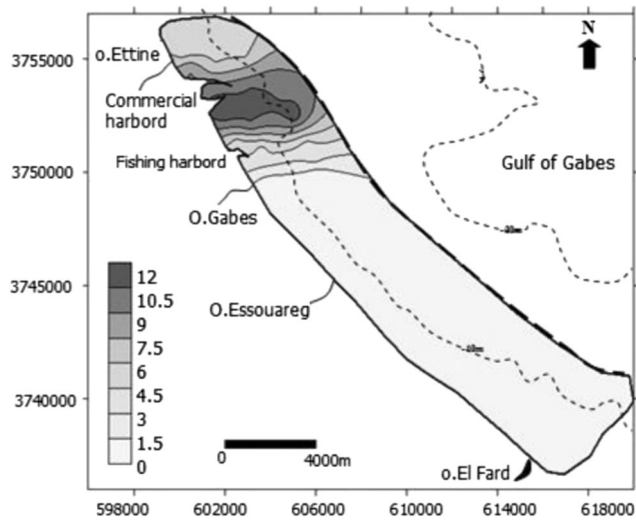
Fig. 4 Spatial distribution of fluor (%) in surface sediments of the study area

to be "moderate" for the sampling taken in the zone I and "considerable" to "very high" for the sampling taken between the two harbors. Similarly, C_f is low for Zn, Cu, and Pb and moderate for Cd in core C1 considered as a reference core. The contamination factor values are very high for Pb and Cd, moderate to low for Cu, and between very high and considerable for Zn in core C2. Core C3, collected between fishing and commercial harbor, indicates a "very high" C_f for Pb, between "very high" in the tops of the core and "moderate" in the bottom for Zn, "low" factor for Cu, and "very high" for Cd.

The calculated results for Igeo indicated that the surface sediments taken between the commercial and the fishing harbors can be considered as from "strongly contaminated" to "moderately/strongly contaminated" for Pb, Cu, and Cd. In the rest of the study area, the sediments can be considerate to be "uncontaminated" to "moderately contaminated" sediments. The Igeo results for Zn showed that all samples were
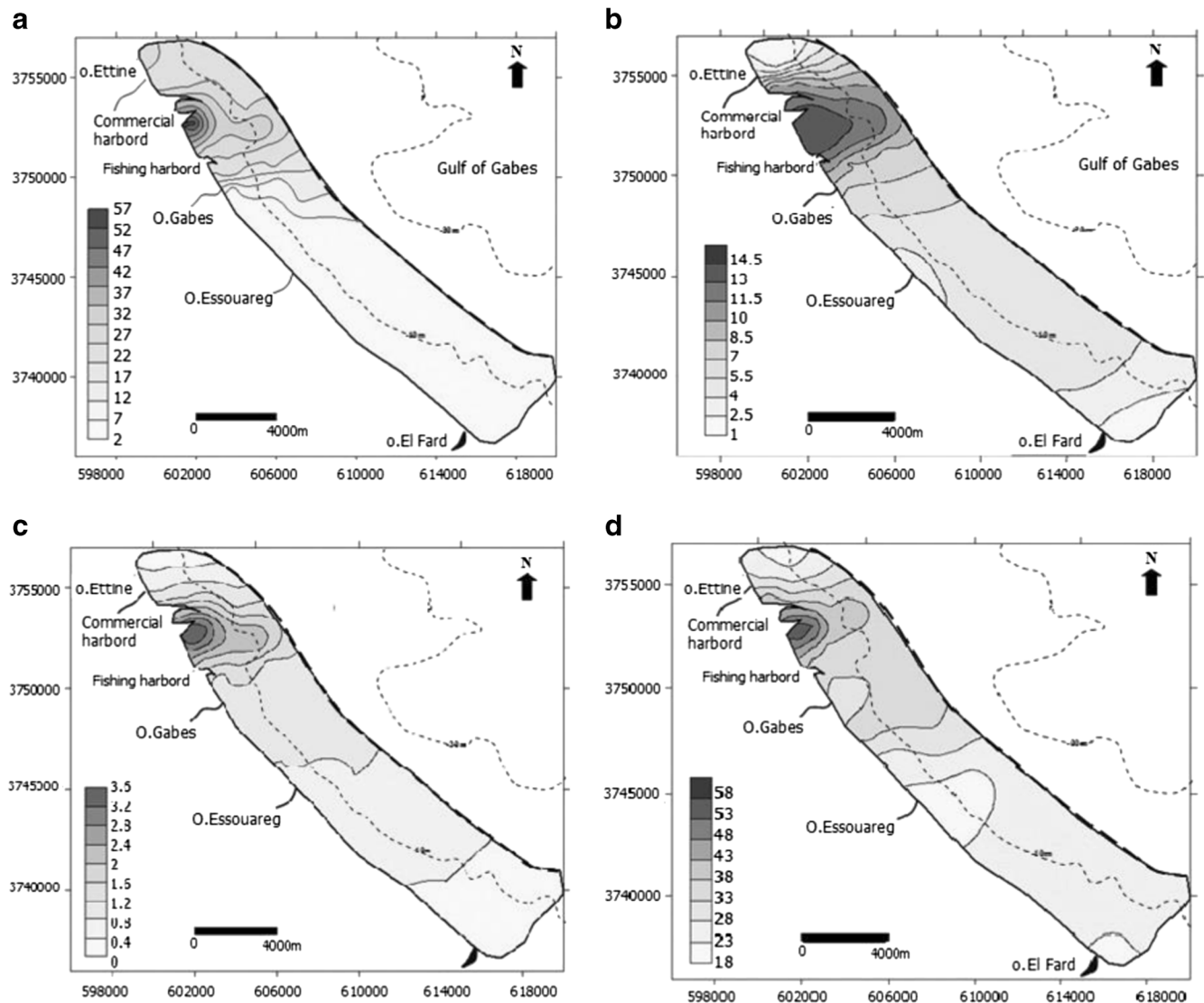
Fig. 5 Spatial distribution of concentrations of **a** cadmium, **b** copper, **c** lead, and **d** zinc (μg/g) in surface sediments of the Ghannouch-Gabes coast

Fig. 6 Vertical distribution of CaO and SO₃ (%) in the core sediments of the Ghannouch-Gabes coast
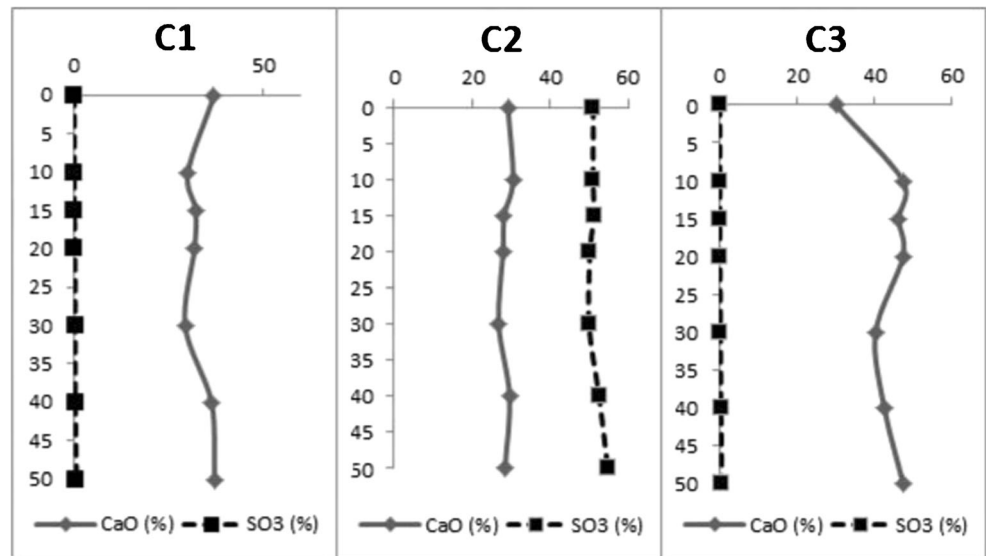
"uncontaminated." The Igeo values indicated that core C1 being a reference is moderately contaminated for Pb and

uncontaminated for Zn, Cu, and Cd. For core C2, Igeo is moderate in the first 25 cm, uncontaminated in the bottom

Fig. 7 Vertical distribution of trace elements Pb, Cu, Zn, and Cd (μg/g) in Ghannouch-Gabes sediment cores
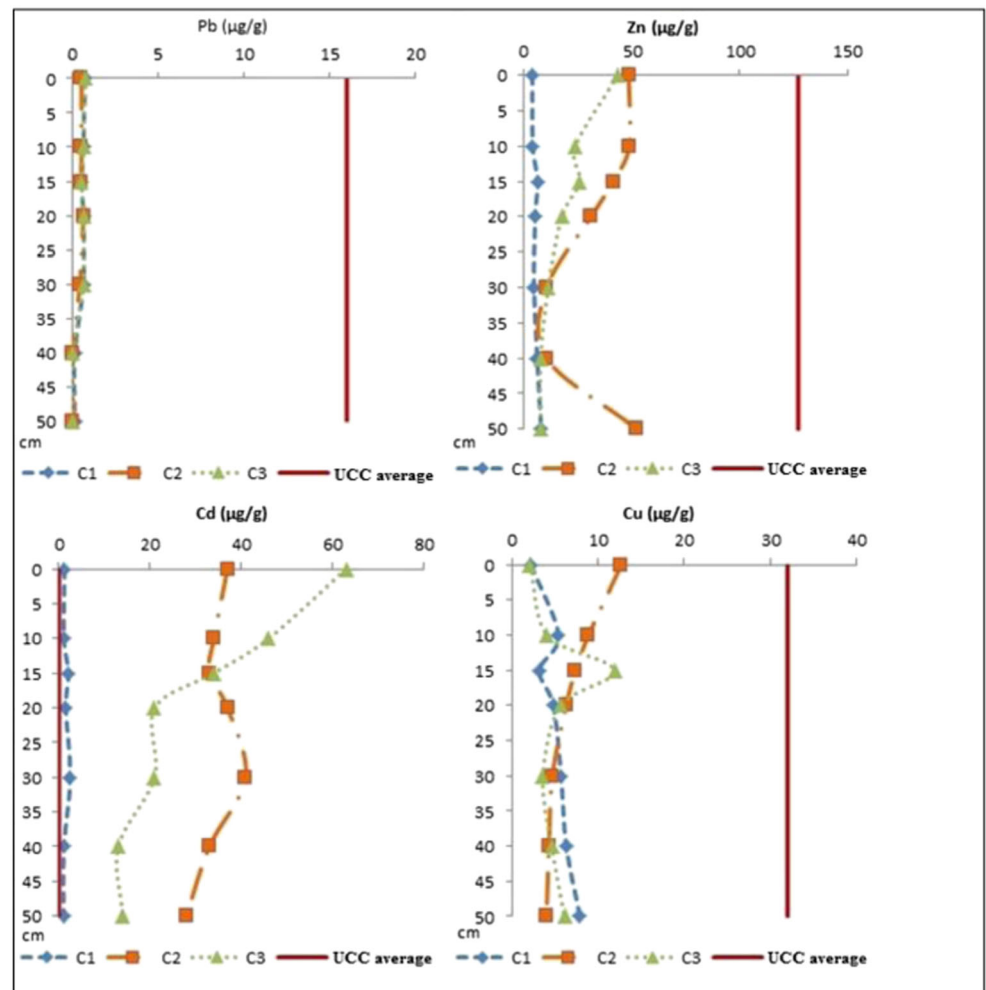
Table 8 Pearson coefficient for Cd, Cu, Zn, Pb, F, CaO, and SO₃ in surface sediments of the Ghannouch-Gabes coast

	Cd (μg/g)	Cu (μg/g)	Pb (μg/g)	Zn (μg/g)	F (%)	CaO (%)	SO₃ (%)
Cd (μg/g)	1	0.64	0.61	0.37	0.90	0.61	0.26
Cu (μg/g)		1	0.51	0.70	0.72	0.67	0.37
Pb (μg/g)			1	0.50	0.47	0.49	− 0.10
Zn (μg/g)				1	0.41	0.41	0.16
F (%)					1	0.61	0.33
CaO (%)						1	0.39
SO₃ (%)							1

for Pb, strongly to extremely strongly contaminated for Cd, and uncontaminated for Cu. All subsamples of C3 are uncontaminated for Cu, moderate to strongly contaminated for Pb, between strongly and extremely contaminated for Cd, and moderately to uncontaminated for Zn.

Basing on the interpretation suggested by Tomlinson et al. (1980), the calculated PLI values in all sites were > 1. All surface and bottom sediments collected in the coast of GG can be classified as "polluted," the high value of PLI characterizes samples taken in the inter-harbor zone, near the PG discharge, indicating that the sediments of this area are strongly polluted by the examined heavy metals, specially by cadmium.

Basing on the different pollution indices (C_f, PLI, and Igeo), the zone situated between the commercial harbor of Ghannouch and the fishing harbor of Gabes is the most polluted area both for surface and core sediments. However, a downward trend was observed from surface to bottom sediments, from the north to the south of study area according to the littoral drift. This is due essentially to the continuous PG discharge in this zone. In addition, the two harbors limited the

extension of PG into the open sea and transformed the inter-harbor zone into a confined area, where the harbor piers (which extend more than 1.5 km) acted as barriers and limited the dispersion of the PG discharge.

Statistical analysis

In order to establish the relations between elements in GG coastal sediments, PCA was used. Only surface sediments were considered, as shown as Table 8, and most of the analyzed elements were correlated positively with each other.

The significantly positive correlation of Cu ($r = 0.64$; $n = 44$), Pb ($r = 0.6$; $n = 44$), and F (0.90; $n = 44$) with Cd and of Zn ($r = 0.70$; $n = 44$), F ($r = 0.72$; $n = 44$), and CaO ($r = 0.67$; $n = 44$) with Cu could indicate the similar origin. The elements that have high correlation in sediments may have common origins, the same behaviors during the different phases of transformation and migration (Wang et al. 2012). Therefore, Cd, Cu, Pb, Zn, F, and CaO may have a common source, which is most probably PG discharged from a chemical group.

Fig. 8 Loading plots of the two factors obtained with corresponding analysis
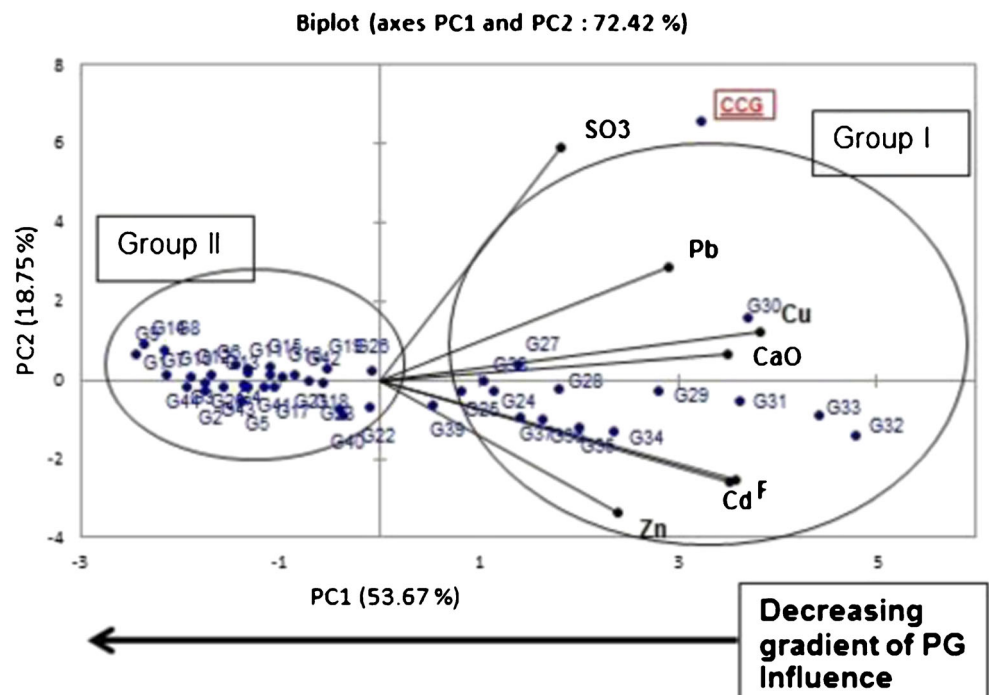
Corresponding analysis was applied to all sampling stations (44) and studied variables (7), in order to illustrate relations between stations and parameters. This analysis identified two principal factors (F1 and F2) that explained 73.7% of the total inertia of observations. The loading plot (Fig. 8) of the two factors showed the presence of two groups:

– The first group included the stations G24 to G41 (located between the two harbors) and are dominated by high concentrations of ETM, F, and CaO. It testified the impact of the industrial sources.
– The second group included the stations between G1 to G23 and are independent of all pollutants indicating their non-contamination.

The compiled results of grain size distribution and mineralogical, geochemical, and statistical analysis showed that the coast of GG can be divided into two zones: the first zone located in the inter-harbor zone, the surface and bottom sediments of this area are the most polluted by trace metals and fluorine and are the most charged by calcium and sulfate. This area receives a high level of industrial waste discharge, especially PG. The finest grain size distribution promotes the accumulation of trace metals, and the presence of harbor piers acted as barriers for the dispersion of industrial waste. The northern area of the chemical complex is protected from contamination by the commercial harbor piers and the north-south coastal currents. The surface and bottom sediments of the second zone, located in the southern area, are less polluted than those in a previous area. This spatial distribution is probably due to the grain size distribution where sand was dominating. In fact, large amounts of trace metals are bound in the fine-grained fraction (< 63 mm) of the sediments, mainly because of its high surface area-to-grain size ratio (Seshan et al. 2010; Armstrong-Altrin et al. 2017). Mineralogically, quartz and calcite are predominant, and this is probably due to the source rock supply and to the deeper sediments' contribution.

Conclusion

Different useful tools, indices, and approaches (mineralogical analysis, CaO (%), SO$_3$ (%), some trace metals (Cd, Pb, Zn, and Cu), numerous indexes, and statistical approaches) have been employed for the evaluation of sediment contamination of the GG coastline. This paper has clearly shown that the coast of GG is polluted by high concentrations of several pollutants. To assess the pollution status of trace metals, concentrations of Cd, Cu, Zn, Pb, F, CaO, and SO$_3$ have been estimated in surface and core sediments. The order of mean concentrations of these metals was Zn > Cd > Cu > Pb. The highest values of metal concentrations and all environmental indices studied were found in the zone between the two harbors, both for

surface and deep sediments, which receive a high level of waste discharge, especially PG. The northern and southern zones of the studied area are protected by the harbor piers. This spatial distribution of pollutants is mainly under the influence of harbor installations, coastal currents (principal current drift from north to south), and the grain size distribution. The results of vertical distribution, which showed an enrichment of Cd, Pb, and Zn in the uppermost layers of core sediments which remain lower than crust average for Pb and Zn, suggested that was due to an excess of industrial loadings in the recent past and that it will continue. Multivariate analysis identified two components (F1 and F2), which are noted with Cd, Pb, Zn, Cu, F, and CaO content and are related to the anthropogenic origin. A significant positive correlation is observed among these pollutants, indicating that these pollutants result from similar sources and moving together.

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