



Assessment of metallic pollution in the waters, suspended particulate matter, and surface sediments of the central coastal area of the Gulf of Gabès, Mediterranean Sea

Dorra Gargouri^{1*}, Neila Annabi-Trabelsi^{2**}, Qusaie Karam³,
Mohammad Ali³, and Habib Ayadi²

*1*Laboratoire de Modélisation des Systèmes géologiques et Hydrologiques (LR16ES17), Université de Sfax, Route Soukra Km 3.5, B.P. 1171, CP 3000 Sfax, Tunisia

*2*Laboratoire Biodiversité Marine et Environnement (LR18ES30), Université de Sfax, Route Soukra Km 3.5, B.P. 1171, CP 3000 Sfax, Tunisia.

3 Environment and Life Sciences Research Center, Kuwait Institute for Scientific Research, 13109 Safat, Kuwait

*Corresponding author, Email address: dorrargargouri@yahoo.fr

**Corresponding author, Email address: neila.trabelsi@isbs.usf.tn

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Abstract

Concentrations of trace metals (Cd, Cu, and Zn) in seawater, suspended particulate matter (SPM), and sediments near the coasts of the Gulf of Gabès were measured in spring 2014. The trace metal concentrations were confirmed to be high, especially in surface sediments and seawater. The levels of contamination and spatial distributions of the selected metals in these phases are mainly influenced by the anthropogenic impact concentrated in the urban area and the influence of hydrodynamic conditions. In the surface sediments and SPM, the high significant positive correlation between the selected trace metals confirms their similar behavior. In seawater, Cd presents a specific behavior attested by its different distributions. This result, which is confirmed by the lower correlations between Cd and the other trace metals (Cu and Zn), could be explained by the influence of pH and salinity on this metal. Partition coefficients $\log(K_d)$ between dissolved and particulate phases showed that Cu metal had a strong affinity for SPM. The high significant positive correlations between the partition coefficients indicate the similarity of partition behaviors of dissolved and particulate phases for these metals.

1. Introduction

Industrialization along the coastal areas in the developed and developing countries is a serious threat to the marine ecosystems and their biota [1]. Wastewater discharged to the sea containing large quantities of trace metals have become a subject of many environmental studies, because of their environmental persistence, biochemical recycling, and potential ecological risks [2,3]. In the aquatic environment, trace metals are detected in several forms: water-soluble species, suspended, and sediment phases [4]. Many aquatic organisms assimilate dissolved metals directly while particulate or sedimentary metals are accumulated by filter-feeding organisms [5-7]. In coastal environments, trace metals can interact in different phases and present different biochemical behavior due to their response to environmental changes. Therefore, it is important to investigate the levels and distribution of trace metals in seawater, suspended particulate matter (SPM), and sediment.

Several studies have focused on trace metal contamination in both seawater and sediments in the Gulf of Gabès [8-16]. Other studies were concerned about the impact of trace metals on marine fauna and flora in the same water mass [17-22]. Also, there are several studies concerned with possible solutions for the reduction of metal contamination in the marine ecosystem in the same water mass [23-26]. However, these investigations did not consider the correlation of trace metals in seawater, SPM, and sediments.

The concentrated industrial development close to the shoreline in Gabès City has generated a high anthropogenic pressure, causing a severe level of pollution [10]. The industrial activities are various, including food, mining, energy production unit, building material, leather tanning, textiles, and phosphate and fertilizer processing. These industries are mainly located at the site of the Tunisian Chemical Group (TCG), which is near the commercial harbor. The phosphate fertilizer industry, in which phosphate rock is treated by sulfuric acid to produce phosphoric acid is the main activity responsible for metal contamination of the seawater in the Gulf of Gabès. This process generates a large amount of phosphogypsum (PG) which is a solid residue. Phosphogypsum contains a high concentration of trace metals [10]. The liquid and solid effluents are discharged by TCG in the same channel that collects urban wastewater. The channel discharges the wastes into the sea at the southern dike of the commercial harbor (Figure 1). The present study aims to quantify the levels and distributions of three selected trace metals; cadmium (Cd), copper (Cu), and zinc (Zn) in these three phases, their relationships, and related environmental parameters in Gabès coast (Central area of the Gulf of Gabès, Figure 1).

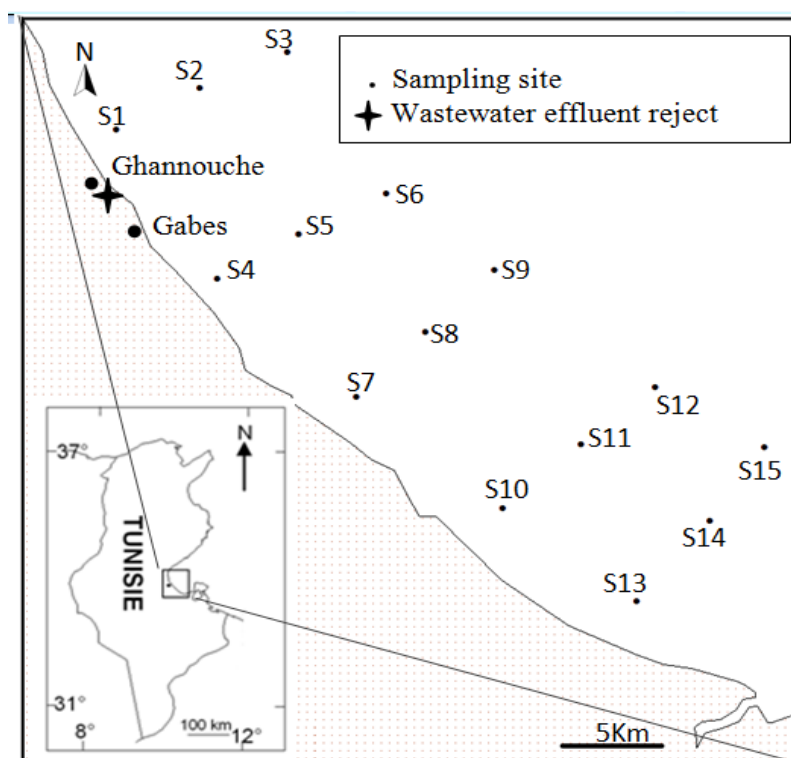


Figure 1. Location of sampling sites in Gabès coast

2. Material and Methods

2.1 Sampling and pretreatment

Surface sediments, SPM, and surface seawater were collected in 2014 from 15 stations in Gabès coastal waters (Figure 1). Sampling was performed in 5 km spacing sites through 5 radials perpendicular to the coastline, which were spaced about 10 km. Immediately after collection, samples were placed in

polyethylene bags, refrigerated, and transported to the laboratory. SPM was recovered by filtration. Sediment and SPM samples were dried in an oven at 60°C.

2.2 Environmental parameters of the surface seawater

Temperature, salinity, and pH were measured at each station immediately after sampling using a multi-parameter kit (Multi 340 i/SET). The concentration of suspended matter was determined by measuring the dry weight of the residue after filtration of 0.5 l of seawater with a Whatman GF/C membrane filter. Concentrations of nutrients (nitrite, nitrate, ammonium, and orthophosphate) were determined with a Bran and Luebbe type 3 autoanalyzer.

2.3 Metal analysis

The dry samples (< 63 µm sized grains of sediments) were digested with HNO₃–H₂O₂–HCl according to the Environment Public Authority of the United States (US EPA) method 3050B [27] then filtered, and made up to volume with deionized water. Afterwards, the selected trace metals were determined by an air–acetylene flame Atomic Absorption Spectrometry (AAS, Perkin Elmer model). Metal concentrations were expressed by mg per kg dry weight (DW) for sediment and SPM and micrograms per a liter of water.

2.4 Geographic Information System (GIS)

The distribution patterns of the nutrients and the selected trace metals in surface sediments, seawater, and SPM were depicted through the inverse distance weighed (IDW) interpolation using Arc Map 10.3.

2.5 Statistics

Pearson's correlation analysis was adopted to evaluate potential relationships between the levels of the selected trace metals in seawater, SPM, and sediments and their correlations with environmental parameters.

3. Results and Discussion

3.1 Environmental parameters

Salinity of seawater varied from 36 to 40 psu and decreased southward. Unlike salinity, pH of the surface seawater did not significantly vary (7.68 to 7.83). The SPM concentrations ranged from 5.9 to 20.9 mg L⁻¹. The significant negative correlation between salinity and SPM suggests essentially terrigenous origin (sediment resuspension). High levels of nitrites, nitrates, ammonium, and phosphates indicate the eutrophic state of the seawater in this region. These nutrients show similar spatial distributions (Figure 2). Higher values recorded for nutrients near the wastewater effluent attest to their anthropogenic origin. The significant extension of nutrients to the south confirms the influence of the dominant longshore current originated in the south [10].

3.2 Trace metal in different phases

3.2.1 Trace metals in marine surface sediments

The mean concentrations of the trace metals in surface sediments decreased from Zn to Cd and then to Cu (Table 1). To provide more information about contaminant toxicity of the studied sediments, the measured metal concentrations were compared with the most widely used sediment quality guidelines (SQGs) established by the National Oceanic and Atmospheric Administration [28]. Effects range low (ERL) and effects range medium (ERM), were proposed to determine whether the metals in sediments pose a threat to the aquatic ecosystems [28, 29, 30].

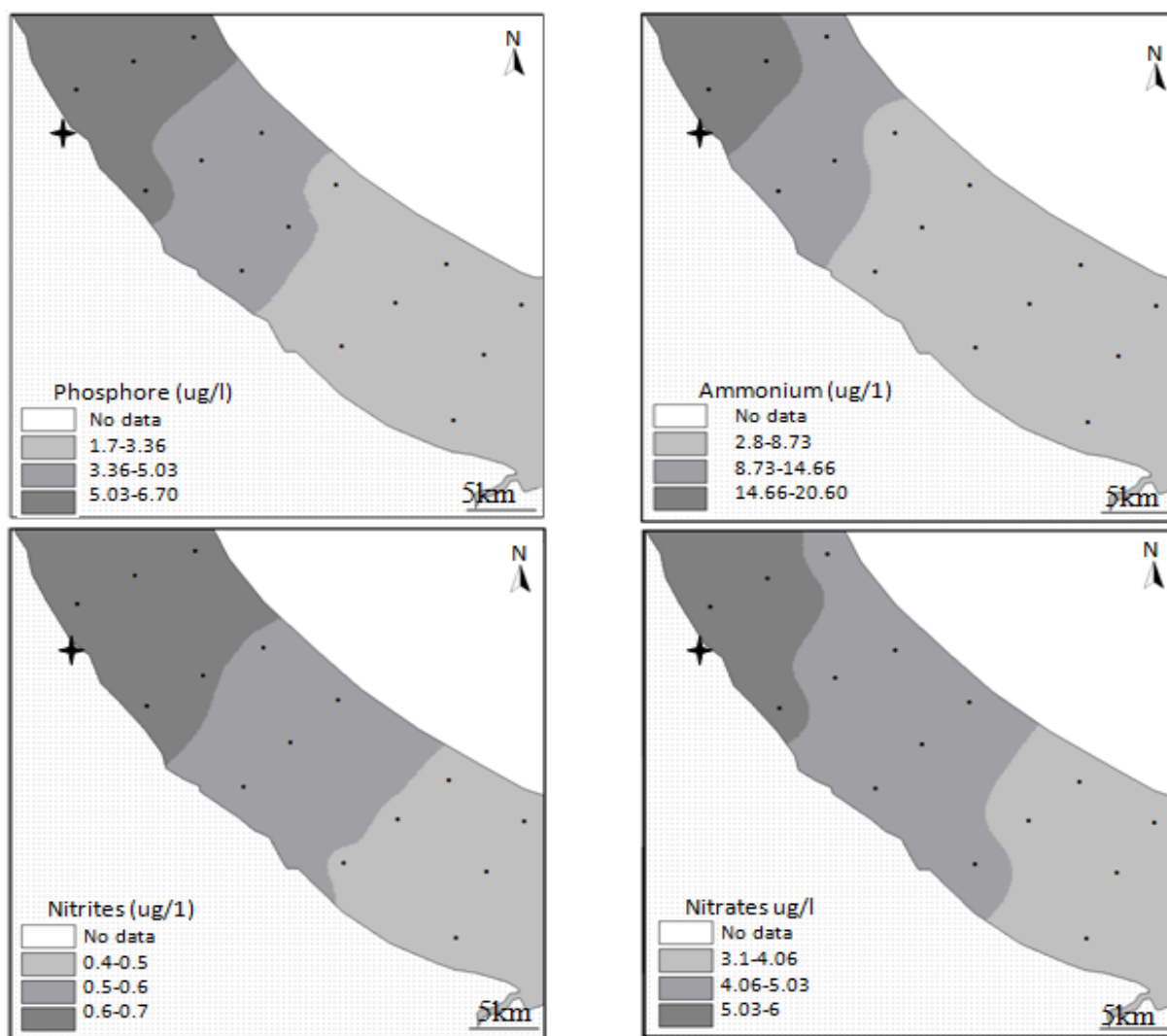


Figure 2. Distribution maps of nutrients (Phosphore, Ammonium, Nitrites, Nitrates)

The trace metals concentrations of the majority of the collected samples were higher than the effects range low (ERL) (Table 2). For comparison purposes, the range concentrations of trace metals in surface sediments reported in some worldwide coastal surface waters are shown in Table 2. Results showed that the coastal waters of the Gulf of Gabès are highly contaminated. This might be related to the discharge of large amounts of untreated PG into the sea. The spatial distribution of selected trace metals in surface sediments presents a similarity (Figure 3), which is confirmed by the high significant positive correlation between trace metals (> 0.9). These results highlight the similar source and behavior of the selected trace metals in this study.

Table 1. Mean, max and min values and standard deviation (SD) of heavy metals in surface sediment (mg kg^{-1} DW), seawater ($\mu\text{g l}^{-1}$) and SPM (mg kg^{-1} DW) in Gabes coast.

	Sediment			Water			SPM		
	Min	Max	Mean \pm SD	Min	Max	Mean \pm SD	Min	Max	Mean \pm SD
Cd	20	732	198.7 \pm 198.6	17.1	201.8	48.1 \pm 47.2	1	5	3.16 \pm 0.9
Cu	20	490	136.5 \pm 136.3	29	160	70.5 \pm 42.1	2.2	32	26.3.5 \pm 3.2
Zn	200	3920	1140.4 \pm 1050.5	60	660	259.3 \pm 190.4	104	157	128.4 \pm 16.2

Table 2. Trace metal concentrations (mg kg^{-1} dry weight) in surface sediment of Gabès coast compared to other coastal areas compared to the American sediment quality guidelines.

Location	Cd	Cu	Zn	References
This study	20-732	20-490	200-3920	
Sfax Kerkennah plateau (Tunisia)	0.11-22	3.25-22	34.98-181	[31]
Tetouan Coast (North of Morocco)	0.1-0.3	2.8-29.11	63.7-115.3	[32]
Gadiz bay (Spain)	0.02-0.4	34.4-61.8	21.7-161	[33]
Yellow sea (China)	0-8.21	5.93-25.7	21.9-96.2	[34]
SQGs (ERL-ERM)	1.2 -9.6	34-270	150-410	[28]

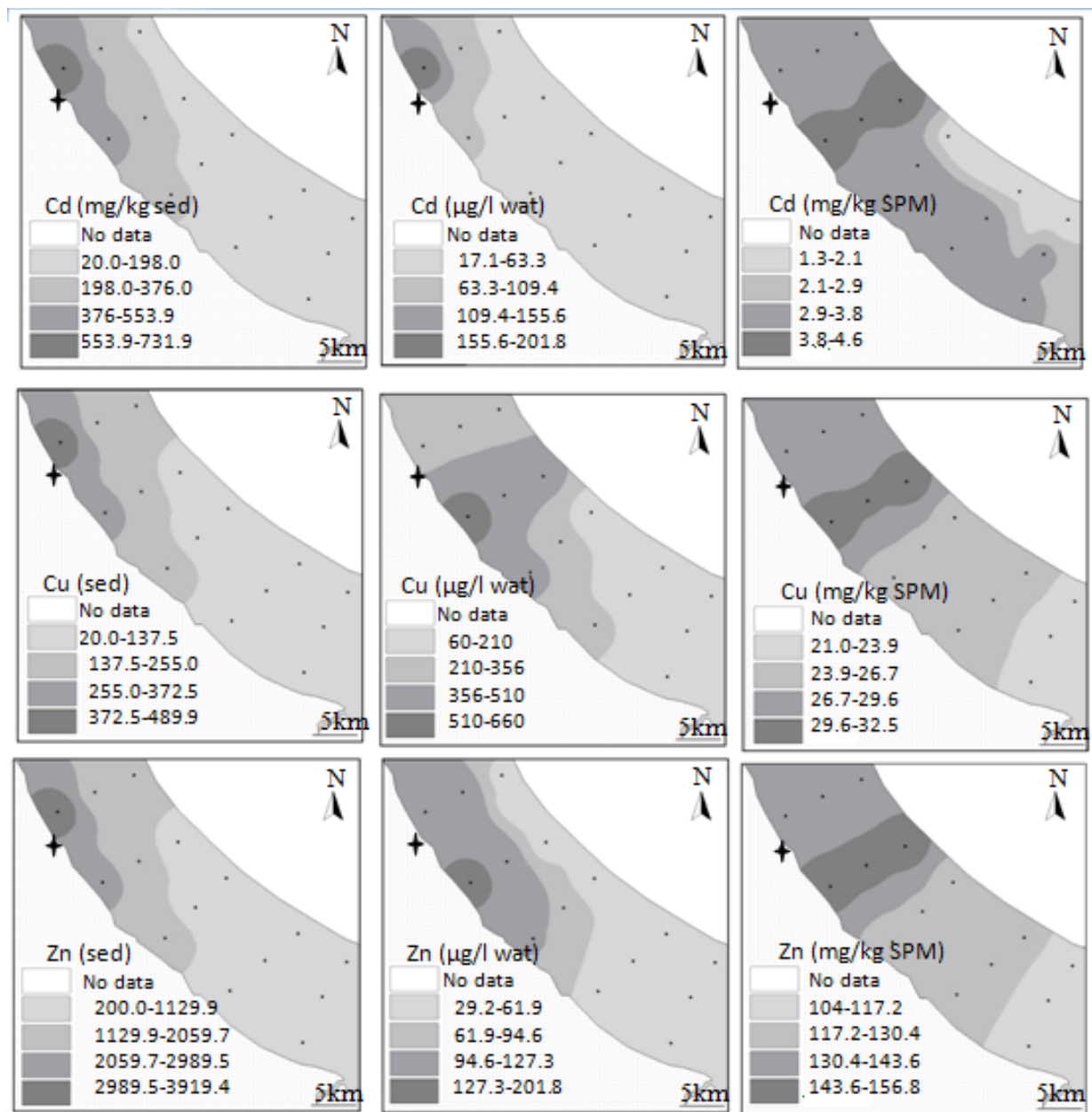


Figure 3. Distribution maps of the selected heavy metals in surface sediment, seawater, and SPM

Spatial variations of sediment trace metal contamination were correlated to two major factors. The first corresponds to the impact of a major pollution source which corresponds to the wastewater effluent reject. Higher values, recorded near the wastewater effluent, indicate their anthropogenic origin. The

second factor is the influence of hydrodynamic conditions. The significant extension to the south is influenced by the dominant longshore current oriented toward the south [10].

3.2.2 Trace metals in surface seawater

The concentrations of trace metals in the seawater are listed in Table 1. The mean concentrations of the studied trace metals decreased in the order of Zn, Cu, and Cd. Copper and Zinc concentrations exceeded the USEPA seawater quality criteria for the protection and survival of aquatic fauna and flora and their uses [27], (Table 3). However, the majority of the samples had relatively low Cd concentrations. Results obtained in this study exceeded by far those obtained in other coastal areas worldwide (Table 3). The spatial distribution of Cu and Zn showed similarity for both metals. This is confirmed by a high correlation coefficient (0.8). Cadmium presented a different distribution from those of Cu and Zn confirmed by the lower correlations between Cd and Cu, on one hand and Cd and Zn, on the other hand. These results suggest that Cd responds differently to environmental conditions.

Table 3. Trace metal concentrations ($\mu\text{g l}^{-1}$) in surface seawater of Gabès coast compared to other coastal areas based on the American seawater quality criteria.

Location	Cd	Cu	Zn	References
This study	17-201	29-160	60-660	
Sfax Kerkennah Plateau (Tunisia)	0.11-3	0.8-5	0.3-17	[31]
Malaga Bay (Spain)	ND-0.48	ND-2.92	-	[35]
Aughinish Bay (Ireland)	0.015-0.061	0.64-3.84	0.35-2.22	[36]
Safety limits ^d	42	4.8	9	[27]

ND: Not Detectable

3.2.3 Trace metals in SPM

The concentrations of trace metals in the SPM are listed in Table 1. The mean concentrations of the studied trace metals decreased in the order of Zn, Cu, and Cd. Unlike the other fractions, particulate metals were generally more or less high compared to those obtained in other coastal areas worldwide (Table 4). The spatial distribution of selected trace metals in SPM presented good functional similarity, which is certified by a significant correlation between the three trace metals. However, the correlation observed between the trace metals in the particulate phase is lower than the correlation observed in the sediment samples (0.69-0.77). The correlation outcome suggests the contribution of biological processes in the distribution of trace metals.

Table 4. Trace metal concentrations ($\mu\text{g g}^{-1}$) in SPM of Gabès coast compared to other coastal areas worldwide

Location	Cd	Cu	Zn	References
This study	1- 5	2.2 - 32	104-157	
Thermikos bay (Greece)	0.6- 66	32-81	60-244	[37]
Zhanjiang Bay (China)	11.96-39.54	15.6-47.17	120.5-482.6	[38]
Uyo, Niger Delta (Nigeria)	0.45	18.36	15.04	[39]
Seven Estuary (United Kingdom)	0.32-0.49	29.8-37	191-238	[40]

3.3 Partition of trace metals between dissolved and particulate phases.

The partitioning of a metal between particulate ($> 0.45 \mu\text{m}$) and dissolved ($< 0.45 \mu\text{m}$) phases is commonly quantified by the distribution coefficient K_d . This coefficient is the ratio of the particulate metal concentration ($\mu\text{g.g}^{-1}$) to the dissolved metal concentration (mg.l^{-1}) [41-43]. A higher log (K_d)

value indicates a stronger affinity between the metal and suspended particles, and a lower $\log(K_d)$ value indicates that more metal exists in the dissolved phase [44]. The high mean $\log(K_d)$ value for Cu was 26.5, which affirms that this metal has a strong affinity for SPM while Cd and Zn, with low $\log(K_d)$ values (-1.02 and -0.20, respectively), thus, had low affinities for SPM. Environmental parameters can explain this speciation. Furthermore, high significant positive correlations (> 0.85) were found among the partition coefficients of the selected trace metals, indicating that the partition behaviors are similar between dissolved and particulate phases for these metals. The positive correlation between partition coefficients of trace metals and pH can be explained by the fact that acidic pH favors the dissolution of trace metals.

3.4 Relationships among sedimentary metals and environmental parameters

The significant positive correlation between the trace sedimentary metals and nutrients (nitrite, nitrate, ammonium, and orthophosphates) verifies their common origin, which corresponds to the wastewater effluent. The correlation between the selected trace metals with pH and salinity was not significant.

3.5 Relationships between dissolved metals and environmental factors

Same results were observed for dissolved metals except for Cd, which showed a significant negative correlation with salinity and pH. This result confirms that Cd has different behavior than Cu and Zn.

3.6 Relationships among particulate metals and environmental parameters

The significant positive correlation of particulate Cu and Zn with nutrients (nitrite, nitrate, ammonium, and orthophosphate) attests their anthropogenic origin. The absence of a significant correlation between particulate Cd and nutrients confirms the specific behavior of this metal. On the other hand, there is no significant correlation between the selected trace metals and the other environmental parameters.

4. Conclusion

High levels of nutrients indicate the eutrophic state of the coastal waters of the Gulf of Gabès. The concentrations of trace metals in sediments and seawater are higher than that of the American standards (US EPA, 1999). These contaminants were introduced to the environment by the wastewater effluent. Industrial activity and hydrodynamic conditions are the major factors that influence the level of contamination and spatial distributions of the selected metals in sediment, water, and SPM in the coastal waters of the Gulf of Gabès. The similar distribution and the high significant positive correlation between Cd, Cu, and Zn in sediments showed their similar mechanisms of metal binding to sediment surfaces. The spatial distribution of the three trace metals in SPM also presents a good similarity. Noticeably, the lower correlation between particulate trace metals suggests the contribution of biological processes in the distribution of trace metals. Unlike sediment and particulate phases in seawater, Cd presents a different distribution from those of Cu and Zn. These results, which are confirmed by the lower correlations between Cd and the other two metals (Cu and Zn), could be explained by the specific behavior of Cd concentration of which is controlled by pH and salinity. Furthermore, the strong affinity between Cu and SPM can be explained by environmental parameters which could favor bioaccumulation of this metal. Consequently, it was demonstrated that metals in the different phases interact with many factors with different intensities. Therefore, further studies of trace metals in the three phases are recommended in such environments because it provides a better comprehension of their behavior. This should facilitate the search for possible solutions that would allow the reduction of contamination in the marine environment.

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