ASSESSMENT OF TRACE METAL CONTAMINATION AND PHOSPHORUS DYNAMIC IN SEDIMENTS OF MONASTIR BAY (TUNISIA)

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Abstract – Located in the eastern coast of Tunisia, the Monastir bay is considered as a fragile area due to its weak water renewal and its high anthropogenic discharge, which influence the physico-chemical quality of the water as well as the sediment. To establish the state of sediment ‘s pollution of Metallic Trace Elements (MTE), Total Organic Carbon (TOC), Total Phosphorus (Total P), Phosphorus particulate fractions, and granulometric distribution were analyzed at sediment sampling stations. The purpose of this research is to evaluate the contamination of the surface sediment and to discuss the sediment phosphorus dynamic: is sediment a sink or a source of phosphorus? Then the geochemical data and the statistical analysis highlighted a moderate pollution of the sediment, in particular by Mn, Ni and Cu, in the northern part of the bay. Overall, the MTE concentrations are much lower than those reported in the Gulf of Tunis and Gabès. Although the sediments of the bay of Monastir do not seem to be affected by a strong inorganic pollution even with the large impact of human activity. Furthermore the sediment phosphorus distribution seems to be controlled by the phosphorus bound to refractory organic matter (Residual Organic Phosphate, ROP) and bound to Calcium carbonate (P-$CaCO_3$). The ROP fraction concentration is very high and can be interpretate by the enrichment with seagrass. Furthermore, the PCA results show two trends: surface sediment enriched with iron (Fe) and TOC versus an other one enriched with carbonate but with weak concentration Fe and TOC. The phosphorus dynamic is controlled by environmental conditions, (i) oxic sediment with basic pH conditions enhance the FeOOH formation and consequently the phosphorus fixation on FeOOH or CaCO3 in sediment (P-FeOOH, P-CaCO3) whatever the sediment lithology and (ii) anoxic and acidic sediment enhance the formation of ROP (Residual Organic Phosphorus) in correlation with the presence of aquatic plants (Posidonia meadow) in the sediment.

Introduction

Coastal ecosystems represent 6 % of earth surface and 8,5% of marine biomes, they are among the most productive in the world. Mediterranean Sea holds a wide biodiversity, even with its little surface area, which is 0,7 % of global marine waters [14], [16]. Scientific community is interested in the sensitivity of coastal zones to anthropogenic discharges [18], [32]. Indeed, Mediterranean ecosystem has been affected by natural and anthropic factors,

Referee List (DOI 10.36253/fup_referee_list)
FUP Best Practice in Scholarly Publishing (DOI 10.36253/fup_best_practice)
Amina Ben Mefteh, Valérie Mesnage, Sarra Ben Jeddi, Mohamed-Amine Helali, Noureddine Zaaboub, Jean-Marie Barrois, Walid Oueslati, *Assessment of trace metal contamination and phosphorus dynamic in sediments of Monastir Bay (Tunisia)*, pp. 378-389 © 2022 Author(s), CC BY-NC-SA 4.0, 10.36253/979-12-215-0030-1.34

especially wastewater rejects. The latters influence chemicophysical quality of marine sediments [4].

Monastir bay is considered as a vulnerable area on account of its reduced hydrodynamic [36]. The bay has been studied in order to determine pollution levels in (i) organic matter and MTE [34], (ii) in hydrocarbons [39], (iii) in PCB et PBDE [27]; these studies lead to the conclusion that Monastir bay surface sediments are enhanced with an organometallic pollution. Monastir bay is used for aquaculture, so it is regularly dragged to pull out sediment stocks charged in nutrients. Khniss drain, built in early 1970's, collects domestic and industrial wastewaters of nearby cities. It permits the drainage of Monastir sebkha water to sea during rise period [34]. Consequently, Khniss drain is considered as the most important source of nutrients and MTE of the bay [6]. All these anthropogenic activities generate organic pollution that causes eutrophication. This leads to chronic anoxia of the Bay's surface waters as well as the emission of solid waste and polluted water [19]. These pollutants come from many sources such as wastewater treatment plants of Khniss, Ksibet El Mediouni, Lamta-Sayada Monastir cities which are discharged into the bay, the Khniss drain and the fishing harbours.

The aim of this research study is to discover the state of contamination of the sediments surface and to discuss the sediment phosphorus dynamic: is sediment a sink or a source of phosphorus? Thus Metallic Trace Elements (MTE), Total Organic Carbon (TOC), Total Phosphorus (Total P), Phosphorus particulate fractions, and granulometric distribution have been measured in surface sediment.

The results allowed us to answer the following scientific questions: (i) Is the pollution of Monastir bay's sediments similar to other marine environments in Tunisia? (ii) Is the Monastir bay surface sediment polluted according to the sediment quality standards? The scientific interest is in the chemical characterisation, by fractioning the forms of particulate phosphorus to discuss the dynamics of sedimentary phosphorus in relation to other sediment characteristics (MTE, TOC, Carbonate).

Material and methods

Monastir bay (35°47'N et 35°37'N; 10°45'E et 11°50'E) is closed, to the north by a rocky escarpment of 17 m high (Cap Monastir) and to the south by the high ground of Teboulba extending as far as the Kuriat islands (Figure 1). This marine environment is characterised by a bathymetric slope in its north-eastern direction: the -1 m isobath is located at a distance of 1 mile from the shore. This particularity is at the origin of the formation of an extensive flat area [1], [28]. The bottoms closest to the coast are generally sandy-muddy to muddy and are largely covered by a black muddy film with various seagrass beds favourable to many organisms [33], [12]. The prevailling winds are characterised by a speed between 1 and 5 m/s and come from three main directions of W, NNE and E [36] – figure 1. Socio-economic context of this bay is characterised by strong development, one of the most intense that Tunisia has undergone in the last twenty years. The textile industry predominating, covering 79 % of the regional industrial fabric [19]. This region concentrates 12 % of Tunisia's industrial activity [7] and 44 % of the national fisheries production comes from fishering and aquaculture in the bay with four fishing harbours equipped with refrigeration complexes [11] – Figure 2. Geological context, described in the works of [23] and [31], is composed of Mio-Plio-Quaternary formations at the level of the Monastir cliff and Tyrrhéniennes formations (Ksibet Mediouni) at the level of the hills bordering the Bay. Watershed belongs to the Sahel platform and is formed by the Moknine tableland and the two NE-SW anticlines of Bir and Taieb and Zéramdine, separated by the broad syncline of Jammel, which in turn passes to the NE at the anticline of Bodeur. The Monastir Sebkha is boundes by the Skanes-Khnis fault to the east and the Sahline flexure to the west. The sedimentary formations spread out from the Miocene to the Quaternary, are essentially formed of sandy-clay continental sediments, with in the Lower Pliocene and Tyrrhenian, marine carbonate deposits, of a sandy to gravelly nature that are more or less consolidated, oolitic or calcarintic. The bay is therefore composed mainly of fine and medium sands including a variety of minerals, especially quartz and calcite [9].

Surface sediment sampling stations were sampled in April 2017 with a Shipek-type sediment grab (Figure 2). The measurement of Total Organic Carbon (TOC) was carried out using the Walkley-Black method [10]. The determination of carbonates was carried out by the calcimeter method of Bernard. For the MTE analyses sediment samples were digested in teflon bombs using a mixed solution (*v*/*v*) 20 mL $HClO_4$ and 10 mL HF, method following the protocol of [29]. Then, MTE (Fe, Mn, Pb, Zn, Cu, Ni, and Co) were analyzed by flame atomic absorption spectrometry (SOLAAR). Phosphate fractionation was carried out according to the chemical method of [20]. The inorganic fraction phosphate: iron bound-phosphate, Fe(OOH)-P and calcium-bound-phosphate, $CaCO_3$-P, have been extracted with chelating agents (NTA and EDTA). Acid Soluble Organic Phosphate (ASOP) has required a single extraction with H_2SO_4 0.25 M. Then followed a 2 M NaOH (90°C) with heating step to extract the Residual Organic Phosphate (ROP). The GIS spatial representation has been performed with Surfer software [2]. Statistical analyses, with a Principal Component Analysis (PCA) and inter-elemental correlation have been performer by PCA R-software.

Results

Organic carbon content (TOC - Table 1) fluctuates from 1 to 5.8 %; highest concentrations are near the Monastir Harbour and in the aquaculture area (sediment sampling stations 2, 7, 8 – Figure 2). High concentrations of CaCO3 from 32 to 65 % (table 1) in sediments can be easily explain by the sedimentary carboneous origin of the watershed around the Monastir Bay.

The spatial variation of MTE concentrations in the surface sediments are presented in figures 3, 4 and 5. The copper and nickel concentrations vary between 16 and 58 ppm and between 9 and 44 ppm respectively (figure 3), with maximum concentrations in the sediments of the Monastir and Sayada Harbours' (Figure 3). Measured copper and nickel values have increased since 2015 [13] and then remained stable between 2017 and 2020 [7]. The copper concentrations in the sediments of Monastir bay are slightly higher and those of nickel of the same order of magnitude as in other Tunisian coastal ecosystems studied, in Sfax [21], in the El Melah lagoon [22] and in the Golf of Tunis [38] and Gabès [3], [15].

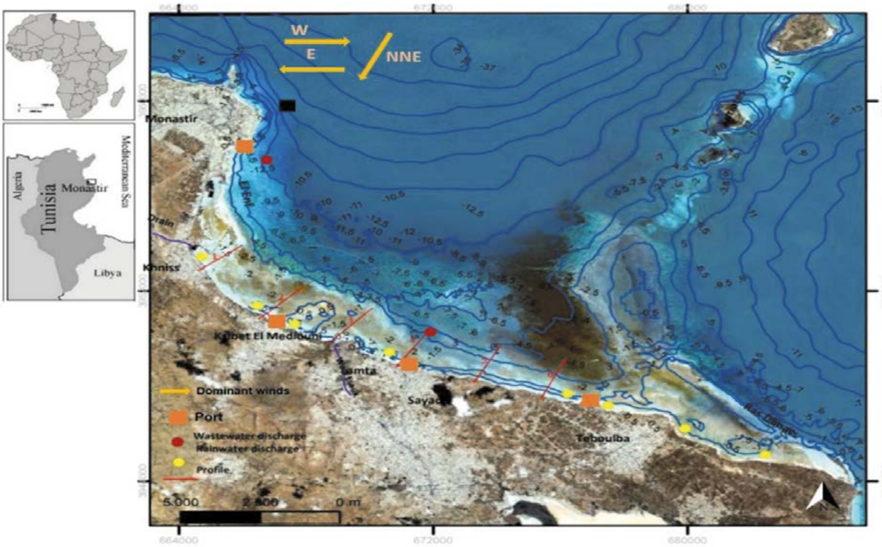

Figure 1 – Location and bathymetry of the study area: The Monastir Bay in Tunisia.

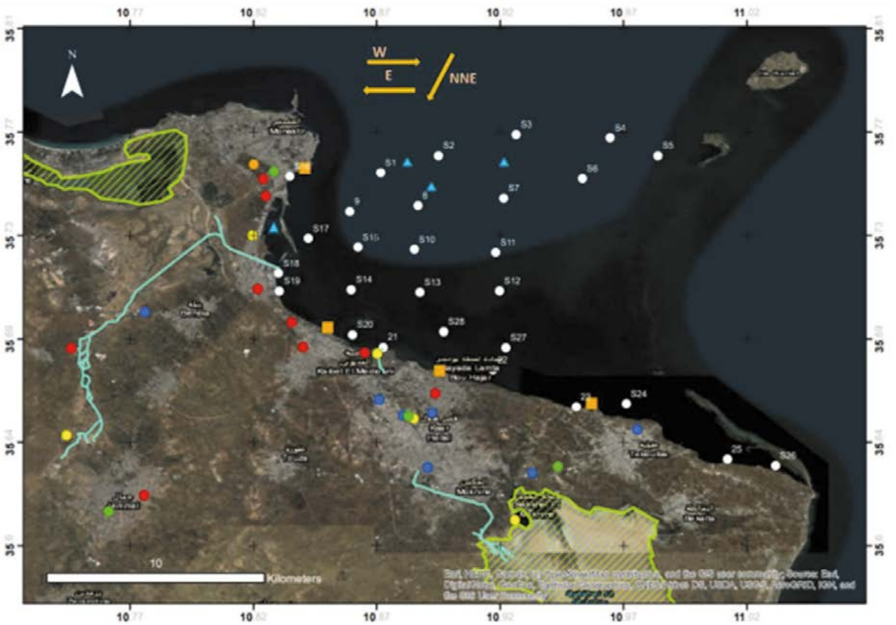

Figure 2 – Location of the sampling sites (white circles) and the anthropogenic activities and of the Monastir Bay: aquaculture (blue triangle), sewage treatment (yellow circle), textile factory (blue circle), chemical factory (red circle), car factory (green circle), harbour (orange square), drains (blue line), sebkra (green hatching).

Table 1 – Granulometric and geochemical characteristics of sediment.

Sample stations	**1**	**2**	**6**	**7**	**8**	**9**	**10**	**11**	**12**	**14**	**15**	**16**	**17**	**18**	**24**	**25**	**28**
Lat.	35° 45’ 20.08’’	35° 45’ 45.12’’	35° 45’ 11.47’’	35° 44’ 42.56’’	35° 44’ 32.02’’	35° 44’ 23.48’’	35° 43’ 26.50’’	35° 43’ 22.22’’	35° 42’ 25.37	35° 42’ 27.21’’	35° 43’ 29.83’’	35° 45’ 14.81’’	35° 43’ 42.75’’	35° 42’ 51.27’’	35° 39’ 38.54’’	35° 38’ 16.89’’	35° 41’ 25.24’’
Long.	10° 52’ 7.79’’	10° 53’ 33.73’’	10° 57’ 7.87’’	10° 55’ 11.04	10° 53’ 3.26’’	10° 51’ 21.92’’	10° 52’ 58.10’’	10° 54’ 59.16’’	10° 55’ 4.92’’	10° 51’ 24.19’’	10° 51’ 34.30’’	10° 49’ 52.60’’	10° 50’ 20.27	10° 49’ 35.70’’	10° 58’ 13.43’’	11° 0’ 43.93’’	10° 53’ 41.81’’
Granulometry (< 63 µm) (%)	27.6	25.93	41.84	32.58	47.29	38.44	19.54	15.92	61.58	15.68	nm	8.51	27.33	27.6	13.97	9.76	55.08
MgO (ppm)	1.68	1.95	2.79	2.82	1.92	1.59	1.96	2.13	3.02	2.52	nm	1.33	2.31	2	1.15	1.31	3.15
O.M. (%)	7.44	22.53	19.56	22.1	19.1	17.2	14.86	nm	nm	8.23	18.6	nm	14.28	9.85	nm	nm	18.46
T.O.C (%)	1.4	5.5	4.37	4.14	3.3	3.04	2.43	5.54	5.8	2.1	3.3	0.97	2.1	3.38	1.5	1.82	2.9
$CaCO_3$ (%)	44.25	31.99	48.27	36.45	36.85	41.93	51.31	nm	54.64	61.53	41.12	nm	63.53	36.77	nm	nm	65.26
Zn (ppm)	37	68.53	48.11	123.44	61.7	88	64.18	69.51	86.5	41.75	nm	117.3	57.94	45.46	38.63	47.44	41.42
Pb (ppm)	19.65	56.44	25.18	30.14	37.2	25.72	26.2	53.76	35.23	3.94	nm	47.96	19.2	21.5	26.96	19.09	11.45
Cd (ppm)	0.17	0.82	0.72	0.33	0.4	0.5	0.81	0.68	0.76	1.4	nm	0.62	0.64	0.13	0.71	0.68	121.43
Cu (ppm)	18.83	29.1	24,95	26.04	23.75	23.56	22.36	34.6	24.24	21.3	nm	58.45	28.72	20.75	26.2	34.6	16.23
Ni (ppm)	16.7	22.5	28.9	28.44	27.28	22.81	22.06	40.6	16.8	30	nm	44.5	28.1	16.64	20.25	18.76	9.43
Fe (%)	0.63	1.3	1.36	1.63	1.56	1.15	0.74	1.82	0.74	0.52	nm	1.14	0.66	0.96	0.63	0.35	0.59
Mn (ppm)	105.76	103	84.4	121.95	90.63	85.56	85.71	110.25	75.2	117.9	nm	308.32	76.91	120.21	115.94	103.1	88.3
Sr (%)	0.54	0.38	0.47	0.36	0.43	0	0.6	0.34	0.51	0.69	nm	0.31	0.7	0.34	0.3	0.55	0.54
Cr (ppm)	48.75	62.55	113.5	88.3	88.9	72.85	63.03	70.75	79.5	106.6	nm	94.31	133.6	99.63	76.14	63.6	80.4
P-FeOOH ($\mu gP.g^{-1}$)	24.4	111.9	16.8	43.6	30.7	66.9	41.5	nm	nm	100.3	34.4	167.6	66.6	19.7	39.3	nm	69.4
P-$CaCO_3$ ($\mu gP.g^{-1}$)	161.4	144.5	82.2	149	121.6	194.1	81.6	nm	nm	219.4	89.5	311.6	107	98	79.8	nm	169.8
P-OM ASOP ($\mu gP.g^{-1}$)	15	17.8	17.1	22.2	23	17.6	12.3	nm	nm	8	18.8	40	12	13.7	17.5	nm	11.3
P-OM ROP ($\mu gP.g^{-1}$)	156.2	234.5	216.2	245.1	235.7	245.1	199.1	nm	nm	156.16	206.6	112.4	238.1	186	159.8	nm	218.5
Total P ($\mu gP.g^{-1}$)	357.6	511.9	332.5	461.2	412.9	527.4	337.1	nm	nm	487	351.1	642.5	425.9	318	298.7	nm	470.8

nm: no measurement.

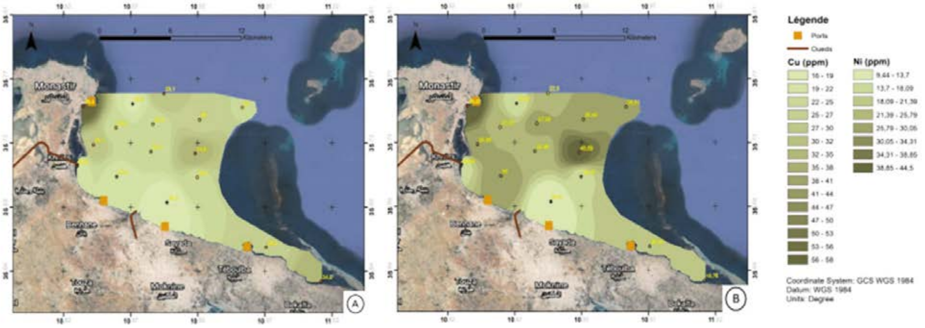

Figure 3 – Spatial distribution of MTE in Monastir Bay surface sediments : (A) Cupper (Cu in ppm), (B) Nickel (Ni in ppm).

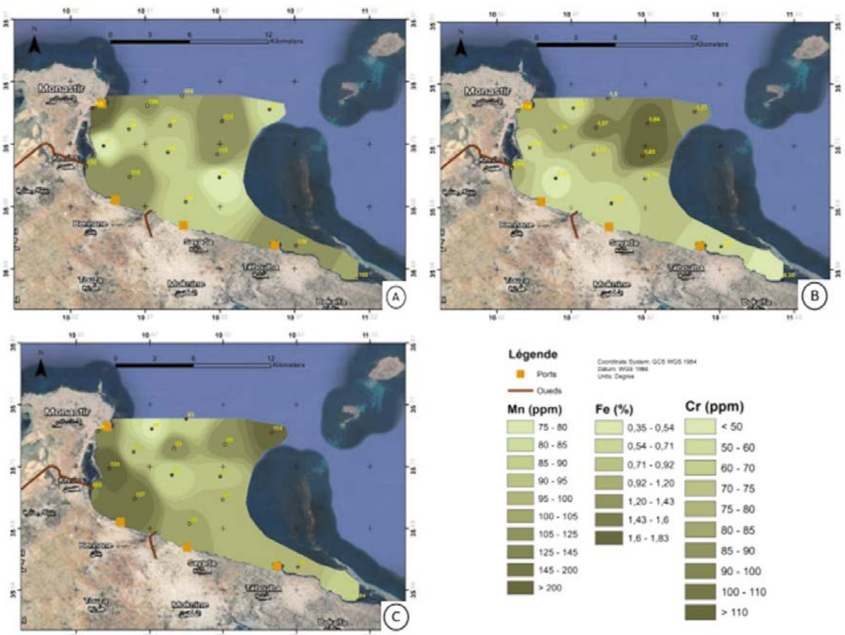

Figure 4 – Spatial distribution of MTE in Monastir Bay surface sediments: (A) Manganese (Mn in ppm), (B) Iron (Fe in %), (C) Zinc (Zn in ppm).

The manganese, chromium and iron concentrations vary between 75 and 308 ppm, 49 and 134 ppm and 0.3 and 1.8 % (i.e. between 3 000 and 18 000 ppm) respectively (Figure 4). The highest concentrations measured in the sediments of Monastir's Harbour and off this area forming a gradient of enrichment of the sediments towards the east of the Bay, of these three metals. The sediments in line with the Khniss drain also have the highest concentrations of Mn, Cr and iron in this bay and they are more polluted in these MTE than in other Tunisian coastal areas.

The concentrations of lead (4 - 56 ppm) and zinc (37 - 123 ppm) - figure 5 - are also maximum in the sediments of the port of Monastir and offshore with an accumulation gradient to the east. Their concentration in sediments were high in 2015 [13], and then decreased and are of the same order as the measurements of [7]. The lead values in the sediments of the Bay of Monastir are comparable to those obtained along the coast of Sfax; higher than near the coast of Gabes and slightly lower than in the Gulf of Tunis. The zinc concentrations are similar to those of the Sfax coast (39 - 117 ppm) but seem much lower than those of the Gabes coast in 2013 and the Gulf of Tunis where the maximum values reach 7165 ppm and 451 ppm respectively.

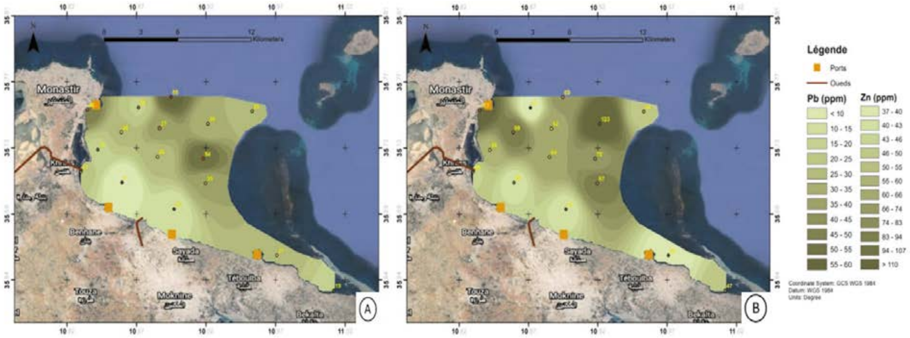

Figure 5 – Spatial distribution of MTE in Monastir Bay surface sediments: (A) Lead (Pb in ppm) and Zinc (Zn in ppm).

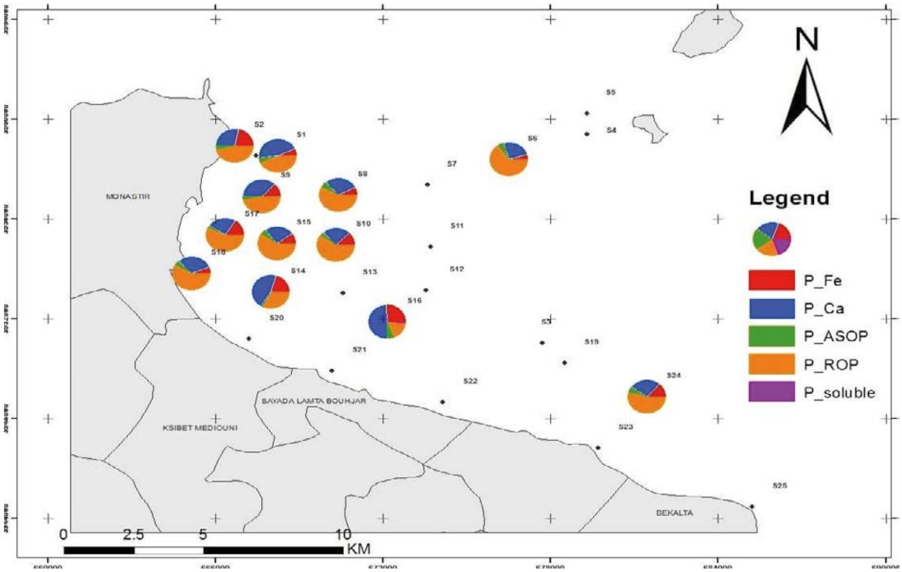

Figure 6 – Spatial distribution of phosphate particulate fractions in Monastir Bay surface sediment.

The total phosphorus concentrations in the surface sediments of the Bay of Monastir vary between 300 and 643 µg/g (Table 1), values in the ranges of those measured in other coastal ecosystems in Tunisia: Tunis Lake and the Korba [30] or Bizerte Lagoons [26]; but much stronger than in other Mediterranean coastal ecosystems in Italy (397 tot. P µg/g, [35]) or in Greece (205 tot. P µg/g, [25]).

Chemical fractionation of phosphorus has identified four main forms of particulate P.: inorganic P. bound to iron hydroxides (P-FeOOH) and carbonates (P-CaCO3) and organic P. bound to biodegradable MO (ASOP) and refractory (ROP) – Figure 6 – It appears that the dominant form is ROP (52 %) and P-CaCO3 (32 %), this distribution of forms of particulate P. is related to geochemical characteristics sediments: richness in OM (high TOC and loss on ignition contents) and in carbonates. The spatial distribution of the ROP is explained by the high concentrations of MOP at the level of the Port, discharges from the Khniss Canal and north of the Bay (aquaculture basins) and the presence of the aquatic plants (Posidonia meadow).

The PCA results (Figure 7), loading plots of the F1 factor highlight two groups of sediment sampling stations: the first one relatively enriched with P-FeOOH and P-CaCO3 fractions and relatively poor in ROP and TOC, the second one, at the opposite: poor with P-FeOOH and PCaCO3 fractions and enriched in ROP and TOC.

In contrast, the loading plots of the F2 factor demonstrate a trend with sediment stations enriched with organic matter and total phosphorus in opposite with others sediment stations poor in organic matter and total phosphorus. Then organic matter is clearly the phosphorus source. To summarize, the PCA results show two trends: surface sediment enriched with Fe and TOC versus an other one enriched with carbonate but with weak concentration of Fe and TOC.

	P-CaCO$_3$	ASOP	ROP	TP	Silt-Clay	OM	TOC	CaCO$_3$	Fe
P-FeOOH	0.441	-0.137	-0.760	0.624	-0.159	0.021	-0.187	0.288	-0.081
P-CaCO$_3$		-0.111	-0.907	0.311	0.173	-0.275	-0.351	0.175	-0.369
ASOP			0.013	0.165	-0.310	-0.167	-0.305	-0.317	0.181
ROP				-0.533	-0.009	0.211	0.379	-0.217	0.278
TP					0.147	0.440	-0.181	-0.128	0.255
Silt-Clay						0.484	0.511	-0.007	0.223
OM							0.576	-0.323	0.446
TOC								-0.336	0.613
CaCO$_3$									-0.495

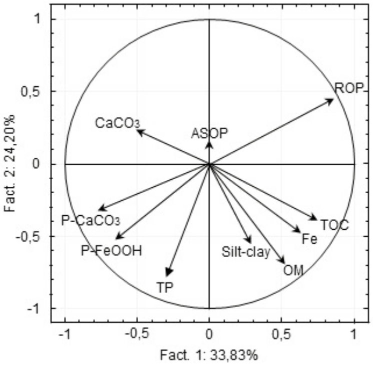

Figure 7 – The Principal Component Analyse: Loading plots of the two factors (F1 and F2) and inter-elemental correlation table.

Discussion

The sediments of the coastal areas, by their very adsorbent silto-organic nature, are a sink of MTE. They therefore constitute markers of the various human activities of this Bay [11], [37]. Indeed, the chemical and metallurgical industry, present all along the coast, from Monastir to Ksibet El Mediouni, explains the high concentrations of chromium and iron [34].

On the other hand, the harbour's activity which uses fuels, anti-corrosion and anti-rust paints (antifouling), explains the accumulations of Pb, Zn and Sr in the sediments of the four harbours of this Bay. The textile industry, with factories located at Oued Essouk, produces Cu, Zn, Pb and Sr which accumulate in the surface sediments. The determination of the degree of pollution of the surface sediments in the bay, by comparison with a standardized standard [4], shows that the sediments, in 2017-2020, present acceptable MTE concentrations, below this international sediment quality standard (Dutch criteria, [4]).

However the strong increase in MTE in 5 years (2015-2020) is worrying because it denotes the exponential increase in MTE discharges following activities industries and harbours in full expansion in the Bay of Monastir. Furthermore, the PCA results demonstrate the environmental sediment conditions necessary for the phosphorus dynamic (i) well oxic sediment with basic pH conditions enhance the FeOOH formation and consequently the phosphorus fixation on FeOOH or CaCO3 in sediment (P-FeOOH, P-CaCO3) whatever the sediment lithology; (ii) anoxic and acidic sediment enhance the formation of ROP (Residual Organic Phosphorus) in correlation with the presence of algae (Posidonia, [8]) in the sediment.

Conclusion

This geochemical study shows that the spatial distribution of particulate organic matter and MTE in the sediments, can be explain by discharges (wastewater and harbours activities) but also by morphological (shallow bay) and hydrodynamic (weak current sailor) specific. This confinement of the Bay accelerates the storage of pollutants in the sediments. Even if the concentrations of MTE remain within acceptable thresholds in view of international legislation; their sharp increase in a few years, from 2015 to 2022, suggests that this metal pollution will soon reach a critical state.

Acknowledgements

The authors would like to thank Khaled Nasri (University Tunis El Manat, Tunisia) and David Moussa (University of Rouen-Normandy-France) for granulometric, geochemical and phosphorus fractionation analysis. We thank also Zoe Reignier for helping in the english writing. We acknowledge particularly the anonymous reviewers for their helpful recommendations.

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